International Conference on
HYPERFINE INTERACTIONS STUDIED
IN NUCLEAR REACTIONS AND DECAY
Uppsala, Sweden, June 10–14, 1974

CONTRIBUTED PAPERS

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CONTRIBUTED PAPERS

Editors:
E. Karlsson and R. Wippling
Editors' comments:

By the last hour for delivery of manuscripts to the printers we had accepted a total of 136 contributions for the conference. Their large number and high quality form a manifestation of the present activity in the field and the power of hyperfine interaction methods in solving problems and displaying new phenomena in central areas of physics.

The task of editing these contributions in a few days' time has not been easy, particularly in cases where they contain elements of interest both for the nuclear physicist and the solid state physicist. In the assignment of contributions to the different sections we have tried to follow the general philosophy of the conference program:

1. an introductory part dealing with the basic mechanisms behind hyperfine interactions in free atoms and in the condensed states
2. a section devoted to physical processes occurring around probe nuclei in experimental situations specific for nuclear radiation detection methods (in this area we include the problems in interpretation of the effects observed and the comparability with non-radiative methods)
3. a main area - to be discussed when interpretational problems (hopefully) have been settled - with the extraction of
   a) nuclear moment information
   b) solid state or other extranuclear information from the measured data
   and the implications of these results for our theoretical descriptions of the respective fields
4. a novelty section, where development of methods as well as new applications of nuclear radiation methods in hyperfine interactions are included.

A specific problem has been to assign a number of papers containing information of type (2) and (3b) to appropriate sessions, since they are partly of a methodological nature and partly form studies of solid state environments. In case of hesitation such contributions have been placed in the first of these two categories. We apologize to the authors if we have misinterpreted their intentions with these and other contributions and hope that mistakes can be corrected at the time of the conference. As an example of a complex situation we may mention the contribution IV. 57, which deals with diffusion, magnetic relaxation and quadrupole moments.

The conference committee is aware that late contributions may reach us even after to-day's date; such post-deadline papers will be made available to the participants at the beginning of the conference.

We have had the privilege of being the first ones to look at the collected contributions. From our impressions at this point of survey we expect an exciting exchange of ideas within this truly interdisciplinary field and we hope that the conference will be profitable to all participants!

Uppsala, 19 April 1974

Erik Karlsson
Roger Weppling
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- A NEW SOURCE OF INFORMATION
I. INTRODUCTION TO μSR

Muons originate and decay in the maximally parity-violating reactions $\bar{u}\nu$ and $u\bar{e}v$. The former reaction produces muons of definite helicity, which can be collected into beams which are nearly 100% polarized. In the latter reaction the $\bar{e}^-$ is preferentially emitted along (for $\bar{u}^+$) or opposite (for $\bar{u}^-$) the muon spin, in a pattern of the form $D(\theta) = 1 + a \cos \theta$, where $\theta$ is the angle between $\bar{u}^+$ and $\bar{e}^-$ direction, and $a$ is an asymmetry parameter—a function of $\bar{e}^-$ energy—whose average over energy is $1/3$. The weak interaction thus provides experimenters with a source of highly polarized Dirac particles and a convenient way to detect their polarization.

A typical experimental technique is to stop an energetic muon beam in a target, starting a "clock" as a muon enters through scintillation counters, and stopping the "clock" when an electron is counted in a second scintillator array. When a magnetic field is applied transverse to the muon polarization, its spin will precess at the Larmor frequency, causing the electron detection probability to become an oscillatory function of time as the muon spin sweeps past the electron telescope. The histogram of measured times is generally fitted to the form

$$N(t) = N_0 \left[ e^{-t/\tau} \left( 1 + A e^{-t/\tau_2} \cos \left( \omega_L t + \phi \right) \right) + B \right], \tag{1}$$

where $N_0$ is a normalization factor, $\tau$ is the muon lifetime ($2.2$ usec for $\bar{u}^+$ or free $\bar{u}^-$), $A$ is the apparent initial asymmetry, $\tau_2$ is a transverse relaxation time (accounting for slow depolarization by random local fields); $\omega_L$ is the muon Larmor frequency, $\phi$ is the initial phase of the precession, and $B$ is a time-independent background.

This technique, which is clearly analogous to PAC methods, has been dubbed "μSR" (for Muon Spin Rotation/Relaxation) in an attempt to suggest applications analogous to those of NMR and ESR. Each of the fitted parameters in Eq. (1) is of special interest in certain studies, but $T_2$ and $\omega_L$ are most interesting for the study of magnetic materials, since they give a direct quantitative measure of the local field $B_H$ at the site of the muon, and its inhomogeneity $\Delta B_H$:

$$\omega_L = \gamma_H \mu_B H \text{ and } \frac{1}{T_2} = \frac{1}{2} \gamma_H \Delta B_H,$$ \tag{2}

where $\gamma_H = 0.85 \times 10^5$ rad/sec-G (period = nsec at 1 kG).

μ+ SR IN MAGNETIC MEDIA

Prof. Yamazaki will discuss the applications of μ+SR (spin studies with negative muons); we are concerned here with positive muons and μ+SR. The $\mu^+$ behaves in matter as if it were a light proton ($m_\mu/m_p = 8.9$); thus in most crystals it will usually, upon thermalizing, take up an interstitial position. It then experiences a local field which has the following contributions:

$$B_\mu = B_{\text{ext}} + B_{\text{dm}} + B_L + B_{\text{dip}} + B_{\text{HF}},$$ \tag{3}

where $B_{\text{ext}}$ is the external applied field, $B_{\text{dm}}$ is the sample demagnetizing field, $B_L$ is the field due to the induced magnetic charges on an imaginary spherical "Lorentz cavity" centered about the $\mu^+$ site, $B_{\text{dip}}$ is the field due to local dipole moments within the Lorentz sphere, and $B_{\text{HF}}$ is a contribution due to contact interactions with polarized electrons.

In a ferromagnetic crystal below saturation, $B_{\text{ext}}$ is just cancelled by $B_{\text{dm}}$. $B_L$ has the value 3 smaller than the dimensions of one domain. $B_{\text{dip}}$ is usually calculable if the favored interstitial position can be identified. We can then obtain a measurement of $B_{\text{HF}}$, a quantity of basic interest to the physics of ferromagnetism.

μ+ SR IN WICEK

Measurements of $\omega_L$ and $T_2$ for positive muons in ferromagnetic Ni have been made by several groups.2 Over a range of temperatures, yielding the data shown in Fig. 1 for $B_L$ and $\Delta B_L$. The temperature dependence is essentially a Brillouin function with the same shape as the saturation magnetization curve, normalized to a zero-degree limit of $B_L(0) = 1440$ G.

In Ni, the $\mu^+$ can be expected to occupy an octahedral interstitial site with cubic symmetry. The local dipole contributions then cancel, giving $B_{\text{dip}} = 0$. The Lorentz cavity contribution to $B_L$ is just $B_L(0) = 1440$ G, leaving a hyperfine contribution of $B_{\text{HF}}(0) = -660$ G. This field is thought to be due to the contact interaction between the $\mu^+$ and polarized 4s-band conduction electrons.

Neutron diffraction studies3 have indicated an unperturbed interstitial electron polariza-
tation of -17, implying a local interstitial magnetization of $M_{loc} = -79$ G. An uncharged probe would thus experience a contact field of $B_{HF} = \frac{4}{3} M_{loc} = -560$ G. The agreement between this value and the measured field must be fortuitous, since the estimated screening length by 4s-band electrons is $\sim 0.6$ Å, leading to concentration of electron density at the $\mu^+$ by a factor of $\sim 5$ over the unperturbed interstitial electron density. If the conduction electrons contributing to the screening had the same polarization as the band from which they come, one would expect a contact field of $B_{HF} = -3300$ G. To explain the much lower measured value we postulate a simplistic model in which energy minimization favors equalized screening contributions from spin up and spin down electrons.

**$\mu^+$ SR IN IRON**

Unlike Ni, Fe has two electrostatically identical but magnetically different interstitial face-centered sites. One has a $B_{dip}$ contribution of $+18800$ G, while the other has $B_{dip} = -9400$ G and occurs twice as often. Thus at high temperatures the $\mu^+$ diffuses between sites fast enough for $B_{dip}$ to average to zero, but at low temperature this motional narrowing effect is absent, and the $\mu^+$ is rapidly depolarized. Preliminary attempts to "freeze" muons into distinct sites in a single crystal of Fe at 77°K have so far yielded inconclusive results.

High-temperature data for $B_{u}$ vs. $T$ can be extrapolated to $T=0$ to yield a value of $B_{u}(0) = + 4100$ G.

Using $<B_{dip}> = 0$ and $B_L(0) = \frac{4\pi}{3} M_\parallel(0) = + 7100$ G

in Eq. (3) gives $B_{HF}(0) = -3000$ G. Interpretation of this result is complicated by the highly structured spatial distribution of the interstitial magnetization in iron.

**$\mu$SR PROGRAMS**

The work reported here was done by Kossler et al.\(^1\) at SRRL and by the authors at the 184 in. Cyclotron in Berkeley. Related work has been done at JINR in Dubna. Extensive $\mu$SR programs are planned at SIN and TRIUMF in the near future.

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I. 2 POSITIVE MOONS IN SOLIDS: II. IMPURITY STATES IN SEMICONDUCTORS.
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\( \mu^+ \) AS AN IMPURITY

In the preceding paper we discussed the general technique of \( \mu^+ \)SR and the applications of \( \mu^+ \)SR for probing local interstitial magnetic fields in ferromagnets. Here we turn our attention to the behaviour of positive muons in semiconductors, where the \( \mu^+ \)SR technique can be extended to provide new information about impurity states.

The \( \mu^+ \), with its family resemblance to a proton, is perhaps the simplest impurity nucleus one can implant in a semiconductor. There has been some question in the past as to whether the implanted \( \mu^+ \) forms substitutional or interstitial states, and whether it remains a bare \( \mu^+ \), captures an electron to form a muonium (Mu) atom, or perhaps captures two electrons to form a \( \mu^+ \) ion. It has generally been agreed that a significant fraction must form Mu atoms, but it has not been clear whether these atoms would be shallow donors, with their electron wave functions spread over many lattice sites, or deep donors, with the entire atoms fitting into single lattice sites. The time scale for stability of such atoms has also been in question. Recent experimental results have partially resolved these issues.

THE MUON ATOM

In a free Mu atom in its ground state, the \( \mu^+ \) and \( e^- \) spins are coupled by a contact interaction of strength \( \Delta E_{HF}/h = \nu_0 = 4463 \text{ MHz} \), given by the splitting between singlet and triplet total angular momentum states:

\[
\Delta E_{HF} = \frac{g_e \mu_B g_\mu}{3} |m| \left( \mu^+ + e^- \right)
\]

where \( r \) is the mean radius of the atom, \( g_e, g_\mu \) are electron and muon g-factors, and \( \mu_B \) are electron and muon magnetons.

One can think of \( \Delta E_{HF} \) as the energy of interaction of the electron's magnetic moment with the "contact field" due to the muon, \( B_c = 1593 \text{ G} \). In weak applied fields \((B \ll B_c)\), the triplet state of Mu will precess at a frequency \( \nu_0 \). Since \( \nu_0/F = 1 \) has essentially the magnetic moment of an electron, but has twice its angular momentum, \( \nu_0 \) is about 1/2 of \( \nu_0 \), the free \( \mu^+ \) Larmor frequency — or about 103 times \( \nu_0 \), the free \( \mu^- \) Larmor frequency. This muonium precession can be observed directly by the \( \mu^+ \)SR method in fields up to \( \approx 200 \text{ G} \).

In stronger fields \((B \gg B_c)\), and in all fields to some extent, competition between the Zeeman couplings and the hyperfine coupling causes \( \nu_0 \) to split into two frequencies, equivalent to the transition frequencies \( \omega_1 \) and \( \omega_2 \), between energy eigenstates described by the Breit-Rabi formulae:

\[
\omega_1 = \frac{\omega_0}{4} + \omega_c, \quad \omega_2 = \frac{\omega_0}{4} + \omega_c
\]

where \( \omega_c = 1/2(\omega_e \pm \omega_\mu) \) and \( \omega_c = 2 \omega_\mu \). In low fields, the splitting between \( \omega_1 \) and \( \omega_2 \) is given by

\[
\Omega = \frac{1}{2} (\omega_1 - \omega_2) = \omega_c/\omega_e.
\]

DEEP-DONOR MUONIUM

Observations of the above-mentioned "two-frequency muonium precession" in pure, cold (77 K) single crystals of Ge and Si have proven that positive muons stopped in semiconductors do, under some circumstances, form long-lived, deep-donor interstitial Mu atoms. The splitting frequency \( \Omega \) has been measured and used in conjunction with Eq. (3) to extract the hyperfine frequency for deep-donor Mu in these crystals. The results are

\[
\nu_1(\text{Ge})/\nu_e(\text{vac}) = 0.56 \pm 0.01 \text{ and } \nu_1(\text{Si})/\nu_e(\text{vac}) = 0.45 \pm 0.02.
\]

Recalling Eq. (1), we see that Mu atoms in these crystals have radii about 1.2 times larger than the radius of Mu in vacuum; thus they are clearly deep-donor impurities. The "swelling" of the atom is due to screening by valence band electrons from neighbouring lattice sites: its magnitude gives new information about the crystals.

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Figure 1 shows two power spectra of precession frequencies, obtained by squaring Fourier transforms of experimental time histograms (with background and exponential decay removed). The upper spectrum is for fused SiO_2 at room temperature, and the lower spectrum is for mildly p-type Si at 77 K. Both are taken in an external field of 100 G. Units are arbitrary but consistent; the Si spectrum to the right of the dashed line has been vertically magnified by a factor of 10.

The peak near zero frequency in each spectrum is due to free \( \mu^+ \) Larmor precession.
the rightmost pair of peaks represent two-frequency precession of deep-donor Mu. Due to the weaker hyperfine coupling in Si compared to SiO₂, where \( v_\text{Si}(\text{SiO}_2) = v_\text{Si}(\text{vac}) \), the splitting is visibly larger in Si than in SiO₂.

**SHALLOW-DONOR QUANTUM**

Also evident in the Si spectrum are two "anomalous" peaks at intermediate frequencies. At first these signals caused great confusion, but studies of their field dependence revealed that they correspond to transitions \( \omega_{11} \) and \( \omega_{12} \) between Breit-Rabi energy levels of a modified Mu atom in which both \( v_\phi \) and \( g_\phi \) are quite different from their vacuum values. It was also found that \( v_\phi \) was anisotropic. The fitted values were: for the (111) crystal axis along the field, \( v_\phi/v_\phi(\text{vac}) = 0.0198 \pm 0.0002 \); for (100) along the field, \( v_\phi/v_\phi(\text{vac}) = 0.0205 \pm 0.0003 \); in both cases, \( g_\phi = 13 \pm 3 \).

The preferred interpretation of these results is that the two "anomalous" frequencies are due to shallow-donor Mu atoms, in which the electron wave functions must be superpositions of conduction band states which may have \( f \neq 0 \) and small, anisotropic effective masses.

**QUASI-FREE \( \mu^+ \)**

Deep- and shallow-donor Mu precession have only been observed in mildly p-type Si and Ge at low temperature; however, in all semiconductor crystals there appears a precession signal of some strength at the free \( \mu^+ \) Larmor frequency. Whether this signal is due to substitutional \( \mu^+ \), Mu⁻ (where the two electrons pair up to make a diamagnetic system), or some diamagnetic bound state of a \( \mu^+ \) with lattice defects (perhaps created in its stopping) is not known. However, the \( \mu^+ \) precession signal in p-type Si at 77 K and 4400 G appears to have two components: one long-lived (the same as is usually observed) and one which dies out within \( \sim 30 \) nsec. These two components must represent distinct products of the thermalization process, as explained below.

**SUMMARY**

All the precession signals mentioned above are present simultaneously in the Si spectrum shown in Fig. 1. Since these motions are out of phase with each other within a few nsec after the muons stop in the target, they must each represent an independent and isolated long-lived product of the original thermalization process, as indicated graphically in Fig. 2.

The amplitudes of the various precession signals are proportional to the respective fractions \( h, f', f'' \), and \( f'' \), which should sum to unity but are all functions of doping and temperature. Studies of their behaviour may reveal much about the formation and behaviour of hydrogen-like impurity states in semiconductors.

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I. 3

**Effective Hyperfine Fields on Muons in Dysprosium and Gadolinium Metals.**

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W. J. Resler, The College of William and Mary, Williamsburg, Virginia

Friedel has pointed out that the time is ripe for detailed theoretical and experimental studies of light impurities in metals. Hydrogen is the simplest such impurity, and positive muons which are ideal for time differential PAC studies, behave in many ways like a light isotope of hydrogen. Foy et al., have begun studies of positive muons in nickel and iron and obtained effective hyperfine fields of about 1500 G and 4000 G respectively. These results are in reasonable agreement with simple conduction electron polarization calculations.

The case of the rare earth ferromagnets is more complex because of the deep discrete levels giving rise to their ferromagnetic properties. We have begun studies of dysprosium and gadolinium metals using polarized muons from the SNL synchrocyclotron. Figure 1 shows decay position time spectra from muons stopped in dysprosium at room temperature and at 77°K. Both spectra were obtained in an external field of 1600 G. Figures 2a and b are Fourier transforms of these data after correcting for exponential decay and a constant background. In paramagnetic dysprosium the line at the applied field is observed. In ferromagnetic dysprosium at 77°K (T< Tc) a second broadened field about 200 G less than the external field is observed. This is interpreted as an effective hyperfine field on the muon of -200 G, which, correcting for the temperature dependent magnetization, yields H_{eff} (77°K) ≈ -305 G. The negative sign is opposite to that observed in iron and nickel, and the reduced magnitude relative to the transition metal ferromagnets is indicative of reduced conduction electron polarization in the rare earth case.

Figure 1c is a Fourier transform for gadolinium at room temperature. Fields in this case are observed at the applied field (1605 G) and reduced by about 100 G. A broad reduced line at a higher field is also apparent. These results may indicate some residual ferromagnetism above Tc as well as possible multiple sites. The results are of sufficient interest to warrant further study at various temperatures and applied fields.

4. Space Radiations Effects Laboratory, supported in part by the NSF and the Commonwealth of Virginia.

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**Figure 1.** Time dependent position emission from μ⁺ stopped in dysprosium. The upper curve is at 77°K, the lower at room temperature.

**Figure 2.** Fourier transforms of reduced precession curves (a) Dy at room temperature (b) Dy at 77°K. (c) Gd at room temperature.
I. 4. MUON PRECESSION AS A PROBE OF MAGNETIC FIELD DISTRIBUTIONS IN TYPE II SUPERCONDUCTORS.
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Positively charged muons are ideal microscopic probes of magnetic field distributions in solids in the range of a few gauss to some tens of kilogauss. The relevant muon properties are its large magnet moment (2.48), relatively long lifetime (2.2 μsec), zero quadrupole moment, and the large decay antiferromagnetic spin flip probability (μ^− → e^− + ve + vμ). In addition, muons are isomorphic to protons, the simplest possible impurity; can be implanted at vanishingly low concentrations and leave no residual contamination.

Type II superconductors, like type I materials totally exclude magnetic fields at low applied fields (The Meissner effect) but at intermediate fields remain superconducting while allowing field penetration as quantized magnetic filaments with vortices of persistent electrical currents flowing around them. As the fluxoid lattice spacing is large, muons randomly distributed in a superconducting lattice should sample a calculable spatial variation of magnetic field intensity.

Our initial experiments were performed using polarized muons from the SREL synchrocyclotron stopped in planar samples of niobium or lead-indium maintained at low temperature and aligned at an angle of 30° to an applied magnetic field. The time dependence of positron emission from stopped muons was measured at 0° and 90° to the incident beam. After correcting for the exponential decay and a constant 'background, the resulting precession spectra, such as those of Figure 1, are Fourier analyzed to obtain effective magnetic field distributions such as are shown in Figure 2.

Data taken as a function of external field and sample temperature have produced the following major observations:

(1) In all cases a Fourier component corresponding to the applied field is observed in the superconducting state, but greatly reduced in power from the normal state.

(2) The power at the applied field is almost independent of field for niobium, but is field dependent for Pb.90In.10

(3) Fourier components expected for a triangular fluxoid lattice are only observed for Pb.90In.10

(4) The spectra show some dependence on the magnetic field - cooling cycle.

Models to explain these observations are under study. The most satisfactory, thus far, involve rapid muon diffusion. Deduced diffusion coefficients are consistent with recent quantum theories of diffusion and the observed isotope effect on hydrogen and deuterium in niobium.

1. I. G. Ivanov and V. P. Smilga, Soviet Physics, JETP 26, 286 (1968).
2. Space Radiation Effects Laboratory, Newport News Va., supported in part by the NSF and the Commonwealth of Virginia.

Figure 1. Reduced time spectra for Pb.90In.10 in an applied magnetic field of 8000gauss. The left hand curve is at 11°K, the right hand curve at 3°K.
Figure 2. Fourier transforms of the spectra of Figure 1. Note the change of power from the normal to the superconducting state. The peak at 0.52 is due to the cyclotron extraction frequency.
In 1928 Breit\(^1\) predicted that the electron bound to a nucleus must possess a g-factor which is different from the free electron g-factor due to the relativistic motion of the bound electron. This Breit effect was first observed in 1961 by the Columbia group\(^2\) through accurate determinations of g-factors of bound muons in light elements up to \(Z = 16\), and their results were successfully interpreted by detailed calculations by Ford et al.\(^3\). Since the Breit effect is much larger in heavier nuclei, it is interesting to extend the g-factor measurements to high-\(Z\) elements. Three diamagnetic metals Zn, Cd and Pb were used as targets, as the muon polarization can supposedly be preserved during its lifetime in such diamagnetic metals.

The method employed is the uSR (Muon-Spin-Rotation) method, in which a time differential Larmor pattern of decay electrons in a transverse magnetic field was observed. A pion beam from the 184\(^{\mathrm{th}}\) cyclotron at LBL and a decay-in-flight muon beam were separated through a polyethylene absorber of appropriate thickness. A 2.5\(^\prime\) x 2.5\(^\prime\) x 8 to 9 g/cm\(^2\) target placed at the center of a 9\(^\prime\) Varian magnet was bombarded with the muon beam thus extracted. The time interval between the stopped muon and decay electrons was measured by means of a digital clock counter.

The bound muon in a diamagnetic metal of atomic number \(Z\) feels the effective field given by \(H_{\text{eff}} = H_0 \times (1 + K)(1 + \sigma)\), where \(K\) and \(\sigma\) are the Knight shift correction and diamagnetic correction for a \(Z' = Z - 1\) impurity, respectively. These two corrections were carefully estimated, and their details are listed in Table I. The experimental g-factor deviations from the free muon g-factor are plotted in Fig. 1, together with the calculated values. The dotted curve shows the relativistic correction for the point nucleus. The solid curve shows the calculated values by Ford et al., where the finite-size effect of the nucleus is taken into account. (The difference between the solid and broken curves is the nuclear polarization effect taken from Ref. 3.) From the figure we see that the g-factor deviations are appreciably large for high-\(Z\) elements. The observed results are well explained by the calculations by Ford et al.

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**Fig. 1** Summary of the present experiment.

The black-closed circles are the present results. The open-circles are taken from Ref. 2. Experimental data after the corrections for the Knight shift and diamagnetic shielding are plotted.

**Table I** Knight shift and diamagnetic shielding

<table>
<thead>
<tr>
<th>Target</th>
<th>Knight shift ((%))</th>
<th>Diamagnetic shielding ((%))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zn</td>
<td>0.55 ± 0.10(^a)</td>
<td>-0.26(^c)</td>
</tr>
<tr>
<td>Cd</td>
<td>0.77 ± 0.10(^a)</td>
<td>-0.56(^c)</td>
</tr>
<tr>
<td>Pb</td>
<td>1.93 ± 0.01(^b)</td>
<td>-1.63(^c)</td>
</tr>
</tbody>
</table>

\(^a\) The Knight shift of impurity \(B\) in a material \(A\) is related with the Knight shift of \(B\) in \(B\) through the equation of
\[ K(B) = \frac{K(A)}{N_A/N_B} \]

\(^b\) Experimental value of a dilute Ti atom in a Pb metal.

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II. PHYSICAL PROCESSES IN HFS-EXPERIMENTS
II. 1

HFI OF $^{18}\text{O}(2^+)$ AS PROBE FOR ATOMIC CHARGE-EXCHANGE PROCESSES


Following inelastic scattering of $^{18}\text{O}$ ions on a $^4\text{He}$ gas target, the HFI of the first $2^+$ state in ions recoiling backward (in c.m.) were studied as function of gas pressure by the time-integral version of the PAC method.

In these conditions the projectile energy is lowered by the nuclear collision from $\sim 45$ to $\sim 15$ MeV, following which the mean ionization at equilibrium is reduced from $\sim 7.5$ to $\sim 5.5$ units of charge by ionic collisions with other gas atoms. The PAC technique selects, however, only the first few such collisions as the nuclear probe has a short lifetime ($\tau_\text{n} \approx 3.8$ ps). As the only appreciable perturbation is caused by the $7^+$ ion in its ground state $^1$, the data at any gas pressure reflect essentially the effective occupation probability for this state over the nuclear lifetime.

For a quantitative data analysis a microscopic description of the ionic processes is needed. With the Ansatz that the new charge equilibrium is established within two collisions by single-electron capture processes and neglecting all other collision types, the pressure dependence can be related to the electron-pickup cross-sections:

$\sigma(8^+ \rightarrow \gamma^\text{ground})$ and $\sigma(7^+ \rightarrow 6^+)$. All other parameters ($g$-factor $^2$, lifetime, HFI field $^3$ and equilibrium charge-state distributions $^4$) are available from external data. In fitting the results it was further assumed that these cross-sections are related as: $\sigma(8^+ \rightarrow \gamma^\text{ground})/\sigma(7^+ \rightarrow 6^+) = 2$, as the Pauli principle would indicate. The cross-section derived:

$\sigma(7^+ \rightarrow 6^+) = (7 \pm 2) \times 10^{-17}$ cm$^2$

agrees in order of magnitude with data from beam-gas experiments $^5$.

Coincident gamma-ray anisotropy as function of gas pressure. The curve is a least squares fit for the perturbation mechanism described in the text.

References

II. 2 A POSSIBLE WAY TO DETERMINE THE SIGN OF THE MAGNETIC MOMENT OF $^{18}_0(2^+)$

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In PAC measurements involving HFI with polarized single-electron systems of isolated ions in their ground configuration ($1s, 1^2S_{1/2}$), a net rotation of the angular correlation is expected, the direction of which depends on the sign of the nuclear magnetic moment. Such a polarization might be attained by letting the ions recoil into vacuum through a saturated ferromagnetic foil.

Calculations of the expected experimental effect have been performed for an $I^0.2 \rightarrow 0$ gamma transition and a realistic ionic charge distribution (see figure). It has been assumed that the moving ion picks up electrons indiscriminately from the ferromagnet, which in the case of iron implies that some 81% of the single-electron ions would be polarized. Velocity-matching arguments would, however, indicate a lower value. The calculations show that the $^{18}_0 2^+$ state is particularly well suited to such a measurement, as the mean integral precession angle is close to the optimal value of:

$$\omega T \approx \pi \text{rad.}$$

As charge equilibrium is established in a very thin layer of matter ($1-5 \mu g/cm^2$), maintenance of a clean ferromagnetic surface throughout the measurement is of crucial importance. This aspect of the problem appears to have been satisfactorily solved experimentally. Further measurements are in progress.

References
3. "The Magnetic Moment of $^{18}_0(2^+)$", contribution to this conference.
The conservation of the nuclear alignment is one of the central problems in perturbed angular distribution experiments on isomeric states populated and aligned via nuclear reactions. Up to now only very few experiments [1] have been performed on states with lifetimes in the ns or µs region in a gaseous environment. Recoil into gas measurements usually employ probes with lifetimes of less than 1 ns [2]; violent perturbations of the nuclear alignment due to changing electronic configurations have been found.

We have observed the spinrotation of the 9/2⁺ state (T₁/₂ = 1 µs) of ⁸⁵Rb excited and aligned by the ⁶⁴Kr(d,n) reaction over several microseconds. A gaseous Kr target (of 2.25 atm pressure) was bombarded by 6.2-MeV deuterons from the 7-MV Van de Graaff. The beam was pulsed at a repetition time of 4 µs with a pulse duration of approximately 10 ns. The modulation spectrum R(t) = (W(t,+45°)−W(t,−45°))/[W(t,+45°)+W(t,−45°)] of the 514-keV γ rays deexciting the isomeric state is shown in Fig. 1. A magnetic field of 4.8 kG was applied perpendicular to the beam-detector plane. From the modulation frequency the g-factor of the 9/2⁺ state of ⁸⁵Rb of g = +1.350 ± 0.014 is derived. This value is just compatible with the one determined by Heubes et al. [3] as g = +1.386 ± 0.012 using the (d, pn) reaction on a metallic Rb target when a Knight shift correction of 0.65 a.u. is applied to their result.

Following the nuclear reaction the Rb-atoms recoil through the Kr gas starting with a maximum recoil energy of about 0.6 MeV. They are slowed down by atomic collisions to thermal energies within 1 ns. During the slowing down process charge changing collisions and electron excitations occur and rapidly fluctuating hyperfine magnetic fields up to several MHz due to paramagnetic states of the Rb-ions may interact with the nuclear moment and lead to a reduction of the nuclear alignment. On the other hand taking the mean time between two atomic collisions at the correlation time τ_c we get approximately τ_c = 10⁻¹⁴ sec at the maximum recoil energy and approximately τ_c = 10⁻¹¹ sec at a recoil energy of 1 eV; both correlation times are short enough to randomize the expected hyperfine fields. Measured charge state

![Figure 1: Spinrotation spectrum of ⁸⁵Rb.](image-url)
distributions on similar alkali-noble gas-systems like Na-Ne and K-Ar [5] and theoretical estimates of stripping and capture cross sections [6] for energies comparable with the recoil energy in our case indicate that about 90% of the Rb ions are in the 1+ charge state at energies of several tens eV. Once the ions are slowed down to these energies charge transfer times have become large compared with the us lifetime of the isomeric 85Rb state yielding a static charge distribution with most of the ions in the diamagnetic 1+ state.

The short correlation time which provides randomization of hyperfine fields of several MG and the predominant population of the 1+ charge state after the slowing down process explain that we observe a nuclear alignment of several percent constant over the nuclear lifetime.

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1972, p. 77
II. 4 DEPENDENCE of NUCLEAR DEORIENTATION on NUCLEAR SPIN for RECOILS in VACUUM

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Time-integral measurements have been made of the vacuum deorientation of the $2^+$ and $4^+$ states in $^{150}$Sm and the $6^+$ and $8^+$ states in $^{156}$Gd Coulomb excited by $133$ MeV $^{35}$Cl ions. The $^{150}$Sm results were deduced from $\gamma$ ray angular distributions measured in coincidence with backscattered $^{35}$Cl ions. In the $^{156}$Gd measurements, a fixed counter array and a sandwich target were used to measure directly the differences between the $\gamma$ ray angular distributions from nuclei recoiling in vacuum and the unperturbed $\gamma$ ray angular distribution from nuclei stopped in a thick $^{156}$Gd target.

Table 1 shows the measured deorientation time constants in ps. The $8^+$ data were also analyzed assuming $\tau_\gamma / \tau_4 = 10/3$ as predicted by the Abragam-Pound theory for $J \geq 2$ essentially independently of the degree of quadrupole admixture.

It is seen from Table 1 that all of the $\tau_\gamma$ values measured in this work are indeed equal within experimental uncertainty. This is in contrast to the results of Nordhagen et al. who found the deorientation of high spin states in the light Er nuclei populated by the $(H_1, xn)$ reaction to be less than that expected on this model. They proposed that the reduced deorientation of the high spin states resulted from the fact that $\langle J \rangle$ of the recoiling ions (electron spin) was small compared to the nuclear spin $I$. Making appeal to Alder’s theoretical treatment of deorientation for the static case, Nordhagen et al. were able to formulate a model in which the deorientation decay constants $\tau_\gamma$ and $\tau_4$ are a function of the spin ratio $J/I$. Their Er data were accounted for with $J = 3$ which on their treatment give the $\tau_\gamma$ vs $I/J$ dependence shown by the upper solid curve in Figure 1. The present data require $J \geq 5$ as seen from the figure.

In the light of the present experiment, it seems likely that the Nordhagen results should be interpreted in terms of reduced g-factors in the back bending region of the three Er nuclei considered. In any event, the present results show that future measurements of g-factors for high spin states may still be possible using the recoil into vacuum technique.

Table 1

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Transition</th>
<th>$\bar{\tau}_\gamma$ (ps)</th>
<th>$\bar{\tau}_4$ (ps)</th>
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<tbody>
<tr>
<td>$^{150}$Sm</td>
<td>2-0</td>
<td>0.279 ± 0.027</td>
<td>0.15 ± 0.02</td>
<td>69 ± 2.5 *</td>
<td>27 ± 4</td>
</tr>
<tr>
<td>4-2</td>
<td>0.60 ± 0.06</td>
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<td>14 ± 1.3 *</td>
<td>21 ± 6</td>
<td>8 ± 4</td>
</tr>
<tr>
<td>$^{156}$Gd</td>
<td>6-4</td>
<td>0.517 ± 0.025</td>
<td>0.25 ± 0.06</td>
<td>22.8 ± 0.6 *</td>
<td>24 ± 3</td>
</tr>
<tr>
<td>8-6</td>
<td>0.80 ± 0.08</td>
<td>0.54 ± 0.20</td>
<td>6.7 ± 0.4 *</td>
<td>27 ± 18</td>
<td>10 ± 16</td>
</tr>
<tr>
<td>8-6 *</td>
<td>0.77 ± 0.05</td>
<td>0.51 ± 0.08</td>
<td>6.7</td>
<td>23 ± 10</td>
<td>10 ± 6</td>
</tr>
</tbody>
</table>

\* Analyzed with $\tau_4 = (3/10) \tau_2$
\* a) Ref. 3
\* b) Present value from Coulomb Ex. cross section.
\* c) Ref. 4
\* d) Ref. 5

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\* a) Ref. 3
\* b) Present value from Coulomb Ex. cross section.
\* c) Ref. 4
\* d) Ref. 5
Figure Caption

Plot of the measured deorientation times $\tau_2$ as a function of nuclear level spin $I$ for $^{150}\text{Sm}$ (open points) and $^{156}\text{Gd}$ (solid points). Assuming a constant g-factor then according to Ref. 1, $\tau_2$ should increase with $I$ for small values of $J$(atomic) as shown by the curves labelled $J=3$ and $J=5$. The present results, interpreted on this model, suggest that $J > 5$.

References

A time differential technique has been employed to observe the time dependence of an angular correlation perturbed by hyperfine interactions. The measurement was performed on the $2^+\rightarrow 0^+$, 122 keV transition in $^{152}$Sm. The excited nuclei recoiled into vacuum with velocities up to $v/c = 0.0023$.

Such a perturbation was originally observed with a microwave timing system, the resolution of which was relatively poor. However, in a later measurement performed with a plunger technique, improved time-resolution enabled the observation of oscillations in the perturbation which suggest the action of static hyperfine fields.

At present we report on refined measurements with a plunger technique. The $2^+$ level in $^{152}$Sm was coulomb excited by alpha beams, the $\gamma$-radiation being detected by two Ge(Li) detectors at 45° and close to 90°. If $G_K(t)$ is the attenuation coefficient in a distribution $W(\theta) = 1 + \Sigma A_K G_K(t) F_K(\cos\theta)$, the $\gamma$-intensity for a given recoil angle of the Sm nuclei, $\theta_R$, is

$$I(\theta_R, \theta) = \int_0^{\infty} e^{-t'/\tau} \left[ 1 + \Sigma A_K G_K(t) F_K(\cos\theta) \right] dt +$$

$$\int_{t'}^{\infty} e^{-t'/\tau} \left[ 1 + \Sigma A_K G_K(t) F_K(\cos\theta) \right] dt,$$

where $t'$ is the corresponding flight time to the stopper foil, and $\tau$ is the nuclear mean life.

The actual calculation of the $\gamma$-intensity, integrated over all $\theta_R$, involves knowledge of kinematics, differential cross sections for all recoil angles, the associated normalised statistical tensors, stopping power in the target and information about the residual ionic systems. The $G_K(t)$ can then be introduced as free parameters. At high recoiling angles, the Sm nuclei can be stopped in the target. This part adds a time-independent component to the gamma radiation but on the other hand narrows the kinematical spread involved. The calculation shows that for a static case where

$$G_K(t) = \Sigma_{F,F'} \left( \frac{(2F+1)(2F'+1)}{2J+1} \right) I F F K \{ I I J \} \begin{bmatrix} 2 & -1 \omega & FF \end{bmatrix} t,$$

the temporal behaviour of the oscillating component is not strongly dependent on the target thickness.
Fig. 1 exhibits the results of two measurements carried out with 3 and 4 MeV alpha beams. What is shown is the ratio of the measured intensities at 45° and 90° as function of the plunger distance. The oscillating pattern suggests static components in the hyperfine interaction. This is to some extent surprising in terms of the simple picture of "hot" versus "cold" ionization (4), since the atomic spectra for the possible Sm ions produced in the recoil are very dense throughout. Moreover, by normalising the 3 MeV data, assuming that only the J = 1/2 ground state of Sm$^{41}$ is causing the perturbation, we arrive at a calculated oscillation amplitude that is roughly in agreement with the measured one.

Under this assumption the magnetic field at the nucleus $H(0)$ from the two measurements is

- $H(0) = 2.9 \pm 0.3$ MG, 3 MeV data
- $H(0) = 3.1 \pm 0.4$ MG, 4 MeV data.

These results, however, can be considered as upper limits only: while being essentially independent of target thickness, a thin carbon or oxygen layer will modify the kinematics, affecting the yield of Sm$^{41}$ ions, yielding an apparent higher field than in reality (compare ref. 2). From the two measurements we therefore conclude that

$H(0) \leq 3.0 \pm 0.3$ MG.

The value of $H(0)$ for Sm$^{41}$ ($^{8}F_{1/2}$) was calculated on the basis of optical spectroscopic information on rare earths, as well as on coupling parameters fitted to the known energy spectrum of Sm$^{41}$ (5). The calculated result $H(0) = 2.6$ MG is in agreement with the above measured limit.

1) R. Armbruster et al., in Hyperfine Interactions in Excited Nuclei, Eds. Goldring and Kalish, Vol. 3 (1971) 973
2) Y. Dar et al.
   Z. Naturforschung 27a (1972) 562
3) K. Alder
4) G. Goldring
   Proceedings of the Munich Conference (1973)
5) J. Bauche
   Private Communication.
II. 6. TEMPERATURE DEPENDENCE OF THE SPIN ROTATION AMPLITUDE FOR $^{71}$As AND $^{76}$As IN GERMANIUM METAL

by H. Bertschat, O. Echt, B. Haas, F. Pleiter, E. Recknagel, E. Schlodder and B. Spellmeyer

Hahn-Meitner-Institut Berlin and Freie Universität Berlin, Fachbereich Physik

A number of isomeric states covering a wide range of lifetimes are known in As isotopes that can be populated from Ge by ($p,n$) and ($d,n$) reactions. Their magnetic moments have been measured by DPAD and SCPAD in recent years. These states offer a good possibility for the investigation of radiation damage. Such effects have been observed in a SCPAD measurement on $^{73}$As [1]. Since the system As in Ge is particularly interesting from the solid state physics point of view, we report here on some further results, obtained with the DPAD method, that are not fully understood yet.

We have measured spin rotation spectra of the 23.6 ms level in $^{71}$As($E = 999$ keV, $g = +1.35(10)^3_j^3$) and of the 2.6 ms level in $^{76}$As($E = 45$ keV, $g = +0.558(4)^3_j^3$) in different magnetic fields. The isomeric levels were populated through the reactions $^{70}$Ge($d,n$) and $^{76}$Ge($p,n$) by a pulsed 6.5 MeV deuteron ($t_{on} = 2$ ns, $t_{off} = 100$ ns) and proton ($t_{on} = 25$ ns, $t_{off} = 8$ ps) beam, respectively. The measurements were carried out at temperatures ranging from 80 K up to the melting point of germanium. Fig. 1 shows some typical spin rotation spectra for the case of $^{76}$As. They were fitted to a function of the type

\[ R(t) = \frac{a_2}{3} \int_{-\infty}^{\infty} \cos \left[ 2(\gamma t + \frac{1}{2}) \right] e^{-\lambda t} \]

Some of the measured amplitudes had to be corrected for finite time resolution.

The most striking result displayed in Fig. 1 is that $a_2$ rather than $\lambda$ shows a strong temperature dependence, a fact also found for $^{71}$As.

Obviously the observed phenomena cannot be explained on the basis of the Abragam and Pound model, as previously assumed [1]. Apparently the non-stationary nature of the processes immediately following the stopping of the recoil ion in the solid has to be included in any treatment of the new effects observed. Since in the $^{71}$As data no significant damping of the amplitude, attributable to time dependent interactions, is observed more than 20 ns after the nuclear reaction, these processes would have to last less than $10^{-8}$ s. Together with the pronounced effect on $a_2$ this complicates an interaction of $\omega_{eff} > 500$ MHz.

Such a high frequency would favour an explanation based on a magnetic hyperfine coupling. If one compares the effective anisotropy coefficients $a_2$ plotted for both levels in Fig. 2, however, one finds that the stronger change in $a_2$ is measured for the state with the smaller g-factor, in sharp contrast with what one expects on the basis of such an assumption.

It is hoped that experiments employing other isomers will help to clarify the situation. A measurement of the quadrupole moments of the states involved would be particularly important.

\[ \text{Fig. 1: Examples of the spin rotation spectra observed for } ^{76}\text{As in germanium at different temperatures and for different magnetic fields.} \]


![Graph](image)

**Fig. 2:** Spin rotation amplitudes $a_2$ as a function of temperature obtained for $^{71}$As and $^{75}$As in germanium. The $^{71}$As data are preliminary results.
II. 7 MÖSSBAUER AND CHANNELING EXPERIMENTS ON IMPLANTS OF $^{119}$Sn IN GROUP IV ELEMENTS

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*Institut für Atom- und Festkörperphysik, Freie Universität Berlin

Hyperfine investigations with impurity atoms implanted into solids are often hampered by radiation damage and lack of information regarding the final location of these atoms. However, these problems can be studied by combining Mössbauer and channeling experiments. Mössbauer measurements provide information on the bonding and electronic configuration of the impurity atom in the host lattice from the determination of the Debye-Waller-factor and the isomer shift. These parameters are almost entirely determined by interaction with the nearest atomic neighbours so that radiation damage can be studied on a microscopic scale (even in heavily damaged crystals). Channeling experiments locate the impurity atoms in the host lattice and reveal macroscopic radiation damage.

Radioactive $^{119m}$Sn was implanted into diamond, silicon and germanium single crystals by means of an isotope separator. (Tin is expected to be an ideal substitutional impurity in these crystals because it is isoelectronic to the host atoms.) The implantations were carried out with an energy of 60 keV at room temperature and $400^\circ$C and at doses between $10^{13}$ - $10^{17}$ atoms/cm$^2$. Mössbauer and channeling experiments were in all cases undertaken on the same samples. The results can be summarized as follows:

1) Low dose ($\leq 10^{14}$ atoms/cm$^2$). Implantations at $400^\circ$C in Si and Ge show negligible radiation damage effects. Tin ends up entirely ($\geq 90\%$) on substitutional lattice sites in an undisturbed surrounding.

2) High dose ($\approx 10^{15} - 10^{17}$ atoms/cm$^2$). Implantations at $400^\circ$C show that sufficient radiation damage is produced to wipe out the channeling effect at doses $\approx 10^{16}$ atoms/cm$^2$. The Mössbauer data indicate that the microscopic surroundings of the impurity atoms are essentially undisturbed. Therefore, the radiation damage is probably of "long range type", i.e. from lattice strain or distortion in the implantation volume. The distribution of tin atoms in the host lattice is the same as that of the distribution of the host atoms.

3) No channeling effect is observed for room temperature implantations and doses $\geq 10^{14}$ atoms/cm$^2$. The Mössbauer spectra for these cold implantations differ significantly from those for hot implantations and indicate that local microscopic distortions of the lattice become important. For high dose ($\approx 10^{15}$ atoms/cm$^2$) implantations line broadenings and reductions of the recoilless fraction of $\approx 20\%$ compared to hot implantations are observed.

For the substitutional tin atoms, the recoilless fraction may be calculated with the Debye-model by introducing an effective Debye-temperature for the impurity atoms, which is given by $\theta_{eff} = (\mu_h/\mu_{Sn})^{1/2} \theta_D$, where $\mu_h$ and $\mu_{Sn}$ are the masses of the host and Sn-atoms, respectively, and $\theta_D$ is the Debye-temperature of the host. Fig. 1 shows a comparison of the calculated (solid line) and experimentally observed values. (Debye temperature $\theta_D$ from low temperature specific heat measurements are adopted). The experimental points follow a straight line as required by the Debye-model. The deviation of the calculated and observed lines, however, seems to indicate a weakening of the bond between impurity and host atoms compared to those between host atoms. However, the Debye-model does not provide a good approximation for the phonon spectrum of these semiconductors. The result of a calculation from Lehmann and de Wames' on a Sn-impurity in germanium based on a phonon spectrum calculated from neutron scattering data by Phillips' is included in Fig. 1 as an asterisk and agrees remarkably well with the experimental value.
Certain qualitative comments can be made regarding the observed isomer shifts. The α-tin is known to exhibit covalent hybridized bonds of approximately $sp^3$ configuration which are typical for all group IV elements with diamond structure. Thus it can be assumed that the valence electron density outside the core region reproduces the electronic configuration of the replaced host atom. On the other hand, the electron density at the tin nucleus is not too dissimilar from that of α-tin since the interaction of the electrons with the nucleus in its close neighbourhood is predominant. The influence of the neighbouring atoms on the contact density can be considered as a small perturbation and the corresponding change can be calculated. It turns out that the change of contact density is negative and its numerical value is proportional to the difference of the atomic cores of the tin and host atoms. It happens despite the fact that the diamond-type lattice contracts considerably from α-tin to diamond so that the volume available for the tin atoms decreases (which might cause an increase in electron density at the nucleus) that the net electron densities decrease. One way of plotting this effect, characteristic of a substitutional implant in group IV elements, is shown in Fig. 2 in the linear relationship between the bond length of the host atoms and the observed isomer shifts.

1) G.W. Lehmann and R.E. Wames, Phys. Rev. 131, 1008 (1963)
II. 8 IN-BEAM OBSERVATION OF ELECTRIC QUADRUPOLE INTERACTION OF $^{111}$Cd RECOIL-IMPLANTED INTO CUBIC PALLADIUM

by H. Bertschat, H. Haas, F. Pleiter, E. Rickenagel, E. Schloeder and B. Spellmeyer
Hahn-Meitner-Institut Berlin and Freie Universität Berlin, Fachbereich Physik

Several in-beam DPAD experiments show that most of the recoiling ions come to rest at regular lattice sites. Some of the experiments, however, additionally indicate local perturbations of the extra-nuclear environment created by the recoiling ions during the stopping process [1,2]. These radiation damage effects have been attributed to static and/or dynamic electric quadrupole interactions though no detailed picture exists up to now.

Conventional methods for studying lattice defects, such as the observation of the change of the residual resistivity with temperature [3], give only global and time-averaged information. Earlier reported PAC measurements [4,5] are also of time-averaged character. The DPAD method itself offers the possibility for investigating the perturbation of the neighbourhood of the probe nucleus during the time of interest, i.e. the time interval after the nuclear reaction. A proper system to study a radiation induced hyperfine quadrupole interaction of the recoil ions is a nonmagnetic cubic host lattice. In-beam experiments on $^{111}$Cd recoil-implanted into the cubic (fcc) Pd-lattice were therefore carried out at temperatures ranging from 20 K up to 1200 K.

The isomeric $\frac{5}{2}^+$ state in $^{111}$Cd ($E_x = 123$ keV, $Q = 0.5$ b [6]) was populated by means of the reaction $^{108}$Pd($\alpha$,n) using the pulsed $\alpha$-beam of the Van-de-Graaff-accelerator at the Hahn-Meitner-Institut Berlin ($E = 13$ MeV; pulse repetition time = 1 s; pulse width = 8 ns). Typical radiation doses were $10^{16}-10^{17}$ ions/cm$^2$. The experimental setup was similar to the one described earlier [6].

Fig. 1 shows the time dependence of the perturbation amplitude $A(t)$ observed at three typical temperatures. In the temperature range from 20 K up to 320 K, apparently a small fraction $f$ of the excited nuclei experiences a unique electric field gradient (EFG) giving rise to a sharp quadrupole frequency $\omega_Q^0$, whereas most nuclei interact with EFGs broadly distributed. This distribution causes a strong damping of the perturbation factor $A(t)$. The solid lines represent least-square fits to the experimental points using an equation of the form

$$A_2(t) = A_2 \sum_{n} s_n \cos \omega_Q^0 t + (1-f)e^{-n5t}.$$  

For the anisotropy coefficient we measured the value $A_2 = (15 \pm 3)$ %.

The fraction of the nuclei that feel the unique EFG was found to be $f = (3.4 \pm 1.2)$ %. Below 410 K this value was constant within the limits of error. At $T = 410$ K, however, the sharp frequency has disappeared completely. Under the assumption of an axially symmetric EFG the temperature independent interaction constant is $e^2Qg/h = 20 \omega_Q^0/6\pi = 87 \pm 2$ MHz.

For the broad frequency distribution (around zero frequency) a Lorentzian shape was preferred, since a Gaussian gave worse fits in all cases. The width $\Delta(e^2Qg/h) = 20 \omega_Q^0/6\pi$ of the interaction constant distribution was found to depend on temperature as shown in fig. 2. An interpretation of the obtained results may be attempted in the light of the present view of defects in fcc-metals.

At the lowest temperatures employed in this experiment interstitials and vacancies do not show thermally activated motion. Radiation damage effects should, therefore, be frozen in and a static EFG distribution can be assumed. Using the approximation of point charges randomly distributed over lattice
sites, the perturbation factor was calculated by a Monte-Carlo technique including effects of EFG asymmetry. In this model one obtains a sharp frequency due to the nearest neighbours and a broad distribution around zero-frequency, justifying the fitting function used. The closest resemblance of the calculated results to the measured curves is obtained for a concentration of 0.5 \%. The measured unique quadrupole interaction constant is of the right order of magnitude for a point defect in nearest neighbour positions.

The sharp drop in the width of the frequency distribution (fig. 2) at about 150 K indicates that above this temperature one contribution to the nuclear perturbation disappears in a time range less than $10^{-8}$ sec. We identify it with the annealing stage I, generally attributed to free interstitial migration. This was observed at 35 K in a damage study on Pd \[[3]\], i.e. above this temperature the interstitials anneal in less than $10^{-3}$ sec, the typical annealing time for resistivity measurements. From these two data, one can estimate the activation energy for interstitial migration as $E = 0.1$ eV. Because Cd impurities distort the Pd lattice, interstitials can be trapped by Cd and thus will anneal at a higher temperature. As the point defects giving rise to the unique quadrupole frequency observed in our experiment disappear at about 400 K, it is tempting to identify them with trapped interstitials. The axially asymmetric EFG assumed in our fits is in agreement with such a picture. The fraction $f$ corresponding to the unique interaction frequency does not increase drastically above 150 K. Therefore, one can conclude that the trapping occurs immediately after the stopping of the recoil.

In this description the frequency distribution observed at temperatures above 400 K is due to defects with lower mobility. These should then also be stable at room temperature and lead to damping effects similar to those observed by Behar and Steffen \[[5]\] in a DPAC experiment on $^{111}$Cd in Ag. A number of possible explanations have been summarized by these authors.

A calculation of the influence of the EFG distribution on magnetic spin rotation spectra predicts damping effects that agree with experimental data.

A single crystal DPAC or DPAD study could give more information about the structure of the defects observed here.

\[\text{[2]}\ J. \text{Bleck, R. Butt, H. Haas, W. Ribbe and W. Zeitz, Phys. Rev. Lett. 29 (1972) 1371}
\[\text{[5]}\ H. \text{Behar and R. M. Steffen, Phys. Rev. 7 (1973) 788}
As part of a program to determine the sign and magnitude of the quadrupole interaction (QI) coupling constant $e_q Q$ of the 247 keV level of $^{111}$Cd after radioactive ion-implantation into a number of non-cubic metals, we have performed a series of experiments in Be metal. Normally one expects the QI frequency to be independent of the type of parent nucleus which populates the level. The present experiments show that $e_q Q$ for this level in Be differs by more than a factor of 3 depending on whether the implanted parent nuclide is $^{111}$Ag on the one hand or $^{111}$Cd and $^{111}$In on the other. Such a result has not been encountered before and remains, at the present time, a puzzle.

The 247 keV level of $^{111}$Cd can be populated by the $\beta^-$ decay of $^{111}$Ag($7d$), the isomeric $\gamma$-decay of $^{111}$Cd in $49 \text{ min}$., or by the electron-capture (E.C.) decay of $^{111}$In($2.8d$), permitting $\beta^-\gamma$ ($^{111}$Ag) or $\gamma-\gamma$ ($^{111}$Ag, $^{111}$Cd, $^{111}$In) perturbed angular correlation (PAC) experiments on the $^{111}$Cd level. The activities mentioned were produced by reactor irradiation of $^{113}$Cd or $^{3}He$ bombardment of $^{113}$Cd or $^{115}$Ag foil in the Princeton University cyclotron and implanted with an energy of 90 keV using the Princeton University Isotope-Separator facility into a single crystal or polycrystalline foil of Be metal. Experiments with $^{111}$Ag sources utilized the $^{111}$Ag($7d$), $^{111}$In($2.8d$) cascades for $\beta^-\gamma$ PAC measurements in the single crystal while the $^{113}$Cd($93$-$247$) keV cascade was employed for the $\gamma-\gamma$ PAC experiments with mono- or polycrystalline sources. The $^{150}$-$247$ and the $^{173}$-$247$ keV cascades were utilized for $\gamma-\gamma$ PAC measurements following $^{113}$Cd and $^{111}$In decays, respectively. All implants and PAC measurements were performed at room temperature and hence were used without any further treatment after the ion-implantation. To make a clear comparison of the results with implants of the three parents, the same single crystal was used in all three cases, the residual activity being etched away after each experiment. The QI constants obtained are listed in Table I, and typical PAC spectra are displayed in Figs. 1 and 2. The main features of the results can be summarized as follows:

a) The QI of the $2^-$ level in $^{111}$Cd in Be metal, measured after $^{111}$Ag implantation is more than a factor of 3 smaller than that observed after $^{111}$Cd or $^{111}$In implantation.

b) The PAC quadrupole precession spectra are very well-defined in each case, indicating that the great majority of atoms are at unique final sites, regardless of the implanted parent species.

c) The single crystal experiments show that the axis of symmetry of the QI is apparently along the c-axis of the crystal, regardless of the type of the parent species.

d) The fact that the same frequency is observed with $^{111}$Cd (isomeric $\gamma$-decay) or with $^{111}$In (E.C. decay) rules out the possibility that E.C. after effects play a role. The "anomalous" result, at present, appears to be that obtained with $^{111}$Ag.

e) The dependence of QI coupling constant on the nature of the parent species is observed so far only in Be. Measurements with $^{111}$Ag and $^{111}$In in Ti, Sn, and Cd show the same QI frequency with either parent. The anomaly appears to be linked to the impurity-host pair Ag-Be only.

A satisfactory understanding of the principal result, viz. observation a) above, is lacking at this time. We are pursuing two main lines of investigation to solve this puzzle. (a) A difference in the nature of the final sites of Ag in Be as contrasted to Cd and In; and (b) vacancy-associated field-effects preferentially produced at the Ag sites. Channeling experiments, to elucidate the lattice location and investigations on the annealing behavior of the QI observed with each parent species, are currently in progress.

*Supported in part by the N.S.F. Resident Visitor at Bell Labs.

1. E. N. Kaufmann, P. Raghavan, R. S. Raghavan, E. J. Ansaldo and R. A. Neumann, Contribution to this Conference.

2. P. Raghavan, R. S. Raghavan and E. N. Kaufmann, to be published.

<table>
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<tr>
<th>Parent</th>
<th>Decay Mode</th>
<th>Crystalline form of Be</th>
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<td>single crystal</td>
<td>$\gamma-\gamma$</td>
<td>$\pm17.1(7)$</td>
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<tr>
<td>$^{111}$Cd</td>
<td>$\gamma$</td>
<td>single crystal</td>
<td>$\gamma-\gamma$</td>
<td>$\pm2.5(15)$</td>
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<tr>
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<td>E.C.</td>
<td>single crystal</td>
<td>$\gamma-\gamma$</td>
<td>$\pm4.6(5)$</td>
</tr>
<tr>
<td>$^{111}$In</td>
<td>E.C.</td>
<td>Polycrystal</td>
<td>$\gamma-\gamma$</td>
<td>$\pm4.9(5)$</td>
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</table>
Fig. 1 Quadrupole precession curves for $^2_+^+$ level of $^{117}$Cd in a Be single crystal following implantation of $^{117}$Ag. Upper curve: $\beta-\gamma$ PAC. Lower curve: $\gamma-\gamma$ PAC with c-axis perpendicular to detector plane.

Fig. 2 Quadrupole precession curves for $^2_+^+$ level of $^{117}$Cd in a Be single crystal (c-axis perpendicular to detector plane) by $\gamma-\gamma$ PAC following $^{117}$In implantation (upper curve) and $^{117}$MgCd implantation (lower curve).
After a nuclear reaction the recoiling nucleus produces a number of point defects in the target lattice during its slowing down process. Recent investigations in AuGa$_2$ have shown that the migration energy of these defects is of the order of half an eV (ref. 1). Therefore the defects are expected to be frozen in the neighbourhood of the excited nuclei at lower temperatures ($T \lesssim 300$ K) causing an electric field gradient in a cubic lattice.

In the present experiment the quadrupole interaction of such field gradients with the isomeric nuclei $^{71}$Ge$^{(5/2^-)}$, $E_1 = 175$ keV, $T = 122$ ns and $^{69}$Ge$^{(9/2^+)}$, $E_1 = 398$ keV, $T = 4$ µs) was investigated. The isomers were populated by the $^{69}$Ga(p,n)$^{69,71}$Ge reaction using a target of cubic polycrystalline AuGa$_2$ and an 8 MeV proton beam.

The time differential perturbation of the angular correlation function for the $5/2^-$ state in $^{71}$Ge is shown in fig. 1 for three different target temperatures ($T_m = 765$ K for AuGa$_2$). The solid lines are least squares fits calculated under the assumption of an uniform axially symmetric electric field gradient, which for instance could be produced by a single defect in a defined distance to the excited nucleus. The averaged quadrupole frequency from several measurements at 110 K is $15 \pm 2$ MHz, and there seems to be a decrease in frequency with increasing temperature, as expected for quadrupole interaction (see e.g. ref. 2).

A similar measurement was carried out with the long lived $9/2^+$ state in $^{69}$Ge. The measured perturbation function at 110 K is plotted in fig. 2. The least squares fit for an axially symmetric field
gradient and an exponential attenuation of the amplitude yields a quadrupole frequency of 24 $\pm$ 3 MHz.

The ratio of the two observed interaction frequencies $\omega_o^{(71 \text{Ge})}/\omega_o^{(69 \text{Ge})}$ is in good agreement with the known ratio of the quadrupole moments (ref. 3). Taking for the quadrupole moments 0.28 b ($^{69}\text{Ge}$) and 0.06 b ($^{71}\text{Ge}$), an electric field gradient of about $10^{22}$ V/m$^2$ is obtained for AuGa$_2$. An electric field gradient of the same order of magnitude was measured in a recent experiment for Cd recoils implanted into a cubic Ag lattice (ref. 4) as well as for Cd in Pd metal (ref. 5).

The initial alignment observed in the case of the short lived $^{71}\text{Ge}$ agrees with the one obtained in a magnetic TDPAD measurement, whereas the amplitude in the case of $^{69}\text{Ge}$ is reduced by a factor of about 3. This indicates that only a fraction of approximately 10% of the nuclei experiences a sharp electric field gradient. The remaining part is exposed to a smeared out distribution of field gradients, which shows no effect during the short lifetime of $^{71}\text{Ge}$ but reduces the amplitude for the long lived case of $^{69}\text{Ge}$.

References:
1) G. Hempel et al. (this conference)
2) P. Heube et al. (this conference)
5) E. Recknagel, private communication

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II. 11. STOCHASTIC THEORY OF VACANCY INDUCED QUADRUPOLE INTERACTION RELAXATION EFFECTS
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Interaction of nuclei in isomeric states with the electric field gradient (efg) of the surroundings has received renewed interest in connection with in-beam methods or ion implantation techniques. In non-cubic hosts an additional quadrupole interaction is set up in many cases due to radiation damage of the lattice. In a cubic environment the pure defect-induced efg can be observed. Even if the nuclear quadrupole moment is known, proper interpretation of the experimental findings is hampered by uncertainties about the origin of the efg. Assuming that the final position of the probes is reasonably well known, which requires information from complementary experiments, like channeling, the complex defect structure of irradiated solids opens various possibilities for explaining the efg. In view of these uncertainties model calculations will help to distinguish between the variety of efg sources or perhaps to show that unique results cannot be expected.

We have chosen vacancy-induced quadrupole interaction as an example to illustrate its effects on the perturbation factors $G_{kk'}(t)$ describing extranuclear interactions of nuclei in excited states. Static interactions due to external magnetic fields or efg's in noncubic materials have been incorporated into the theory, but numerical results will only be shown for pure quadrupole interaction in cubic crystals. To cover the wide range from quasistatic to rapidly fluctuating quadrupole interaction, in the sense of Abragam and Pound, Blume's stochastic theory of hfi in its general form is most appropriate. Since in our model dynamic hfi results from diffusive motion of vacancies on the lattice, (quasistatic interactions being a natural limiting case) we have from the very beginning aimed at linking Blume's approach of dynamic hfi to the theory of (generalized) random-walk processes. This leads at once to an extension of known applications of Blume's theory, since vacancy jumps to nearest-neighbour positions change, in general, direction as well as strength of vacancy-induced efg. Also the number of stochastic states increases considerably if discrete-lattice calculations are performed. Usually, only three- or four-site jump models are treated numerically. Let us think of investigating dynamic quadrupole interactions by time-differential perturbed angular distribution (TDPAD). We then need to know the perturbation factors

\begin{equation}
G_{kk'}^{(i)}(t) = \langle k' | (\Omega^q_{k} \cdot \{ \Sigma \})_{kk'} | k \rangle
\end{equation}

which are most conveniently expressed as matrix elements of the stochastically averaged evolution operator $H^{q}_{f}(-i\overline{\Delta})$ with respect to irreducible tensor operators $\eta_{k}^{q} | k \rangle$, $\eta_{i}^{q} (-i\overline{\Delta})$ is a stochastic operator, because the quadrupole part of the total Hamiltonian $\hat{H} = \hat{H}^{q} + \hat{H}^{q}$ (or the Liouvillian $\hat{\Omega}$ associated to it)

\begin{equation}
\chi_{n}(i) = \sum_{j=1}^{2} \left( \begin{array}{c} \alpha \beta \\ i \end{array} \right) \beta_{j}(Q) \chi_{n}^{(j)}(j)
\end{equation}

depends on the stochastic variable $x^{(j)}(t)$ attributed to the location of a monovacancy $V_{j}$ at site $j$ and time $t$. Attributing a stochastic state $| j(t) \rangle$ to $x^{(j)}(t)$, the time evolution of which can for homogenous Markov processes be written as

\begin{equation}
| j(t) \rangle = e^{(W+t_{1})} | j(t_{0}) \rangle,
W = \nu (M_{i j} - 1);
\end{equation}

$\nu$ = vacancy jump frequency,
$(M_{i j})_{11} =$ transition probability matrix for single direct jump $i \rightarrow j$.
Blume's restricted average takes the following form

\begin{equation}
\langle \Omega^q_{k} \cdot \{ \Sigma \} \rangle_{kk'} = \sum_{j=1}^{2} \left[ \begin{array}{c} i \beta \\ j \end{array} \right] \exp \left\{ \left[ t_{0} \right] \left[ \{ t_{0} \} W + i \{ t_{0} \} \Sigma \right] \right\} \{ j \} p_{j}(t_{0}).
\end{equation}

$p_{j}(t_{0})$ (the probability of finding $V_{j}$ at $x_{j}$ initially), the jump frequency $\nu$, the transition probabilities $(M_{i j})_{11}$, and the efg associated with a vacancy at site $j$ are the basic parameters of the theory.

Some illustrative results of the behaviour of $G_{kk'}^{(i)}(t) = (1/5) \sum_{i=1}^{5} G_{kk'}^{(i)}(t)$ for nuclear spin $I=1$ and various jump frequencies are shown in the figure. The parameters $\nu \cdot t_{1}$ refer to $t_{1}$ = $2\pi/3\Omega_{q}(1)$, $\omega_{q}(1)$ being the quadrupole frequency due to a single vacancy in the 1st shell.
Higher shell quadrupole frequencies are calculated from a simple unscreened point-charge model. The curves labelled by squares and triangles correspond to a four-site jump model of a vacancy in the first shell and to a three-site jump model in the second shell (which explains the difference in the oscillation frequency). A priori probabilities $p_i = 1/4 (i = 1, 2, 3, 4)$ respectively $p_3 = 1/3 (i = 1, 2, 3)$ were used in the calculations. The transition probability matrix is merely determined by the number of nearest-neighbours accessible to the random walker.

Discrete lattice-calculations indicate that the contribution of the second shell is overestimated in the present calculations, at least if stationary distribution functions $p(t)$ would enter the expression. The first shell effects dominate for large vacancy-impurity binding energies. The heavy curves of Fig. 1 would, of course, also be influenced by re-reduction of second shell effects. As it stands they display the result of offering the random-walker shells plus an environmental shell to which vanishing quadrupole interaction was attached for simplicity.

With the lower set of curves we have reached the Abragam-Pound regime ($v t = 10$ corresponds to $\omega_0 t = 0.2$). The influence of higher shells to which the vacancy can jump is obvious. In going from the upper set of curves (quasistatic interaction) to the lowest ones (rapid diffusive motion of vacancies) an intermediate regime is crossed which requires more elaborate treatment along the lines shown in the present note. Again pronounced deviations from low-site models can be noticed.

5. C. Abromeit and H. Gabriel (this conference)
II. 12  HYPERFINE INTERACTION STUDIES IN SOLIDS WITH POINT DETECTS -
THE INFLUENCE OF THE INITIAL STATE

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Hyperfine interaction studies are inevitably linked to the complexities of radiation
damage whenever initial state preparation is accomplished by nuclear reactions, Coulomb
excitation or ion implantation into solid hosts. In contrast to conventional techniques
of sample preparation (e.g. diffusion, alloying) the nuclear probes experience interactions
with a nonequilibrium environment.

It is the purpose of the present note to elucidate the importance of properly defining
the initial state of the system at the onset of the measurement as well as the necessity
of averaging over the recovery process of the initially highly perturbed lattice if the
duration of the experiment is sufficiently long. To be specific, we have chosen the case of
vacancy-induced nuclear quadrupole interaction in metals. We feel that oversimplifying the
complex problem is more helpful than trying to solve too many questions at the same time.

Let us consider impurity implantation into a solid host under conditions which allow us
to neglect interaction of vacancies with one another and with vacancy-impurity complexes.
We assume that the majority of incident impurities comes to rest at a substitutional site
(interstitial positions could be incorporated if wanted), and that, when irradiation ceases
at time $t = 0$, a given number of vacancies is randomly distributed over the lattice. The
number of implanted impurities $N_I$ is supposed to be small compared to the number of mono-
vacancies ($V_I$) $N_V < N_M$, where $N_M$ is the total number of lattice sites. Recovery due to
free-vacancy annihilation (recombination with interstitials, migration to sinks, formation
of impurity-vacancy pairs) can be treated within the chemical reaction scheme or by dis-
crete-lattice calculations as was done for the last two processes by Koivu.

The intimate connection between discrete-lattice calculations of the annealing process
and Blume's stochastic theory of hfi is obvious. Recovery processes change the concentra-
tion of free vacancies and impurity-vacancy complexes and therefore also the electric
quadrupole interaction induced by vacancies. Moreover, dynamic hfi is caused by random walk
of vacancies on the lattice at sufficiently high temperatures. Here we are interested in
the time scale of the recovery process compared to the lifetime of the excited state $\tau$, the
beginning, and the duration of the run at times $t_0$ and $t_d$, respectively.

Consider an implanted impurity (I) on a substitutional site taken as the centre of shells
with a discrete number of lattice points for a particular lattice structure. The probability
$p_j(t)$ of finding a monovacancy ($V_j$) at any of the sites of the j-th shell at time $t$ is cal-
culated from:

$$ \frac{d}{dt} p_j(t) = \sum_{i} W_{ji} p_i(t=0). $$

The transition probability matrix $W$ for $V_j$ jumps between nearest-neighbour sites depends on
various factors, e.g. jump frequency $\nu$, $V_j$ migration energy $E_M$, $V_j$-I binding energy
(responsible for the complex formation), the lattice structure, particularly the number of
sites on each shell and the number of nearest-neighbours seen from an arbitrary site. Only
a finite number of shells can be considered in discrete-lattice calculations. The outer
region of the crystal is approximated by an "environmental" shell over which all the re-
maining lattice sites are homogeneously distributed. Within the range of validity of the
present approach ($N_I << N_V << N_M$) a variation of the vacancy concentration $x = N_V/N_M$
can be achieved by changing the number of shells (thereby changing the dimension of $W$) at the
sites of which one single vacancy can be placed. In a fcc lattice the number of lattice
sites up to the $6^{th}$ ($11^{th}$) shell are 86 (200), corresponding to $x = 1.0(0.5)$ %.
Numerical solutions of the random-walk equations are shown (Figs. 1-2) for a fcc lattice (10 and 11 shells) at temperature $T = 300K$ and with $E_{V} = 1.0\,\text{eV}$, $E_{g} = 0.05, 0.1\,\text{eV}$. A random initial distribution of vacancies over the $n_{i}$ sites of the $i$th shell was assumed $(p_{i}(t=0) = n_{i}/\sum n_{i})$. The curves in Fig. 1 (2) display how the probability of finding mono-vacancies on one of the first six (eleven) shells changes with time. The concentration of sinks was taken to be zero in all cases except in the lowest set of curves of Fig. 1.

Whenever vacancy-induced hfi is dominant, some features of the curves shown should be regarded in the interpretation of Mössbauer or TDPAD experiments. The initial distribution of vacancies is usually not known and is hard to determine experimentally with sufficient accuracy. If the observation time $(t_{r}-t_{0})$ covers the flat short-time regime, false weights will be given to the partial vacancy-induced field gradients, whenever the $p_{i}(t=0)$ are chosen unrealistically. The long-time behaviour, however, is insensitive to the initial distribution and sampling in this regime is more reliable. Figs. 1 and 2 show marked deviations of the stationary solution from a random initial distribution. As expected, the probability of finding a vacancy on the $i^{th}$ shell is increased for higher complex binding energies.

Notice the small probability for 2nd shell occupancy and the relatively high probabilities for vacancy location on shells with $j$ around 10 (Fig.2). This fact will become important if one sharp quadrupole frequency (1st-shell vacancy) as well as a distribution of field-gradients around a certain mean value is needed to explain the experimental data. The presence of sinks manifests itself in a slow decrease of the $p_{i}(t)$'s. This process can most easily be described within a chemical-reaction scheme into which discrete-lattice calculations merge in the regime after the rapid decrease of the free-vacancy concentration due to complex formation.

If the transient regime $10^{-2} < vt < 10^{2}$ lies within the interval $t_{0}$ and $t_{d}$, effective perturbation factors

$$c_{kk}'(\text{eff}) = \frac{(t_{d}-t_{0})^{-1}}{t_{d}} \int_{t_0}^{t_d} c_{kk}(t',t_0)dt'$$

will be observed. The absolute position on the time scale depends on the magnitude of $v$.

The latter quantity also determines whether vacancy-induced quadrupole interaction is quasi-static ($\omega_{QV}^{-1} \gg 1$) or affected by relaxation effects. The situation is most complicated if the transient regime is located such that $10^{-2} < vt < 10^{2}$, since then transient recovery and relaxation effects overlap.

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[1] A.C. Damask and G.J. Dienes "Point Defects in Metals" (Gordon and Breach, N.Y. 1963)
Spin-lattice relaxation processes in Li-metal have been studied several times with classical NMR techniques. In these experiments, however, the necessary application of a resonant rf field implies some difficulties: restriction of the accessible temperature and magnetic field ranges, skin effect, heating of small particles, etc.

In the case of Li the polarized neutron capture \( ^{6}\text{Li} \rightarrow 1.2\text{sec} \) nuclei have been produced in \( ^{7}\text{Li} \)-metal, enclosed in a stainless steel box, by polarized neutron activation. The \( ^{6}\text{Li} \) decay asymmetry was used to detect the nuclear polarization. \( T_1 \) was measured in two ways: (i) by direct observation of transients of the decay asymmetry \( a = a_0 \exp\left(-t/T_1\right) \) and (ii) by observation of the steady state asymmetry \( a = a_0\left(1 - T_1/T_d\right)^{-1} \) for continuous activation (an example is shown in Fig. 1).

In pure solid metals the essential contributions to the measured relaxation rate are due to the hyperfine interactions of the nuclei with the conduction \( \ell \)-electrons and the fluctuating nuclear dipole-dipole interaction caused by the diffusional motion of the atoms: \( 1/T_1 = 1/T_{10} + 1/T_{1d} \). According to the Korringa relation \( T_{10} \) is proportional to \( 1/T \) (apart from effects of thermal expansion) and contributes over the whole temperature range. On the contrary, \( T_{1d} \) is finite only if the Larmor frequency \( \omega_0 \) is comparable with the mean jump frequency \( 1/\tau \) of the diffusing atoms. So \( T_1d \) and by that \( T_1 \) pass through a minimum when \( \omega_0 > 1/\tau \).

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**Fig. 1.** \( \beta \) decay asymmetry \( 2a \) of \( ^{6}\text{Li} \) in \( ^{7}\text{Li} \)-metal as a function of temperature at \( H_0 = 3450 \text{ Oe} \).
The mean time between jumps of a nucleus is generally assumed to obey the Arrhenius relation \( t = \exp(E_A/kT) \), where \( E_A \) is the activation energy of the self-diffusion. Minima of \( T_1 \), i.e., of the \( B \) decay asymmetry, have been observed in the temperature range \( T = 300 \ldots 370 \) K, corresponding to \( H_0 = 100 \ldots 7000 \) Oe.

From the measurements of \( T_1 \) we obtained \( T_{1d} \) by separating the electronic contribution. In Fig. 2 minima of \( T_{1d} \) are shown for three different values of the magnetic field. The shape and precise \( \omega_0 \) value of these minima depend on the underlying diffusion mechanism. The analysis of these minima in terms of a monovacancy model \(^2\) is in progress and treats questions concerning the correlation of subsequent jumps, diffusion parameters, etc.

References

1) A. Seeger, H. Mehrer, in: Vacancies and Interstitials in Metals, p. 11, North-Holland, Amsterdam 1969; and References given there.

The temperature dependence of the nuclear spin relaxation time of isomeric nuclei produced and aligned by a nuclear reaction has been observed first by Bertschat et al.\(^1\) for isomeric \(^{73}\)As nuclei in solid Ge by the stroboscopic method. Here we want to report on similar studies concerning the influence of the temperature and of the magnetic field strength on the time dependent attenuation of the \(\gamma\)-ray anisotropy of the three isomeric nuclei \(^{69}\)Ge \((T = 4.2\) ps\), \(^{71}\)Ge \((T = 0.12\) ps\) and \(^{73}\)As \((T = 8.1\) ps\) in solid cubic AuGa\(_2\). These isomers were produced by the reaction \(^{69}\)Ga(p,n)\(^{69}\)Ge and \(^{71}\)Ga(p,n)\(^{71}\)Ge with the 10 MeV proton beam of the Erlangen tandem accelerator and by the reaction \(^{71}\)Ga(\(\alpha,2n\))\(^{73}\)As with the 30 MeV \(\alpha\)-beam of the Hamburg cyclotron. In the case of \(^{69}\)Ge and \(^{73}\)As the spin relaxation time was extracted from stroboscopic resonance measurements (SOPAD) carried out between room temperature and the melting point \((T_m = 492^\circ\)C\) of the AuGa\(_2\) target. The relaxation behaviour of the \(^{71}\)Ge nuclei was investigated with the spinrotation method (TIDPAD).

A possible explanation for the experimentally determined temperature dependence of the relaxation time is obtained by the assumption that the attenuation of the \(\gamma\)-ray anisotropy is caused by statistically fluctuating field gradients, which are produced by the diffusion of radiation-induced lattice defects. This situation can be described by the relaxation theory of Abram and Pound\(^2\) (AP), which gives a relationship between the nuclear spin relaxation time \(T_{rel}\) and the temperature \(T\):

\[
\ln(T/T_{rel}) = \frac{E_m}{kT} + \ln(const \cdot \omega_0^2)
\]

where \(E_m\) denotes the migration energy of the lattice defects and \(\omega_0\) the quadrupole interaction frequency. Equation (1) is valid only in the temperature region where the magnetic Larmor frequency \(\omega_L\) and the correlation time \(\tau_c\), given by

\[
\tau_c \sim c_{irr} \exp(\frac{E_m}{kT})
\]

\((c_{irr}\) concentration of the radiation-induced lattice defects\), obey the condition

\[
\left(\omega_L \tau_c\right)^2 \ll 1.
\]

The measured temperature dependence of \(T_{rel}\) is shown in fig. 1. With regard to equ.(1) a plot of \(\ln(T/T_{rel})\) against \(1/T\) was chosen. The solid lines in fig. 1B and 1C show the least squares fits of equ.(1) to the experimental data in the temperature range from 280\(^\circ\)C to 450\(^\circ\)C. From the slope of the lines one obtains a migration energy of \(E_m = (0.5\pm0.2)\) eV for the two magnetic fields 7.5 kOe and 17.7 kOe. This value is of the right order of magnitude. However, a lack of macroscopic data on AuGa\(_2\) does not allow to
decide on the type of the diffusing defects (vacancies, interstitials, divacancies, diinterstitials, etc.).

The much steeper slope in the SOPAD measurements just below the melting point cannot be explained by the theory of AP. Because application of this theory would yield a migration energy of 1.6 eV, and this value leads to a correlation time of the corresponding temperatures, which is too large to satisfy condition (3). However, time differential measurements carried out at two temperatures (fig. 1B) show that the initial alignment which is assumed to be constant in the SOPAD measurements, is decreasing with decreasing temperature. The perturbation responsible for this attenuation which has not been observed until now, is effective within less than 10 ns after the excitation of the isomers. If the temperature dependence of the initial anisotropy is taken into account, the experimental data can be explained by eq.(1) in the total temperature region with a migration energy of about half an eV. In this case the term \(\ln(\text{const} \cdot \omega_0^2)\) of eq.(1) gives a quadrupolar interaction frequency of \(\omega_0 = (27 \pm 10) \text{ MHz}\), if one assumes a defect concentration \(c_{\text{irr}}\) of 0.03 within a region of twenty nearest neighbours of the isomer. This value of \(\omega_0\) is in agreement with the static quadrupole frequency measured at lower temperatures. The stroboscopic data of \(^{73}\text{As}\) (fig. 1A) also show a steep slope just below the melting point which is equal to that observed in the \(^{69}\text{Ge}\) case. The strong relaxation in this case may be due to a larger quadrupolamoment of the isomeric \(^{73}\text{As}\) state.

A completely different behaviour is shown by the relaxation time of the \(^{71}\text{Ge}\) nuclei (fig. 1D). The temperature independent relaxation time of about 1 \(\mu\)s is in strong contrast to the prediction of the theory of AP indicated by the solid line. Since the ratio between the electric and the magnetic interaction energy is much smaller for \(^{71}\text{Ge}\) than for \(^{69}\text{Ge}\), this behaviour may be due to a decoupling of the fluctuating electric field gradients by the magnetic field of 21 kOe.

Fig. 2 shows the initial alignment taken from time differential measurements as a function of the applied magnetic field \(H_0\) for \(^{69}\text{Ge}\). The observed initial \(\gamma\)-ray anisotropy \(b_{2}^{\text{eff}}\) is remarkably smaller in solid AuGa\(_2\) than in the liquid phase. The values \(b_{2}^{\text{eff}}\) seem to be constant above 3 kOe whereas below 3 kOe a significant decrease is observed.

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2) A. Abragam and R. V. Pound, Phys. Rev. 92, (1953) 943
3) D. G. Doran, Radiation Effects 2, (1970) 249
4) P. Heubes et al., this conference
II. 15 Observation of relaxation in \( \text{Rb}_2\text{HfF}_6 \) and \( \text{Cs}_2\text{HfF}_6 \) due to vacancy motion

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We have measured the time dependence of the perturbed angular correlation of the 133-482 keV cascade in \( ^{181}\text{Ta} \) following the \( \beta \) decay of \( ^{181}\text{Hf} \) in \( \text{Cs}_2\text{HfF}_6 \) and \( \text{Rb}_2\text{HfF}_6 \) as a function of temperature between 20°C and 550°C. The behaviour of the two compounds is qualitatively similar, showing at low temperature a pure static quadrupole interaction, passing at intermediate temperatures to a mixture of a pure relaxation spectrum and a pure static quadrupole interaction, and at the highest temperatures measured to a pure relaxation spectrum.

The measurements on \( \text{Rb}_2\text{HfF}_6 \) are described in more detail in ref. 1. The production and measurements for the Cesium compounds were qualitatively similar. The perturbation factors for both compounds were analyzed as being a mixture of static and time dependent spectra in the form

\[
G_2(t) = \alpha G_2(\text{Static}) + (1-\alpha) G_2(\text{Relaxation})
\]

where \( G_2(\text{Static}) \) is the well known expression for a static quadrupole interaction in a crystalline powder, and depends on the interaction frequency \( \omega_0 \), the asymmetry parameter \( \eta \) and the frequency distribution factor \( \delta \), and \( G_2(\text{Relaxation}) = e^{-\omega^2 t} \).

We have interpreted these results in terms of a model in which the time dependent interaction is supposed to be due to the hopping of a trapped vacancy around the \( ^{181}\text{Ta} \) impurity ion. We imagine that during the 17.8 µs mean life of the 615 keV state which is populated by the \( \beta \) decay of \( ^{181}\text{Hf} \) a certain fraction of the 5+ Ta impurities which replace the 4+ Hf ions in the lattice succeed in trapping a Rb or Cs vacancy to neutralize their charge difference. Due to the presumable high mobility of the Rb or Cs vacancies in the rather open lattice structures of these compounds these vacancies may be attracted from a considerable distance during the long life time of the 615 keV state, and the probability of trapping thus depends both on the mobility and the vacancy density at a given temperature, making it difficult to derive the excitation energy for vacancy production from the temperature dependence of \( \alpha \).

We have attempted to apply Blume's model\(^2\) for a time dependent quadrupole interaction with fixed value and a finite number of possible directions. An analysis was carried out for two and for three different orientations using numerical techniques to invert the Laplace transform of the relaxation operator. The results agree with those in ref. 3 obtained by a different method. We have used these results as a qualitative guide to the interpretation of the observed spectra. In particular it is possible to predict from these formula that the relaxation constant \( l_2 \) is proportional to the inverse of the frequency \( W \) that characterizes the hopping of the vacancy from one lattice site to another.

Using the fact that \( W = \exp(-E_{\text{exc}}/kT) \) where \( E_{\text{exc}} \) is the excitation energy for the vacancy to jump from one lattice site to the neighbouring site, we may deduce values of the excitation energy for this process directly from the temperature dependence of the relaxation constant \( l_2 \). The results obtained for \( \text{Rb}_2\text{HfF}_6 \) were \( E_{\text{exc}} = -27±0.7 \text{ eV} \), and for \( \text{Cs}_2\text{HfF}_6 \).
$E_{exc} = .48 \pm .28 \text{ eV}$. In the latter case more points at higher temperatures would significantly reduce the error.

We are presently working on extending our calculations with Blume's theory to obtain results for a more realistic model, as well as attempting to formulate analytically the expected temperature dependence of the factor $a$. We feel however that these results already demonstrate the power of perturbed angular correlations in studying diffusion processes in insulators.

Table I - Measured Parameters

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<th></th>
<th>$\omega_0$ (MHz)</th>
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<th>$\lambda_2$ (ns$^{-1}$)</th>
<th>$a$</th>
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<td>.01</td>
<td>.07</td>
<td>.45</td>
<td></td>
<td>49.1</td>
<td>.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
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<td>-</td>
<td>-</td>
<td>.04</td>
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</table>

Typical Errors: $\pm 2$, $\pm .05$, $\pm .02$, $\pm .05$, $\pm .05$

* These results are qualitative only.

REFERENCES

Nuclear relaxation measurements are one of the few possibilities to investigate liquid metals microscopically. Of special interest are quadrupolar relaxation rates $\tau^{-1}_Q$, because these rates are expected to be sensitive to the ionic and electronic structure of liquid metals and its dependence on time, so that complementary knowledge to the work done by x-ray and neutron scattering may be derived from such experiments.

Hitherto, all $\tau^{-1}_Q$ data in monoatomic liquid metals which were measured accurately and in wide temperature ranges, e.g. in Bi [1], Ga [2], In, Hg [3], can be attributed to the diffusion based mechanism leading to a temperature dependence $\tau^{-1}_Q \propto \exp (Q_A / k_B T)$, where the temperature dependence of the macroscopic diffusion is parametrized by an activation energy $Q_A$.

In this paper we report quadrupole relaxation phenomena of the $13/2^-, 47 \mu s$ state of $^{207}$Po in lead below and above the melting point up to 1200K. The $\tau^{-1}_Q$ data in liquid Pb cannot be explained by the diffusion based mechanism alone. We assume that fluctuating field gradients due to vibrational modes in liquid lead may contribute to the observed relaxation rates.

The $\tau^{-1}_Q$ data in solid lead can be qualitatively assigned to time dependent quadrupole interaction due to diffusion of lattice defects which are produced by the nuclear reaction.
Relaxation times were measured as a function of the target temperature by the pulsed beam time differential perturbed angular correlation method in an external magnetic field. The isomer was produced by the $^{206}$Pb($\alpha,3n$) reaction with 38 MeV $\alpha$-particles from the Karlsruhe cyclotron using pulses of 3 us width every 200 us. Relaxation rates $\tau_1^{-1}$ were extracted from the 816 keV $\gamma$-ray modulation spectra measured with a two detector set-up. From experiments we know that the $\gamma$-ray angular distribution is characterized by $k_{\text{max}} = 2$. The results are presented in Fig. 1.

As a first step of a qualitative discussion of $\tau_1^{-1}$ we estimate the magnetic relaxation $\tau_M^{-1}$. Because Knightshift- or magnetic relaxation data for Po impurities in Pb are unknown we used as an approximation the corresponding data for $^{207}$Pt recalculated by the Korringa relation for the g-factor $g = -0.140$ of the $13/2^+$ state in $^{207}$Po. The result is described by curve (a) in Fig. 1. It follows with high probability that the measured rates over the whole temperature range are of nearly pure quadrupolar origin.

**Discussion of $\tau_0^{-1}$ in solid lead:** The strong increase of $\tau_0^{-1}$ in solid cubic Pb with decreasing temperature may be interpreted to be due to dynamic quadrupole interaction due to radiation induced lattice damages, as was suggested for quadrupole relaxation effects of $^{73}$As impurities in cubic Ge [6]. With high probability the $^{207}$Po recoil comes to rest at a normal lattice site but the vicinity of the stopped recoil may be disturbed by lattice defects. Due to diffusion processes these lattice defects create fluctuating field gradients at the site of the probe nucleus thus giving rise to relaxation phenomena. From the temperature dependence of $\tau_0^{-1}$ the diffusion energy of the lattice defects may be derived.

**Discussion of $\tau_0^{-1}$ in liquid lead:** The measured temperature dependence of $\tau_0^{-1}$ differs appreciably from self diffusion data in Pb [7] which are marked by curve (b) in Fig. 1 (right hand scale). Especially at high temperatures a large amount of the rate seems not to be diffusion controlled. This discrepancy can be removed by introducing a second quadrupole relaxation mechanism in liquid Pb. Warren and Wernick [8] suggested the possibility that part of the quadrupole relaxation in liquid AuGa$_2$ and AuIn$_2$ alloys may be caused by vibrational ionic modes. They assumed for the temperature dependence $\tau_0^{-1} \propto T^2$, by extrapolating the behaviour known in solids. With both the diffusion based mechanism and the vibrational modes based mechanism it should be possible to explain the observed temperature dependence of $\tau_0^{-1}$. Such an interpretation is supported by inelastic neutron scattering experiments in liquid lead [9] which yield evidence for co-operative ionic modes over wide temperature ranges.

3. N. Bräuer et al., Contribution to this Conference

This work was supported by the Bundesministerium für Forschung und Technologie.
II. 17 PERTURBED ALPHA-GAMMA ANGULAR CORRELATION STUDIES OF RADON RECOILS INTO ALUMINIUM

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The technique of using the recoil momentum from alpha decay to implant nuclei into different host-materials has become an important tool for the study of nuclear moments and hyperfine fields among the translead elements. Because of the very short range of alpha-recoils, the hyperfine interaction of nuclei stopped already in the source or in the oxide layer of the backing material complicates, however, the interpretation of the results of experiments with evaporated sources.

Activities $^{224}$Ra and $^{226}$Ra (about 0.05 μCi for $^{226}$Ra) were vacuum evaporated onto the different backing foils of plates of pure 99.9 % aluminium which had previously been anodically oxidised according to the method described by Davies et al. /1/. The radium thickness of the thin sources were in the order of 0.5-1 μg/cm$^2$ deduced from Rutherford scattering measurements with a 25 MeV $^{16}$O beam from the tandem accelerator.

The experimental apparatus was the IMPACT set-up at the tandem accelerator in Uppsala /2/. The measurements for each run were performed from -115° to +115° (0° is opposite the α-detector) in steps of 5 or 10 degrees. The data were fitted by a least squares procedure to the angular correlation equation:

$$\mathcal{W}(\theta) = 1 + A_2^{\text{exp}} \cos 2\theta + A_4^{\text{exp}} \cos 4\theta$$

where $A_k^{\text{exp}} = Q_k G_k A_k (k = 2, 4)$

The factors $Q_2$ and $Q_4$ are solid angle corrections. The unperturbed angular correlation factors ($A_2^P$ and $A_4^P$) are equal to 0.714 and -1.714, respectively. The attenuation factors $G_2$ and $G_4$ take into account the interaction between the nucleus and its environment.

The values obtained for $G_2$ and $G_4$ are presented in fig. 1 where also the loci of the different types of interactions /3,4/ are shown. We have studied how the axial asymmetry of the field gradient influences the quadrupole interactions by calculations for different asymmetry parameters $\eta$ and one typical result ($\eta = 0.4$) is indicated in the figure. The time dependent electrical interaction does not change with $\eta$, however. From the results of the measurements it can be clearly seen that the hyperfine interaction for the various oxide layers is of electric quadrupole type.

If one studies the results in fig. 1 more closely there is indication of an "oxide thickness effect" between 40 Å and 1000 Å for both $^{226}$Ra decay (d and e) and $^{224}$Ra decay (d' and e'). The values of the perturbation coefficients are consistent with the two different lifetimes of the 2+ level. This fact is valid for both 40 Å and 1000 Å oxide layer. An estimate of the field gradient from the static electric interaction can be done for eg. 1000 Å thick oxide layers: $^{222}$Rn(e) gives an average field gradient of $6.8(1.0) \cdot 10^{18}$ V/cm$^2$ and $^{220}$Rn(e') $7.2(1.0) \cdot 10^{18}$ V/cm$^2$ using the lifetimes and quadrupole moments from Bell et al. /5/. The values agree within the errors and the order of magnitude is what would be expected for Rn at a site of low symmetry in a solid /6,7/.

The range of Rn-ions after α-decay in Al$_2$O$_3$ can be estimated to about 160 Å /8,9/. 300 Å oxide layers [6'] is thus sufficient to stop almost all Rn-recoils. Backings of naturally oxidated aluminium have also been investigated (a, a', b and c). According to Samov's results for Al-films /10/ the oxide thickness is in the order of 20 Å. No accurate information about these thin oxide layers can be obtained from present measurement, because of the small percentage which stop in the oxide environment (2.4 % for 20 Å oxide layer and zero source thickness).

Estimates of α-recoil range /8,9/ and preliminary Rutherford scattering studies with the oxygen beam did not give enough thick sources to explain the present deviation from the measurements on isotope implanted $^{224}$Ra sources, where no perturbation was found.
To avoid source thickness and backing surface oxide problems the activity was diffused into the Al-metal and the result is presented in fig. 1 (c).

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Fig. 1. The experimental results for the following Al backings are presented in the $G_2 - G_4$ Plot.

- a) Al-foil, natural oxide
- b) evaporated Al, natural oxide
- c) Al-foil, diffused source
- d) 40 Å Al$_2$O$_3$ + Al
- e) 1000 Å Al$_2$O$_3$
- f') 300 Å Al$_2$O$_3$ + Al
- g') thick source +1000 Å Al$_2$O$_3$
II. 18 PERTURBATIONS OF THE $\gamma$ RAY ANGULAR DISTRIBUTIONS OF $^{222}$Rn FOLLOWING $\alpha$-IMPLANTATION INTO n-TYPE, p-TYPE AND INTRINSIC Si.

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The immediate effects which implantation may have on the hyperfine interactions of impurity nuclei implanted into various materials are not yet well understood. There are several different physical processes associated with the implantation that may give rise to non-vanishing hyperfine fields acting on the implanted nuclei for some time after the implantation. The radiation damage caused by the recoiling ions may induce electric fluid gradients at the nuclei, which may live for times of the same order, or longer than, the lifetime of the nuclear state under study. 1) The recoiling ions may have a net electric charge, either because of the recoil process or because of a preceding charged particle nuclear decay ("after effects"), which may induce fields on the nucleus which interact with the nuclear moments.

One could expect that the above mentioned phenomena would depend on the implantation conditions and on the impurity-host combination. Different backing materials, in which the recoiling ions come to rest, and different recoiling ions could give rise to hyperfine interactions differing in nature (electric quadrupole or magnetic dipole), in strength and in characteristic relaxation times. Indeed, very weak perturbations of the decay $\gamma$ ray angular distributions of excited nuclei implanted (following nuclear reactions or $\alpha$ decay) into metals were observed, whereas rather large perturbations, mostly but not all of electric quadrupole nature, were measured for excited nuclei implanted into non-conducting materials. 2,3

In the present work use is made of the ($\approx 100$ KeV) recoil, following the $\alpha$ decay of $^{226}$Ra, to implant excited $^{222}$Rn nuclei into various semiconducting backings. The anisotropy of the 186 KeV ($2^+ \to 0^+$ $T_{1/2} = 0.32$ nsec) gamma rays, in coincidence with $\alpha$ particles preceding the $\gamma$ decay, were measured for implantation into Cu, and into intrinsic ($10^{14}$ cm$^{-3}$) n-type ($\approx 10^{10}$ cm$^{-3}$) and p-type ($\approx 10^{19}$ cm$^{-3}$) Si. Full angular distributions were measured in some cases in order to extract the exact coefficients of the angular distributions. Preliminary results are shown in figures 1 and 2. As can be seen from the figures only slight differences, if any, in anisotropy can be observed among the various samples; nuclei recoiling into Cu are somewhat less perturbed than those recoiling into intrinsic p and n type Si. The perturbations seem mostly to be of electric quadrupole type, with typical field gradients of the order of $10^{19}$ Volt/cm$^2$.

Since all the sources used were approximately of equal thickness ($\approx 4 \mu g/cm^2$ RaCl$_2$) and care was taken to avoid oxidation of the samples, it is reasonable to assume that most of the recoiling Rn nuclei came to rest well inside the backing material. The measured results should therefore be related to the properties of the various backings. The crystalline structure of Cu is cubic and the backing used was polycrystalline, whereas single Si crystals (diamond structure) were used for the semiconductor implantations. The electrical properties of the backings used are quite different. The mobility and concentration of carriers vary drastically between Cu, an excellent conductor, and the semiconducting Si with carrier concentrations of $10^{19}$ cm$^{-3}$ for the p and n types, and $10^{14}$ cm$^{-3}$ for the intrinsic silicon. It was pointed out in various papers dealing with perturbations on the $\alpha$-$\gamma$ angular correlation following implantation into conductors, insulators and vacuum, 21 that it is hard to estimate the hf interactions for different implantation conditions. Perturbations of quadrupole nature measured in insulators were suggested as being caused by damage tracks left behind the recoiling ion or by amorphous regions in the vicinity of the stopped atom, whereas magnetic perturbations in insulators and in vacuum seem to have to do with the recovery times of electronic shells. The average "survival time" $\langle \tau \rangle$ for a charged-impurity in a substance with dielectric constant $K$ having $n$ carriers per cm$^3$ with mobility $\mu$ is given by $\langle \tau \rangle = \frac{K}{\mu n}$ (ref. 4). Using this expression one can calculate $\langle \tau \rangle = 10^{-14}$ sec for the p and n type samples, and $\langle \tau \rangle = 10^{-9}$ sec for the intrinsic Si. The fact that no significant difference between the three Si backed sources could be observed over the nuclear lifetime of $T_{1/2} = 0.32 \times 10^{-9}$ sec leads to the conclusion that either there are no hf perturbations associated with some disrupted charge distributions around the recoiled nuclei, or that the associated recovery times are shorter than
- 0.3 $10^{-9}$ sec. Measurements of the temperature dependence of $\bar{\rho}(\theta)$, which are under way, should clarify this point because of the strong dependence of $\bar{\rho}$ on the sample temperature.

* Shmuel Kochan fell in action in the Yom Kippur War (October 1973).

**References**

**Fig. 1:** Integral attenuation coefficients for implantation of $^{222}$Rn into Cu and Si samples.

**Fig. 2:** Anisotropies in Y ray distributions following implantation into Cu and various Si samples.
In $^{103}$Rh there are excited states at 360 keV with $J^\pi = 5/2^+$ and a lifetime $\tau_{360} = 85.0$ psec and at 296 keV with $J^\pi = 3/2^+$ and a lifetime $\tau_{296} = 11.1$ psec [1]. These levels were populated by Coulomb excitation with 30 MeV $^{16}$O ions. The experimental setup was similar to that generally used in IMPAC-technique [2]. The backscattered $^{16}$O ions were detected with an annular detector at 180° in coincidence with the 360-keV and 296-keV photons respectively. To vary the velocity of the recoiled $^{103}$Rh ions sandwich targets consisting of layers of Rh/Cu/Fe were used. The iron layer (25 μm) was magnetized by an external field perpendicular to the reaction plane. The perturbed angular correlations were measured for opposite directions $H_\perp$ and $H_\parallel$. As it is shown in ref. 3 one gets for the precession angle:

$$\omega \tau = -g \frac{\mu_N}{\hbar} (H_\parallel \tau + T(v))$$

(1)

where $g$ is the g-factor and $\tau$ the lifetime of the level, $\mu_N$ being the nuclear magneton, $H_\parallel$ being the internal static magnetic field and $T(v)$ is the time integrated transient field for the entrance velocity $v$. The value for the internal field $H_\parallel$ has been taken from ref. 4 to $H_\parallel = 543 \pm 11$ kG. If one measures the precession angle for different levels and for different entrance velocities one gets from eq. (1) a system of equations where the unknown quantities are the g-factors and the transient fields. From the analysis of our data we get the g-factors to $g_{360} = 0.52 \pm 0.08$ and $g_{296} = 1.55 \pm 0.35$. The results for the transient fields are shown as a function of the entrance velocity $v$ in fig. 1. The solid line is the theoretical description following Lindhard and Winther [5]. The parameters $v_p$ and $C$ are taken to $v_p = v_0$, the Bohr velocity, and to $C = 1.25$ in agreement with ref. 5. Cameron et al. [6] bombarded targets of Rh-Fe-alloys. Since in ref. 1 there are no values for the average recoil velocity, with the exception of 7 MeV α-particles, these measurements are not included in fig. 1. Our result for the highest entrance velocity is compared with other experimental values on different nuclei in fig. 2. The precession angle $\psi$ due to the transient field divided by the g-factor is shown as a function of the atomic number $Z$. The solid line is the prediction following ref. 5 with the same parameters as in fig. 1. As one can see there is fairly good agreement between our results and other experimental values and with the theoretical calculations too.

**Fig. 1:** Time integrated transient field as a function of the entrance velocity.

**Fig. 2:** The precession angle $\psi$ due to the transient fields in iron divided by the g-factor as a function of the atomic number $Z$. 
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4.) D.A. Shirley, S.S. Rosenblum and L. Matthias

5.) J. Lindhard and A. Winther
II. 20  Problematies of Temperature Spikes Following Coulomb Excitation
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Central Research Institute for Physics, Budapest, Hungary

For the successful extraction of the nuclear magnetic moment from a perturbed angular correlation or distribution measurement utilizing hyperfine fields, it is necessary to know the correct value of the hyperfine field acting on the excited nucleus of interest during its lifetime. When the investigated nuclear state is populated by nuclear reaction or Coulomb excitation, the nucleus recoils and generally leaves its equilibrium position in contrast with a measurement performed with radiative nuclei where no significant momentum transfer is involved. Because the excited nucleus moves in the first part of its lifetime the assumption of a constant hyperfine field acting on it is not valid. Two main phenomena can be expected. During the slowing down process the so-called transient field can occur, on the other hand the stopping atom can cause a thermal spike so probably some time is required for rebuilding the static hyperfine field at the end of the track of the recoiled atom.

The expected duration of both effects is in the order of picoseconds so very short living nuclear states are suitable for checking them. For this purpose we have measured the integral rotation angle of the angular distribution of the gamma rays coming from the first 2+ state of the $^{56}$Fe isotope excited by Coulomb excitation with protons of 2.5 MeV energy. The lifetime of this state is about 9 psec. In this case of low bombarding energy, the transient field does not exist so that the comparison of the obtained rotation with the rotation measured among static circumstances can show experimental evidence for the thermal spike effect.

The experimental arrangement was published elsewhere. Because of the expected small rotation a careful beam bending and shift measurement was performed by bombarding a pure rhodium target. The experimental results, $\epsilon = -2.8/3$ mrad, $\phi = 1.8/8 \times 10^{-3}$ cm, are in agreement with the numerical calculations. The angular distribution coefficients of the 847 keV $\gamma$-rays were measured to be $b_2=0.221/5$ and $b_4=-0.066/6$, which indicate some interference of nuclear reactions. The integral rotation angle was found to be $\omega = 0.0070/10$. The weighted average of the measured static rotations is $\omega = 0.0005/10$. The relatively high experimental errors do not permit us to draw strict conclusions, even though they force us to improve the precision of both values, especially the static one.

II. 21  RADIATION DAMAGE EFFECTS IN IMPLANTATION PERTURBED ANGULAR CORRELATION EXPERIMENTS

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and D. E. Murnick
Bell Laboratories, Murray Hill, New Jersey, USA

Integral in-beam implantation PAC experiments are unique in that the hyperfine interaction is measured within nanoseconds or less of the implantation. In addition, the interaction region is generally within the stream of a primary particle beam. Nevertheless, for cases where the implant is heavier than the host there are numerous data indicating a unique (or almost unique) effective hyperfine field consistent with that measured using conventional static techniques.1 There are also many cases of disagreement, including disagreements between ostensibly similar experiments.2 Models to explain the disagreements have included insufficient polarization and "conical" magnetic field effects.3

In the course of a study of the measurement of the g factor of the 1.4 ps state of 56Fe we have discovered an effect dependent on the primary particle beam. A brief summary of many careful experiments follows: 1) For iron recoil implanted into iron with a 36 MeV 10O beam or a 29 MeV 12C beam the effective hyperfine field at the implanted nucleus is reduced from its static value. 2) 9 MeV alphas yield an unperturbed field. 3) The oxygen effect is independent of whether the primary beam stops in, or escapes from the target. 4) The effective field is partially restored if single crystals are used or if the target is heated. 5) Subsequent Mössbauer effect measurements showed no difference between irradiated, or unirradiated samples. In addition, a careful reanalysis of IMPAC data of Heestand et al.4 using known g factors and transient field effects yields new values for the effective magnetic fields on various impurities in iron. In the cases listed below (Table I) it is seen that the IMPAC average hyperfine fields are reduced from static measurements.

Observation 3) rules out oxygen contamination effects via back diffusion. Using a value of 110 eV to produce a Frenkel pair, the defects produced by iron recoils themselves is calculated to be too small to appreciably perturb the local environment. For the various primary ion beams we have calculated a parameter

\[ D = \frac{\text{Number of Collisions} \times \text{Number of defects}}{\text{Collision}} \]

in the region of the stopped iron atoms. D = 35 for our oxygen experiments, 20 for the carbon case and 1.5 for alphas. This implies, for our experimental conditions with oxygen, a 10% defect concentration in the region of interest after 4 hours of running and that only 4% of the implanted atoms experience a normal environment within the first two nearest neighbor shells. The damage annealing rate can be estimated from the alpha experiments which yielded the normal hyperfine field and is about 2x10^{11}/sec. This rate is also consistent with the Mössbauer results, i.e., the beam induced damage had self annealed post irradiation.

We conclude that on line recoil implantation experiments are sensitive to the primary beam mass, intensity and energy. Our evidence suggests that radiation damage in iron due to the ion beam may be controlled by limiting the defect production rate in the region of interest to less than 3x10^{11} defects/sec.


**TABLE I**

<table>
<thead>
<tr>
<th>Solute</th>
<th>IMPAC</th>
<th>Other Methods</th>
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<tr>
<td>Pd</td>
<td>-409(36)</td>
<td>-573(20)</td>
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<tr>
<td>Ru</td>
<td>-359(44)</td>
<td>-490(10)</td>
</tr>
<tr>
<td>Cd</td>
<td>-238(131)</td>
<td>-348(10)</td>
</tr>
<tr>
<td>Te</td>
<td>+342(163)</td>
<td>+606(20)</td>
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Deduced in-beam (10O) hyperfine fields on impurities in iron and static measurements.

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Hyperfine Interaction of Cd-111 in Nickel after Recoil Implantation
by A. Andreeff* H.-J. Hunger, S. Unterricker
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The polished surface of a nickel foil of 0.13 mm thickness was evaporated with silver (w=1500 A) and then irradiated by 27 MeV a-particles. The In-11 nuclei produced by a (a, 2n)-reaction were implanted with a recoiled energy of about 14 MeV in the nickel backing. The magnetic hyperfine interaction was measured after the irradiation by TDPAC using the "reversal-field" method. Fig. 1 shows the ratio R(t) together with its Fourier transform F(w) for a sample before annealing. The peaks I, II, III and IV in the Fourier transform are caused by different magnetic fields at the Cd-nuclei. The corresponding frequencies, amplitudes, phases and hyperfine fields are given in the following table.

<table>
<thead>
<tr>
<th>2ω /10^6 s^-1</th>
<th>amplitude b_{II}</th>
<th>phase</th>
<th>B_{eff}/kG</th>
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<tr>
<td>75 ± 10</td>
<td>0.018</td>
<td>τ</td>
<td>-(27 ± 4)</td>
</tr>
<tr>
<td>132 ± 10</td>
<td>0.030</td>
<td>τ</td>
<td>-(65 ± 3)</td>
</tr>
<tr>
<td>280 ± 30</td>
<td>0.043</td>
<td>0</td>
<td>+(90 ± 4)</td>
</tr>
<tr>
<td>530 ± 50</td>
<td>0.02</td>
<td>?</td>
<td>?(170 ± 20)</td>
</tr>
</tbody>
</table>

The existence of other fields cannot be excluded. After an annealing procedure at 500°C for 20h the same sample showed a normal TDPAC-curve with the well known CdNi-field of -(65 ± 3) kG /1,2/ and an amplitude of 0.088 ± 0.006. This means that after the annealing most of the Cd-nuclei were substituted in nickel. This is a contrary behaviour to the cases of Ta(Ef) /3/ or Lu(Yb) /4/ implanted in Ni or Fe. The reason is that Cd(In) is better soluble in Ni than Ta(Hf) or Lu(Yb) in Ni or Fe.

In fig. 2 is shown the amplitude b_{II} to the main field (B_{eff} = -65 kG) as a function of the tempering temperature. At each temperature the sample was kept for two hours. After slowly cooling the measurements were carried out at room temperature. Because the amplitude b_{II}(T) is proportional to the number of In-111 nuclei sitting at the most stable lattice site after tempering at the temperature T, b_{II} - b_{II}(0) (b_{II}(0) is the saturation value) is a measure for the lattice imperfection. Therefore we compared the measured values with a theoretical annealing isochrone. The drawn curve was calculated for a reaction order of 2 and an activation energy of 0.52eV. A reaction order of 3 and an activation energy of 0.79eV also gives a good agreement. From the annealing isochrone we cannot decide, which reaction order is the most probable. But the annealing isochrone shows that after 9h at 500°C the annealing is nearly finished. This can only explained by a reaction order of approximately two.

Our conclusions from these experiments are the following: After implantation the In-111 nuclei can at least take place at four different but definite lattice sites. Ni has a fcc-lattice and therefore besides the substitutional position three stable interstitial positions for a Cd-atom. The space for such an interstitial occupation is smaller than that for substitution.
From this we can qualitatively understand the change of the field caused by overlap of the 5s-electron wave functions from the Cd atoms with the 3d-wave functions of the neighbouring Ni-atomes.

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*present address: TH Dresden, Sektion Physik
The location and hyperfine interaction of Se implanted into Fe and Co, determined by channelling, nuclear orientation and NMR/OK

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The use of ion implantation as a technique for sample preparation of dilute alloys intended for hfs investigations necessitates some independent investigation of the implant lattice location in order that a proper interpretation of the hyperfine interaction measurement can be made. In this paper such a combined experiment to measure the hyperfine interaction for Se in Fe and Co is reported. The magnetic hyperfine interaction for 4s-4p impurities in ferromagnetic metal hosts may be expected to show a similar behaviour to the striking change from negative to positive fields exhibited by analogous 5s-5p impurities. Because of their general insolubility in the ferromagnetic metals, the final members of the 4s-4p series can only be studied in alloys formed by ion implantation. Recent channelling experiments on Br\(^{(1)}\) implanted into Fe indicate a distribution of sites with only a fraction of implanted atoms substitutional. Direct measurement of the magnetic hyperfine field at substitutional Br is thereby complicated, although average field measurements have been made\(^{(2)}\). Any estimate using systematics depends greatly on an accurate determination of the hyperfine field at substitutional Se in Fe.

In the present experiment active samples of Fe\(^{75}\)Se and Co\(^{75}\)Se intended for hfs measurements were prepared by implanting \(^{75}\)Se at 120 keV and at a dose of \(1 \times 10^{14}\) cm\(^{-2}\) into polycrystalline Fe and h.c.p. Co. Atom location was performed by the channelling and backscattering of 3.5 MeV \(^{14}\)N from polished single crystals of Fe and h.c.p. Co. These crystals had been implanted with stable Se atoms at 120 keV but with a dose of \(5 \times 10^{14}\) cm\(^{-2}\), the minimum at which location experiments could be satisfactorily performed. In the single crystal implantations the beam was inclined at \(-8^\circ\) to the nearest major crystal axis to avoid an anomalous implant depth distribution. The energy of backscattering of the N beam from the Se nuclei indicated that the implanted Se had a mean projected range of 350 ± 40Å. Figure 1(a) shows the result of a complete angular scan across the \langle110\rangle axis of the \((5 \times 10^{14}\text{ cm}^{-2})\) Fe crystal in a tilting plane at \(14^\circ\) from a \{211\} plane. All counts have been normalized to the random level. The Se yield closely follows the Fe yield indicating a substitutional Se location. This conclusion is substantiated by minimum yield (\(X_0\)) measurements for the \langle111\rangle and \langle100\rangle axes and \{100\} plane for which the Se and Fe yields agree within experimental error. A \(c\)-axis scan (shown in figure 1(b)) indicated that implanted Se has a similar substitutional location in Co.

For the magnetic hyperfine interaction determination, implanted \(^{75}\)Se nuclei were polarized through the hyperfine interaction by thermal equilibrium nuclear orientation. The magnetized Fe and Co hosts were cooled in separate experiments to \(-0.014\) K by contact with a demagnetized chrome alum salt pill. The \(\gamma\)-ray distribution from the polarized nuclear spins exhibits an anisotropy described by the expression

\[
W(\theta) = \sum_{\nu} \text{min}(2I,2L) \left( \frac{A_{\text{INT}}}{B_{\nu}} \right) P_\nu (\cos \theta) \quad \nu \text{ even}
\]

where \(I\) is the nuclear spin and \(L\) the observed transition multipolarity. The other terms have been described elsewhere\(^{(3)}\). Figure 2(a) shows the observed change in axial \(\gamma\) count rate for the 136 and 280 keV transition in the \(^{75}\)As daughter for the case of \(^{75}\)Se oriented in Fe. In another part of the experiment modulated r.f. was applied and a change in the axial \(\gamma\) count rate was observed as the populations of the 2I+1 sublevels in the hyperfine
split ground state were altered by resonant r.f. absorption at the Larmor frequency (NMR/ON).

Figure 2(b) shows the NMR/ON signal observed at 142.1 MHz using the 136 keV γ-ray anisotropy as an indicator.

Since the resonant frequency is given by \( \frac{\mu}{H_{\text{ext}}} = \frac{\mu_0}{H_{\text{hf}}} + \frac{\mu_0}{H_{\text{ext}}} \), it is possible to independently determine \( \mu \) and \( H_{\text{hf}} \) by observing the change in resonant frequency under increasing applied field, \( H_{\text{ext}} \). In a shift experiment up to 20.9 kG, the following quantities were determined:

\[
\begin{align*}
\mu_0 & = 141.6 \pm 0.1 \text{ MHz (at } H_{\text{ext}} = 0) \\
\mu_0 & = 0.67 \pm 0.04 \text{ } \mu_0 \\
H_{\text{hf}} & = 690 \pm 50 \text{ kG}
\end{align*}
\]

A more accurate determination of \( \mu_0 \) by any means will fix \( H_{\text{hf}} \) more accurately.

Using the resonant frequency for Fe-75Se and taking this to be the hyperfine splitting for the 100% substitutional nuclear population (as determined by the channelling experiment) one may fit the 75Se γ-anisotropy to yield information about the nuclear decay through the \( U_\gamma \) and \( F_2 \) parameters. The fit to the 116 keV anisotropy shown in figure 2(a) determines the electron capture decay to be pure \( \frac{1}{2} \) (Fermi) while the fit to the 280 keV transition yields an E2/HI admixture amplitude of \( \frac{1}{2} (\frac{1}{2}) = 0.35 \pm 0.03 \) or \( 1.4 \pm 0.1 \) in good agreement with other workers.

The observed Co-75Se anisotropy was fitted to give a value for the hyperfine field at Se in h.c.p. Co of \( H_{\text{hf}} \) (CoSe) = -420 \( \pm \) 40 kG or -440 \( \pm \) 40 kG. NMR/ON was looked for but not observed, a result which may be attributed to quadrupolar line broadening due to a crystal electric field gradient at the non-cubic lattice site.

The combined atom location - NMR/ON experiment on implanted Selenium shows the interdependence of the techniques. The necessity for implanting the Se atoms was confirmed by a nuclear orientation experiment on a thermally prepared sample of Fe-75Se in which the observed γ-anisotropies were significantly lower, indicating a hyperfine field \( \sim 75\% \) on average of the substitutional value. This discrepancy in location between diffused and implanted sources is anomalous since previous implantations on other systems have produced totally substitutional alloys only when the impurity was in any case able to be placed on the lattice site by thermal diffusion.

References
3. R.J.Blin-Toyle and M.A.Grace, Handbuch der Physik 42 (1957) 555
Recent channelling experiments$^1$ have shown that $^{61}\text{Br}$ atoms implanted into Fe at an energy of 80 keV and a dose of $5 \times 10^{14}$ cm$^{-2}$ have a site distribution with 10% of impurity atoms substitutional and the remainder in a site of (specified) lower symmetry. This site the authors called an X-site. The location was found to be independent of dose over two orders of magnitude. An example of one of the six axial and planar scans used in the interpretation of the channelling experiment referred to is shown in figure 1. This shows an angular scan across the Fe $\{100\}$ planar channel using a 3.5 MeV $^4\text{He}$ beam along with the calculated impurity backscattering yields (solid line - Monte Carlo, dashed line - analytic) for the 60k X-site, 40k substitutional Br distribution$^1$.

The existence of a two site distribution for implanted Br in Fe greatly complicates the interpretation of hyperfine interaction measurements. However the quantitative determination of the site occupancies allows one to fit, for a two site hyperfine Hamiltonian, the temperature dependent $\gamma$-anisotropy measured in a nuclear orientation experiment on radioactive Br implanted into Fe. In the nuclear orientation experiment reported here, $^{82}\text{Br}$ (moment 1.6261 m.m. and spin 5$^1/2$) was implanted into a single crystal of iron (at $\approx 7^\circ$ to the $\{110\}$ axis normal to the surface) under the same conditions as for the location study samples. The crystal was connected by its copper backing strip to the cold finger of a chrome alum salt pill and cooled by salt demagnetisation to $\approx 11$ mK.

Figure 2 a) shows the axial $\gamma$-anisotropy measured with a Ge(Li) detector for the pure E1 554 keV transition in the decay of the polarized $^{82}\text{Br}$ when the Fe crystal was magnetized along a $\{110\}$ direction. Similar data was obtained for the 828 keV and 1044 keV transitions (pure E1 and pure E2) and in interpreting the data, all three sets of data were used.

An estimate of the substitutional hyperfine field at Br in Fe can be obtained by extrapolation from known hyperfine field systematics at the end of the 4p shell. The adjacent known field for $^{85}\text{Se}_{\text{Fe}} + 690(50)\text{kG}$ strongly points to a Br substitutional field of about 1000 kG, of similar magnitude to that in the S-p shell.$^6$ In fitting the $^{82}\text{Br}_{\text{Fe}}$ $\gamma$-anisotropy vs temperature data, it is useful to propose a purely magnetic substitutional hyperfine interaction (because of cubic symmetry) and an interstitial Hamiltonian $\mathbf{H}_i$, containing both magnetic dipole and electric quadrupole terms. The anisotropy is then described by an equation of the form

$$\eta(\theta) = \sum \varepsilon \varepsilon' \mu^2 \varepsilon' \left( x_5^2 \varepsilon \varepsilon' \left( \varepsilon \varepsilon' \frac{\mu^2}{kT} \right)^2 + (1 - x_5) \varepsilon \varepsilon' \left( \varepsilon \varepsilon' \frac{\mu^2}{kT} \right) \right)$$

where $x_5$ is the substitutional fraction and $H_5$ the substitutional magnetic field.

Figure 1 a) shows a single field fit to the 554 keV data with $x_5 = 1.0$ and $H_5 = 390$ kG but it is clear that the temperature dependence is poorly represented by such a model. Also shown in a two field fit where $x_5$ is taken as 0.4 (from the channelling experiment) and $H_5$ is taken to be zero. In this case the best fit for $H_5$ is 780 ± 40 kG. If $H_i$ is allowed to include a non zero magnetic term then other fits for $H_5 \leq 780$ kG are possible and such curves would be intermediate to those shown.

Although the "780 kG + 0 kG" model seems to fairly well reproduce the observed temperature dependence for the 554, 828 and 1044 keV anisotropies, the substitutional field used is considerably lower than that predicted by systematics. A fit has therefore been made using $H_5 = 1145$ kG (the Fe field$^4$) but even when the 60k Interstitial nuclei have $H_i = 0$, the $\gamma$-anisotropies calculated lie well above the data indicating that on this model the interstitial bromine must contribute $\gamma$-anisotropy of the opposite sign. Such sign reversal can only be explained by the inclusion of a quadrupole term in the interstitial hyperfine Hamiltonian. Figure 2 b) shows the 554 keV "interstitial anisotropy" calculated for each data point by assuming $H_5 = 1145$ kG. A simple mixed magnetic dipole plus electric quadrupole Hamiltonian is then used to fit the interstitial anisotropy data.

This has the form

$$H_i = -x_5 H_5 + \frac{3 e Q V x_5}{41(2I+1)} (I_{1z}^2 + \frac{1}{3}(I_{1z})^2) \left( \frac{3 e Q^2 2^1}{2} \right)$$

where Q is the nuclear electric quadrupole moment of $^{82}\text{Br}$ (0.7e x $10^{-28}$ m$^2$). Two extreme best fits are shown. The first has $H_i = H_5$ and $\frac{3 e Q V x_5}{41(2I+1)} / (x_5 H_5^2) = 0.196$ and the second has $H_i = 0$ and $\frac{3 e Q V x_5}{41(2I+1)} / (x_5 H_5^2) = 0.03$. The best fit for all the transitions is obtained for the low magnetic field interstitial Hamiltonian.

The present model, while it uses the evidence of the channelling experiment, rests on an assumption about systematic trends in hyperfine field data. The indications given about the character of the interstitial hyperfine interaction do however appear sensible. For a dominantly conduction electron magnetic hyperfine interaction one might expect (for the positive substitutional field case) that an increase in the core polarization...
contribution off the lattice site would lead to a less positive interstitial magnetic hyperfine field. Furthermore the low symmetry of the X-site suggests the existence of a hyperfine quadrupole term. As an order of magnitude calculation, an estimate of the electric field gradient $V_{zz}$ can be obtained by assuming a single unscreened charge at the lattice site nearest the X-site. For such a model $\frac{1}{4}(21^{-1}) = 0.02$ when $H_s = 1145$ kG, which is similar to the value of 0.05 used to fit the data when the interstitial magnetic field is taken to be small.

References


![Figure 1](image1.png)

![Figure 2](image2.png)
The Mössbauer effect provides a powerful method for studying the nature of the various sites at which implanted impurity atoms may be found in a crystal lattice. Since inequivalent sites generally produce different hyperfine spectra, the number of spectral components observed is an index of the number of inequivalent sites occupied by the implanted ions. The recoilless fraction of each component can be measured by varying the source temperature, thus allowing a determination of the relative site populations. In the case of xenon impurities in iron, the implantation-induced damage gives rise to the formation of at least three spectral components in the Mössbauer effect measurements\(^1\) with different magnetic splittings, isomer shifts and recoilless fractions.

The sites were called high-field sites (h-sites), intermediate-field sites (i-sites) and low-field sites (l-sites) according to their respective magnetic hyperfine interactions. Apparently, h-sites correspond to substitutional implanted xenon atoms surrounded by 8 iron atoms (confirmed by channeling measurements\(^3\)). i-sites correspond to xenon atoms associated with one vacancy and l-sites to xenon atoms associated with two or more vacancies.

In the present investigation the Mössbauer effect of the 81 keV transition in \(^{133}\text{Xe}\) was used to study the lattice location of \(^{133}\text{Xe}\) implanted in iron at temperatures ranging from 90 K to 480 K. Isochronal annealing experiments were done with sources implanted at 90 K and at room temperature (RT). Sources of 50 to 1000 \(\mu\)Ci were prepared with the Groningen isotope separator\(^4\) using gaseous xenon diluted with natural xenon as the charge material. The implantation energy was 140 keV. The concentration of implanted impurity atoms was kept low by beam sweeping (<0.1%, typical doses of \(10^{14}\) atoms/cm\(^2\) or less). A single line 460 mg/cm\(^2\) CoCl absorber, was used. All spectra were taken with source and absorber at 4.2 K in transmission geometry. Details of the experiments and the data analysis are given in ref. 5. Fig. 1 shows Mössbauer spectra of \(^{133}\text{CaFe}\) after implantations at 90 K, room temperature, and 480 K respectively.

In the present report we concentrate on the behaviour of the relative site populations of the spectral components as a function of the implantation and annealing temperatures and therefore we confine ourselves to the results for the relative absorption depths of the various components. Average values for a number of measurements are given in columns 2-5 of table I. The results show that the low-field component increases with increasing implantation temperature while the high-field component decreases, but the total percentage absorption depth hardly changes. Because the recoilless fraction of the low-field component is much smaller than that of the high-field component \((f_h = 0.16 \pm 0.02, f_i = 0.12 \pm 0.03\) and \(f_l = 0.05 \pm 0.01\) at 4.2 K\(^5\)) the increase of the low-field component can not be due only to the decrease of the high-field component. The same abnormal increase of the low-field component is observed in the annealing experiments. The physical interpretation of the l-sites (i.e. xenon atoms associated with two or more vacancies) provides a solution to this problem: The visible l-component in the spectra consists only of xenon atoms surrounded by a few vacancies (typically from 2 to 4), while xenon atoms with a large number of vacancies (say 5 or more) have a very small recoilless fraction (certainly \(f < 0.01\)) and therefore hardly contributes to the Mössbauer absorption. A division of the l-component in two parts is therefore proposed. One part, consisting of clusters formed by xenon atoms associated with a small number of vacancies is (again) called the l-component. The second part, consisting of xenon-vacancy clusters with a large number of vacancies is called the s-component (recoilless fraction effectively zero). The large increase of the l-component can now be explained.
as follows: at moderate temperatures (250-500 K) the $x$-sites are not the most stable sites: it is energetically favourable for the large clusters to lose one or perhaps more vacancies e.g. by annihilation with migrating interstitials. Apparently, the most stable type of clusters are small ones, belonging to the $1$-fraction. A similar behaviour for this type of clusters as a function of implantation (or irradiation) temperature in molybdenum and rhenium has been indicated by Brimhall et al. $^6$). The difference in site populations found for the source implanted at 480 K and the source annealed to the same temperature after implantation at a lower temperature is due to the rather different equilibrium conditions during these implantations. At the higher implantation temperatures vacancies are mobile and a direct flow to the large xenon atoms (resulting in extra formation of the most stable clusters: $1$-sites) takes place, whereas at the lower implantation temperatures many of the vacancies have been annihilated by migrating interstitials before the annealing starts. Applying the four-component model just proposed to our measurements we can deduce site populations for several implantation temperatures and annealing temperatures, using the formula $a_{j} = \frac{r_{j}^{rel}}{f_{j}^{rel}}$ $^7$ where $a_{j}$ is the fractional occupation of site $j$, $r_{j}^{rel}$ is the relative absorption depth of site $j$, $f_{j}$ is the recoilless fraction of site $j$ and $f_{j}$ is the (known) resonantly absorbed fraction of the 81 keV $\gamma$-rays. This formula holds for the $h$-, $i$-, and $1$-sites; $a_{x}$ has to be determined from $a_{x} = 1 -(a_{h} + a_{i} + a_{1})$, because $f_{x} = 0$. The results are shown in columns 6-9 of table I.

References

4. S. A. Drentje, Nucl.Instr. and Meth. 59 (1968) 64.

**TABLE I**

Average values for relative absorption depths and site occupations of xenon in iron, implanted at and annealed to various temperatures with a typical dose of $1 \times 10^{14}$ at/cm$^2$ and an implantation energy of 140 keV.

<table>
<thead>
<tr>
<th>impl. or ann. temp. ($\text{K}$)</th>
<th>absorption depth (%)</th>
<th>site occupations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$a_{h}$</td>
<td>$a_{1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>3.3$^{\pm}0.02$</td>
<td>0.9$^{\pm}0.15$</td>
</tr>
<tr>
<td>300</td>
<td>3.6$^{\pm}0.02$</td>
<td>1.0$^{\pm}0.15$</td>
</tr>
<tr>
<td>500</td>
<td>3.8$^{\pm}0.02$</td>
<td>1.2$^{\pm}0.15$</td>
</tr>
<tr>
<td>700</td>
<td>3.0$^{\pm}0.02$</td>
<td>1.5$^{\pm}0.15$</td>
</tr>
<tr>
<td>90</td>
<td>3.3$^{\pm}0.02$</td>
<td>1.6$^{\pm}0.15$</td>
</tr>
</tbody>
</table>
THE LATTICE LOCATION AND HYPERFINE FIELD
OF IMPLANTED Yb IN Fe AS A FUNCTION OF ANNEALING TEMPERATURE
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The importance of lattice location (channeling) studies for the interpretation of hyperfine interaction measurements on implanted sources has been recognized for some time. However, additional information is often required to understand the physical origins of the hyperfine field (hf). Earlier work on implanted Yb in Fe showed that both the lattice location of the Yb and the average hf changed upon annealing. To investigate the nature of the hf, both the location and hf have been determined as a function of annealing temperature.

Channeling and PAC measurements were made on Fe single crystals implanted with $2 \times 10^{14}$ Yb ions/cm$^2$ at an energy of 60-80 keV, and annealed in vacuo at temperatures up to 600°C. For the channeling experiments stable $^{172}$Yb was implanted. Using backscattering of 1.5-3.5 MeV $^{12}$C and $^{14}$N ions, channeling angular distributions were measured about the major crystal axes. For the PAC experiments radioactive $^{169}$Yb (decaying to excited states in $^{171}$Tm) was implanted. The anisotropy and rotation for both the 177-131 keV $\gamma$-$\gamma$ cascade (7/2$^+$ state) and the 198-110 keV $\gamma$-$\gamma$ cascade (5/2$^+$ state) were determined, at both room temperature and 140°C (after each anneal) to check on effects from electronic relaxation.

It is found from the channeling measurements that initially 58% of the implanted Yb occupies substitutional sites in the Fe lattice, while the remainder does not occupy any specific crystallographic site. On annealing Yb migrates from substitutional to non-substitutional sites; no atoms remain substitutional after a 600°C anneal. Typical results of the angular scans are shown in Fig. 1. The small, broad hump seen near the centre of the Yb yield curves is associated with the channeling flux-pecking effect, and confirms the 'random' distribution of the non-substitutional Yb atoms.

The PAC pattern for $^{169}$Yb in Fe can be described using a combined static and time-dependent magnetic interaction. The ratio of the measured relaxation parameters $\lambda$ for the 7/2 and 5/2 states is consistent with this assumption. By measuring the correlation patterns for both cascades as well as the measurements at two different temperatures after each anneal, the average hf $H_{\text{eff}}$ is overdetermined and the behaviour of Yb in physically different surroundings can be studied. Under the assumption that there is only a magnetic and a non-magnetic Yb site, the observed correlations can be fitted to yield the fraction of Yb atoms in each site. However, a better fit to the data is obtained using a correlation of the form $W(\theta) = W(0) H_{\text{eff}}(\theta^0)$, where $H_{\text{eff}}$ and $\lambda$ are the only fitting parameters. Here $H_{\text{eff}}$ and $\lambda$ represent some type of average value of the hf and relaxation parameter, respectively. Both $H_{\text{eff}}$ and $\lambda$ are found to decrease with annealing temperature, and the hf disappears after a 600°C anneal. The results for both the 7/2 and 5/2 states are consistent.

For comparison with the channeling results, a quantity

$$F = \frac{H_{\text{eff}}(T)}{H_{\text{eff}}(0)}$$

can be defined, where $H_{\text{eff}}(T)$ is the average magnetic field after annealing at temperature $T$ and $H_{\text{eff}}(0)$ before annealing (i.e. after implantation). In a two-site model, $F$ is the normalized fraction of atoms in high-field sites. Values of the parameter $F$, together with the normalized fraction of Yb atoms in substitutional sites from the channeling measurements, are shown in Fig. 2. A definite correlation is seen between the location and hf results, the high magnetic field being associated with the substitutional site. It appears that Yb atoms migrate from magnetic to non-magnetic sites on annealing.

Mössbauer experiments$^2$ indicate a very similar annealing behaviour for implanted $^{151}$Gd in Fe. The isomer shift shifting a 600°C anneal was found to correspond to $\text{Gd}_2\text{O}_3$, suggesting that Yb implanted in Fe may also reside in an oxide after annealing at 600°C. It has been suggested in other work$^3$ that the Yb may form precipitates on annealing. To investigate this, the relaxation parameter $\lambda$ was measured for sources of $^{169}$Yb$_2\text{O}_3$ and activated $^{169}$Yb in Yb metal. The values of $\lambda$ for the oxide source agree with those obtained on the 600°C-annealed $^{169}$Yb in Fe sample, while those for the activated Yb source are significantly lower.

Taken together with the location results, this is very strong evidence that the non-substitutional Yb is present as an oxide in the Fe host. As the backscattering measurements indicate that the implanted Yb does not reside in the surface oxide layer (< 40 Å thick), it is concluded that the non-substitutional Yb is trapped by interstitial oxygen in the Fe crystals. It is well known that rare earths have a strong affinity for oxygen.
The results of this study may therefore be relevant to hf measurements on other rare earths implanted in Fe.

References
(3) F. Abel, M. Bruneaux, C. Cohen, H. Bernas, J. Chaumont and L. Thome, ibid.

Fig. 1. Channeling angular scans across a (111) axis in 172Yb-implanted Fe single crystals after annealing at different temperatures.

Fig. 2. Normalized fraction of 172Yb atoms in substitutional sites (channeling measurements), and normalized fraction of 169Yb atoms in high-field sites (PAC measurements), as a function of annealing temperature.
Recent experiments measuring the anisotropic gamma-ray distribution from oriented nuclei have shown that the mixture of small electric quadrupole and magnetic hyperfine interactions can significantly change the angular distribution of gamma-rays following the population of relatively short-lived excited states in the nuclear decay. The normal situation in nuclear orientation experiments is that the angular distribution of gamma radiation is directly related to the nuclear ordering of the long-lived parent state in the hyperfine interaction of its environment. No reorientation in the intermediate nuclear states populated in the decay will occur if (a) the intermediate state lifetimes are sufficiently short that no nuclear spin-lattice relaxation occurs, or (b) if the axis of quantisation for the hyperfine interaction is the same for all states. It is the latter condition that does not hold when the relative magnitudes of magnetic dipole and electric quadrupole hyperfine interactions differs in the parent state and intermediate states. This is a different situation to that discussed by Daniels and Misra where strong transient fields following K-capture are responsible for attenuations of gamma-ray anisotropies from nuclei with this decay mode.

In contrast to the time required for nuclear reorientation by spin-lattice relaxation, the change in quantisation axis for an intermediate state can produce substantial reorientation in a time comparable with the nuclear Larmor precession period. A situation in which nuclear reorientation of this type can give a large change in the subsequent gamma-ray angular distribution has been found to exist for two isotopes of bismuth present as dilute impurities in iron and nickel. Measurements of many gamma transitions in $^{206}$Pb, $^{207}$Bi in Fe and Ni can be interpreted in terms of the parent state orientation and known decay sequences. However all transitions following decay of the 120 μs 2.2 MeV isomer of $^{206}$Pb and the 0.9 ± 1.63 MeV isomer of $^{207}$Pb show attenuated anisotropies which cannot be ascribed to uncertainties in the decay scheme, and also which vary with the impurity environment. These attenuations have been quantitatively explained in terms of an electric quadrupole contribution to the hyperfine interaction not co-axial with the magnetic dipole term. The existence of the electric quadrupole interaction is attributed to the non-cubic symmetry of the bismuth impurity environment in a non-substitutional site in the host lattice.

The populations of the magnetic substates of the isomeric levels at the instant of formation, $P_{k,0}$, are calculated from the appropriate angular momentum coupling to the oriented parent state. The bismuth isotopes both present a situation where the electric quadrupole interaction is only a small perturbation on the large magnetic dipole hyperfine interaction of the parent state, but is comparable in magnitude with the very much reduced magnetic interaction of the isomeric states in lead.

The nuclei are considered as subject to a Hamiltonian of the form

$$\hat{H} = \frac{-\hbar}{2} \sum_{l} \sum_{I_k} \left[ a_{l,k}^{\text{M}} \right]^{2} + \sum_{l} \sum_{m} \hbar \omega_{l,m} \left[ a_{l,m}^{\text{E}} \right]^{2} + \sum_{l} \sum_{m} \hbar \omega_{l,m} \left[ a_{l,m}^{\text{M}} \right]^{2} + \sum_{l} \sum_{m} \hbar \omega_{l,m} \left[ a_{l,m}^{\text{E}} \right]^{2}$$

where $\omega_{l,m}$ is the relative strength of the electric quadrupole and magnetic dipole components in the hyperfine interaction. The $a_{l,m}^{\text{M}}$ are to a lesser extent sensitive to the isomeric state spin and the degree of orientation...
of the parent nucleus. In the limit, \( X \rightarrow \infty \), for a pure quadrupole interaction, the parameters \( C_k \) represent simply a rotation or axis through the angle \( \phi \). In polycrystalline material all values of \( \phi \) will occur, and taking a weighted average we obtain

\[
\overline{C_k} \left( \lim X \rightarrow \infty \right) = \int_0^\pi \left[ F_k(\cos \phi) \right]^2 \sin \phi d\phi = [2k + 1]^{-1}.
\]

Analysis of the measurements on \(^{208}\text{Bi}\) and \(^{207}\text{Bi}\) has shown that the magnitude of the electric field gradient at the impurity site is the same in f.c.c. nickel and the f.c.c. phase of iron, frozen in by quenching from a temperature above the b.c.c./f.c.c. phase transition of iron. These results suggest a unique non-substitutional site for dilute bismuth impurities in these hosts. Implantation of bismuth in the b.c.c. phase of iron shows a slightly larger magnetic interaction, but an electric field gradient more than twice that for the bismuth site in the f.c.c. hosts.

This work was supported by grants from the Science Research Council.

References

Figure Captions

**Fig. 1:** The variation of the polycrystal attenuation parameters \( C_k \) as a function of the ratio of the magnetic dipole and electric quadrupole interaction strengths in an intermediate state of spin 7. The difference between the curves for positive and negative values of \( X \) is associated with the degree of orientation of the parent nucleus.

**Fig. 2:** The temperature dependence of the axial \( 516 \text{ keV} \) gamma-ray intensity from \(^{206}\text{Bi}\) nuclei oriented in the hyperfine field from a nickel host for various values of \( X \) in the 2.2 MeV state. The data points show the experimental results with a value \( X = 0.112 \). The curve for \( X = 0.086 \) is very similar.
As a result of the nuclear transformation which precedes the Mössbauer gamma emission a variety of phenomena may be observed in Mössbauer source experiments [1,2]. In the study of these aftereffects much attention has been particularly devoted to the study of charge states. Although there is no convincing experimental evidence for lifetime effects of charge states which result from the change in the configuration of the Mössbauer atom as a response to the change in nuclear charge, it is generally assumed that rearrangement among the configurations may be quite slow in insulators. Since the same rearrangement between configurations is known to be fast in metals, in the past it has usually been assumed that the Mössbauer atom in a metallic source reaches its thermal Boltzmann equilibrium in times short compared to the lifetime of the excited Mössbauer state.

Here we wish to discuss Mössbauer source experiments of dilute 4f impurities in metals with low lying Stark or Zeeman levels for which the rearrangement among these intraconfigurational levels is slow enough to have important effects even when the rearrangement between configurations is very fast. In source experiments of such systems a non-equilibrium population of the lowest electronic energy levels might result as a consequence of the beta decay leading to a marked difference as compared to the corresponding absorber experiment. It will be shown that the problem of intraconfigurational rearrangement is closely connected to the problem of relaxation effects in the Mössbauer effect.

Mössbauer studies of rare earth (RE) impurities in metals need to be carried out as source rather than absorber experiments in order to achieve a high enough resonance signal at high dilutions. Since the solubility of the RE elements in the cubic host metals increases across the 4f series the most suitable candidates for Mössbauer studies are found to be $^{166}$Er and $^{170}$Yb. Both isotopes have a simple 2$^+$ to 0 Mössbauer transition of about 80 keV. The radioactive parents are $^{166}$Ho ($T_{1/2}=27$ h) and $^{170}$Tm ($T_{1/2}=130$ d) which directly feed the excited nuclear $2^+$ state by means of a $\beta^-$-decay. This $\beta^-$-decay starts both the Mössbauer $\gamma$-emission and the electronic rearrangement in response to the change in nuclear charge and will therefore fix the zero on our time scale.

Since the $\beta^-$-decay increases the nuclear charge by one, the electronic shell will change its configuration from $4f^N$ to $4f^{N+1}$. The time for this process may be estimated to be of the order of $10^{-14}$ sec. This time needs to be compared with the shortest characteristic time associated with the Mössbauer spectrum, i.e. the nuclear correlation time $(\omega_{hf})^{-1} \approx 10^{-10}$ sec, where $\omega_{hf}$ is the total hyperfine (hf) splitting. Thus rearrangement between configurations is indeed fast on the Mössbauer time scale and the excited charge state $4f^{N+1}$ cannot be seen in Mössbauer source experiments in metals.

Next we need to consider the rates with which the rearrangement proceeds within the $4f^{N+1}$ ground configuration. For an impurity in a metal this rearrangement will be produced by the coupling of the impurity to the conduction electron bath in the sense of a generalized a-f interaction. This coupling should be thought of as to provide transitions among all levels within the ground configuration and in second order perturbation theory one obtains for the rate of such a transition between a state $i$ and another state $f$ /5/:

$$K_i^f = 2\pi^2 |I_c^f g(\epsilon_f)|^2 \frac{\omega_{hf}^2}{\exp(\omega_{hf}/kT)-1}$$

(1)

Here $I_c^f$ stands for a combination of coupling constants which should be approximately of the same order of magnitude for all transitions within $4f^{N+1}$, and $g(\epsilon_f)$ is the density of conduction electron states. For rearrangement between an excited state $i$ and a lower state $f$ which are separated by an energy $|\omega_{hf}|$ large compared to kT, $\omega_{hf}$ is negative and from equation (1) it is seen that the rearrangement decays proceed at a rate which is approximately proportional to $|\omega_{hf}|$. The factor $2\pi^2 |I_c^f g(\epsilon_f)|^2$ may be estimated to be $10^{-3}$ for Er and $10^{-2}$ for Yb impurities /4,5/. This implies that intraconfigurational rearrangement can be regarded fast on our time scale when

$$|\omega_{hf}| > 10^{13} \text{sec}^{-1} \approx 0.01 \text{eV} \approx 100 \text{K} \text{ for Er and } |\omega_{hf}| > 10^2 \text{K} \text{ for Yb. For } 4f \text{ elements excited } [3l] - 3$$
states lie typically 0.1 to 10eV above the ground $[5L]-J$ manifold and it can therefore be assumed that the decay of the electronic shell into the $[5L]-J$ ground manifold of the ground configuration $4^2S^1$ is fast. However, our estimates show that decays of the CEF levels within the lowest $[5L]-J$ manifold whose energies may be only a fraction of 100K are not necessarily fast.

The implications of our considerations can be understood best by comparing a source with a hypothetical absorber experiment. In the absorber experiment one has a thermal-equilibrium Boltzmann population of electronic energy states as the initial preparation of the system. In a source experiment the above estimates indicate that the decay of the electronic shell into its $[5L]-J$ ground manifold following the beta decay is so fast as to be quasi-instantaneous. This cascade through higher energy levels results in an initial population which can be represented quite accurately as an equal population of all states within the $[5L]-J$ ground manifold. The further electronic rearrangement which occurs over the time of Mössbauer $\gamma$-emission is then described by the same equations of motion which are used to describe the Mössbauer relaxation phenomena. These equations are the same as would be used for an absorber experiment, except that a different initial population of the CEF levels is assumed. The relaxation rates which enter the relaxation theory are explicitly given by eq. (1). Although an equal population of the CEF levels within the free ion ground state is assumed as the initial distribution, this does not mean that all these levels contribute equally to the Mössbauer hf spectrum. The higher CEF levels will tend to decay to the ground state before the gamma emission is complete, but their contributions are more prominent than would be in the corresponding absorber experiment.

Our theoretical calculations are found to be in good agreement with experimental results on the $^{166}$Er Mössbauer resonance of $^{166}$Ho:Au alloys /6/, where two excited CEF levels ($\gamma_2$ and $\gamma_3$) are known to be close (~20K) to the ground state $\gamma_4/4$/. In our spectra /6/ a rather broad central structure is always observed in addition to the well resolved hf structure of the $\gamma_4$ doublet, and it is striking that it remains at a constant finite value in the low temperature limit. According to equation (1) the relaxation rate from an excited CEF level to the CEF ground state approaches a finite limiting value for temperatures small compared to the CEF energy difference. Since the initial populations are also temperature independent, the experimental observation of a finite contribution from excited CEF states in the low temperature limit is in full accord with the theoretical prediction. More experimental evidence for the effectiveness of our theoretical considerations comes from studies of the $^{170}$Tb resonance in $^{170}$Tm:Au alloys in magnetic fields between 20 and 50KG. In this case the contributions from the upper Zeeman level of the well isolated $\gamma_4$ ground doublet are found to be much more pronounced as would be allowed by Boltzmann statistics.

Finally, we mention that all rearrangement effects discussed above are typically much slower in insulators. Intracorfigurational rearrangement, eg. between $[5L]-J$ manifolds, will often be able to proceed only via inefficient multiphonon processes and one may thus obtain a large number of contributions from excited intraconfigurational levels. It appears that this mechanism has been neglected in the interpretation of chemical aftereffects /2/. The implications of our discussion for perturbed angular correlation experiments should also be pointed out.

The knowledge of the ultimate charge state of nucleogenic impurities is of paramount interest for radiation damage studies and may give fundamental clues to the nature of chemical bonding. Though the charge state of atoms of decaying nuclei in conductors can be predicted, this would not be the case in insulators, where the mechanism of charge compensation is not well understood.

In this paper we present results on investigations of the "fate" of a nucleogenic Te impurity produced by the electron capture decay of $^{125}\text{I}$ in the insulator $\text{MnI}_2$. By means of the hyperfine interactions in antiferromagnetic $\text{MnI}_2(T_N = 3.4^\circ\text{K})$ and by applying the Mössbauer method in $^{125}\text{Te}$ we were able to identify the formal ionic state of the Te impurity and deduce its electronic structure. It will be shown that from the quadrupolar interactions below and above $T_N$ we could conclude, unambiguously, that a Te$^0$ species following the $^{125}\text{I}^+ \rightarrow \text{Te}^0$ nuclear transformation is formed and stabilized.

Halogeous iodide has a hexagonal layer structure of the CdI$_2$ type (see insert in Fig. 1). Neutron diffraction carried out by Cable et al.\textsuperscript{(1)} proposed a magnetic structure of the helical type where the moments on the (307) planes are ferromagnetically aligned, rotating by $2\pi/16$ in successive (307) planes. Measurements were performed with a $\text{Hg}^{125}\text{I}$ source and a single line ZnTe absorber at 77, 4.2 and at 1.4 K. The source was prepared from carrier-free $^{125}\text{I}$, by oxidizing the iodide to I$_2$ and allowing it to react with very pure Mn metal in vacuum at 700°C. The quality of this preparation was checked with a $^{129}\text{I}$ absorber\textsuperscript{(2)}.

In Fig. 1a and 1b we present spectra taken at 1.4 K and 4.2 K respectively.

The spectrum at 1.4 K was analysed by solving for the eigenvalues and eigenvectors of the following spin-hamiltonian, namely:

$$\hat{H} = \frac{\alpha q}{4I(I+1)} [3I_z^2 - I(I+1)] + \mu_{\text{hf}} [I_x \cos \theta + (I_x + I_y) \sin \theta]$$

where $\theta$ is the angle formed by the internal magnetic field $-\mu_{\text{hf}}$ and the z-axis of the axially symmetric electric-field-gradient (c.f.g.). The solid line through the experimental points is a least-squares-fit curve.

![Fig. 1. The emission spectrum of $\text{Mn}^{125}\text{I}_2$ (c.e. $^{125}\text{Te}$) at 1.4 K (a) and 4.2 K (b). The solid lines through the experimental points are theoretical curves obtained by the least-squares-fit method (see text).](image-url)
based on the following parameters:

\[ h_f = 250 \pm 2 \text{ kHz} \]
\[ e^2 Q = -19.2 \pm 0.5 \text{ mm/sec} \]
\[ 2 \gamma = 9.2 \text{ mm/sec} \]
\[ \theta = 73^\circ \pm 2^\circ \]

The relevant parameter is the quadrupole coupling constant, its magnitude and sign. The sign of \( e^2 Q \) is positive, and since the sign of the quadrupole moment \( -eQ \) of the first excited state of \( _{125} \text{Te} \) is negative, the sign of the e.f.g. \( -eQ \) is negative. Thus, the e.f.g. is produced by an unpaired p-electron in the Te outer shell. The magnitude of \( e^2 Q \) coincides with that produced by a p-electron in Te calculated by using the value of \( r_p \) of Barnes and Smith. The only configuration which fits to the above conclusions is \( 5p_x^1 5p_y^1 5p_z^2 \), namely the outer shell of a Te atom.

Further evidence for the existence of a neutral Te atom weakly bound to its \( I^{1-} \) and \( I^{2+} \) neighbours we deduce from the behavior of the quadrupole interaction at high temperatures. At 77 K a single line spectrum with a line-width of 8.9 mm/sec, the same value as obtained with a \( 125\text{I(Cu)} \) source, is observed. At 4.2 K the line is broadened (see Fig. 1b) with a "line-width" of 12.5 mm/sec. The lack of static quadrupole interaction and the significant broadening at the lower temperature leads us to interpret this spectrum as due to quadrupole relaxation. The origin of this relaxation is the degeneracy in the 5p levels where the extra p-electron hops among the \( p_x, p_y, \) and \( p_z \) subshells. The relaxation time for this process at 4.2 K was found by applying the method of Tjone and Blume, assuming equal probability for the three components of \( -eq \). Using a least-squares fitting method we obtained the solid line shown in Fig. 1b. The mean relaxation time deduced was 18.4 usec.

We proceeded to discuss the possibility of other charge states formation such as \( I^{1-} \) and \( I^{2+} \) and how they would have been characterised by the Mössbauer spectra. Concerning charge conservation, the immediate product of the \( I^{1-} \) decay is a \( I^{1+} \) ion. This ion has one p-hole in its outer shell - similar to atomic I - resulting in a negative \( e^2 Q \). Thus, at 1.4 K a mirror image spectrum would have been observed following the inversion of the nuclear sub-levels by the negative quadrupole coupling constant. At \( T > T_N \) one would have observed a doublet due to a static quadrupole splitting. The radius of \( I^{1+} \) is 2.5 \( \AA \) which is significantly larger than that of atomic Te (1.43 \( \AA \)); therefore a sizeable overlap with its \( I^{1-} \) and \( I^{2+} \) neighbours would be expected. This well defined bonding would have made the quadrupole relaxation phenomena very improbable.

Our present results suggest that a \( I^{1+} \) species in such an ionic crystal is energetically unstable. The possibility of a \( I^{2+} \) formation can be easily ruled out from the fact that \( I^{2+} \) has a closed shell; therefore at \( T < T_N \) one would have observed a six-line symmetric pattern due to the absence of quadrupole interaction; at \( T > T_N \) a single narrow line spectrum would have appeared.

From the limited number of measurements taken in this work we cannot determine the reason for the quenching of the quadrupole relaxation at 1.4 K. Probably the onset of the long range magnetic order at \( T > T_N \) removes the degeneracy of the p-sublevels thus forming a well defined orientation of \( q \) with respect to the direction of the magnetic moments. The slow relaxation rate at 4.2 K could be due to residual short-range ordering partially affecting the degeneracy of the electronic states. In conclusion, we have shown that from the sign and magnitude of the quadrupole coupling constant of \( _{125} \text{Te} \) we were able to identify the final product of a \( _{125}^{125}\text{I}^{1-} \) e.c. decay in \( \text{Mn}_2 \) as atomic Te. This observation is consistent with the occurrence of quadrupole relaxation at \( T > T_N \).

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III. NUCLEAR MOMENTS
DETERMINATION OF THE CHANGE OF THE NUCLEAR CHARGE RADII FOR ROTATIONAL TRANSITIONS IN $^{178}$Hf AND $^{182}$W

III. 1

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The interest in Mössbauer measurements to determine the change $\Delta\langle r^2 \rangle$ of the mean square nuclear charge radius of deformed even-even nuclei undergoing rotational transitions has been stimulated by the success of recent calculations (1) based on the self-consistent cranking model, that can, at least qualitatively, explain the small and sometimes even negative values observed for $\Delta\langle r^2 \rangle$. We wish to report the determination of $\Delta\langle r^2 \rangle$ for the 100 keV and 93 keV transitions in $^{182}$W and $^{178}$Hf, respectively. In these cases the isomer shifts are of the order of, at best, a few percent of the natural linewidths. Therefore the experiments were performed with a Mössbauer spectrometer capable of recording the spectra of three source-absorber combinations at the same time (2). Since in this spectrometer the sinusoidal motion of all three sources is provided by a single velocity drive, the experimental uncertainties in the relative shift between the three spectra recorded at a time are reduced to practically the statistical error. The isomer shifts were measured between sources containing the $^{178}$W and $^{182}$Ta activities as dilute impurities in cubic transition metals. Metallic tungsten (48 mg/cm$^2$) and Hf (55 mg/cm$^2$) served as single-line absorbers. The systematics of isomer shifts in dilute alloys of transition metals have recently been established (3,4) sufficiently well for an evaluation of such data in terms of $\Delta\langle r^2 \rangle$. By the use of different sources with always the same absorbers, it was possible to avoid inaccuracies that may occur due to the dispersion term in the resonance cross-section (5-7), if isomer shifts are measured between absorbers of different Mössbauer thickness. Only cubic host metals were used for the sources in order to avoid shifts of the Mössbauer line that can be caused if samples with an unresolved quadrupole splitting cannot be made without some texture or have an anisotropic recoilless fraction. In the $^{182}$W measurements one

of the three sources used at a time always consisted of Ta metal, which was used as a reference standard for all $^{182}$W isomer shifts. Due to the smallness of the isomer shifts, the second-order Doppler shift has to be taken into account. The second order Doppler shifts for the individual Mössbauer sources were calculated in the Debye approximation from experimental values of the recoilless fraction. For the 100 keV line of $^{182}$W second order Doppler shifts between -0.0043(11) mm/s for the W matrix and -0.0046(8) mm/s for the Pd matrix were obtained and subtracted from the experimental values for the shifts of the Mössbauer lines. The resulting isomer shifts are plotted in fig. 1 versus the isomer shifts for the 6.2 keV Mössbauer line in the respective host lattices (4).

The correlation between the $^{182}$W and the $^{181}$Ta isomer shifts exhibited by this plot is not unexpected if one considers the behaviour found for other Mössbauer isotopes as dilute impurities in transition metals (3,4). In particular, isomer shift results for $^{57}$Fe, $^{93}$Ru, $^{180}$Os, $^{193}$Ir, $^{195}$Pt, and $^{197}$Au show that generally the electron density $\psi(0)$ at an impurity nucleus in a 4d host is larger than in the homologous 5d metal, and still larger in the homologous 3d host. This behaviour has recently been used to derive a value of $\Delta\langle r^2 \rangle = -50 \times 10^{-3}$ fm$^2$ for the 6.2 keV transition in $^{181}$Ta (4). The correlation between the $^{181}$Ta and $^{182}$W isomer shift (fig. 1) shows that the sign of $\Delta\langle r^2 \rangle$ for the 100 keV transition in $^{182}$W is the same as that for $^{181}$Ta. In order to obtain the magnitude of $\Delta\langle r^2 \rangle$ for tungsten, we assume that a sufficiently accurate estimate for the ratio of the electron density differences for W and Ta as impurities in the same transition metal hosts can be obtained from Dirac-Fock-Slater calculations for free ion 5d$^2$Te$^0$ and 5d$^2$Se$^2$ electron configurations of both elements. For these configurations one calculates (8) $\Delta\phi(0)_W/\Delta\phi(0)_Ta = 1.08$. Using this value, the mean of the experimental values for the ratio of the isomer shifts of $^{181}$Ta and $^{182}$W in 4d and 5d hosts, and $\Delta\langle r^2 \rangle = -50 \times 10^{-3}$ fm$^2$ for the 6.2 keV transition in $^{181}$Ta, we obtain $\Delta\langle r^2 \rangle/\beta = -0.18 \times 10^{-4}$ for the 100 keV transition in $^{182}$W.

The measurements with the 93 keV transition in $^{178}$Hf are still being continued. So far,
Isomer shifts have been measured for Ni, Pd, and Pt host lattices. Using these data and a similar procedure for the electron density calibration as for $^{182}$W, we obtain $\Delta <r^2>/<r^2> = +0.15 \cdot 10^{-4}$ for the 93 keV transition in $^{178}$Hf. The errors of the $\Delta <r^2>/<r^2>$ values originating from the errors of the isomer shift results are about 10% for the $^{182}$W and 20% for $^{178}$Hf. The uncertainties in the estimate of the electron-density ratio between Hf and Ta or W and Ta, as well as in the error of the $\Delta <r^2>$-value for $^{181}$Ta, however, are difficult to estimate and probably much larger.

The result for $^{182}$W is in good agreement with the value of $\Delta <r^2>/<r^2> = -0.16 \cdot 10^{-4}$ that has been obtained (9) independently from measurements in tungsten compounds. For $^{178}$Hf, however, the present value even differs in the sign from the value of $\Delta <r^2>/<r^2> = -1.3 \cdot 10^{-5}$ recently obtained by Boolchand et al. (10) from measurements with Hf metal and Hf(C$_5$H$_5$)$_2$ absorbers.

The calculations of Meyer and Speth (1), who adapted the self-consistent cranking model for the calculation of nuclear charge radii, yield values of $\Delta <r^2>/<r^2> = +0.11 \cdot 10^{-4}$ and $+0.07 \cdot 10^{-4}$ for $^{178}$Hf and $^{182}$W, respectively. For $^{178}$Hf the calculations agree with the experimental value. For $^{182}$W the experimental value and the calculated one differ in the sign. This disagreement is not astonishing since, particularly in cases where $\Delta <r^2>$ is very small, the calculations sensitively depend on the position of individual single-particle states near the nuclear fermi level. Thus, in its present state of refinement (1), the self-consistent cranking model can explain the order of magnitude and the occurrence of negative $\Delta <r^2>$ values, but does not seem to be capable of reliably predicting the sign of $\Delta <r^2>$ in cases where the magnitude of $\Delta <r^2>/<r^2>$ is of the order of a few times $10^{-5}$.

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An analysis of the experimental data on the $0^+$ states and E0 transitions over the range $A = 150-190$ is performed. It is pointed out that the experimental results are inadequate and inaccurate for a detailed comparison with the present theoretical treatments. Lack of a unified description of the varied $0^+$ excitations in the transitional and heavily deformed nuclei is indicated. The E0 component in the $2^+ \rightarrow 2^+$ in $^{192}$Pt is investigated by measuring $\phi_{4\gamma}$ correlation, the absolute and relative internal conversion coefficients. The values obtained are $\phi(E0/\beta_2) = +0.04 \pm 0.05$ with $A = -4.5 \pm 3.5$. The experimental $\phi(E0) = +0.004 \pm 0.005$ agrees with the value predicted by Kumar-Baranger, $\phi(E0, 2^+ \rightarrow 2^+) = +0.006$. The nature of the $2^+$ level in $^{192}$Pt is discussed.

E0 transitions are considered to be one of the most interesting kinds of de-excitation of excited nuclear states. The probability of such transitions is related to the change in the r.m.s. nuclear radius between the initial and final states at the quantum transition moment. The nuclear matrix element $\phi(E0)$ is determined exclusively by wave functions of the nucleus. Studies of monopole transitions make it possible to obtain important information on the details of the nuclear structure. The presence or lack of an E0 transition, the magnitude and sign of the nuclear matrix element enable to judge of the nature of the level under investigation. A comprehensive review of the experimental data on the $0^+$ states and E0 transitions in atomic nuclei is presented in ref. 2/.

Most of the experimental data over the range $A < 170$ have been obtained from measurements of the internal conversion coefficient and the region of $A \sim 150$ and $A \sim 190$ was investigated using angular correlations of conversion electrons. All the E0 transitions investigated occur between the positive parity integral spin levels in even-even nuclei. In all cases known to date, the experimental values of $\phi(E0)$ are much less than the single-particle estimate $\phi(E0) = 1$. The greatest values of $\phi(E0) = 0.2 \pm 0.4$ hold for the transitions from the lower excited states close to $\beta$-vibrational levels in nature. However, the experimental data available are inadequate and inaccurate and a higher accuracy of measurements of $\phi(E0)$ is required to check on the model representations of the nuclear monopole excitations. Angular correlation measurements of conversion electrons are particularly important because both the magnitude and sign of $\phi(E0)$ can be determined in this experiment. Of particular interest are the E0 transitions between non-integral spin or negative parity levels. The only E0 transition of this type known with assurance is that between the $5/2^+ \rightarrow 5/2^+$ (124 keV) in $^{132}$Os /2/.

Comparison between the experimental data on the $0^+$ levels and the probability of E0 transitions shows that at present there is no unified satisfactory description of the varied $0^+$ excitations over the range $A = 150-190$. One can hardly hope to describe the E0 transitions within the framework of the phenomenological models, the probability of an E0 transition being dependent on the particulars of the nuclear structure. More accurate calculations within the framework of the microscopic approach are required.

The author investigated the E0 component in the $2^+ \rightarrow 2^+$ (296 keV) transition in $^{192}$Pt. Measurements were made of the angular correlation of K- and L- conversion electrons using a spectrometer with semiconductor Ge(Li) and Si(Li) detectors as well as of the absolute and relative I090 in the $^{192}$Ir $\rightarrow 192$Pt transitions and values of $\phi_{4\gamma}$ were determined.

Use of the $\phi_{4\gamma}$-angular correlation data for an analysis of the E0 component within the transition under study enabled to eliminate one of the two experimentally observed ranges
of the amplitude ratio of $E0$ and $E2$ conversion transitions, $\varphi(E0/E2)$, and penetration parameter $\lambda$ of the $M1$ transition (see the accompanying figure). The result obtained is $\varphi = +0.04 \pm 0.05$ with $\lambda = -4.5 \pm 3.5$. The reduced nuclear matrix element $\varphi(E0) = +0.004 \pm 0.005$ agrees with the theoretical value $\varphi(E0, 2^+_2 \rightarrow 2^+_1) = +0.006$ and is inconsistent with $\varphi(E0, 2^+_2 \rightarrow 2^+_1) = -0.089$ calculated as a semi-microscopic approximation by Kumar and Baranger/4/. The experimental value of $\varphi(E0, 2^+_2 \rightarrow 2^+_1)$ may suggest that the 612 keV level in $^{192}$Pt from which the 296 keV transition occurs, is two-phonon or gamma-vibrational and that the admixture of other states at the expense of which the $E0$ transition is possible in this level is small. Comparison between the experimental value of $\chi(E0/E2) = (1.3 \pm 1.4)$ and the calculated $\lambda$ ones is a qualitative support of this conclusion. The nuclear matrix element $\varphi(E0, 2^+_2 \rightarrow 2^+_1)$ for $^{192, 194, 196}$Pt isotopes increases with $\lambda$ which points to the fact that the admixture of other states in the $2^+_2$ level is greater for the nuclei more remote from the region of deformation.


Fig. The $\chi - \lambda$ plane for the 296 keV $K$-conversion transition in $^{192}$Pt.
The magnetic moment of the $^1_-$ state at 397 keV ($\tau = 6.5 \pm 0.5$ ps) in $^{16}\text{N}$ has been measured using the time differential recoil into vacuum technique employed in a similar measurement on $^{16}\text{O}$ (1).

This state was populated through a resonance at 37.5 MeV in the reaction $d(\text{He},p)^{16}\text{N}$ and its decay was followed by observing the 276 keV transition to the $^0_-$ state. The $^{16}\text{N}$ recoil ions were stopped in a Ni foil and the $\alpha$-particles to the low-lying states in $^{16}\text{N}$ were detected at $^0_-$ by a silicon surface barrier detector. Gamma-rays in coincidence with these $\alpha$-particles were detected also at $^0_-$ in a Ge(Li) detector. The $^{16}\text{N}$ ion recoil velocity was determined from the energy separation of the stopped and Doppler-shifted 276 keV $\gamma$-rays, giving the result $\gamma/C = 0.0455 \pm 0.0010$. At this velocity one expects approximately 45% one-electron ions and 45% two-electron ions. (2) Previous experiments have shown that such one-electron ions are predominantly in Is atomic ground states (1), producing a hyperfine field of 57.5 MHz, and that the effects of two-electron systems at these recoil velocities are negligible in the determination of the $g$-factor.

The coincidence yield of 276 keV $\gamma$-rays as a function of time of flight is shown in the figure, corresponding to flight paths of between 20 cm and 60 cm in target-stopper separation. These data were fitted with a function of the form:

$$I(t) = A(1+q-e^{-t/\tau})+Be^{-t/\tau} \cos \omega t - \frac{1+q}{1+(\omega t)^2}$$

with

$$1+q = (\frac{1+\beta}{1-\cos \theta_c}) \left(\frac{\epsilon_f}{\epsilon_0}\right)$$

where $\beta = \gamma/C$

$\theta_c$ = half-angle subtended by $\gamma$-detector in lab.

$\epsilon_f/\epsilon_0$ = photopeak efficiency ratio for flight to stopped peak in $\gamma$-detector.

The first term in the above product is an approximation to the relativistic correction to the effective solid angle of the $\gamma$-detector for decays from nuclei in flight. (4) $A$ and $B$ are constants determined by the percentage of one electron ground state systems and the unperturbed angular correlation coefficient ($A_2 = +0.28 \pm 0.03$).

This fit gave a hyperfine precession frequency $\gamma = 1.44 \pm 0.10$ rad/ps, resulting in $|g| = 1.74 \pm 0.12$ for the 397 keV state in $^{16}\text{N}$. The lifetime used in the above expression was $\tau = 6.5 \pm 0.5$ ps as determined by a recoil-distance analysis of the data.

If this state arises from a $p_{1/2}$ proton hole and an $s_{1/2}$ neutron, its $g$-factor, whether determined from the Schmidt limit or empirically from neighbouring odd-A nuclei , is -2.2. In order

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**Figure**

Time differential $\alpha$-$\gamma$ coincidence yield for the decay of the $^1_-$ state in $^{16}\text{N}$ ions recoiling into vacuum.
to bring this down nearer to the experimental value, a more extensive calculation is required. A recent
calculation using 2-particle – 2-hole and 3-particle – 3-hole components in the wavefunctions predicts
g(1) = -1.67 and also \(\tau(1') = 6.95\) ps., in good agreement with experiment.

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Recent investigations have shown that the HFI of the 3⁻ state in $^{16}$O ($\tau \approx 26$ ps) in isolated ions of low charge ($q \approx 4^+$) is dominated by electrons in the 2s subshell. Indeed, these data are consistent with the assumption that the K shell is closed in such ions and that the other electrons are statistically distributed in the n=2 shell.

We report here a time-integral PAC measurement on the $^{16}$N(1⁻) state in similar ions, performed with the aid of the $^{14}$C($^3$He,p) reaction at $E_{^3$He} = 2 MeV. The 276 keV gamma rays from the 1⁻→0⁻ transition were registered by 25 cc Ge(Li) counters in coincidence with protons detected at backward angles.

As the observed attenuation is rather small, a careful comparison of anisotropies on recoil into vacuum and gold was carried out, varying the bombarding energy over a 200 keV range. This yielded for the attenuation coefficient a value of:

$$G_2(\omega) = 0.915(15)$$

With the nuclear lifetime of $\tau = 10(2)$ ps (as recently measured using the recoil-distance method), and invoking the above-mentioned HFI mechanism, one obtains a lower limit for the g-factor (within one standard deviation limits on $G_2$ and $\tau$) of:

$$|g| > 1.7$$

In order to impose a reliable upper limit, more sensitive experiments are needed. This is due to the fact that one cannot rule out the possibility of a certain fraction of the ions being formed in excited electronic configurations (for which the HF fields are, in general, considerably smaller). However, the quoted lower limit is itself rather close to the Schmidt value of:

$$g = -2.2$$

for a $(\gamma P_{16} P_{16}^{-1})$ shell-model configuration.

![Diagram](image_url)

Time-integral attenuation coefficient plotted against nuclear g-factor and lifetime. The curves represent theoretical values for the HFI mechanism mentioned in the text. They are calculated for nuclear spin $I=1$, using HF field values from a spin-unrestricted Hartree-Fock calculation and empirical charge state probabilities ($^{16}$N recoil velocity was $\approx 0.015c$).

References

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Recent investigations \cite{1,2} of highly ionized light atoms have shown that the HFI for short-lived nuclear states ($\tau \approx 10^{-11}$ sec.) are predominantly associated with the single-electron systems in their ground configuration. Due to this simple general feature, it was possible in a previous measurement (performed on $7^+$ oxygen ions using the time-integral PAC method) to impose certain experimental limits on the $g$-factor in question ($0.2 < |g| < 0.36$) \cite{3}.

We report here a PAC measurement in which the HFI in such ions was decoupled by means of an external magnetic field. $^{18}$O ions of $\approx 45$ MeV from the Köln FN Tandem accelerator were inelastically scattered off a target of $^{4}$He gas. The 1.98 MeV gamma rays were registered in coincidence with forward-recoiling alpha particles. The magnetic field was applied along the beam direction (which coincides with the symmetry axis of nuclear alignment) with the aid of a 700 KG superconductive coil. By measuring the angular correlation as function of the external field strength (up to values corresponding essentially to Paschen-Back coupling), the $g$-factor was determined as:

$$|g| = 0.25 \pm 0.02$$

This technique has the merit of being sensitive to the HF fields associated with the atomic configurations involved, but not to their occupation probabilities in the ionic ensemble. The influence of subsequent collisions of the scattered ions with gas atoms during the nuclear lifetime is discussed in another contribution.
III. 6 THE MAGNETIC MOMENT OF THE 4+ 3.55 MeV LEVEL OF 18O

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Recent measurements (1,2) have shown the g-factor of the first 2+ 1.98 MeV level of 18O to be:

\[ |g| = 0.26 \pm 0.05 \]

This value deviates strongly from the value \( g = -0.76 \) for a pure \( \left( \frac{d_3/2}{s_1/2} \right)^2 \) configuration, suggesting admixtures of \( d_3/2 \) and \( d_3/2 \) \( s_1/2 \) configurations. In view of this, the present measurement for the 4+ 3.61 MeV level was initiated to obtain more information on the configuration mixing governing the structure of the low levels of 18O. The 4+ level is well suited for this purpose as the possible s-d shell admixtures reduce in this case to the \( d_3/2 d_3/2 \) configuration.

The magnetic moment was measured by means of the recoil-into-gas (\( H_2 \)) PAC technique, and comparison with a calibration measurement - the 3+ 6.13 MeV level of 16O. The 4+ level was populated via the

\[ ^{18}O(p,p')^{18}O \]

reaction at \( E_p = 6.0 \) MeV. The target was a self-supporting 50 \( \mu \)g/cm\(^2\) Si 18O foil. \( \gamma \)-rays were detected by two 5"x5" NaI crystals situated at 45° and 90° in coincidence with protons scattered into an annular surface barrier detector subtending 160° < \( \theta < 175° \). The 500 \( \mu \)g/cm\(^2\) Ni gas foil was placed behind the annular particle counter rather than in front as was the case in previous measurements thus eliminating the problem of intense elastic scattering from the gas foil into the particle detector. Under these experimental conditions, the 4+ 3.55 MeV level was not completely resolved from the 0+ 3.63 MeV level.

As the cross sections for populating the 4+ and 0+ levels, as well as the angular correlation of the \( \gamma \)-rays from the 4+ might depend on bombarding energy, the proton beam energy was adjusted at each pressure to compensate for energy loss in the \( H_2 \) gas so as to maintain a constant energy on target.

The 3+ level of 16O at 6.13 MeV excitation was chosen for comparison and calibration. This level has a known g factor (refs. 3, 4) and a mean life \( \tau = 27 \) ps (ref. 3) which is close to the mean life of the 4+ 3.55 level of 18O: \( \tau = 26 \) ps (ref. 5). A measurement similar to the above was performed for the 3+ level. The level was excited in the

\[ ^{19}F(p,a)\ ^{16}O \]

reaction at the \( E_p = 1.375 \) resonance in the experimental arrangement described in ref. 3. In both the 16O and the 18O measurements the recoil velocity was close to 0.01 c.

An extensive study of the HFI (ref. 3) elucidated in some detail the nature of the ionic environment that prevails in these measurements, allowing a direct estimation of the g factor of the 4+ level from the 16O measurements alone. The close comparison with the 3+ level reduces to a large extent the dependence of this evaluation on the atomic parameters and considerably enhances the reliability and accuracy of the measurement.

Fig. 1 shows the ratio of 16O gamma counts at 45° to counts at 90° as a function of \( H_2 \) gas pressure for the sum of the 4+ and 0+ levels. The relative intensity of the 4+ and 0+ level excitations was determined by observing the \( \gamma \)-rays in a 50 c.c. Ge-Li detector and independently by a gaussian fitting procedure applied to the partially resolved peaks in the particle spectrum. The relative intensity was found to be:

\[ N(4^+)/N(0^+) = 1.97 \pm 0.03 \]

The experimental results were corrected for the isotropic contribution of the 0+ level using this measured number. The unperturbed angular correlation was measured (at \( H_2 \) pressure of 0.8 At.) at three angles 22°, 45°, 90° and was found to be:

\[ A_2/A_0 = 0.56 \pm 0.02 \]
\[ A_4/A_0 = -0.360 \pm 0.02 \]

which is fully consistent with a pure \( m=0 \) substate population. The g-factor was deduced by evaluating the time integral perturbation using the electronic ensemble of Ref. 3 and by an Abragam-Pound procedure using the 16O.

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III. 6 THE MAGNETIC MOMENT OF THE 4+ 3.55 MeV LEVEL OF $^{18}_0$


Department of Nuclear Physics, The Weizmann Institute of Science, Rehovot, Israel

Recent measurements (1,2) have shown the g-factor of the first 2+ 1.98 MeV level of $^{18}_0$ to be:

$$|g|=0.26±0.05.$$  This value deviates strongly from the value

$$g ≈ -0.76$$  for a pure $(d_{5/2})^2$ configuration, suggesting admixtures of $d_{5/2}$ $d_{3/2}$ and $d_{5/2}$ $s_{1/2}$ configurations. In view of this, the present measurement for the 4+ 3.61 MeV level was initiated to obtain more information on the configuration mixing governing the structure of the low levels of $^{18}_0$. The 4+ level is well suited for this purpose as the possible s-d shell admixtures reduce in this case to the $d_{5/2}$ $d_{3/2}$ configuration.

The magnetic moment was measured by means of the recoil-into-gas ($H_2$) PAC technique, and comparison with a calibration measurement - the 3+ 6.13 MeV level of $^{16}_0$. The 4+ level was populated via the $^{18}_0(p,p')^{18}_0$ reaction at $E_p = 8.0$ MeV. The target was a self-supporting SO ug/cm$^2$ Si $^{18}_0$ foil. $\gamma$-rays were detected by two 5"x5" NaI crystals situated at 45° and 90° in coincidence with protons scattered into an annular surface barrier detector subtending $160°<_\theta<175°$. The 500 ug/cm$^2$ Ni gas foil was placed behind the annular particle counter rather than in front as was the case in previous measurements thus eliminating the problem of intense elastic scattering from the gas foil into the particle detector. Under these experimental conditions, the 4+ 3.55 MeV level was not completely resolved from the 0+ 3.63 MeV level.

As the cross sections for populating the 4+ and 0+ levels, as well as the angular correlation of the $\gamma$-rays from the 4+ might depend on bombarding energy, the proton beam energy was adjusted at each pressure to compensate for energy loss in the $H_2$ gas so as to maintain a constant energy on target.

The 3+ level of $^{16}_0$ at 6.13 MeV excitation was chosen for comparison and calibration. This level has a known g factor (refs. 3, 4) and a mean life $\tau=27$ ps (ref. 3) which is close to the mean life of the 4+ 3.55 level of $^{18}_0$: $\tau=26$ ps (ref. 5). A measurement similar to the above was performed for the 3+ level. The level was excited in the $^{19}_F(p,\alpha)^{16}_0$ reaction at the $E_p = 1.375$ resonance in the experimental arrangement described in ref. 5. In both the $^{16}_0$ and the $^{18}_0$ measurements the recoil velocity was close to 0.01 c.

An extensive study of the HFI (ref. 3) elucidated in some detail the nature of the ionic environment that prevails in these measurements, allowing a direct estimation of the g factor of the 4+ level from the $^{18}_0$ measurements alone. The close comparison with the 3+ level reduces to a large extent the dependence of this evaluation on the atomic parameters and considerably enhances the reliability and accuracy of the measurement.

Fig. 1 shows the ratio of $^{18}_0$ gamma counts at 45° to counts at 90° as a function of $H_2$ gas pressure for the sum of the 4+ and 0+ levels. The relative intensity of the 4+ and 0+ level excitations was determined by observing the $\gamma$-rays in a 50 c.c. Ge-Li detector and independently by a gaussian fitting procedure applied to the partially resolved peaks in the particle spectrum. The relative intensity was found to be:

$$N(4+)/N(0^+)=1.97±.03$$

The experimental results were corrected for the isotropic contribution of the 0+ level using this measured number. The unperturbed angular correlation was measured (at $H_2$ pressure of 0.8 At.) at three angles 22°, 45°, 90° and was found to be:

$$A_2/A_0=0.56±.02$$  $$A_4/A_0=-0.360±.02$$

which is fully consistent with a pure $m=0$ substate population. The g-factor was deduced by evaluating the time integral perturbation using the electronic ensemble of Ref. 5 and by an Abragam-Pound procedure using the $^{16}_0$

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data as a calibration (Fig. 2). These two procedures respectively yield: 

\[ |g| = 0.62 \pm 0.12 \quad \text{and} \quad |g| = 0.63 \pm 0.15 \]

The wave function for the \(4^+\) level can be written in the framework of the s-d shell as:

\[ \psi(4^+) = (d_{5/2}^2 + \alpha d_{3/2}^2 3/2) (1 + \alpha^2)^{-1}. \]

Excitations of the core are negligible for the \(2^+\) and \(4^+\) levels as has been shown in the weak coupling model calculation of Ellis and Engeland (Ref. 6).

The measured g-factor yields \(\alpha = 0.2 \pm 0.13\) indicating very little admixture of the \(d_{5/2}^2 3/2\) configuration in the \(4^+\) wave function and is in good agreement with the value of \(\alpha = 0.36\) obtained by Ellis and Engeland.

The authors wish to thank Professor I. Talmi for many helpful discussions.

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2) M.E. Goldberg, private communication.

Fig. 1. The ratio \(N(45^0)/N(90^0)\) as a function of \(H_2\) gas pressure for \(\gamma\) rays de-exciting the \(4^+\) and \(0^+\) levels of \(^{18}O\). The solid line represents the best fit obtained with an Abragam-Pound procedure.

Fig. 2. The ratio \(N(0^0)/N(90^0)\) as a function of \(H_2\) gas pressure for \(\gamma\) rays de-exciting the \(3^+\) level of \(^{16}O\). The solid line represents the best fit obtained with an Abragam-Pound procedure.
THE MAGNETIC MOMENT OF THE 2.211 MEV STATE IN $^{22}\text{Na}$ BY HYPERFINE INTERACTION

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Centre de Recherches Nucléaires de Strasbourg - France

A time differential technique has been employed to observe the time dependence of an angular correlation perturbed by hyperfine interactions. The measurement was performed on the transition between the $J^\pi = 1^-$, 2.211 MeV and the $J^\pi = 0^+$, 0.657 MeV states in $^{22}\text{Na}$, on excited nuclei recoiling into vacuum with a velocity $v/c = 0.0092$.

The present measurement follows in technique, as well as in the intermediate ionization approach, that was applied by the Rehovot group. Under this assumption the perturbation is mainly caused by the contact field of an unpaired 2s electron, a field that is modified by the presence of a few p electrons. In the present case, the total contribution of the 2p electrons can be neglected for the first 60 ps. The measurement in $^{16}\text{O}$ suggests the action of static configurations, resembling "cold" ionization. However, unlike the extreme and simple case of a hydrogenic atom with $J = 1/2$, here a manifold of terms with different $J$-values produces a complex series of attenuation frequencies of the angular correlation. The properly weighted contributions, when summed up, finally result in theoretical attenuation coefficients $G_K(t)$ which are parametrized by values of the nuclear magnetic moment. (There is some evidence, that a simple attenuation picture, suggesting the action of a static field, has been observed in conditions suitable for "hot" ionization.)

The $J^\pi = 1^-$ state in $^{22}\text{Na}$ was populated by $\alpha$-particles in the $^{19}\text{F}(\alpha,n)$ reaction near the threshold at $E_\alpha = 5,170$ MeV ($E_{\text{th}} = 5,036$ MeV). The angular correlation of the 1.554 MeV $E1$ $\gamma$-ray transition is of the form $W(\theta, t) = 1 + A_2 Q_2 Q_2(t) P_2(\cos\theta)$. Two Ge(Li) detectors at 125° and 0° are located at $P_2 = 0$ and $P_2 = 1$, respectively. This choice provides normalization as well as sensitivity, the detector at $\theta = 0$ simultaneously providing an additional overall check by remeasuring the lifetime of the $E_x = 2.211$ MeV state.

The temporal behavior was analyzed by means of a plunger arrangement. A 60 $\mu$g/cm$^2$ CaF$_2$ target was deposited on a 1 mg/cm$^2$ gold foil, the stopper being a 3 mg/cm$^2$ gold foil stretched on a special holder. The maximum Doppler shift, which amounts to 15 keV, is enough to allow separation between the stopped and shifted peaks. If $I_0(\theta, t)$ is the stopped $\gamma$-intensity measured at $\theta$ and corresponding to a plunger separation $d = vt$, we define

$$ R(t) = \frac{I_0(0^\circ, t)}{I_0(125^\circ, t)} $$

$G_2(t)$ can now be expressed as

$$ G_2(t) = \left[ \frac{R(t)}{R(0)} - 1 \right] \left[ \frac{1 + A_2 Q_2}{A_2 Q_2} \right] + 1 $$

The value of $A_2 Q_2$ was determined from $W(\theta, 0)$ by reversing the target and backing to be $A_2 Q_2 = 0.78 \pm 0.01$.

The calculation of $G_2(t)$ follows the lines given in ref. 1. The charge state fractions $f_K$ and the associated contact fields $H_K(0)$ are tabulated in Table 1.

<table>
<thead>
<tr>
<th>Charge State</th>
<th>1$^+$</th>
<th>2$^+$</th>
<th>3$^+$</th>
<th>4$^+$</th>
<th>5$^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_K$</td>
<td>0.06</td>
<td>0.27</td>
<td>0.40</td>
<td>0.24</td>
<td>0.03</td>
</tr>
<tr>
<td>$H_K(0), \text{MG}$</td>
<td>-</td>
<td>9.4</td>
<td>9.7</td>
<td>10.0</td>
<td>10.4</td>
</tr>
</tbody>
</table>

Table 1
Fig. 1 presents calculated $G_2(t)$ functions for three values of the nuclear g-factor, as well as the measured time spectrum. From a $\chi^2$ analysis, the g-factor of the $E_\gamma = 2.211$ MeV state in $^{22}$Na, deduced under the above assumptions, is determined to be $g = 0.49 \pm 0.04$. The mean life of the same level was simultaneously measured to be $\tau = 22.0 \pm 0.8$ ps.

Refined calculations and the present calculation in detail are expected to be reported in the near future.

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III. TRANSIENT FIELD MEASUREMENTS OF $g$-FACTORS FOR $^{24}\text{Mg}$ AND $^{26}\text{Mg}$

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The $g$-factors of the first excited $J^p = 2^+$ states of $^{24}\text{Mg}$ and $^{26}\text{Mg}$ have been measured with the ion-implantation perturbed angular correlation technique (IMPAC). The measured precession of the angular correlation on recoil into a magnetized Fe backing for such short-lived nuclear states ($\tau = \approx 1$ ps) in this mass region is caused predominantly by the transient magnetic field that occurs due to scattering of the polarized electrons during the slowing-down of the recoiling nucleus. For the Mg isotopes the transient field reaches a value of 200 T and yields small average precession angles of about 1.5 mrad.

The levels were excited by the $\text{Mg}(\alpha,\gamma)$ reaction with $\alpha$-particles of about 7.5 MeV energy from the Utrecht tandem accelerator. Targets of 150 $\mu$g cm$^{-2}$ of isotopically pure $^{24}\text{Mg}$ and $^{26}\text{Mg}$ evaporated onto 10 $\mu$g cm$^{-2}$ Fe foils were used for the precession experiment. The Fe foils were annealed in a $N_2$ atmosphere and the magnetization as function of the external magnetic induction was measured. The Fe was found to be saturated for a magnetic induction $B > 0.07$ T with an average value for the number of polarized electrons per atom $\xi = 2.02(8)$ (10% lower than the full saturation value of $\xi = 2.2$). Beam bending effects, due to the magnet fringing field, were measured with Cu-backed targets. The targets were maintained in a regularly reversed external field of 0.15 T. Four 12.7 cm x 12.7 cm NaI(Tl) crystals at angles $\pm 72^\circ$ and $\pm 108^\circ$ were used to detect $\gamma$-rays in coincidence with the detection of outgoing $\alpha$-particles at $\theta_\alpha = 180^\circ$. Since the measured precession angles were very small, special care was taken, especially in the electronics net-up and in the statistical analysis, to minimize and to check for possible systematic errors.

The mean precession angle $\delta \theta$ in terms of the relative change of $\gamma$-ray yield $N$ measured at a certain angle $\theta$ is given by

$$\frac{\Delta N}{N} = \left| \frac{dW}{d\theta} \right| \delta \theta,$$

where $W(\theta) = k \cdot \theta^2 \cdot \phi \cdot \Phi(k \cdot \theta)$. The precession angle was determined from the ratio of counts $N_1$ and $N_2$ accumulated in one pair of detectors with magnetic field up and down. The effect $c$ is defined by

$$c = \left( \frac{N_1}{N_2} \right) + \left( \frac{N_2}{N_1} \right) - 1 = \frac{4\Delta N}{N}.$$

The measured precession and beam bending effects are given in Table 1.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>detector pair</th>
<th>precession</th>
<th>beam bending</th>
<th>$\Delta \theta$ = $\Delta \theta_p - \Delta \theta_b$</th>
<th>$g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{24}\text{Mg}$</td>
<td>1,2</td>
<td>3.6(3)</td>
<td>2.0(2)</td>
<td>0.8(4)</td>
<td>0.4(2)</td>
</tr>
<tr>
<td></td>
<td>3,4</td>
<td>3.2(3)</td>
<td>1.8(2)</td>
<td>1.1(4)</td>
<td>0.6(2)</td>
</tr>
<tr>
<td>$^{26}\text{Mg}$</td>
<td>1,2</td>
<td>4.2(5)</td>
<td>2.4(3)</td>
<td>1.0(5)</td>
<td>0.6(2)</td>
</tr>
<tr>
<td></td>
<td>3,4</td>
<td>4.8(5)</td>
<td>2.7(3)</td>
<td>1.8(5)</td>
<td>1.0(3)</td>
</tr>
</tbody>
</table>

To extract $g$-factors from the observed precessions the Lindhard-Winther theory is applied as follows.

1) Correction for decay in flight leads to an average precession $\Delta \theta$ due to both transient and static fields

$$\Delta \theta = \frac{1}{\tau} \int_0^\tau \left( \tau t \right) e^{-t/\tau} \frac{d}{dt} \sum_i \frac{p_i^2}{m_i} \tau e^{-t/\tau} dt,$$

where $\tau$ is the mean lifetime of the state.

$$\Delta \theta = \frac{1}{\tau} \int_0^\tau \left( \tau t \right) e^{-t/\tau} \frac{d}{dt} \sum_i \frac{p_i^2}{m_i} \tau e^{-t/\tau} dt,$$

(3)
where \( \tau_m \) and \( \tau_e \) denote the mean life and stopping time, respectively, of the recoiling nuclei, \( \nu_s \) is the Larmor frequency due to the static hyperfine field and \( \phi(t) \) is the transient field precession.

ii) The predictions of the theory are about a factor of two lower than the experimental values, but the general trends of experiment (the velocity\(^2\) and nuclear charge dependence\(^3\)) are well described. To obtain quantitative agreement, the polarized electron velocity \( v_p \) was fitted to the experimental results in the higher \( Z \) region. The experimental points and the fitted curve are shown in Fig. 1. The value of \( v_p \) obtained this way \([0.40(8)]\) is now no longer a parameter with a strictly physical meaning.

In the analysis, corrections for finite target thickness were applied. In the error calculus all possible sources of errors have been included, the most important being the statistical error \([14\%]\), the uncertainty in \( v_p \) \([20\%]\), the error in the stopping power \([20\%]\) and uncertainties in target thickness \([30\%]\) and lifetime \([5-10\%]\). The values for the g-factors are displayed in Table 1. The result for \( ^{24}\)Mg is in excellent agreement with a recent measurement\(^4\) using the time-differential recoil-into-vacuum technique.

This value can be used to obtain a calibration of the transient field precession for Mg. This calibration point is inserted in Fig. 1 and falls on the line fitted to the higher values (the error is smaller than the size of the drawn circle).

References

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3) J.L. Eberhardt, R.E. Horstman, H.W. Heeman and G. van Middelkoop, Nucl. Phys. to be published
The g-factors of excited states in $^{41}K$ and $^{51}Cr$ were found from differential perturbed angular correlations (DPAC) in an external magnetic field. The measurements were made in an external cyclotron beam. Since in the case of question the $\gamma$-ray angular distribution anisotropies were small, measurements were taken to avoid distortions due to the beam structure.

The 1290 keV state in $^{41}K$ was excited by inelastic scattering of 6.8 MeV protons. The potassium target enriched to 90% of $^{41}K$ was obtained by evaporation onto the platinum backing. The results of the measurements in an magnetic field of $21.24\pm0.16\text{ kOe}$ are given in Fig.1a.

The field strength was determined from the simultaneous measurements of precession frequency of the $^{197}$Hg nucleus in the 197 keV excited state ($g=1.442\pm0.003$). By the least squares method a value for $g=1.290\pm0.034$ was found, from which $g=1.290\pm0.034$. This is in good agreement with $\frac{1}{2}$. The shell model calculations with a realistic interaction $\sqrt{2}$ and with effective nucleon g-factors calculated in the frame of the finite $\sqrt{3}$ Fermi-systems give a very close value $g=1.275$.

To excite the 760 keV state of $^{51}Cr$, reactions $^{48}Ti(n,\gamma)^{51}Cr$ and $^{50}Cr(d,p)^{51}Cr$ at 27 MeV energy were used. The field strength was $29.20\pm0.18\text{ kOe}$. The results are shown in Fig.1b and 1c, correspondingly. The precession frequencies obtained were $(7.26\pm1.25)\times10^{7}$ and $(8.85\pm0.90)\times10^{7} \text{ rad/sec}$. The targets were titanium metal and chromium oxide, the latter being antiferromagnetic up to 307°C. The chromium atoms have a small intrinsic magnetic moment of the order $0.4\mu_B$ (polymetallic chromium is an antiferromagnetic up to $-360°C$) but due to their being surrounded by the titanium atoms, both their magnetic moment and point Neel are essentially lowered. A slight difference in precession frequencies might be attributed to a weak paramagnetism of chromium atoms knocked out in the reaction from the chromium oxide target at 320°C.

An average of two precession frequencies gives $g=0.57\pm0.06$. The sign is negative.

A theoretical explanation of the g-factors for nuclei with unfilled $1p_{3/2}$ shell is of interest because in this case deviations from the Schmidt line are particularly large. For $^{51}Cr$ one obtains from the shell model with configuration mixing $g=0.59$. However this agreement does not seem satisfactory because the $3/2^-$ state seems to have a more complicated structure. Both the one-particle level $2p_{3/2}$ and the $3/2^-$ level obtained due to
the nucleon rearrangement in the $127/2$ shell $1/2$ are lying much higher than the observed level, while their mixing results in a decrease of the excitation energy.

The g-factor of the $^{197}$Hg 133 kev state was measured by the conversion electron -- $\gamma$-quantum DPAC at the $^{197}$Hg isomer decay in the hyperfine field on Hg nuclei inside the Ni matrix. The source was made by an evaporation of $^{197}$Hg atoms, built up in a gold foil by the $^{197}$Au(d,2n)$^{197}$Hg reaction, onto a nickel backing upon which a little nickel was additionally deposited and the whole array annealed.

The differential angular correlation (Fig.2) in this case shows the frequency distribution, and the anisotropy is essentially less than that measured by the integral method. It is difficult to say whether this is due to the presence of several fields on Hg nuclei inside the Ni matrix (including B=0) because the method of source preparation itself allows the Hg atoms to fill the positions with $B < B_{\text{max}}$.

The precession frequency obtained is $(18.6\pm0.4)\times10^7$ rad/sec. Using for $B(\text{Hg in Ni})$ a value of $-86\pm22$ kOe $^{4/4}$ we obtain $\mu=1.12\pm0.25$ n.m. On the other hand, with the above value for $\omega$ and a g-factor measured by a DPAC of $\gamma$-rays in an external field $^{5/5}$ one obtains for $B(\text{Hg in Ni})$ a bigger value $102\pm9$ kGs.

References
The g-factor of the \( \frac{7}{2} \)\(^{6+} \) state in \(^{42}\text{Ca}\) plays an important role in the determination of the \( \frac{7}{2} \) neutron g-factor and the understanding of deformed wave function admixtures. Because of the existing experimental uncertainties, the g-factor of this 3189-keV \( 6^+ \) state in \(^{42}\text{Ca}\) has been remeasured. A significant improvement in accuracy has been achieved by the utilization of a large magnetic field, 60,940.4 kG, provided by an in-beam superconducting magnet. The g-factor measurements were carried out by time-differential NAA using the \(^{42}\text{Ca}(a,p)^{42}\text{Ca}\) reaction induced by a 15 MeV pulsed beam on a thick \( \text{KI} \) target and the \(^{40}\text{Ca}(a,2p)^{42}\text{Ca}\) reaction with a 23 kV pulsed a beam and a thick \( \text{Ca} \) target. Both NaI and Ge(Li) \( \gamma \) detectors positioned at \( \pm 45^\circ \) with respect to the effective beam direction were used to detect the 1228 keV \( (\frac{7}{2} \rightarrow \frac{5}{2}) \) \( \gamma \) ray. The ratio data \( R(t) = \frac{[N(-45^\circ)-N(+45^\circ)]/[N(-45^\circ)+N(+45^\circ)]}{[N(-45^\circ)+N(+45^\circ)]} \) for the \(^{42}\text{Ca}(a,p)^{42}\text{Ca}\) reaction are shown in Fig. 1 along with the least squares fits. The results for the \(^{42}\text{Ca}(a,2p)^{42}\text{Ca}\) reaction yielded Larmor frequencies in agreement with those in Fig. 1, although the uncertainties were larger because of an increased background and large neutron contributions near the prompt peaks. The magnetic field was calibrated with in-beam g-factor measurements of the \(^{210}\text{Po} \frac{7}{2}^+ \) state and the \(^{198} \frac{5}{2}^+ \) state along with additional flip-coil measurements. An initial set of data analyses including careful background considerations, yields a g-factor of \( g(\frac{7}{2}) = -0.50 \pm 0.02 \), a value very close to the Steidt value \( -0.51 \pm 0.03 \). This result agrees with agreement with previous integral measurement of Homura et al.\(^1\) of \( -0.52 \pm 0.03 \) but is in disagreement with the time-differential measurement of Homura et al.\(^2\) of \( -0.50 \pm 0.02 \).\(^\text{a} \). \(^\text{b}\)

Considerable theoretical interest has been generated over these previous g-factor measurements; the present theoretical situation of this problem has been recently summarized by T. Erikson.\(^3\) Using their wave function including the deformed \((3p-2h)\) components for the \(^{42}\text{Ca} \frac{7}{2}^+ \) state, the present result gives an effective g-factor of the \( \frac{7}{2} \) neutron of \( g(\frac{7}{2}) = -0.52 \pm 0.02 \), a value very close to the Schmidt value \( -0.546 \). If this wave function is correct, the present measurement indicates that the anomalous orbital g-factor of the \( \frac{7}{2} \) neutron is small. A comparison of the present g-factor results with that for the \(^{42}\text{Ca} \) ground state suggests that the deformed admixtures in this region are not completely understood. For example, a 5% decrease in the \((3p-2h)\) deformed component in \(^{42}\text{Ca}\) makes the two g-factor measurements compatible. The present experimental result for the \(^{42}\text{Ca} \frac{7}{2}^+ \) state thus points to a need for further theoretical examination of the effective moments and the nuclear structure of these \( \frac{7}{2} \) nuclei.

\(^a\)Supported in part by the National Science Foundation.
\(^\text{b}\)On leave from Tata Inst. of Fund. Research, Bombay, India.
\(^\text{c}\)Alfred P. Sloan Research Fellow.


Fig. 1
III. NEW MAGNETIC MOMENTS BY IN BEAM OPTICAL PUMPING

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The measurements of magnetic moments of short-lived β emitters by Optical Pumping (OP)[1-3] at the Karlsruhe Isochronous Cyclotron have been continued. The technique of OP in beam applies preferably to alkalies with half-lives between some 100 ms and some sec. The lower limit, which has been extensively studied in the cases of $^{20}$Na $(T_1/2 = 145$ ms)[2] and $^{36}$K $(T_1/2 = 340$ ms)[3], is due to the recombination of recoil ions produced by the nuclear reaction, and to the OP process. The upper limit arises from wall diffusion. Compared to this, the half-lives of Rb isotopes A 82 are rather long.

$^{80}$Rb $(T_1/2 = 30$ sec) was produced by proton impact on an enriched $^{80}$Kr target by the reaction $^{80}$Kr$(p,n)^{80}$Rb.

200 Torrs of He were added to the 20 Torr Kr target in order to increase the diffusion time. The $^{80}$Rb atoms were polarized by direct OP with D1 line of a Rb lamp. Fig. 1 shows the time dependence of the polarization detected by β-decay asymmetry after a short activation pulse. The asymmetry disappears at the time constant of wall diffusion. Thus, 5 sec after activation, all $^{80}$Rb atoms adhere to the walls, and decay unpolarized. hfs Zeeman transitions were detected by a destruction of the asymmetry. Fig. 2a shows the spin measurement, performed by irradiation the δF = 0, δF = ±1 transition frequencies corresponding to different possible spin values in weak magnetic field. At higher field (Fig. 2b) these frequencies are sensitive to the magnetic moment via the hfs separation δν. The results are I($^{80}$Rb) = 1, I($^{80}$Rb) = 234.0 (1.0) MHz which corresponds to $\mu_2^{\text{Rb}} = -0.0034(3)$ n.m. if the ratio δν/δμ is taken from the stable $^{87}$Rb. The unknown hfs anomaly may shift this value by at most 10%. The result yields the most probable shell model configuration for $^{80}$Rb, is $t_9^2(|t_5^2/2)^2$.

$^{82}$Rb $(T_1/2 = 75$ sec): In order to continue the series of Rb isotopes we have investigated $^{82}$Rb whose spin could be determined to be I = 1. The magnetic moment will allow to decide in the nuclear configuration.

$^{25}$Na $(T_1/2 = 60$ sec): This light nucleus was investigated by the spin exchange polarization method used previously for other Na and K isotopes. It was produced by a 10 MeV α beam in 700 Torr $^{22}$Ne target: $^{22}$Ne$(α,p)^{25}$Na. By the β decay asymmetry of 0.8% we detected Zeeman transitions, and thus confirmed the spin assignment I = 5/2. Measurements of the magnetic moment are in progress.

References:

2) J. Bonn, E.W. Otten, Ch. von Platen, W. Linder, H. Müller and H. Schweickert; Proc. Int. Conf. on the properties of nuclei far from the region of stability, Leyzyn, 1970
4) E.W. Otten; private communication
The nuclear structure of the Mn isotopes is well described by the shell model. The low-lying levels of $^{53}$Mn may be ascribed to the proton configuration $f_{7/2}^{-3}$ with a doubly magic $^{50}$Ni core. In $^{54}$Mn, the 29th neutron occupies the $p_{3/2}$, $f_{5/2}$ and $p_{1/2}$ shells. Excitations across the closed shell occur at low excitation energy.

$^{53}$Mn and $^{54}$Mn are excited in the $^{53,54}$Cr(p,n) reaction at 3 MeV bombarding energy. Further levels of higher spin in $^{54}$Mn are excited in the $^{51}$V(n,n) reaction at 8 MeV. We have used these reactions in alloy targets containing 80% iron to measure integral rotations for a number of short-lived excited levels. The hyperfine fields at Mn nuclei in these alloys are inferred from nuclear orientation and Mössbauer measurements to be -200 kOe and -160 kOe at 70 K and 300 K respectively. Gamma-ray angular distributions and their integral rotations were measured using Ge(Li) detectors at back angles. The results are shown in Table I.

The 377 keV Level of $^{53}$Mn:

The mean lifetime of this level is 169 ± 9 ps. We therefore conclude that the g factor is $g_{5/2} = 1.15 ± 0.17$. This appears to be somewhat smaller than the ground state g factor, $g_{7/2} = 1.44 ± 0.002$. If only the $f_{7/2}^{-3}$ proton configuration entered the wave functions of the two states, the g factors would be equal. An admixture in the $5/2^-$ level of the proton configuration $f_{7/2}^{-4}$ $f_{5/2}^{-5}$ would produce the observed effect. A similar ratio of $g_{5/2}$ to $g_{7/2}$ is found in the particle-hole conjugate nucleus $^{51}$V.

Levels of $^{54}$Mn:

No definitive lifetime measurements are available. The g factors indicated in Table II have been calculated from shell model wave functions for the levels and are relatively insensitive to the form of the residual interaction used. The wide variation in lifetimes thus deduced for the low-lying levels may be attributed to constructive and destructive interference between M1 transition matrix elements for seniority-1 and seniority-3 proton wave functions. The longevity of the 1072 keV level is consistent with the interpretation that it arises from the $f_{7/2}$ neutron hole and is therefore analogous to the ground state of $^{52}$Mn. In $^{55}$Fe, the $7/2^-$ hole state occurs at 1408 keV and decays with an M1 rate of $6 \times 10^{-3}$ W.u.

References:
3) A. W. Gibb, Private Communication
### Table I

<table>
<thead>
<tr>
<th>$E_n$ (keV)</th>
<th>$J_n$</th>
<th>$E_\gamma$ (keV)</th>
<th>$A_2$</th>
<th>$A_4$</th>
<th>$\omega T$ (mmrad)</th>
<th>$\tau$ (ps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{53}$Cr(p,n)$^{53}$Mn</td>
<td>377</td>
<td>$3/2^-$</td>
<td>377</td>
<td>-0.12(1)</td>
<td>-0.01(1)</td>
<td>193(28)</td>
</tr>
<tr>
<td>$^{54}$Cr(p,n)$^{54}$Mn</td>
<td>55</td>
<td>$2^+$</td>
<td>55</td>
<td>-0.08(2)</td>
<td>-0.02(3)</td>
<td>119(38)</td>
</tr>
<tr>
<td>156</td>
<td>$4^+$</td>
<td>156</td>
<td>-0.21(1)</td>
<td>-0.04(2)</td>
<td>328(57)</td>
<td>340(60)</td>
</tr>
<tr>
<td>408</td>
<td>$3^+$</td>
<td>251</td>
<td>-0.13(2)</td>
<td>-0.02(3)</td>
<td>-6(14)</td>
<td>&lt;10</td>
</tr>
<tr>
<td>$^{51}$V(a,n)$^{52}$Mn</td>
<td>368</td>
<td>$5^+$</td>
<td>211</td>
<td>-0.07(2)</td>
<td>-0.03(3)</td>
<td>57(34)</td>
</tr>
<tr>
<td>1072</td>
<td>$5^+$</td>
<td>705</td>
<td>-0.12(3)</td>
<td>-0.00(4)</td>
<td>117(60)</td>
<td>150(80)</td>
</tr>
</tbody>
</table>

### Table II

**Table II**

**Transition Rates in $^{54}$Mn**

| $E_\gamma$ (keV) | $J_{x}$ $\rightarrow$ $J_{y}$ | $\tau$ (ps) | $B$(M1) ($|v|^2$) | $T_\gamma$ (W.u.) |
|---|---|---|---|---|
| 55 | $2^+ \rightarrow 3^+$ | 1.8 | 70 | 4 | 2 |
| 156 | $4^+ \rightarrow 3^+$ | 0.9 | 400 | 0.04 | 0.025 |
| 211 | $5^+ \rightarrow 4^+$ | 0.7 | 100 | 0.06 | 0.04 |
| 251 | $3^+ \rightarrow 4^+$ | 1.2 | 10 | 0.3 | 0.2 |
| 705 | $6^+ \rightarrow 5^+$ | 0.5 | 300 | 0.001 | 0.0005 |
The integral perturbed angular correlation technique has been used to measure the g-factors of the 264 keV and 632 keV states in $^{77}$As. The measurement of the 264 keV level was performed in an external magnetic field of 30 kOe. The 632 keV state, having a shorter lifetime, was studied with the help of the large hyperfine magnetic field present at As nuclei in an Fe matrix.

The decay scheme of $^{77}$As, showing the relevant levels and transitions is presented in fig. 1. A conventional automated coincidence spectrometer with two NaI(Tl) detectors and a standard water cooled magnet have been used. The g-factor of the 264 keV ($5/2^-$) state was determined through the 367-264 keV gamma cascade. The radioactive $^{77}$Ge sources were produced by neutron irradiating 10 mg of GeO$_2$ enriched to 74% in $^{76}$Ge. Previous measurement of angular correlations with a liquid source$^1$ have shown that attenuations due to extranuclear interactions are negligible for the solid GeO$_2$ source.

Experimental values of $\omega \tau$, the spin precession angle in the magnetic field, were obtained from the ratio

$$R(\theta) = \frac{W(\theta,+H) - W(\theta,-H)}{W(\theta,+H) + W(\theta,-H)} = \frac{2b_4 \omega \tau}{1 + (2\omega \tau)^2}$$

measured at angles of 135° and 225° for three sources ($b_4 < b_2$). Values of $\omega \tau(225°) = (-2.36 \pm 0.60) \times 10^{-2}$ rad, $\omega \tau(135°) = (+2.45 \pm 0.54) \times 10^{-2}$ rad and $\omega \tau(225°) = (-2.93 \pm 0.51) \times 10^{-2}$ rad respectively were obtained and $\omega \tau = (2.25 \pm 0.32) \times 10^{-2}$ rad was deduced from the weighted average of the measurements. The lifetime of the 264 keV level in $^{77}$As has been recently reported by Chopra et al$^2$ to be $\tau = 500 \pm 30$ ps. Using a fast coincidence system with Pb loaded plastic scintillators we also measured the lifetime of this state. Our result (to be published elsewhere) is $\tau = 400 \pm 30$ ps.

The average value of $\omega \tau = (2.25 \pm 0.32) \times 10^{-2}$ rad with $H = 30$ kOe and $\tau = 450 \pm 30$ ps yields the g-factor of the 264 keV state: $g_{5/2}^- = +0.33 \pm 0.05$.

The g-factor of the short-lived 632 keV level was measured making use of the hyperfine field present at As nuclei in a dilute solid solution (1 at %) of Ge as impurity in Fe. The $^{77}$Ge activity in metallic form was deposited in a pot made of pure Fe. A tapered pin was driven in the pot under vacuum and the sealed pot was melted in an induction coil in presence of Argon gas. After melting, the alloy was annealed for 2 h at 800°C in Ar atmosphere and placed in a polarizing field of 5 kOe. The ratio R(8) was then measured at $\theta = 135°$ and 225°C for both states, 264 keV and 632 keV, in the same source through the 367-264 and 558-417 keV gamma sequences respectively.

The results are shown in table 1, giving average values of $\omega \tau(264$ keV) = $0.204 \pm 0.014$ rad and $\omega \tau(632$ keV) = $0.185 \pm 0.030$ rad, under the same hyperfine magnetic field. Using the known lifetimes (table 1) and the measured g-factor of the 264 keV state in an external field, the g-factor of the 632 keV state is obtained to be $g_{5/2}^- = +1.42 \pm 0.32$. 

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**III. 13**

**G-FACTORS OF THE 264 keV AND 632 keV STATES IN $^{77}$As**


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São Paulo, Brasil

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The decay scheme of $^{77}$As, showing the relevant levels and transitions is presented in fig. 1. A conventional automated coincidence spectrometer with two NaI(Tl) detectors and a standard water cooled magnet have been used. The g-factor of the 264 keV ($5/2^-$) state was determined through the 367-264 keV gamma cascade. The radioactive $^{77}$Ge sources were produced by neutron irradiating 10 mg of GeO$_2$ enriched to 74% in $^{76}$Ge. Previous measurement of angular correlations with a liquid source$^1$ have shown that attenuations due to extranuclear interactions are negligible for the solid GeO$_2$ source.

Experimental values of $\omega \tau$, the spin precession angle in the magnetic field, were obtained from the ratio

$$R(\theta) = \frac{W(\theta,+H) - W(\theta,-H)}{W(\theta,+H) + W(\theta,-H)} = \frac{2b_4 \omega \tau}{1 + (2\omega \tau)^2}$$

measured at angles of 135° and 225° for three sources ($b_4 < b_2$). Values of $\omega \tau(225°) = (-2.36 \pm 0.60) \times 10^{-2}$ rad, $\omega \tau(135°) = (+2.45 \pm 0.54) \times 10^{-2}$ rad and $\omega \tau(225°) = (-2.93 \pm 0.51) \times 10^{-2}$ rad respectively were obtained and $\omega \tau = (2.25 \pm 0.32) \times 10^{-2}$ rad was deduced from the weighted average of the measurements. The lifetime of the 264 keV level in $^{77}$As has been recently reported by Chopra et al$^2$ to be $\tau = 500 \pm 30$ ps. Using a fast coincidence system with Pb loaded plastic scintillators we also measured the lifetime of this state. Our result (to be published elsewhere) is $\tau = 400 \pm 30$ ps.

The average value of $\omega \tau = (2.25 \pm 0.32) \times 10^{-2}$ rad with $H = 30$ kOe and $\tau = 450 \pm 30$ ps yields the g-factor of the 264 keV state: $g_{5/2}^- = +0.33 \pm 0.05$.

The g-factor of the short-lived 632 keV level was measured making use of the hyperfine field present at As nuclei in a dilute solid solution (1 at %) of Ge as impurity in Fe. The $^{77}$Ge activity in metallic form was deposited in a pot made of pure Fe. A tapered pin was driven in the pot under vacuum and the sealed pot was melted in an induction coil in presence of Argon gas. After melting, the alloy was annealed for 2 h at 800°C in Ar atmosphere and placed in a polarizing field of 5 kOe. The ratio R(8) was then measured at $\theta = 135°$ and 225°C for both states, 264 keV and 632 keV, in the same source through the 367-264 and 558-417 keV gamma sequences respectively.

The results are shown in table 1, giving average values of $\omega \tau(264$ keV) = $0.204 \pm 0.014$ rad and $\omega \tau(632$ keV) = $0.185 \pm 0.030$ rad, under the same hyperfine magnetic field. Using the known lifetimes (table 1) and the measured g-factor of the 264 keV state in an external field, the g-factor of the 632 keV state is obtained to be $g_{5/2}^- = +1.42 \pm 0.32$. 

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**1** Chopra et al.

**2** Chopra et al.
Recently Chopra and Tandon\(^5\,6\) measured the g-factor of both levels, 264 keV and 632 keV, using the hyperfine field on As in a Ge\(_7\)Fe alloy. In the analysis of their data Chopra and Tandon used the hyperfine field obtained from a NMR experiment by Koi et al.\(^7\) in a AsFe alloy. Our value for the g-factor of the 264 keV is in good agreement with their result of \(g_{5/2}^- = +0.30 \pm 0.03\) (with \(\tau = 500\) ps), but they obtain a lower value of \(g_{5/2}^- = +0.79 \pm 0.17\)

### Table 1 - Values of \(\omega t\) for the \(^{77}\)As states of 264 keV and 632 keV in a Ge\(_7\)Fe alloy

<table>
<thead>
<tr>
<th></th>
<th>264 keV</th>
<th>632 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\omega t) (225°)</td>
<td>-0.196\pm0.018 rad</td>
<td>-0.190\pm0.045 rad</td>
</tr>
<tr>
<td>(\omega t) (135°)</td>
<td>+0.212\pm0.020 rad</td>
<td>+0.181\pm0.040 rad</td>
</tr>
<tr>
<td>(\omega t) a</td>
<td>0.204\pm0.014 rad</td>
<td>0.185\pm0.030 rad</td>
</tr>
<tr>
<td>(\tau) (ps)</td>
<td>450\pm30 ps</td>
<td>95\pm8 ps</td>
</tr>
<tr>
<td>(g)</td>
<td>+0.33\pm0.05 b</td>
<td>+1.42\pm0.32</td>
</tr>
</tbody>
</table>

a) Average value of \(\omega t\) at \(\theta = 135°\) and 225°.
b) Measured with an external field of 30 kOe.
c) Weighted average from refs. 3 and 4.

for the 632 keV state. Our result of \(\mu = +3.55 \pm 0.30\) n.m. agrees reasonably with the calculated value of 4.3 n.m. for a 5/2\(^+\) state obtained by coupling of the \(g_{9/2}\) proton with the first 2\(^+\) state of the neighbouring nuclei.

### REFERENCES

The nature of the 632 keV (5/2+) level in $^{77}$As

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There has been considerable theoretical interest\(^1-4\) in the odd-\(A\) isotopes of arsenic whose energy levels cannot be described by simple single particle or collective models. These isotopes are characterized as having a ground state of \(3/2^-\) and apart from a low lying triplet \(1/2^-, 3/2^-, 5/2^-\), also a low lying \(3/2^+\) and \(5/2^+\) doublet in the level spectrum\(^5\). The occurrence of these low lying even parity states is also characteristic of the odd-\(A\) isotopes of Ga, Br and Kr. Recent calculations of Scholz and Malik\(^3\) could account for these states using the statically deformed collective model with the inclusion of the Coriolis coupling and a residual interaction of the pairing type. Though lot of experimental investigations have been made on the odd parity states of arsenic isotopes\(^6-10\) very little information is available about the \(5/2^+\) states.

The \(g\) factors of the \(9/2^+\) states in the arsenic isotopes have been very well explained using the single particle model after taking into account the core polarization effects\(^11\). We report here the \(g\) factor measurement of the \(5/2^+\) state in $^{77}$As using perturbed angular correlation technique. The large hyperfine fields present at arsenic site in iron host are utilised for the measurements\(^12,13\). The short half life of\(^14\) this state, \(T_{1/2} = (60 \pm 6) \text{ms}\), necessitates the requirement of high magnetic fields for the measurements.

Levels in $^{77}$As are populated in the \(\beta^-\)-decay of \(11.33 \text{m}^{77}\text{Ge}\). High purity germanium metal was irradiated by neutrons for a period of 30 m. The activity besides having $^{77}$Ge and its decay product $^{77}$As also contained a large amount of $^{75}$Ge(80 m). The $^{75}$Ge activity was allowed to decay for 10-36h before starting the measurements. The alloys of Ge and Fe, containing 3 at.\% Ge, were made by melting, in vacuum, radioactive Ge and Fe in the required proportions. In an earlier experiment\(^10\), it was shown that the field at \(1/2^\) in Fe was about the same for Ge concentrations of 1, 2 and 3 at.\%.

The \(g\) factor measurements were done using a conventional two channel coincidence system using NaI(Tl) detectors. The 588-617 keV gamma cascade through the 632 keV level was used for the investigations. The directional correlation of this cascade measured both in the 'free' and in the 'magnet' geometry gave identical results. The correlation coefficients obtained (corrected for finite size of the detectors) are \(A_2 = 0.123 \pm 0.006\) and \(A_4 = -0.008 \pm 0.010\). These values are in good agreement with a recent measurement of Gould et al.\(^15\) who have used Ge(Li) - NaI(Tl) coincidence system. The value reported by them is \(A_2 = 0.140 \pm 0.013\) and \(A_4 = -0.035 \pm 0.017\). The fractional change in coincidence counting rate upon field reversal \(\pm 1\) was measured with counters at \(\pm 125^\circ\) and in a polarizing field of 10 kG. The value of \(\pm 1\) obtained is \(R = 0.053 \pm 0.002\). From this the average rotation of the correlation \(\omega_C\) was obtained as \(\omega_C = 0.162 \pm 0.019\). Using \(T_{1/2} = (60 \pm 6) \text{ms}\)\(^14\) and \(H_{eff} = H_{app} = 121.3\) kG, the \(g\) factor is obtained to be \(g = +1.01 \pm 0.14\). The spin of the 632 keV state being \(5/2^\), its magnetic moment is \(\mu = +2.53 \pm 0.60\) n.m.

The observed \(g\) factor of this state rules out \(5/2^+\) to be a single particle \(9/2\) state \((g = 1.9)\). The \(g\) factor of a \(9/2^-\) state obtained by coupling of a \(9/2^+\) state \((g_{9/2} = 1.31\)\(^11\)) to the \(3^+\) state of the core \((g_{3/2} = 0.4)\) is \(g = 1.0\). This value being well outside the experimental results, such configuration of the \(5/2^+\) state is also ruled out. The measured values, however, agree well with the Nilsson estimates for \(\gamma_2 = 3\) assuming this state to be \(5/2^+\) \([415]\) \((g = 1.24)\). The existence of these positive parity \(9/2^+\) and \(5/2^+\) states could only be explained using positive deformation for arsenic isotopes\(^3\) which is further borne out by the present experiment.
References

1. L.S. Kisslinger and R.A. Sorensen, Rev. Mod. Phys. 35 (1963) 853
   (Rotterdam University Press 1971) p.543
8. R.C. Chopra and P.K. Tandon, Pramana 1 (1973) 70
15. J.M. Gualda et al., (Preprint 1973)
A study of the system $^{103}\text{RhFe}$ is interesting for both solid state physics (formation of a local moment at the Rh atom in Fe (1), and for nuclear physics: The $7/2^+$ metastable "intruder" state, which is not explained by the simple shell model, has been predicted by the extended quasi-particle phonon coupling model of Sherwood and Goswami (2). An alternative description of the state might be expected from the Coriolis coupling model of Imanishi et al. (3). To date, the magnetic moments of only two of these intruder states near $A = 100$ have been determined ($^{99}\text{Tc}$ and $^{103}\text{Ag}$). Further magnetic moment measurements should provide a more sensitive test of the above models than is given by level spectra alone. As a preliminary step to an NMR-ON experiment we have measured the magnetic moment of the ($7/2^+$, 57 min) state in $^{103}\text{RhFe}$. Two sources were prepared as follows: $^{103}\text{Pd}$ activity was electroplated onto 6 - 12 $\mu$ Fe foils, reduced in $\text{H}_2$-atmosphere at 800 °C for 3 hrs and diffused in vacuum for 14 hrs at 1000 °C. The 0.1 mCi sources (atomic concentration $3 \times 10^{-4}$) were cooled to about 40 mK in a $\text{He}^3$-$\text{He}^4$ dilution refrigerator, using the $\gamma$-anisotropy of $^{58}\text{CoFe}$ and $^{60}\text{CoFe}$ for temperature determination.

The 40 keV $\gamma$-ray from $^{103}\text{Rh}$ was detected by a planar Ge(Li) detector with a Be entrance window; the thermometer $\gamma$-ray anisotropy was measured using a coaxial Ge(Li) spectrometer. The value of the hyperfine field at RhFe was taken to be -54.3 T, based on an NMR measurement on stable $^{103}\text{RhFe}$ (4).

Fig. 1 gives a graphical representation of our results for the moment $\mu(7/2^+)$ at various applied fields $H_0$ and 37 mK (circles). The triangle represents data taken with the first source at 50 mK using the $^{58}\text{CoFe}$ thermometer. Errors shown are statistical. All values were calculated under the assumption of complete alignment of the local fields. Although this assumption is incorrect for $H_0 < 0.2$ T (5), the observed deviation of the 40 keV $\gamma$-anisotropy is nearly compensated by the simultaneous deviation of the measured temperature, so that no systematic variation in the derived moment values is seen even for small applied fields. Nevertheless, the points below $H_0 = 5$ T were considered to be less reliable because of the incomplete magnetic saturation of the source and thermometer foils, and were not included in the final value. Also, the point at 1.0 T was corrected for a shift (10 %) due to the stray-field sensitivity of one of the $\gamma$-ray detectors and was thus omitted from the final average. We then obtain

$$\mu = (4.78 \pm 0.1) \text{ nm}$$

as our final result.
This work was supported in part by the Bundesministerium für Bildung und Wissenschaft.

References

The magnetic hyperfine splitting constants for $^{105}$RhFe and $^{105}$RhNi were determined as $539.608 \pm 0.020$ MHz and $218.070 \pm 0.030$ MHz, respectively. With the assumption that hyperfine anomalies can be neglected the nuclear g-factor of $^{105}$Rh is deduced as $1.2650 \pm 0.0036$. The hyperfine field of Rh in Fe is determined to be $H_{\text{HH}} = -226.15 \pm 0.65$ kG.

From decay scheme studies the ground state of $^{105}$Rh($T_{1/2} = 39.5$ h) is assumed to have $J = 7/2^+$. This could be explained by a seniority 3 proton configuration. This assumption can be tested by a measurement of the nuclear g-factor. The NMR-ON technique is well suited for this purpose.

The hyperfine field of Rh in Fe is known to be $-559.6 \pm 1.6$ kG. The $^{105}$Rh activity can be produced by thermal neutron capture on $^{104}$Ru and subsequent beta decay to $^{105}$Rh. Sources of $^{105}$Rh in Fe and Ni were prepared by melting enriched $^{104}$Ru metal powder with Fe and Ni of high purity. The samples were cold-rolled with intermediate annealing steps, until a final thickness of $0.5 \ldots 1.0$ um was obtained. After an irradiation of 3 days in a thermal neutron flux of $9 \times 10^{13}$ sec$^{-1}$cm$^{-2}$ the sources were cooled to $0.01$ K, and NMR-ON spectra were obtained. Details of the experimental procedure are described in ref. /3/. Typical resonance curves are shown in Fig.1. Fig.2 shows the linear dependence of the resonance frequency on the applied external field. From a least squares fit we obtain the hyperfine splitting constants $539.608 \pm 0.020$ MHz for $^{105}$RhFe and $218.070 \pm 0.030$ MHz for $^{105}$RhNi. With the known hyperfine field of Rh in Fe we derive /2/ the g-factor of the ground state of $^{105}$Rh as $1.2650 \pm 0.0036$. Hyperfine anomalies are neglected in this derivation. This experimental result is close to the known $g_{9/2}$ factor in this mass region and thus supports seniority 3 ground state configuration mentioned in the introduction.

The hyperfine field of Rh in Ni is determined as $H_{\text{HH}} = -226.15 \pm 0.65$ kG, in good agreement with the extrapolated ($T = 0$ K) value of $-223 \pm 3$ kG obtained from ref. /4/.

/3/ E. Hagn, P. Kienle, G. Eska "NMR-ON on $^{52}$MnFe and $^{58}$CoFe", this conference
The purpose of this investigation was to determine the unknown $g$-factors of the first $106$ keV/ and second states in $^{151}$Gd $I=5/2^-$ and $3/2^-$ respectively/ and to continue our former measurements. It was investigated by the IPAC technique with magnetic field $H_{nf}$ at room temperature of Gd implanted in Fe by means of an isotope separator with the implantation energy of $70$ keV/. To check the influence of the relaxation processes, the attenuation factors $G_2$ of the angular correlations in $^{151}$Gd in the Fe were measured without external magnetic field at room at liquid-nitrogen temperature. The $g$-values were deduced using the value of $H_{nf}/200 \pm 50 / kG$ at room temperature from. The 108 keV and 395 keV levels in $^{151}$Gd has been interpreted in as members of the multiplet resulting from the coupling of the $f_{7/2}$ single neutron orbit of the ground state to the $2^+$ collective excitation of the $^{150}$Gd even-even core. Within the de Shalit model we have deduced the theoretical $g$-factors, using for the ground state the Schmidt $g$-value in $g_1$ and the Airmé-Horie $g$-value in $g_2$.

### Table 1.

| Source | Temperature | Cascade /keV/ | $A_2/A_4=0/ | G_2$
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Aqueous solution of $^{151}$Gd in HCl</td>
<td>room</td>
<td>287-108</td>
<td>$-0.240^{+0.017}_{-0.017}$</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>room</td>
<td>444-287</td>
<td>$-0.161^{+0.015}_{-0.015}$</td>
<td>1</td>
</tr>
<tr>
<td>$^{151}$Gd implanted in Fe</td>
<td>room</td>
<td>287-108</td>
<td>$-0.253^{+0.046}_{-0.046}$</td>
<td>unattenuated</td>
</tr>
<tr>
<td></td>
<td>liquid-nitrogen</td>
<td>287-108</td>
<td>$-0.155^{+0.022}_{-0.022}$</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>room</td>
<td>444-287</td>
<td>$-0.229^{+0.079}_{-0.079}$</td>
<td>unattenuated</td>
</tr>
<tr>
<td></td>
<td>liquid-nitrogen</td>
<td>444-287</td>
<td>$-0.121^{+0.050}_{-0.050}$</td>
<td>0.75</td>
</tr>
</tbody>
</table>

### Table 2.

<table>
<thead>
<tr>
<th>Level /keV/</th>
<th>$T_2$ /ns/</th>
<th>Cascade /keV/</th>
<th>$H/H = (12_1/4+12_2)sin(4\alpha)$</th>
<th>$N_0$</th>
<th>$g$ exp.</th>
<th>$g$ theor.</th>
<th>$g$ theor.</th>
</tr>
</thead>
<tbody>
<tr>
<td>108</td>
<td>4.33$^{+0.15}_{-0.15}$</td>
<td>287-108</td>
<td>$+0.065^{+0.010}_{-0.010}$</td>
<td>40°</td>
<td>$-0.69^{+0.17}_{-0.17}$</td>
<td>$-0.69$</td>
<td>$-0.36$</td>
</tr>
<tr>
<td>395</td>
<td>0.45$^{+0.05}_{-0.05}$</td>
<td>444-287</td>
<td>$+0.122^{+0.022}_{-0.022}$</td>
<td>19° or 26°</td>
<td>$-0.90^{+0.27}<em>{-0.27}$ or $-1.49^{+0.41}</em>{-0.41}$</td>
<td>$-1.55$</td>
<td>$-0.96$</td>
</tr>
</tbody>
</table>

### References.

III. 18

NUCLEAR MAGNETIC RESONANCE OF THE $^2$ EMITTER $^{108}$Ag($^1/2$ - 2.4 m) IN SILVER CHLORIDE
by H. Ackermann, D. Dubbers, M. Grupp, P. Haitjema, and H.J. Stückmann
I. Physikalisches Institut der Universität Heidelberg and Institut Laue-Langevin, Grenoble.

The nuclear magnetic moment of $^{108}$Ag ($^2$) had been studied before by atomic beam resonance yielding the results $4.2(5)$ n.m. and later $2.79(2)$ n.m. $^1$. According to shell model arguments both results seem to be reasonable. An exact result for $\mu_2^{108}$Ag may be obtained from an NMR measurement of $^{108}$Ag in a crystal with cubic symmetry using the polarized-neutron capture asymmetry technique. An experiment of this type has been performed by Gulko et al. $^3$ using a rolled $^{108}$AgCl single crystal at 4.2 K. However no NMR signal could be observed. This was probably due to the strong creation of lattice defects by the neutron capture process which are completely frozen at the applied low temperature. Indeed, it turned out in the present experiment that the resonance effect was very small.

Our experiment was performed at the High Flux Reactor Grenoble. A polycrystalline AgCl sample $^4$ (containing 2g of 99.9$^{107}$Ag) was irradiated with polarized thermal neutrons at 9K. The beam intensity was $5 \times 10^7$ polarized neutrons per cm$^2$ sec at the target. The polarization and the NMR of the activated $^{108}$Ag nuclei were monitored via the $\beta$ decay asymmetry. In order to search the weak signal a frequency modulated rf field (depth of modulation 90kHz) was irradiated which resulted in a quickly detectable NMR curve of about 150 kHz width. For the determination of the magnetic moment an unmodulated rf field was used (Fig. 1). The resonance position was determined relative to that of the NMR signal of $^8$Li in LiF taken at the same values for temperature and magnetic field. The result for the ratio of their gyromagnetic ratios is

$$\frac{\gamma_{^{108}\text{Ag in AgCl}}}{\gamma_{^8\text{Li in LiF}}^0} = 3.2341(7)$$

at 9K.

Inserting for $\gamma_{^8\text{Li in LiF}}$ at 9K the value measured at room temperature $^3,^5$ we obtain

$$\frac{1}{2^2} \mu_2^{^{108}\text{Ag}} = 2.0374(11)$kHz/0e or \(\mu_2^{^{108}\text{Ag}} = 2.6728(15)$n.m.

without diamagnetic correction.

The magnetic moment of $^{108}$Ag deviates only by -1.4 % from that of $^{110}$Ag(24s)$^6$. Thus both isotopes possess nearly the same configuration which has been proposed to be $^{110}$Ag(24s)$^6$ of the $^1f$ coupling model predicts for this configuration the empirical magnetic momentum value $3.2$ n.m.

---

Fig. 1. NMR of $^{108}$Ag in AgCl shown by change of the $\beta$-decay asymmetry at 9K target temperature. The magnetic field was $H_0 = 4981$ Oe, the rf field strength about $2H_{\text{rot}} = 0.4$ Oe. The linewidth is 5kHz.
References

1) G.K. Rochester, K.F. Smith, Phys. Letters B 266 (1964)
4) We are indebted to Dr. P. Wolf, KFZ Karlsruhe, for the preparation of the sample.
We have measured the following g-factors:

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>State</th>
<th>$T_{1/2}$</th>
<th>Reaction</th>
<th>Energy (MeV)</th>
<th>g-factor (uncorrected)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{113}$Sn</td>
<td>$11/2^-$</td>
<td>89 ns</td>
<td>$^{113}$In($p,n$)</td>
<td>7</td>
<td>-0.235(4)</td>
</tr>
<tr>
<td>$^{132}$Xe</td>
<td>$10^+$</td>
<td>8.4 ns</td>
<td>$^{130}$Xe($a,2n$)</td>
<td>35</td>
<td>-0.195(5)</td>
</tr>
<tr>
<td>$^{205}$Po</td>
<td>$13/2^+$</td>
<td>640 μs</td>
<td>$^{204}$Pb($a,3n$)</td>
<td>41</td>
<td>-0.144(7)</td>
</tr>
</tbody>
</table>

All measurements were performed by the pulsed beam time differential perturbed angular distribution method using a proton beam from the Berlin Van de Graaff and an α-particle beam from the Karlsruhe cyclotron. In the first two cases the targets were kept liquid to avoid static quadrupolar interaction. In liquid Te the relaxation time of $^{132}$Xe is about 1.5 ms at 500°C [1]. The $^{205}$Po isomer was measured in a solid Pb target heated close below the melting point because the relaxation time at this temperature is larger by a factor of 2 than in liquid Pb [2]. It is important in this case to decrease the attenuation of the alignment, since the nuclear lifetime of the isomer considerably exceeds the relaxation time ($T_R=70\mu$s).

$^{113}$Sn: The measured g-factor of the $11/2^-$ state in $^{113}$Sn fits well into the systematics recently interpreted for g-factors of neutron $11/2^-$ states in Cd and Sn isotopes [3,7]. Comparing the measured transition rate [4] of the $11/2^- + 7/2^+$ decay of this state with a single particle estimate including the reduction factor resulting from pairing theory [5], an agreement within 30% is obtained, which holds for transitions in other Sn isotopes, too. For the pairing reduction factors experimentally determined occupation

Time spectrum of the 773 keV γ-ray decaying from the $10^+$ isomer in $^{132}$Xe
probabilities from spectroscopic factors were used. Thus the assumption that the 11/2\(^{-}\) states in the Sn region should have a rather pure \(v_{h_{11/2}}\) configuration is supported.

\(^{132}\text{Xe}\): It has been concluded \cite{6} that the 10\(^{+}\) isomer in \(^{132}\text{Xe}\) has a quite pure quasiparticle \((v_{h_{11/2}})^{2}\) configuration. Thus the magnetic moment of the \(h_{11/2}\) neutron in this nucleus can be derived to be \(\mu = -1.07 (3)\) n.m. This moment is close to known moments of other \(h_{11/2}\) states in Cd and Te isotopes, which all agree within the range \(-1.0 \pm 0.1\) n.m., and may be interpreted in terms of core polarization effects including pairing correlations \cite{3,7}.

\(^{205}\text{Po}\): The 640 \(\mu s\) state of this nucleus belongs to the series of \(v_{13/2}^{+}\) isomers in the neutron deficient isotopes \(^{207}\text{Po}\) through \(^{201}\text{Po}\). Within the experimental error the measured moment equals that of the 13/2\(^{+}\) state in \(^{207}\text{Po}\) \cite{8}. These and other known \(v_{13/2}\) moments in the nuclei \(^{193,195,197,199}\text{Hg}\) \cite{9}, \(^{205,206}\text{Pb}\) and \(^{205,208}\text{Bi}\) \cite{10} offer the chance to investigate systematically the influence of the neutron configuration on the moments in this mass region.

This work was supported by the Bundesministerium für Forschung und Technologie.

References:

\cite{1} J. Hadjuana et al.: contribution to this conference
\cite{2} F. Dimming et al.: contribution to this conference
\cite{3} D. Riegel et al.: Phys. Lett. \textbf{A63}, (1973), 170
\cite{5} S. Kisslinger and R.A. Sorensen: Rev. Mod. Phys. \textbf{25}, (1963), 553
\cite{6} A. Kerek et al.: Nucl. Phys. \textbf{A172}, (1971), 603
\cite{7} R.E. Silverans et al.: Nucl. Phys. \textbf{A210}, (1973), 307
\cite{8} D. Riegel et al.: Phys. Lett. \textbf{A48}, (1973), 456
\cite{9} R.J. Heinem and M.N. McDermott: Phys. Rev. \textbf{TC}, (1973), 2066
\cite{10} N. Bräuer et al.: contribution to this conference, and references therein
Even-even isotopes of Xe can be studied over a range from $A=124$ to $A=136$ and offer therefore, a unique possibility to study effects of closing the neutron $^{11/2}$ shell which is finished at $^{136}$Xe. Here we report measurements of the g-factors of the first 2+ states in $^{126, 128, 130, 132}$Xe, for which we have obtained results so far.

All stable Xe-isotopes are accessible to measurement by the IMPAC technique. Such data were obtained for the four isotopes mentioned above and in addition a measurement was performed on the 2+ state in $^{126}$Xe using radioactive sources of $^{126}$I. The latter experiment was made for calibration purposes, since the IMPAC data, which were taken with Xe on Fe-foils, depend on the average hyperfine field $B_{HF}$ for Xe-ions stopped in Fe as well as the transient field. The average hyperfine field is not well known in this case and may depend on implantation conditions since more than one site is populated [1]. The net transient precession, $\omega/g = (-36\pm3)$ mrad as taken from the systematics is on the other hand relatively well known and acts in this case in the same direction as the precession in the hyperfine field.

The IMPAC-measurements were performed at the Uppsala tandem accelerator using $^{16}$O beams of 30-49 MeV energy on targets prepared by shallow implantation of Xe in a mass separator. Stable targets containing 3-90 pg/cm$^2$ of the Xe isotopes could be obtained using low current and very long implantation times [8]. The IMPAC equipment consisted of four 3" x 3" NaI detectors and a 300 mm$^2$ annular silicon detector for backscattered particles mounted in an electromagnet run at 4.5 KG. Corrections were applied for beam bending (2 mrad).

The results for $\omega t$ are shown in Fig. 1. For $^{128}$Xe and $^{132}$Xe they agree with data reported by D. Gordon et al. [2] but for $^{130}$Xe the result differs significantly. The life-times used for deriving the g-factors were in part taken from ref. [3] and partly from B(E2) data obtained at the Uppsala tandem accelerator [4]. The $^{126}$Xe calibration experiment was made on $^{126}$I sources prepared by irradiating enriched $^{128}$Te by 30 MeV protons in the Gustaf Werner synchrocyclotron. Measurements were performed on chemically separated iodine in the form of AgI. The highly anisotropic but relatively weak 2++2++0+ cascade in $^{126}$Xe was used in a 4-detector system described earlier [5] with an external magnetic field of 50 KG. The result obtained for the Larmor precession angle, $\omega t$, of the first 2+ level was $\omega t = 5.3\pm1.0$ mrad corresponding to a g-factor of

$$g = +0.37(7)$$

The effective hyperfine field for the Xe atoms recoil implanted into Fe could now be ob-
tained by taking the ratio between the two \( \omega - \) values for \(^{126}\text{Xe}(2^+)\) and assuming \( \psi / g = -36(3) \text{ mrad} \) as mentioned above. The result is a mean \( B_{hf} = +730(80) \text{ kG} \). This can be compared to the value for substitutional sites, \( 1510(160) \text{ kG} \) as reported in a nuclear orientation measurement \([1]\) and \( 1510(200) \text{ kG} \) in a Mössbauer experiment \([6]\), and the average value for \(^{127}\text{Xe}\) implanted at 50 keV acceleration voltage measured by nuclear orientation \([1, 7]\), which is \(-1000 \text{ kG}\).

The g-factors of \(^{128}\text{Xe}(2^+)\), \(^{130}\text{Xe}(2^+)\) and \(^{132}\text{Xe}(2^+)\) could now be derived from the measured data. The results of all g-factors are presented in Table 1.

| PAC \(^{126}\text{Xe}\) & IMPAC & PAC \(^{128}\text{Xe}\) & PAC \(^{130}\text{Xe}\) & PAC \(^{132}\text{Xe}\) |
|-----------------|-------|-----------------|-----------------|-----------------|
| \( \omega \) (mrad) | 5.3(10) | 89(7) | 51(2) | 32(3) | 21(3) |
| \( \tau \) (ps) | 59.6(20) | 34(3) | 15(2) | 6.7(7) |
| \( g \) | 0.37(7) | 0.33(7) | 0.36(7) | 0.35(5) |

Over this range of isotopes all 2+ g-factors agree with the "collective" value given by Greiner \([9]\). It remains to see whether any drastic change will appear on approaching \(^{136}\text{Xe}\) and a continuation of this series of experiments is therefore planned.

References

5. B.E. Karlsson, Arkiv Fysik 22 (1962) 1
8. A. Arnesen, T. Noreland and L.-O. Norlin (to be published)

* On leave from Institute of Nuclear Physics, Krakow, Poland.
Some years ago Prof. F.I. Shapiro proposed the method of measuring the magnetic moments of high energy nuclear states excited by neutron resonance capture. The method is based on the observation of the shift of the resonance due to the hyperfine interactions between the magnetic moment of compound state and an internal magnetic field. For this purpose one needs a polarized neutron beam or nuclear oriented target technique. In the last case the shift \( \Delta E \) is given by:

\[
\Delta E = -\frac{\hbar}{2} H_i (m_J - m_1), \quad \Delta E = -\frac{\hbar}{2} H_i (m_J^+ - m_1)
\]

where \( H_i \) - the hyperfine magnetic field, \( f_N \) - nuclear polarization in magnetic domains, \( I \) and \( J+ = I \pm 1/2 \) - the spins of the target and compound states, \( f_I = 1/(2I + 1) \) (\( I+1 \)), \( m_J \) and \( m_1 \) - the magnetic moments of corresponding states.

The first results were obtained in /2,5/ for Erbium, here we are reporting the preliminary results for Dysprosium.

Dy-metal is normal ferromagnet below 85°C with \( H_i = 5.2 \times 10^6 \) oe /4/. Magnetic moments of the ground states \(^{161}\)Dy and \(^{163}\)Dy are equal to \(-0.47\mu_B\) and \(+0.56\mu_B\).

At temperature \( T \approx 0.05\)K the internal field \( H_i \) orients the spins of the nuclei along the direction of easy magnetization in domains (axes in the basal plane). The calculated values of the \( f_N \) are 0.84 \( (^{161}\)Dy) and 0.92 \( (^{163}\)Dy). The absence of the effective average polarization of the target (in the zero field) makes the shift observation easier. However, the expected shift (\( \approx 10^{-6} \) eV assuming \( m_J - m_1 \approx 1\mu_B \)) is small in comparison with the width of the resonance \( \approx 10^{-7} \) eV. This circumstance complicates the measurements.

Unlike the first experiment /3/ performed at pulse reactor IBR-30, new measurements were carried out at IBR-30, which worked in Lina mode of operation with the 20nsec pulse of the 30 meV Linac. The neutron beam was transmitted through the \((200 \times 60 \times 0.6 \) mm\) plate of Dysprosium inside the \(^3\)He - \(^4\)He refrigerator. The transmission of the sample versus the neutron energy was measured by the time-of-flight method \((E=58.5keV)\) with the help of the 200 \( \ell \) liquid scintillator detector /3/ and the mini-computer TPA /5/, which worked as the fast time analyzer.

The example of the time-of-flight spectrum (6 hour, 2.5 nsec channel width) is shown by Fig 1. The resonance energies are given in the top of Fig.1. The target was heated from 0.03K to 0.5K periodically every 12 hours and the resonance shifts were measured by the least square fitting. The systematic shift which was not connected with the temperature change was measured using the resonances of other samples outside the cryostat: 6.24 and 15.4 eV Antimony, 1.30 eV Iridium.
As an example, Fig. 2 gives the frequency distribution of obtained $\Delta E$ values for 59 runs in 1.71 eV resonance. The smooth curve is Gaussian with the experimental dispersion. The preliminary results for all measured resonances are given in the Table. The large value of the magnetic moment (2.6nm) discovered for Dy – the isotope with the great neutron number.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$^{164}$Dy</th>
<th>$^{162}$Dy</th>
<th>$^{165}$Dy</th>
<th>$^{162}$Dy</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E$(eV)</td>
<td>1.71</td>
<td>2.72</td>
<td>3.69</td>
<td>4.35</td>
</tr>
<tr>
<td>$E$(meV)</td>
<td>$-24.7 \pm 6.4$</td>
<td>$+1.5 \pm 1.0$</td>
<td>$-34.5 \pm 12.5$</td>
<td>$+15.5 \pm 17.2$</td>
</tr>
<tr>
<td>$m_J$(nm)</td>
<td>$+2.6 \pm 0.5$</td>
<td>$-0.4 \pm 0.8$</td>
<td>$-1.7 \pm 1.0$</td>
<td>$+0.8 \pm 1.4$</td>
</tr>
</tbody>
</table>

These results are the first experimental data for the magnetic moments of the Dy-compound states at nearly 8 meV excitation energy. They are in agreement with the theoretical conception about the single particle order of the magnitude for the magnetic moments of such nuclear level /6/.

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III. 22 NMR-study of oriented $^{181}$Re(19h) and $^{182}$Re(64h) nuclei after recoil implantation into Fe

E. Hagn, P. Kienle, Phys. Department- TU München
G. Ew,, A. Zentralinstitut für Tieftemperaturforschung, Garching

Using the NMR-ON technique, we have determined the magnetic hyperfine splitting constants of $^{181}$ReFe and $^{182}$ReFe as $729.08 \pm 0.15$ MHz and $231.34 \pm 0.04$ MHz, respectively. With the hyperfine field of $-760 \pm 15$ kG the g-factors of the ground states are deduced to be $1.297 \pm 0.026$ and $0.399 \pm 0.008$, respectively.

Because of the relatively short half lives of $^{181,182}$Re(19h, 64h), the sources could not be prepared as described in ref. /1/ for $^{184}$ReFe. It has been shown, however, that the NMR-ON method is applicable in cases where the activities are produced in thin Fe foils by nuclear reactions /2/.

Taking this into account, the recoil implantation technique has been combined with the NMR-ON method.

a) $^{182}$ReFe

Sources of $^{182}$Re (64h) in Fe were prepared in the following way: A target stack consisting of 6 Ta foils (4 mg/cm$^2$) each of which was followed by a Fe foil (0.8 mg/cm$^2$) was bombarded with 36 MeV/$\alpha$-particles at the Karlsruhe Cyclotron. $^{182}$Re is produced via the reaction $^{181}$Ta($\alpha$,3n)$^{182}$Fe and implanted into the Fe catcher foils. The most active parts of the iron foils were cut out and cooled to temperatures as low as 0.01 K. Using two Ge-Li detectors (25 cm$^3$), which were placed at 0° and 50° with respect to the direction of the external magnetic field, the anisotropy of several $\gamma$-rays at 0.01 K was roughly analyzed. Thus the hyperfine splitting $g_{\text{HF}}$ was estimated to be $250 \pm 50$ MHz. For the NMR experiment two 3" $\phi$ x 3" NaJ (Tl) detectors were used to get better counting statistics. Although it was not possible to resolve the different $\gamma$-rays due to the complexity of the $^{182}$Re decay scheme the use of scintillation detectors turned out to be a good procedure, since the corresponding $\gamma$-ray spectra showed regions of positive and negative anisotropies at 0.01 K.

These regions were selected by single channel discriminators and the corresponding counts were stored in a multichannel analyzer as a function of the frequency of the applied rf field. Fig. 1 shows a plot of the number of counts versus the frequency for a region of negative $\gamma$-ray anisotropy measured in the direction of the polarizing field with an external magnetic field of $H_0 = 1.07 \pm 0.01$ kG. Fig. 2 shows the linear dependence of the resonance frequency on the external field $H_0$. A least squares fit yields $g_{\text{N}}^H = 231.34 \pm 0.04$ MHz and $g_{\text{N}}^H = 0.318 \pm 0.025$ MHz/kG.

Using a hyperfine field of $-760 \pm 15$ kG /3/ and neglecting hyperfine anomalies which should be small, we get $g = 0.399 \pm 0.008$.

b) $^{181}$ReFe

The sources of $^{181}$Re (19h) in Fe were prepared in a similar manner as described before, with the only exception that the $\alpha$-energy was chosen to 45 MeV. From systematics of the odd Re isotopes with $A = 183, 185, 187$, which have $I = 5/2^+$ ground states like $^{181}$Re and known magnetic moments, the region of search was chosen from 700 to 750 MHz. The lower half of Fig. 1 shows the anisotropy of the 368 KeV $\gamma$-ray versus the frequency for $H_0 = 1.07 \pm 0.01$ kG. In this experiment it was impossible to reduce the deviation of the FM below $\pm 1$ MHz, probably because the short spin lattice relaxation time connected with the high frequency causes a small depolarization effect only. For this reason the shift...
of the resonance with $H_0$ (fig. 2) could not be measured very accurately. A least squares fit yields $g_{N_{H_0}} = 729.08 \pm 0.15$ MHz. From this the $g$-factor of the $^{181}\text{Re}$ ground state is deduced to be

$$g = 1.297 \pm 0.026.$$ 

We want to point out here that the errors given for the $g$-factors are essentially due to the error of the value of the hyperfine field. Neglecting hyperfine anomalies, we can calculate more accurate values for the ratios of the $g$-factors of $^{181,182,184}\text{Re}$:

$$g(^{181}\text{Re}) / g(^{182}\text{Re}) = 3.1518 \pm 0.0010$$
$$g(^{184}\text{Re}) / g(^{182}\text{Re}) = 2.0858 \pm 0.0006$$

1/ T. Butz et al. Z. Physik 254, 312 (1972)
2/ E. Hagn, P. Kienle, G. Eska "NMR-ON on $^{57}\text{MnFe}$ and $^{59}\text{CoFe}$" this conference
The $g$-factor of the $^{184}$Re ground state has been determined to be $g = 0.833 \pm 0.017$ by the NMR-ON method, at which the anisotropy of the $\gamma$-radiation is used as a detector for nuclear magnetic resonance.

The experiment was done in the following way: Sources of $^{184}$Re ($T_1/2 = 32$ days) and $^{183}$Re ($T_1/2 = 716$ days) were produced via the $(d, 2n)$ reaction by irradiating tungsten metal of natural abundance with deuterons at the Karlsruhe Cyclotron. As described in detail in ref. 1, the rhenium activities were separated from tungsten. The carrier free activity was electrolytically deposited on an iron foil, which was then melted in an electron beam furnace. The alloy was coldrolled to a thickness of about 1 µm and annealed for 12 hours at 604°C under a hydrogen atmosphere. Two samples of $8 \times 8 \text{mm}^2$ were soldered to both sides of the copper fin system of an adiabatic demagnetization cryostat and cooled to a temperature of 0.01 K.

In a previous $\gamma$-anisotropy measurement /2/ the hyperfine splitting of $^{184}$Re in Fe was determined to be $485 \pm 15$ MHz. Therefore the resonance was sought between 470 and 500 MHz. The 904 keV $\gamma$-rays were detected by two NaI(Tl) detectors placed at 0° and 30° with respect to the external magnetic field $H_0$ and stored in a multichannel analyzer as a function of the oscillator frequency. The temperature of the source was monitored simultaneously by observing the anisotropy of the 162 keV $\gamma$-rays of Re. In fig. 1 the number of counts is plotted versus the frequency for the 904 keV $\gamma$-rays observed in the direction of the polarizing field for the two different fields $H_0 = 1.07(1)$ kG and 2.94(3) kG. The increase of the background towards higher frequencies

is due to a frequency-dependent heating arising from a resonance in the rf circuit. For the fits of the resonance curves a linear background was taken into account. This assumption is justified by the simultaneously measured $\gamma$-anisotropy of $^{183}$Re, which indicates that the temperature increases linearly in this frequency region.

The best procedure to distinguish NMR signals from rf resonances is to measure the resonance frequency as a function of the external field. In first order /3/ the NMR frequency is given by:

$$\nu = g \mu_N (H_{HF} + H_0)$$

Fig. 2 shows the linear dependence of the resonance frequency on the external field $H_0$ and confirms the negative sign of $H_{HF}$. A least squares fit yields

$$g \mu_N H_{HF} = 482.54 \pm 0.07 \text{ MHz}$$

and

$$g \mu_N = 0.622 \pm 0.039 \text{ MHz/kG.}$$

Taking the hyperfine field of -760 ± 15 kG /4/ and neglecting hyperfine anomalies which should be small, we get $g = 0.833 \pm 0.017$. The accuracy of this value is essentially limited by the poor knowledge of the hyperfine field. A previous measurement of the temperature dependence of the $\gamma$-anisotropy of $^{184}$Re in Fe /2/ yielded $g = 0.84 \pm 0.05$ under the assumption of spin 3 and $g = 1.08 \pm 0.05$ for spin 2. The first value is in good agreement
with this measurement. Taking this into account, one can unambiguously assign spin 3 to the $^{184}\text{Re}$ ground state. According to Gallagher /5/ this spin can be understood as resulting from the coupling of a $5/2^+ [402]$ proton ($^{183}\text{Re}$) and a $1/2^- [510]$ neutron ($^{183}\text{W}$). Then the g-factor is expected to be

$$g = \frac{1}{\Omega_f} \left( g_{k_1} \cdot k_1 + g_{k_2} \cdot k_2 + g_R \right)$$

Taking $g_{k_1,2}$ from the known magnetic properties of the neighbours and $g_R = 0.4 \pm 0.1$ one gets $g = 0.83 \pm 0.03$ /6/, which is in good agreement with our experimental value.

/2/ T. Butz et al. Z. Physik 254, 312 (1972)
/3/ E. Hagn and G. Eska, "Shift of NMR-ON resonances with the external magnetic field", this conference
The ground state hyperfine structure of the neutron-deficient thallium isotopes, \(^{193}\text{Tl} - \text{Tl}^{232}\), has been investigated using the atomic-beam magnetic resonance method. The ground state activities were obtained as daughter products of the corresponding lead isotopes; the latter produced by \((p, \alpha n)\)-reactions using stable thallium as target material in the synchro-cyclotron at the Gustaf Werner Institute in Uppsala. The proton energies chosen ranged from 50 - 140 MeV, and the current was about 0.4 μA. Since the isomeric states in the doubly-odd thallium isotopes are not fed by the decay of lead, we used samples produced by bombarding stable gold with \(a\)-particles in the cyclotron at AIF, Stockholm.

The atomic-beam experiments were performed in the \(^{2s}_{1/2}\) electronic ground state of thallium, indicating that the magnetic dipole hyperfine interaction would be observable. The results of the present investigation are given in the table together with some data previously known. The nuclear ground state spin values being \(I = 1/2\) of all the odd isotopes and \(I = 2\) of all the doubly-odd ones caused some experimental trouble, but the isotopes giving rise to the resonance signals could be unambiguously identified by analysing the radioactive decay curves.

By observing the deviation from the linear Zeeman-effect in successively stronger external magnetic fields, the hyperfine structure separations in the doubly-odd thallium isotopes were determined. More accurate values were obtained in \(^{198}\text{Tl}\) and \(^{200}\text{Tl}\) for which also the direct transitions were measured in weak magnetic fields. The transition \((F_2, m_F) = (3/2, -1/2) \rightarrow (3/2, -1/2)\) in \(^{198}\text{Tl}\) was followed up to 107 gauss giving the electronic g-factor for thallium: \(g_e = -0.66578(4)\). Compared to the doubly-odd isotopes, the odd thallium isotopes all have very large hyperfine structure separations and nuclear moments.

In the latter isotopes, the nuclear g-factors were determined directly as an deviation from the electronic g-factor for the double-quantum transition \((1, 1) \rightarrow (1, -1)\) in external magnetic fields up to 175 gauss.

The nuclear dipole moments evaluated from the experimental data are given in the last column of the table. Thallium has atomic number \(Z = 81\), and according to the nuclear shell model the odd proton assignment should be \(s_{1/2}\). The theoretical dipole moment for this state using \(g_s = 0.6 g_{s_{1/2}}\) is 1.68 n.m., in agreement with the experimental values. The decreasing dipole moments for smaller mass numbers may be attributed to the increase of collective effects.

Although the hyperfine structure separations in the doubly-odd nuclei have been accurately determined, the errors of the nuclear dipole moments are rather large due to the uncertainty introduced by the hyperfine structure anomaly. An upper limit of the absolute error arising from a combination of the wave functions \(2s_{1/2}v_{5/2}\) and \(2s_{3/2}v_{3/2}\) is shown to be as large as ±0.01 n.m.

In the figure are plotted the ground state dipole moments of the doubly-odd thallium isotopes \(^{194}\text{Tl} - \text{Tl}^{204}\); the value of \(^{204}\text{Tl}\) being previously measured. Since the signs of the dipole moments have not been determined, both alternatives are given in the figure. However, from theoretical considerations it can be shown that the moments connected by the line are the most probable ones. For \(g_s = 0.6 g_{s_{1/2}}\) we obtain \(\mu_{1/2}(2s_{1/2}v_{5/2}) = -0.35\) n.m. and \(\mu_{3/2}(2s_{1/2}v_{3/2}) = 0.53\) n.m. In the neighbouring odd-neutron nuclei, the \(f_{5/2}\) level accounts for the ground states of nuclei with mass numbers larger than those accounted for by the \(f_{3/2}\) level. A mixture of the configurations \(2s_{1/2}v_{5/2}\) and \(2s_{1/2}v_{3/2}\) with increasing amplitude of the configuration \(2s_{1/2}v_{5/2}\) with mass

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number would thus nicely reproduce the magnetic dipole moments connected by the line in the figure. Collective effects which would be significant for the lower mass numbers have so far not been considered.

The magnetic dipole moment of the $7^+$ isomeric state in $^{198}\text{Tl}$ was determined to be $0.64(8)$ n.m. This value, being a factor of ten larger than that obtained from an optical hfs measurement, is in good agreement with that expected from the configuration $7^+(\approx s_{1/2}^1I_{13/2}^1)$.

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Ground state nuclear magnetic moments of doubly-odd thallium isotopes.

| Isotope | $I/2$ | Measured spin $I$ | Measured hfs constant $|a|$ MHz | Measured nuclear g-factor $g^*_I$ | Nuclear dipole moment $\mu_I$ n.m. |
|---------|------|------------------|------------------|--------------------------|------------------|
| $^{193}\text{Tl}$ | 23 min | 1/2 | | | ±0.135(3) |
| $^{194}\text{Tl}$ | 33 min | 2 | 443(9) | 1.79(14)$\times10^{-3}$ | 1.65(12) |
| $^{195}\text{Tl}$ | 1.2 h | 1/2 | 228.8(6) | 1.79(14)$\times10^{-3}$ | 1.65(12) |
| $^{196}\text{Tl}$ | 1.8 h | 2 | 3.9499(6) | ±0.0012066(2) |
| $^{197}\text{Tl}$ | 2.8 h | 1/2 | 599(69) | ±0.640(74) |
| $^{198}\text{Tl}$ | 5.3 h | 2 | 1.63(10) |
| $^{198m}\text{Tl}$ | 1.9 h | 7 | 1.78(11)$\times10^{-3}$ | ±0.035685(3) |
| $^{199}\text{Tl}$ | 7.4 h | 1/2 | 116.814(8) | ±1.68(11) |
| $^{200}\text{Tl}$ | 26.1 h | 2 | 185.1(7) | ±0.0566(2) |
| $^{201}\text{Tl}$ | 73.5 h | 1/2 | 40.135(3) | 1.68(11) |
| $^{202}\text{Tl}$ | 12.4 d | 2 | 1.63(10) |

The electronic g-factor measured in $^{198}\text{Tl}$: $g_J(Tl) = -0.66578(4)$

*Previously measured
+ Evaluated using the relation $\mu_I = \mu_{1205} \frac{a_{I}}{a_{205}}$ (hfs-anomaly neglected)

$^*_{205} = 1/2$

$a_{205} = 21310.833946$ MHz (R.F. Lacey, Metrologia 3 (1967)70)

$\mu_{205} = 1.62754$ n.m. (I. Lindgren, Table of Nuclear Spins and Moments, in Alpha, Beta, and Gamma Ray Spectroscopy, Appendix 4, edited by K. Siegbahn, 1954).
MAGNETIC MOMENT OF THE 458 ns $^9_+ = 9^-$ ISOMER IN $^{200}\text{Pb}$


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The neutron-deficient even isotopes of Pb contain low-lying negative parity states which arise from the coupling of $|l^2_{13/2}|$ with $|p^2_{1/2}|$, $|f^2_{5/2}|$ and $|p^3_{3/2}|$ neutron quasiparticle- hole orbitals. If these states the $|l^2_{13/2}|$ $9^-$ states have the purest wavefunction (W.N. True, Phys. Rev. 168 (1968) 1258).

We have applied the standard time-differential perturbed angular distribution method to measure the $g$-factor of the 2227 keV, 480 ns $^9_+$ isomer in $^{200}\text{Pb}$. The isomer was populated and aligned by the $^{136}\text{Xe}^{(3,2n)}$ reaction using the 27 MeV pulsed beam from the Munich MP tandem accelerator. Liquid mercury targets enriched in $^{199}\text{Hg}$ were used to avoid relaxation of the nuclear alignment. Delayed x-rays were observed in two Ge(Li) spectrometers at $\pm 162^\circ$ and $-162^\circ$, respectively. The ratio of counting rates, $R(t) = (N_{162^\circ}) - N_{-162^\circ})$, for the combined data of the $7/2^- \to (5/2^-)$ cascade is shown in Fig. 1. The results which includes uncertainties in the Knightshift and the diamagnetic shielding factor is

$$g_{\text{exp}}[9^-]=-0.0285\pm0.0011$$

Using for the $g$-factor of the $\frac{7}{2}^{-}$ orbital

$$g[\frac{7}{2}^{-}] = 0.192\pm0.005$$

and assuming a pure configuration for the $9^-$ state, we calculate $g[\frac{7}{2}^{-}] = 0.292\pm0.014$. This value agrees with time-integral measurements of the $\frac{7}{2}^{-}$ $g$-factor in $^{207}\text{Pb}$, which may be averaged to $g[\frac{7}{2}^{-}] = 0.304\pm0.024$. The above interpretation may however be altered by small admixtures, particularly of $\frac{11}{2}^{-}$ configuration. Conversely, from the present experiment and from the time-integral value for $g[\frac{7}{2}^{-}]$ an upper limit of $\leq 1\%$ can be obtained for the intensity of this component in the wavefunction of the $9^-$ state.

**Figure 1**

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THE g-FACTORS OF THE 9+ AND 12+ ISOMERIC STATES IN 209Pb.

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The g-factors of the 9+ state (2.237 MeV, I=7/2+) and the 12+ state (13.1 MeV, I=23/2+) in 209Pb contain a sensitive dependence on the anomalous orbital contribution $\Delta g_\pi$ for the neutron and on additivity violations, respectively. These g-factors were measured by time-differential PAD in an external 10 kG field of an on-line superconducting magnet. The large magnetic field was essential for the measurement of the small g-factor of the 9+ state. The 9+ state in 209Pb was studied with the $^{197}$Au($^7$Li,4n)$^{200}$Pb reaction using a 4 MeV pulsed $^7$Li beam on a thick Au target. This reaction did not populate the 12+ state significantly. The 12+ state which was studied with the $^{197}$Au($^6$Li,4n)$^{200}$Pb reaction using a 14 MeV pulsed $^6$Li beam was populated only weakly. Two Ge(Li) detectors of 1% relative efficiency were positioned at 90° to the beam direction. A partial decay scheme including these levels is shown in Fig. 1. The 6.6-kMeV $\gamma$ ray ($h\nu=7\pi$) of the stretched cascade following the decay of the 9+ state was used for the 9+ g-factor measurement while for the 12+ state, the 777 keV cascade $\gamma$ ray (10.3-9.4) was used. The ratio data

$R(t) = \frac{[I(20.3\pi)-I(4.5\pi)]/I(4.5\pi)}{[I(10.3\pi)-I(4.5\pi)]/I(4.5\pi)}$ and the least squares fits are shown in Fig. 2. The resulting uncorrected g-factors are $g(9^+) = -0.030 \pm 0.003$ and $g(12^+) = -0.157 \pm 0.007$. The influence on the amplitude and phase of $R(t)$ for the 9+ state by the precession of the 9+ state (I=63 ms) has been carefully considered. The lack of knowledge about this phase makes the sign (not the magnitude) of the 9+ g-factor somewhat uncertain as indicated by the brackets. The estimated diamagnetic and Knight-shift corrections of 0.0510, 0.2 do not significantly change these results.

The 9+ state is believed to be of a pure (11/2$^-,\frac{5}{2}^+$)$^{12}$ neutron configuration with less than a 1% (11/2$^-,\frac{3}{2}^-$) configuration. This configuration is also consistent with the present g-factor measurement. The sensitivity of this result to $\Delta g_\pi$ stems from the fact that in $\langle 1\pi, 2\gamma \rangle$ and $\langle 1\pi, 3\gamma \rangle$ stretched configurations, the spin moments and the corresponding core polarizations are nearly cancelled, which leaves for two neutrons essentially only the $\Delta g_\pi$ contribution. Using first order core polarization calculations for the $1\pi, 2\gamma$ and $1\pi, 3\gamma$ neutron states, the neutron $\Delta g_\pi$ deduced from the present 9+ g-factor result is small, $\Delta g_\pi < 0.015$ in contrast to the larger value of $\Delta g_\pi = 0.151 \pm 0.02$ for the proton.

The present g-factor for the 12+ state in 209Pb which involves an ($1\pi, 2\pi$)$^{12}$ configuration is in excellent agreement with the 12+ state in 208Pb and the 13/2$^+$ state in 209Pb. The lack (<1%) of any violation of additivity, $g(J^+) = g(I^+)$, suggests that contributions from core-polarization blocking which depends on the number n of $11/2$ holes and any additional non-additive (J,v) state dependent effects are not observed.

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Fig. 1.  Fig. 2.
We have measured the g-factor of the $^{[3]}_{9/2}, ^{31}_{13/2}$, 10$^-$ particle hole state at 1.57 MeV in $^{208}$Bi. The derived magnetic moment of the $^{13}_{13/2}$ neutron hole in the $^{208}$Pb core will be discussed together with known values from neighbouring nuclei.

The g-factor was determined by the NMR-PAC method. The isomer was excited by the $^{208}$Pb(d,2n) reaction with a 18 MeV deuteron beam from the Bonn cyclotron which was pulsed with a pulse width of 150 μs and a repetition rate of 100 Hz. The target was kept molten at 375 °C. The perturbation of the angular distribution of the 321 keV γ-rays by the rf-field was observed in the first 600 μs with respect to the beam pulse because the relaxation time $T_1$ of the 10$^-$ state in liquid lead is expected to be much smaller than the lifetime ($T_2 \approx 300$μs, $T_1 = 3.61$ μs).

The resonance is shown in Fig. 1. After correction for diamagnetic and Knight shifts we obtain $g(10^-) = 0.2666(27)$. Using the value $g(3/2) = 0.314$ the additivity relation gives $\mu_{\text{exp}}(31/2) = -0.066(10)$ n.m. In Table 1 this value is compared to moments of other $^{13/2}$ states in $^{205}$Pb, $^{207}$Po, $^{206}$Pb and $^{208}$Bi (Ref. 1). Assuming mesonic corrections to be equal for all $^{13/2}$ states listed, differences between the moments can result from admixtures to the wavefunction $\delta\mu_{\text{adm}}$ and core polarization effects $\delta\mu_{\text{CP}}$. These corrections are defined by

$$\mu(31/2) = \mu_{\text{exp}} + \delta\mu_{\text{adm}} - \delta\mu_{\text{CP}}$$

where $\delta\mu_{\text{CP}}$ denotes the deviation of the core polarization compared to the $^{13/2}$ hole in the $^{208}$Pb core.

Fig. 1 NMR-PAC resonance of the 10$^-$ state in $^{208}$Bi. The rf-field parameters used were $\nu = 65.52$ kHz and $H_1 = 5.1$ G.
The main admixtures to the wavefunction of the $33/2^+$ state arise from couplings of the $f_{7/2}$ neutron hole state to the low lying collective states in $^{208}$Pb:

$$|33/2^+> = a|\nu_{13/2}^{-1} \cdot 0^+> + \sum_i b_i|\nu_{7/2}^{-1} \cdot i^+>$$

With this wavefunction one obtains approximately $\delta u_{adm} = (1-a^2) \mu(\nu_{13/2}^{-1})$. Hamamoto [2] and Klemt and Speth [3] obtain $a^2 = 0.93$ and 0.87, respectively. For the calculation of $\mu(\nu_{13/2}^{-1})$ in the table we assume therefore $\delta u_{adm} = -0.10$ n.m.

The difference in the core polarization correction arises mainly from the $3p_{1/2}$, $3p_{3/2}$, $1^+$ excitation which is only present in the nuclei $^{205}$Pb, $^{207}$Po, and $^{206}$Bi. We neglect the effects due to the different fillings of the $h_{11/2}$ and $\nu_{13/2}$ shells. With the wavefunction

$$|0^+, 206Pb> = a|\nu_{1/2}^{-2}> + b|\nu_{3/2}^{-2}> + c|\nu_{5/2}^{-2}> + ...$$

one obtains $\delta u_{CP} = (a - 1/2 b)2\mu'$, where $\delta u'$ is the core polarization correction neglecting pairing effects. Average values for the amplitudes from several published calculations are $a = 0.80$ and $b = 0.35$. The single particle core polarization value has been estimated in reference [5] as $\delta u' = 0.4$ n.m. Using 1.7 \"e\" for the excitation energy of the $1^+$ state in $^{205}$Pb we obtain $\delta u_{CP} = 0.06$ n.m.

The corrected moments are given in the last column of the table. We adopt the moment of the single $i_{13/2}^+$ neutron hole in the $^{208}$Pb core as $\mu(\nu_{13/2}^+) = -1.11 \pm 0.05$ n.m. This value is in good agreement with calculations of Bauer et al. [6] within the framework of the theory of finite Fermi systems. Using an effective moment operator including mesonic exchange effects they calculate the moment of the $i_{13/2}^+$ neutron hole state in $^{207}$Pb to be $\mu(\nu_{13/2}^+) = -1.14$ n.m.

This work was supported by the Bundesministerium für Forschung und Technologie.
Some 30 spontaneously fissioning isomeric states have been identified to date in nuclides ranging from $^{236}\text{U}$ to $^{245}\text{Bk}$. Because of the very large nuclear deformation associated with these states, a more detailed spectroscopic study can provide unique information about nuclear structure. An attempt has been made to measure the g-factors of such states in an external magnetic field. Due to the fact that the actinides are paramagnetic, also a study of the hyperfine fields is necessary before reliable interpretation of such measurements can be made.

Two fission isomeric states are known in $^{237}\text{Pu}$ with half-lives of 100 ns and 1.1 μs, respectively (see fig.1). The 1.1 μs state lies 300±50 keV above the shorter-lived isomer and is known to have the higher spin. The predominant decay of the 1.1 μs state is by direct fission rather than by gamma emission to the lower-lying state, as evidenced by the very different anisotropies for the two isomers $1.4\pm0.14$ and $0.58\pm0.16$ for the 1.1 μs and 100 ns isomers, respectively. The angular distribution data indicate that the long-lived and short-lived states decay via fission channels having $K<1$ and $K>1$.

The precession frequencies have been measured of the two states in a transverse magnetic field by observing the time-dependent angular distribution of delayed fission fragments. Results for the 1.1 μs state have been reported earlier. The plutonium nuclei were formed and aligned in the reaction $^{235}\text{U}(a,2n)^{237}\text{Pu}$ using a pulsed beam with $E_a=25$ MeV. The recoils were stopped in Pb to minimize perturbation effects from quadrupole fields. In view of the small (≈1 pb) cross section, sandwich targets consisting of alternate layers of 35 μg/cm$^2$ $^{235}\text{U}$ and 65 μg/cm$^2$ nat Pb were used. Altogether 40 layers of each were sputtered onto a Cu backing. Two Si surface barrier detectors were placed at 135° and 225° with respect to the beam direction, each subtending an angle of ±19°. Measurements were made with external magnetic field strengths ranging from 1.0 to 5.5 kGauss.

Fig.2 shows two examples of spin rotation patterns obtained in the same counter placed at 225° for the 1.1 μs state (upper part) and the 100 ns state (lower part). Since the $P^J(\cos \theta)$ terms having $\alpha>2$ contribute negligibly to the angular distributions, the observed frequencies are simply twice the respective Larmor frequencies $\omega_L$.

Using the relation $\omega_L = -\mu_B B_{\text{ext}}$ where $\mu_B$ is the paramagnetic correction factor, we obtain $g\cdot B = +1.3\pm0.1$ and $+0.41\pm0.03$ for the 100 ns and 1.1 μs states, respectively. The positive sign is in each case extracted from the phase of the oscillations and the knowledge of the unperturbed fragment anisotropies. Assuming that $B_{\text{ext}} \ll B_{\text{ff}}$ and that within the same $J$ the electronic level splitting $\Delta E \ll K_T$, the $\beta$ factor may be approximated as inversely proportional to the temperature of the target. With this in mind we have measured the temperature dependence of the spin rotation frequency in the case of the 1.1 μs isomer. The results are shown in fig.3.

Theoretical estimates of $\beta$ have also been made. In addition to the applied external field, the magnetic field at the nucleus is composed of an orbital part

$$B_{\text{orb}} = B_{\text{ext}} 2e\langle r^2 J \rangle \langle J \parallel N \rangle (J+1)/(3K_T) \quad (1)$$

plus a core polarization component

$$B_{\text{core}} = -B_{\text{ext}}(g_J^{-1})6.25\times10^{-5} \mu_B g_J(J+1)/(3K_T) \quad (2)$$

plus the Fermi contact part (which we shall neglect). The most probable ionic states of Pu in a Pb environment are $3^+$ and $4^+$. Assuming Russell-Saunders coupling and neglecting crystal field effects we calculate $\beta(3^+) = 1.36$ and $\beta(4^+) = 2.86$ at room temperature. It must be pointed out, however, that possible crystal field effects, which could in principle be rather important, would lower $\beta$.

Fig.3 shows the $\beta$-value calculated for $4^+$ ions to be in reasonable agreement with the experimental trend, although the data actually favour a somewhat lower value. The present method does not prove sufficiently sensitive to clarify whether crystal field effects are indeed negligible. We have assumed $\beta_{\text{room}} = 2.86$, obtaining $g = +0.44\pm0.05$ and $+0.14\pm0.02$ for the short- and long-lived states, respectively.

The anisotropies of the fission fragment angular distributions are readily extracted from the amplitudes of the observed oscillations, after correction for the finite solid angle of the detectors and the finite length of the beam burst. Values for the anisotropy...
$\frac{W(0^0)}{W(90^0)}$ of 0.4±0.1 and 2.5±0.4 are deduced for the short- and long-lived states, respectively. The true anisotropies are somewhat more pronounced since possible attenuation from perturbing extranuclear fields are neglected. Furthermore we have consistently observed that the oscillatory patterns in the decay curves disappear after a bombardment of approximately 500 µC of beam (beam diameter ≈2.5 mm). A very likely explanation is that the Pb cubic structure undergoes radiation damage and that crystal fields from the lattice begin to play an important role.

From the measured anisotropies we can conclude that $I > \frac{7}{2}$ for the 1.1 µs isomer and $5/2 < I < 9/2$ for the 100 ns isomer.

The absolute value and sign of the $g$-factor can be compared to the theoretical expectations

$$g = (1 - \frac{\chi^2}{\chi^{2} + 1}) g_R + \frac{\chi^2}{\chi^{2} + 1} g_n, \quad K \neq \frac{1}{2}$$

(3)

where $g_R = g_A <A> + g_A <S>$. Here $g_R$ is the collective $g$-factor and $g_n$ that associated with the unpaired particle, while $\alpha, \Lambda$ and $I$ are the Nilsson quantum numbers. Using values $g_R = 0.35 \pm 0.04$, $g_A = -0.06 \pm 0.02$ and $g_n = 0.7x g_n^\text{free}$, where $g_n^\text{free} = -3.82$, we see from equation (3) that $g > 0$ requires $<\alpha>$ to be negative, implying that the unpaired neutron spin and orbital angular moments are antiparallel ($\Pi = \frac{1}{2}$). This means that earlier speculations that the 1.1 µs isomeric state could be connected to either the [505]11/2- or the [615]11/2+ orbitals now seem ruled out. The theoretical $g$-factors for various values of $I$ are $g_\frac{5}{2} = +0.43 \pm 0.02$, $g_\frac{7}{2} = +0.32 \pm 0.02$ and $g_\frac{9}{2} = 0.25 \pm 0.02$.

The most likely spin for the 100 ns isomeric state combining the measured anisotropy and magnetic moment is 5/2. Recent single particle calculations by I.Hamamoto and W.Ogle$^6$ place the [633]5/2 state close to the Fermi level for 143 neutrons. For the 1.1 µs isomeric state the situation is at present uncertain. Since states with spin values 9/2 and 11/2 and $\Lambda = 5$ and 6, respectively, are predicted to lie 3 and 15 MeV above the Fermi surface, the highest spin value to the isomeric state is 7/2. The predicted magnetic moment for this spin is, however, twice as large as measured. An alternative explanation could be that the isomer has $K = \frac{1}{2}$ with a large negative decoupling parameter which would make the 7/2 member of the rotational band the lowest lying level. A theoretical investigation of the various possibilities is being carried out$^5$.

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6) I.Hamamoto and W.Ogle, to be published
The considerable amount of data accumulated for the last few years has shown that $2^+$ states of vibrational as well as transition nuclei have large static quadrupole moments. Most of these data have been obtained by using the reorientation technique based on Coulomb excitation cross section measurements. A new type of reorientation measurement has been developed recently, the precession technique. The principle is when during the Coulomb excitation process, the excited state is polarized, reorientation manifests itself as a precession of the angular distribution of the deexcitation gamma rays. This can be qualitatively understood as arising from the precession of the nuclear quadrupole moment $Q$, interacting with the strong electric field gradient of the incoming ion. It is therefore a first order effect, proportional to the quadrupole moment, and in principle insensitive to interference from higher order transitions. When the interaction strength is large, however, even this method becomes somewhat sensitive to higher order transitions. This sensitivity has been studied systematically for the even-even nuclei $^{148}$Sm, $^{150}$Sm and $^{152}$Sm, ref. 5.

The purpose of this work is to extend these investigations to odd-A nuclei, where it is very difficult to get reliable results from conventional reorientation measurements. The lowest two excited states in $^{103}$Rh were chosen for this investigation. They are $3/2^+$ at 295 keV and $5/2^+$ at 358 keV. Since these states can be thought of as being a $2p_{1/2}$ particle coupled to the $2^+$ core state of $^{102}$Ru, we have also measured the quadrupole moment of this state.

The experiment consisted of measuring the symmetry angle, $\theta_s$, of deexcitation gamma rays after excitation by particles scattered through 90°. This must be done with high precision. The technique used is the same as that described in ref. 4, except for the addition of an extra pair of gamma counters, each $\pi/4$ degrees outside of the original pair. The experimental value of $\theta_s$ is then compared to the symmetry angle derived from the output of the de Boer-Winther Coulomb excitation program. The value of $M_{22}$ is then varied until $\theta_s$ is matched.

While the reorientation precession technique is less sensitive to interference from higher order transitions than the standard technique, a knowledge of the predominant transition matrix elements is essential. It has been shown that, at least for even-even nuclei, at low bombarding energies, the symmetry angle is rather insensitive to the inclusion of higher excited states. In such a case only the inclusion of the $4^+$ state has a noticeable influence. The inclusion of quantum mechanical corrections for $\eta$-values encountered with an $^{16}$O beam, can lower the quadrupole moment by 10-20%. At the present state of this investigation, no quantum mechanical corrections have been carried out. Neither has the transient electric field been taken into account; this correction is calculated to be small (<0.1°). The $^{102}$Ru enriched target was prepared by sputtering on 2 mg/cm$^2$ thick Cu; it was 0.1 mg/cm$^2$ thick. The $^{103}$Rh target was prepared from natural Rh by rolling. The $^{16}$O beam energy used in both cases was 45 MeV. In the case of the thick Rh target, the effective energy range was 32-45 MeV. Several digital windows were set on the particle spectrum, and gamma spectra coincident with each window were taken. Since the statistical accuracy of the data coming from each window was rather low, we quote here only a weighted average of all windows.

The Coulomb excitation of even Ru nuclei has been investigated by McGowan et al. 7; states up to $4^+$ have been found and most matrix elements have been deduced. These matrix elements have been used with signs corresponding to the vibrational model. The experimental value of the symmetry angle for $^{102}$Ru is found to be $\theta_s = 39.16 \pm 0.22$ which yields a quadrupole moment of $Q = -1.06 \pm 0.4$. This should be compared to $Q = -0.37 \pm 0.24$ obtained by Nolan et al. 8 who used a conventional reorientation technique. With the opposite sign of the interference term, our result becomes $Q = -0.99 \pm 0.4$.

The $^{103}$Rh nucleus has a fairly complex structure with both even and odd parity states. In Coulomb excitation the latter are populated exclusively. The negative parity levels can be approximately described by core excitation. The $^{102}$Ru core is coupled to a $2p_{1/2}$ proton, therefore the $2^+$ core state is split into a $3/2^-$, $5/2^-$ doublet. The center of gravity of the doublet does not coincide with the $2^+$ state and its splitting is rather large.
(134 of the $2^+$ energy), therefore the core-particle coupling is hardly weak. The negative-parity states of $^{103}$Rh have been studied by Coulomb excitation by Sayer et al.\textsuperscript{11}. Six excited states have been populated and ten off-diagonal matrix elements have been determined. The matrix element $M_{23}$ between the doublet states is unfortunately missing; this will introduce ambiguities in the final interpretation of the data. The measured symmetry angles are $\theta_3 = 40.0^\circ \pm 0.20$ for the $3/2^-$ level and $\theta_5 = 38.69^\circ \pm 0.20$ for the $5/2^-$ level. If we assume $K_{23} = 0$, the values of $Q$ are $Q_{3/2} = -0.57^\circ \pm 0.4$ and $Q_{5/2} = -1.65^\circ \pm 0.4$. Preliminary experiments on the angular distribution of the 5/2-3/2, 63 keV gamma rays indicate that the transition is not purely M1. Since the $Q$-values, especially for the 3/2 state, are rather sensitive to $M_{23}$, it is possible that the values given above should be adjusted following an accurate measurement of this matrix element. Sign ambiguities of some matrix elements can also lead to further uncertainties of $Q$. These signs are not always unequivocally defined by fitting cross sections and henceforth model depend criteria are sometimes used. Subject to these large uncertainties, it appears that the sequence $Q_{3/2} < Q_{\text{core}} < Q_{5/2}$ is followed, in agreement with de Shalit's weak coupling model\textsuperscript{10}.

In the figure the calculated symmetry angle is shown as a function of bombarding energy. Two sets of curves are shown for both the $3/2^-$ and the $5/2^-$ level. One set is calculated using a three level system while the other includes seven levels. As can be seen, in this case one must include the higher excited states to get a reliable result. So we must conclude that while it is possible to measure odd-A nuclei by this method, great care must be taken with respect to inclusion of higher excited states in the calculations.

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III. 30 MEASUREMENT OF Q(2+) IN 110Pd USING THE REORIENTATION PRECESSION TECHNIQUE

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Static electric quadrupole moments are determined by observing their interaction with an electric field gradient. Electric field gradients of the magnitude $10^{33}$ V/cm are produced in Coulomb excitation when the charged particle is passing near the nucleus and is deflected. Furthermore, the electric field gradients, produced in this way, can easily be calculated. The time of interaction is however very short ($<10^{-15}$ s).

Breit et al. (1) considered the reorientation of the nuclear axis caused by the electric field of the bombarding particle after it had excited the nucleus. The change in the nuclear spin direction affects the angular distribution of the gamma rays. This approach for determining electric quadrupole moments of excited states, hereafter referred to as the reorientation precession technique, was successfully applied to the first $2^+$-states of $^{150}$Sm and $^{197}$Au by Grodzins et al. (2). All earlier methods were based on a second-order effect in the differential cross-section in Coulomb excitation. These methods have been of great importance for the measurements of the quadrupole moments of the first excited $2^+$-states in nuclei. They are referred to as the reorientation technique and have been reviewed in a number of articles (3-7). The reorientation precession technique, however, is a first order effect, and moreover it is less sensitive to interferences involving higher order transitions than the conventional reorientation technique. We have applied the reorientation precession technique in the measurement of the quadrupole moment of the first excited state in $^{110}$Pd.

The targets have been prepared by rolling enriched (97%) $^{110}$Pd on a 10 mg/cm$^2$ Cu-backing. The use of a Cu target will prevent perturbations of the gamma ray distribution as the recoiling nuclei are stopped in the backing material. The experiment was performed using 160 ions accelerated in the Van de Graaff at the Uppsala Tandem Laboratory. The effective energy of the 30 KeV $^{16}$O beam was found to be 28.8 KeV. This energy is equivalent to a distance of closest approach of 24.5 fm, and will thus be a safe bombarding energy ($\gamma$).

Our focusing and collimator systems give a spot with a diameter of 1 mm on the target and the current during this experiment was in the order of 25 nA ($\mu$A).

As a result of the present measurement the following ratio of the number of coincidences between the two combinations of detectors was obtained:

$$ R = \frac{N(22.5^\circ)}{W(157.5^\circ)} = 1.402 \pm 0.015 $$

The error is a pure statistical error representing one standard deviation, plus an estimated geometrical error.

The theoretical ratios as a function of $M_{22}$ were calculated using the de Boer-Winter program (8). Six levels were taken into consideration in this analysis. The matrix elements were taken from $B(E2)$-values measured by Robinson et al. (9). The theoretical ratio of the number of coincidences between the two combinations of detectors as function of $M_{22}$ for 28.8 MeV $^{16}$O in $^{110}$Pd is shown in fig. 2.

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Fig. 1 A schematic cross-section of the experimental setup showing the gamma-ray distribution $(2^+\rightarrow 0^+)$ for particles scattered 90° in the LAB-system. The precession of the gamma-ray distribution due to the interaction between a field gradient and a negative quadrupole moment of the nucleus is indicated in the figure.

As a result of the present measurement the following ratio of the number of coincidences between the two combinations of detectors was obtained:

$$ R = \frac{N(22.50)}{W(157.50)} = 1.402 \pm 0.015 $$

The error is a pure statistical error representing one standard deviation, plus an estimated geometrical error.

The theoretical ratios as a function of $M_{22}$ were calculated using the de Boer-Winter program (8). Six levels were taken into consideration in this analysis. The matrix elements were taken from $B(E2)$-values measured by Robinson et al. (9). The theoretical ratio of the number of coincidences between the two combinations of detectors as function of $M_{22}$ for 28.8 MeV $^{16}$O in $^{110}$Pd is shown in fig. 2.

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Fig. 2 $W(22.50)$ as a function of $W(157.50)$ for 28.8 MeV $^{16}$O on $^{110}$Pd scattered 90° in the LAB-system. The matrix elements used in the calculation are given in table 1. The experimental ratio (with errors) is marked in the figure and the experimentally deduced values of $M_{22}$ can be read.
The maximum interferences have been taken into account by putting the values for $M_{13}$ and $M_{15}$ either both positive or both negative. This figure is used to determine $M_{22}$ from our experimental ratio. The result is

$$M_{22} = 0.69^{\pm}0.25$$

for $M_{13} = +.11$ and $M_{15} = +.06$

$$M_{22} = 0.74^{\pm}0.25$$

for $M_{13} = -.11$ and $M_{15} = -.06$

The quantum correction is in this case small and has not been applied. Influences of uncertainties in the matrix elements used in the analysis have been examined. As a result we find that there is no influence on the ratio from $M_{13}$ and $M_{15}$ but a minor influence from $M_{12}$ of 0.1%.

A summary of the experimentally determined values for the static electric quadrupole moment of the first excited 2$^+$-state in $^{110}$Pd is given in table I. The error in the quadrupole moment deduced from the present measurement is not fully representative for the reorientation precession technique. Under more favorable experimental conditions this error can be decreased by a factor of at least two. Due to the large error there is an agreement with both ref. 10 and ref. 11 even though these values do not agree with each other. As we have mentioned earlier interferences from excitations involving higher excited states have greater influence when "conventional" techniques are applied for the determination of quadrupole moments than when the reorientation precession technique is used. This is clearly demonstrated in table I.

| Table 1. Experimentally determined values of the static electric quadrupole moment for the first excited 2$^+$-state in $^{110}$Pd. |
|------------------|------------------|------------------|
|                  | $Q(2^+, 110_{Pd})$ constructive interference | $Q(2^+, 110_{Pd})$ destructive interference |
| Ref. 9           | -.83±.19 eb      | -.45±.08 eb      |
| Ref. 10          | -.72±.08 eb      | -.26±.049 eb     |
| Ref. 11          | -.48±.049 eb     | -.36±.12 eb      |
| Mean ref. 7      | -.60±.12 eb      | -.36±.12 eb      |
| This work        | -.56±.20 eb      | -.52±.20 eb      |

A more detailed report on the present measurement is given in ref. 12.

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12. L. Hasselgren, F. Falk, B.S. Chumman, C. Fahlander and J.E. Thun, UUIP-839
An increasing number of measurements of the quadrupole interaction of excited states in non-cubic metals, performed by γ-γ angular correlations and perturbed angular distribution experiments following nuclear reactions were reported in the past few years ([1,2] and Refs. therein). Generally, the interpretation of the measured interaction frequencies suffers from the fact that the nuclear quadrupole moment and the electric field gradient (efg) at the site of the impurity are both poorly known.

We have measured the quadrupole interaction of the 11/2−, 89 ns excited state of 113Sn in metallic Cd time-differentially following the reaction 112Cd(α,4n). In this case, the efg for the system SnCd can be estimated with some reliability. Systematic investigations of quadrupole interaction of impurities in Cd and In metal suggest the conclusion that the efg at the site of the impurity is essentially caused by the host metal if mass radius and charge of the impurity are similar to those of the host [1,3,4]. Thus using the efg for the system 117InCd the quadrupole moment of the 11/2− state in 113Sn can be deduced from our measurement.

The experiment was performed at the Stockholm 225 cm cyclotron with a pulsed beam of 51 MeV α-particles to populate and align the isomeric state. A fast external pulsing system skipped four out of five of the natural beam bursts of the cyclotron thus providing pulses of 3 ns width and 562 ns repetition time. The target was a 100 mg/cm² foil of isotopically enriched polycrystalline Cd metal, which could be heated to reduce radiation damage effects. Time spectra of the 661 keV γ-rays were detected by two NaJ(Tl) scintillators at a target temperature of about 500K. The result is shown in Fig. 1.
The theoretical curve is calculated with the expression
\[ G_{22}(t) = \sum_{n} \frac{S_{2n}}{2n} \cos(n \omega q t), \quad \omega q = \frac{e^2 q}{\hbar n^2 (21-2)} \]
for the time dependent attenuation factor \[5\], thus assuming a uniform axially symmetric field gradient and \[ K_{\text{max}} = 2 \] for the tensor degree of alignment. The quadrupole coupling constant obtained is \[ e^2 q / \hbar = 95(2) \text{MHz} \]. From the undisturbed structure of the measured pattern, it must be concluded that a large amount of the recoil nuclei was stopped at substitutional sites with the nearest neighbours remaining undisturbed.

The measured temperature dependence \[7\] and band structure calculations \[6\] of the efg in Cd-metal both lead to the conclusion that the efg is essentially determined by the conduction electron part. Thus possible differences in the antiscreening factor for In and Sn in Cd, acting on the lattice part of the efg, have only a small influence on the total efg. Assuming that the band structure contributions at the Sn- and In-site in Cd metal are equal within 20% (this uncertainty may be caused by different charge screening effects), the quadrupole moment of the \(11/2^+\) state in \(^{113}\)Sn follows as \[ Q' = 0.48(10) \text{ b} \]. Here the efg of \(^{117}\)In \((3/2^+\) state, \[ Q = 0.63(4) \text{ b} \) in Cd \[14\] was used.

The \(11/2^+\) state in \(^{113}\)Sn, as well as other \(2J + 1/2\) states in the Cd, Sn and Te isotopes, are considered to be rather pure single quasiparticle states \[1\]. Quadrupole moments of \(2J + 1/2\) states are measured or can be calculated from B(E2) values in the isotopes \(^{111,113,115}\)Cd \[8\], \(^{116,118}\)Sn \[9\] and \(^{119}\)Sn \[11\], ranging from \(-0.85 \text{ b}\) for \(^{115}\)Cd to \(-0.13 \text{ b}\) for \(^{119}\)Sn. Therefore the sign of the quadrupole moment of the \(11/2^+\) state in \(^{113}\)Sn can be assumed negative. In terms of the nuclear shell model this trend can be interpreted as due to the successive filling of the \(h_{11/2}\) neutron shell, the quasiparticle and \(P\)-force contribution to the quadrupole moment being proportional to the quantity \(U_1^2 - U_0^2\) \[11\]. \(U_1^2, U_0^2\) are the probabilities for occupation and nonoccupation of the \(\frac{1}{2}\) shell respectively. Taking experimental values of \(U_1^2\) from spectroscopic factors \[12\] and keeping all other parameters constant, the trend of the quadrupole moments of the states in the aforementioned Sn isotopes can be reproduced within the error bars of the experimental values. Because the \(Q\)-value of \(^{113}\)Sn fits well into the given systematics, the crucial assumption that the efgs of SnCd and InCd are equal within 20% is strongly supported.

Two of us (F.D. and D.R.) gratefully acknowledge the support of the Deutsche Forschungsgemeinschaft (SFB 161).

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For a nuclear shell model orbit of angular momentum $j$ and seniority $\Omega=1$ the quadrupole moments $Q(j^n)$ depend strongly on the number of particles $n$ occupying this state: $Q(j^n) = \frac{2j+1-2\Omega}{2j}Q(j)$, where $Q(j)$ is the single particle quadrupole moment. The known $^{4}\gamma$-factors indicate that the $5^-$ states in $^{116}$Sn, $^{118}$Sn, and $^{120}$Sn are rather pure $(h_1^{11/2} s_1^{5/2})5^-$ neutron configurations, where according to the shell model $n$ takes the values $n=1, 3, 5$ for $^{116}$Sn, $^{118}$Sn, and $^{120}$Sn, respectively. Configuration mixing in these states is probably small since the Sn-isotopes have the magic proton number $Z=50$ and since the $h_1^{11/2}$ neutron orbit has opposite parity to the other orbits within the major shell $50 \leq N \leq 82$. Therefore the above relation can be tested by measuring the quadrupole moments of the $5^-$ states in the even Sn-isotopes. The known $^{4}$ quadrupole moment of the 23 keV state in $^{119}$Sn together with a recent determination $^{5}$ of the quadrupole interaction frequency of $^{119}$Sn in Sn-metal provides a calibration of the effective field gradients at the Sn-site for such investigations.

In our experiment enriched $^{118}$Sn and $^{116}$Sn foils were irradiated with 18 MeV deuterons in order to produce the radioactivities $^{118m}$Sb and $^{116m}$Sb directly in Sn environment.

In figs. 1 and 2 the spin rotation curves obtained by time differential perturbed angular correlation technique using suitable $^1_1$-cascades via the $5^-$ states in $^{118}$Sn and $^{116}$Sn are shown. The solid lines are least squares fitted curves of the theoretical perturbation function $G_2(t)$ to the experimental data. In the case of $^{116}$Sn the points near $t=0$ have been omitted in the fits due to disturbances due to prompt background. The quadrupole frequencies $\omega_0 = \frac{3eQV_z^2}{2I(2I-1)}$ were fitted as $\omega_0(5^-, ^{116}$Sn$) = 2.62 \pm 0.007$ MHz and $\omega_0(5^-, ^{118}$Sn$) = 1.63 \pm 0.11$ MHz. The derived quadrupole moments for $^{116}$Sn and $^{118}$Sn are given in the table.

The second value for $^{118}$Sn and the value for $^{120}$Sn were determined $^{6,7}$ from quadrupole rotation experiments in $(K(C_4H_8Sb(OH_2))1/2 H_2O)$. The agreement of the two experimental quadrupole moments for $^{118}$Sn indicates that the calibration of the electric field gradient in this insulator is reliable. The errors quoted for the absolute moments are due to the uncertainty.
of the quadrupole moment of the 23 keV state in $^{119}$Sn used for the field gradient calibration. The errors of the ratios of the quadrupole moments are given by the statistical accuracies.

The experimental quadrupole moments agree with the single particle estimates where an effective charge $q_{\text{eff.}}^n = e$ was assumed. However, from the $B(E2, \gamma^+ - 5^-)$ transition probabilities in the even Sn-isotopes it is known that pairing correlations are important. The influence of the pairing effects on the quadrupole moments were investigated using the parameters of Ref. 8) for $h_{11/2}$ states in the odd Sn-isotopes. The theoretical values for the quadrupole moments including the pairing correlations are slightly smaller than the single particle estimates but are still in agreement with the experiments. We conclude that the quadrupole moments in the even Sn-isotopes show the expected systematic variation with the number of neutrons in the $h_{11/2}$ shell model orbit.

Table: Quadrupole moments in $^{116}$Sn, $^{118}$Sn, and $^{120}$Sn

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$Q_1$</th>
<th>$2j+1-2n$</th>
<th>Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{116}$Sn</td>
<td>$0.165(60)$</td>
<td>1.0</td>
<td>-0.235</td>
</tr>
<tr>
<td>$^{118}$Sn</td>
<td>$0.10(4)$</td>
<td>0.6</td>
<td>-0.140</td>
</tr>
<tr>
<td>$^{120}$Sn</td>
<td>$0.094(11)$</td>
<td>0.5</td>
<td>-0.426</td>
</tr>
</tbody>
</table>

References:
2) Cooperation of the angular correlation groups of Bonn and Hamburg; Nucl. Phys. 89(1966) 505
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III. 33 ELECTRIC QUADRUPOLE INTERACTION OF $^{206}\text{Po}(8^+)$ AND $^{208}\text{Po}(8^+)$ STATES IN HEXAGONAL THALLIUM

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Sektion Physik der Universität and *Technische Universität, München

The nuclei near the $^{208}\text{Po}$ doubly closed shell provide an excellent testing ground for the usefulness of the effective charge concept. Measurements of E2 transition probabilities indicate that rather large changes of the E2 effective charge occur in nuclei with changing neutron or proton number (J. Blomqvist, J. Phys. Soc. Jap. 34 (1973) 223). It appears then to be of some interest whether or not similar observations can be made on static nuclear quadrupole moments in this mass region.

We have applied the time-differential perturbed angular distribution method to study the quadrupole interaction of the 212 ns $^{206}\text{Po}(8^+)$ and 380 ns $^{208}\text{Po}(8^+)$ isomers in hexagonal Tl. The isomeric states were populated and aligned by the $^{203,205}\text{Tl}(^7\text{Li},4n)$ reactions using the 36 MeV pulsed $^7\text{Li}$ beam from the Munich MP tandem accelerator. The natural Tl-target of 75 mg/cm$^2$ thickness was vacuum evaporated onto a copper backing cooled to 77$^\circ$K. Deexcitation gamma-rays of the stretched $E2$ $6^+\rightarrow4^+\rightarrow2^+\rightarrow0^+$ cascades for both isomers were clearly resolved in two Ge(Li) spectrometers located at $\theta = 0^\circ$ and $90^\circ$, respectively. The ratio of counting rates, $R(t)=W(0^\circ)/W(90^\circ)-1$, measured at three different temperatures, is shown in Fig. 1.

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The time distributions could be fitted well under the following assumptions:

a) the interaction in hcp Tl is characterized by a randomly oriented, axially symmetric field gradient and a corresponding undamped frequency component $\omega_0$.

b) in all cases a second, considerably faster frequency component, $\omega_1 \sim 2.2 \omega_0$, with a Gaussian frequency distribution was required to fit the data. This component, whose intensity increases rapidly at lower temperatures (~70% at T=77°K and ~20% at T=273°K) may be attributed to interstitial lattice positions of $^{115}\text{In}$ in Tl.

The undamped frequency component, $\omega_0$, is found to depend strongly on temperature, $((\omega_0(T=273°K)-\omega_0(T=293°K))/\omega_0(T=293°K)) \sim -0.3$. The magnitude of the electric field gradient, $V_{zz}$, which corresponds to $\omega_0$, can be obtained after estimating $Q(\delta^+)$ from observed $E2$ transitions in Po isotopes. Using the values of $Q(\delta^+)$ derived in Table 1 from the $8^+\rightarrow 6^+$ transitions, we calculate $V_{zz}(373°K) \sim 0.5 \times 10^{-17} \text{V/cm}^2$.

The ratio of quadrupole moments obtained in the present experiment, $|Q(206\text{Po},\delta^+)/Q(208\text{Po},\delta^+)| = 1.3\pm0.1$, compares favorably with the value, 1.34\pm0.03, obtained from the $E2$ transition rates. This agreement suggests that the effective charge is a valid concept for both $E2$ transitions and static quadrupole moments in the $(h_9/2^+)^2$ states of $^{206,208}\text{Po}$.

Table 1

Quadropole Moments in Po-Isotopes from E2-Transitions
Assuming $(h_9/2^+)^2$ Quasiparticle Configurations

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Transition</th>
<th>$E_\gamma$ (keV)</th>
<th>$T_{1/2}$ (ns)</th>
<th>B(E2) ($e^2\text{fm}^4$)</th>
<th>$Q(\delta^+)$ (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{210}\text{Po}$</td>
<td>$8^+-6^+$</td>
<td>83.7</td>
<td>110±8</td>
<td>74±6</td>
<td>0.53</td>
</tr>
<tr>
<td></td>
<td>$6^+-4^+$</td>
<td>46.5</td>
<td>38±5</td>
<td>247±33</td>
<td>0.62</td>
</tr>
<tr>
<td></td>
<td>$4^+-2^+$</td>
<td>245.3</td>
<td>1.8±0.2</td>
<td>285±32</td>
<td>0.55</td>
</tr>
<tr>
<td>$^{208}\text{Po}$</td>
<td>$8^+-6^+$</td>
<td>a)</td>
<td>380±10</td>
<td>106</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>$6^+-4^+$</td>
<td>177</td>
<td>4</td>
<td>469</td>
<td>0.78</td>
</tr>
<tr>
<td>$^{206}\text{Po}$</td>
<td>$8^+-6^+$</td>
<td>a)</td>
<td>212±5</td>
<td>189</td>
<td>0.85</td>
</tr>
</tbody>
</table>

a) assumed to be between 3.85 keV and 13.81 keV, the binding energies for $M_2$ and $L_3$ electrons
IV HYPERFINE INTERACTION IN
THE CONDENSED STATE
IV. 1 MAGNITUDE AND DIRECTION OF HYPERFINE FIELDS FOR A Cd IMPURITY IN Ni AT LOW AND HIGH MAGNETIZING FIELDS

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Methods for determination of hyperfine fields at impurities in ferromagnets can be classified as zero external field and applied field methods. Most Mössbauer and many DPAC and NMR experiments belong to the first category, whereas NO and time-integral perturbed angular correlation (IPAC, IMPAC) require a magnetically polarized sample. Recent NO-data on impurities in Fe[1] indicate that very high external fields are necessary for saturation of the effective field at the impurity. To investigate if this is a general phenomenon we have undertaken a detailed study of Cd impurities in Ni by the DPAC-method using applied fields in the range 0-10 kG. In DPAC there are possibilities to extract the spatial distribution as well as the magnitude of the local field. For Cd in Ni the hyperfine field is comparable in magnitude to applied fields available in the laboratory.

The measurements were made at room temperature using a conventional two-channel apparatus. External fields were obtained from an electromagnet and applied 90° to the detector plane. The field strength was measured with a small test coil and a fluxmeter. This assembly was calibrated to a nuclear resonance arrangement. The overall accuracy of the field measurements corrected for inhomogeneity, was found to be better than 2%.

Coincidences from the 173-247 keV γ-γ cascade in the radioactive decay of 111In to 111Cd were detected at 180° and 90° and \( R(t) = \frac{G(0,1)}{G(1,0) + G(2,0)} \) (1) was calculated. The 111In activity was obtained by irradiating natural Cd with 50 MeV protons in the synchrocyclotron at GWI, Uppsala. After chemical separation the In activity was reduced on a Ni foil by hydrogen gas and the sources were melted and cooled to small spheres of polycrystalline Ni. The sphericity was better than 3%.

The perturbation factor for a local magnetic field at the nucleus making the angle \( \beta \) to z-axis of the lab system is given by

\[
G_{k_k}^{\text{MM}} = G_{k_k}^{\text{MM}} = \left( \frac{2k_k!}{(2l+1)!} \right)^{\frac{1}{2}} \sum_{n-p} \left( \frac{\bar{I}}{n-p} \right)^{(l)} \left( \frac{\bar{I}}{n-p} \right)^{(l)} \frac{B_{k_k}^{(k_k-1)}}{B_{k_k}^{(k_k-1)}} \exp \left[ i \frac{\pi}{2k_k} (E_n - E_0) t \right]
\]

where \( E_n - E_0 = (n-n')\omega_B = P w_B \) and \( \omega_B \) is the Larmor frequency. The product of the \( B_{k_k}^{(k_k-1)} \) functions can be written as

\[
B_{k_k}^{(k_k-1)} = B_{k_k}^{(k_k-1)} \left( \frac{k}{\omega_B} \right) \left( \frac{k}{\omega_B} \right) \left( \frac{k}{\omega_B} \right) \left( \frac{k}{\omega_B} \right)
\]

The perturbation factor then reduces to

\[
G_{k_k}^{\text{MM}} = \sum_{n-p} \frac{B_{k_k}^{(k_k-1)}}{B_{k_k}^{(k_k-1)}} \exp \left[ -i P \omega_B t \right]
\]

For the geometry above, \( R(t) \) is given by

\[
R(t) = \frac{2B_{2a}^{2a} + (B_{22}^{22} + B_{2a}^{2a}) \cos(\omega_B t) + (B_{22}^{22} + B_{22}^{22}) \cos(2\omega_B t)}{\sqrt{3}a_{2a}^2 + 2B_{2a}^{2a} \cos(\omega_B t) + 2B_{22}^{22} \cos(2\omega_B t)}
\]

If the local field displays a normalized spatial probability density \( f(\omega_B \beta) \) the Legendre polynomials in (3) may be replaced by

\[
R(\beta) = \int f(\omega_B \beta) \cos(\omega_B \beta) \sin \beta \ d\beta
\]

The ratio of the intensities, of the single and double frequency components in the nominator of eq (5), will vary from 1, for random distribution, to 0, for a polarized sample. This can be seen in Figs. 1a, b, c. The experimental R-values were LSQ-fitted to (5). The Larmor frequencies obtained are plotted in Fig. 2 versus applied field. Below the saturation of a Ni-sphere (2.2 kG), \( \omega_B \) is constant and above that \( \omega_B \) decreases with applied field. Points above 3 kG were fitted to a straight line whose slope agrees with the earlier measured g-factor, -0.318(7) [2]. Using the convention of Shirley et.al. [3] this g-factor and the zero field value of \( \omega_B \), gives a
local field \((B_{hf} + B_{Lorentz})\) of \((-)164(2)\) kG. The constancy in the unsaturated region can be explained by shielding effects. In the saturated region the local field is given by
\[
B_{local} = B_{ext} - DM + B_{Lorentz} + B_{hf}
\]
(7)

The Lorentz field for cubic symmetry is \(+ \frac{1}{3} M\), \((2,00 \text{ kG})\) and the demagnetisation factor for a sphere is \(1/3\), hence \(B_{local} = B_{ext} + B_{hf}\). Extrapolation to \(B_{ext} = 0\) gives a field, which agrees with the classical Lorentz field. Corrections due to a contact interaction [4] at the impurity site therefore seem to be small in this case.

The spatial distribution of the local fields for some applied fields are given in table I. \(B\) is a cone angle fitted to (5) assuming a discrete angle. \(\beta^I_{\max}\) resp. \(\beta^{II}_{\max}\) denote values obtained by assuming the distribution of angles:
\[
I \left\{ \cos \beta \right\} = \begin{cases} 
\frac{1}{2} \left( 1 - \cos \beta \right) & \beta \leq \beta_{max}^I \\
0 & \beta > \beta_{max}^I
\end{cases}
\]
\[
II \left\{ \cos \beta \right\} = \begin{cases} 
\frac{2(\cos \beta - \cos \beta_{max}^I)}{\left( 1 - \cos \beta_{max}^I \right)^2} & 0 \leq \beta \leq \beta_{max}^I \\
0 & \beta > \beta_{max}^I
\end{cases}
\]

The same \(\chi^2\) were obtained for the three different models. Comparing these results with those predicted by Aharoni, [5] for the case of \(W\) in \(Ni\) \((-27^\circ\) for \(700 \text{ G}\) above saturation), we conclude that the magnetostriction-effects in our case are quite small and negligible above \(B_{ext} = 3 \text{ kG}\) although the difference in metallic radii [6] between \(Ni\) and \(Cd\) is greater than between \(Ni\) and \(W\).

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5. A. Aharoni, in Hyperfine Interactions in Excited Nuclei, (ed) Goldring, Kalish
IV. 2 Shift of NMR-ON resonances with an external magnetic field

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and G. Eska, Zentralinstitut für Tieftemperaturforschung, Garching

The NMR frequencies of radioactive impurities in ferromagnetic Fe and Ni lattices have been measured as a function of the applied external magnetic field. It could be shown that especially in IrNi systems the simple additive superposition of the hyperfine field and the external field seems not to be fulfilled.

Introducing the parameter \( t \) , the resonance condition is given by

\[
\hbar \nu = g \mu_B \left( H_{HF} + H_O (1 + t) \right)
\]

\( t \) depends on the host lattice and on the type of the impurity. A compilation of our experimental data is given in the table below.

Especially in the IrNi systems \( 1 + \frac{t}{t} \) deviates significantly from 1.0. To exclude systematic errors connected with the source geometry and the field calibration a source with \( ^{60}\text{Co} \) and \( ^{191}\text{Ir} \) was used. The resonance shift of \( ^{60}\text{CoNi} \) and \( ^{191}\text{IrNi} \) was thus measured simultaneously. The corresponding values are denoted with an asterix in the table. Typical resonance curves are shown in fig. 1.

The relatively large deviation of \( 1 + \frac{t}{t} \) for \( ^{181}\text{ReFe} \) is probably connected with the fact, that the resonance was lying on the tail of a strong resonance of the rf circuit.

The value of 0.948 for \( ^{105}\text{RhFe} \) is a subject of further investigations.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Number of spectra</th>
<th>Hyperfine splitting constant (MHz)</th>
<th>Frequency shift (MHz/kG)</th>
<th>( t = \frac{t}{t} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{60}\text{CoFe} )</td>
<td>256</td>
<td>165.957 ± 0.005</td>
<td>taken for calibration</td>
<td>1.0</td>
</tr>
<tr>
<td>( ^{60}\text{CoNi} )</td>
<td>86</td>
<td>69.231 ± 0.010</td>
<td>-0.591 ± 0.008</td>
<td>1.032 ± 0.015</td>
</tr>
<tr>
<td>( ^{58}\text{CoCo} )</td>
<td>64</td>
<td>124.979 ± 0.009</td>
<td>-0.576 ± 0.008</td>
<td>1.005 ± 0.010</td>
</tr>
<tr>
<td>( ^{52}\text{MnFe} )</td>
<td>67</td>
<td>441.726 ± 0.036</td>
<td>-1.496 ± 0.023</td>
<td>0.992 ± 0.015</td>
</tr>
<tr>
<td>( ^{198}\text{AuFe} )</td>
<td>69</td>
<td>88.700 ± 0.015</td>
<td>-0.359 ± 0.007</td>
<td>0.952 ± 0.024</td>
</tr>
<tr>
<td>( ^{182}\text{ReFe} )</td>
<td>72</td>
<td>239.857 ± 0.012</td>
<td>-0.207 ± 0.007</td>
<td>0.920 ± 0.040</td>
</tr>
<tr>
<td>( ^{184}\text{ReFe} )</td>
<td>20</td>
<td>231.34 ± 0.06</td>
<td>-0.318 ± 0.025</td>
<td>1.046 ± 0.085</td>
</tr>
<tr>
<td>( ^{181}\text{ReFe} )</td>
<td>39</td>
<td>402.54 ± 0.07</td>
<td>-0.622 ± 0.039</td>
<td>0.976 ± 0.080</td>
</tr>
<tr>
<td>( ^{191}\text{IrNi} )</td>
<td>91</td>
<td>729.08 ± 0.15</td>
<td>-0.73 ± 0.11</td>
<td>0.74 ± 0.11</td>
</tr>
<tr>
<td>( ^{191}\text{IrFe} )</td>
<td>191</td>
<td>389.690 ± 0.010</td>
<td>-0.957 ± 0.014</td>
<td>1.144 ± 0.023</td>
</tr>
<tr>
<td>( ^{192}\text{IrNi} )</td>
<td>197</td>
<td>389.713 ± 0.017</td>
<td>-0.962 ± 0.015</td>
<td>1.150 ± 0.025</td>
</tr>
<tr>
<td>( ^{192}\text{IrFe} )</td>
<td>18</td>
<td>1175.05 ± 0.06</td>
<td>-0.776 ± 0.050</td>
<td>0.928 ± 0.066</td>
</tr>
<tr>
<td>( ^{105}\text{RhNi} )</td>
<td>82</td>
<td>167.643 ± 0.023</td>
<td>-0.448 ± 0.011</td>
<td>1.248 ± 0.031</td>
</tr>
<tr>
<td>( ^{105}\text{RhFe} )</td>
<td>50</td>
<td>504.192 ± 0.035</td>
<td>-0.358 ± 0.016</td>
<td>0.991 ± 0.045</td>
</tr>
<tr>
<td>( ^{105}\text{RhNi} )</td>
<td>18</td>
<td>218.070 ± 0.030</td>
<td>-0.944 ± 0.018</td>
<td>0.979 ± 0.019</td>
</tr>
<tr>
<td>( ^{105}\text{RhFe} )</td>
<td>182</td>
<td>539.608 ± 0.020</td>
<td>-0.913 ± 0.011</td>
<td>0.948 ± 0.011</td>
</tr>
</tbody>
</table>
Fig. 1: NMR-ON resonances of $^{60}\text{Co}^{191\text{m}}\text{Ir-Ni}$
IV. 3 DILUTE IMPURITY HYPERFINE FIELDS IN FERROMAGNETIC Fe, Co and Ni HOSTS
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All the experimental data reported so far on the hyperfine fields for impurities in the host matrices of Fe, Co, Ni and Gd have been tabulated. The best dilute impurity hyperfine field values are suggested. From the plot of hyperfine field vs the electronic configuration of the impurity for the different hosts we point out many interesting systematic trends of these fields. The magnitudes of the hyperfine fields are often assumed to be approximately proportional to the respective host moments. From our studies we conclude that deviations are usually larger than 10%, and therefore this assumption is valid only to a first approximation.

The experimental hyperfine field values are compared with the existing hyperfine field models as briefed below:

In the Daniel, Fridel, Campbell model the contact contribution is given by

$$E_{\text{CEP}} = A(2)P$$

where $$A(2)$$ is free atomic hyperfine field due to one unpaired outer s-electron and $$P$$ is the polarization at the impurity site. For transition impurities

$$E_{\text{CEP}} = A(2)P_h \left[ 0.6 + 0.4 \mu_1/\mu_h \right]$$

where $$\mu_1$$ is the local moment at the impurity site and $$\mu_h$$ is the host moment, $$P_h$$ is polarization at the host site. Wherever the measured $$\mu_1$$ values are available, the experimental and the predicted values are compared.

Shirley and Westenbarger and later Shirley, Rosenblum and Matthias suggested a model for the solute hyperfine fields. In this model the main contribution in the case of nd impurities is thought to arise from conduction electron polarization (CEP) and the core polarization (CP). From our comparisons, we conclude that for nd impurities the predictions of this model are in good agreement with the experimental values. For sp impurities, it was shown by Shirley that the transferred hyperfine structure in ferromagnets because of overlap of 3d electrons of the host and 5s electrons of the solute can result in observed large hyperfine fields.

Recently Stearns has shown that the volume overlap effect is important for the non transition series elements and the observed fields may be explained by means of a suitably normalized volume effect. Yet another model was suggested by Balabanov and Delyagin.

The best experimental dilute impurity hyperfine field values for the Fe, Co and Ni host have been compared with the various models mentioned above and their relative validity and possible explanation for the observed discrepancies are discussed.

REFERENCES
Hyperfine fields at sp impurities in the
ferromagnetic metals has attracted considerable
attention since it has been shown experimentally that
these fields are systematically negative for elements
in the first half of sp series and positive for elements
in the second half. For $^{69}\text{Ga}$ the absolute values of the
hyperfine fields in Fe and Co were measured using the
NMR-method by Kontani and Itoh [1]. In the reported work
the 43.9 keV level in $^{65}\text{Ga}$ was used to measure the sign
and value of the hyperfine field for Ga in Fe, Co and Ni.
The $2.2\text{h}$ $^{66}\text{Ge}$ activity was produced in $^{59}\text{Co}(^{11}\text{B},4\text{n})^{66}\text{Ge}$
and $^{56}\text{Fe}(^{12}\text{C},2\text{n})^{66}\text{Ge}$ reactions with 65 MeV $^{11}\text{B}$ and
40 MeV $^{12}\text{C}$ ions from the HI Cyclotron of the JINR in
Dubna. Measurements were performed by the TDPAC-method
using (182 ± 190) keV - 44 keV gamma-gamma cascade.
The $A_2$ angular correlation coefficient for this
cascade was found to be $-0.12 \pm 0.05$. From the

registered at the angle of $135^\circ$, for two field directions the $R(t)$ function was
determined. The polarizing field was about 1.5 kG. The measurements were performed at
room temperature. Fig.1 shows examples of $R(t)$ function for $^{66}\text{Ga}$ in Co and Ni. The values
of the obtained Larmor frequency are $\omega_L = (0.23 \pm 0.01) \times 10^9$, $(0.126 \pm 0.005) \times 10^9$
and $(0.030 \pm 0.005) \times 10^9$ s$^{-1}$ for $^{66}\text{Ga}$ in Fe, Co and Ni, respectively. Using $g = -0.504$ [2]
the effective fields for Ga in Fe, Co and Ni are equal to $-94 \pm 5$, $-52.2 \pm 2.4$ and
$-12.5 \pm 2.1$ kG, respectively. These values are in good agreement with the values
reported in Ref.[1].

2. P.Heubes, H.G.Johann, W.Klinger, W.Kreische, W.Lampert, W.Loeffler, G.Schatz,
IV. 5  

**FAST PRECESSIONS OF $^{57}$Fe IN IRON BY TIME-DIFFERENTIAL PERTURBED ANGULAR CORRELATIONS**  

by H. C. Benski and J. Berthier  

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The use of $^{57}$Fe as a hyperfine field probe with Time-Differential Perturbed Angular Correlations (TDPAC) has been hampered by poor time resolution at low $\gamma$ energies and the inherently small nuclear anisotropy, $A_2$, of the 122-14.4 keV cascade. It is possible to do TDPAC on the 136.4 keV state but in addition to having a much smaller lifetime than the 14.4 keV state, the former must be created by Coulomb excitation. The TDPAC technique allows for a Larmor precession measurement over several lifetimes of the intermediate state thus making the interpretation of the equivalent linewidth easier than in the corresponding Mössbauer resonance case. This fact has been recognised before and the fast Larmor precession of $^{57}$Fe in nickel and the slow quadrupole interaction frequency in a Cu$_{0.55}$Ni$_{0.47}$ alloy were measured using TDPAC. These experiments made use of the first photoelectron triggering principle to overcome the problem of poor time resolution using classical leading edge timing discriminators. We have used the recently developed noise rejecting timing discriminators to measure the fastest precessions of the 14.4 keV state of $^{57}$Fe observed to date by TDPAC. These discriminators can resolve the photomultiplier noise pulses from the small 14.4 keV pulses by rejecting the pulse signals which decay in less than a predetermined time. Since the NaI(Tl) crystals used for $\gamma$ ray detection produce pulse tails which are much longer than noise pulses, true first photoelectron triggering is easily achieved without appreciable loss in the counting rate. The time resolution attainable with a 2.5x2.5 cm NaI(Tl) crystal for the 122 keV $\gamma$ ray and a 2.5x0.1 cm NaI(Tl) crystal for the 14.4 keV $\gamma$ ray, coupled to XP2020 photomultipliers, was determined with a $^{88}$Y $\gamma$-X ray prompt source. The full width at half maximum (FWHM) of the resolution curve was 3.85 nsec.

The sample studied was a Fe$_X$(Fe) source prepared by diffusing the $^{57}$Co activity (~1 $\mu$Ci) into a 9N purity iron foil under a hydrogen atmosphere for one hour at 900 °C. The Larmor precession frequency of $^{57}$Fe in iron was measured with an otherwise conventional three counter slow-fast coincidence system. An external field, $H_{ext}$, of 210 G was applied perpendicular to the plane of the counters with a C-type magnet, clamping the foil between its pole pieces. Two "Start" detectors were positioned at 135 ° and 225 ° with respect to the "Stop" detector axis. The output of an ORTEC 457 Biased Time to Pulse Height Converter was routed to the two halves of a 400 channel multichannel analyser. The ratio:

$$\frac{1}{2} R(t) = \frac{C(3\pi/4, t) - C(3\pi/4, t)}{C(3\pi/4, t) + C(3\pi/4, t) - 2B}$$

was calculated, where $C(0, t)$ is the time-dependent coincidence rate and $B$ is the accidental coincidence background. For a static magnetic interaction, the nuclei precess with the Larmor frequency $\nu_L$ and:

$$R(t) = \frac{3}{2} A_2 \gamma_a \gamma_t \sin(4\nu_L t)$$

was calculated, where $\gamma_L = -g\mu_N H_{eff}$, where $g = -0.135$ for the 14.4 keV state, $H_{eff}$ is the effective field at the nucleus, (external plus hyperfine field) and $\gamma_a$ and $\gamma_t$ are the angular and time resolution attenuation factors.
From (2) we see that the period observed in $R(t)$ is actually half the Larmor period. The experiment resulted in a value of $(1/2\omega_0) = 19.2 \pm 0.15$ nsec, leading to $H_{\text{eff}} = -331.5 \pm 2.6$ kOe which after correction for the external field yields $H_{\text{hf}} = -331.3 \pm 2.6$ kOe for the hyperfine field of $^{57}$Fe in iron. This is in excellent agreement with the very accurate result obtained by Mössbauer effect measurements which gave $H_{\text{hf}} = -330.4 \pm 0.3$ kOe as well as with other previous similar experiments.

The Fourier transform of the autocorrelation function of $R(t)$ is shown in Fig. 1. The oscillatory component $2\nu_0$ is clearly seen. Our result can be directly compared to the 26 MHz resonance curve obtained by Nuclear Magnetic Resonance (NMR) in the $I = 3/2$ state of $^{57}$Fe in iron. As a further application, we note that the observation of rf-induced transitions between the magnetic sublevels of this state by the Mössbauer-NMR technique is believed to be complicated by acoustic-magnetostrictive sidebands. In contrast, these effects should not affect the $^{57}$Fe TDPAC experiments, (although acoustic vibrations could, in other particular cases, perturb the angular correlation), thus giving the possibility of suppressing the undesired acoustic sidebands from excited state NMR data.

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References


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The NMR-ON method was applied to $^{58}\text{CoFe}$ and $^{52}\text{MnFe}$, yielding the magnetic hyperfine splittings 441.736 ± 0.036 MHz and 88.700 ± 0.015 MHz, respectively. The following upper limits are deduced for the hyperfine anomalies

$$^{58}\text{Co} : 60\text{Co} \quad \Delta 60 \quad 2 \times 10^{-3}$$
$$^{52}\text{Mn} : 55\text{Mn} \quad \Delta 55 \quad 3 \times 10^{-3}$$

Besides the determination of the hyperfine anomaly, these experiments were essentially done in order to check whether the NMR-ON method is applicable to sources which are prepared by direct recoil implantation of radioactive impurities into thin Fe foils by the nuclear reaction. This technique can be applied to many isotopes which have so short half lives that the usual source preparation (chemical separation, melting, rolling etc.) is not applicable. One of the problems of all implantation techniques is the assumption of a unique hyperfine field, i.e. the question to what percentage the impurity atoms are substituted on regular lattice places. This has to be known for common NO experiments, where the anisotropy of the $\gamma$-radiation is measured as a function of the temperature. In NMR-ON experiments, however, only the magnitude of the resonance effect is influenced if a certain number of impurity atoms sees a "wrong" hyperfine field.

Activities of $^{58}\text{Co}$ in Fe were produced by $\alpha$-irradiation of thin Fe foils (0.8 and 2.0 mg/cm$^2$) at the Karlsruhe Cyclotron. The most active parts of the foils were cut out and soft soldered to both sides of the copper fin system of an adiabatic demagnetization cryostat without further annealing. Using Cr-K alum as cooling salt, temperatures of 0.01 K were obtained. The external magnetic field $H_0$, which is necessary to align the ferromagnetic domains and to establish a unique direction of the hyperfine field, was produced by a pair of superconducting split coils. Perpendicular to $H_0$ the rf field was applied, with both fields acting in the plane of the foils. With the g-factor of $^{58}\text{Co}$ $g = 1.998 \pm 0.006$ /1/ and the hyperfine field of Co in Fe $H_{HF} = -238.98 \pm 0.62$ kG /2/ the resonance frequency for $H_0 = 0$ is expected at 441.6 ± 1.6 MHz. One typical frequency spectrum (number of counts in the direction of $H_0$ versus frequency) for $H_0 = 0.40 \pm 0.01$ kG is shown in the upper part of fig.1. It has been measured in the following way: The applied rf field was 1 kHz sinusoidally frequency modulated with a deviation of ~ 300 kHz (FM), while the center frequency (CW) was swept continously from 436 to 446 MHz with a sweep rate of 5 kHz/sec. The 810 keV $\gamma$-ray was detected by two 3" $\uparrow$ x 3" NaI(Tl) detectors, which were placed at $0^\circ$ and $90^\circ$ with respect to the direction of $H_0$. The corresponding counts were stored in a multichannel analyzer as a function of the CW. Exact frequency-channel-correspondence was accomplished by controlling the CW with a frequency counter which itself synchronized the FM. In order to avoid a shift of the center of the resonance due to the nuclear spin lattice relaxation time two successively measured spectra with opposite sweep direction were always added together. Fig.2 shows the shift of the resonance frequency with the external field. Each point in this plot results from the summation of several spectra which were taken during the warming up of the salt pill. From a least squares fit we get $g\mu_B H_{HF} = 441.733 \pm 0.036$ MHz. With the hyperfine splitting of $^{60}\text{CoFe}$ of 165.957 ± 0.005 MHz /2/ we calculate the ratio of the
The ratio of the magnetic moments has been determined as $\frac{\mu_{58}^{\text{Co}}}{\mu_{60}^{\text{Co}}} = 1.0645 \pm 0.002$ by EPR. Thus the hyperfine anomaly

$$\frac{\Delta_{58}^{\text{Co}}}{\Delta_{60}^{\text{Co}}} = \frac{\Delta}{58} \frac{\mu_{58}^{\text{Co}}}{\mu_{60}^{\text{Co}}} - 1$$

can be estimated as $58 \Delta_{58} = 2 \times 10^{-3}$.

b) $^{52}\text{MnFe}$

The measurements on $^{52}\text{MnFe}$ were performed in a similar manner as described before. Because of the smaller linewidth the deviation of the FM was chosen to $\pm 100$ kHz.

A typical spectrum (counting rate of the $744$ keV $\gamma$-ray at $0^\circ$ with respect to $H_0$) is shown in the lower half of fig. 1. The shift of the resonance with $H_0$ is shown in fig. 2. Our result for the hyperfine splitting is $g_J^{\text{MnFe}} H_{\text{HF}} = 88.700 \pm 0.015$ MHz. Taking the hyperfine frequency of $^{55}\text{MnFe}$ as $239.99$ MHz /3/ and the magnetic moments from ref. /4-5/ the hyperfine anomaly can be estimated as $\Delta_{55} = 3 \times 10^{-3}$.

/2/ E. Hagen, G. Eska, "Shift of NMR-ON resonances with the external magnetic field", this conference
The uncertainty in the distribution of nuclei over sites with different hyperfine fields is a possible source of error when deriving both nuclear properties and the hyperfine fields themselves from a conventional nuclear orientation experiment. N.O./N.M.R. however offers the possibility of obtaining both unique values of the hyperfine fields and a lower limit to the percentage of nuclei in each particular site. This paper reports the application of N.O./N.M.R. to a case of current interest (1) that of I\textsubscript{Fe}. Samples of two isotopes have been obtained, 8 day I\textsubscript{31} and 2 hour I\textsubscript{32}, the former by ion-implantation and the latter by thermal diffusion of 3 day I\textsubscript{32} into Fe.

For the I\textsubscript{31} experiment carrier free I\textsubscript{31}, obtained from the Radiochemical Centre, Amersham, Bucks., was implanted at 80 keV into polycrystalline iron by the E.M. separator group at A.E.R.E. Harwell. Prior to implantation copper backing strips were soldered to the sample. After implantation these copper strips were soldered to the cold finger of a chrome alum salt pill, cooled to 15 K and polarized with an external field of 2.2 kOe. The 364 keV γ-ray was observed using a 3\textdegree\times 3\textdegree NaI detector to have an anisotropy of +1\%. Taking the moment of I\textsubscript{31} as 2.738 ± 0.001 n.m. (2), the spin as 7/2, and the present value of the hyperfine field of I in Fe, 1.13 ± 0.05 (3) kOe a resonance was expected in the range 650 to 700 MHz. With a frequency modulation amplitude of 500 kHz a resonance was observed near 680 MHz. The response at each frequency was measured as the difference in anisotropy between a frequency modulated and pure carrier R.F. field. Fig. 1 shows the resonance observed, the computed line shape being for a gaussian inhomogeneously broadened line of FWHM 8.3 MHz and 42\% integrated destruction of anisotropy, the centre frequency of the resonance is 683.3 ± 1.0 MHz. Fig. 2 shows the effect on the resonant frequency of different applied fields on a single crystal sample, the positive slope indicating that iodine has a positive hyperfine field. Using the above value of μ and allowing for an applied field of 2.2 kOe we obtain H = + 1143.5 ± 1.7 kOe.

In the I\textsubscript{32} experiment 60 Cl/gm I\textsubscript{32} activity was obtained as sodium tellurite from the Radiochemical Centre, Amersham. A few drops were dried on a 30 μm polycrystalline iron foil and diffused for 12 hours at 1000°C in a hydrogen atmosphere. The foil was rubbed to 20 μm to remove surface activity, lightly etched and then rolled to 1.8 μm, the resulting foil having a local tellurium concentration of 10\textsuperscript{-5} atomic \%. Again a 3\textdegree\times 3\textdegree NaI detector was used to monitor the 670 keV γ-ray and at 15 kOe an anisotropy of -20\% was observed.

Fig. 1 shows the resonance response obtained in an external field of 1.3 kOe with a frequency modulation.
amplitude of 1 MHz. Taking the moment of $^{132}$I as $3.084 \pm 0.007$ m.m., its spin as 4 and allowing for the external field, the resonance at 674.0 ± 1.0 MHz gives us $H = +1445.3 \pm 2.6$ kOe. Combining both results we obtain $H = 1444.0 \pm 2.5$ kOe.

The line width observed for the implanted $^{131}$I sample is more than twice that for the thermally diffused $^{132}$I sample. This extra width could be due either to damage during implantation or to a small quadrupole splitting as $^{131}$I has a quadrupole moment four times that of $^{132}$I. Experiments are proceeding in single crystals to clarify this point.

The computed line shape for $^{131}$I represents an integrated destruction of 42% so that at least 42% of the iodine nuclei must be in this high field site. Unfortunately with the present experimental set up it was not possible to saturate the resonance i.e. as more R.F. power was applied the integrated destruction increased and showed no sign of approaching a fixed value at the highest R.F. power available to us. However, if we assume a two site model in which the nuclei either experience our high field or zero field then conventional nuclear orientation gives 70% in the high field site.

The computed line shape for $^{132}$I represents 11% integrated destruction of anisotropy. Again it was not possible to saturate the resonance, this sample having the additional complication that its thickness, 1.6 mm, is somewhat larger than the skin depth at these frequencies, approximately 0.5 mm. The skin depth is not a problem in the implanted source as the iodine atoms are within 120° of the surface. Conventional nuclear orientation on this source gave 60 to 80% in this high field site again assuming a two site model.

This work was supported by S.R.C. grants.

References

We have implanted radioactive Gd\(^{151}\) into polycrystalline iron foils, and used Mössbauer spectroscopy to determine the hfs of the Eu\(^{151}\) decay product. The spectra observed show 2 components (Fig. 1) of roughly equal intensity. From the behavior under annealing, and comparison with backscatter channeling measurements on similar systems, the site yielding the split spectrum can be identified as substitutional. That site has an isomer shift of \(-3.4 \text{ mm/sec} \) vs. the EuF\(_3\) absorber, corresponding to the charge density which would be obtained for an Eu\(^{3+}\) ion with an additional contribution from conduction electrons. Eu\(^{3+}\) has 6 4f electrons in the configuration \(^7F_0\), and this ground state is "non magnetic", having \(J=0\). However, an applied magnetic field or the presence of an exchange field mixes low lying states of \(J=1,2,\ldots\), and these do produce a hf field. (This second-order mixing process produces the well-known Van Vleck temperature-independent susceptibility in Eu\(^{3+}\).) The hf field may then be written \(B(T) = B_{\text{cep}}(T) + B_{\text{ex}}(T)\), where \(B_{\text{cep}}\) is the component arising from the conduction electron polarization, which should be proportional to the host magnetization, and thus vary only slowly with temperature. \(B_{\text{ex}}\) contains the admixture term, and has been shown to be describable as \(B_{\text{ex}}F(T)\), where \(B_{\text{ex}}\) is the exchange field, and \(F(T)\) is a complex function determined entirely by the known energy splittings and matrix elements of the 4f\(^6\) manifold. Thus, the observed temperature dependence (Fig. 2) can be fitted to a theory with only 2 free parameters, whose values are \(B_{\text{cep}} = (-0.80 \text{ MG})\), and \(B_{\text{ex}} = (-0.25 \text{ MG})\). The negative sign of \(B_{\text{cep}}\) is determined from systematics, and that of \(B_{\text{ex}}\) from the fact that the 4f and cep contributions must be additive to produce the observed temperature dependence. \(B_{\text{cep}}\) should vary only slowly with \(Z\) for the lanthanides. Thus, the accurate value obtained here for the substitutional Eu site should help to understand the wide range of values that have been reported for other rare-earth impurities in iron. The value of the exchange field, \(B_{\text{ex}}\), is considerably smaller than those determined for other rare earth ions dissolved in iron, and Re-iron intermetallic compounds. The unsplit component of the spectrum has an isomer shift of about \(-0.8 \text{ mm/sec} \) vs. EuF\(_3\), similar to that observed for Eu\(_2\)O\(_3\).
Fig. 1: Eu$^{151}$ Mössbauer hfs from Gd implanted into iron. The spectrum can be fitted with two components as shown. The split spectrum is identified as coming from the substitutional Eu ions primarily because its anneal behavior is similar to that observed (by channeling backscatter measurements) for substitutional Yb implanted into iron.

Fig. 2: Temperature dependence of the hfs field for the substitutional component of EuFe. The data have been fitted with the two-parameter model discussed in the text, and the calculated hfs field is shown by the solid line. The broken line shows the temperature dependence of the iron sublattice magnetisation.
IV. 9 Mössbauer effect of Er impurities implanted in iron
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The hyperfine interaction of rare earth nuclei in ferromagnetic metals is a subject of considerable interest, both from the nuclear and from the solid state point of view. Although a number of experiments have been performed, the general picture is unsatisfactory. Precise implantation measurements at room temperature\(^{1,2}\) give field values which are roughly a factor of four lower than the expected interaction field of the "ionic" hyperfine field (dominated by the \(\text{Fe}^{3+}\) contribution).

The isotope \(^{166}\text{Er}\) has been used for a Mössbauer study of Er impurities in iron. The effect is measured on the \(2^{+}-0^{+}\) transition (energy 30.5 eV). The parent isotope \(^{164}\text{Ho}\) (half-life 37 h.) was produced by irradiating \(\text{Ho}_{2}\text{O}_3\) during 3 h in a neutron flux of \(5 \times 10^{12} \text{n/s cm}^2\), giving a specific activity of 8 Ci/g. About 0.5% of the irradiated \(\text{Ho}_{2}\text{O}_3\) was used in the implantation, which was done with the EUL method\(^3\), at a stable beam current of \(\approx 30 \mu A\). An efficiency of 3% was achieved. The \(^{166}\text{Er}\) ions were implanted in a \(99.999\%\) pure iron foil with an energy of 150 keV; the resulting source had an initial strength of \(459 \text{ mCi}\) and the total implanted dose was estimated to be \(2 \times 10^{15} \text{ at/cm}^2\).

The source was mounted on a lucite rod moving vertically in the liquid \(\text{He}\) in the cryostat. The absorber consisted of \(185 \text{ mg/cm}^2\) \(\text{ErAl}_2\), connected to the liquid nitrogen shield. The spectrometer operated in the constant acceleration mode at a trapezoid frequency of \(8 \text{ Hz}\). The considerable background curvature due to the moving source was corrected to a large extent by means of a background measurement with a \(^{133}\text{Ba}\) source in the same geometry. Velocity calibration was done with the Mössbauer technique\(^7\). The folded spectrum can be seen in the figure. It was fitted by a sum of five lorentzians with equal widths \(\Gamma\), the positions of which were calculated from the following Hamiltonian

\[
H = -\epsilon_0\eta\eta^* + \frac{e^2\hbar}{4} \left( 3 I_z^2 - I(I + 1) \right)
\]

\(I = 2\) being the spin and \(\eta_0 = 0.346 \pm 0.008\) the nuclear \(g\)-value of the excited state. \(e^2\eta_0\) is the usual quadrupole coupling constant. The results are summarized in the table.

<table>
<thead>
<tr>
<th>Experimental values</th>
<th>Theoretical estimates</th>
</tr>
</thead>
<tbody>
<tr>
<td>(E_{4f}) (keV)</td>
<td>(E_{4f}) (keV)</td>
</tr>
<tr>
<td>(E_{4f}) (cm/s)</td>
<td>(E_{4f}) (cm/s)</td>
</tr>
<tr>
<td>(I) (cm/s)</td>
<td>(I) (cm/s)</td>
</tr>
<tr>
<td>(H_{hf}(4f)) (M(\text{o}))</td>
<td>(H_{hf}(s)) (M(\text{o}))</td>
</tr>
<tr>
<td>2.85(3)</td>
<td>7.64(17)</td>
</tr>
</tbody>
</table>

Measurements on \(\text{DyFe}\)\(^{9}\) and \(\text{TbFe}\)\(^{10}\) have been explained by assuming the ground state of the rare earth ion (for \(\text{Er}^{3+}\) this is \(4f^{15/2}\)) to be split by the exchange interaction between the \(4f\) local moment and the polarized conduction electrons. Using a simple molecular field approach this interaction can be written as

\[
E_{\text{exch}} = g_0\frac{\hbar}{2} \sum_{\mathbf{S}_{4f}} S_{4f} = (g_0 - 1) g_\text{hf} \sum_{I} \frac{1}{2} I(I + 1)
\]

For the electronic ground state \(\langle J_z \rangle = 3\) and \(\langle S \rangle = 5\). \(g_{hf}\) is of the order of \(\approx 5 \text{ M}\(\text{o}\)). Apart from this exchange interaction the \(\text{Er}^{3+}\) experiences a crystalline...
electric field from its neighbors. For a substitutional Er$^{3+}$ ion thin field has cubic symmetry, its influence on $|S>$ and $|S'>$ in the ground state.

The $4f$ contribution to the hyperfine interaction in the electronic ground state, neglecting off-diagonal elements of the quadrupole interaction, can be written as:

$$\mathcal{H}_{hf} = 2 \zeta \frac{e^2 q}{4 \pi \epsilon_0} \left( \frac{3}{2} \left| \langle J_2 | S_2 \rangle \right|^2 \left| \langle J_2 | S_2 \rangle \right|^2 \right)$$

Apart from the $4f$ contribution to the hyperfine field there will be a component $H_{hf}(s)$ arising from the $s$ electrons. This contribution consists of three terms, which can be estimated using the analysis of Hüffer for the case of Gd$^{3+}$ and the known difference between $H_{hf}$ for Gd$^{3+}$ and Gd$^{2+}$:

a) core polarization + relativistic effects $= 170$ kOe, b) conduction electron polarization due to the $4f$ moment $= 120$ kOe, and c) due to the iron host $= 270$ kOe. The sum of $H_{hf}(s)$ and $H_{hf}(300 K)$ is seen to be 0.7 kOe higher in absolute value than the experimental result. If this is interpreted as a reduction of $<J_2>$ due to crystalline field effects, one can calculate the effect of this reduction on the quadrupole coupling. The calculated value agrees very well with the observed interaction.

The NMR value for the hyperfine field in Er$^{3+}$, measured at room temperature, must be corrected to $-3.8$ kOe by assuming the rest of the Ho ions occupy substitutional sites; nevertheless we see only one hyperfine field. This contradiction can be removed by assuming that the Ho ions occupy sites which are associated with vacancies. In that case a large axial electric field in present which together with the exchange interaction gives rise to an electronic ground state with $|J_2>$ $= J$. The hyperfine field of both fractions thus differ by a small amount, resulting only in some linebroadening.

References:

14. B.I. Deutch, in ref. 12, p 137.
IV. 10 THE TEMPERATURE DEPENDENCE OF HYPERFINE MAGNETIC FIELDS AT $^{150}$Sm AND $^{146}$Nd RECOIL IMPLANTED INTO POLARIZED Fe AND Ni HOSTS

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D. E. MUEHICK
Bell Laboratories, Murray Hill, New Jersey 07974

The large hyperfine magnetic fields observed at rare earth (RE) nuclei implanted into Fe and Ni at $T = 300$K are generally attributed to the sum of three effects: a contact interaction involving the polarized RE ionic core (CP), a contact interaction involving polarized conduction electrons (CEF), and the net orbital moment of the partially filled 4f shell. The CP and CEF effects are of order several hundred kOe or about 10% of the observed fields at Sm and Nd recoils in Fe and Ni at 300K. The dominant contribution to observed hyperfine magnetic fields is the splitting of the RE ionic ground state due to exchange interactions with the ferrimagnetic host. Studies of the temperature dependence of a hyperfine magnetic field can help elucidate the nature of its components, since the temperature dependence of the CP and CEF contributions is expected to be similar to that of the host magnetization, whereas, the contribution of the 4f shell orbital moment is proportional to a Brillouin function and is expected to approach the value of the free ion field at $T = 0$K in the absence of additional level-splitting mechanisms such as crystalline electric fields (CEF). We have performed measurements of the temperature dependence of the hyperfine magnetic fields at $^{150}$Sm and $^{146}$Nd nuclei recoil implanted into polarized Fe and Ni hosts from $T = 4.2$K to 552K using the IMPAC technique. The results have been analyzed in terms of the interactions of the respective RE ionic ground states in the exchange fields of the Fe and Ni hosts. The recoil IMPAC technique has the advantages of allowing studies of the splitting of ionic ground states in Fe and Ni hosts using the lowest RE impurity dose of any other available technique and the possibility of using short nuclear lifetimes. These advantages result in simplifying the interpretation of the experimental results by limiting the number of observed effects.

A beam of 38 MeV $^{16}$O$^+$ ions was used to Coulomb excite and recoil implant the target nuclei into Fe and Ni hosts which were polarized by an external field of $-2.5$ kOe. In order to insure that de-excitation y-rays corresponded to proper implantation and alignment of the target nuclei in their $J = 2^+$, $m = 0$, first excited states, coincidence was required with oxygen particles backscattered into an annular surface barrier detector. Angular correlations were measured for 12 or more angles for opposite external field directions at all of the observation temperatures. The results were least-squares fitted to the usual expression for the time integral particle-γ correlation including the transient field effect, from which $H_{\text{Total}}$ was extracted using $\gamma = 0.20 (4)$, $t = 31.8 (3.2)$ psec. for $^{146}$Nd and $\gamma = 0.273 (2.4)$, $t = 69.1 (1.5)$ psec for $^{150}$Sm.

The resultant values of $H_{\text{Total}}$ were corrected for $H_{\text{CP}}$ and $H_{\text{CEF}}$ contributions and the remainder, $H_{\text{EX}}(T)$ (see fig. 1), was analyzed in several ways. The values of $H_{\text{EX}}$ were first fit to a Brillouin function (of appropriate ground state spin) for the 4f local moment in an exchange field (see Table 1). Note that the ratio of the fitted exchange fields ($H_{\text{EX}}$) in Fe and Ni for either implant equals the host moment ratio. Attempts to fit $H_{\text{CEF}} + H_{\text{EX}}$ with a Low model yielded inconclusive results for the former but still the same ratio for the exchange fields. CEF contributions were then added to the Hamiltonian. Assuming that a pure exchange interaction would lead to saturation of $H_{\text{CEF}}(T = 0)$ at the free ion value (3.3 kOe for Sm and 4.2 kOe for Nd), CEF contributions to the splitting of the ionic ground state appear to explain the lower saturation values observed. CEF symmetry appropriate to lattice sites in Fe and Ni was used. Exchange mixing of the first excited ionic state ($J = 7/2$) of the Sm$^{+}$ ion does not seem necessary to explain the results.

<table>
<thead>
<tr>
<th>IMPLANT</th>
<th>$H_{\text{EX}}(T=0)$</th>
<th>$H_{\text{CP}}$</th>
<th>$H_{\text{CEF}}$</th>
<th>$H_{\text{EX}}(T=0)$</th>
<th>$H_{\text{CP}}$</th>
<th>$H_{\text{CEF}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{150}$Sm($5/2^+$)</td>
<td>2.86(35)</td>
<td>-3.12(42)</td>
<td>0.161</td>
<td>0.133(13)</td>
<td>1.50(17)</td>
<td>-0.79(12)</td>
</tr>
<tr>
<td>$^{146}$Nd($13/2^+$)</td>
<td>2.80(20)</td>
<td>-6.1 (8)</td>
<td>0.111</td>
<td>0.133(13)</td>
<td>1.52(9)</td>
<td>-1.66(32)</td>
</tr>
</tbody>
</table>

Table 1. Results of RE IMPAC Experiments

*a) In units of kOe.
Fig. 1. a) SmFe, b) SmNi, c) NdFe, d) NdNi. Solid line is best fit Brillouin function.

References

1. Supported in part by the National Science Foundation.
IV. 11 ANOMALOUS TEMPERATURE DEPENDENCIES OF HYPERFINE FIELDS FOR TRANSITION ELEMENT IMPURITIES IN Fe AND Ni

By

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The magnetic hyperfine field $B_{hf}$ at the nucleus of an impurity atom in a ferromagnet normally has a temperature dependence proportional to that of the host magnetization $M$. For 3d, 4d and 5d transition metal impurities, deviations from the proportionality has been observed in a few cases [1,2,3], whereas others show a normal behaviour. In a series of experiments we have obtained data for $B_{hf}(T)$ for a number of 4d- and 5d elements in iron and nickel in order to find out under what circumstances temperature anomalies occur.

The following impurity-host combinations were investigated. All measurements were performed by time-integral PAC in the respective isotopes. Some selected data have been published earlier [3], but have now been improved and complemented.

<table>
<thead>
<tr>
<th>Host</th>
<th>4d</th>
<th>5d</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>101Ru $\phi$</td>
<td>106Pd $\phi$</td>
</tr>
<tr>
<td>Ni</td>
<td>95Mo $\phi$</td>
<td>102Ru $\phi$</td>
</tr>
<tr>
<td>Fe</td>
<td>101Ru $\phi$</td>
<td>106Pd $\phi$</td>
</tr>
<tr>
<td>Ni</td>
<td>188Os $\phi$</td>
<td>191Ir $\phi$</td>
</tr>
</tbody>
</table>

The samples were prepared by melting the mother elements together with the host metals followed by heat treatments at appropriate high temperatures for periods of 1-50 days. They were placed in a polarizing field of 20 kG and the $\omega$ was determined in a 4-detector system at temperatures from 4K up to the Curie points.
A survey of the $B_{hf}(T)$ data obtained here is presented in Figs. 1 and 2 together with relevant data from the literature (filled circles) for near-by elements.

In addition to the well-known anomalous case of MnFe in the 3d-series it is found that RuFe and OsFe are anomalous cases whereas the elements above and below in each period show no or only minor anomalies. For the Ni-based alloys the well-known anomaly in RuNi was confirmed, now with the short-lived probe level in $^{101}$Ru (0.79 ns). In addition, there are indications that OsNi is anomalous at high temperatures.

These data should be compared to those of Vincze et al [4] on the hyperfine fields of host atoms near 4d and 5d impurities. They show temperature anomalies for the same elements and in the same temperature range as the impurity hyperfine fields. This indicates that the origin of the anomalies are local spin disturbances around the impurity elements, which are very sensitive to temperature variations. The model of Campbell [5] based on the Friedel scattering theory explains the anomalous cases as transition cases with a "critical" screening charge for the impurity. Such impurity-host combinations may have a high-temperature, low spin-density phase and a low-temperature, high spin-density phase for the impurity. The correlation with other data such as the increase of resistivity for these impurities [6] is also satisfactory.

References
4. I. Vincze, Solid State Comm. 10 (1972) 341
Measurements of the critical exponent $\delta$ in ferromagnets have been performed both with macroscopic and microscopic experimental methods. Microscopic methods such as Mössbauer and perturbed angular correlation measurements have one main advantage; they can be done without applying external magnetic fields. The basic assumption for these microscopic measurements says that, at least in the critical region, the magnetic hyperfine field should closely follow the spontaneous magnetization as a function of temperature

$$H_{hf}(T) \sim M(T) \sim \left(1-T/T_C\right)^{\delta} \left|T-T_C\right|^\delta.$$  

Up to now all microscopic measurements for $\delta$ have been done with impurity atoms. The present measurement is the first to be done with lattice atoms.

There is only one isomeric level in all of the nickel isotopes that can be used as a probe for measuring $\delta$. This level - the $5/2^-$ level at 87.2 keV in $^{63}$Ni - can be reached by the (d,p) reaction on $^{62}$Ni. The lifetime and g-factor are $\tau = 2.48(4)$ ms and $g = 0.301(1)$ [ref.1] which, like $^{109}$Rh, allow the determination of hyperfine fields of $\approx 1$ kG. The experiments were performed at the 7 MeV Van-de-Graaff-accelerator of the Hahn-Meitner-Institute using the well known time-differential perturbed angular distribution of $\gamma$-radiation following nuclear reaction (TDPAD) as the experimental method. The technique and apparatus have been well described elsewhere [2,3]. Two different targets were used, each consisting of 98% enriched $^{62}$Ni. The metal powder was first reduced in a hydrogen atmosphere within a quartz tube, then melted and pressed into disks. After that they were annealed in argon at 600°C for five hours and in vacuum over a period of three hours at 400°C. The targets were heated in an electron beam furnace with fast temperature regulation [4], which gave a sufficient temperature stability even with unstable accelerator beams.

One severe problem connected with the nuclear reaction method arises from radiation damages which cause a damping of the spinrotation pattern even at room temperature. Assuming that pure magnetic fluctuations are responsible for that damping rather than quadrupole interactions, we can use the calculations of Gabriel on PAC in the Markoffian limit [5]. These calculations yield the perturbation factor

$$G^{QW}_{kk}(t) = e^{-\frac{t}{T}} \left|u_L - \frac{\lambda_{QW}}{2}\right|^2 (t>0)$$

with $u_L$ being the Larmor frequency in the static magnetic field. This formula was used for the fit to the data. The damping factor

$$\lambda_{QW} = \frac{k(k+1)}{\hbar^2} \left(\frac{T_2}{T_1}\right) q^2$$

is different from the Abragam-Pound solution [6] because $T_1T_2$ and gives only a term proportional to $q^2$ as the electron Larmor frequency is much greater than the nuclear Larmor frequency [7]. As an example, three spinrotation modulations for different target temperatures are shown in fig. 1.

In addition, we tried to fit the data in different ways, allowing the frequency $u_L$ to have a Gaussian or a rectangular distribution as well as

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**Fig. 1:**
Modulation of $\gamma$-ray time spectra showing Larmor precessions of excited $^{63}$Ni nuclei in randomly oriented internal magnetic fields in nickel at various temperatures below $T_C$. 

Hahn-Meitner-Institut Berlin and Freie Universität Berlin, Fachbereich Physik
having combined magnetic and quadrupole interactions, but the fits with magnetic fluctuations gave better values for $\chi^2$.

The results for $\delta$ are obtained from a three parameter least squares fit of the function $H_{hf}(T)/H_{hf}(0) = B \delta^\gamma$, $\gamma = 1 - T/T_C$ to the temperature dependent hyperfine fields $H_{hf}(T)$ which have been extracted from the Larmor frequencies $\omega_c(T)$. Free parameters were $B$, $\delta$, and $T_C$. The final experimental results, which agree within the errors for the two targets, is $\delta = 0.32(3)$. The different results for $T_C$ (fig. 2) can be explained by the different fraction of copper impurities in the targets. Our result is in serious disagreement with experimental values obtained by hyperfine measurements with impurity atoms in nickel (tab. 1) but is in agreement with the three dimensional Ising model [12], $\delta = 0.312$ and with recent results of kink point measurements on Ni, $\delta = 0.346(7)$ and on NiCu alloys ($\delta = 0.34(1)$) [13]. The result reported in this paper casts serious doubts on the validity of Eq. (1) for microscopic measurements done with impurity atoms. These doubts are confirmed by our recent experiments with $^{66}$Cu and $^{67}$Zn impurities in Ni [14].

We thank Dr. Malow of the Hahn-Meitner-Institut for the electron probe microanalysis of the target.

\[ \begin{array}{|c|c|c|} 
\hline
\text{probe} & \delta_{\text{exp}} & \text{ref.} \\
\hline
^{57}\text{Fe} & 0.38(1) & [8,9] \\
^{100}\text{Rh} & 0.385(5) & [10] \\
^{161}\text{Ta} & 0.417(10) & [11] \\
^{63}\text{Ni} & 0.32(3) & \text{this work} \\
\hline
\end{array} \]

Table 1: Experimental Hyperfine Critical Exponents in Nickel.

IV. 13

HYPERFINE CRITICAL EXPONENT $\beta$ FOR $^{111}$CD IN A NICKEL HOST

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In the study of magnetic critical phenomena, the critical exponent $\beta$ is defined in terms of the asymptotic form

$$\lim_{H_{\text{a}} \to 0} \sigma(T, H_{\text{a}}) = \sigma_0(T) = D(1 - C_1/T_{\text{c}})^{\beta}$$

(1)

where $\sigma$ is the reduced magnetization, $T_{\text{c}}$ the Curie or Neel temperature, $H_{\text{a}}$ the applied field, and $D$ a constant. Since the work of Heller and Benedek on $^{57}$Fe measurements of the reduced hyperfine field $h_{hf}(T) = H_{hf}(T)/H_{hf}(0)$ have been used to determine $\beta$ in a variety of cases. For anti-ferromagnets, this makes accessible the staggered magnetization. For ferromagnets it permits determination of the spontaneous magnetization without assumptions about the magnetic equation of state near $T_{\text{c}}$. Use of $h_{hf}$ for measurements requires that $h_{hf}(T)/\sigma_0(T)$ is constant near $T_{\text{c}}$. Hyperfine critical exponents may be criticized because this condition can not be proven, and is in fact known to fail for several cases well below $T_{\text{c}}$. In this situation one may seek empirical proof of the desired condition. Available information is discussed elsewhere in this conference. Here, as a contribution to this information, we present new results on $^{111}$CdNi.

Our measurements constitute an extension of earlier work into the critical region. Unlike $^{100}$RhNi and $^{57}$FeNi, which have also been investigated near $T_{\text{c}}$, $^{111}$CdNi is characterized by a hyperfine field that is largely produced by conduction electron polarization. It is in this way comparable to $^{161}$TaNi, for which $\beta$ has been recently reported.

Our sources utilized carrier free 2.81 day $^{111}$In, which after co-plating from a NiSO$_4$ solution, were diffused into 0.0025 cm thick Si foil at 1050 X in vacuum. Both the NiSO$_4$ and the foil were of 4- nines purity. The In concentration after diffusion was estimated at $10^{-5}$. Two types of measurements were performed, both with the source foil perpendicular to the counter plane. (1) With two counters and a magnetized foil, we measured

$$R(t) = \left[ C_{\text{mag}}(3\pi/4, t) - C_{\text{mag}}(3\pi/4, t) \right]/\left[ C_{\text{mag}}(3\pi/4, t) + C_{\text{mag}}(3\pi/4, t) - 2B \right]$$

(2)

(2) With three counters, and the source foil demagnetized, we measured

$$R(t) = \left[ C(\pi, t) - C(\pi/2, t) \right]/\left[ C(\pi, t) + 2C(\pi/2, t) - 3B \right]$$

(3)

Here $C(\pi, t)$ is the 171-245 keV coincidence rate as a function of counter angle and time, $B$ is the accidental background, and $\phi$ and $\phi'$ denote source foil magnetization parallel and anti-parallel to the source plane. Only demagnetized spectra were used close to $T_{\text{c}}$, thus satisfying the limit of Eq. (1). To double the time range available near $T_{\text{c}}$, start and stop channels were gated to accept both 171 and 245 keV gamma rays.

Assuming only static magnetic hyperfine interactions, and neglecting the anisotropy coefficient $A_{44}$, we expect for two geometries described above that

$$R_{\text{mag}}(t) = \frac{\gamma_a}{A_{22} \left(4 + A_{22} \right)^{\frac{3}{2}} \sin 2\omega_L t}$$

and

$$R(t) = \frac{\gamma_a (A_{22} / 10) (2 \cos \omega_L t + 2 \cos 2\omega_L t + 1)}{4 \times 10^{-3}}$$

Here $A_{22}$ is the usual anisotropy coefficient, $\gamma_a$ describes attenuation due to finite counter angle, and $\omega_L$ is the Larmor frequency. For the magnetized geometry and $(T - T_{\text{c}}) > 10^{-2}$ we obtain 90% of the amplitude prescribed by Eq. (4) and no other time dependence. We attribute the reduced amplitude to Compton scattering in the source and the oven walls, and conclude that nearly all of the $^{111}$In atoms occupy equivalent lattice sites. For the demagnetized geometry, we observe the signal prescribed by Eq. (5), but with amplitude reduced by 20-30%. The reduction was temperature independent, and for $(T - T_{\text{c}}) > 4 \times 10^{-3}$ showed no visible loss of coherence in time. We attribute the amplitude discrepancy to (a) increased Compton scattering effects, produced by the wide energy channels used; and (b) possible partial decoupling of the angular correlation due to greater than random alignment of domains in one of the counter directions.

Examples of demagnetized spectra are shown in Fig. 1 for three typical temperatures in the critical region. To provide the best possible estimate of the frequency for
The signal was arbitrarily fitted with an "effective relaxation" constant multiplying the right side of Eq. (5). The effect of the relaxation correction to the frequency was in all cases less than 10%. As in previous work on $^{100}$RhNi, it is impossible to attribute the observed loss of coherence to a specific cause, such as inhomogeneities or critical fluctuations. The temperature control system was as described previously, and provided long term stability and reproducibility to within 0.02 K, and gradients across the source of less than 0.1 K.

Well below $T_c$, we confirm the results of earlier measurements; i.e., $\beta(T/T_c) = 0.383 (4)$, $T_c = 632.16 (6)$, $D = 1.31 (2)$. If the temperature range is diminished to $3 \times 10^{-4} \leq (1-T/T_c) \leq 1.5 \times 10^{-2}$ the result is $\beta = 0.375 (15)$, $T_c = 632.12 (3)$, $D = 1.28 (3)$. Our results agree within statistics with two previous measurements on $^{100}$RhNi and $^{57}$FeNi, as well as with various bulk measurements. Our results differ markedly with recent measurements on $^{181}$TaNi, where $\beta = 0.417 (10)$ was obtained. The possible slight decrease in $\beta$ for $^{111}$CdNi as the temperature range is reduced is consistent with the model of Lovesey and Marshall. A possible explanation of the $^{181}$TaNi result in terms of this model is mentioned elsewhere in this conference.

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IV. 14

ARE HYPERFINE CRITICAL EXPONENTS PROBE INDEPENDENT?

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At one time it was proposed that atatic critical exponents are described by "universality"; that they depend only on lattice and spin dimensionality. Recently Sutherland, arguing from certain powerful inequalities, has suggested that critical exponents in general may depend on energy parameters; Aharony and Fisher, using the exact renormalization approach, have shown that critical exponents undergo small changes when dipolar interactions are added to the Hamiltonian. Universality is therefore at best naive, and small differences in observed exponents may have physical significance. For experimentalists this situation poses a two-fold challenge: (a) to look closely for small differences in critical behavior; and (b) to be concerned more than ever with the validity of their methods. Here we discuss the validity of hyperfine critical exponent measurements.

Since the work of Heller and Benedek, reduced hyperfine fields $h_{hf}(T) = h_{hf}/H$ have been used to determine the critical exponent $\beta$ where $\beta$ is defined by

$$\lim_{T \to T_c} \sigma(T)/H = \sigma^\beta_0(T)$$

and $\sigma^\beta_0(T)$ and $H$ are the reduced magnetization, Curie or Neel temperature, and applied field, respectively. The use of $h_{hf}(T)$ for $\beta$ measurements requires that near $T_c$

$$M(T) = h_{hf}(T)/\sigma^\beta_0(T) = \text{constant}$$

For antiferromagnets the use of $h_{hf}$ makes accessible the staggered magnetization; for ferromagnets it permits determination of the spontaneous magnetization without assumptions about the magnetic equation of state near $T_c$. Hyperfine measurements are criticized because Eq. (2) can not be proven near $T_c$, and in fact, known to fail in many cases well below $T_c$. The most dramatic failures occur for certain impurities dissolved in ferromagnets, but failure also occurs in pure systems. For anti-ferromagnets, failure of Eq. (2) is a moot question since $h_{hf}$ and $\sigma^\beta_0(T)$ can not be independently measured.

As one test of Eq. (2), I have evaluated $M(T)$ in the critical region for three representative models for impurities in ferromagnets, with results given in Fig. 1. For magnetic impurities I have used the model of Shirley, Rosembium and Matthias and the model of Campbell, both applied to one of the most anomalous cases, RuNi. For non-magnetic impurities I have used the model of Lovesey and Marshall as applied to Ni. All three models are seen to have only a weak temperature dependence in $M(T)$ near $T_c$. For magnetic impurities the corresponding error in $\beta$ would be less than 0.5% for $10^{-3} < 1 - T/T_c < 10^{-2}$. For non-magnetic impurities, the possibility of a 5% deviation exists. All available models use a molecular field approximation for the impurity-host interaction, while leaving the host-host interaction unspecified. While this permits non-classical behavior for the...

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Fig. 1: Behavior of $M(T)$ as calculated for three models, and comparison to $\sigma^\beta_0(T)$, the magnetization of Ni.

(a) Lovesey-Marshall model: the behavior of $M(T)$ represents the maximum possible effect, and occurs only if $h_{hf}$ is produced by nearest neighbor host atoms. (b) Shirley, Rosembium and Matthias extension of the model of Jacarino, Walker and Wertheim, with parameters chosen to fit the case of RuNi. (c) Campbell's model, with parameters chosen to fit the case of RuNi.
host, it is not clear to what extent the molecular field assumption introduces error.

Therefore, as a further test of Eq. (2), we take a strictly empirical approach and ask (a) are exponents obtained with different hyperfine probes equal to each other and (b) are they equal to exponents obtained in bulk measurements? Available information is summarized in Table 1. For FeF₃, an insulating anti-ferromagnet, hₕf measurements exist both for Fe and F sites, and are in excellent agreement. For CrBr₃, an insulating ferromagnet, hₕf and bulk measurements exist and agree perfectly. For Ni, a metallic ferromagnet, there are five hₕf measurements using four impurity probes, and five bulk results. Except for one bulk (kink-point) and one hₕf measurement, all β values are again amazingly consistent. Given the extrapolation difficulties of the kink point method, it is possible to discount reference 23, especially in view of other bulk data to the contrary. The deviant hₕf result is for a non-magnetic impurity system, 111Cd, and is roughly consistent with the maximum deviation expected in the Lovesey Marshall model. As a test of the model, it would be interesting to re-fit the 111Cd data for a smaller temperature range: the effective value of β should decrease. A slight effect of this kind is observed in extending to 10⁻¹⁸ is not anomalous. If hₕf arises largely from long range contributions this is quite consistent with the model.

In summary, I conclude that available theoretical and experimental evidence supports probe independence of hyperfine critical exponents to within a possible 5%. deviation.

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IV. 15 HYPERFINE FIELDS OF TRANSITION ELEMENT IMPURITIES IN GADOLINIUM

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The systematics of hyperfine fields for impurities in iron and nickel is now fairly complete. It has been possible to explain most of the variation of \( B_{hf} \) with impurity atomic number \( Z \) in terms of the Friedel theory for scattering of polarized conduction electrons against the impurity potentials [1]. For d-element impurities where the variation with \( Z \) is strong in these d-band hosts, there are also indications that certain temperature anomalies, \( B_{hf} (T) \), for impurities with "critical" screening charges can be explained by the same theory [2,3], as well as the well-known behaviour of the moment variations of such alloys, \( du/dc \).

For impurities in gadolinium only incomplete information exists so far and practically all of this comes from the hyperfine fields, since the solubility of most elements other than rare earths is very limited. The hyperfine field data existing so far have been collected from nuclear orientation, PAC (perturbed angular correlation) and Mössbauer source experiments. The \( B_{hf} (Z) \) curve for the sp-impurities Ag - I has earlier been shown to be roughly symmetric around the middle of the period which according to Campbell [4] indicates that the position of the Fermi level would be near the middle of the sp-band. The present work is a preliminary account of work on d-element impurities, which has started with the 4d-elements, Mo, Ru, Rh and Pd. A value for the hyperfine field of Nb in Gd has earlier been reported. [5].

Experiments on \(^{95}\)Mo and \(^{106}\)Pd were carried out with IPAC techniques (time-integral PAC) on the 204 and 512 keV states, respectively, from which one obtains the sign as well as the magnitude of the effective hyperfine field. Alloys were prepared by melting the mother elements (\(^{95}\)Mo and \(^{106}\)Ru decaying through \(^{106}\)Rh) with the purest available Gd-metal followed by various heat treatments (1-30 days). Since in a two-phase sample the IPAC method yields only the average \( B_{hf} \), the concentration of the impurities were varied over a large interval in order to find out whether the data represented true single-phased solid solutions in Gd-metal. Typical concentrations were around \( 10^2 \) per cent. The experiments on \(^{99}\)Ru (reported earlier [6]) and \(^{100}\)Rh were performed by the DPAC method using the 90 and 75 keV levels, populated in the decays of \(^{99}\)Rh and \(^{100}\)Pd respectively. Similar metallurgical procedures as mentioned above were performed, although in DPAC one can resolve nuclei precessing in a magnetic hyperfine field from those of possible undissolved atoms yielding no field in the Gd-alloys.

The DPAC experiments were carried out without external field, except for an experiment on Rh in Gd which was specifically designed to determine the sign of \( B_{hf} \). The sign of \( B_{hf} \) for Ru in Gd was difficult to obtain by the radioactive method, but an IMPAC experiment with \(^{102}\)Ru and \(^{104}\)Ru on cooled Gd foils was carried out [7], which indicates a net positive hyperfine field, \( B_{hf} = 65(25) \) kG and a transient field precession of \( \omega/g = 74(10) \) mrad. All experiments in the ferromagnetic region were carried out at 77 K and for the longlived nuclear states in \(^{95}\)Mo, \(^{99}\)Ru and \(^{100}\)Rh additional experiments were performed above \( T_C \) to find out the magnitude of the quadrupole interaction. The results are summarized in Fig. 1 together with some other relevant data for the 3d impurities. The contribution of d-electrons are expected to lie on a background contribution from sp-electrons. The results for GdMo have not been reproducible, but fall into two groups: samples showing a net negative \( B_{hf} = -(5-10) \) kG and samples showing positive \( B_{hf} = +20(30) \) kG (after prolonged heat treatments). The mother activities in this case (Gd-Tc) may have a tendency to form intermetallic compounds with Curie temperatures above 77 K (compare Gd-Mn-alloys) and the positive values probably correspond to segregation of GdTc\(_2\) in the samples.
It is evident that the 4d impurities in Gd show a similar trend in their hyperfine fields as the corresponding impurities in Fe and Ni. The signs of the specific d-contributions are however opposite, which can be understood in terms of Friedel theory since Gd as a transition metal host is near the beginning of the series while Fe and Ni are near the end.

The present experiments, together with the scattered data on hyperfine fields which exist for 3d- and 5d impurities, indicate that the screening charge at the impurities is largely of d-character and that the d-electrons are magnetically polarized. Other experimental evidence that the conduction electron moment in gadolinium metal is carried to a large extent by electrons of d-character comes from the analysis of contributions to the hyperfine field in Gd-metal itself taken together with the fact that the total conduction electron moment is $0.55u_B$. It is found that the net $B_{hf}$ from conduction electrons is very small [8], although electrons of s-character should produce a strong contribution to $B_{hf}$ if they were polarized.

It seems, therefore, that there is strong support for the predictions by the band calculations (Dimmock and Freeman, [9]) that the bands are s-d hybrids with a high density of states near the Fermi surface. The mechanism leading to ferromagnetism in Gd may therefore well be similar to that in the 3d ferromagnets, i.e. that the f-electrons polarize their local d-electrons and the d-electron moments couple ferromagnetically.

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IV. 16

INTERNAL HYPERFINE FIELD ON ARSENIC IN GADOLINIUM

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Earlier we reported our measurements of the internal hyperfine fields on arsenic in cobalt and nickel hosts. Hyperfine field on arsenic in iron has been measured previously by Kol [2] using NMR technique and recently Chopra et al. [3] also reported their measurement. We have also measured the arsenic field in iron and our measured value $H_{\text{int}}(\text{As-Fe}) = +0.05 \mu$Oe is in agreement with their values. In continuation of our investigations of the hyperfine fields, we have chosen gadolinium as the host material. Gadolinium is known to be ferromagnetic below $T_c = 293 \text{K}$ [4]. Here we report our measurement of the internal hyperfine field on arsenic in gadolinium using the integral reversed field method of perturbed angular correlation technique.

In the present work the rotation of the 281.1-279.6 keV $\gamma$-cascade in $\text{As}^{75}$, populated in the decay of $\text{Se}^{75}$, together with the known g-factor of the 279.6 keV state of $\text{As}^{75}$ have been used to determine the hyperfine field.

The radioisotope $\text{Se}^{75}$ obtained from BARC, Bombay in the form of sodium seleno sulphate in neutral solution was converted to selenium bromide and selenium activity electroplated on a specpure gadolinium wire. It was diffused in argon atmosphere firstly at 100°C for 30 hours and then at 200°C for 50 hours. The source was melted and annealed for 6 hours at 400°C.

A conventional fast slow coincidence set up with two identical NaI(Tl) detectors was used for the present measurements. For rotation measurements, metallic source was immersed in liquid nitrogen and measurements were performed at $77 \text{K}$. The arrangement for performing experiment at liquid nitrogen temperature was designed so that drilling a hole in the magnet was avoided and the temperature was checked to be constant at the source. The details of the arrangement are given elsewhere [3].

An external polarizing field of 5 kOe was applied perpendicular to the plane of the detectors, the detectors were kept at a distance of 10 cm from the source and at 135° with respect to each other. For measurement of $\omega C$, the quantity $R$ defined as $R = 2(C_{\text{up}} - C_{\text{down}})/(C_{\text{up}} + C_{\text{down}})$ was calculated from the coincidence counts collected after correcting for chance coincidences. The quantity $R$ is related to $\omega C$ as

$$R = \frac{4 C_2 \omega C}{4 + 2 \omega C}$$

$C_2$ was taken to be unity because of the absence of any time dependent perturbations.

The measured value of $R = 0.037 \pm 0.003$ corresponds to a rotation $\omega C = 0.135 \pm 0.011$. The internal hyperfine field acting on arsenic nuclei was extracted from the relation

$$\omega C = \frac{s}{2} \mu_{\text{n}} H_{\text{eff}} \tau / h$$

where $s = 0.36 \pm 0.04$ and $\tau = (4.67 \pm 0.43) \times 10^{-10}$ sec are the nuclear g-factor and the lifetime of the 279.6 keV level of $\text{As}^{75}$, g-factor and lifetime values are the weighted averages.
of the previous measurements\textsuperscript{1}). Since the lifetime of the nuclear state is small, electrical interactions are neglected in this case. The value of the hyperfine field was evaluated to be

\[
H_{\text{int}} (\text{As} = \text{Gd}, 7\gamma K) = - (166 \pm 27) \text{ koe}
\]

Campbell\textsuperscript{6)} has extended the Daniel-Friedal theory of conduction electron polarization and predicted the hyperfine fields on s-p impurities in Gd. For 5 s-p impurities in gadolinium, the comparison of experimental results and Campbell's predictions are given in ref. (7). Arsenic belongs to a class of s-p impurities in the 4 s-p region. Besides our measurement reported here, there is one more measurement available in this region i.e., for selenium in gadolinium for which \(H_{\text{int}} (\text{Se-Gd}) = 271 \pm 63 \text{ koe}\). It is difficult to comment on the systematic trend with only two measurements in this region. For better understanding further measurements in this region will be helpful.

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The production of large hyperfine fields at the nucleus of atoms dissolved in a ferromagnetic lattice is by now well established. For the rare earth elements, the problems of solubility associated with using an iron host lattice may be avoided if this is replaced by gadolinium. It is expected that all rare earth elements, with the possible exception of europium and ytterbium, should form a continuous range of solid solutions with gadolinium. Furthermore, the magnetic hyperfine interaction may be represented by a simple axial magnetic field which may be as large as 7 Mgauss. This is ideally suited to nuclear orientation and perturbed directional distribution experiments.

In a preliminary experiment $^{177}$Lu was dissolved in gadolinium and cooled to 10 mK. The anisotropy of the 208 keV $\gamma$-ray of $^{177}$Hf, in the direction of the 6 kgauss applied field, is shown as a function of temperature in Figure 1. The Figure includes theoretical curves calculated with the assumptions of allowed Gamow-Teller $\beta$-decay to the 321 keV level and an E1 $\gamma$-transition. Curve (2) is the best fit to the data for a pure magnetic interaction. With a magnetic moment of 2.24 n.m. (Petersen and Shugart, 1962) it corresponds to a hyperfine field of 256 kgauss; in excellent agreement with 260(25) kgauss measured by Zorra et al. (1969).

The other theoretical curves contain a small quadrupole interaction which may arise from the interaction between the electric field gradient due to the hexagonal gadolinium lattice and the nuclear quadrupole moment of 5.51(6) barn (Petersen and Shugart, 1962). The measurement shows that the average effective field gradient is less than $1.8 \times 10^{21}$ V.m.$^{-2}$. Restricting the hyperfine field to 260(25) kgauss would limit this to less than $0.6 \times 10^{21}$ V.m. $^{-2}$. A value of $0.9 \times 10^{21}$ V.m$^{-2}$ has been measured by Göring (1972) for pure gadolinium. The present measurement is likely to be considerably smaller since it deals with an average, due to the polycrystalline nature of the gadolinium lattice.

The agreement with earlier results indicates that a homogeneous solid solution of lutecium in gadolinium has been formed and that thermal contact is good down to 10 mK. It further shows that the applied field of 6 kgauss is sufficient to polarize the gadolinium domains.
The hyperfine interactions expected for most other rare earths in gadolinium are about an order of magnitude larger than that for lutecium since they possess an unfilled shell of 4f electrons. If the exchange field in gadolinium is sufficient to create an electronic ground state for the rare earth impurity, well separated from other electronic levels, with \( J_z = J \), then the hyperfine interaction will be similar to that observed in a free rare earth ion. Its nuclear Hamiltonian will then be axial and of the form

\[
K = -g_P n_H \mu_B + \mathbf{P} \cdot \mathbf{I}^2 - \frac{1}{2} \mathbf{I} \cdot \mathbf{I} + 1),
\]

where \( g_P n_H \) is the nuclear magnetic moment of the rare earth impurity. Values of the hyperfine magnetic field and quadrupole interaction observed for free rare earth ions are given in Table 1. Kobayashi et al. (1967) have measured the hyperfine interaction at several rare earth nuclei dissolved in gadolinium. The measured magnetic fields are also included in Table 1, the difference of about 10% may be attributable to fields arising from other causes.

| Ion   | \( J \) | Hyperfine field \( M \) gauss | \( |\mathbf{I} \cdot (\mathbf{I}-1)\mathbf{P} \times 10^{18}\) | Hyperfine fields as measured by Kobayashi et al. (1967), M gauss |
|-------|-------|-----------------------------|--------------------------------|---------------------------------------------------------------|
| Ce\(^{3+}\) | 5/2   | 1.9                         | 1.11Q                          | Table 1: Hyperfine fields and quadrupole interactions as measured for the free rare earth ions. The values measured by Kobayashi et al. (1967) for the ions in gadolinium are also tabulated. |
| Pr\(^{3+}\) | 4     | 3.14                        | 1.27Q                          | |
| Nd\(^{3+}\) | 9/2   | 4.30                        | 0.60Q                          | 3.62 |
| Pm\(^{3+}\) | 4     | 4.18                        | -0.26Q                         | 3.52 |
| Sm\(^{3+}\) | 5/2   | 3.36                        | -1.25Q                         | 3.03 |
| Dy\(^{3+}\) | 6     | 3.14                        | 2.57Q                          | 5.70 |
| Ho\(^{3+}\) | 8     | 7.25                        | 1.20Q                          | 7.28 |
| Er\(^{3+}\) | 15/2  | 7.64                        | -1.31Q                         | 6.31 |
| Tm\(^{3+}\) | 6     | 6.71                        | -3.53Q                         | |
| Yb\(^{3+}\) | 7/2   | 4.13                        | -3.87Q                         | |

The polarization of \(^{160}\)Tb has been achieved by dissolving the activity in gadolinium and cooling to 10 mK as was done for \(^{275}\)Lu. The measured anisotropy as a function of temperature down to 10 mK can be fitted with a theoretical curve which contains a magnetic hyperfine field of 3400 gauss and a quadrupole interaction, \( \mathbf{P} \) of \( 2.0 \times 10^{-18} \) erg. These values are in disagreement with the results of Kobayashi et al. on \(^{160}\)Tb if \( \mu = 1.485(3) \) n.m. and \( Q = 3.0(5) \) barn (Easley et al., 1968) are used for \(^{160}\)Tb. The latter measurements predict values of

\[
H = 3030(3) \text{ gauss} \quad \text{and} \quad \mathbf{P} = 1.3(2) \times 10^{-18} \text{ erg}
\]

Work is continuing to clarify the source of this discrepancy.

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Using $^{111}\text{Cd}$ as a probe, DPAC measurements of the hyperfine interaction in gadolinium metal have been carried out previously by this group [1]. The g-factor $g = 0.32$ of the 247 keV state ($I = 85\text{ns}$) gives a magnetic interaction frequency $\omega_0 = 154 \text{ Mrad/s}$ per 100 kG of magnetic field, enabling the observation of several spin-rotation periods for an interaction of the magnitude expected in the rare earths. Also, the $^{111}\text{In}$ activity was found to be easily soluble in gadolinium metal. These facts encouraged us to start a series of measurements of the hyperfine interaction for $^{111}\text{Cd}$ in the rare earth metals.

The magnetic structure of the heavy rare earth metals is in general of helical type and with the exception of Gd there are at least two temperature regions with different magnetic orderings. Terbium metal has one of the least complicated structures [2]. In the antiferromagnetic region (226-216 K) Tb-metal has the simple helical structure shown in Fig. 1., with the moments lying in the basal plane, turned through an angle from plane to plane. The turn angle is $-20^\circ$, varying a few degrees with temperature. The transition to ferromagnetic structure is of first order. In the ferromagnetic phase the direction of easy magnetization is in the b-direction $\langle110\rangle$. In this case the symmetry axis of the EFG ($c$-axis) lies at $90^\circ$ to the magnetization.

The $^{111}\text{In}$ activity was obtained by irradiating natural Cd metal with 50 MeV Protons in the Uppsala synchrocyclotron. After chemical separation, the activity was evaporated in vacuum onto terbium foils (purity 99.9%). The foils were melted into small spheres of approx. 1 mm diameter by means of an electron gun. The samples were annealed at 1150 K for 20 hours. For the measurements in the ferromagnetic region the spherical terbium samples were used directly. The samples used in the antiferromagnetic region were filings from the spheres. This technique minimizes the influence of texture effects. The possible presence of texture was monitored by observing the electric quadrupole interaction at room temperature. At room temperature, the experimental time spectrum was fitted to the theoretical expression $G_{22}(t)$ for pure quadrupole interaction, yielding $\omega_E = 4.23$ (6) Mrad/s. Fechner et al obtained $\omega_E = 4.07(7)$ Mrad/s [3]. No definite indication of $\eta \neq 0$ was found, but an attenuation of the amplitude due to a small frequency spread was visible (Fig. 2). The $G_{22}(t)$ is very sensitive to texture [4] and samples showing such effects were discarded.

In the ferromagnetic region the combined electric and magnetic interaction [5] for $\eta = 0$, $\beta = 90^\circ$ is expected. A $G_{22}(t)$ curve taken at 77 K is shown in Fig. 3. The fitted parameters at 77 K are $\omega_B = 384(1)\text{Mrad/s}$, $\omega_E = 4.35(19)\text{Mrad/s}$. Runs were made in the ferromagnetic region at the temperatures shown in Fig. 5. The hyperfine field at 15 K is $-264(5)\text{kG}$. The sign was determined in an external field measurement.

In the antiferromagnetic region $G_{22}(t)$ was obtained at one temperature in the middle of the 216-220 K range. This curve shows a very strong damping (Fig. 4).
The absolute value of $B^{\text{hf}}_{ij}$ at low temperatures can be compared to $B^{\text{hf}}_{\text{Gd}} (= -314 \text{ kG})$. These two field-values are roughly proportional to the $(g_j - 1)$-values for the hosts. The temperature dependence in the ferromagnetic state is compared to a Brillouin function for $I = 6$ in Fig. 5. The local spin density at the impurity is proportional to the magnetization. In the antiferromagnetic state the data can be interpreted in terms of a broad distribution of magnetic frequencies.

If the spin-polarization mechanism is of short range the hyperfine field of a non-magnetic impurity is sensitive to the distribution of neighbouring host spins. For a long-range mechanism a zero net magnetic hyperfine field should be expected in the a.f. region.

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IV. 19 The Temperature Dependence of the Magnetic Hyperfine Field at a

$^{111}$Cd Impurity in Ferromagnetic Holmium

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The magnetic hyperfine (hf) field at impurity sites in ferromagnetic metals has been investigated in the past mostly in the 3d ferromagnets Fe, Co and Ni. Only recently the studies have been extended to the Rare Earth (R.E.) metals (1, 2). Most of the 4f-elements exhibit a rather complex magnetic order below room temperature and are thus well suited hosts for further investigations of impurity hf fields.

In this contribution we report about a TDPAC measurement of the temperature dependence of the magnetic hf field acting on $^{111}$Cd impurities in metallic Holmium.

As all heavy R.E. metals, Ho has a hcp crystal structure with a c/a-ratio smaller than the ideal value of 1.633 for closest packing. At low temperatures two phases of different spontaneous magnetic order have been found: Up to the Curie-temperature $T_C \approx 200K$ one observes a conical ferromagnetic screw-type structure, whereas between $T_C$ and the Neel-temperature $T_N \approx 1320K$ the order of the 4f-moments is of the antiferromagnetic spiral type (3).

An impurity on a regular substitutional lattice position of magnetically ordered Ho will experience both a magnetic hf field and an axially symmetric electric field-gradient (EFG), which is caused by the surrounding lattice ions and the conduction electrons. In the case of Ho, the magnetic hf field and the EFG are not collinear, but form a finite angle $\beta$, which should be close to the angle between the direction of the 4f-moments and the c-axis. The perturbation of an angular correlation by a combined electric and magnetic hf interaction in a polycrystalline sample has been treated theoretically by Boström et al.(4).

For an axially symmetric EFG the perturbation factor $G_{kk}(t)$ depends only on the angle $\beta$ and the magnetic and electric interaction frequencies $\omega_B = g \mu_B H / h$, $\omega_Q = eQV_{zz}/4I(2I-1)h$.

In our experiment solid solutions of metallic Ho and radioactive, carrierfree $^{111}$In were prepared by electron gun melting of the two components. The radioactive impurity concentration was less than $10^{-5}$ at. %.

A four detector apparatus (5) was used to measure the time dependence of the angular correlation of the 73-247 keV cascade of $^{111}$Cd which is populated by the EC decay of $^{111}$In. The source was mounted in a He cryostat which allowed a controlled variation of the source temperature. Fig. 1 shows as an example the time spectrum of $A_{22}(t)$ measured at 79°K. The intersection parameters $\omega_B$, $\omega_Q$ and $\beta$ were derived from the measured perturbation factors by a least squares fit program which was based on the theoretical expression for $G_{kk}(t)$ derived in ref.(4).

In the analysis of the data it turned out, that a satisfactory fit to the time spectra $A_{22}(t)$ could only be obtained by assuming that at all temperatures the Cd impurities occupy two distinct sites in the Ho lattice, where they are subject to different magnetic hf fields. At 4.2°K the field at the two sites, which are about equally populated, is 159(3) kG and 139(3) kG, respectively. The observation of two sites for Cd in Ho is unexpected as in all experiments with impurities in Ho (1) and other comparable heavy R.E. metals (2) one has found only one impurity position. Our data do not permit a definite conclusion about the nature of the two sites. Most probably one is a regular substitutional lattice position with undamaged neighbourhood. The other might be either an interstitial position or a regular site with a dislocation nearby.

Fig. 2 shows the relative temperature dependence of the magnetic hf field at the high and the low field site. The values of $\omega_Q$ and $\beta$ obtained from the analysis shall not be discussed here. At both impurity sites the magnetic hf field has a nearly linear temperature
Fig. 2: Temperature dependence of the hf field of $^{119}$Sn in Ho (open circles and dashed line in fig. 2) deviates even stronger from the magnetization curve, which indicates an influence of the impurity properties on the temperature dependence of the hf field.

Since Cd is a closed shell impurity, it is improbable, that the observed anomalous temperature dependence is due to the existence of a local moment at the impurity. One possible explanation for the anomaly would be a decrease of the spin susceptibility of the conduction electrons with increasing temperature, caused either by changes in the density of states near the Fermi surface or by an increased electron-electron scattering, which can lead to a disordering of the spins. The temperature dependence of the interlayer turn angle in Ho indicates the existence of such an effect, its size however, is at present difficult to estimate.

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IV. 20 TDPAC STUDY OF THE MAGNETIC HYPERFINE INTERACTIONS FOR CALCIUM IN HEUSLER ALLOYS

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The hyperfine fields in Heusler alloys of the type $X_{2}MnY$ have been extensively studied in a number of laboratories using mainly nuclear magnetic resonance and Mössbauer techniques. In this contribution we present results of the magnitude and sign of magnetic hyperfine fields at the sp Y sites in the Heusler alloys: $Ni_{2}MnIn$, $Cu_{2}MnIn$, $NiMnSb$, $Pd_{2}MnSb$, $PdMnSb$, $Hi_{2}MnSb$ (Table 1).

The samples were prepared by melting the constituents of 3N purity at about 1100°C in evacuated quartz tubes. The products were then crushed and again sintered at: 850°C (50 hours) for $Ni_{2}MnIn$, 535°C (2 days) for $Cu_{2}MnIn$, 650°C (50 hours) for $NiMnSb(In)$, 800°C (50 hours) for $Pd_{2}MnSb(In)$ and $PdMnSb(In)$, 750°C (60 hours) for $Hi_{2}MnSb(In)$. The X-ray diffraction analysis performed for the powdered samples showed the desired cubic phase with the following lattice constants (in Å): 6.076(5), 6.187(5), 5.923(5), 6.42(1), 6.267(5) and 6.010(5) for the above alloys, respectively.

The time-differential perturbed angular correlation technique TDPAC was used for polarized and unpolarized sources. The $A_{2}G_{2}(t)$ coefficients calculated from the measured angular correlations for the 173 - 247 keV cascade, in the decay of $^{111}In$, were then fitted (Fig. 1) with an appropriate theoretical function which accounted for the prompt integrated $G_{2}(t)$ factor for the magnetic interaction smeared by a Lorentzian distribution. The sign of the magnetic hyperfine fields was obtained by applying a polarizing external magnetic field of 5 kGs, perpendicular to the detector plane (Fig. 2).

The results can be compared with recent calculations of Blandin and Campbell [1] for the $CuMnIn$ host, with the Cd as the Y site atom, for which the calculated $H$ is equal to -180 kGs. As pointed out in Ref. 1, the fields for the Y site atoms in the $Ni_{2}MnSb$ and also $Pd_{2}MnSb$ hosts should be identical to those in $Cu_{2}MnIn$. Our results are in satisfactory agreement with their predictions (see Table 1).

![Fig. 1. $A_{2}G_{2}$ coefficient for $^{111}Cd$ at Y site in $Cu_{2}MnIn$. The full line is the least squares fit with the best parameters.](image-url)
Table 1

<table>
<thead>
<tr>
<th>Host</th>
<th>Y site atom</th>
<th>$T_c$(°K)(^a)</th>
<th>$T$(°K)</th>
<th>$H_r$(kGs)</th>
<th>$H_o$(kGs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni(_2)MnIn</td>
<td>Cd</td>
<td>323 ± 10</td>
<td>107 ± 4</td>
<td>-150.0 ± 2.1</td>
<td>-155.0 ± 2.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>296 ± 2</td>
<td></td>
<td>-70.3 ± 1.6</td>
<td>-171.0 ± 30</td>
</tr>
<tr>
<td>Cu(_2)MnIn</td>
<td>Cd</td>
<td>533 ± 10</td>
<td>124 ± 4</td>
<td>-210.4 ± 3.0</td>
<td>-213.1 ± 3.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>295 ± 4</td>
<td></td>
<td>-183.0 ± 2.2</td>
<td>-216.4 ± 2.5</td>
</tr>
<tr>
<td>NiMnSb</td>
<td>Cd</td>
<td>720 ± 10</td>
<td>95 ± 4</td>
<td>-210.8 ± 4.1</td>
<td>-211.3 ± 4.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>293 ± 2</td>
<td></td>
<td>-194.3 ± 3.8</td>
<td>-207.9 ± 4.1</td>
</tr>
<tr>
<td>Ni(_2)MnSb</td>
<td>Cd</td>
<td>381 ± 5</td>
<td>85 ± 4</td>
<td>222.8 ± 3.0</td>
<td>225.1 ± 3.1</td>
</tr>
<tr>
<td>Pd(_2)MnSb(^b))</td>
<td>Cd</td>
<td>247 ± 10</td>
<td>90 ± 4</td>
<td>223.9 ± 4.7</td>
<td>235.7 ± 5.3</td>
</tr>
<tr>
<td>PdMnSb(^b))</td>
<td>Cd</td>
<td>500</td>
<td>90 ± 4</td>
<td>255 ± 8</td>
<td>256 ± 10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>297 ± 3</td>
<td></td>
<td>194 ± 8</td>
<td>235 ± 10</td>
</tr>
</tbody>
</table>

\(^a\) Values of $T_c$ were taken from Ref. 2.

\(^b\) Preliminary results

Fig. 2. Experimental $R(t)$ curves for polarized Heusler alloy hosts and water solution of an InCl\(_3\) source used for callibration.

IV. 21 SPIN-FLOP TRANSITION IN MnCl₂·4H₂O STUDIED BY NUCLEAR ORIENTATION
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It has been demonstrated that low temperature nuclear orientation can be used to study the spin-flop transition in an antiferromagnetic crystal. On cooling the crystal to low temperatures, the nuclear spins will be oriented by hyperfine interaction, and the nuclear orientation can be measured by observing the γ-ray intensity from radioactive nuclei doped into the sample. The nuclear spin quantization axis is defined by the orientation of the electron spins so that any change in direction of the latter will cause a change in the observed γ-emission. We report here an experiment investigating this effect in MnCl₂·4H₂O using ⁵¹Mn.

The crystal structure of MnCl₂·4H₂O is monoclinic and its Néel point is in the vicinity of 1.61 K. The easy (or preferred) axis of magnetization is usually taken to be the c-axis although recent measurements indicate that, in fact, it is tilted from this axis by an angle of 11° in the ac plane. The low temperature heat capacity of the salt has been measured by Miedema et al. and a hyperfine field of \( H = 650 \pm 12 \text{kOe} \) can be deduced from their data. These authors present a very useful discussion of the magnetic properties of the salt.

In the molecular field model for the uniaxial antiferromagnet, which has two sublattice magnetizations \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \), the exchange interaction is represented by a field \( \mathbf{H}_\text{ex} = \lambda \mathbf{M} \), where \( \lambda \) is a constant. The anisotropy energy is represented by terms \( K/2 \cos^2 \theta \), where \( K \) is a constant and \( \theta \) is the angle between \( H \) and the easy axis. It is convenient to define an 'anisotropy field' of magnitude \( H_\text{A} = K/M \) and fields \( H_1 = 2H_2 = H_\lambda \).

When a magnetic field, \( H \), is applied along the easy axis, it can be shown that the antiferromagnetic state \( (\theta = 0; \varphi = \pi) \) is stable up to a field \( H_\text{c} = H_\lambda \). This is the zero-frequency resonance field, sometimes erroneously taken as the spin-flop field. The spin-flopped phase \( (\theta = \pi; \varphi = \pi) \) is stable for fields up to \( H_\text{c} = H_\lambda \). The field in which the antiferromagnetic and spin-flopped states have equal energy is

\[
H_\text{c} = \frac{1}{2} \left[ H_\lambda \right]^2,
\]

and this is the field at which the spin-flop transition occurs in the thermodynamically stable system. An analysis of the spin-dynamics shows that in the spin-flopped phase, increasing the applied field causes the magnetizations \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \) to change their orientation so that

\[
\cos \theta = H/H_\text{c}.
\]

The system remains in this phase until \( H > H_\text{c} \), when \( \mathbf{M}_1 \neq \mathbf{M}_2 \) with \( \theta = \varphi = 0 \). In MnCl₂·4H₂O the spin-flop transition is reported to occur at \( H_\text{c} = 7.0 \text{kOe} \) at 1 K and \( H_\text{c} = 7.4 \text{kOe} \) at 0.13 K.

In an antiferromagnet, nuclear orientation can be produced by utilizing the hyperfine interaction and cooling to low temperatures. Daniels et al. have demonstrated this effect in a number of salts, including MnCl₂·4H₂O. The γ-ray intensity distribution for ⁵¹Mn is given by

\[
W(\theta) = 1 + A_1 B_1 P_2(\cos \theta) + A_2 B_2 P_4(\cos \theta).
\]

Here \( \theta \) is the angle between the axis of orientation (defined by the electronic magnetization) and the direction of observation. \( A_1 \) and \( A_2 \) are radioactive decay parameters, \( B_1 \) and \( B_2 \) are measures of the degree of order in the nuclear spin ensemble and \( P_2 \) and \( P_4 \) are the Legendre polynomials. In the past, nuclear orientation has been used to obtain information concerning the decay scheme through the \( A \) terms and to measure g-factors and hyperfine fields through the \( B \) terms. Also the technique has been used for thermometry. The novelty of the present experiment is the change in \( \theta \) observed through the phase transition, allowing determinations of \( H_\text{c} \) and the new directions in which the electron spins point.

A single crystal of MnCl₂·4H₂O doped with 15 µCi of ⁵¹Mn was glued to a copper cold finger connected to a chrome potassium alum pill in a demagnetization cryostat. The crystal was oriented so that the c-axis was vertical. A number of counters (3 or 4) detected the γ-radiation emitted in various directions in a number of runs. A superconducting solenoid produced fields up to 10 kOe along the c-axis.

After demagnetization the copper cold finger cooled to about 10 k. The crystal cooled to about 70 mK in approximately 10 minutes, after which time it cooled at a very slow rate (about 4 mK per hour). The large temperature difference between cold finger and specimen, and the slow cooling rate can be explained by the thermal barrier at the copper-crystal interface and the relatively huge thermal capacity of the crystal. The dominant contribution to which is that of the ⁵¹Mn spins. The effects of spin-lattice relaxation may also be important. Unfortunately we were unable to resonate the ⁵¹Mn spins in order to

![Fig. 1. A typical run through the spin-flop transition. Counters A, B and C measured the normalized intensity of γ-radiation, \( W(\theta) \), in various directions. In this run C was aligned along the c-axis while A and B were in the equatorial a-b plane with angles to the b axis of 130° and 38° respectively.](image-url)
measure the relaxation rate. The observed cooling was greater than that achieved by Daniels et al.

Measurements of $W(0)$ in zero applied field indicate that indeed the easy axis is tilted from the $c$-axis. We determine the angle of inclination to be $12 \pm 6^\circ$ with the easy axis lying in the $ac$ plane to within $\pm 10^\circ$. We intend to improve the accuracy of these measurements in future experiments. As the applied field is increased, the electronic magnetization remains aligned very near the $c$-axis (the angle between $H$ and easy axis is very small) until the spin-flop occurs. This was indicated by a dramatic change in the count rates. Figure 1 shows a typical run in which the magnetic field was increased through the spin-flop transition.

We measure $H_c = 6.70 \pm 0.1$ kOe with a width $\pm 0.2$ kOe. It should be emphasized that this measurement is effectively for $T=0$, an important merit of this technique. We can also determine the orientation of the electron spins after the transition. At $H = 7.0$ kOe, they lie within $\pm 5^\circ$ of the $bc$ plane, making an angle of $\phi = 71 \pm 5^\circ$ with the $c$-axis. At $H = 9.2$ kOe, $\phi = 66 \pm 5^\circ$. The errors in $\phi$ include the statistical error of $1.5^\circ$ and a systematic error in counter alignment of $4^\circ$.

Using molecular field theory we deduce $H_p = 11.7 \pm 0.6$ kOe and $H_s = 2.0 \pm 0.1$ kOe, in good agreement with the values of $12.8$ kOe and $2.2$ kOe quoted by Miedema et al.

References
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Nuclear Orientation in Concentrated AuCo Alloys

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The binary alloy AuCo is an example of a system in which the magnetic behavior of the impurities is strongly influenced by impurity-impurity interactions. This system has been studied extensively using macroscopic methods (specific heat, magnetization)\(^1,2\). The authors of these studies proposed a model for the effect of impurity interactions, which assumed groups of three or more Co nearest-neighbor atoms to have a local magnetic moment. Nearest neighbor pairs and isolated Co atoms were taken to be non-magnetic, with enhanced local susceptibilities and spin fluctuation temperatures \(T_{sf}\) of ca. 22.5K and 190K, respectively. This model, interpreted in terms of spin magnetism alone, led to contradictions although it was able to account for the major features of the experimental results. Hyperfine interactions measurements\(^3-5\), in which only the isolated Co atoms were studied, indicated that orbital magnetism also plays an important role. Theoretically, it is expected that the orbital susceptibility should be important in the non-magnetic limit\(^6\). As the magnetic instability is approached, the orbital susceptibility becomes nearly constant while the spin susceptibility increases rapidly. Thus for the relatively non-magnetic isolated Co impurities, a significant orbital contribution is not surprising. The orbital contribution is in any case overweighted in a hyperfine measurement due to the large orbital hyperfine field constant. Taking the presence of both spin and orbital magnetism into account, the authors of (2) have obtained a more consistent explanation of their data, although it appears that a simple model assuming independent spin and orbital contributions is inadequate for this case\(^4,5\).

To provide a further test of this hypothesis of an increasing dominance of spin magnetism with increasing Co-Co interactions, we have performed nuclear orientation measurements on a series of concentrated AuCo alloys (0.9-11 \% At. Co) in applied fields of 1.5-7.2 Tesla and at temperatures in the range 7-30mK. Samples were prepared by melting followed by rapid quenching and were activated with \(^60\)Co either by diffusion and quenching or by direct neutron irradiation. Both \(\gamma\)-ray and \(\beta\)-particle angular distributions from the oriented \(^60\)Co were measured.

The \(\beta\)-particle studies give the sign of the average hyperfine field while the \(\gamma\)-ray results give the magnitude of the field. Quantitative interpretation of the \(\beta\)-particle results is not possible because of magnetic focussing of the electrons in the applied fields, but a strong trend towards negative hyperfine fields with increasing Co concentration was observed indicating that spin magnetism is indeed dominant for the interacting Co atoms.

Fig. 1 shows the gamma ray results, interpreted on a naive model which assigns a unique hyperfine field to each type of Co site (isolated atoms = \(N_3\), pairs = \(N_2\), etc.) and assumes the alloys to be homogeneous. The dashed curve labelled \(N_3\) was known from the previous work on dilute alloys. Two features of these curves are immediately apparent: first, the hyperfine field of the "magnetic" sites (\(N_3, N_4\)) is not saturated even at the highest applied fields. (Examination of the data shows also a strong positive temperature dependence for these fields). This is an indication that a randomly oriented magnetic interaction (random molecular field)\(^8\) is preventing complete alignment of the local moments and reducing the apparent hyperfine fields. The second anomaly in Fig. 1 is the fact that the curve for the pairs (\(N_2\)) does not \(\beta\)\(\gamma\) to rise smoothly from 0; it has either a large initial slope followed by a slow rise at higher applied fields, or a non-zero intercept at zero applied field. In any case a straightforward interpretation of this curve indicates either a large negative hyperfine field or a small negative Knight shift for the pairs; the assumption of a positive field leads to the unphysical result of a hyperfine field which decreases with increasing applied field.
We have performed calculations based on various random molecular field models, which indicate that the raw anisotropy data can be fit satisfactorily in this manner, although quantitative agreement with the individual hyperfine field curves was not obtained with any of the simple models. We conclude that the increasing role of spin magnetism with increasing Co-Co interactions is verified, with a dominant spin contribution appearing on the Co pairs. The slow saturation of the magnetic sites is in disagreement with the interpretation of bulk magnetization measurements², and the apparent non zero intercept for the pair hyperfine fields remains unexplained (a similar effect was seen in nuclear specific heat measurements⁷).

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7) P. Costa-Ribeiro, Thèse, Université de Grenoble (1973), unpublished
Much progress has recently been made in Mössbauer studies of Rare Earth (RE) impurities in metals (1,2). Because of their well defined electronic energy states, the RE elements lend themselves to the study of the static and dynamic hyperfine (hf) interactions. Here we wish to report the influence of small external magnetic fields (<1 KG) on the hf structure and relaxation of dilute Yb impurities in Au as seen through the Mössbauer effect.

From previous studies (1,3) the crystalline electric field (CEF) ground state of Yb impurities in Au is known to be a $\Gamma^2$ Kramers’ doublet well separated from the excited CEF states $\Gamma_3$ and $\Gamma_5$ which lie more than 80K higher. Thus the magnetic isotropy of the $\Gamma^2$ CEF ground state is unaffected by weak magnetic fields. Furthermore in the liquid helium range only the ground state contributes to the hf spectrum, which in the presence of an external field $H_{\text{ext}}$ is then described by:

$$\mathcal{H} = A_B (S_z^2) + g_s g^* \mu_B H_{\text{ext}} S_z$$

Here $S=1/2$ is the effective spin, $A_B$ is the effective spin hf coupling constant and $g$ is the $\Gamma^2$ g-factor. For the system under consideration $A_B=13.3$ mm/sec (646 Gauss) and $g=3.34$ are known (4,5).

For the $2\rightarrow 0$ Mössbauer transition of $^{170}$Yb two resonance lines at $a_2$ and $-3/2a_2$ are obtained in the zero field limit with relative intensities 12 and 8, respectively. The labels $i\leq j$ correspond to transitions between excited hf states $| i \rangle$ ($i=1...10$) and ground hf states $| \rangle$ ($\langle i \rangle$). In analogy to atomic hf spectroscopy we will refer to Fig. 1a as the Breit-Rabi diagram.

For $H_{\text{ext}}=0$ Fig. 1a provides the two line asymmetric spectrum mentioned above. For $H_{\text{ext}}<1000G$ the transition energies split into three groups: at high energy, at low energy and at $-1<2E$ respectively. In this field region all transitions have comparable intensities, although the outermost lines become rapidly weaker with increasing field (cp. Fig. 1b). Above 1000G only the ten central lines are present while the intensities of the outer lines vanish. At still higher fields the ten central lines, which are all equally intense, merge into 5 lines indicated by arrows in Fig. 1a. This is the well known “effective field” spectrum which is commonly observed for magnetically ordered systems. The variation of the hf spectrum can be understood with the help of Eq. (1) and its eigenfunctions, which are found to have the general form

$$| i \rangle = a_{1/2} | 2I_z = 1/2 \rangle + b_{-1/2} | 2I_z = -1/2 \rangle$$

$$| j \rangle = 0, 1/2, 1, 3/2 \rangle$$

In raising the field the diagonal Zeeman term gradually “quenches” the off-diagonal elements of the term $A_B (S_z^2)$. Thus the wavefunctions $| i \rangle$ of Eq. (2) approach pure angular momentum eigenstates ($a_{1/2}$ or $b_{-1/2}$). As a result the respective intensities $a_{1/2}^2$ or $b_{-1/2}^2$ which belong to the outermost lines in Fig. 1a vanish.

In a Mössbauer experiment with the external field applied along the direction of gamma ray propagation only transitions with $\Delta I_z = 1$ are observed because of gamma ray polarization. This leaves eight Mössbauer lines which are labeled $7+, 6+, 5+, 4+, 3-, 2-, 1-$ and $0-$ in Fig. 1. It is striking that the polarization selection rule $\Delta I_z = 1$ is in effect even for very small fields, due to the complete magnetic isotropy of the system.

In Fig. 2 the Mössbauer spectra of Yb impurities in Au is shown at 1.4K in $H_{\text{ext}}=0$ and 400G. Like in previous studies (1,2) the alloys, containing 400ppm of impurities, were investigated as the source, where the decay of $^{170}$Yb ($T_{1/2}=130d$) feeds the $84.3$ keV $2\rightarrow 0$ Mössbauer transition in $^{170}$Yb. The spectra clearly show a marked variation in changing the external field and the line positions and intensities can be well accounted for by the Breit Rabi diagram. In the analysis we have allowed for a finite relaxation rate. The solid curves through the data points represent such a fit using a microscopic relaxation theory (3,4). Both spectra yield a unique value for the relaxation rate, independent of field. Such a field independent rate for this system is indeed expected since for small fields the Zeeman and hf splitting of the doublet is much smaller than the bath temperature leading to a Korringa type of behavior.

References:
The Sm$^{3+}$ ion (4$f^5$, 6$^2_1$), unlike most other rare earth ions, exhibits unusual magnetic properties, which arise because of its closely spaced multiplet levels. The energy separation between the ground ($J = 5/2$) level and the first excited ($J = 7/2$) level is only about 1400 K, as a result of which, the applied and the exchange fields admix these levels giving rise to anomalous behaviour of the magnetic susceptibility, the Knight shift, and the 4f induced hyperfine field at the samarium nuclear site in paramagnetic compounds. When embedded in a crystalline lattice, the crystalline electric fields also admix higher $J$ levels of Sm$^{3+}$ into its ground level introducing further new features in the magnetic behaviour of Sm$^{3+}$ ions (4-6). Recently Malik and Vijayaraghavan (7) have shown that the hyperfine field $<E^f_{\text{av}} >$ at the samarium nuclear site produced by the paramagnetism of the 4f electrons may become negative because of strong (cubic) crystal fields and exchange interactions. Such an effect can be easily verified by measuring, using perturbed angular correlation technique, the paramagnetic correction factor $\beta$, which is the ratio of the field $H_{\text{eff}}$ seen by the nucleus to the applied field $H$. Neglecting small contributions to the field such as those arising from the core polarisation and the conduction electron polarisation, we can write $\beta = 1 + <E^f_{\text{av}} > / H$.

In this paper we report the measurement of $\beta$ for Sm$^{3+}$ ions at various temperatures in paramagnetic Eu$\text{II}_2$ lattice. The measurements were made using $^{152}$Eu radioactive isotope (containing about $10\%$ $^{154}$Eu as impurity) which decays to levels in $^{152}$Sm. The $^{154}$Eu activity did not interfere with the measurements. Eu$\text{II}_2$ was chosen for these measurements because $^{152}$Eu activity can be produced at the lattice sites by neutron irradiation, and moreover, from other measurements the crystal field interactions are known to be appreciable in Rare Earth-Al$_2$ compounds (6). The 1408-122 keV gamma cascade ($2^+ \rightarrow 2^+ \rightarrow 0^+$) was used in the measurements.

The directional correlation in the presence of time dependent perturbations can be written as $H(\theta) = 1 + G_2P_2(\cos\theta) + G_4P_4(\cos\theta)$; where $G_2$ and $G_4$ are the integrated attenuation coefficients. The directional correlation and the mean precession angle $\omega \tau$ were measured at various temperatures. An external field of 19.4 kOe was used for $\omega \tau$ measurements. At 85 K the measurement was also made in an external field of 10.0 kOe. The experimental results are summarised in the table below.

<table>
<thead>
<tr>
<th>Temp (K)</th>
<th>Source form</th>
<th>$\Delta_2$</th>
<th>$G_2$</th>
<th>External Field kOe</th>
<th>$\omega \tau$ rad. sec</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>300 EuCl$_3$(aq.)</td>
<td>0.203 $\pm$ 0.006</td>
<td>1 (b)</td>
<td>19.4 $\pm$ 3.59 $\pm$ 0.28</td>
<td>-72.9 $\pm$ 5.6</td>
<td>$\pm$ 1.10 $\pm$ 0.10</td>
<td></td>
</tr>
<tr>
<td>300 Eu$\text{II}_2$</td>
<td>0.100 $\pm$ 0.006</td>
<td>0.79 $\pm$ 0.04(c)</td>
<td>19.4 $\pm$ 1.05 $\pm$ 0.23</td>
<td>-58.8 $\pm$ 7.6</td>
<td>$\pm$ 0.79 $\pm$ 0.11</td>
<td></td>
</tr>
<tr>
<td>300 Eu$\text{II}_2$</td>
<td>0.149 $\pm$ 0.007</td>
<td>0.74 $\pm$ 0.04(c)</td>
<td>19.4 $\pm$ 0.15 $\pm$ 0.17</td>
<td>-5.4 $\pm$ 6.2</td>
<td>$\pm$ 0.08 $\pm$ 0.10</td>
<td></td>
</tr>
<tr>
<td>85 Eu$\text{II}_2$</td>
<td>0.112 $\pm$ 0.004</td>
<td>0.55 $\pm$ 0.03(c)</td>
<td>19.4 $\pm$ 0.15 $\pm$ 0.22</td>
<td>+65.4 $\pm$ 16.2</td>
<td>-1.45 $\pm$ 0.25</td>
<td></td>
</tr>
<tr>
<td>85 Eu$\text{II}_2$</td>
<td>0.112 $\pm$ 0.004</td>
<td>0.55 $\pm$ 0.03(c)</td>
<td>10.0 $\pm$ 0.15 $\pm$ 0.18</td>
<td>+65.4 $\pm$ 11.9</td>
<td>-2.38 $\pm$ 0.40</td>
<td></td>
</tr>
</tbody>
</table>

(a) Corrected for finite size of the detectors. In this case $\Delta_2 > A_4$. (b) Assuming no extranuclear perturbations in the liquid source. (c) $G_2 = A_2(\text{obs}) / A_2(\text{aq})$. (d) $R_{\text{III}}^{\text{II}} = 2 \text{Coine(up)} - \text{Coine(down)}$. (e) Calculated using $\omega = 28 = 0.345 \pm 0.03$. The experimentally determined values of $\beta$ for Sm$^{3+}$ in Eu$\text{II}_2$ lattice are plotted as a function of temperature in the figure. The corresponding values of $\beta$ in an aqueous solution of EuCl$_3$ at 300 K is also shown and agrees well with the value of 1.15 for free Sm$^{3+}$ ions (3). The $\beta$(Sm$^{3+}$) values in Eu$\text{II}_2$ are less than...
the free ion values at all temperatures of measurements implying that $\langle S_{\text{eff}} \rangle_{\text{av}}/\mathbf{B}$ is negative. Moreover, the $\beta$ factor becomes negative at low temperatures and has a value $\beta = -1.5$ at 85 K. Thus at 85 K, not only is the $\mathbf{A}$ induced hyperfine field negative, but also much larger in magnitude than the applied field. This, we believe, is the first observation of this kind which verifies the theoretical prediction\(7\).

The large negative values of $\langle S_{\text{eff}} \rangle_{\text{av}}/\mathbf{B}$ can be explained in terms of the crystal fields and exchange effects following the calculations of Malik and Vijayaraghavan\(7\). The $\text{Sm}^{3+}$ ions occupying europium sites in $\text{BaAl}_2$ experience crystal fields of cubic symmetry, the strength of which is determined by $A_4 \langle s^4 \rangle$ and $A_6 \langle s^6 \rangle$, the coefficients of the fourth and sixth degree terms, respectively, in the crystal field Hamiltonian. The Hamiltonian consisting of the spin orbit term $\lambda L S$ ($\lambda/\hbar = 410$ K), the crystal field and the Zeeman term (for $\mathbf{H} \parallel \mathbf{a}$) is diagonalised in the lowest three multiplet levels to obtain the energy eigenvalues and eigenfunctions. The effect of exchange interaction between $\text{Sm}$ and $\text{Eu}$ ions is included as a perturbation. The quantity $\langle S_{\text{eff}} \rangle_{\text{av}}/\mathbf{B}$ is calculated by taking the expectation value of the operator $S_{\text{eff}}^2$ over each level and subsequently Boltzmann averaging over all the levels. The $\beta$ values could be fitted for several combinations of $A_4 \langle s^4 \rangle/\lambda$, $A_6 \langle s^6 \rangle/\lambda$ and the exchange constant $\Delta_{\text{eff}} /x$. Some of the fits obtained are shown in the figure where the curves are labelled with the values of $A_4 \langle s^4 \rangle/\lambda$, $A_6 \langle s^6 \rangle/\lambda$ and $\Delta_{\text{eff}} /x$ in Kelvin. Although $\text{BaAl}_2$ orders antiferromagnetically (negative $\Delta_{\text{eff}}$ for $\text{Eu}-\text{Eu}$ interaction) we used positive $\Delta_{\text{eff}}$ for $\text{Sm}-\text{Eu}$ interaction in order to explain the results. It has been shown earlier\(6\) that in compounds of the type $\text{R}_2\text{BaAl}_2$ ($\text{R}^3$ = rare earths) the exchange interaction between $\text{R}$ and $\text{R}^3$ favours ferromagnetic alignment of the spins (with exceptions). Thus a positive value of $\Delta_{\text{eff}}$ ($\text{Sm-Eu}$) is justified.

REFERENCES

IV. 25 TEMPERATURE DEPENDENCE OF THE KNIGHT SHIFT AT IMPURITIES IN PALLADIUM

by H. Bertschat, H. Haas, F. Pleiter, E. Recknagel, E. Schlodder and B. Spellmeyer
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Hyperfine fields and Knight shift (KS) in the transition metals palladium and platinum and in some of their alloys have been investigated intensively by Mössbauer- und NMR-techniques. For the pure metals the KS was found to be large and temperature dependent \([i,2]\). It could be explained by a simplified two-band model of s- and d-electrons. The contributions to the KS are given as follows:

\[
KS(T) = a_s x_p^s + a_d x_p^d(T) + \delta x_{\text{ov}}
\]

where \(a_s x_p^s\) denotes the contribution arising from contact hyperfine interaction of s-electrons \(a_d x_p^d\) the d-electron induced core polarisation and \(\delta x_{\text{ov}}\) the orbital contribution. The temperature dependence of the shift was shown to be linearly related to the susceptibility arising from the d-electrons. The KS for palladium has a maximum at 80 K and was measured to be -4.5\% \([1]\).

The application of the method of time-differential observation of perturbed angular distribution following a nuclear reaction (DPAD) may help to get more information about the KS providing the advantages of a wide temperature range which can be used and of infinite dilution as a consequence of the nuclear reaction, if the KS at an impurity site is investigated. To study the different contributions to the KS in a transition metal, the temperature dependence of the KS at an impurity embedded in palladium was measured.

Via the \((p,n)\) and \((d,p)\) reactions it is possible to create Ag and Cd impurities in the palladium metal host.

Silver in palladium: An isomeric state in \(^{108}\text{Ag}\) (215 keV; \(2^+; \tau = 77(7)\) ns \([3]\) was populated by \(^{108}\text{Pd}(p,n)\) and \(^{107}\text{Ag}(d,p)\) reactions. Observing the Larmor frequencies of the isomeric state in palladium and silver, the KS in palladium could be extracted directly from the comparison of the two frequencies, since the KS of Ag in silver is known to be \(KS(\text{AgAg}) = 0.522(2)\%\) \([4]\). The shift at 80 K turned out to be \(KS(\text{AgPd}) = -1.4(3)\%\) \([5]\) in agreement with a value extrapolated from NMR measurements on Ag-Pd alloys \([6]\).

Cadmium in palladium: The KS measurements on the CdPd system were performed by DPAD and DPAC methods using the isomeric \(5/2^+, 248\) keV state in \(^{111}\text{Cd}\) (\(\tau = 123\) ns). A remeasurement of the g-factor for this state yielded \(g = -0.306(1)\) \([7]\). From this value and the Larmor frequencies extracted from the DPAD measurement at different temperatures the KS of Cd in palladium was calculated. At 80 K the shift was found to be \(KS(\text{CdPd}) = -0.9(3)\%\). This value was confirmed by a DPAC measurement on a \(^{111}\text{In}\) source diffused into palladium.

In addition a measurement of the KS at Cd impurity in platinum was performed by the DPAC method on \(^{111}\text{In}\) source diffused into Pt metal foils. As a preliminary result at 300 K we give \(KS(\text{CdPt}) = -1.0(6)\%\), which is to be compared with \(KS(\text{PtPd}) = -4.0\%\) \([2]\). Though the result is not very accurate, it clearly shows the negative sign of the shift.

In fig. 1 the KS versus susceptibility diagram is shown for the three systems PdPd, AgPd and CdPd. The data of PdPd are taken from ref. \([7]\). As in the Pd case it is obvious that the KS is linearly related to the susceptibility within the experimental accuracy. With increasing atomic number \(T\) the temperature dependence becomes less pronounced, but from the negative sign even in the CdPd system it is seen that the core polarisation contribution is still dominant. For high temperatures one would expect the KS from the hyperfine contact interaction of the s-electrons to be identical to that in the simple metal (points a, b and c in fig. 1): a) \(KS(\text{PdPd}) = +0.72\%\) \([7]\), this includes the d-orbital contribution; b) \(KS(\text{AgAg}) = +0.522(2)\%\) \([4]\); c) \(KS(\text{CdCd}) = +0.412(4)\%\) \([8]\). Though for the silver data our experimental results are in good agreement.
with this expectation, there is a discrepancy of about 0.6 % for Cd, somewhat larger than
the estimated error of 0.4 % in the absolute shift, resulting from the experimental uncer-
tainty, the error of the Cd g-factor and the field calibration.

It should be noted that the hyperfine fields of Pd, Ag, Cd in nickel [9] are proportional
to the temperature dependent part of the KS for these elements in palladium.

The influence of radiation damage on the KS as derived by the DPAD method has been neglect-
ed, since for AgPd the DPAD and NMR measurements yielded the same result and for CdPd the
DPAD and DPAC data were consistent. However, the DPAD data for CdPd at very low temperatures
show a larger negative shift, which may be attributed to magnetic impurities in the target
material.

Literature

to be published
to be published
IV. 26 HYPERFINE FIELD AT $^{181}$Ta in Zr$_{1-x}$Hf$_x$Zn$_2$

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The compound ZrZn$_2$ is ferromagnetic, with a moment of about 0.16 $\mu_B$/Zr atom and a transition temperature near 26°K. Both the magnetization and Curie point are sensitive to the method of sample preparation and to impurities. In particular, the substitution of Hf for Zr decreases $T_c$ while Ti increases it. 1) Nuclear orientation of Zr and Nb have established low upper limits for the hyperfine field at the Zr site. 2) Magnetic neutron scattering suggests that much of the spin density lies away from the Zr atoms. 3)

Using TDPAC of the 133-482 KeV cascade in $^{181}$Ta following the decay of $^{181}$Hf as the Zr site, we have investigated the effects of stoichiometry and substitution. The appropriate solution of Zr Hf was first formed by melting. The ingot was filed to powder, cleansed with formic acid and superoxyl to remove iron, and then neutron irradiated to produce $^{181}$Hf. Powdered Zn was added and a cylindrical pellet formed under pressure. This was then heated in an evacuated sealed quartz tube for a few days. The resulting material was found by X-ray analysis to have the ZrZn$_2$ structure (Fig. 1). For the TDPAC measurements, the samples were mounted in a small permanent magnet (<5 kOe) and placed in a helium cryostat. Fig. 2 shows the results for one sample at 4°K and 300°K. Table 1 contains the results for a number of alloy samples.

The effect of reducing the nominal Zn concentration was noted by others to stabilize $T_c$. 4) The rapid decrease in $T_c$ with Hf substitution (roughly as 1-5c) appears to be followed much more slowly in $H_{\text{eff}}$.

References:
2) T. Huidoch, C. E. Ohsen and W. A. Steyert, Phys. Lett. 44A (1973) 413.

<p>| Table 1 |</p>
<table>
<thead>
<tr>
<th>Nominal Composition</th>
<th>$H_{\text{eff}}$ (kOe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr$<em>{0.98}$Hf$</em>{0.02}$Zn$_{2.0}$</td>
<td>$-15.7 \pm 0.6$</td>
</tr>
<tr>
<td>Zr$<em>{0.98}$Hf$</em>{0.02}$Zn$_{1.9}$</td>
<td>$-19.9 \pm 0.5$</td>
</tr>
<tr>
<td>Zr$<em>{0.9}$Hf$</em>{0.1}$Zn$_{1.5}$</td>
<td>$-17.9 \pm 0.6$</td>
</tr>
<tr>
<td>Zr$<em>{0.85}$Hf$</em>{0.15}$Zn$_{1.9}$</td>
<td>$-17.6 \pm 0.4$</td>
</tr>
</tbody>
</table>

Fig. 1. Crystal structure of ZrZn$_2$
Dark circles Zr, open circles Zn.

Fig. 2. TDPAC spectra of Zr$_{0.98}$Hf$_{0.02}$Zn$_{2.0}$
at (a) 350°K (b) 4.2°K.

References:
2) T. Huidoch, C. E. Ohsen and W. A. Steyert, Phys. Lett. 44A (1973) 413.
IV. 27  

The Concentration Dependence of the Hyperfine Field at Cd-111 in Fe_{1-x}Al_x  

by A. Andreeff, H.-J. Hunger, S. Unterricker  

Bergakademie Freiberg, Sektion Physik

The hyperfine field at Cd-111 nuclei in Fe_{1-x}Al_x alloys with different Al-concentrations was measured by TDPAC. Our result for the hyperfine field in pure Fe(99.999%) is (361 ± 8) kOe in good agreement with /1,2/. Fig. 1 shows the hyperfine field for different aluminium concentrations. The strong decrease of $H_{hf}$ between 15 and 20 at% aluminium is connected with the formation of the Fe$_3$Al ordered structure. The TDPAC-curves at 20.4 at% for slow cooled specimens show only one frequency. This means that a two phase region between the disordered α-phase and the Fe$_3$Al-structure could not be observed. In contrast to this the quenched specimens show two frequencies corresponding to the disordered α-phase and a phase with Fe$_3$Al ordering tendency.

The decrease of $H_{hf}$ connected with increasing aluminium concentration up to 15 at% Al can be fitted by

$$H_{hf}^{Cd}(x) = H_{hf}^{Cd}(0)(1-0.50\cdot x).$$

$H_{hf}^{Cd}(0)$ is the hyperfine field in pure iron. From Mössbauer measurements M.B. Stearns /3/ assumed that the decrease of $H_{hf}$ is caused by a dilution effect. Fig. 2 shows the temperature dependence of the reduced hyperfine field in Fe$_3$Al together with the reduced magnetisation. The proportionality of the hyperfine field and the lattice magnetisation is a direct proof that no localised moment exists at the Cd-nuclei.

A simple dilution effect is not able to explain why $H_{hf}$ decreases weaker than the Fe concentration. Therefore we assume that $H_{hf}$ at the Cd-nuclei is produced partly by the conduction electron polarisation which provides a negative hyperfine field and partly by overlapping of the 5s-wave functions of the impurity with the 3d-wave functions of Fe-atom, which provides a positive hyperfine field /3/. The increase of the aluminium concentration produces not only a dilution but also a dilatation of the lattice. Such a lattice dilatation causes a decrease of the overlap term to the hyperfine field. With a very simple model which takes into account the lattice spacing measured by Taylor and Jones /4/ and the dilution effect, we obtain

$$H_{hf}^{Cd}(x) = H_{CEP}^{Cd}(0) \cdot (1-x) + H_{hf}^{Cd}(0) \cdot (1-2.15 \cdot x)$$

From the CEP-field in pure iron measured by M.B. Stearns /3/ ($H_{CEP}^{Cd}(0) = -145$ kOe) we calculated $H_{CEP}^{Cd}(0) = -530$ kOe by using the hyperfine coupling constants of Fe and Al given in /5/. Comparing this with the measured value of $H_{hf} = -360$ kOe in pure iron we get overlap field $H_{hf}^{Cd}(0) = -170$ kOe. From this phenomenological model we obtain a theoretical concentration dependence

$$H_{hf}^{Cd}(x) = H_{hf}^{Cd}(0) \cdot (1-0.46 \cdot x)$$

in good agreement with the experimental result.

/2/ J.I. Cisneros et al., Arkiv för Fysik 38 (1968), 363

*present adress: TU Dresden, Sektion Physik
Fig 1: Concentration dependence of the magnetic hyperfine field in Fe-Al alloys.

Fig 2: Temperature dependence of the reduced hyperfine field (h) at the Cd-III moment in Fe₃Al and the reduced magnetisation (M) of the same sample (T/Tₐ=Curie temperature).
An earlier study of the compound \( \text{Cu}_2\text{FeSn}_3 \) (stannite) by CAMEL, HERMON and SHTRIKMAN\(^{(1)} \) has shown the presence of an antiferromagnetic transition at 7°K. The \( \text{Fe}^{2+} \) Mössbauer resonance above this temperature consists of a doublet (Q.S. = -2.7 mm/sec) and the \( \text{Sn}^{2+} \) resonance of a singlet (J.S. = 1.8 mm/sec with respect to \( \text{BaSnO}_3 \)). The line width of the \( \text{Sn}^{119} \) resonance at 9°K (\( \gamma \) 1.0 mm/sec) indicates cubic site symmetry at the tin atom lattice position. Below the magnetic ordering temperature, the \( \text{Fe}^{57} \) resonance splits into a sextet, from which \( H_{\text{int}} \approx 205 \) Koe, with the internal field perpendicular to the major axis of the efg tensor. The \( \text{Sn}^{119} \) resonance at 4.2°K shows considerable broadening (\( \Gamma_{\text{exp}} \approx 3.3 \) mm/sec) from which \( H_{\text{int}} \approx 20 \) Koe, although the individual components of the magnetic hyperfine spectrum assumed by the authors, could not be resolved.

We have examined the \( \text{Sn}^{119} \) Mössbauer resonance in the related synthetic compound \( \text{Cu}_2\text{MnSn}_3 \) in greater detail, both at room temperature and in the vicinity of the magnetic ordering temperature. At ambient temperature, the \( \text{Sn}^{119} \) resonance consists of a singlet (J.S. (\( \text{SnO}_2 \)) \( \approx 1.37 \) mm/sec, \( \Gamma_{\text{exp}} \approx 1.30 \) mm/sec), and similar parameters are observed at 78°K (J.S. (\( \text{SnO}_2 \)) \( \approx 1.48 \) mm/sec, \( \Gamma_{\text{exp}} \approx 1.49 \) mm/sec), indicating essentially cubic site symmetry at the tin atom lattice site at these temperatures. At 4.2°K, the spectrum shows the presence of a magnetic hyperfine interaction, although the (presumed) six components of the resonance pattern again could not be individually resolved. Initial estimates of the strength of the magnetic field at the tin nucleus indicate \( H_{\text{int}} \approx 12 \) Koe. Further reduction of the sample temperature to 1.6°K did not significantly increase the magnitude of the hyperfine interaction, and from these data it is concluded that essentially the full transferred magnetic hyperfine field has been reached at liquid helium temperature. From the temperature dependence of the \( \text{Fe}^{57} \) resonance data for \( \text{Cu}_2\text{FeSn}_3 \) in the temperature range 1.6° < \( T < 7° \)K, and known ratio of the excited \( \text{Fe}^{57} \) state magnetic moments of \( \text{Fe}^{57} \)\(^{(4)} \) and \( \text{Sn}^{119} \)\(^{(5)} \) \( \left( \frac{\mu_{\text{Fe}^{57}}(\frac{2}{3})}{\mu_{\text{Sn}^{119}}(\frac{2}{3})} = 0.226 \right) \) it is possible to calculate the temperature dependence of the transferred magnetic hyperfine field at the tin site in stannite, and compare this to the observed temperature dependence of \( H_{\text{int}} \) at the tin site in this compound. The significance of these data will be discussed in detail. The temperature dependence of \( H_{\text{int}} \) in \( \text{Cu}_2\text{MnSn}_3 \) below the magnetic ordering temperature and its relationship to the results of a neutron diffraction study\(^{(2,6)} \) will also be discussed. Moreover, the significance of the observed extreme sharpness of the onset of the hyperfine transition with a lowering of the sample temperature near 5°K will be considered.

\( ^{119} \)J.C. and R.H.H : CPH/Groupe Interactions Hyperfines
\( ^{119} \)M.W : Laboratoire de Diffraction Neutronique
\( ^{119} \)Permanent address : School of Chemistry, Rutgers University, NEW BRUNSWICK, NEW JERSEY 08903, U.S.A.

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(6) M. WINTENBERGER, to be published.
Figure 1: Mossbauer spectrum of Cu$_2$MnSnS$_4$ at 77°K and 4.2°K.
IV. SIGN OF THE QUADRUPOLE INTERACTION AT 111Cd IN NON-CUBIC METALS BY β-γ PERTURBED ANGULAR CORRELATIONS.

by E. N. Kaufmann, P. Raghavan and R. S. Raghavan
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and S. J. Ansaldo and R. A. Neumann
Princeton University, Princeton, N. J.

Knowledge of the sign of the nuclear electric quadrupole interaction (QI) is vital to the understanding of electric field gradients in solids. This is especially true for metallic systems in which the sign may be largely determined by the conduction electrons. A significant advance in the methods available to measure the sign of the QI has been the recent introduction of the ρ-γ perturbed angular correlation (PAC) technique, which simplifies the measurement considerably. An important extension of this technique was the demonstration that it is applicable to the 24.7 keV level of 111Cd, which has been utilized to measure the largest number of QI constants by PAC experiments. This opens the way to the determination of the sign of the QI in most of these cases.

In this work we report the sign of the QI at 111Cd embedded in several non-cubic metals. The unique first-forbidden (UFF) β-decay (ΔI=2, yec) of 111Ag (7-day) populates the β-γ cascade

$$\frac{1}{2}^- \rightarrow \frac{3}{2}^+ γ(247), \frac{1}{2}^+ \rightarrow \frac{3}{2}^+ γ(247)$$

through the 24.7 keV (T1/2 = 64 ns) level of 111Cd. As a result of parity non-conservation in β-decay, the emission of the first radiation leaves the 247 keV level polarized. Because only one nuclear matrix element contributes to a UFF transition, the degree and sign of the nuclear polarization is uniquely determined by the β decay direction. The quadrupole precession of the nucleus in a single crystal has the special property of converting the polarization to an alignment, the sign of which is determined by the sign of the QI. This alignment can then be observed as an anisotropy in the emission of the succeeding γ-ray relative to the β-particle direction and the crystalline symmetry (c-axis). The sign of the anisotropy leads directly the sign of the QI coupling constant, e^2q0.

When the β-active nuclei are embedded in only one face of the host crystal, the most convenient detection geometry is that shown in Fig. 1a. The β-particles are observed normal to the c-axis which is in the plane of the crystal and the γ-rays are detected alternately at the positions denoted by $φ = ±π/2$ in the figure. The theory shows that the asymmetry ratio

$$A(t) = \frac{2[W(-π/2)-W(π/2)]}{[W(-π/2)+W(π/2)]},$$

which can be computed from the observed β-γ delayed coincidence spectra, is given by

$$A(t) = a \sin 2φ + b \sin 3φ$$

for the cascade in question, where a and b are positive and

$$ω_φ = \frac{3π}{10} (e^2q0/ℏ).$$

Thus the sense of measured A(t) spectra directly reveals the signs of e^2q0.

111Ag sources were produced in the reaction 110Pd(n,γ)111Ag, 111Ag and 111Pd. In the first application of this technique, chemically separated 111Ag was diffused into a Cd single crystal and the A(t) spectrum is reproduced in Fig. 1b. For subsequent experiments, in order to insure that all of the activity was actually in the host lattice while still sufficiently close to the crystal surface to minimize β scattering and absorption, the 111Ag was implanted at 90 keV energy using an isotope separator. The resulting spectra for Sn, Ti and Be host crystals are shown in Figs. 2a, b, c. It can be seen from the sign of the initial excursion of A(t) near t = 0 that for the cases of Cd, Sn and Ti (Fig. 1b and Figs. 2a, b) that e^2q0 > 0 and that for Be (Fig. 2c) that e^2q0 < 0. From nuclear systematics on generically related 5+ levels in Cd isotopes it is believed that Q > 0; thus the sign of the field gradient, e^2q, is determined. Point ion lattice sum calculations for these metals indicated that e^2q (lattice only) is positive for Ti and Be and negative for Cd and Sn. The effect of the electronic contribution to e^2q, therefore, overwhelms and reverses the sign of the lattice field in Cd (as has been theoretically predicted) and in Sn, whereas not in Ti. Be must be omitted from this consideration since, unlike in the other three hosts, a very interesting anomaly between this measurement and previous γ-γ PAC measurements in Be has been observed which is the subject of another contribution to this Conference.

Resident Visitor from Rutgers University, New Brunswick, N. J.; supported in part by the N.S.F.

2. P. Raghavan, R. S. Raghavan and E. N. Kaufmann, to be published.
Fig. 1a Experimental geometry showing the emission directions of the $\beta$ and $\gamma$ radiations relative to the single crystal symmetry ($c$) axis.

Fig. 1b $A(t)$ spectrum from Ref. 2 for the case of $^{111}\text{Ag}$ diffused into a single crystal of hexagonal close packed Cd.

Fig. 2a $A(t)$ spectrum for $^{111}\text{Ag}$ implanted into a single crystal of tetragonal B (white) Sn.

Fig. 2b $A(t)$ spectrum for $^{111}\text{Ag}$ implanted into a single crystal of hexagonal close packed Ti.

Fig. 2c $A(t)$ spectrum for $^{111}\text{Ag}$ implanted into a single crystal of hexagonal close packed Be.
The detection and measurement of electric quadrupole splittings, as weak as \(10^{-6}\) smaller than a dominant magnetic dipolar splitting, by NMR/ON

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The technique of resonance detection through changes in the anisotropy of radioactive emissions from oriented nuclei (NMR/ON) has been most extensively applied to the study of impurities in ferromagnetic metal hosts. The dominant hyperfine interaction is magnetic dipole, but recently interest has developed in the weaker electric quadrupole interactions which can arise either from the proximity of vacancies or lattice defects, from magnetostrictive effects\(^{(1)}\), or from effects of spin-orbit coupling, which combines with spin polarisation to give a non-zero quadrupole interaction at the site of a magnetised impurity\(^{(2)}\). The purpose of this paper is to show that NMR/ON can be applied to study such quadrupole interactions, not only when they are large enough (> 0.1 % of the dipole term) to give a resolvable multiplet structure to the observed resonance, but also when they are far weaker than the typical linewidths, which are of order 1 MHz. Recent theoretical investigations of the phenomenon of adiabatic fast passage, as detected by NMR/ON, have shown that the nuclear magnetic sub-level population inversion which is associated with conventional adiabatic fast passage, is measurably perturbed in the presence of quadrupole interactions which are of the order of the interaction strength of the rf field at the nuclei thus reducing the minimum detectable quadrupole splitting to of order 0.1 kHz or \(10^{-6}\) of the magnetic dipole interaction\(^{(3)}\).

In this outline presentation of the theory we proceed from the pure Zeeman case as observed in adiabatic fast passage, through increasing magnitude of quadrupole interaction to the simple resolvable multiplet case which has already been reported in IrNi\(^{(4)}\).

1. Pure Zeeman splitting. The observation of adiabatic fast passage involves moving once through the resonance line in a time fast compared with the nuclear relaxation time \(T_1\) but slowly enough for the nuclei to follow the effective field \(H_{\text{eff}}\) acting upon them. \(H_{\text{eff}}\) is compounded of the static interaction \(H_0\) and the radio frequency field \(H_1\). True fast passage is most easily understood in terms of a transition to a frame of reference rotating with \(H_1\), when at resonance the levels have a minimum splitting \(\Delta H\). On passing through resonance the population distribution among the magnetic sub-levels is inverted. The Hamiltonian describing this situation is

\[
\hat{H}_{\text{eff}} = \gamma \mathbf{H}_0 \mathbf{I} - \frac{\gamma \mathbf{H}_1}{\omega_c} (\mathbf{I} - \mathbf{I}')
\]

where \(\mathbf{H}_{\text{eff}} = (H_0^2 - \frac{\gamma^2 H_1^2}{\omega_c^2}) \mathbf{I} + \mathbf{I}'\) and \(\mathbf{I}'\) are spin operators in the frame of \(\mathbf{H}_{\text{eff}}\). Full inversion (which requires no transitions between \(\mathbf{I}'\) states) is observed only when \(\frac{\gamma H_1}{\omega_c}\) is small enough to be a negligible perturbation.

2. \(H_{\text{eff}} < H_1 < \gamma H_0\). We define \(P = 3eQ V_{zz} (3 \cos^2 \alpha - 1)/4I(2I-1)\) where \(V_{zz}\) is an electric field gradient, taken for simplicity here to have cylindrical symmetry, along an axis \(z\) at angle \(\alpha\) to the axis of \(H_0\). This small electric quadrupole interaction gives rise to an additional non-oscillatory term in \(\hat{H}_{\text{eff}}\) frame

\[
I_{\text{quadrupole}} = \frac{P}{2} \left(3 \cos^2 \alpha - 1\right) \mathbf{I}'^2 \cos^2 \theta + \mathbf{I}'^2 \sin^2 \theta - \left(I_x' I_y' + I_y' I_x'\right) \cos \theta \sin \theta
\]

Using time dependent perturbation theory the effect of this term on the change immediately after single passage in the orientation parameters \(B(5)\) is shown in Fig. 1 for \(I = 5\). The horizontal axis is a parameter \(\left(\gamma H_1^2/4I_0\omega_c^2\right)\) which increases as the conditions for adiabatic inversion are fully satisfied. It is seen that for \(P/\gamma H_1 < 0.1\) the variation in the \(B_2\) term is three times that for \(P = 0\) for an adiabatic parameter of 5. This is readily observable, as is also the variation in the subsequent time evolution of the emission anisotropy as the partially inverted spin system regains equilibrium with the lattice. Given that \(y_{12}\) is typically of order 1 kHz (r.f. amplitude of 1 G, after enhancement, acting on 1 \(\mu_\text{N}\)), this gives the ability to detect and measure \(P/\gamma H_1\) of order 0.1 kHz.

3. \(y_{12} \ll P/\gamma H_1 < y_{12}\). We have also analysed the situation in which the quadrupole interaction is large enough to split the nuclear sublevels that they are no longer simultaneously on resonance. The situation then corresponds to a succession of resonances between pairs of sublevels with the result that, if the adiabatic parameter is large, the sublevel populations invert in sequence. The overall result is that the populations of the sublevels are cyclically moved by one place either upwards or downwards depending upon the sign of \(P\) relative to the sweep direction of the r.f. frequency.

The change in the orientation parameters is shown in Fig. 2 for \(I = 5\) and experimental data are given in the following paper. We note that in this regime \(P/\gamma H_1 \approx 100\text{ kHz}\) observable effects are sensitive to the sign of \(P\) only.
In this situation the quadrupole term is detectable without resort to fast passage studies as the resonance is split into \((2I + 1)\) resolvable components separated by \(\omega_{\text{Q}}\). The situation differs from conventional NMR only in that the nuclear polarization leads to unequal changes in the orientation parameters for different component resonances. In this regime \((\gamma_{\text{n}} > 100 \text{ kHz in metals})\) both the sign and magnitude of \(P\) may be measured.

This work was supported by grants from the Science Research Council.

References

Figure 1

Figure 2
IV. OBSERVATION OF LOCAL TEMPERATURE DEPENDENCE IN THE IMPURITY ELECTRIC QUADRUPOLE INTERACTION.

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Measurements of the nuclear electric quadrupole interaction (QI) at impurities in non-cubic metals have been reported by several groups recently. It is hoped that such information will shed light on the electronic structure of the metal as well as on the specific impurity-host interaction. To this end, the temperature dependence of the QI has been measured in several cases where it was found that for a given host the magnitudes of the QI varied from one impurity to another but the relative variation of the QI with temperature was independent of the specific impurity. Such observations are consistent with the notion that the temperature variation arises mainly from the thermal expansion of the host lattice and from thermal redistribution of host conduction electrons among states of different spatial symmetry and in therefore a property only of the host characteristics.

We report here the first observation of a markedly different temperature dependence at the impurities in the same host. Using the time-differential perturbed angular correlation technique on the 187-lm and 187 keV level of 187Ta, the QI was measured as a function of temperature for these impurities in a Ti metal host. The parent activities, 113In (produced by 104Ag n, 2n)114In and 184Hf (from 184Hf (n,γ) 184Hf), were implanted at 90 and 70 keV energy, respectively, into a freshly cleaved Ti single crystal using the Princeton and Bonn isotope separators. Unique QI frequencies were found in each case and were assigned to impurities at regular lattice sites. This assignment is based on a previous channelling measurement on 187Ta (for the Cd case) and on the agreement of the QI frequency observed with TiHf alloys3 (for the Ta case). The room temperature values for the QI frequency νQ = eQe2Q/h are 27 ± 0.3 MHz for 113Cd and 345 ± 5 MHz for 187Ta. The temperature variations normalized to 4.2°K are shown in Fig. 1 (where some data from Ref. 3 are included).

The substantial difference between vQ(T) for Cd and Ta impurities must be attributed to distinctly local differences in the way each impurity interacts with the Ti host. Local lattice distortions as well as impurity charge screening mechanisms will probably play an important role in the explanation of this effect.

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Fig. 1 Values of vQ normalized to their respective values at 4.2°K plotted against temperature for the Cd probe (○), the Ta probe (□), and the data from Ref. 3 (●). (The points at 77°K have been slightly displaced horizontally for the sake of clarity.) The solid lines have been drawn to guide the eye.
The electric field gradient at an impurity in a metal is usually decomposed into a contribution from the positive lattice ions and a part arising from the nonuniform distribution of the conduction electrons around the impurity. The temperature dependence of the EFG can yield information on the different contributions and the role played by the impurity itself.

We measured $V_{zz}^{(T)}$ between 77°K and 5/3°K for fluorine in zinc, using as a probe the 5/2, 128 ns state of $^{19}$F at 197 keV. Contrary to the other measured cases of metallic impurities in zinc fluorine has a very dissimilar electronic structure than the host lattice.

A 45 MeV $^{19}$F beam from the Stony Brook Fh Tandem was Coulomb excited to the 5/2$^+$ level and strongly polarized by scattering from a gold foil into a cone centered at 60° from the beam direction, and implanted through vacuum into a Zn single crystal. A thin film detector in the flight path generated the timing signals for the observation of the nuclear spin precession in the EFG with a four ka! setup. The delayed time distributions taken at different angles were fitted with the theoretical distribution function calculated using even and odd rank statistical tensors from the Windier-OeBoer Coulomb Excitation code.

The measured temperature dependence of $V_{zz}^{(T)}$, normalized to the room temperature value, is shown in Fig. 1 together with the $^{111}$Cd, $^{67}$Zn and $^{55}$Fe data and the theoretical lattice contribution calculated from ref. 6). The amplitude of about 50% of the theoretical value is consistent with one half of the implanted $^{19}$F ions experiencing a unique field gradient. The agreement between observed and calculated time distributions for different orientations of the $c$ axis supports the assumption that the fluorine ions come to rest on regular lattice sites or sites with the same symmetry of the EFG.

To parametrize the temperature dependence of the EFG the following approximate relation is used:

$$V_{zz}^{total} = V_{zz}^{latt} \left( 1 - \gamma_m \right) \left( 1 + F_{CE}^{ZZ} \right)$$

where $\gamma_m$ is the Sternheimer factor and $F_{CE}^{ZZ}$ is a (anti)shielding factor describing the contribution of the conduction electrons, the + and - signs applying to the case where the sign of the measured $V_{zz}^{total}$ is the same or opposite to the sign of $V_{zz}^{latt}$. Fig. 2 shows these experimental shielding factors obtained with the following quadrupole moments and Sternheimer factors: $^{19}$F(0.11b, -42.2), $^{111}$Cd(0.9b, -29.3), $^{67}$Zn(0.2b, -12.3). Whereas for $^{111}$Cd and $^{67}$Zn in Zn theoretical considerations favor $F_{CE} < 0$ and therefore only $F_{CE}^{ZZ}$ is shown in Fig. 2, the known positive sign of $QV_{ZZ}$ for $^{19}$F in Zn $^{11}$) together with the theoretically predicted $Q < 0$ leads to $V_{zz}^{total} < 0$ and results in a shielding factor $F_{CE}^{ZZ}$ which is inversely and much more strongly temperature dependent than the other cases which can be qualitatively explained by Fermi-surface contributions to $V_{zz}^{latt}$.

One is thus led to consider also a possible anomalous temperature dependence of the lattice part of the electric field gradient, a fact which has not found much attention so far. In the case of fluorine in Zn$^{2+}$ the difference in valence is shielded by the conduction electrons with the screened potential of the (presumably) F$^{1-}$ reaching out over a few lattice constants $^{7}$ and therefore changing the equilibrium positions of the nearest Zn$^{2+}$ neighbors. As $V_{zz}^{latt}$ depends strongly on their contributions the use of the bulk c/a ratio and of its temperature variation to calculate $V_{zz}^{total}(T)$ may not be sufficient. Clearly calculations of the lattice structure around impurities of different valence are needed.
Fig. 1: Temperature dependence of the quadrupole interaction frequency, normalized to room temperature.

Fig. 2: Temperature dependence of the electronic shielding factors for impurities in Zn.

References:

IV. 33 TEMPERATURE DEPENDENCE OF THE ELECTRIC QUADRUPOLE INTERACTION OF $^{111}$Cd IN Sn AND In

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The static electric quadrupole interaction in metals has been studied recently by several groups using the time differential perturbed angular correlation technique with radioactive sources (TDPAC) and as well as in-beam (TDPAD). A strong variation of the quadrupole interaction frequency with temperature was observed in all cases. We have investigated the temperature dependence of this interaction on excited $^{111}$Cd nuclei ($I^\pi = 5/2^-$, 247 keV) in an environment of metallic In and Sn, in order to get more systematic information on the variation of the electric field gradients (EFG).

In the present experiment the isomeric level of $^{111}$Cd ($T_{1/2} = 84$ ns) was populated in the 2.8 d decay of $^{111}$In. The radioactive $^{111}$In nuclei were electrolytically deposited on indium or tin foils; then the metals were melted to get an uniform distribution of the activity. No effects of different annealing procedures applied to the sources were observed. The 150-247 keV $\gamma-\gamma$ coincidences were detected by three NaI(Tl) detectors and conventional slow-fast electronics. High stability of the time scale was obtained by using a digital TAC. The intensity ratio

$$R(t) = \frac{N(180^\circ) - N(90^\circ)}{N(180^\circ) + N(90^\circ)} \sim \frac{A_2}{A_4}g_2(t)$$

is shown in fig. 1 for $^{111}$Cd in Sn for different source temperatures. The solid lines are least squares fits to the experimental data assuming a static quadrupole interaction with an axial symmetric field gradient. No significant relaxation of the perturbation amplitudes or distribution of the EFG was observed. All results are summarized in the following table.

<table>
<thead>
<tr>
<th>$^{111}$Cd in In</th>
<th>Quadrupole frequency $\frac{eQV_{zz}}{\hbar}$ (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature T (K)</td>
<td></td>
</tr>
<tr>
<td>1.2 (.1)</td>
<td>24.95 (.15)</td>
</tr>
<tr>
<td>1.5 (.1)</td>
<td>25.02 (.18)</td>
</tr>
<tr>
<td>1.8 (.1)</td>
<td>25.21 (.22)</td>
</tr>
<tr>
<td>2.7 (.1)</td>
<td>25.19 (.11)</td>
</tr>
<tr>
<td>4.2 (.1)</td>
<td>25.24 (.08)</td>
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<tr>
<td>77 (.2)</td>
<td>24.62 (.15)</td>
</tr>
<tr>
<td>194 (.2)</td>
<td>21.32 (.19)</td>
</tr>
<tr>
<td>273 (.2)</td>
<td>18.59 (.12)</td>
</tr>
<tr>
<td>384 (.3)</td>
<td>15.32 (.13)</td>
</tr>
<tr>
<td>413 (.4)</td>
<td>11.75 (.27)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$^{111}$Cd in Sn</th>
<th>Quadrupole frequency $\frac{eQV_{zz}}{\hbar}$ (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature T (K)</td>
<td></td>
</tr>
<tr>
<td>1.2 (.1)</td>
<td>43.56 (.25)</td>
</tr>
<tr>
<td>4.2 (.1)</td>
<td>43.40 (.29)</td>
</tr>
<tr>
<td>77 (.2)</td>
<td>42.69 (.26)</td>
</tr>
<tr>
<td>194 (.2)</td>
<td>39.91 (.21)</td>
</tr>
<tr>
<td>273 (.2)</td>
<td>37.44 (.20)</td>
</tr>
<tr>
<td>323 (.3)</td>
<td>35.45 (.20)</td>
</tr>
<tr>
<td>373 (.4)</td>
<td>33.81 (.22)</td>
</tr>
<tr>
<td>422 (.4)</td>
<td>32.01 (.20)</td>
</tr>
<tr>
<td>478 (.4)</td>
<td>30.02 (.26)</td>
</tr>
</tbody>
</table>
The mean quadrupole frequency $\Omega = eQ_{zz}/\hbar$ is plotted in fig. 2 over $T^{3/2}$ as a first empirical attempt for a better understanding of the temperature dependence of the EFG. The data of the present experiment for $^{111}\text{Cd}$ in In and Sn (filled circles) as well as the data given by Hewitt$^6$ for $^{115}\text{In}$ in In (open squares) are well reproduced by a simple $T^{3/2}$ relation. The results of previous measurements performed by Bodenstedt$^2$ for $^{111}\text{Cd}$ in In (filled squares), by Haas$^7$ for $^{111}\text{Cd}$ in Sn and In (open circles), by Raghavan$^1$ (open circles) and by Bleck$^3$ (filled triangles) for $^{111}\text{Cd}$ in Cd metal are also shown in fig. 2. The small deviations of the in-beam data (Bleck) at low temperatures are explained by an additional EFG caused by radiation damage effects$^3$. All data, however, indicate that the temperature variation of the EFG can be reproduced by a $T^{3/2}$ dependence.

At the present state no theoretical explanation of the temperature variation of the EFG has been reported. We hope, however, that this empirical relation may stimulate theoretical investigations of this problem.

References

5) R. M. Steffen and H. Frauenfelder, in Perturbed Angular Correlations (Amsterdam 1964)
7) H. Haas and D. A. Shirley, Journ. of Chem. Phys. 58 (1973) 3339
The measurement of the temperature dependence of the nuclear quadrupole interaction by the time differential perturbed angular correlation technique (TDPAC) proved to be a handy method to disclose the relative importance of the various contributions to the electric field gradient (EFG) in metals.

It was the aim of this work to extend the existing hyperfine studies on tantalum in transition metals [1,2,3,4] to group IIIb hosts. The samples were produced by melting Sc, Y, and Lu metal with neutron irradiated Hf metal (≈250 ppm) in an argon arc furnace. The experimental setup consisted of two 1/2'' x 2'' NaI(Tl) scintillation crystals mounted on XP2020 photomultipliers and a conventional fast-slow coincidence circuit.

The angle between the detectors was 180° in all experiments. The data were analysed by least squares fitting. A typical spectrum for 181TaSc is shown in fig.1. The system 181TaLu yielded similar spectra. Results on 181TaV have been reported earlier [5].

In all three host lattices we found uniform and axially symmetric field gradients. Using the known quadrupole moment of the 482 keV state of 181Ta, \( Q_{181}\) = 2.5(15) barns [1], the EFGs at room temperature have the values: \( Q_{181} \times 10^{-17} = 5.1(2), 5.9(3) \) and \( 5.0(3) \) V/cm² for Sc, Y, and Lu, respectively. The measured interaction frequencies \( \omega = \frac{e^2 q \gamma}{\hbar} \) at various temperatures between 4.2 K and 450 K are presented in fig.2, the data points being connected with dashed lines. The solid lines represent lattice EFG calculated in the point-charge-plus-uniform-background model [6] with the lattice parameters of refs. [7,8,9]. These EFG values were converted to frequencies and multiplied by constant factors to agree with the experimental values at 300 K. This normalization implies the conduction electron contribution to the EFG to be proportional to the lattice EFG [10]. From fig.2 it is evident that the temperature de-
dependence of the EFG in the system $^{181}\text{Ta}_{\text{Lu}}$ is fully accounted for by the lattice expansion in the temperature range between 4.2 K and 450 K. On the other hand, a thermal repositioning of conduction electron states at $^{181}\text{Ta}$ in Sc and particularly in Y is strongly indicated.

Unfortunately, Zeeman is not sensitive to the sign of the interaction. But, since $^{181}\text{Ta}$ is a high resolution Mössbauer effect isotope as well, both methods combined could yield detailed information about EFGs in a large number of host lattices. The observation of the Ta resonance, however, is easily prevented by crystal inhomogeneities. In this respect the absence of a damping worth mentioning in the spectra presented here should encourage to attempt Mössbauer experiments on $^{181}\text{Ta}$ in group IIIb metals in order to determine the sign of the EFGs.

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7. Schmitz-Fragh, H. and Dümmer, Ph. (1968) Z.Metallkde 59, 377
For investigations of the quadrupole interactions in metals Zn and particularly Cd are hosts of special interest for the following reasons: 1) Experimentally large effective field gradients are expected due to the large deviations of the ratios c/a of the lattice constants from the ideal value 1.632; 2) Theoretically the wavefunctions needed to calculate the electronic contributions to the field gradient can be tested, at least for Cd, since Knight shift and cyclotron resonance data are available.

Experiments have been performed with Cl in Zn, Zn in Zn, and Rh in Zn. In order to extend the systematics of the quadrupole interactions in these lattices to other impurities, the quadrupole rotations of the 165 keV e⁻ - 134 keV angular correlation of ¹⁹⁷Hg were observed. As an example the data for ¹⁹⁷Hg in Zn are plotted in Fig.1. The solid lines are results of least squares fits of the theoretical perturbation factor for polycrystalline lattices $G_2(t) = \sum_n s_n^2 \cos n\omega_0 t$ to the experimental data. The thus obtained values of $\omega_0 = \frac{6eQVz}{4I(2I-1)h}$ are given in the table.

<table>
<thead>
<tr>
<th>Temperature (°K)</th>
<th>¹⁹⁷Hg in Zn</th>
<th>¹⁹⁷Hg in Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>105</td>
<td>33.3(6)</td>
<td>24.8(6)</td>
</tr>
<tr>
<td>207</td>
<td>32.5(6)</td>
<td>24.0(6)</td>
</tr>
<tr>
<td>242</td>
<td>31.6(6)</td>
<td>24.0(9)</td>
</tr>
<tr>
<td>293</td>
<td>31.2(6)</td>
<td>21.7(6)</td>
</tr>
<tr>
<td>332</td>
<td>30.5(6)</td>
<td>---</td>
</tr>
</tbody>
</table>

Due to the short half life of the 134 keV state ($T_{1/2} = 7$ ns) and its small quadrupole moment, we observed only a fraction of the perturbation period. Therefore it is difficult to detect possibly partial alignments of the host foils or frequency distributions. We investigated these effects in further fits by treating $s_n^2$ as free parameters and by including frequency distributions of finite width. The absolute values of the quadrupole interaction frequencies scatter somewhat for these different fits (±20%). However, the relative changes of the frequencies with temperature seem not to be affected. In Fig.2 the ratios $\frac{\omega(T)}{\omega_{0}(293K)}$ for the Zn host are plotted as function of temperature. The normalization to 1 at room temperature eliminates the unknown quadrupole moments and allows to include in the same plot the data for other impurities in this lattice. These normalized frequencies which are equal to the normalized effective field gradients...
\( \psi_{zz}^{eff} (T) / \psi_{zz}^{eff} (293^\circ K) \) follow nicely a common curve. We assume the effective field gradient \( \psi_{zz}^{eff} \) to be proportional to the lattice part (\( \psi_{zz}^{eff} = \alpha \psi_{zz}^{lat} \)). The factor \( \alpha = (1 - \psi_{zz}^{lat} \beta) \) takes the electronic contributions to the field gradient into account, and arises from the Sternheimer antishielding effect \(^6\), the local moment contributions \( \psi_{\text{local}} \) according to the model of Watson \(^7\), and the enlargement of the effective charges of the lattice ions \( \beta \) as proposed by Das and Ray \(^8\). The lattice parts \( \psi_{zz}^{lat} \) for Cd and Zn hosts could be calculated by use of the formula given by Das and Pomerantz \(^9\) since the lattice constants are known as a function of temperature. These contributions normalized to 1 at 293\(^\circ\)K are also plotted in Fig. 2 for Zn. The ratio of both functions gives the temperature dependence of the electronic contribution

\[
\frac{\alpha (T)}{\alpha (293^\circ K)} = \frac{\left[1 - \psi_{zz}^{lat} (T) + \kappa_0 (T) \right]}{\left[1 - \psi_{zz}^{lat} (293^\circ K) + \kappa_0 (293^\circ K) \right]} \beta (T)
\]

The dependence seems to be linear and the variation is quite large. The same analysis of the data for Cd gives almost identical results. If local contributions by the different impurities were important, quite different temperature dependences of the quadrupole frequencies should be expected.

For Cd in Cd in a theoretical calculation \(^{10}\) it was derived that the p-electrons contribute the major part to the electronic field gradient. From a theoretical interpretation of the Knight shift anomaly \(^1\) in Cd it was shown that with rising temperature the p-electron density decreases whereas the s-electron density increases. From these two calculations it follows that a decrease of the electronic contribution to the field gradient with rising temperature should be expected at least for Cd metal in agreement with our experimental results. It would be very interesting if calculations of the electronic field gradient for different temperatures would reproduce the observed linear variation.

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*) On leave from the University of Lisbon with a fellowship from the Instituto de Alta Cultura of Portugal

**) Fellow of the Consejo Nacional de Investigaciones Científicas y Tecnica de la República Argentina

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3) P. Raghavan, R.S. Raghavan, E.N. Kaufmann, K.Krien, and R.A. Naumann; to be published in J. of Phys. F (Metals)
A combined study of the pressure and the temperature dependence of the quadrupole interaction (QI) allows to separate the explicit temperature dependence which helps to clarify the role of the conduction electron contribution to the QI in a metal. This has been first demonstrated by Raghavan et al.[1] for Cd metal. Here we report TDIAC measurements of the QI at $^{187}$Ta in m-Zr at 0 kbar and 25 kbar. The source was produced by arc melting Zr metal with neutron irradiated Hf (≈250 ppm). The quasi-hydrostatic pressure was produced with two Bridgman anvils with the source being encapsulated in boron epoxy. The phase transition of Zr at 25.4 kbar served for the pressure calibration. The experimental TDIAC setup has been described earlier[2]. Fig.1 shows TDIAC spectra measured at room temperature. In contrast to the results reported in ref.[3] no deviation of the QI from axial symmetry was found. Our measurements yield an increase of the QI from $e^2q_0/H = 314.3(4) \text{kHz}$ at 0 kbar to $345.0(1.7) \text{kHz}$ at 25 kbar.

In analyzing our data we follow the procedures of ref.[4], use the compressibility and thermal expansion coefficients of refs.[4,5] and take the temperature dependence of the QI at $^{187}$Ta in m-Zr of ref.[5]. This yields the explicit temperature coefficient of the QI at constant lattice parameters a and c as $(\Delta \ln(q_{0}/T))_{a,c} = -3.3(1.0)_{-0.6}^{+0.4} \text{kHz/K}$. This result shows a thermal redistribution of the orbital character of the conduction band.

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IV. 37 QUADRUPOLE INTERACTION FOLLOWING NUCLEAR REACTIONS STUDIED BY THE RECOIL IMPLANTATION TECHNIQUE

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One of the aims in the field of the investigation of hyperfine interactions is to get full variability in selecting the nuclear probe and its surrounding. Many off-line experiments done in this direction in the last years are performed by PAC-technique with the help of chemical preparation of radioactive samples or implantation via isotope separators.

Recoil implantation following nuclear reactions combined with the high accuracy of the time-differential perturbed angular distribution method (DPAD) is another favourable tool for these studies, if one takes advantage of the linear momentum transferred to the excited nuclear probes. Especially in the case of electric quadrupole interactions, systematic implantation of nuclear probes in different monocubic host metals should help to get a better understanding of the origin of the electric field gradients (EFGs). We have performed such experiments studying the EFGs acting on the probe $^{67}$Ge cycles in $^{70}$nsec, $I^+=9/2^-$, $E_V=734$ keV [1] in the polyvalent s-p-metals Cd, Zn, Sn and Sb and on the probe $^{67}$Zn cycles in $^{734}$keV $^{70}$nsec, $I^+=9/2^-$, $E_V=605$ keV [2] in Cd metal. In addition, measurements of the temperature dependence of the quadrupole coupling constants were started.

The nuclear probes are excited via the $^{64}$Zn($n$,n)$^{67}$Ge and $^{64}$Ni($n$,n)$^{67}$Zn reactions with 13-MeV n-particles from the 7-MV Van-de-Graf. The recoil energy of the final nuclei is rather low (< 1 MeV) compared with that available from heavy ion Coulomb excitation usually employed in recoil implantation experiments [2]. This requires a very thin target layer to allow the recoil nuclei to reach the implantation matrix. To compensate for this, we constructed sandwich-type targets with a total of 80 layers ($^{67}$Ge) and 160 layers ($^{67}$Zn). Layers of ZnS (=50 ug/cm$^2$ for $^{67}$Ge) and $^{64}$Ni-acetylacetonate (=30 ug/cm$^2$ for $^{67}$Zn) were evaporated by turns with the proper host metal (=120 ug/cm$^2$) onto a gold backing. As known from the literature [4], the natural polycrystalline structure should be obtained in the evaporation process. All experiments were performed with heated targets to reduce possible radiation damage effects.

Results of these experiments are shown in Figs. 1 and 2. The ordinates indicate the ratio $R(t) = (W(t,0°)-W(t,90°))/W(t,0°)+W(t,90°))$. The solid lines are $\chi^2$ fits assuming an axially symmetric field gradient and a polycrystalline probe. The fit of the modulation spectra for $^{67}$Ge in Cd and In could be improved when a second quadrupole interaction (the same for both hosts) was added. This frequency could be ascribed to Ge nuclei coming to rest in the ZnS layers. In all spectra no frequency distribution can be seen within the limits of the accuracy of our measurements. Table 1 summarizes the extracted quadrupole coupling constants at the respective temperatures.

The analysis of the modulation spectra suggests that the nuclei under study occupy substitutional lattice sites in agreement with other experiments [1,5]. The results for the quadrupole coupling constants of $^{67}$Ge (see Table 1) together with the previously obtained values of Ge in Zn and Ga [1] directly reflect the relative variation of the magnitude of the EFGs at the impurity site. A comparison of these values with the respective data measured in the pure host metals should give insight into the contribution of conduction electrons to the EFG [6].

**Fig. 1**: Quadrupole interaction of recoil-implanted $^{67}$Ge.
Table 1

<table>
<thead>
<tr>
<th>Probe</th>
<th>Matrix</th>
<th>T [K]</th>
<th>$\Delta^2Q_2/h$ [MHz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{67}$Ge</td>
<td>Cd</td>
<td>383</td>
<td>56.3(6)</td>
</tr>
<tr>
<td>In</td>
<td>328</td>
<td>22.6(6)</td>
<td></td>
</tr>
<tr>
<td>Sn</td>
<td>448</td>
<td>39.8(5)</td>
<td></td>
</tr>
<tr>
<td>Sb</td>
<td>469</td>
<td>20.3(3)</td>
<td></td>
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<tr>
<td>$^{67}$Zn</td>
<td>Cd</td>
<td>373</td>
<td>39.0(4)</td>
</tr>
<tr>
<td>Cd</td>
<td>310</td>
<td>39.5(4)</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2: Quadrupole interaction of recoil-implanted $^{67}$Zn in Cd at two different temperatures.

Literature

[3] e.g.: J. M. McDonald, P. H. S. Lesser, and D. B. Fossan, Phys. Rev. Lett. 28 (1972) 1057
IV. PERTURBED ANGULAR CORRELATIONS WITH $^{111}$Cd IN THE $\alpha$ AND $\delta$ PHASES OF INDIUM-TIN

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In the present contribution, we examine the distortion of the static quadrupole TDPAC pattern caused by valency impurities, In$^{0.9}$Sn in the $\alpha$ phase and Sn in the $\delta$ phase of the In-Sn system.

Data-reduction preliminaries. Under the circumstances, the perturbation factor is

$$G_k(\tau) = \sum_{n=0}^{N} a_n \exp(-\frac{1}{2} \tau^{2} \cdot n^2) \cos(n\gamma)$$

with the Laplace condition $\langle V^2 \rangle = 0$. In order to find the EFG components $V_x', V_y', V_z'$ and possibly also $\langle V^2 \rangle$, it is reasonable to choose a distribution which is symmetric in all of the three EFG components. Thus, for example,

$$P(V_x', V_y', V_z') = \exp\left(-\frac{\Delta V^2}{2\sigma^2}\right) \cdot \exp\left(-\frac{\Delta V^2}{2\sigma^2}\right) \cdot \exp\left(-\frac{\Delta V^2}{2c^2}\right).$$

The distribution for the individual components then becomes gaussian with $\langle V_x'^2 \rangle = \langle V_y'^2 \rangle = \langle V_z'^2 \rangle = \frac{\sigma^2}{2}$. As the actual experiments were performed with crystals of tetragonal structure, we restrict the discussion to the case $\tau = (V_x' - V_y') / V_z' = 0$. Defining $\delta = (\Delta V_x'^2) / V_z'^2$ and introducing the asymmetry parameter $\eta$, we find in the limit $\delta = 0$

$$P(\frac{\Delta V_x'^2}{V_z'^2}) = \exp\left(-\frac{\Delta V_x'^2}{V_z'^2}\right) \cdot \exp\left(-\frac{\Delta V_y'^2}{V_z'^2}\right).$$

so that $\langle \tau^2 \rangle = 3\sigma^2$. With Eq.(3), the integration of Eq.(1) yields

$$G_k(\tau) = \sum_{n=0}^{N} a_n \exp(-\frac{1}{2} \tau^{2} \cdot n^2) \cos(n\gamma).$$

The discussion above suggests the measured time spectra to be fitted to Eq.(4), yielding $V_z'^2$ and $\langle \tau^2 \rangle = \delta \cdot V_z'^2$. The procedure has been tested, simulating the case $\tau = 0.5/2$ by numerical integration of Eq.(2) and then performing least-squares fit to Eq.(4). For $\langle \tau^2 \rangle = 0.5V_z'^2$, excellent fits are always found with the starting values well reproduced (1.00±0.02) and reproducible (1.00±0.02) for $\delta > 1.00$. The ratio $\tau / \delta$ turns out to be sensitive from $2/3$ to 1.50 (cf. Fig. 1). With axial symmetry assumed in Eq.(4), an overestimate of $V_z'^2$ of more than 10% results.

Experimental Results and discussion. We have measured $G_k(\tau)$ of the 5/2$^+$ state of $^{111}$Cd at the In sites for $0-99$% Sn in In and $0-32$% In in Sn. The quantities $\tau$ and $\langle \tau^2 \rangle$ derived as discussed above are presented in Fig. 1. The ratio $\tau / \delta$ is nearly constant with a value of $\tau = 2.3$, somewhat higher than predicted from the distribution (Eq.(2)), and the values seem to saturate at high-impurity concentrations. It is not surprising that the EFG distribution (2) is too crude since the spread in the EFG arises from impurity atoms at discrete lattice positions. A more quantitative analysis may be performed following Kohn and Vosko [2]. The contribution to the EFG from an impurity ion is explained by the screened charge created around the solute atom. This charge distribution is shown to have a $1/r^3$ dependence, more specifically

$$V_z'^2 = e^2 \frac{8n^2}{3} \frac{\cos(2kF_{z'}\tau + t)}{r_{z'}^3},$$

where the $z'$ axis defines the direction from the probe to the solute ion and $r_{z'}$ their distance. The amplitude $A$ and the phase $\delta$ can be calculated either from "first principles" [3] or semiprimarily [4]. The two methods give nearly identical values for $A$, and since the EFG distribution is rather insensitive to $\tau$, we may use $\tau = \pi$ and $A = 2\pi e^{2}/(4\pi)\delta$, where $Z'$ is the difference between host and solute and $Z = 2kF_{z'}$. The enhancement factor,

$$\alpha = \frac{\int [\sin^2(2kF_{z'}\tau + t)]^{2} P(\cos\theta) \sin(\pi \tau) \sin^2 \theta \tan^2 \theta}{\int (\cos^2(2kF_{z'}\tau + t) P(\cos\theta)^2 \sin^2 \theta \tan^2 \theta) d\theta d\tau}$$

accounts for the Sternheimer antisheilding ($\delta_{\text{eff}}$) and for the fact that the conduction-electron wavefunction $\phi_k$ are Bloch functions rather than plane waves. We shall treat $\alpha$ as a parameter to be estimated from the experimental data. By computer simulation, impurity atoms are placed randomly in the crystal lattice, yielding the total EFG tensor which, in turn, is diagonalized. This procedure must be repeated until the two-dimensional distribution $P(V_x', V_y')$ can be established. Then, at last, the resulting $G_k(\tau)$ can be fitted to Eq.(4). Although the $V_z'^2$ distribution turns out to be not quite gaussian (cf. Fig. 1), the fits obtained are good. The results of the analysis are compared to the experimental data on the left-hand part of Fig. 2, which also shows the variation of $V_z'^2$ compared to the expectation from the change in lattice constants. The $\sigma$ values (see table) for Cd in In are consistent with $\alpha = 100(20)$ found for In in the $\alpha$ phase at 4.3 K [5] and the measured relation between $\tau / \delta$ and $\epsilon$ is also well reproduced in our model calculations as seen from Fig. 1. This is, of course, not a crucial test of Eq. (5), but merely a consequence of the fact that the spatial distribution of the impurity ions is properly taken into account. Both theory and experiment show that large $\eta$ values can be found when measurements are made in imperfect crystals despite their axial symmetry. Although the model (Eq. (5)) may be crude and the uncertainties are quite large, the comparison (cf. table) indicates a correlation between $\alpha$ and the conduction-electron contribution to the EFG, suggesting that the same physical effect causes the deviation of both these quantities from their plane-wave values.
In 295\(^\circ\)K  In 77\(^\circ\)K  Sn 295\(^\circ\)K

| \(|V_{zz}| \times 10^{16} \text{V/cm}^2\) | \(|V_{zz}\text{(latt)}| \times 10^{16} \text{cm}^2 \text{a)│} | \(F_{\text{cond}}\) |
|-----------------|-----------------|-----------------|
| 9.57            | 13.3            | 19.6            |
| 4.51            | 5.11            | 5.05            |
| 1.12            | 1.60            | 2.88            |

\(\alpha = \frac{9 \times 10^6}{10^6(60) \times 300(60)}\)

a) Using \(\gamma_{\text{m}}(\text{Cd}^{++}) = -29.3\) and \(Q_0/2^{111}\text{Cd}^+ = 0.77\) [6]

b) Defined by \(V_{zz} = V_{zz}\text{(latt)} + V_{zz}\text{(el)} \times (1 + F_{\text{cond}}) V_{zz}\text{(latt)}\)

References

[1] M. Forker, Nucl. Instr. Methods 106 (1973) 121 consider a somewhat different distribution, which also leads to Eq. (4), however with the relation \(\gamma_{\text{m}} = \delta^2\).


**Fig. 1**

Experimental and computer simulated relationship between \(\delta\) fit and \(\delta\) fit.

**Fig. 2**

Left-hand part: The variation of \(\delta\) with impurity concentration compared to simulated curves treating \(\alpha\) as a free parameter (see text).

Right-hand part: The relative variation of \(V_{zz}\) with impurity concentration. For comparison is given the variation of \(V_{zz}\text{(latt)}\) due to the change in lattice parameters [5] and the experimentally observed variation for \(111\text{Sn}\) in using NMR [4].
TDPAC Measurements of the Electric Fieldgradient at Impurity Sites in the Heavy Rare Earth Metals

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Institut für Strahlen- und Kernphysik der Universität Bonn

In this contribution we communicate the first results of a program for the investigation of the electric fieldgradient (EFG) in the heavy Rare Earth (R.E.) metals. The aim of this program is to gain further information on the conduction electron contribution to the EFG in noble cubic metals. With the exception of Yb all heavy R.E. metals from Gd to Tm crystallize in a hcp lattice with a c/a-ratio smaller than the ideal value for closest packing. Consequently, a finite EFG is to be expected at the lattice sites. In electric quadrupole interaction (EQI) studies aiming at information on the conduction electron contribution to the EFG it is unfavourable to measure the EQI with the R.E. nuclei themselves, as the EFG acting on the R.E. nuclei is dominated by the large ionic contribution from the unclosed 4f shell.

We introduced $^{111}$Cd and $^{181}$Ta impurities into the R.E. metals Gd, Tb, Dy, Ho and Er and determined the EQI at the impurity site by means of the TDPAC technique. An electron gun was used to alloy the R.E. with radioactive $^{111}$In and $^{181}$Hf, which decay to $^{111}$Cd and $^{181}$Ta, respectively. The radioactive impurity concentrations were $10^{-3}$ at. % $^{111}$In and 0.3 at. % $^{181}$Hf. The time differential behaviour of the angular correlations of the 173-247 keV cascade of $^{111}$Cd and the 133-482 keV cascade of $^{181}$Ta was measured at room temperature with standard TDPAC equipment. Fig.1 shows the time spectra of $\frac{A}{A_{22}(t)}$ for $^{181}$Ta in the different R.E. hosts. The spectra for $^{111}$Cd are given in ref. (1). The quadrupole frequency $\omega_Q$ and the asymmetry parameter $\eta$ were determined by fitting the theoretical perturbation factor for EQI in polycrystalline sources: $G_{kk}(t) = \omega_Q + \Sigma \sigma_{kn} \exp(-0.5 \omega_n^2 t^2) \cos(\omega_n t)$ to the measured spectra. The exponential factor describes the influence of lattice imperfections on the PAC pattern. Inspired by the hexagonal symmetry of the R.E. lattices the fits yield in all cases a small but finite asymmetry parameter. This is probably due to the fact, that not all the impurity atoms occupy substitutional lattice positions, which is evident from the damped oscillation amplitudes of the time spectra in fig.1. The values of $\omega_Q$, $\eta$ and $\delta$ obtained from the fits are listed in table 1. A calculation of the effective EFG $\nu_{eff}$ from the measured $\omega_Q$ was only possible for $^{181}$Ta, where the quadrupole moment of the 482 keV state is known (2). In the case of $^{111}$Cd the quadrupole moment is still uncertain.

<table>
<thead>
<tr>
<th>$^{181}$Ta</th>
<th>$^{111}$Cd</th>
<th>Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\omega_Q$ (MHz)</td>
<td>$\eta$</td>
<td>$\delta$</td>
</tr>
<tr>
<td>Gd</td>
<td>62.8(5.6)</td>
<td>0.30(5)</td>
</tr>
<tr>
<td>Tb</td>
<td>57.3(1.0)</td>
<td>0.17(3)</td>
</tr>
<tr>
<td>Dy</td>
<td>62.8(1.1)</td>
<td>0.23(3)</td>
</tr>
<tr>
<td>Ho</td>
<td>64.1(1.2)</td>
<td>0.25(2)</td>
</tr>
<tr>
<td>Er</td>
<td>72.4(1.4)</td>
<td>0.20(3)</td>
</tr>
</tbody>
</table>

Table 1: Experimental results for $^{181}$Ta and $^{111}$Cd in the different R.E. metals

Furthermore table 1 contains the theoretical values of the lattice EFG $V_{lat}^{zz}$, which is produced by the 3+ ions of the hexagonal R.E. matrix. The values have been determined by inserting the lattice parameters $c$ and $a$ into the formula for $V_{lat}^{zz}$, which Das et al. (3) derived by a lattice sum calculation.

In the upper part of fig.2 we have plotted the calculated values of $V_{lat}^{zz}$ and the experimental values of $\nu_{eff}^{zz}$ for $^{181}$Ta and $^{111}$Cd as a function of the R.E. atomic number. Because of the uncertainty of the quadrupole moment it is not possible in the case of $^{111}$Cd to obtain absolute values of $\nu_{eff}^{zz}$ from the measured $\omega_Q$. Therefore the values of $\nu_{eff}^{zz}$ ($^{111}$Cd R.E.) were normalized such that: $\nu_{eff}^{zz}$ ($^{111}$Cd Gd) = $\nu_{eff}^{zz}$ ($^{181}$Ta Gd). The plot shows that $V_{lat}^{zz}$, $\nu_{eff}^{zz}$ ($^{181}$Ta) and $\nu_{eff}^{zz}$ ($^{111}$Cd) depend in completely different ways on the R.E. atomic number. While $V_{lat}^{zz}$ increases continuously, $\nu_{eff}^{zz}$ ($^{111}$Cd) goes through a maximum at Dy and $\nu_{eff}^{zz}$ ($^{181}$Ta) through a minimum at Tb.

The lower part of fig.2 shows the ratio $\alpha \equiv |\nu_{eff}^{zz}/V_{lat}^{zz}|$ between the measured EFG and the
lattice EFG for both impurities as a function of the R.E. atomic number. Again the values of $\alpha(111\text{Cd R.E.})$ were normalized in such a way that: $\alpha(111\text{CdGd}) = \alpha(181\text{TaCd})$. It is a remarkable result, that the ratio $\alpha$ between the total EFG and the lattice EFG is not constant throughout the R.E. series. While $\alpha(111\text{Cd})$ decreases nearly linearly with increasing atomic number, $\alpha(181\text{Ta})$ shows a parabola-like behaviour. According to the current theoretical concepts on the EFG in non cubic metals, the enhancement factor $\alpha$ may be written as: $\alpha = ((1-\gamma_\text{se}) + \gamma_\text{ce})\beta$. $\gamma_\text{se}$ is the Sternheimer correction, $\gamma_\text{ce}$ has been introduced by Watson et al.\cite{4} to describe the enhancement of the lattice EFG by electrons localized at the nucleus under consideration and $\beta$ describes the renormalization of the ion charges of the lattice due to the presence of the conduction electrons, as discussed by Das et al.\cite{5}.

According to arguments discussed in detail in ref.\cite{1} it may be expected that both the Sternheimer factor and the renormalization factor $\beta$ decrease with increasing R.E. atomic number. This could explain - at least in a qualitative way - the observed decrease of $\alpha(111\text{Cd})$, but not the completely different behaviour of $\alpha(181\text{Ta})$. One possible reason for the difference between $\alpha(111\text{Cd})$ and $\alpha(181\text{Ta})$ is that the contribution to the total EFG from electrons localized at the impurity - described by $\gamma_\text{ce}$ - is probably larger for $181\text{Ta}$ than for $111\text{Cd}$. With respect to the $3^+$ ions of the R.E. lattice, $\text{Ta}^{5+}$ appears as a positive ($2^+$) impurity, while $\text{Cd}^{2+}$ is a negative ($1^-$) impurity. This should affect the charge distribution around the impurity and thus the local conduction electron contribution to the EFG, as the conduction electrons of the R.E. metals are to some extent of $5d$ character.

Fig.1: TDPAC spectra for $181\text{Ta}$ in the different R.E. metals

Fig.2: $\nu_{\text{lat}}, \nu_{\text{eff}}(181\text{Ta})$ and $\nu_{\text{eff}}(111\text{Cd})$ as a function of the R.E. atomic number

References:

(1) J.B.Fechner et al. Z.Physik 265, 197(1973)
(2) G.Netz et al. Nucl.Phys. A208, 505(1973)
IV. 40 QUADRUPOLE INTERACTION OF TUNGSTEN NUCLEI IN TELLURIUM AND ZINC SINGLE CRYSTALS AFTER RECOIL IMPLANTATION

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The quadrupole interaction of the first $2^+$ states in $^{182,184,186}\text{W}$ implanted into tellurium and zinc single crystals by 40 MeV $^{16}\text{O}$ ions from the Tandem Accelerator at the ENE in Zürich were studied by perturbed angular correlation after Coulomb excitation. Coincidence spectra between the backscattered particles and de-excitation gamma rays observed with a 30 ccm Ge/Li detector were measured. True and random coincidence spectra were recorded simultaneously. Furthermore, two particle gates were used in order to obtain coincidence spectra with particles implanted into different depths of the crystals. The accumulation and analysis of the data was carried out with a PDP-15 on-line system. Carefully prepared tellurium and zinc single crystal backings with different orientations of the $c$ axis were used. In order to determine the $A_{22}$ and $A_{44}$ angular correlation coefficients, measurements with the $c$ axis lying along the beam direction were made. In this case the correlation function should be theoretically unperturbed / see ref. 1 for geometrical considerations /.

Several series of crystal-axis dependence of the angular correlation were measured. In fig. 1 an example for $^{186}\text{W}$ in tellurium is presented and fig. 2 shows an example for $^{184}\text{W}$ in zinc single crystal. The interaction parameters determined from these experiments were $\omega_2^T = 0.45 \pm 0.08$, $0.39 \pm 0.05$, $0.36 \pm 0.04$ for $^{182,184,186}\text{W}$ in tellurium and $\omega_2^T = 0.51 \pm 0.08$, $0.42 \pm 0.04$, $0.33 \pm 0.04$ for $^{182,184,186}\text{W}$ in zinc respectively.

From these data the effective electric field gradients acting on W nuclei in Zn and Te were calculated, using $Q_{zz}$ values evaluated from experimental $B(E2)$ transition probabilities. The results are presented in Table I, together with other known values for W. The value for tellurium represents the $\frac{\text{eff}}{\text{lat}} + \frac{\text{velo}}{\text{local}}$ contributions.
### TABLE 1
Effective field gradients for W obtained in IMPAC experiments

<table>
<thead>
<tr>
<th>Host</th>
<th>$v^{exp}_{zz}$</th>
<th>$v^{latt}_{zz} (1 - j'_e)$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>/ in $10^{17} \text{ V}^2/\text{m}^2$/</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gd</td>
<td>$5.24 \pm 1.00$</td>
<td>$+0.65$</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>$6.35 \pm 0.60$</td>
<td>$+0.65$</td>
<td>6</td>
</tr>
<tr>
<td>Zn</td>
<td>$7.27 \pm 0.57$</td>
<td>$-8.18$</td>
<td>this work</td>
</tr>
<tr>
<td>Cd</td>
<td>$7.8 \pm 1.2$</td>
<td>$-6.7$</td>
<td>6</td>
</tr>
<tr>
<td>Te</td>
<td>$6.40 \pm 0.40$</td>
<td>$12.4 #$</td>
<td>this work</td>
</tr>
</tbody>
</table>

# References

6. L. Ph. Roesch at all. preliminary results and to be publ.
Perturbed angular correlation measurements for the first 2\(^+\) levels in \(^{152,154}\text{Sm}\) implanted into tellurium single crystals after Coulomb excitation were performed. Measurements for geometries in which the angular correlation should be unperturbed showed an attenuation, which was attributed to magnetic relaxation. The data were fitted with attenuation factors of the form presented in eq. 1, where \(\omega_m\) is the magnetic interaction frequency and \(\tau_c\) the correlation time. Different geometries were used in order to find out whether the time dependent magnetic interaction is isotropic according to the crystal orientation. From the fits the values \(0.052 \pm 0.015\) and \(0.070 \pm 0.015\) for \(^{152}\text{Sm}\) and \(^{154}\text{Sm}\) respectively were obtained in comparison with \(0.17 \pm 0.03\) and \(0.27 \pm 0.04\) obtained for \(^{152,154}\text{Sm}\) in copper from control measurements.

\[
G_k = \sum_n S_{kn} \frac{S_k}{1 + n\omega_m \tau_c}^{1/3}, \quad \text{where} \quad S_k = \frac{1}{1 + 1/3 + k (k+1) \omega_m \tau_c} \quad /1/
\]

In the limit of error the magnetic interaction was found to be isotropic. Fig. 1 shows an example for the crystal axis dependence of the angular correlation. The two curves were obtained simultaneously by two Ge/Li/ detectors. The data were fitted to a correlation function perturbed by a static quadrupole interaction in a single crystal and an isotropic time dependent magnetic interaction. The possible polycrystalline interaction was treated as a constant background. The quadrupole interaction parameters determined from these measurements are \(\omega_m \tau_c = 0.13 \pm 0.03\) and \(0.22 \pm 0.02\) for \(^{152}\text{Sm}\) and \(^{154}\text{Sm}\) respectively. Using the B(22) values for the quadrupole moment, this implies an electric field gradient \(V_{zz} = (1.75 \pm 0.40) \cdot 10^{17} \text{ V/cm}^2\), which is 25\% lower from the value reported by Grodzins at all. /1/.

REFERENCES

1. L. Grodzins in Nuclear Reactions Induced by Heavy Ions /North Holland Publ. Co., Amsterdam 1970/.
Electric Field Gradient in Disordered Binary Cubic Alloys.
E. Bozek, J. Golczewski, J. Kajfoss
Institute of Nuclear Physics, Cracow, Poland

The impurity atoms introduced into the alloy lattice perturb the charge symmetry of the original lattice, hence the electric field gradient can be produced even in alloys with cubic crystal structure. Assuming that the impurity atoms occupy randomly distributed substitutional sites in the host lattice, the EFG components can be calculated using simple lattice summation:

$$V_{ij} = \sum_{nm} q_i \left( \frac{3x_i x_j}{r_{nm}^3} - \delta_{ij} \right) / r_{nm}^3$$

where $q_i$ is an effective charge of impurity atom (point charge approximation). Using a similar lattice summation and replacing $q_i$ with $q_i = \sum_j |\psi(x_j)|^2$ defining the conduction electron density, the contribution to EFG due to the conduction electron screening of the impurity charge will be calculated.

In the present work both contributions to EFG have been calculated. The Monte Carlo method was used for simulation of random impurity distribution. The considered vicinity of a probe site depended on the impurity concentration and always contained at least 10 impurity atoms. The electron density distribution was described by Friedel's formula:

$$q_{ea} = \alpha \cos (2k_F r + \phi) / r^3$$

The results of the performed calculations are presented in fig.1.

Fig.1. The EFG distribution calculated for CP and CE contributions for different impurity concentrations. (in CE contribution $\phi = 170^\circ$)

Fig.2. The CE contribution of EFG calculated for constant impurity concentration $c = 3\%$ and different $\phi$ values.
Because of the random distribution of impurity atoms in the vicinity of the probe site the broad spectrum of $V_{zz}$ values was obtained. It is remarkable that these spectra are different for the charge point and conduction electron contributions and in both cases are strongly dependent on impurity concentration. The common feature is that for low impurity concentration $c < 10\%$ some separated peaks in the $V_{zz}$ spectrum are observed which could be connected with the distinct positions of the considered impurity atoms. The distribution of EFG for charge point approximation (CP) should be independent of the impurity type because of weak changes of lattice constant within the given range of concentration; only the strength of EFG can be different owing to different impurity charges. On the contrary, the distribution of the conduction electron part (CE) could be very different because of changes in $\varphi$ value connected with phase shifts describing the scattering of conduction electrons on impurity potential (see fig. 2).

The comparison of the measured distribution of EFG with the presented calculations could provide information about conduction electron density and its contribution to EFG in binary cubic alloys.

References:
IV. 43 Investigation of EFG Contributions in Disordered Cubic Alloys.
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The PAC method has been extensively applied in measurements of hyperfine interaction in solid states. These measurements could not be simply used for determination of the electron or crystal structure of the investigated samples without additional information about the localization of PAC probe nuclei. However, this information is not needed in the case of disordered binary alloys with cubic crystal structure, because the PAC effect averaged over random positions of the probe nuclei follows exactly the physical situation when the hf field originates from random distribution of impurity atoms in the host lattice. Recently the TD-PAC method was applied for investigation of hf interaction in Ag based binary alloys [1]. In the present work the same technique was used for measurements in Cu based alloys (Cu; Mn, Ni, Pd, In) and AgMn dilute alloy. The comparison of time dependent perturbation factors $C_2(t)$ measured in different alloys is presented in fig. 1. In all cases a static quadrupole interaction due to the EFG of the order $10^{17}$ V/cm$^2$ was determined. For interpretation of the experimental data the superposition of different interaction frequencies was assumed [1] which was a rough approximation only.

![Fig. 1. The time dependent perturbation measured in different alloys with constant impurity concentration $c = 1\%$.](image-url)
However, the obtained results were in qualitative agreement with the calculation of EFG [2] which means that the TDPAC method is essentially sensitive to EFG distribution. The correct data treatment would be the fitting of the experimental results with the theoretical $G_2(t)$ factor following calculated EFG distribution. The $G_2(t)$ patterns for the CE contribution to the EFG spectrum calculated for a constant impurity concentration $c = 1\%$ and different values of the $\varphi$ parameter describing the density distribution of conduction electrons $q_{el} = \frac{A}{r} \cos(2\pi r + \varphi)$ are presented in fig. 2. The calculated EFG is of the order $10^{15} \text{V/cm}^2$ similarly for both contributions (CP and CE) [2]. The agreement with experiments is possible only with the help of an enhancement factor of the order $10^2$. Taking into account the arguments given in the preceding paper [3], the changes observed in the shape of perturbation factors measured in alloys with the same impurity concentration may lead to the conclusion that the enhancement factor for the CE contribution is much larger than for the CP contribution and EFG in binary alloys is dominated by the effect of conduction electron distribution. In the present stage a decisive conclusion about the value is difficult, owing to the insufficient accuracy of the experimental data. However, assuming that the main contribution to EFG in cubic alloys is due to the CE contribution, the TDPAC method can be applied for experimental determination of parameters $\lambda$ and $\varphi$. In such measurements an impurity concentration in the range $3 - 6\%$ would be more favourable because then the EFG distribution as well as the $G_2(t)$ pattern are sensitive to changes of $\varphi$ value.

Fig. 2. The theoretical patterns of $G_2(t)$ calculated for CE part of EFG with different $\varphi$ values and constant impurity concentration $c = 1\%$

References:
1. E.Botek et al. IFJ Reports 865/PM and 866/PM (submitted to Phys. Lett. A)
2. J.Klijfoss, IFJ Report 856/PM
3. E.Botek et al. contribution to this Conference.
IV. 44 AN INVESTIGATION OF THE QUADRUPOLE INTERACTION OF IMPURITY IODINE NUCLEI IN CRYSTALS
OF TERNARY DIAMOND TYPE SEMICONDUCTORS BY PERTURBED ANGULAR CORRELATIONS

by K. V. Makarininas and B. K. Makarionien

Inst. of Phys. and Mathematics, Academy of Sciences of Lithuanian SSR, Vilnius, USSR

In this work perturbed angular correlations (PAC) were used to study a quadrupole interaction (QI) of $^{127}$I impurity nuclei in diamond type crystals of the compounds

$$A^V_{II}B_{III} ^{Te_2}$$ and $A^V_{II}B_{IV} ^{Te_3}$ (the ternary analogs of the binary compounds of the type of CdTe) and $A_{II} ^{Te_2}B_{IV} ^{Te_4}$ (the ternary analogs of the binary compounds with defective crystal lattice structure of In$_2$Te$_3$ type). The following question was raised: can a magnitude of the QI be used to characterize a crystallochemical structure and a type of the electrical activity of the impurity center in crystals of complex composition?

The experiments were performed using the radioactive isomer $^{127}$Te. The QI induced attenuations of the integral angular correlation of the 360-516 keV cascade gamma rays emitted by excited $^{127}$I nuclei after a $^{127}$Te beta decay were studied. The results are presented in the table. The calculations of the PAC attenuation coefficients $G_2$ and of the effective values of the electric field gradients (EFG) at the $^{127}$I nuclei were performed for the unperturbed correlation coefficient value $A_2 = 0.35$. The lifetime and the quadrupole moment of the $^{127}$I 57.6 keV level were taken equal to 2.79 ns and -0.1 barn respectively, and the asymmetry parameter of the EFG at $^{127}$I nuclei was assumed equal to 1 in $A^V_{II}B_{III} ^{Te_2}$ and 0 in $A^V_{II}B_{IV} ^{Te_3}$ and $A_{II} ^{Te_2}B_{IV} ^{Te_4}$ (ref. in /1/).

<table>
<thead>
<tr>
<th>Compound</th>
<th>$G_2$</th>
<th>$V_{zz1}$</th>
<th>$\Delta_{Te}/1-4/ 10^{18}$</th>
<th>$\Delta_{Te}/1-4/ \text{mm/s}$</th>
<th>Compound</th>
<th>$G_2$</th>
<th>$V_{zz1}$</th>
<th>$\Delta_{Te}/1-4/ 10^{18}$</th>
<th>$\Delta_{Te}/1-4/ \text{mm/s}$</th>
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</thead>
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<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>CdTe</td>
<td>$1.11 \times 10^{-3}$</td>
<td>$0.00$</td>
<td>$0.00$</td>
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<td>In$_2$Te$_3$</td>
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<td>$2.1 \times 10^{-3}$</td>
<td>$3.5 \times 10^{-3}$</td>
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<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$0.67 \times 10^{-3}$</td>
<td>$1.6 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>ZnAl$_2$Te$_4$</td>
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<td>$3.5 \times 10^{-3}$</td>
</tr>
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<td>$1.7 \times 10^{-3}$</td>
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<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>ZnGa$_2$Te$_4$</td>
<td>$0.61 \times 10^{-3}$</td>
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</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$1.0 \times 10^{-3}$</td>
<td>$2.1 \times 10^{-3}$</td>
<td>$3.1 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>ZnIn$_2$Te$_4$</td>
<td>$0.60 \times 10^{-3}$</td>
<td>$2.5 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$1.2 \times 10^{-3}$</td>
<td>$2.4 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>CdAl$_2$Te$_4$</td>
<td>$0.62 \times 10^{-3}$</td>
<td>$2.6 \times 10^{-3}$</td>
<td>$3.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$1.4 \times 10^{-3}$</td>
<td>$3.1 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>CdGa$_2$Te$_4$</td>
<td>$0.64 \times 10^{-3}$</td>
<td>$3.6 \times 10^{-3}$</td>
<td>$4.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$1.6 \times 10^{-3}$</td>
<td>$3.4 \times 10^{-3}$</td>
<td>$4.1 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>CdIn$_2$Te$_4$</td>
<td>$0.6 \times 10^{-3}$</td>
<td>$3.8 \times 10^{-3}$</td>
<td>$4.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$1.8 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
<td>$4.4 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>ZnGa$_2$Te$_4$</td>
<td>$0.64 \times 10^{-3}$</td>
<td>$3.7 \times 10^{-3}$</td>
<td>$4.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>$A^V_{II}B_{III} ^{Te_2}$</td>
<td>Cu$_2$Te$_2$</td>
<td>$2.0 \times 10^{-3}$</td>
<td>$4.1 \times 10^{-3}$</td>
<td>$4.7 \times 10^{-3}$</td>
<td>$A^V_{II}B_{IV} ^{Te_3}$</td>
<td>ZnIn$_2$Te$_4$</td>
<td>$0.66 \times 10^{-3}$</td>
<td>$4.4 \times 10^{-3}$</td>
<td>$5.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

A comparison of these results with the available data of the quadrupole splitting of the $^{125}$Te Mössbauer spectra /1-4/, which are presented in the last column of the table, shows the following regularities:
1) the QI of the $^{127}$I nuclei in the isomorphous rows of the compounds $AIB_{III}Te_2$ and $A_{IV}^{127}$Te changes in the same manner as the QI of the $^{125}$Te nuclei;

2) the QI of the $^{127}$I impurity nuclei is almost constant in the compounds $A_{II}^{127}$Te, the slight change of the QI of the $^{127}$I nuclei, if it exists, is of the opposite direction than that of the $^{125}$Te nuclei; the QI of the $^{127}$I nuclei in these compounds is approximately equal to that in the analogic binary compound In$_2$Te$_3$.

The first one of these experimental facts becomes clear if the assumption is made that the $^{127}$I atom in the $AIB_{III}Te_2$ and $A_{IV}^{127}$Te crystals forms four tetrahedral bonds, which are similar to those of the substituted atom of $^{125}$Te, and an additional electron is given to the conduction band. Such conclusion for analogic binary compounds ZnTe and CdTe, is confirmed by electrical measurements, which show that the I is the donor impurity /5/.

The results obtained for the compounds $A^{127}IB_{III}Te_2$ show that chemical bonds of the impurity I atoms are not identical to those of the Te atoms. The QI results for binary analogs of these compounds, Ga$_2$Te$_3$ and In$_2$Te$_3$, can be explained, if the assumption is made that some of the bonds disrupt when the impurity I atom replaces the Te atom /6/. The I atom instead of the disrupted bonds obtains the lone pairs of valence electrons. The appearance of the free bonds in the crystal can result the hole type impurity electrical conductivity. Such conductivity in the In$_2$Te$_3$ has been observed by electrical measurements (ref. in /6/). A similar situation, apparently, exists also in the crystals of the ternary compounds with defective structure.

The results of this investigation thus show that PAC data can be useful for the prediction of the electrical activity type of the impurity atoms in the compounds of complex constitution.

REFERENCES

IV. 45 Behaviour of Electric Field Gradients in PbZrO$_3$ and PbHfO$_3$ near the Curie Point

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University of California, Los Alamos Scientific Laboratory, New Mexico, U.S.A.

We have investigated the variation of the electric field gradient (EFG) at phase transitions of the perovskites PbHfO$_3$ and PbZrO$_3$. In these compounds Hf, Zr were replaced by radioactive Hf$^{181}$ and the 480, 133 keV cascade of Ta$^{181}$ was used for differential perturbed angular correlation (DPAC). The samples were prepared by pressing PbO, HfO$_2$ or ZrO$_2$ with 1% neutron irradiated HfO$_2$ and sintering at 800°C. With a specially constructed furnace whose stability was better than ±0.1K for periods of a week we made DPAC measurements between 450K and 550K.

For this case (polycrystalline sample, nuclear spin I = 5/2 and one EFG) general PAC theory simplifies to

$$N(\theta, t) = 1 + A_2 G_{22}(t) P_2(\cos \theta)$$

where

$$G_{22}(t) = \sum_{n=1}^{\infty} a_{2n} \cos \omega_n t \exp(-\frac{\nu^2}{2} \omega_n^2 t^2).$$

The quadrupole interaction frequency $\nu = eQV/h$ and the asymmetry parameter $\eta$ derived from the perturbation function $G_{22}(t)$ determine the electric field gradient at the Ta$^{181}$ site in PbHfO$_3$ or PbZrO$_3$. With more than one EFG the weighted sum of the corresponding $G_{22}(t)$ functions must be used. In Fig. 1 typical DPAC measurements in PbZrO$_3$ at different temperatures are shown.

From macroscopic measurements PbHfO$_3$ is known to undergo transitions from one antiferroelectric phase to another (AFE I - AFE II) at 435K and to become paraelectric (AFE II - PE) at 488K. We reported previously the existence of one EFG changing continuously with temperature in the AFE I phase. At 430K, the transition point, a second EFG occurs with $\nu = 75$ % and $\eta = 50$ % of their values for the first EFG. The relative fraction of the second EFG increases with temperature and within Hf both fractions are equal. New measurements confirm that this 50:50 ratio remains unchanged in the whole AFE II phase. The occurrence of this second EFG corresponds to a change of the lattice structure at the transition. Half of the Hf ions occupy a new site while the rest remain unchanged. At the AFE-PE transition (see Fig. 2) a small temperature hysteresis of one degree is observed (transition temperature 467K). Since a single measurement at one temperature requires typically 3 days, this hysteresis is stable at least for two weeks.

First γ-γ DPAC measurements in polycrystalline PbZrO$_3$ indicated the existence of one EFG in the low temperature AFE phase. This EFG changes continuously with temperature and decreases abruptly at the Curie point at 506K. Recently an intermediate ferroelectric (FE) phase just below the Curie point was detected, which should manifest itself in structure changes.

In searching for a temperature hysteresis in PbZrO$_3$ we heated the sample monotonically with a tolerance of 0.1K. Results are shown in Fig. 2. Up to 480K the Ta$^{181}$ is acted on by one EFG, which is in good agreement with other measurements. On further heating, a second EFG occurs with $\nu = 25$ % and $\eta = 50$ % of the values for the first EFG. This indicates a transition to the FE phase. With increasing fraction of the FE-gradient the AFE gradient disappears. Just below the Curie point only the EFG of the FE phase is present. On cooling the FE-AFE transition region lasts from 495K to 450K. This means a distinct AFE-FE temperature hysteresis is observed which is stable on the order of weeks. The FE-PE transition in PbZrO$_3$ shows a small temperature hysteresis indicated by the arrows in Fig. 2 similar to that in PbHfO$_3$.

This work has been supported by Bundesministerium für Forschung und Technologie.

3) M. Forker and A. Hammersfahr, Z. Physik, 255, 196 (1972)
Fig. 1 γ-γ angular correlation measurements in PbZrO$_3$ at temperatures in AFE-FE transition region.

Fig. 2 Phase transitions in PbHfO$_3$ and PbZrO$_3$:
- PbHfO$_3$ antiferroelectric-paraelectric (AFE-PE), PbZrO$_3$ antiferroelectric-ferroelectric (AFE-FE), the FE-PE transitions (508K, 506K) are indicated by vertical arrows.
Failure of the point-charge model for quadrupole hyperfine interactions in organo-tin compounds

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Due to the wide variety of coordination numbers and the accessibility of compounds in which the metal atom occupies a site of varying symmetry, the electric quadrupole split hyperfine spectra of the $^{119}$Sn resonance in organo-tin compounds have been interpreted by the application of a point-charge model in an attempt to elucidate the structure and bonding in such compounds. Among the fundamental assumptions of this model are: (a) a knowledge of the partial electronic charge residing on the metal and the ligand; (b) a knowledge of the internuclear distance between metal and ligand; and (c) a knowledge of the bond angles between the various ligands bonded to the central metal atom. Since usually, the absolute values of these parameters (in the absence of detailed single crystal X-ray diffraction data) are not known, the point-charge model has been used to account for the relative magnitude of the Q.S. interactions in $^{119}$Sn spectra of two structurally related compounds, and to use such data to derive "partial quadrupole splitting" parameters for a given group of ligands bonded to tin in a wide variety of organo-tin compounds.

One of the predictions of the point charge model is that for quasi-tetrahedral compounds of the type $R_jSnX$ and $HSnX^2$ (where $R$ is an organic moiety such as CH$_3$, C$_2$H$_5$, C$_3$H$_7$, etc.; and $X$ is an electron pair donor moiety such as a halogen, pseudohalide or chalcogen) the absolute value of Q.S. will be identical, while the sign of the e.f.g. tensor will be opposite for the two compounds. This prediction has been examined in detail for the compounds summarized in Table 1, in which the data refer to samples at $< 82 \pm 2^\circ$K. In order to eliminate the possible contribution of crystal stacking forces and preferential sample orientation effects, these compounds have also been studied in frozen glassy matrices, and these data are included in the summary, Table 1.

It is clear from the $^{119}$Sn and $^{129}$I data for $(CH_3)_3SnI$ and $CH_3SnI_2$, as well as the related data for the $^{119}$Sn resonance in similar organo-tin chlorides and bromides, that the point-charge model cannot be applied to the detailed quantitative interpretation of the Q.S. interaction in these materials. The reason for the breakdown of this model and the fundamental assumptions involved in the formulation of "partial quadrupole splitting parameters" will be discussed in detail.

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Références

3. I am indebted to Dr. A. Marks and Prof. R.S. Drago not only for the compounds in question, but also for fruitful discussion of some of the ideas presented in this paper.
### TABLE 1a - $^{119}$Sn Mössbauer Results at Liquid Nitrogen Temperature

<table>
<thead>
<tr>
<th>Absorber</th>
<th>Form</th>
<th>$T_\text{a.} , ^\circ\text{K}$</th>
<th>I.S. (a) mm/sec.</th>
<th>Q.S. (b) mm/sec.</th>
<th>$\Gamma_{\text{ave}}$ mm/sec.</th>
<th>$R$ (d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(CH$_3$)$_3$SnI</td>
<td>neat liquid</td>
<td>81.5</td>
<td>1.434</td>
<td>2.908</td>
<td>0.218</td>
<td>1.06</td>
</tr>
<tr>
<td></td>
<td>0.86 M BuBz(e)</td>
<td>82.5</td>
<td>1.418</td>
<td>2.690</td>
<td>0.883</td>
<td>0.97</td>
</tr>
<tr>
<td>CH$_3$SnI$_3$</td>
<td>neat solid</td>
<td>86</td>
<td>1.582</td>
<td>1.683</td>
<td>0.069</td>
<td>0.936</td>
</tr>
<tr>
<td></td>
<td>0.46 M BuBz(e)</td>
<td>83</td>
<td>1.564</td>
<td>1.614</td>
<td>0.943</td>
<td>0.56</td>
</tr>
<tr>
<td>(C$_6$H$_5$)$_3$SnCl</td>
<td>neat solid</td>
<td>83</td>
<td>1.394</td>
<td>2.479</td>
<td>0.080</td>
<td>0.932</td>
</tr>
<tr>
<td></td>
<td>0.82 M BuBz(e)</td>
<td>83</td>
<td>1.301</td>
<td>2.399</td>
<td>0.873</td>
<td>0.94</td>
</tr>
<tr>
<td>(C$_6$H$_5$)SnCl$_3$</td>
<td>neat liquid</td>
<td>83.5</td>
<td>1.187</td>
<td>1.726</td>
<td>0.034</td>
<td>0.972</td>
</tr>
<tr>
<td></td>
<td>1.1 V/V BuBz(e)</td>
<td>82</td>
<td>1.205</td>
<td>1.692</td>
<td>1.032</td>
<td>0.97</td>
</tr>
</tbody>
</table>

(a) With respect to the center of the room temperature BaSnO$_3$ absorption spectrum ; ± 0.015 mm/sec.
(b) ± 0.020 mm/sec.
(c) Uncorrected for thickness broadening.
(d) Intensity ratio ; $R = I_+ / I_-$ , where $I$ is the total area under each component of the resonance doublet ; ± 0.03.
(e) BuBz = n-buty1benzene

### TABLE 1b - $^{129}$I Mössbauer Results at 4.2$^\circ\text{K}$

<table>
<thead>
<tr>
<th>Absorber</th>
<th>Form</th>
<th>I.S. mm/sec.(a)</th>
<th>$e^2Q(127\text{I})$</th>
<th>$\Gamma$ mm/sec.</th>
<th>$\eta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(CH$_3$)$_3$SnI</td>
<td>0.03 M CH$_3$C$<em>6$H$</em>{11}$</td>
<td>- 0.15 ± 0.05</td>
<td>- 740 ± 20</td>
<td>0.80 ± 0.04</td>
<td>0.02 ± 0.02</td>
</tr>
<tr>
<td>CH$_3$SnI$_3$</td>
<td>0.037M CH$_3$C$<em>6$H$</em>{11}$</td>
<td>- 0.26 ± 0.04</td>
<td>1270 ± 20</td>
<td>0.84 ± 0.04</td>
<td>0.02 ± 0.02</td>
</tr>
</tbody>
</table>

(a) with respect to the Zn$^{129}$Te source
Numerous hexammine salts of the type $\text{M(NH}_3\text{)}_6\text{X}_2$ undergo a phase transition accompanying the hindrance of rotational motion of $\text{NH}_3$ molecules at low temperature. The investigation of the hyperfine interactions at the $\text{M}(=\text{Co, Fe, Ni, Cd, Zn})$ ions as well as at the $\text{X}(=\text{I})$ sites using Mössbauer effect or PAC would provide considerable microscopic information on such phase transitions. In this contribution we report the Mössbauer study in a temperature range from 1.6 to 300°K of some of these hexammines using $^{57}\text{Fe}$, $^{127}\text{I}$ and $^{129}\text{I}$ probes.

These compounds belong to the space group $\text{O}_h$ and this implies disorder within the ammonia groups. The absence of any quadrupole interaction at the M site above the phase transition temperature ascertains this disorder to be a dynamical one. This has been confirmed from the resonance spectra of $^{57}\text{Fe}$ occupying M sites in above hexammines. Below a critical temperature, however, the ammonia molecules tend to become static through a cooperative electrostatic interaction between $\text{NH}_3$ rotors in the crystal. This lowers the site symmetry of both the M and X sites from a cubic point group, and should manifest in a quadrupole interaction at both these sites. In Fig.1 we show this phenomenon observed from the Mössbauer spectra of $^{57}\text{Fe}$ atom in $\text{Fe(NH}_3\text{)}_6(\text{BF}_4)_2$. The value of $T_c$ has been found to be 108, 45, 47, 156, 140, 96, 20, 20, 25°K for (M, X) = (Fe, Cl), (Fe, Br), (Fe, I), (Fe, ClO$_4$), (Fe, BF$_4$), (Co, Cl), (Co, I), (Ni, I), (Mn, I) respectively. These values agree with the ones obtained from macroscopic investigations, like the specific heat, thermal conductivity, etc... on some of these compounds.

The hyperfine interaction at the X site was investigated from the Mössbauer resonance of $^{129,127}\text{I}$ in some of the salts. No measurable quadrupole interaction is observed even below $T_c$ established from M site resonance experiments. The rather small distortion and the highly ionic character of iodine as deduced from the isomer shift accounts for this observation.

The sudden decrease in the Mössbauer resonance fraction above $T_c$ is tentatively understood in terms of presence of liberons (quantum of tortional oscillations) in the high temperature phase, which scatter the acoustic phonons. The cooperative nature of the transition is corroborated by the hysteresis in the quadrupole interaction (Fig.1). The details of this investigation leads one to conclude that the transition is of second order but turns to the first order near $T_c$.
HIGH-RESOLUTION MÖSSBAUER STUDY OF HYDRIDED TANTALUM.

by A. Heidemann, G. Kaindl, D. Salomon, and O. Wortmann

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Even though considerable emphasis has been devoted in recent years to the study of hydrogen in metals /1,2/, there is still a serious controversy on the electronic structure, the different models ranging from a pure protonic as far as to a pure anionic picture for the interstitially dissolved hydrogen atoms /2/.

The present paper reports on a room-temperature study of the α-phase of the tantalum-hydrogen system, using the 6.2-keV gamma resonance of $^{181}$Ta /3/. As a function of hydrogen concentration c, isomer shift S and linewidth W of the gamma resonance line were studied. With increasing c up to about 8 at.%, a large and approximately linear increase of S over a range of about 5 mm/s, corresponding to about 40 experimental linewidths, was observed.

Previous Mössbauer investigations of metal-hydrogen systems have suffered from very limited resolution as well as from the fact that the Mössbauer atoms ($^{57}$Fe and $^{119}$Sn) could only be introduced as impurities into the metal-hydrogen host lattice /4/. These drawbacks are completely eliminated in the present work.

From the phase diagram of the Ta-H system, a room-temperature solubility limit of about 25 at.% hydrogen in the α-phase can be expected /5/. In the present work, hydrogen concentrations up to 8 at.% were achieved by an electrolytic method, using a 0.01 normal $\text{H}_2\text{SO}_4$ solution and typical current densities of 100 mA/cm$^2$. Before hydriding, the Ta foils (about 4 mg/cm$^2$ thick; 99.995 % nominal purity) were degassed in ultra-high vacuum by a method described previously /3/. The hydrogen concentration was determined from the relative change of the lattice constant $\Delta a/a$, measured by X-ray diffraction technique; in the studied concentration range a linear relationship between $\Delta a/a$ and c can be assumed, $d\Delta a/dc = 5.3 \times 10^{-2}$ /6/, where c is defined here and throughout the paper as the H/Ta atom ratio.

Three typical absorption spectra are shown in Fig. 1. At a hydrogen concentration of 8.2 at.%, the resonance line is shifted by about 5 mm/s to higher transition energies. This is a very strong effect, which can be measured with great precision. In addition, an increase of the experimental linewidth with c is observed. It should be noted, that the Mössbauer spectra as well as the respective $\Delta a/a$ measurements were taken several weeks after hydriding, when stationary conditions were reached.

The experimental results for S and W are plotted in Fig. 2. Within the limits of error, S increases linearly with $\Delta a/a$ (and with c), while W tends to saturate at higher concentrations. From a least-squares fit of a straight line to the isomer shift data a value of $dS/dc = 62$ mm/s is obtained.

The observed increase in transition energy with c reflects a decrease of the total electron density $\rho(0)$ at the nucleus, since the change $\Delta \langle r^2 \rangle$ of the mean-squared nuclear charge radius has negative sign for the 6.2-keV

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\gamma \text{ transition, } \Delta \langle \rho^2 \rangle = -(5 \pm 2.5) \times 10^{-2} \text{ m}^2 \left/ 3^1 \right.

For the following discussion the total isomer shift change will be separated into two contributions

\[ \frac{dS}{dc} = \left( \frac{dS}{d\rho} \right) \left( \frac{d\rho}{dc} \right) + \left( \frac{dS}{d\rho} \right) \]

The first term accounts for the volume change, while the second term describes the explicitly concentration dependent part caused by changes in the electronic structure of tantalum due to interstitially dissolved hydrogen atoms. In principle, the influence of lattice expansion on \( \rho(0) \) (first term in eq. 1) is difficult to estimate in the present case. To get at least an upper limit for the volume effect, we assume that the effect of volume expansion on \( \rho(0) \) due to small amounts of dissolved hydrogen atoms is identical (but of opposite sign) to the one caused by the application of hydrostatic pressure on pure tantalum metal, \( \left( \frac{dS}{d\rho} \right) \left( \frac{d\rho}{dc} \right) = 140 \pm 15 \text{ mm/s} \left/ 3^2 \right. \). Then a value of 22 mm/s is obtained for the first term in eq. 1 (where we have used \( \frac{d\rho}{dc} = 3 \frac{d\rho}{dc} / 0.159 \left/ 3^3 \right. \)). This means that the lattice expansion alone can account for at most about one third of the observed value for \( dS/dc \) (see Fig. 2a). For the second term a lower limit of \( \left( \frac{d\rho}{dc} \right) \geq 40 \pm 5 \text{ cm/s} \) is then obtained, corresponding to an explicit decrease of \( \rho(0) \) with \( c \).

This can be accounted for if one assumes that the number of d-band conduction electrons increases with increasing \( c \), assuming an electron configuration of 5d\( ^{6} \)6s\( ^{5} \)5p\( ^{0} \) for tantalum metal and using electron densities extrapolated from Dirac-Fock calculations for a few free-ion configurations of tantalum \( /7^1 \), we may conclude that not more than about 5 % of the hydrogen electrons can be donated to 6s states of the conduction band, while at least 50 % up to 100 % have to fill up the empty 5d states of tantalum, where the exact number depends mainly on the value taken for \( \Delta \langle \rho^2 \rangle \). This result should, however, not be taken in support of the simple "proton model" for transition metal-hydrogen systems, since it does also not contradict a more realistic picture as developed for example in Ref. \( /8^2 \).

The authors would like to thank H. Pfeiffer and H. Letzger for valuable experimental assistance and advice.

1. Proc. Int. Conf. on Hydrogen in Metals, Jülich, Germany (1972); JIL-CONF-6, Vol. 1 and 2.
7. J. B. Mann, Los Alamos Scientific Laboratory, private communication (1972).
The isomer shift (IS) as measured by the Mössbauer effect is

\[ IS = \frac{\hbar}{2} \sum c^2 R_{\Delta R} |\Psi(0)|^2 = C |\Psi(0)|^2 , \]  

(1)

where \( |\Psi(0)|^2 \) is the difference of the electron density of the nucleus in different chemical environments. \( \Delta R \) is the change of the nuclear radius during the \( \gamma \)-transition. \( R \) is related to the mean square charge radius by \( R^2 = \frac{5}{3} \langle r^2 \rangle \). In order to evaluate \( |\Psi(0)|^2 \) from IS-data one has to know the calibration constant \( C \) for the corresponding Mössbauer isotope.

The present work describes an experimental determination of \( C \) for \( ^{119}\text{Sn} \). For \( \chi \)-transitions Pauli \(^1\) has shown that the \( s \)-electron densities of the various shells are proportional to the corresponding internal conversion coefficients. Detailed relativistic HFS-calculations for \( ^{119}\text{Sn} \) \(^2\) show that the influence of the chemical environment on the radioactivity decay constant may be written as

\[ \Delta \lambda{\lambda} = \frac{\alpha}{\alpha + 1} \frac{|\Psi(0)|^2}{|\Psi(0)|^2} \]  

(2)

where \( \alpha \) is the total and \( \alpha_s \) the \( s \)-electron conversion coefficient. \( |\Psi(0)|^2 \) is the total \( s \)-electron density of the shells contributing to the conversion. (In \( ^{119}\text{Sn} \) \( \alpha_x = 0 \), therefore \( |\Psi(0)|^2 \) must not be included.) Equations (1) and (2) combined give:

\[ C = \frac{\alpha}{\alpha + 1} \frac{1}{|\Psi(0)|^2} \text{ IS} \quad \text{and} \quad \frac{\Delta R}{R} = \frac{5}{4 \pi 2 e^2 \hbar^2} . \]  

(3)

(\( R = 5.90 \text{ fm}, \alpha = 5.12, \alpha_s = 4.75, |\Psi(0)|^2 = 24400 \text{ a.u.} \) \(^2\))

In order to determine \( C \) the IS and \( (\Delta \lambda{\lambda}) \) were measured for various source combinations.

Four sources were chosen: \( \text{Sn(SO}_4\text{)}_2, \text{SnS}_2, \text{Sn}, \) and \( \text{SnCl}_2 \). To avoid time filtering effects \(^3\) due to resonance absorption in the sources special care was taken in the preparation of thin sources. For the same reason the sources chosen had a low Debye-Waller factor at room temperature.

The IS of the sources were determined at 80 K by conventional Mössbauer technique. It can be shown that shifts due to the second order Doppler effect may be neglected.

The decay constants were measured with a time-to-amplitude converter (TAC) \(^b\). The 26-keV X-ray from the highly converted \( \chi \)-transition which populates the 23.6 keV level in \( ^{119}\text{Sn} \) served as a start, the 24 keV \( \gamma \)-ray as a stop pulse for the TAC (see insert Fig. 1). As scintillator \( \text{NaI(Tl)} \) crystals were used. Single channel analyzers and a slow coincidence circuit selected the proper transitions. The events were stored as a function of time in the four quadrants of a multichannel analyzer. A typical example is shown in Fig. 1. Since the detectors cannot distinguish between start and stop pulses, the spectrum is symmetric. The four sources were interchanged automatically every ten minutes to average out drifts in the electronics. The exact time scale was de-
determined with a 100 MHz quartz 'time calibrator'. The two halves of the symmetrical decay curve were least square fitted to \( N(t) = a + b \exp(-\lambda t) \). The central part of the spectrum up to ± 10 ns which may contain some prompt coincidences was discarded. The time resolution of the spectrometer as determined with a \(^{125}\)I source (27 keV) was 6 ns FWHM.

In Fig. 2 the decay constants of the four sources are plotted against the IS. The calibration constant \( C \) and \( \Delta R/R \) are obtained in the Mössbauer unit \( \text{mm/s} \) from the slope of the straight line

\[
C = +\left(9.3 \pm 1.5\right) \times 10^{-2} \text{ mm/s}
\]

\[
\frac{\Delta R}{R} = +\left(1.74 \pm 0.30\right) \times 10^{-4}
\]

The half life of the 24 keV level determined is

\[
T_{1/2} = (18.29 \pm 0.10) \text{ ns}
\]

Compared with earlier measurements \(^1\) the main advantage was the symmetry in the time spectrum and the very simple decay scheme. No corrections were necessary for nonlinearities of the background and counting rate influences on the TAC. Since no indications for systematic errors exist the quoted error is purely statistical.

The value \( \Delta R/R \) may be compared with some of the newer results. Mössbauer measurements combined with Knight shift data \(^5\) give \( \Delta R/R = (1.8 \pm 0.4) \times 10^{-4} \). IS measurements of tin atoms isolated in rare gas matrices \(^6\) give \( \Delta R/R = 0.74 \times 10^{-4} \). A reevaluation of conversion electron data by Emery and Perlman \(^7\) gives \( \Delta R/R = (1.8 \pm 0.4) \times 10^{-4} \).

References:
Previous treatments of paramagnetic relaxation effects in P.A.C. were limited to the case of very short electronic relaxation times $T$. We wish to show that by adaptation of the relaxation theory devised by Hirst for Mössbauer spectra, it becomes possible to compute the perturbation factors for arbitrary $T$.

Let us consider a paramagnetic impurity with a radioactive cascade $I \rightarrow I \rightarrow I'$, and assume that the intermediate state $I$ observed in P.A.C. exhibits a hyperfine interaction $H = A \cdot \mathbf{S}$, where $\mathbf{S}$ is the effective spin of the lowest electronic level of the impurity. Coupling of $\mathbf{S}$ with the lattice gives rise to an electronic relaxation characterized by $T$. When $T$ is very short, this relaxation gives rise to an attenuation of the correlation which was computed by Abragam and Pound, assuming a fluctuating Hamiltonian $H_S$ with correlation time $T_{\text{cor}}$.

In order to treat this last region, we have adapted to this problem the perturbation theory developed by Hirst for Mössbauer spectra. In this approach relaxation takes place between the levels of the coupled system $(I, S)$ and is due to the coupling of $S$ with the lattice, represented by a Hamiltonian $H_S$ acting only on $S$. This relaxation Hamiltonian has a correlation time which is of the order of a lattice fluctuation time, i.e., very short and this ensures the validity of the perturbation treatment. On the other hand, as in [1], we drop the "secular approximation", in order to be able to handle the region where $(\Delta \omega T) \approx 1$, and we assume that the relaxation of $\mathbf{S}$ has "spherical symmetry" ("extreme narrowing" and isotropic instantaneous fluctuations).

The perturbation factor can be written as [2]:

$$G_{k', k}^{r, s}(t) = \text{Tr} \left\{ T_k^{r} (I) \Lambda (t) T_k^{s} (I) \Lambda^{\dagger} (t) \right\}$$

where $A$ is the evolution operator in level $I$ and $T_k^{r} (I)$ is a tensor operator which can be expressed in terms of the eigenstates $|m\rangle$ of $I$:

$$T_k^{r} (I) = \sum_{m'=m} (-1)^{m-m'} \sqrt{2k+1} \begin{pmatrix} m & k & m' \end{pmatrix} |m\rangle \langle m'\rangle$$

Let us define $F = I + S$; $F = I + S$, ..., $I - S$. The eigenstates of $A$ are $|F\rangle$. One can construct a new set of tensor operators adapted to the coupled system $(I, S)$ [3]:

$$V_k^{r} = \sum_{F} (-1)^{F+k} \sqrt{2F+1} \begin{pmatrix} F & k & F' \end{pmatrix} |F\rangle \langle F'\rangle <F'|F>$$

The $T_k^{r} (I)$ can be expressed in terms of the $V_k^{r}$:

$$T_k^{r} (I) = \sum_{F} \rho_k V_k^{r}$$

where $\rho_k = (-1)^{F+k} \sqrt{(2F+1)(2F'+1)} \begin{pmatrix} F & k & F' \end{pmatrix}$. This leads to a new expression of the perturbation factor:

$$G_{k', k}^{r, s}(t) = \text{Tr} \left\{ \sum_{F} \rho_k V_k^{r} \rho_{k'} V_{k'}^{s} \left\{ \rho F V_{k'}^{s} (t) \right\}^{\dagger} \right\}$$

As in ref [2], we may relate $G$ to the evolution of the density matrix $\mathbf{V}$ of the level $I$.

Let us expand $\mathbf{V}$ in terms of the $V_k^{r}$:

$$\mathbf{V} = \sum_{k} \sum_{k'} \rho_{k} V_{k}^{r} \left( \rho_{k'} V_{k'}^{s} (t) \right)$$

$\mathbf{V}$ is related to the $\mathbf{V}$ by an equation of the form:

$$\rho_{k} V_{k}^{r} (t) = \sum_{k'} \rho_{k'} V_{k'}^{r} (t) \int_{0}^{t} g_{k' \rightarrow k} (\tau) \rho_{k'} V_{k'}^{s} (\tau) \, d\tau$$
and it can easily be shown by identification of corresponding terms that:

\[ G_{kk}^{q_q}(t) = \frac{1}{2\pi^4} \sum_{\xi\xi'} \rho_{\xi'} \rho_{\xi} a_{\xi} a_{\xi} g_{\xi} \rho_{\xi} \rho_{\xi} \xi(k_{\xi} q_{\xi} q_{\xi} \xi) \xi(k_{\xi} q_{\xi} q_{\xi} \xi) (t) \]

Hence \( G \) will be obtained by integration of the equation of evolution of \( T \).

In order to compute the relaxation effects due to \( T \) acting on \( S \), it is interesting to use an intermediate step a decoupled tensor basis:

\[ k_{\xi} k_{\xi} U_{k}^{q} = \sum_{q_{\xi} q_{\xi}} \langle k_{\xi} k_{\xi} q_{\xi} q_{\xi} \xi | k_{\xi} \rangle T_{k_{\xi}}^{q}(j) \xi T_{k_{\xi}}^{q}(j) \xi \]

The \( U_{k}^{q} \) and \( V_{k}^{q} \) are related by equations involving 9 \( j \) symbols [3]:

\[ F_{k_{\xi}} U_{V_{k_{\xi}}}^{q} = \sum_{k_{\xi} k_{\xi}} \sqrt{(2J+1)(2J'+1)(2k_{\xi} + 1)} \left\{ \begin{array}{ccc} \xi & \xi & k_{\xi} \\ S & S & k_{\xi} \\ F & F & k \end{array} \right\} k_{\xi} k_{\xi} U_{k_{\xi}}^{q} \xi \]

Assuming that the relaxation of \( S \) has spherical symmetry a straight-forward transposition of the results obtained in [6] leads to:

\[ \frac{d}{dt} T_{k_{\xi}}^{q}(j) = -\lambda_{k_{\xi}} T_{k_{\xi}}^{q}(j) \]

where \( \lambda_{k_{\xi}} \) is given by (in the notation of eq (29) of [5]):

\[ \lambda_{k_{\xi}} = \sum_{r_{\xi} r_{\xi}} \frac{1}{4} \langle k_{\xi} k_{\xi} q_{\xi} q_{\xi} \xi | k_{\xi} \rangle T_{k_{\xi}}^{q}(j) \xi T_{k_{\xi}}^{q}(j) \xi \left( 1 - (2J+1) \text{V}(S k_{\xi} S S S) \right) \]

where \( S(k_{\xi}) \) being the order of the tensor interaction \( k_{\xi} \) responsible for the relaxation.

Making use of all these results one gets:

\[ \frac{d}{dt} T_{k_{\xi}}^{q}(j) = -\lambda_{k_{\xi}} (E_{F} - E_{G}) T_{k_{\xi}}^{q}(j) - \sum_{F \neq G} \beta_{k_{\xi}} (FG \leftrightarrow F'G') T_{k_{\xi}}^{q}(j) - \sum_{F \neq G} \gamma_{k_{\xi}} (FG \leftrightarrow F'G') T_{k_{\xi}}^{q}(j) \]

where \( E_{F} \), \( E_{G} \) are eigen values of \( A \) 's and where:

\[ \beta_{k_{\xi}} (FG \leftrightarrow F'G') = \sum_{r_{\xi} r_{\xi}} \left\{ \begin{array}{ccc} \xi & \xi & k_{\xi} \\ S & S & k_{\xi} \\ F & F & k \end{array} \right\} k_{\xi} k_{\xi} U_{k_{\xi}}^{q} \xi \]

We have solved this set of equations for \( I = 2 \) and \( S = 1/2 \). In this special case there is only one electronic relaxation constant: \( \lambda_{k_{\xi}} = 1/T_{1e} \). The corresponding results for \( G_{1e} \) and \( G_{1o} \) exhibit a damped oscillating behaviour when \( T_{1e} N/A \) and a purely damped behaviour when \( T_{1o} N/A \) (Abragam and Pound limit).

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According to the well-known model of Abragan and Pound [1] for randomly fluctuating extranuclear fields the perturbed angular correlation attenuation coefficients turn out to be noteworthy simple if two restricting conditions are introduced: (1) The width \(\sigma^2\) of the distribution of the field strength and/or the correlation time \(\tau_0\) has to be sufficiently small such that \(\sigma^2/\tau_0 \ll 1\); (2) The typical time of observation of the angular correlation has to be large compared to the correlation time, i.e., \(t \gg \tau_0\) in a time-differential experiment or \(\tau > \tau_0\) in a time-integral measurement, where \(\tau\) is the lifetime of the nuclear state. The first condition secures the irrelevance of the probability distribution of the perturbing fields. The second condition means that, strictly speaking, the model does not describe a situation where good time resolution allows to observe the attenuation in the region \(t \sim \tau_0\). The Abragan and Pound expression is a "steady state solution" and the transition to the region \(t \ll \tau_0\), where most nuclei actually witness static fields, is not correctly described.

In a more detailed stochastic model it is possible to drop the second condition while the much simplifying first condition can be retained. The result of such a calculation within the framework of a rather general stochastic model [2] is given below. The model assumes successive reorientation of the axis of interaction of the extranuclear field in a random time sequence, described by Poisson statistics, and spherical symmetry of the fields at any time. The statistics of possible simultaneous changes of the interaction strength is in the limit \(\sigma^2/\tau_0 \ll 1\), fully described by the r.m.s. width \(\sigma^2/\tau_0\) of an arbitrary probability distribution of the perturbing fields. For fluctuating classical magnetic field \(H\) of this type, the following time-differential attenuation coefficients are obtained (with the superscript \(M\) for "magnetic"):

\[
\langle\nu_{kk}(t)\rangle = \exp\left[-\frac{t}{2\tau_0}\right] \cos\left(\frac{x^2}{2\tau_0}\right) + \frac{1}{2}\sin\left(\frac{x^2}{2\tau_0}\right); \quad (1)
\]

\[
\nu_k = \sqrt{1 - 4c_k^2} \langle\nu_{kk}(0)\rangle; \quad \langle\nu_{kk}(0)\rangle = \left(\frac{\sigma^2}{4(2\pi)^2}\right) \langle\nu_{zz}(0)\rangle;
\]

\[
\nu_k = \sqrt{2} \sum_{N=1}^\infty \frac{2}{(2k+1)} \frac{1}{2(k+1)} \langle\nu_{zz}(0)\rangle < 1.
\]

For the interaction of the nuclear quadrupole moment \(Q\) (spin \(I\)) with a fluctuating (classical) electric field gradient \(\nu_{zz}\) the following parameters have to be used in eq. (1) (superscript \(E\) "electric"):

\[
\nu_k = \left(1 - 4c_k^2\right) \langle\nu_{zz}(0)\rangle; \quad \langle\nu_{zz}(0)\rangle = \left(\frac{\sigma^2}{4(2\pi)^2}\right) \langle\nu_{zz}(0)\rangle;
\]

\[
\nu_k = \sum_{m=\pm I} g_m^2 (m^2 - m^2)^2 \left\langle\delta_{k-m,0} \delta_{m,0}\right\rangle \frac{1}{2}(k+1)[2I(I+1)-k(k+1)-1].
\]

The time-integral attenuation coefficients for magnetic or electric interaction are found from eq. (1) to be

\[
\langle\nu_{kk}(t)\rangle = \left(1 + \frac{\nu_{kk}(0)}{1 + \tau_0/\tau}\right)^{-1} \quad \langle\nu_{zz}(0)\rangle < 1. \quad (2)
\]

Eqs. (1) and (2) reduce to the attenuation coefficients of the Abragan and Pound model

\[
\langle\nu_{kk}(t)\rangle_{A.P.} = \exp\left(-\frac{\nu_{kk}(0)}{\tau_0}\right) \tau_0,
\]

if the additional condition \(t/\tau_0 \gg 1\) or \(\tau_0/\tau \gg 1\) is introduced.

In fig. 1 two examples of eq. (1) are compared to eq. (5). The more general attenuation coefficients always start with vanishing slope (similar to static interactions!) and there is always only one point of intersection with eq. (5). The maximum relative deviation

\[
\Delta_{max} = \frac{\langle\nu_{kk}(t)\rangle_{A.P.} - \langle\nu_{kk}(t)\rangle_{A.P.}}{\langle\nu_{kk}(0)\rangle_{A.P.}},
\]

turns out to occur at a time \(t\), which coincides with the point of inflection of eq. (1). In fig. 2 the dependence of \(\nu_{kk}(t)\) on the parameter \(\sigma^2/\tau_0\) is shown for the magnetic case, together with \(\Delta_{max}\).

In the region of interest the deviation is several per cent.

In the first order treatment of Abragam and Pound [1] for the attenuation coefficients of angular correlations perturbed by randomly fluctuating extranuclear fields two parameters are introduced: the second moment $\langle\omega^2\rangle$ of the probability distribution of Larmor-frequencies in the sample and the correlation time $\tau_c$. The theory applies in the limit $\langle\omega^2\rangle^{1/2}<<<<\tau_c$ only, but as a compensation, no details of the probability distribution generating the second moment $\langle\omega^2\rangle$ need be known.

In an attempt to extend the scope of the theory to arbitrary strengths of interaction $\langle\omega^2\rangle^{1/2}>>>\tau_c$, one faces the general fact that the moment-generating distribution is no longer irrelevant. Instead one has to cope with the stochastic details of the fluctuating interaction. In order to meet this mathematical requirement either the shape of the distribution has to be specified or all its moments $\langle\omega^n\rangle$, $n = 1, 2, \ldots$, must be introduced as parameters (the even moments only are relevant in a spatially isotropic interaction). The first possibility has been fully described in a recent paper [2]. The attenuation coefficients can always be calculated, at least numerically, if the analytic form of the distribution of the perturbing extranuclear fields is introduced into the theory. From the experimental point of view, however, this approach appears to be rather inconvenient because usually the stochastic details of the interaction are crudely known only. The moment method, on the other hand, offers the possibility of performing a higher order treatment of the attenuation coefficients. As has been shown in ref. [2] a series expansion in powers of $\langle\omega^2\rangle^{1/2}$ can be generated (in the Laplace space). The first order attenuation coefficients (which correspond to the Abragam and Pound model) are obtained by neglecting all the terms with $n > 1$. Results are discussed in ref. [3].

The higher order attenuation coefficients will obviously comprise the moments $\langle\omega^n\rangle$ as additional parameters. As a simple example the calculation of the second order attenuation coefficients ($n = 2$) is sketched below. For a randomly fluctuating magnetic field $H$ of the type assumed in ref. [3], the Laplace transformed attenuation coefficients are found to be (superscript $M$ for "magnetic"):

$$\langle G_{kk}(p) \rangle = \frac{\tau_c k^2}{c - p^2 + c \langle\omega^2\rangle_{kk}^c + \frac{\tau_c}{c} \langle\omega^2\rangle_{kk}^c - \frac{\tau_c}{c} \langle\omega^2\rangle_{kk}^c - \tau_c^2},$$

(1)

where $\tau$ is the lifetime of the intermediate nuclear state. (From the character of eq. (4) it can be seen that the inclusion of still higher moments simply adds terms according to an obvious rule.)

In this approach the fourth moment $\langle\omega^4\rangle$ appears as an additional parameter which must be fitted experimentally if the mean strength of interaction is supposed to be beyond the Abragam and Pound limit. Stated otherwise: The angular correlation comprises information on the stochastic details of the perturbing fields if the mean strength of interaction is sufficiently high. In the Abragam and Pound limit all such information is lost.

We note that eqs. (3) and (4) are not subject to the restricting condition $\tau >>> \tau_c$ or $\tau >>> \tau_c$ (a discussion of the implication of this point is given in ref. [2]). The second order calculation can easily be extended to the perturbing interaction of the nuclear quadrupole moment $Q$ (spin 2) with a randomly fluctuating electric field gradient $V_{zz}$ of axial symmetry. The parameters in eqs. (1) and (4) must be replaced by (superscript $E$ for "electric"):

$$E = \sum_{n,m} g(n^2 - m^2)^2 \langle I, I, k | A^{I, I, k}_{m', -m', -m'} \rangle^2 ;$$

$$E = \sum_{n,m} g(n^2 - m^2)^2 \langle I, I, k | A^{I, I, k}_{m', -m', -m'} \rangle^2 ;$$

(4)

$$\langle G_{kk}(p) \rangle = \frac{\tau_c k^2}{c - p^2 + c \langle\omega^2\rangle_{kk}^c + \frac{\tau_c}{c} \langle\omega^2\rangle_{kk}^c - \frac{\tau_c}{c} \langle\omega^2\rangle_{kk}^c - \tau_c^2} \exp \left[ \left( \frac{\tau_c}{c} \langle\omega^2\rangle_{kk}^c - \tau_c^2 \right) \right].$$

IV. 53 Spin relaxation phenomena in Mössbauer spectra of Dysprosium impurities implanted in Iron

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Mössbauer effect measurements on the implanted system DyFe using the 26 keV (3/2\(^-\) \rightarrow 5/2^+\) transition of \(^{161}\)Dy were first reported by Inua\(^1\)\(^,\)\(^2\). We have extended these measurements with one source at four different temperatures and a narrower Dy\(^{3+}\) absorber linewidth. In the iron host the energy levels of the Dy\(^{3+}\) ion, with ground state \(J = 15/2\), are split by exchange interaction between the Dy-spin \(S\) and the conduction electron spins \(s\). In a molecular field approximation the interaction Hamiltonian can be written as

\[ H = -J_S \vec{S} \cdot \vec{S}_{\text{exch}} \]

(1)

In the spectrum taken at 5K we observe the whole ionic hyperfine splitting, thus justifying the neglect of the cubic crystalline electric field in calculating the electronic levels. The exchange field gives rise to 16 levels, at equal distances

\[ \Delta = (g_J^1 - 1) \mu_B H_{\text{exch}} \]

(2)

The relaxation phenomena in the spectra taken at higher temperatures are treated by a relaxation model based on the rate-equation methods\(^3\)\(^,\)\(^4\), described in detail by Khurgin et al\(^6\). As is shown by these authors we can describe the intensity distribution of the \(n\)th component of the Mössbauer spectrum by

\[ S(n, \omega) = \Re [H(n, A(n)^{-1})] \]

(3)

and the complete Mössbauer spectrum by

\[ S(\omega) = \sum_{n=1}^{16} S(n, \omega) \]

(4)

In eq. (3), \(I\) is a columnvector of 16 ones, \(W(m) = C(m) \left[ p(J), p(J-1), \ldots, p(-J) \right]\) where \(C(m)\) is the nuclear transition probability and the population densities are given by

\[ p(J) = \exp(-\Delta J / kT) / \sum_{J=-J}^{J=J} \exp(-\Delta J / kT) \]

(5)

The matrix \(A(n)\) is given by

\[ A(m) = -\epsilon (m) I + I (p(m) - I) + \eta \]

(6)

where \(\epsilon(m)\) is the Mössbauer linewidth and \(\eta = \omega / (T + I)\). In the last expression \(\omega_k\) is the frequency of the \(n\)th Mössbauer transition for a given value of \(J\), between the electronic levels are given by

\[ \omega_k = \frac{\left( I + 1 \right) \left( J(J+1) - k^2 + 1 \right)}{2J(2J+1)} \]

(7)

In this expression \(H_{\text{exch}}(J)\) is proportional to \(J\), while \(\omega_k\) is proportional to \(\Delta(J+1)\). Assuming that the Dy electronspin relaxation via the conduction electrons (i.e. through the off-diagonal elements of the s-f interaction\(^9\)) the transition probabilities \(\tau_{kl}\) between the electronic levels are given by

\[ \tau_{kl} = \frac{(2k)\left( J(J+1) - k^2 + 1 \right)}{2J(2J+1)} \exp(\Delta / kT) \]

(8a)

(8b)

This is analogous to the Korringsa relaxation of nuclear spins in a metal but without the approximation that the energies associated with the spin flips are small compared to \(kT\). \(C\), a Korringsa-like constant, given by \(7) \]

\[ C = \frac{4\pi^2 \hbar^2 (g_S - 1)^2}{9 \alpha^2} \]

(9)

is a parameter in the model. Here \(g_S\) is the density of states per atom per spin of the host at the Fermi level. The other parameter is \(\Delta\), the distance between the electronic levels.

We expect \(C\) to be temperature independent, and \(\Delta\) to be proportional to the \(\alpha\) magnetisation. The source for the experiment was obtained by irradiating \(7\) mg. of \(^{160}\)Gd\(_2\)O\(_3\) enriched to 90% in \(^{161}\)Gd for 4 days in a thermal neutron flux of \(2.10^{14} \text{ cm}^{-2} \text{ sec}^{-1}\). The desired activity \(^{161}\)Tb \((I = 7/2)\) at 76 was the decay product of \(^{161}\)Gd\(_2\)O\(_3\) \((I = 5/2)\) and \(^{161}\)Tb was separated from the irradiated oxide in the Groningen isotope separator\(^8\) by means of the CC\(_{13}\)-method\(^10\) and implanted with an energy of 13 keV in a 99.99% pure iron foil. The implanted dose was about 10\(^10\) atoms/cm\(^2\), the source strength 3*0 \(\mu\)Ci. The 11 mg/cm\(^2\) atoms/cm\(^2\). The desired activity \(\Phi\) days in a thermal neutron flux of \(2.10^{20} \text{ cm}^{-2} \text{ sec}^{-1}\). The density of states per atom per spin of the host at the Fermi level was better than 1K.

The Mössbauer spectra are given in the figure. At 5K only the lowest electronic level \(J = 15/2\) of the Dy\(^{3+}\) ion is populated. In that case the spectrum can be fitted by the sum of 16 Lorentzians, the positions of which can be found from Eq.(7) by taking \(J = 15/2\). A good fit was obtained by taking the linewidth \(\Gamma\) of the two central lines in the spectrum smaller than that of the 11 other lines (\(\Gamma\)). The ratio \(\hbar \omega_0 / \Gamma = 1.236 \pm 0.006\% \) was kept constant in the fit. The results are:

<table>
<thead>
<tr>
<th>(T(K))</th>
<th>(\Delta(K))</th>
<th>(C(\text{sec.K}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>159</td>
<td>94</td>
<td>10.10(^{-8})</td>
</tr>
<tr>
<td>273</td>
<td>80</td>
<td>(4.6\times10^{-8})</td>
</tr>
<tr>
<td>363</td>
<td>89</td>
<td>2.5\times10^{-9})</td>
</tr>
</tbody>
</table>

The errors are statistical. There is an uncertainty of about 2% in these values because of difficulties with the absolute velocity calibration. Using the above values theoretical spectra were calculated according to the relaxation model. These spectra were fitted to the measured spectra at 15K, 27K and 36K giving the following values for the parameters \(A\) and \(C\):

\[ T_2 = 0.88 \pm 0.06 \text{ cm/sec} \]

The estimated error in \(\Delta\) is about 10%; that in \(C\) about 50%.
Using (2), we derive from the obtained values of \( \Delta \): 

\[
\langle |c_s| \rangle^2 = 4.0 \pm 0.4 \text{ MOe and}
\]

\[
J_{\text{exch}}[c_s] = 2.3 \times 10^{-2} \text{eV. The value found for } H_{\text{exch}}
\]

can be compared to that found by Bernas and Gabriel\(^3\) for TmFe (2.5 \pm 0.5 \text{ MOe}). Assuming \( J_{\text{ex}} \) to lie between \( +0.1 \) and \( 0.4 \) eV as has been found for Dy\(^3\) in various metals\(^4\) we find for \( \langle |c_s| \rangle \), the absolute value of the conduction electron polarization at the rare-earth site, a value between about \( 0.05 \) and \( 0.20 \). Using the value found for \( C \) at \( 159\text{K} \) we further calculate from (9) \( n^e = 0.25 \pm 1.0 \text{ Ry}^2 \). This is much smaller than the total density of states (30 \text{ Ry}^2)\(^5\), but compares well with the estimated value of the S-density (0.2-0.6 \text{ Ry}^2)\(^6\). From the relaxation model we expect \( C \) to be constant. We see, however, that \( C \) decreases when \( T \) increases.

From the figure it can be seen that there are systematic differences between the observed and calculated spectra. The line broadening observed at high velocities in the 5K spectrum (which is only for a small part due to geometrical effects) indicates that the Dy\(^3\) ions occupy more than one lattice site, each site giving rise to a slightly different hyperfine interaction. This is consistent with channeling experiments on rare earth ions implanted in Fe\(^{17}\) from which it is found that only about 50% of the ions occupy substitutional sites. It is thought that the other fraction occupies sites associated with vacancies, giving rise to a strong axial component of the crystalline electric field and thus to a different electronic energy level spacing. An analysis of the spectra using a simple “two-component” model is in progress.

17. B.L. Deutch in ref.2 p.137.
IV. 54 SPIN-LATTICE RELAXATION MEASUREMENTS BY NUCLEAR ORIENTATION METHODS

Spin-lattice relaxation at very low temperatures (T<0.1K) has been the subject of many investigations, both experimental and theoretical, since the first measurement using the technique of nuclear magnetic resonance on oriented nuclei (NMR/ON). Non-resonant methods may also be used, and it is the purpose of this paper to discuss briefly the merits and difficulties associated with the different methods and draw attention to a serious discrepancy between values of the relaxation rate for *Co spins in iron measured by various groups, including ourselves, using the different techniques.

In the NMR/ON method, the ensemble of nuclei with spin, J, is oriented by hyperfine interaction, and the degree of orientation is altered by application of an rf field with a frequency satisfying the resonant condition hv = ΔE, where ΔE is the separation in energy between magnetic substates characterized by M(±1/2±1). It is well known that at a temperature, T, low enough that hv is not much smaller than KT, the usual high temperature approximation, commonly used in NMR theory, is not applicable and exact theory must be employed. A requirement is that the initial state of the spin system be known. However, in all experiments performed to date on ferromagnets by the NMR/ON method only partial saturation of the resonance has been attained. Also attempts to achieve complete reversal of the magnetization by fast passage methods have been unsuccessful. Consequently, for samples with spin concentrations so low that a common spin temperature cannot be assured (usually the case in NMR/ON experiments), the initial state of the system is not known. It follows that there is some uncertainty in interpreting the experimental data. This point has been discussed for example by Stone. (Actually if all orientation parameters, <M>, <M^2> - 1<[I+1]>, etc. were measured, the population distribution would be completely determined).

This difficulty is obviated in the various non-resonant methods used to determine relaxation times. With the exception of the technique used by Stone et al. which is limited to very few radioactive decays these methods rely on changing the lattice temperature leaving the spin system in a known Boltzmann distribution. In the first experiments the temperature change was produced by eddy current heating incurred on changing the polarizing field on the sample. However, the 'fast demagnetization' method proposed by Barclay et al. appears most attractive. In their experiment, the demagnetizing field on the paramagnetic cooling salt was held at a finite value so that salt and sample cooled to some known initial temperature. The demagnetizing field was quickly reduced to zero and the relaxation of the spins to the known final temperature was monitored through the changing anisotropy of nuclear radiations. In this method both initial and final temperatures are known, although there is some uncertainty concerning the response of the system during the time taken to reduce the field.

Using the fast demagnetization technique, Barclay et al. measured a value for the Korringa constant for *Co in iron in high field of C = 1.75 ± 0.15 sec.K. While being in agreement with their own measurement by the single passage technique, this value differs considerably from previous determinations obtained by resonant and non-resonant methods, including those by conventional NMR on *Co in iron at 1K. These experiments gave C = 2.5 sec.K. Bacon et al. obtained values of 1.75 ± 0.10 sec.K or 2.5 depending on the way they analysed their data. In Table 1 we list the various determinations of C excluding early values deduced using incorrect theory. These values all pertain to the fully magnetized sample since the relaxation rate increases as the magnetic field is reduced. The values obtained from NMR measurements on *Co have been converted to those appropriate to *Co. The discrepancy in C is quite puzzling. Values deduced from the NMR/ON measurements might be suspected because of uncertainty in initial conditions, but on the other hand they are in agreement with values determined by NMR.

We decided to re-measure the Korringa constant for *Co using the non-resonant, fast demagnetization technique. The experiment was performed in a manner very similar to that described above. Various starting fields were utilized so that relaxation through a range of different initial and final temperatures could be investigated. The deduced value of C was 1.75 ± 0.2 sec.K in excellent agreement with that obtained by Barclay et al.

Next we performed an NMR/ON experiment on a sample of iron containing about 0.1 at.% of *Co. In this case, it is reasonable to assume the existence of a spin temperature in the relaxation. It follows that the population distribution among the M states of the spin system can be deduced at any time from a measurement of the y-ray anisotropy, and the relaxation rate determined definitely. In this case, we measured a value of C = 2.5 ± 0.2 in excellent agreement with the values obtained previously by NMR/ON methods.

Thus the discrepancy between the values obtained by resonant and non-resonant methods remains. Because of agreement with NMR one should favour the results of the NMR/ON experiments. A possible explanation for the fast demagnetization method yielding a low value of C might be that the copper fins connecting sample to cooling salt are cooled by nuclear demagnetization. If the iron lattice also were cooled initially below the final temperature, the observed relaxation rate would appear to have a lower value. However, estimates of the cooling effect and the long spin-lattice relaxation time of the copper nuclei make this an unlikely explanation. On the other hand, we can propose no other reason for the discrepancy.
TABLE I
Values of the Korringa constant, C, measured for $^{60}\text{Co}$ in iron by various techniques

<table>
<thead>
<tr>
<th>Method</th>
<th>C (sec.k)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Non-resonant; eddy current heating</td>
<td>2.6 ± 0.2</td>
<td>12</td>
</tr>
<tr>
<td>NMR/ON; fit to tail</td>
<td>2.5</td>
<td>10</td>
</tr>
<tr>
<td>NMR/ON; fit to initial slope</td>
<td>1.76 ± 0.10</td>
<td>10</td>
</tr>
<tr>
<td>NMR/ON; no spin temp.</td>
<td>2.6 ± 0.15</td>
<td>11</td>
</tr>
<tr>
<td>NMR/ON; spin temp.</td>
<td>2.3 ± 0.15</td>
<td>11</td>
</tr>
<tr>
<td>NMR/ON; single passage</td>
<td>1.75 ± 0.15</td>
<td>6</td>
</tr>
<tr>
<td>Non-resonant; fast demagnetization</td>
<td>1.75 ± 0.15</td>
<td>6</td>
</tr>
<tr>
<td>NMR; 1% $^{59}\text{Co}$-Fe powder</td>
<td>2.7</td>
<td>13</td>
</tr>
<tr>
<td>NMR; 1% $^{59}\text{Co}$-Fe foil</td>
<td>2.3 ± 0.2</td>
<td>14</td>
</tr>
<tr>
<td>Non-resonant; fast demagnetization</td>
<td>1.75 ± 0.2</td>
<td>This work</td>
</tr>
<tr>
<td>NMR/ON; 0.1% $^{60}\text{Co}$-Fe foil</td>
<td>2.5 ± 0.2</td>
<td>This work</td>
</tr>
</tbody>
</table>

References
3. B.G. Turrell, in 'Hyperfine Interactions and Nuclear Radiations', edited by E. Matthias and D.A. Shirley (North Holland, Amsterdam, 1968) 803
For a single passage sweep of the nuclear magnetic resonance in a system of polarized nuclear spins, a redistribution of populations $P(m)$ will occur, characterized by the nature of the quadrupole interactions present and the size of the adiabatic parameter $\gamma_H^2 \frac{k^2}{2K}$. For the regime $\gamma_H^2 \ll \gamma K H_0$ (as described in the previous paper) the population redistribution in the successive $|m\rangle, |m+1\rangle$ subresonances can be simply expressed in two limits:

1. $\gamma_H^2 \frac{k^2}{2K} \ll 1$
   \[ \Delta P(m+1) = \frac{1}{2} \left[ P(m) - P(m+1) \right] \frac{\pi}{2K} \gamma_H^2 \frac{k^2}{2} \]
   \[ \Delta P(m) = P(m+1) - P(m) \]

2. $\gamma_H^2 \frac{k^2}{2K} \gg 1$
   \[ \Delta P(m+1) = \frac{1}{2} \left[ P(m) - P(m+1) \right] \frac{\pi}{2K} \gamma_H^2 \frac{k^2}{2} \]
   \[ \Delta P(m) = P(m+1) - P(m) \]

where $k^2 = 1/(1-1) - m(m+1)$ and $K = \frac{3\omega}{\beta K}$. Under conditions of high adiabatic parameter, successive population inversion occurs through the $|m\rangle, |m+1\rangle$ subresonances separated in frequency by $\gamma H_0/2(1-1)$ resulting in an overall cyclic population shift up or down one place depending on the relative sign of $P$ and $\gamma_H^2 \frac{k^2}{2K}$. Any overall population changes in the $|m\rangle$ eigenstates will consequently alter the observed $\gamma$-anisotropy through a change in the $B_i$ parameters so that the NMR signal is given by the fractional change in anisotropy.

Figure 1 shows the result of a single passage sweep through the NMR line of polarized $^{60}$Co in Fe for an adiabatic parameter consistent with full cyclic inversion as described above. The fractional change in $\gamma$-anisotropy is shown for both sweep directions and the subsequent relaxation is plotted against time after sweep. The observed dependence of signal upon sweep direction and the characteristic of the relaxation are consistent with cyclic inversion predictions indicating the existence of an electric quadrupole hyperfine interaction for this system in the range $\gamma H_0/2(1-1)$ to $10^2$ Hz.

Full $\frac{3\omega}{\beta K}$ asymmetry will result only if all nuclei experience an electric field gradient of unique sign with a fixed axis of symmetry. Such would be the case if the gradient were to arise from an unquenched orbital moment in the atomic electron shells. The present experiments were performed in a polycrystal host for which the distribution in microcrystal orientation leads to a variation in the sign and magnitude of $P = \frac{3\omega V_{xx}}{41(21-1)} (3 \cos^2 \alpha - 1)/2$ if $V_{xx}$ is associated with the microcrystal axes. Such a description would be applicable if the quadrupole interaction were to arise from a breaking of cubic symmetry by magnetostriction or lattice defects. If $V_{xx}$ were unique then the number of nuclei for which $(3 \cos^2 \alpha - 1)$ takes positive or negative values is different despite the fact that $\alpha$ ranges over all angles. In the analysis of the $^{60}$Co in Fe experiment, $V_{xx}$ is taken to be positive so that $P$ is positive for a fraction $1 - \frac{3\omega}{2K}$ of nuclei and negative for $\frac{3\omega}{2K}$. These weighting factors are used in the calculation of the $\gamma$-anisotropy changes shown in figure 1 where longitudinal relaxation following passage was simulated by an iterative procedure. The Korringa constant so determined is $2.3 \pm 0.1$ in good agreement with other authors.

Much greater sweep direction dependence is expected for the case of electric field gradients arising predominantly from unquenched orbital angular momentum in the impurity atom electrons. The present model suggests great value in further experiments on high $Z$ impurity atoms where spin-orbit coupling allows such orbital angular momentum to exist.

In liquid Te the changes of the Knight shift and the magnetic relaxation time between the melting point (725K) and 1000K demonstrate an increasing metallic nature (1). The quadrupolar relaxation which would provide independent information about structural and/or dynamic behaviour, can only be studied using radioactive or impurity atoms since the stable Te isotopes have I = 0 or 1/2. We have measured the nuclear spin relaxation of the 8ms excited nuclear state 132Xe in the Te host in this temperature range.

The 1+ state (2753 keV, g = -0.195 from (2)) was produced by the reaction 130Te(a,2n) in an isotopically enriched target at the Karlsruhe cyclotron with an a energy of 35 MeV. Beam pulses of 100ns width every 20 msec were used. The alignment resulting from the nuclear reaction was observed by measuring the precession of the anisotropy of the 770 keV decay γ-ray in a perpendicular field of 8-10 Gauss using two NaI(Tl) detectors. The sample was prepared from 99.98% Te and was contained in a graphite crucible, covered with mica, in a He atmosphere.

The relaxation time $T_R$ was derived from the observed modulation of the γ-ray intensity by

$$W(\theta, t) = 1 + A_\theta e^{-t/T_R} \left[1/4 + 3/4 \cdot \cos(2\theta - 2\omega_L t)\right]$$

The results are presented in the figure. The values below the melting temperature were obtained when the liquid supercooled. In the following, we will concentrate on the temperature dependence of $T_R$.

We consider the magnetic relaxation first. According to the data for $^{125}$Te (1), the relaxation time for the present experiment would be 4ms for $T \geq 800K$ and 5.5ms at 700K if the fluctuating magnetic fields at the Xe nuclear site were equal to those for $^{125}$Te. It is supposed, however, that the magnetic relaxation due to conduction electrons is smaller for Xe in Te than for Te in Te, since it may be expected that the electrons of the closed shell Xe atom will not participate in the Te conduction band. This assumption is suggested by the fact that Xe does not enter chemical bonds except with the most electronegative elements. At lower temperatures, the observed relaxation is then dominated by the fluctuating quadrupole interactions.

The quadrupolar relaxation in liquids has been related to the atomic diffusion in several cases (see e.g. (3), (4), (5)). Along this line we approximate the temperature dependence of $T_R$ below 800K by

$$T_R = T_{R\infty} \cdot e^{-Q/K_BT}$$

which yields an activation energy $Q \approx 4$ kcal/mol (magnetic relaxation neglected). This value fits into the systematic variation of the self-diffusion Q values with melting temperature as observed in pure metals (6). A recent determination (7) of the diffusion constant for liquid Te, however, quotes an anomalously high activation energy of 35.5 kcal/mol in the small temperature range $10^3 K/T = 1.37 \ldots 1.33$, while $Q \approx 0$ (but compatible with typical values of liquid metals) is quoted for the subsequent temperature interval $(10^3 K/T = 1.33 \ldots 1.22)$. 
In ordinary liquid metals, the structural and electronic properties are essentially temperature independent, and the increase of the quadrupolar relaxation time (and of the diffusion constant) with temperature is due to the decrease of the correlation time. In liquid Te, additional effects may contribute to the temperature dependence of the relaxation rate. The electrical conductivity $\sigma$ increases by about 50% between 730K and 1000K ((8), see the figure). It can be argued that this will also result in an increase of $T_R$ - The average number of nearest neighbours $z$ changes from about 2.5 to 3 in this temperature range (9). Cabane and Friedel (10) have interpreted this increase as a transition of the atoms from two- to threefold coordination with a characteristic parameter $n_2 = z-2$ (see the figure). In this picture, the temperature dependence observed would imply a high sensitivity of the quadrupolar relaxation of the Xe impurity atom to the Te coordination number.

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IV. MAGNETIC AND QUADRUPOLE RELAXATION RATES OF EXCITED NUCLEI IN LIQUID IN AND Hg

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General Remarks: Perturbed angular correlation methods were first applied to a systematic investigation of relaxation rates $T_1^1$ in liquid metals in 1972 [1]. This method seems to be well suited for the study of the magnetic relaxation $T_1^1$ of isolated impurities and the mechanism of quadrupole relaxation $T_2^1$ in monoatomic liquid metals, information which is hard to obtain by conventional n.m.r. methods [2]. Usually $T_1^1$ can be well described by the Korringa relation

$$ T_1^1 = \frac{k(k+1)}{2} \frac{1(1^2 - 1^2)}{(1^2 - 1^2)} \cdot \left( \frac{\hbar \gamma_\nu / \gamma_\epsilon}{\gamma_\nu / \gamma_\epsilon} \right)^2 \cdot K(a) \cdot K^2 $$

with $K(a)$: electron-electron correction factor and $K$: Knightshift. The most important contribution to $T_1^1$ was assigned to the diffusional motion of ions. Starting from the general expression

$$ T_1^1 = \frac{3}{8} \frac{k(k+1)}{(1^2 - 1^2)} \cdot \left( \frac{\hbar \gamma_\nu / \gamma_\epsilon}{\gamma_\nu / \gamma_\epsilon} \right)^2 \cdot J(0) $$

Sholl [3] derived that the temperature dependence of $T_1^1$ is essentially given by

$$ T_1^1 \propto 1/DCT $$

with $D$: diffusion constant, $J(0)$: spectral density) whereas the magnitude of $T_1^1$ is sensitive to the fluctuating field gradients which depend on the two- and three-particle correlation functions, the conduction electron contribution and an antisheilding factor.

In this paper we present relaxation data for the systems $\text{SnIn}$ and $\text{PbHg}$ and evaluate information about the Knightshift, the mechanism of $T_1^1$ and quadrupole moments of ms-states from these experiments.

Experiment: Relaxation times of the $\frac{11}{2}$, 160 µs state of $^{115}\text{Sn}$ in liquid In, of the $\frac{7}{2}$, 123 µs state of $^{206}\text{Pb}$ and of the $\frac{13}{2}^+$, 5.5 ms state of $^{205}\text{Pb}$ in liquid Hg were measured by the time differential spin precession method in an external magnetic field. The levels were excited by the $^{115}\text{In(p,3n)}^-, \text{Hg}(\alpha,2n)^-$ and $\text{Hg}(\alpha,3n)$ reactions respectively using pulsed particle beams from the Berlin Van de Graaff ($^{115}\text{Sn}$ case) and from the Karlsruhe cyclotron ($^{206,205}\text{Pb}$ cases). In each case the spin precession spectra all described by $k=2$ were observed as function of the target temperature; the relaxation rates obtained are shown in Fig. 1 and Fig. 2.

Analyses: The data for $^{115}\text{SnIn}$ can be fitted well over the whole temperature range investigated, by assuming that only the mechanisms described by Eq. (1) and (2) contribute to the observed $T_1^1$. The resulting separation into the magnetic and quadrupole part is given by curves (a) and (b) in Fig. 1 respectively.

The data for the system $\text{PbHg}$ can be analyzed rigorously because the g-factors $g(13/2^+, 205\text{Pb}) = -0.153$ and $g(7^-^+, 206\text{Pb}) = -0.0217$ are very different and moreover the latter is extremely small, so that the relaxation for $^{206}\text{PbHg}$ is of almost purely quadrupolar origin. Using the known temperature dependence of $T_1^1$ for $^{206}\text{PbHg}$ it is possible to separate $T_1^1$ (curve (b)) and $T_1^1$ (curve (a) in Fig. 2) for $^{205}\text{PbHg}$ unambiguously.

Discussion A) Magnetic Relaxation $T_1^1$: From the curves (a) in the figures the values $K(a)K^2$ can be calculated by Eq. (1). Inserting for $K(a)$ the values of the pure metals, $K$-values for extremely dilute ($<1/10$ ppm) impurities can be extracted to be 0.68(5) % for Sn in In, and 2.56(15)% for Pb in Hg. These data may be compared to $K$-values of the pure liquid metals $K(\text{In}) = 0.78$ %, $K(\text{Sn}) = 0.73$ %, $K(\text{Pb}) = 1.49$ % and $K(\text{Hg}) = 2.42$ %. Since PbHg has the same anomalous Knightshift at Hg, it is an exception from the trend that K of a solute in a solvent is close to that of the pure solute.

B) Quadrupole Relaxation $T_2^1$: For both systems the temperature dependence of $T_2^1$ follows closely to the reciprocal of the macroscopic self diffusion in the host metals, which are represented by the circles in Fig. 1 and by the triangles in Fig. 2 (right-hand scales).
Theoretically the dependence of $\tau^{-1}$ on diffusion for e.g. \( \text{SnIn} \) is taken as $\tau_{\text{diff}}^{-1} = 2/[\text{D(}\text{InIn}) + \text{D(}\text{SnIn})]$. Usually the diffusion data of solutes with mass, radius and valency similar to the solvent are close to the self diffusion. Thus we conclude that $\tau_{\text{diff}}^{-1}$ in both systems is essentially caused by ionic diffusion. Possible contributions to $\tau_{\text{diff}}^{-1}$ in the system $^{208}\text{PbHg}$ due to the vibrational modes based mechanism, for which a temperature dependence $\tau_{\text{diff}}^{-1} \propto T$ should be assumed [4], can be estimated to be smaller than 1/10 of the measured rates at lower temperatures. Such a mechanism seems to contribute appreciably to $\tau_{\text{diff}}^{-1}$ in the system $^{207}\text{PoPb}$ [5].

Cl. Quadrupole Moments: Information about the Q-moments of the excited states in the Pb-isotopes can be deduced by comparing the $\tau_{\text{Q}}^{-1}$ values with those of the $^{201}\text{Hg}$ ground state in liquid Hg [6]. Correcting for the diffusion data of $^{208}\text{PbHg}$ [7] and using the assumption that the fluctuating field gradients of the systems $^{208}\text{PbHg}$ and $^{207}\text{PbHg}$ are equal within 20 % (this assumption is supported by systematic investigations of the e.f.g. of impurities in solid metals) the values $|Q(7^{-}, \text{Pb}^{206})| = 0.30(6)$ b and $|Q(13/2^{-}, \text{Pb}^{205})| = 0.27(5)$ b result from Eq. 2. The ratio $|Q(7^{-})/Q(13/2^{-})| = 1.10(9)$ follows directly from measurement. Both moments are essentially given by the $vi_{13/2}$ hole configuration and may be compared to predictions of Ring et al. $Q(vi_{13/2}) = +0.368$ b [8] and to $Q(vi_{13/2}) = +0.33$ b derived from the B(E2) value of the $12^{-} \rightarrow 10^{+}$ transition in $^{206}\text{Pb}$.

This work was supported by the Bundesministerium für Forschung und Technologie.

5. F. Dimelings et al., Contribution to this Conference
V DEVELOPMENTS IN METHODOLOGY AND APPLICATIONS
In this paper we report the sign determination for the magnetic interaction \( g \cdot H \) measured in an NMR-PAC experiment using a rf field, \( H_r(t) \), phase-locked to the accelerator beam pulse. In a second experiment we demonstrate the occurrence of higher order effects, the Bloch-Siegert shift and subharmonic resonances in case of rf amplitudes comparable to the dc field \( H_r \) at the position of the main resonance.

The sign of the product \( g \cdot H \) is usually determined in NMR experiments by applying circularly polarized rf. Linearly polarized oscillating fields do not yield the sign. It has been shown [1], however, that one can make use of the time coherence in PAC experiments to measure the sign of \( g \) by keeping the rf phase fixed with respect to \( t = 0 \), the excitation time of the state. The rf phase provides knowledge about \( \text{sign}(H) \) and its periodic change during the decay of the state from which the sign of \( g \) can be deduced.

We have used this method to determine the sign of the \( g \)-factor of the 181 keV, 120\( \mu \)s state in \(^{78}\)Br excited and aligned by the \(^{78}\)Se(p,n) reaction with a pulsed proton beam from the Berlin Van de Graaff accelerator. The target was a molten SeTl alloy. Because the relaxation time of the excited \(^{78}\)Br nuclei in liquid SeTl was known to be 100\( \mu \)s [2], the resonances were measured in a time window from 50\( \mu \)s to 90\( \mu \)s to decrease attenuation of the resonance structure. Electronically, the phase-locking was achieved by triggering the beam pulse (width 2.5\( \mu \)s) with the zero-phase crossing of the rf in time intervals of 380\( \mu \)s.

The 180° phase was achieved by a delay which shifted the beam pulse trigger with respect to the rf. In a proper geometry the resonances were observed for the two phase angles \( \Delta = 0° \) and \( \Delta = 180° \). The result is shown in Fig. 1. The theoretical curves in Fig. 1 were calculated including the influence of the time window on the resonance structures (compare Ref. [3]). Unambiguously, the \( g \)-factor is seen to be positive: \( g = +1.025(3) \), which supports the earlier proposed nucleon configuration [4].

![Fig. 1 Sign determination with phase-locked rf at a frequency \( \nu = 21.04 \text{ kHz} \). \( k_1 \) determines the beam axis, \( k_2 \) the detector position in a coordinate system defined in Ref. [1].](image-url)
Whenever linearly polarized rf is used in NMR experiments the counterrotating component causes, in principle, a shift of the resonance \( |5| \) as well as the occurrence of subharmonic resonances \( |6| \) at frequencies \( \nu_p/(2n+1), (n=1,2,...) \) where \( \nu_p = g \cdot H_r \cdot \nu_0/h. \) As long as \( H_1 \ll H_r \) the shift is negligible and subharmonic resonances are unobservable. However, for \( H_1 \approx H_r \) the semiclassical resonance concept breaks down \( |6| \) since these higher order effects become important. Subharmonic resonance effects have been studied by optical pumping \( |7| \) where the condition \( H_1 \approx H_r \) can easily be realized. It is the purpose of this note to point out that nuclear states with lifetimes in the order of milliseconds are also very well suited to study subharmonic resonances.

The angular distribution of the \( \gamma \)-decay of the \( 9/2^+, 20 \) ns state in \( ^{71}\text{Ge} \) produced by the \( ^{71}\text{Ga(p,n)} \) reaction was perturbed in an NMR-PAC experiment with random phase conditions using the convenient experimental arrangement described e.g. in Ref. \( |4| \). The main NMR-PAC resonance and the g-factor \( |6| = 0.2307 \) of the 20 ns state were reported in Ref. \( |9| \). The resonance structures were observed with a fixed frequency \( \nu = 3.5 \) kHz by sweeping the \( H_0 \)-field over suitable ranges. The results are shown in Fig. 2. With an rf-amplitude \( H_1 = 12 \) G the main resonance at about 17 G is saturated exhibiting the splitting for \( k = 2 \) (Fig. 2a). Compatible with the theory of Capeller, the subharmonic resonance (n=1) becomes broader and the splitting more pronounced on increasing the \( H_1 \)-value from 12 G to 15 G (Fig. 2b,c). All shifts obtained are proportional to \( H_1^2 \).

This work was supported by the Bundesministerium für Forschung und Technologie.

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**Fig. 2**

Main and first subharmonic resonances of the 20 ns state in \( ^{71}\text{Ge} \). The arrows mark the position of the resonances without the Bloch-Siegert shift.

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Nuclear Quadrupole Resonance Detected by Perturbed Angular Correlations (PAC-NQR)

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Quadrupole moments (Q) provide detailed information about the nuclear charge distribution. Therefore a systematic experimental determination of Q is of great interest for nuclear models. For short-lived excited nuclear states Coulomb excitation, Mössbauer effect and perturbed angular correlations (PAC) are the techniques to measure quadrupole interactions. The application of PAC to this problem received great stimulus when it was demonstrated\(^1\) that under certain temperature conditions sharp quadrupole frequencies can be obtained with nuclear reaction-produced isomeric states in thick metallic targets. The time-differential observation of PAC has been so far the commonly used detection technique. It is the purpose of this contribution to illustrate the expected effects of nuclear quadrupole resonance (NQR) on the angular correlation of γ-rays.

Here, we shall only discuss the simplest case of a single crystal with axial symmetry, oriented in such a way that the field gradient is along the z-axis, parallel to an external static magnetic field (which can be zero). A circularly polarized rf-field is applied perpendicular to the z-axis. For this geometry the interaction Hamiltonian in the rotating frame is of the form

\[
\hat{H}_Q = \frac{1}{2} q Q I_z^2 + \omega_Q (I_+ I_- - I(I+1)) \tag{1}
\]

where \(\omega_Q = -\frac{\mu}{\hbar} H_z, \omega_1 = -\frac{\mu}{\hbar} H_z, \) and \(\omega_2 = \frac{eQ_2}{\hbar} I_z^2 - (I(I+1))\)

In the presence of quadrupole interaction it is no longer possible to define an "effective field" along which the Hamiltonian is diagonal or equivalently, diagonalization cannot be accomplished by rotating to a new z'-axis parallel to \(H_{eff}\) as in pure PAC/HKB. The time evolution of the density matrix under the influence of these fields is described by

\[
\frac{d\rho(t)}{dt} + [\hat{H}_Q, \rho(t)] = \hat{H}_Q \rho(t) \tag{2}
\]

The calculation of the perturbation factors is most conveniently done in Liouville formalism\(^2\) by taking matrix elements of the time evolution operator \(\hat{S}_Q(t) = \exp (-i\hat{H}_Q t/\hbar)\) with respect to irreducible tensor operators \(\{kq\}\) yielding:

\[
\hat{G}_{kk'}^q(t) = e^{-i\left(\omega_0 t - \omega_q t\right)} (kq) \hat{U}_Q(t) |k'q'\rangle e^{i\omega_q t} \tag{3}
\]

Restricting to random rf-phase (\(\omega_0\)), which results in \(q = q'\), the time-integrated perturbation factor is of the form

\[
\hat{G}_{kk'}^q = \int (kq)\{\hat{U}_Q(t)|k'q'\} e^{i\omega_q t} dt \tag{4}
\]

For \(\omega_0/\omega_q < 1\) the resonance positions are determined by the relation

\[
[(m-m') - q_j] \omega_{res} = (m+m')\omega_0 + (m+m')\cdot 3\omega_0 \tag{5}
\]

In the figure, only terms with \(q = 0\) are shown because of practical importance. Near resonance \(\hat{G}_{kk'}^q \ll \hat{G}_{kk'}^0\) if \(\omega_{res} \gg 1\). Terms with \(q \neq 0\) give rise to small additional resonances according to eq. (5).

The general calculation must be performed numerically, and a few typical results are shown in Figs. 1 - 3. In Figs. 1 and 2 the nature of the quadrupole resonance and how it develops from the pure magnetic case is shown for spins 1 and 5/2. Although these apply to small \(\omega_q\)-values only they provide information about the complexity of PAC-NQR spectra. To
relay to the experimenter a feeling of how much power is needed to obtain a measurable effect in a realistic case, the 9/2→7/2 transition for an I = 9/2 system is shown in Fig. 3 for various parameters $\omega_1$.

From the curves shown and from eq. (4) and (5) we notice the following points:

1. The sign degeneracy of $m$ makes the quadrupole resonance pattern to be symmetric about $\omega_0$. A magnetic field merely causes a shift of the pattern. The size of the resonances is independent of the ratio $\omega_0/\omega_1$. Hence, an externally applied magnetic field is no essential for measuring quadrupole resonances.

2. There is no multipole structure like in the pure magnetic case; the resonances exhibit Lorentzian shape according to eq. (4). The dominant and broad resonance peaks correspond to $\Delta m = \pm 1$ transitions and are anticrossings in the rotating frame level scheme. Narrower resonances which show a pronounced rf-dependence are $\Delta m = \pm 2$ transitions which come about by coherent superposition of two rf quanta of the same polarization. Their origin are $\Delta m = \pm 2$ crossings in the level diagram.

References

STROBOSCOPIC OBSERVATION OF STATIC ELECTRIC FIELD GRADIENTS

A Theoretical Study)

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The counting rate in a Perturbed Angular Distribution (PAD) experiment will increase considerably and show Lorentzian-like resonance behavior as a function of an external field, if the pulsed-beam excitation technique is applied with pulse frequency $T^{-1}$ large compared to the reciprocal lifetime $\tau^{-1}(\tau/T \ll 1)$. This fact has been utilized in many stroboscopic observations of PAD (SOPAD) to determine nuclear $g$-factors of long-lived isomeric states [1].

However, the work done so far was restricted to the case of pure nuclear Zeeman interaction. Only recently first attempts were made on $^{69}$Ge and $^{111}$Cd to detect a stroboscopic resonance in the presence of "mixed interactions" (i.e. a non-vanishing static nuclear quadrupole interaction besides the Zeeman energy) [2]. In view of the experimental difficulties a theoretical investigation of the counting rate including mixed interactions appeared most helpful in making an optimal choice for the experimental set-up. For this reason we specialized the general SOPAD result derived in [3] to the case of mixed interactions. Moreover, we restricted the calculation to the simple axially symmetric case ($n=0$) displaying the main new features without the additional complexity introduced by a non-diagonal hamiltonian. Starting from

\[ H = -g\mu \hbar I_z + \frac{eQ_V}{4I(I+1)} [3I_z^2 - I(I+1)], \]

and using formula (15) of [3] for the counting rate $Z(T;t)$, we arrive at

\[ Z(T;t) = \text{const} \frac{\exp(-t/\tau)}{1-\exp(-T/\tau)} \left[ 1 + \frac{3/4 a_1 a_2}{1+3/4 a_1 a_2} E(T;t) \right]. \]

Conventionally, $a_2$ and $t$ denote radiation and excitation parameters of multipole order 2, and $t$ is the delay time. The effect function $E(T;t)$ is given by

\[ E(T;t) = \frac{\exp(V_\perp t) - 1}{\exp(V_\perp t) - 1} \sum_{n=1}^{(2I-2)!} \frac{n(n+1)(2I-n+1)}{2I(2I-3)!} \frac{\cos(2\omega_n T + 2\omega_n T \cos(2\omega_n T + 2\omega_n T))}{\cosh(2\omega_n T) - \cos(2\omega_n T)}, \]

(4) \[ 2\omega_n T = -2\pi/h_\perp(t) - (2I-2n)2\pi/h_\perp(h_\perp(t)), \]

with the magnetic and quadrupole resonance fields:

\[ h_\perp(t) = \frac{2\pi}{3\gamma T}, \quad \text{and} \quad h_\perp(V_\perp) = \frac{3\epsilon Q_V}{2\gamma 2I(2I-3)}. \]

In (3) $\theta_0$ denotes the angle between beam axis and $\gamma$-counter and the magnetic field $h$ is assumed perpendicular to the plane spanned by the beam and the counter. Only multipole terms up to $k=2$ have been considered to be of practical importance. For $V_{zz} = 0$ formula (2) reduces to the case studied in [1]. A typical resonance line $E(T;t)$ for SOPAD in the 398 keV-level of $^{69}$Ge (I=9/2, $\tau = 4.2$ $\mu$s) is shown in fig. 1(a). As $V_{zz}$ is increased this single line broadens and finally splits up into (2I-1) constituents according to the sum in (3), fig. 1(b), 1(c). Since $\cosh(T/\tau) > 1$, the denominator in (3) reaches its minimum for $\cos(2\omega_n T) = 1$, i.e. the resonant field values are given by

\[ a_n(T;h_\perp(t)) = b_n(T;h_\perp(t)) - (2I-2n)h_\perp(h_\perp(t)), \]

with integer $k_n$. According to (6) the distance $2h_\perp(V_{zz})$ between two adjacent peaks in fig. 1(c) will be a direct measure of the quadrupole interaction strength if the $g$-factor is known. However, the size of the effect is drastically reduced (roughly speaking by a factor of (2I-1) for well separated lines) by introducing the quadrupole interaction thus lifting
the magnetic degeneracy. Moreover, the lineshape may no longer be assumed to be simply Lorentzian, since from (3) in the vicinity of resonance we have:

\[ E(T) \propto \sum_{n=1}^{2^{1/2}} n \left[ \frac{1}{2} \sin\left( \frac{3}{2} \phi \right) \right] - 2 \left[ \frac{h^2}{T} \frac{T}{h^2} \right] \sin\left( \frac{3}{2} \phi \right) \left[ \frac{1}{2} \exp\left( -\frac{1}{2} \right) + \frac{1}{2} \exp\left( +\frac{1}{2} \right) \right] \]

with the linewidth:

\[ \Gamma = \frac{\hbar / 2 \log_{10}(\tau / \gamma \mu)}{2 \mu \tau} \approx \frac{\hbar}{2 \mu \tau} \text{ if } \tau / \tau \ll 1. \]

The dispersive part in (7) cannot be made to vanish simultaneously for each \( n \) by special choice of \( t \) and \( C \), in general. According to (3) or (7) the effects can be enhanced in two ways:

(I) **SOPAD with small-spin states**

Fig. 2 shows the effect (3) for the 247 keV level of \( ^{111}\text{Cd}(I=5/2, g=0.304, \tau=121\text{ns}) \) in a \( \text{Cd-lattice} \) \( \text{(eVf=125 MHz)} \). Although the height of the peaks has increased due to \( I=5/2 \), the curves 2(a) and (b) exhibit two disadvantages: (1) broad lines due to short lifetime \( (\tau=6\text{K}) \); (2) Identification of peaks is difficult in case 2(a), where \( h_{31}(T=100\text{ns}) \approx 3h_{0}(\nu_{22}) \) leads to an overlap of several patterns of the kind shown in fig. 1(c). The latter problem is avoided in fig. 2(b) by choosing \( T \) very small leading to the expected two peaks \( E(T, t) \) is an even function of \( h \).

(II) **SOPAD with "resonance pulse frequency"**

For large enough field gradients the degeneracy of the pure Zeeman spectrum may be simulated by choosing

\[ T = T_{0} = \frac{2 \pi}{h(\nu+1)} \text{ and } t / T = T / \nu_{0} = k / (\nu+1), \]

which not only leaves the denominator in (3) independent of \( n \) but also prevents destructive interference in the numerator. Conditions (9) have been used in the curves of fig. 2(c) and 3(c). Finally, to get a maximum effect \( 0 \) was chosen according to (N integer):

\[ \phi = \frac{1}{2} \pi k_{n} + \frac{N}{2} \left( \frac{3}{2} \pi k_{n} \right) \text{ for purely Lorentzian, } \]

\[ \phi = \frac{1}{2} \pi k_{n} \text{ for dispersive} \]

Fig. 3(c) demonstrates the sensitivity of the effect to the pulse frequency.

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STROBOSCOPIC OBSERVATION OF QUADRUPOLE HYPERFINE INTERACTIONS*

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The stroboscopic observation of magnetic perturbations of excited nuclei (SOPAD) is a well established method for precision magnetic moment measurements for states with lifetimes between 1 μsec = 10 nsec,†

In these measurements the resonance behaviour of the time dependent γ-ray anisotropy for a given beam-pulse repetition time is observed by changing the external magnetic field, e.g., the Larmor frequency. An extension of this technique to quadrupole perturbations would be useful because of the increased precision and resolution as well as an improved experimental yield. To apply this method to the quadrupole hyperfine interaction, it is necessary to vary the pulsing frequency since the hyperfine electric field gradients are fixed. The resonance behaviour for the quadrupole interaction involves a superposition of harmonics of the basic quadrupole interaction frequency ω0 = (6/41511(1/2))eQ2e/H (Q nuclear quadrupole moment, eQ electric field gradient, 1/2 half integer nuclear spin).

To explore this new application, the quadrupole interaction of 69Ge(9/2+) nuclei in polycrystalline hexagonal zinc metal has been studied. A spin rotation result of ω0 = 9.96(3) MHz at a target temperature of 300(15)°C has previously been measured.‡ For the present study, the 9/2+ state was populated via the reaction 64Cu(α,n)69Ge with a 1.6 MeV α beam from the Stony Brook tandem accelerator. A pulsing system was constructed which allows a variable pulse repetition time with a pulse width of about 40 nsec. A thick enriched 66Zn target was used and maintained at about 350°C (the relative temperature accuracy was ±3°C). The 398 keV decay γ-rays were detected by two NaI scintillators placed at 0° and 90°. In order to achieve the coherent superposition of all of the harmonics, the pulsing frequency ω was varied around ωres = ω0/2 and the delayed γ-rays, electronically selected in a time region at t = π/ω of width Δt = 40 nsec, were observed.

This choice of the time-delay region with respect to the beam pulses leads to the following theoretical expression for the resonance behaviour in the intensity: H(ω) = N(0) * [1 + exp(-π/ω)] [1 + 2G22(ω)F2(cosθ)]

\[ (1) \]

In this two detector experiment, the frequency dependent term exp(-π/ω) is cancelled out by forming the intensity ratio. The present results for the ratio N(0°)/N(90°) are shown in Fig. 1. The solid line is a least squares fit to the data using Eq. 1. In the fit, the polycrystalline S2n for n = 0 to 3 were used with the higher S2n values set to 0, because the pulsed beam width and the finite time region average out these higher harmonics. The fit to the data yields a resonance at ωres = 1.565(5) MHz, which corresponds to a quadrupole frequency ω0 = 3.130(10) MHz. The fitted line width indicates a slight broadening of the natural width. The precision in this successful application of the stroboscopic technique to quadrupole interactions, suggests that it will be a valuable tool for quadrupole moment comparisons and a sensitive probe of electric-field-gradient distributions.


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Fig. 1 Stroboscopic resonance due to quadrupole interactions of 69Ge(9/2+) nuclei in polycrystalline Zn metal.
V. 5

Comparison of the Precession of Angular Correlations Produced by Magnetic Dipoles and Axially Symmetric Quadrupole Interactions

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To determine not only the magnitude but also the sign of an electric quadrupole interaction by means of perturbed angular correlations (PAC) one generally needs aligned electric field gradients (EFG), as e.g., in single crystals, and a (maybe indirect) measurement of a polarization. For a more intuitive interpretation of these effects one often can use the classical precession model for a non-spherical nucleus with spin $I$ subject to an axially symmetric EFG. In contrast to the magnetic case the classical quadrupole precession $\mathbf{q} = -\frac{\mathbf{q}}{2} \mathbf{I} \times (\mathbf{q} \cdot \mathbf{I}) \mathbf{e}_z$ depends on the projection $\mathbf{q} \cdot \mathbf{I}$ of the magnetic quantum number $I$ on the symmetry axis of the EFG ("$q$ axis") (Fig. 1). From this one expects a net effect of rotation (a superposition of rotations of different frequencies and of opposite directions) of a PAC, if the intermediate state $I$ is predominantly polarized in either one direction of the $q$ axis or if through a polarization measurement of the second transition the two subgroups of $I$ with opposite polarization and precession direction can be distinguished.

In the special case of polarization by quadrupole interaction such rotation effects are discussed and experimentally proved in Refs. 2, 3. In the present paper the quadrupole precession will be discussed in a more general context using the PAC formalism of Steffen (1971) and it will be demonstrated how the features of the classical precession model give rise to a special correlation (Fig. 1). From this one expects a rotation effect of the γ emission detected in the second transition.

According to Ref. 4 the correlation can be written

$$W(E_{I1}E_{I2}) = \sum_{k} p_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k) \left| E_{I1}E_{I2}\right|^2$$

where the nuclear orientation at the time of the γ emission is described by the statistical tensor

$$p_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k) \propto \chi_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k)$$

(1)

with

$$\chi_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k) \propto \chi_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k)$$

(2)

where in the symmetry relation the plus (minus) sign has to be taken for $I = \text{align} (\text{polar})$.

In the well-known case of a magnetic dipole precession in a homogeneous magnetic field (parallel to the quantization axis $Z$) the attenuation factors are simply phase factors. The orientation at the time $t$ of the second transition

$$p_k(E_{I1}E_{I2}; I, k) = p_k(E_{I1}E_{I2}; I) \propto \chi_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k)$$

(3)

where $p_k(E_{I1}E_{I2}; I) = 0$, $p_k(E_{I1}E_{I2}; I) = 0$, and $p_k(E_{I1}E_{I2}; I) = 0$. Therefore, if $E_{I1}E_{I2}$ both $k = 0$ and $k = \text{even}$, non-zero complex phase factors similar to Eq. 4 remain and Eq. 6 can be written (if the $p_k(E_{I1}E_{I2}; I)$ are real)

$$p_k(E_{I1}E_{I2}; I) = \sum_{m} \left| p_k(E_{I1}E_{I2}; I) \right| \propto \chi_{E_{I1}E_{I2}}(E_{I1}E_{I2}; I, k)$$

(4)

where $p_k(E_{I1}E_{I2}; I) = 0$ for $k = \text{odd}$. Therefore, if $E_{I1}E_{I2}$ both $k = 0$ and $k = \text{odd}$ cannot both be non-zero. Consistent with the classical vector model (Fig. 6) one gets no rotation terms, but nevertheless the sign of $p_k$ can be measured.

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5. H.J. Reberand and D. Reberand, Z. Physik 162, 195 (1968)
In this contributed paper we describe an investigation of the hyperfine components in ZnO by the frequency modulation of the 93.26-keV γ rays of $^{67}$Zn. Frequency modulation can be obtained by mechanically moving the source with a quartz crystal. The technique has been used before at fixed-modulation frequency as a method for calibration of velocity spectrometers for the 14.4-keV radiation from $^{57}$Fe. Here we sweep the frequency so that it becomes a direct method of spectroscopy.

If $a$, the modulation index, is defined as the ratio of the maximum deviation of gamma-ray frequency to the modulation frequency $\omega_m$, then $a = k x_{\text{max}} v_{\text{max}} / D$, where $k$ is the magnitude of the gamma-ray wave vector, $x_{\text{max}}$ is the amplitude of the motion, and $D$ the piezo-electric constant. An initial Lorentzian emission line at $\omega_0$ is modulated to become a spectrum of Lorentzian carrier plus sidebands at $\omega_0 + n \omega_m$ ($n = 0, \pm 1, \pm 2, \cdots$) with intensities $J_n(s)^2$. In the absence of mechanical resonances, $D$ is independent of frequency, and one can sweep the modulation frequency at constant voltage amplitude and hence constant sideband intensity. The voltage is chosen to obtain maximum intensity of a particular sideband $n$. By sweeping the frequency over the range $\Delta \omega_m$, the transmission through an absorber can be investigated over a range $n \Delta \omega_m$.

The method gives an absolute frequency determination of energy differences, at the expense of a dilution of intensity over unused sidebands. The expense is small, however, because the method allows one to have precise frequency offsets, so that counting is restricted to the desired portion of the spectrum, without loss of precision.

The experiments are a continuation of earlier work on the Mössbauer effect in $^{67}$Zn. They arose initially from a desire to calibrate our piezo-electric velocity spectrometers for the low velocities needed for experiments with this narrow line.

In our experiments the quartz crystal was an X-cut cylindrical disk, 3 mm thick and 4 mm in diameter; the faces were covered with conducting electrodes of silver paint. The sources, which were fastened to the quartz crystal, were $^{67}$Ga in ZnO single crystals. The $^{67}$Ga activity was produced by α or d bombardment of the ZnO crystals at the Argonne cyclotron. The ZnO crystals were cut in the form of cylindrical disks of 0.5 mm thickness and 4 mm diameter. The γ rays were observed along the cylinder axis, which was also the crystalline c axis. The absorber was polycrystalline ZnO, 2.1 g/cm$^2$ thick, enriched to 89.6% in $^{67}$Zn. The γ transition in $^{67}$Zn is $\gamma = (5/2^-)_{\text{exc}} \rightarrow (5/2^-)_{\text{gd}}$. In ZnO the ground state is split into a triplet by electric-quadrupole interaction. Figure 1 shows the transmission of the "carrier" radiation ($\omega_m$) as a function of the driving voltage at a constant modulation frequency $\omega_m$. The solid line represents a fit with a curve of the form $y = y_0 (1 - a) J_0 (b V)$, where the background $y_0$, the fractional effect $a$ and the modulation index per volt, $b$, are the free parameters. The quantity $V$ is the voltage across the quartz crystal. At $V=0$ maximum absorption is observed, since the isomer shift between the source and absorber is negligible. With increasing voltage more intensity is put into the sidebands which suffer no Mössbauer absorption. The counting rate therefore rises. Figure 2 displays a transmission spectrum where the frequency was varied between 149 kHz and 216 kHz, the modulation index being $r = 3.1$. 

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4 The natural width of the 93.26-keV level in $^{67}$Zn is $\Gamma_\gamma = 0.31 \mu$m/sec or 23.4 kHz.
5 The frequency was swept by applying a ramp voltage to a sine wave generator, whose frequency could be voltage controlled.
Fig. 1. Transmission of γ radiation as a function of the driving voltage at the quartz crystal. The modulation frequency was kept constant.

Fig. 2. Transmission as a function of the modulation frequency.

which is close to the 1st maximum of $J_2$. The dip at $180.3 \pm 1.2$ kHz corresponds to a splitting between $|m| = \frac{1}{2}$ and $|m| = \frac{1}{2}$ of $360.6 \pm 2.4$ kHz, corresponding to $4.79 \pm 0.03$ microns/sec. The line width corresponds to twice the minimum observable. We see about the same line broadening in conventional Doppler spectroscopy. Experiments to measure the larger splitting in the multiplet and thereby the asymmetry parameter are in progress.
If an ensemble of optically aligned $^{203}\text{Hg}$-nuclei is subject to the influence of static and r-f-magnetic fields the rate of $\gamma$-quanta emitted by the "rotating" nuclei in the direction $(\phi, \psi)$ and detected by a polarization insensitive counter, is given by

$$W(\phi, \psi, t) = \sum_{Kx} \rho_{Kx}(t) A_K \mathcal{D}_{Kx}^{Kx}(\phi, \psi, t)$$

where the $\rho_{Kx}(t)$ are the irreducible tensor components describing the (time dependent) orientation structure of the ensemble, and where the other symbols have the usual meaning. The values of the coefficients $\rho_{Kx}(t)$ are to be computed by a solution of the dynamic equation

$$\frac{d}{dt} \rho = - \frac{i}{\hbar} \left[ H, \rho \right] + \rho_{\text{incoherent}}$$

which in irreducible tensor representation reads

$$\frac{d}{dt} \rho_{Kx} = -i \left\{ \omega_0 \rho_{Kx} + \sum_{Kx} \left( R(K+1)-R(K-1) \rho_{Kx+1} \cos \omega t + \frac{\omega_0}{2} R(K+1)-R(K-1) \rho_{Kx-1} \cos \omega t \right) \right\} \rho_{Kx}$$

$$+ \frac{1}{\tau_{Kx}} (\rho_{Kx} - \rho_{Kx})$$

where $\omega_0 = \gamma H_0$; $\rho_{Kx} = \rho \cdot \delta_{Kx}$ for pumping by unpolarized light

$\tau_{Kx}$ = relaxation time constants

The solutions of this system of differential equations (1) show a typical resonance behaviour at $\omega = \omega_0$ (main resonance) and also at $\omega = \omega_0/3$, $\omega_0/5$, ... a.s.o. (subharmonic resonances; see later). For $\omega = \omega_0$ the static orientation of the ensemble and - as a consequence - the time integrated anisotropy of the emitted $\gamma$-radiation is diminished. Under the same conditions $1\omega$- and $2\omega$-modulations of the emitted $\gamma$-radiation are observed, the degree and the phase of the modulation being fixed as well by the frequency and the strength of the driving field as by the direction $(\phi, \psi)$ of observation (2,3).

Moreover - in contrast to the well known effects of NMR - resonance phenomena appear not only at the Larmor-frequency, but - for higher values of the strength of the driving field - also at frequencies $\omega = \omega_0/3$, $\omega_0/5$ a.s.o. The appearance of these resonances is typical of the situation, that

1) the driving field is linear polarized,
2) the longitudinal and transversal relaxation times of the spin system are of the same order of magnitude as it is fulfilled if the driven nuclear ensemble is in a gaseous state.

Measurements of the various effects are compared with the predictions of the general theory.

Measurements of the linear polarization of radiation in external magnetic fields

by R. Heusinger, W. Kreische, K. Reuter, K.H. Roth and K. Thomas
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The linear polarization of the radiation emitted from aligned excited nuclei depends on external magnetic fields. In view of the present measurements we discuss the following cases:

1. The direction of observation is perpendicular to the field direction (transverse Zeeman effect). The linear polarization of the emitted radiation is independent of the magnetic field.

2. The direction of observation is parallel to the field direction (longitudinal Zeeman effect).

\(\Delta M_e = \Gamma'\) (\(\Gamma'\) is the natural line width, \(\Delta M_e = g_e \mu_B B\) is the hyperfine splitting of the excited state).

The components with \(\Delta m = \pm 1\) are right-handed (RHC) and left-handed (LHC) circularly polarized respectively. If one cannot detect the two components separated energetically the resultant radiation is unpolarized.

\(\Delta M_e < \Gamma'\).

The RHC and LHC polarized components can interfere, resulting in a linear polarization of the emitted radiation. As the field increases and therefore the Zeeman splitting, the interference decreases and therefore the linear polarization.

The following linear polarization measurements were done with the 197-keV radiation from the second excited state \(J^e = 5/2^e, \tau = 128.8\text{ns}, g_e = 1.476\) of \(^{19}\text{F}\), populated by the reaction \(^{19}\text{F}(p, p')\) at \(E_p = 5.5\text{MeV}\).

The polarimeter used is a conventional Compton polarimeter consisting of a plastic scintillator (P1) and two NaI detectors D1 and D2. The \(\gamma\) quanta are scattered in the plastic scintillator and the scattered quanta are detected in D1 and D2 respectively in coincidence with the Compton electrons in the plastic scintillator. The ratio \(N_1/N_2\) of coincidence counting rates is a measure for the linear polarization of the \(\gamma\) radiation.

For the case that the direction of observation is perpendicular to the beam axis and coincides with the external magnetic field, the dependence of the linear polarization of the emitted radiation on the angle \(\phi\) (see Fig. 1) and the magnetic field \(B\) is given in Refs. 2,3 to

\[ p = p_0 \cos(2\phi - 2\phi_{min}) \sqrt{1 + (2\Delta M_e/\Gamma)^2} \]

with \(\Delta M_e = g_e \mu_B B\), \(\phi_{min} = \frac{\pi}{2}\) arctan \((2\Delta M_e/\Gamma)\).

The hyperfine interaction of the excited level is \(\Delta M_e\), \(\tau\) and \(g_e\) are the lifetime and the \(g\)-factor of this state, \(p_0\) is the linear polarization for zero magnetic field and \(\phi = 0^\circ\) and is defined as \(p_0 = (I_+ - I_-)/(I_+ + I_-)\).

\(I_{\pm}\) are the intensities of the radiation polarized parallel and perpendicular respectively to the reaction plane without magnetic field.

From a measurement of the angular correlation together with the measurement of the polarization distribution one gets \(p_0 = 0.188\) and the polarization sensitivity \(R\) of the polarimeter to \(R = 10\).

In Fig. 1 measurements of the ratio \(N_1/N_2\) as a function of the angle \(\phi\) with different magnetic fields are parameterized.

The angle \(\phi\) is defined in the diagram of the experimental setup.

FIG. 1. The ratio \(N_1/N_2\) measured as a function of the angle \(\phi\) with different magnetic fields as parameter.
fields as parameter are shown. The solid lines are predictions following Eq. (1). Fig. 1 shows for the case of \( \phi = 0^\circ \) an increasing value for \( N_1/N_2 \) and therefore a decreasing polarization for increasing magnetic field. As it is seen from Eq. (1) this dependence is Lorentzian shaped. With increasing magnetic field the minima of the curves are shifted. This means a precession of the plane of polarization. The orientation of the plane of polarization is given by the condition \( \frac{\partial \phi}{\partial \phi} = 0 \). Therefore from Eq. (1) for the precession angle \( \psi_{\text{rot}} \) of the plane of polarization results

\[
\psi_{\text{rot}} = \psi_{\text{min}} = \frac{1}{2} \arctan(2\pi W/\Delta)
\]  

(2)

Using a pulsed beam it is possible to observe the time differential linear polarization. The plane of polarization precesses around the direction of the magnetic field. Such investigations are in progress.

Further measurements were the direction of observation is perpendicular to the beam axis and were the beam axis coincides with magnetic field were done. The results are shown in Fig. 2. The linear polarization is independent of the magnetic field. This behaviour follows immediately since each magnetic sublevel causes a linear polarized component, the observed degree of polarization \( p_0 \) depending on the nuclear alignment.

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Brute Force Orientation of $^{111}$In in Cu

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With very few exceptions\(^1,\) the observation of low temperature thermal equilibrium nuclear orientation has involved systems in which the nuclei are subject to sufficiently strong magnetic or electrostatic hyperfine interactions to produce appreciable ordering at millidegree temperatures. This corresponds to an overall hyperfine splitting of order $10^{-10}$ erg or $20$ MHz as a lower limit. However, if the nuclear polarisation were achieved by a large external magnetic field (Brute Force Polarisation) the lower limit of detectable internal hyperfine interactions may be reduced since they now appear as perturbations upon the basic brute force effect. The increasing availability of laboratory produced fields in the range $5 - 15$ T makes such experiments attractive. This report describes exploratory experiments made with a $7$ T NbSn superconducting solenoid.

For initial investigation $^{111}$In was chosen (g factor 1.23, spin $\frac{9}{2}$ and simple, well-known, decay scheme) to give large effects in the available field. Cu was selected as a cubic, diamagnetic host in which In is soluble and in which the only internal hyperfine interaction would be a Knight shift. The In activity was diffused into a thin foil of nominally $0.99999$ pure Cu foil at $800^\circ$C cooled and etched to remove surface activity. Samples were soldered to a copper cold finger and cooled by contact with demagnetised chrome potassium alum to temperatures of order $14$ mK. Temperatures were measured with $^{60}$CoFe thermometer with due correction for applied field.

The $^{111}$In spectra were recorded in a Ge(Li) detector placed on the axis of polarisation. Fig. 1 shows the observed variation of normalised count rate for the $173$ keV (Mixed M1 $+$ E2) and $247$ keV (pure E2) transitions as a function of $\frac{H}{T}$ where $H$ is the applied field and $T$ the temperature deduced from the $^{60}$CoFe thermometer. The data are for three different samples and for fields in the range $3 - 7$ T and temperatures between $14$ and $25$ mK. For each gamma ray variation of $H$ and $T$ independently yielded a common curve indicated by the dashed lines. In fitting the $171$ keV transition data, the mixing ratio $s (\frac{E2}{M1})$ is important, and the value $0.146(3)$\(^3\) has been used. The $245$ keV angular distribution is less sensitive to this mixing ratio in the $173$ keV decay which precedes it.

For neither of the transitions is the observed anisotropy in agreement with calculations based on the known g-value and decay scheme and the measured values of $H$ and $T$ (full curve). The value of the applied field has been checked with a Hall probe magnetometer. Both transitions show the same attenuation by a constant factor for all values of $\frac{H}{T}$, within experimental error, which can be described in terms of a 'effective' of $0.82(4)$. Such a deviation cannot be explained by a variation in $s$ alone. Experiments to investigate this apparent discrepancy are continuing both by nuclear orientation and NMR/AE.

This work was supported by grants from the Science Research Council.

References
Fig. 1: \(^{111}\)In/Cu Anisotropy vs \(H/T\) for the 171 keV and 245 keV transitions. Full line is calculated for measured \(^{111}\)In g-factor. Dotted line indicates attenuation by factor 0.82.
The extraction of the relevant physical parameters from the experimental Mössbauer spectroscopic data may in some cases be quite difficult, especially when combined electric and magnetic hf interactions are present in the sample examined. This paper deals with that problem for Mössbauer spectra originating from an $I = 1/2 + I = 3/2$ transition. Analytical expressions of the energy eigenvalues, for both the excited state and the ground state of the Hamiltonian describing combined static magnetic and electric hyperfine interactions have been derived. By incorporating these analytical functions into a fitting program a practical way of analyzing Mössbauer spectra is provided.

The hf interaction Hamiltonian $H$ can, in the efg principal axis system, be expressed as:

$$H = H^e + H^m = \frac{eOV_{zz}}{4I(2I-1)} (3I^2 - I(I+1) + \eta (I^2 + I_0^2)) - g_\eta B_{hf} \frac{1}{2}(I_+ + I_-) \cos \phi \sin \theta - \frac{1}{2}(I_+ + I_-) \sin \phi \sin \theta + I_0 \cos \theta$$

The energy eigenvalues of the Hamiltonian for $I = 1/2$ are easily found to be $\pm \sqrt{\gamma}$, while the eigenvalues for $I = 3/2$ cannot be found so easily. The corresponding Hamiltonian matrix (for $I = 3/2$) can be reduced, taking out the factor $\frac{1}{2} g_\eta B_{hf}$, to a matrix involving only 4 variables; $\theta$, $\phi$, $\eta$ and a new parameter $R = \frac{eOV_{zz}}{2 g_\eta B_{hf}}$. The secular determinant equation can thereby be written:

$$\lambda^4 + p \lambda^2 + q \lambda + r = 0$$

where

$$p = -10 - 2R^2 (1 + \frac{1}{2})$$
$$q = -8R (3\cos^2 \theta - 1 + \eta \sin^2 \theta \cos 2\phi)$$
$$r = 9 + 2R^2 (6\sin^2 \phi - 5\eta^2 \cos 2\phi + 4\eta \sin^2 \phi \cos 2\phi + R^2 (1 + \frac{1}{2})^2)$$

The solution of such an equation of the fourth order, can be found in ordinary mathematical textbooks. The analytical result for $q > 0$ is:

$$\lambda_1 = \sqrt{\gamma} + \sqrt{\gamma_2 + \sqrt{\gamma_3}}$$
$$\lambda_2 = \sqrt{\gamma} - \sqrt{\gamma_2 + \sqrt{\gamma_3}}$$

while for $q < 0$ all $\lambda_i$ as given above, should be multiplied with -1.

Here $\gamma$ can be represented by the formula:

$$\gamma = \frac{1}{6} \left[ \sqrt{p^2 + 12r} \cos \left( \frac{(p^2 + 12r)}{3} \right) - p \right]$$

where

$$\cos \phi = \frac{2p^2 + 27r^2 + 72pr}{2 \sqrt{(p^2 + 12r)^3}}$$

The energy eigenvalues of the Hamiltonian are then given as the product $\lambda_i \cdot \frac{1}{2} g_\eta B_{hf}$

By making use of the analytical results given above and of derived analytical results for the corresponding eigenvectors a subroutine has been written, which calculates the lines positions and transitions probabilities for given hf parameters. This routine has been incorporated into the least squares fitting program, written by Agresti et al. [1]. This program originally used for rather special cases could easily be extended to a more general physical situation. The least squares fitting program has been tested on some relevant cases. In Fig. 1 are shown experimental $^{57}$Fe Mössbauer absorption spectra of the hexagonal and antiferromagnetic compound FeGe at four different temperatures ranging from 10 K to 390 K (Tm - 400 K). The fits yielded the results shown in Table 1. The corresponding $R$
values are 0.52(3), 0.75(3), 0.98(3) and 1.55(8) respectively. The value of the electric quadrupole interaction seems to stay nearly constant, as well as the values of \( \theta \), \( \eta \) and \( \phi \) over the temperature range investigated.

In Fig. 2 are shown \(^{119}\)Sn Mössbauer absorption spectra of the antiferromagnetic compound FeSn, isostructural with FeGe. The tin atoms populate two types of position in this crystal, a singlefold position (Sn(1)) and one twofold (Sn(2)). The iron atoms, restricted to the basal plane, are antiferromagnetically coupled in alternating basal planes, with the spin in the planes. The Sn(1) atoms are in the basal plane, while Sn(2) lies between these planes. Both tin positions experience relatively strong electric quadrupole interactions above the Neel temperature (-370 K). Below \( T_N \) the Sn(1) atoms also experience a magnetic hf interaction. The environment of the tin atoms has trigonal symmetry around the c-axis, leading to \( \eta=0 \) and \( V_{zz} \) collinear with the c-direction. Below \( T_N \), at most three different lines originating from Sn(1) atoms could be resolved. Nevertheless the least squares fitting program exhibits good convergence, giving the results presented in Table II. The corresponding R values are -4.4(1), -6.7(3) and -10.8(3). From the table it is evident that \( \theta \) is close to 90° throughout the whole antiferromagnetic region, indicating that the magnetic hf field at Sn(1) lies in the basal plane. Detailed discussions of these Mössbauer studies are presented elsewhere [2]. When either the magnetic dipole or the electric quadrupole hf interaction is dominating, approximate solutions of the eigenvalue equations have been developed. For example the first order perturbation energy \( \delta' \) on the magnetic hyperfine splitted sublevels \((I = 3/2)\), due to a small electric quadrupole hf interaction can be expressed as

\[
\delta' = (-1)\left[\frac{\eta_1}{2} + \frac{\eta_2}{2} - \frac{3}{4} \cos^2\theta \right] + \frac{V_{zz}}{2} \end{equation}

which is an extension of the formula for axially symmetric efg tensor, given by Wertheim [3]. Formulas for the second and third order perturbation energies have also been derived. For \( R > 1 \), i.e. dominating electric quadrupole hf interaction, expressions for the first and second order perturbation energies due to a small magnetic dipole hf interaction are developed and the possibility of determining the sign of \( eQV_{zz} \) will be discussed in connection to these expressions.

References

Table I.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>( B_{hf} ) (Gauss)</th>
<th>( \Delta ) (G)</th>
<th>( \delta ) (%)</th>
<th>( \eta ) (%)</th>
<th>( \phi ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>15.7(1)</td>
<td>-0.56(3)</td>
<td>0.39(1)</td>
<td>0.31(2)</td>
<td>70(25)</td>
</tr>
<tr>
<td>295</td>
<td>11.8(1)</td>
<td>-0.60(2)</td>
<td>0.28(1)</td>
<td>0.31(1)</td>
<td>70(10)</td>
</tr>
<tr>
<td>360</td>
<td>8.7(1)</td>
<td>-0.58(1)</td>
<td>0.23(1)</td>
<td>0.41(1)</td>
<td>80(15)</td>
</tr>
<tr>
<td>350</td>
<td>5.8(1)</td>
<td>-0.55(2)</td>
<td>0.22(1)</td>
<td>0.31(3)</td>
<td>70(16)</td>
</tr>
<tr>
<td>395</td>
<td>-0.60(1)</td>
<td>0.21(1)</td>
<td>-</td>
<td>-1.1(1)</td>
<td>-1.1(1)</td>
</tr>
</tbody>
</table>

Table II.

| \( Sn(1) \) | \( B_{hf} \) (Gauss) | \( \Delta \) (G) | \( \delta \) (%) | \( \eta \) (%) | \( \phi \) (%) | \( |\delta| \) (G) | \( \delta \) (G) |
|-------------|------------------|---------------|----------------|--------------|--------------|-------------|-------------|
| 10          | 15.7(1)          | -0.56(3)      | 0.39(1)        | 0.31(2)      | 70(25)       | 1.58(2)     | 1.92(3)     |
| 295         | 11.8(1)          | -0.60(2)      | 0.28(1)        | 0.31(1)      | 70(10)       | 2.13(4)     | -2.30(3)    |
| 360         | 8.7(1)           | -0.58(1)      | 0.23(1)        | 0.41(1)      | 80(15)       | 3.05(4)     | -2.90(6)    |
| 350         | 5.8(1)           | -0.55(2)      | 0.22(1)        | 0.31(3)      | 70(16)       | 4.08(3)     | 2.08(3)     |
| 395         | -0.60(1)         | 0.21(1)       | -              | -1.1(1)      | -1.1(1)      | 5.08(1)     | 1.85(1)     |
by F. GRANDJEAN and A. GERARD
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We have shown previously the existence of an electronic hopping process in ilvaite (CaFe$_2^+$Fe$^{3+}$[Si$_2$O$_5$]OH) between 300°K and 500°K.

A spectrum with a very complicated hyperfine structure is observed at 4.2°K, by Mössbauer spectroscopy. This structure appears to result from the superposition of at least three six-lines Zeeman patterns, but we have been unable to find any satisfactory combination of 18 lines. Thus, we think that the analysis should involve a larger number of lines. The internal magnetic field deduced from the splitting between the external lines is 500 kOe. At 77°K, the spectrum is similar but the lines are broader, the internal magnetic field amounting to 400 kOe.

Between 90°K and 115°K, an overlapping of the magnetic spectrum with a field identical to the one at 77°K and of the paramagnetic spectrum is observed. The transition is of first-order type, the magnetic state appears to be antiferromagnetic, it is a Mott transition.

The Néel temperature distribution observed may be ascribed to the small variations in Mn content (1) which exist between the mineral grains. This distribution can explain the reduced broadening of the lines observed between 300°K and 400°K.

The variation from 2.3 mm/s to 1.32 mm/s of the quadrupole splitting of one of the iron ions between 200°K and 500°K remains to be explained. We have simulated spectra in a fluctuating electric field gradient on the basis of Tjon and Blume (2) model. A time fluctuating term is introduced in the Hamiltonian of the system:

\[
\frac{1}{2} e^2 Qa \left( 3I_I^2 - I(I+I) \right) f(t) + \delta_a f(t)
\]

where \( \delta_a \) is the 3d$^6$ electron of Fe$^{2+}$ ion contribution to the isomer shift,
\( Q_a \) is the 3d$^6$ electron of Fe$^{2+}$ ion contribution to the electric field gradient,

and \( f(t) \) is a random function of time taking values 0 and 1.

The line shape is given by

\[
R(k) = \frac{2p + 1(\delta_a + \delta^*) + 2W_0 + 2W_1}{p^2 + p(W_0 + W_1) + 1(\delta_a + \delta^*)(p + W_1)}
\]

with \( p = 1 \) (\( \omega - \omega_0 \) - \( \omega_0 \) = 14.4 keV)

\( \omega_0 = E_1 - E_0 = 14.4 \) keV

\( \tau = \) unperturbed linewidth

\( \delta_a = \frac{1}{2} (3m_1^2 - 15) e^2 Q_a \)

\( \delta^* = \frac{1}{2} (3m_1^2 - 15) e^2 Q_a \)

\( W_0 = \) projection of the \( E_1 \) state spin

\( W_1 = \) projection of the quantization axis.

\( W_0 \) and \( W_1 \), the probabilities of hopping from state 0 to state 1 and inversely are taken proportional to the equilibrium populations of the system.

\[
W_0 = \frac{\exp(-u)}{1 + \exp(-u)} \quad W_1 = \frac{C}{1 + \exp(-u)}
\]

\( u \) being connected to \( T \) by the relation \( u = \frac{\Delta E}{kT} \), \( \Delta E \) = activation energy.

Attempts with \( C \) kept constant between \( 10^{-4} \) \( \tau \) and \( 10^2 \tau \) have shown no changes in the spectra.

* Chargé de Recherches du F.N.R.S.
At low temperature, the quadrupole coupling of the 1 state is observed as $\pm 2.3$ mm/s; at high temperature, the mean value of quadrupole couplings of 0 and 1 state is $\pm 1.32$ mm/s. As the quadrupole couplings are not known in the magnetic state it is impossible to determine their signs. If they are of opposite signs, the simple consideration of relative positions of the lines shows that the line at 0.0 mm/s must be shifted and thus, in the complete spectrum, the 3 components of the central line would be resolved. We conclude that the quadrupole couplings have the same sign.

Spectra have been simulated for different values of $u$ (or $T$) with $C=10^{-3}$; $\delta_B=0.34$ mm/s, $\delta_a=1.96$ mm/s, $\delta_B=-0.05$ mm/s, $\delta_a=1.08$ mm/s. The variation of the quadrupole coupling between 2.3 mm/s and 1.32 mm/s as a function of temperature is thus obtained, but with the value of 0.08 eV measured by conductivity for the activation energy, the temperature range where the phenomenon takes place, is too wide, and in order to account for the observed results, we are led to an activation energy as low as 0.005 eV and to more complex relationships between $u$ and $T$ such as:

$$u = \alpha E/k(T-250).$$

This is explained by the fact that the process does not start at 0°K.

We remain however unable to understand the values of the quadrupole couplings of 3d$^5$ and 3d$^6$ electrons. Of course, a 3d$^6$ contribution of order of 3.6 mm/s is expected in the octahedral symmetry as deduced from X-rays spectra.

Only a more complicated model supposing a partial delocalization of 3d$^6$ electrons from 115°K can explain a decrease of quadrupole couplings.

Under these conditions, we find for Ilvalte a behaviour similar to that of Fe$_3$O$_4$: at low temperature up to 115°K, localized electrons, from 115°K up to room-temperature, partially delocalized electrons; above room temperature, delocalized electrons.

Attempts by infra-red spectroscopy and thermogravimetry to check the existence of OH in the formula do not bring about any conclusion, either positive or negative.

Moreover, a system closer to Fe$_3$O$_4$ where electron hoppings exist and where magnetic order takes place only below 100°K, has been studied: solid solutions of spinels $\text{Zn}_{1-x}\text{Ge}_x[\text{Fe}^{2+}_{x}\text{Fe}^{3+}_{1-x}]_2\text{O}_4$.

In Mössbauer spectra, a very broad line is observed between the 2+ and 3+ lines and in infra-red spectra, an absorption band reveals the existence of a conduction band as in Fe$_3$O$_4$.

In conclusion, the Ilvalte presents a Mott transition. Moreover, the electron hopping processes in systems containing Fe$^{2+}$ and Fe$^{3+}$ ions seem to correspond to very similar level schemes despite the difference in crystallographic structures between Ilvalte and the spinels.


Among the macromolecules involved in biological processes, proteins play a dominant role for the function of living cells. The basic elements of these molecules are some 20 different amino acids, which in different combinations can be found in chains linked together by covalent bonds, thus forming molecules with molecular weight ranging from a few thousands to several millions. These chains fold by hydrogen bonding, and for many proteins the folding results in a compact spherical-like structure with the exception of a cavity structured active site, where the protein performs its function. For the subgroup metalloproteins, a metal atom covalently bound to the active site is required for functioning. This means that the hyperfine interaction between the metal nucleus and the protein surroundings could give valuable information about the active site. Such studies have been performed on metalloproteins containing iron by the Mössbauer method\(^1\). In the case of DPAC, the metal site of Zn metalloproteins could be conveniently studied by DPAC if one replaces Zn with the \(^{111}\text{Cd}\) 49 min isomer.

The advantages of the DPAC method is that it is applicable to proteins in solution as well as proteins in crystallized form. Concerning proteins in solution, one has to consider the influence from rotational diffusion on the angular correlation. The relaxation time resulting from this process can for spherical molecules in the Stokes Einstein model be written

\[
\tau_R = \frac{4\pi a^3}{3kT}
\]

where \(a\) is the radius of the protein and \(n\) the viscosity of the solvent. The important parameter for the influence of relaxation on the angular correlation is \(J^0\tau_R\), where \(J^0 = \frac{6\pi}{\sqrt{11}}\) (I half integer), assuming no magnetic interaction. A stochastic model for calculating the influence of rotational diffusion for all correlation times on angular correlation has been developed\(^2\), and from these calculations it follows that in order to deduce a static quadrupole interaction from a DPAC measurement one must have \(\omega_0\tau_R > 4\). If the condition \((\omega_0\tau_R)^{1/2} > 1\) is satisfied one can derive a simple formula for the influence of rotational diffusion on the perturbation factor which in the case of spherical molecules gives\(^3\)

\[
G_X(t) = e^{-t/\tau_K} \left( \frac{\tau_K}{\tau_K} + 1 \right)
\]

Aside from rotational diffusion there might be other local relaxation processes present. It is frequently occurring that proteins in solution might be simultaneously present in two or several forms. This effect might for example come from a proton exchange close to the metal atom. The time spectrum from a DPAC measurement should be sensitive to such processes if the corresponding relaxation time is in the region where \(10^{-1} < \omega_0\tau_R < 10\). The assumption that the process can be described with a transition between two or several quadrupole couplings would then make it possible to use the stochastic model in order to estimate the effect for different \(\tau_R\) on the angular correlation.

The possibility of relating a quadrupole coupling to the local structure of protein would in the case of a covalent metal-protein complex require the calculation of the field gradient tensor. An exact calculation is, however, extremely difficult and instead one can use a simple point model which gives

\[
\nu^2 = \sum_{i} \frac{\left( \theta_i, \phi_i \right)}{r_i^4}
\]

where \(\theta_i, \phi_i\) and \(r_i\) are the polar coordinates for the \(i\)th ligand. The absolute magnitude of \(A\) is then assumed to account for the redistribution of the atomic metal orbital and the finite size of the ligand orbitals. Such a model should be especially suitable in the case of Cd where the atom itself possesses no atomic field gradient. Thus normalizing all \(\nu_i^2\) to one of the components would leave four independent parameters due to the Poisson equation. In order to determine all four parameters one has to measure on a single crystal. In the case of a solution of proteins oriented by for example a strong electric field, only three of the four parameters can be determined, because of the random orientation of the proteins perpendicular to the electric field direction.

DPAC experiments have been performed on the enzyme carbonic anhydrase, which catalyzes the reaction 

\[
\text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{H}^+ + \text{HCO}_3^-
\]

Even though this enzyme has been well studied with many different methods, it has not been possible to determine in which way the enzyme cata-

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**V. 12**

Application of DPAC to the Study of Structure and Function for Proteins

by Roger Bauer and Peter Limkilde, The Niels Bohr Institute, Copenhagen, Denmark

Among the macromolecules involved in biological processes, proteins play a dominant role for the function of living cells. The basic elements of these molecules are some 20 different amino acids, which in different combinations can be found in chains linked together by covalent bonds, thus forming molecules with molecular weight ranging from a few thousands to several millions. These chains fold by hydrogen bonding, and for many proteins the folding results in a compact spherical-like structure with the exception of a cavity structured active site, where the protein performs its function. For the subgroup metalloproteins, a metal atom covalently bound to the active site is required for functioning. This means that the hyperfine interaction between the metal nucleus and the protein surroundings could give valuable information about the active site. Such studies have been performed on metalloproteins containing iron by the Mössbauer method\(^1\). In the case of DPAC, the metal site of Zn metalloproteins could be conveniently studied by DPAC if one replaces Zn with the \(^{111}\text{Cd}\) 49 min isomer.

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DPAC experiments have been performed on the enzyme carbonic anhydrase, which catalyzes the reaction \(\text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{H}^+ + \text{HCO}_3^-\). Even though this enzyme has been well studied with many different methods, it has not been possible to determine in which way the enzyme cata-

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alyzes the above mentioned reaction. The enzyme contains 1 atom Zn per molecule, which is situated in the bottom of the active site cavity\(^4\). The DPAC measurements have all been performed with the \(^{111}\text{Cd} 49\text{ min isomer substituted for Zn. As the enzyme has a molecular radius of about 22Å, and the frequency is of the order of 100 MHz which can be seen in the table below, one has according to eq.(1) that } \omega_0 T_R = 1 \text{ for the enzyme dissolved in water. It is therefore necessary to increase the viscosity of the solution in order to determine a static quadrupole coupling. This is confirmed by the measurement on carbonic anhydrase in a water solution}\(^5\). For this purpose we have added different amounts of sucrose such that the correlation time resulting from eq.(1) is increased approximately a factor of 10. Two measurements of his kind have been performed on human carbonic anhydrase B form, and the conditions and results are listed below.

\[
\begin{array}{l|l|l}
\text{pH} & \text{Sucrose} & \text{Correlation Time} \left(\tau_R\right) \\
6.5 & 12\% & 6 \times 10^{-8} - 3 \times 10^{-8} \\
6.5 & 46\% & 7 \times 10^{-8} - 5 \times 10^{-7}
\end{array}
\]

\(1\) To \(22^\circ\text{C}\) \(\omega_0 = 84 \pm 9 \text{ MHz}\)

\(1\) To \(22^\circ\text{C}\) \(\omega_0 = 108 \pm 12 \text{ MHz}\)

\(1\) To \(0^\circ\text{C}\) \(\omega_0 = 135 \pm 10 \text{ MHz}\)

\(1\) To \(0^\circ\text{C}\) \(n_1 = 0.6 \pm 0.2\)

\(1\) To \(0^\circ\text{C}\) \(n_2 = 0.4 \pm 0.2\)

\(1\) To \(0^\circ\text{C}\) \(\tau_R = 6 \times 10^{-8} - 3 \times 10^{-8}\)

\(1\) To \(0^\circ\text{C}\) \(\tau_R = 6 \times 10^{-8} - 3 \times 10^{-8}\)

\(1\) To \(0^\circ\text{C}\) \(\tau_R = 6 \times 10^{-8} - 3 \times 10^{-8}\)

The data for \(22^\circ\text{C}\) are taken from ref.\(^6\) and the experimental data for \(0^\circ\text{C}\) are shown on fig.1. In the analysis of the data eq.(2) has been used. The most striking difference is the appearance of two different frequencies at \(22^\circ\text{C}\) whereas only one is seen at \(0^\circ\text{C}\). In order to check that Cd binds uniquely to the Zn site, we have measured DPAC on the enzyme without removing the Zn atom, keeping all other conditions the same as in the measurement presented in ref.\(^6\). The result, which can be seen on fig.2, gives that the degree of Cd binding to the Zn containing enzyme must be less than 15%. The two frequencies then seem to result from an equilibrium between two forms of the enzyme. The spectrum taken at \(0^\circ\text{C}\) shows a frequency different from both of the two observed at \(22^\circ\text{C}\). A possible explanation would be the stabilisation of the enzyme in a new conformational state at low temperature. The spectrum might, however, also be interpreted as a damped average frequency, which could be expected if \(\omega_0 T_R\) is below 1. This means that the enzyme could still exist in two forms, but now with the relaxation time for the equilibrium decreased.

DPAC measurement on the 247 keV state in \(^{111}\text{Cd}\) seems then to be a promising method for studying active site in metallo-proteins such as Zn proteins. The method should be able to give information about chemical equilibria, dynamical processes and metal-protein coordination, thereby providing new means of studying structure-function problems for proteins.

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Mossbauer measurements have been carried out in the glass system \( x\text{Na}_2\text{O.6Fe}_2\text{O}_3.(94-x)\text{B}_2\text{O}_3 \) (mole per cent) with \( x \) varying between 10 and 35 to study the boron anomaly. A sharp decrease of isomer shift has been observed in the composition range between 20 and 25 mole per cent \( \text{Na}_2\text{O} \). The results are summarised in Table 1. This behaviour is explained on the basis of the existence of non-bridging oxygen atoms in glasses containing alkali ions beyond the above concentration.

**Table 1**

Mossbauer Results for Glasses in the System \( \text{Na}_2\text{O.6Fe}_2\text{O}_3.(94-x)\text{B}_2\text{O}_3 \) (Mole percent)

<table>
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<tr>
<th>( x )</th>
<th>Isomer Shift (Cu) (mm/sec)</th>
<th>Quadrupole Split (mm/sec)</th>
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<td>0.957</td>
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<tr>
<td>30</td>
<td>0.012</td>
<td>0.948</td>
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<tr>
<td>35</td>
<td>0.012</td>
<td>0.960</td>
<td>0.839</td>
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[The maximum error in the velocity scale is ±0.02 mm/sec]
Investigations with high-resolution Mössbauer nuclides have traditionally been the subject of high interest in Mössbauer spectroscopy. In this respect, considerable progress has been made in the well-known cases of $^{113}$Sn and $^{57}$Fe in recent years. We report here the successful observation of the Mössbauer effect in a new case: the 13.3 keV transition in $^{73}$Ge. This transition, with a mean life of $\tau = 4.3$ usec, has long been considered very attractive because of the extreme sharpness of the resonance ($E/\Gamma _{0} < 10^{-14}$), the conveniently long lifetime of the parent nuclide $^{73}$As ($T_{1/2} = 80$ days) and the low energy of the $\gamma$-ray which assures a high probability for recoilless transitions even at room temperature. The present results have been obtained in conventional transmission experiments at room temperature.

The nuclear spectroscopic data on the decay of $^{73}$As $\rightarrow$ $^{73}$Ge are summarized in the decay scheme in Fig. 1. The principal difficulties in the observation of this resonance arise from a) the high total internal conversion coefficient of this transition ($\Gamma = 1100$), b) the close proximity of its energy to the K-absorption edge of Ge (11.1 keV) and c) the low natural abundance of $^{73}$Ge (77%). These aspects impose severe demands on the strength, radioactive purity and thickness of the source, the thickness and enrichment of the absorber and the energy resolution and the ability to handle high total counting rates of the detection system used to observe the 13.3 keV $\gamma$-ray. The extreme sharpness of the resonance enhances these demands by requiring materials of high crystal quality for the source and absorber.

Mössbauer sources of $^{73}$As have been prepared by two different procedures: In the first, the $^{73}$As activity was produced by the reaction $^{74}$Ge(p, 2n) $^{73}$As using powder targets of enriched $^{74}$Ge. After suitable aging to reduce interference from $^{75}$As contaminants, the $^{73}$As was sublimated on to a 10 $\mu$m thick disk of high purity natural Ge single crystal, and diffused into it by annealing in H$_2$ at $\approx 850^\circ$C. In the second method, a 20 $\mu$m single crystal layer of enriched $^{74}$Ge was grown epitaxially on a Si single crystal substrate and irradiated to produce $^{73}$As in situ. The irradiated target was aged and annealed for 1 hr. at $\approx 500^\circ$C and used as a Mössbauer source. The direct irradiation method resulted in a significant improvement in the Mössbauer source strength as compared to the diffusion method. The absorber was prepared by epitaxially growing a 20 $\mu$m thick single crystal layer of enriched $^{73}$Ge on a Si crystal substrate.

The quality of the detection system is shown in Fig. 1 which depicts the energy spectrum in the Mössbauer geometry with the absorber in place. The mean signal to noise ratio in the window on the 13.3 keV photopeak was 1.0 with the diffused source. The S/N ratio was reduced to 0.7 for the irradiated source because this source was made with a larger thickness. Velocity spectra obtained so far are displayed in Fig. 2. These were recorded with a conventional electromechanical drive system operated in the velocity range at $\pm$88 $\mu$m/sec. Both the upper curve ("diffused" $^{73}$As source) and the lower curve ("irradiated" $^{73}$As source) were measured using the same enriched $^{73}$Ge single crystal absorber. Least squares fits of Lorentzians to these data yield, within errors, identical results for both sources; depth 2% width $2\Gamma = 47(7)$ $\mu$m/sec and center shift $+7(2)$ $\mu$m/sec, after corrections for background, experimental geometry and finite thickness of the absorber. The width observed is $\approx 7$ times the natural linewidth $2\Gamma _{0} = 6.85$ $\mu$m/sec. Despite the broadening, however, this represents the narrowest Mössbauer resonance observed so far at room temperature.

The nearly identical results obtained with the two different types of sources indicate that the major component of the broadening and the center shift could probably be attributed to the $^{73}$Ge single crystal absorber, from inhomogeneous strain introduced during the epitaxy. Experiments are currently in progress with carefully prepared $^{73}$Ge powder absorbers in an effort to reduce the observed line width of the resonance.

Fig. 1 Decay scheme and γ-ray energy spectrum of 75As source in Mössbauer geometry, taken with a Si(Li) detector.

Fig. 2 Mössbauer velocity spectra of the 13.3 keV transition in 73Ge. Upper curve: "diffused" 75As source; lower curve: "irradiated" 73As source. Common absorber: single crystal (10 mg/cm²) 75Ge.
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