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# DEFECT CHARACTERIZATION WITH POSITRON ANNIHILATION*

MASTER

L. Granatelli and K. G. Lynn
Brookhaven National Laboratory
Upton, New York 11973



It has been sixteen years since the positron annihilation technique was first used to study crystal lattice defects in deformed nickel (1). Since that time it has been applied to the investigation of defect mechanisms in most pure metals and several alloy systems. This review will deal only with positron annihilation in metal crystals, although the technique has also been applied to such diverse materials as ionic solids and polymers. The positron annihilation technique has matured to the point where, for example, it is now the preferred method for measuring vacancy formation enthalpies. In addition, it has been quite successful in the study of radiation damage and has been shown to be sensitive to void formation and growth up to diameters where they first become detectable with electron microscopy. To date, positron annihilation has had its greatest success with more complex defects such as dislocations and grain boundaries when it has been used as a compliment to the more traditional methods of studying defects. Positron annihilation is just beginning to be used to study fatigue in metals. Until recently, the high kinetic energy of positrons obtained from radioactive sources has prevented positrons from being used as a surface probe. However, with the development of an energy tunable monoenergetic positron source positron annihilation can now be applied to the study of surfaces and their associated defects.





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## 1. Introduction

In section 1 we present a brief introduction to the positron annihilation technique. In sections 2-5 the ability of the positron technique to perform microstructural characterization of four types of lattice defects is discussed. It is frequently not possible to obtain samples which contain only one type of defect in non-negligible concentrations. Such situations exist for some alloys (section 6) and for fatigued metal samples (section 7). In the final section (8) the current limitations and some future prospects of the technique will be presented.

The behavior of thermalized positrons in most metals is affected by the presence of vacancy-like defects in the crystal lattice. The positron annihilation technique is non-destructive and the positron is very sensitive to local changes in both electron density and momentum produced by these defects. In addition, relatively small samples may be used and the near surface or interior of the sample may be probed by proper choice and configuration of the positron source. For these reasons the positron annihilation technique can be the preferred method for studying crystal lattice defects in certain instances.

Positrons may be produced either in or at the surface of a specimen by the radioactive decay of a positron emitting isotope. Depending upon the isotope used positrons can enter the sample with considerable energy: up to 1.90 MeV with $^{68}Ge$. When positrons are injected into a metal sample they are rapidly thermalized (relative to their lifetime) by interactions initially with core electrons and later with both conduction electrons and phonons (2). Thermalization times are on the order of a picosecond (3,4). Once thermalized the positron will annihilate with an electron. In 99.7% of all annihilation events in metals the resulting energy is divided between two oppositely directed gamma rays each of approximately 0.511 MeV ($= m_0c^2$). The remaining 0.3% consist mainly of three quanta events which are not generally considered in lattice defect studies.

The exclusion principle allows only two conduction electrons (spin 1/2) to exist in the lowest momentum state of a crystal. The remaining electrons must, pairwise, occupy progressively higher levels. In a positron annihilation experiment only one positron exists in a sample at any moment. However, there are a large number of conduction electrons. Thus, since the positron is thermalized prior to annihilation, essentially all momentum effects on the annihilation photons can be considered to be the result of the momentum of the electron. The total energy released during annihilation is derived from the rest mass of the two particles, their mutual binding energy, the binding energy to the solid, and their kinetic energy at annihilation. Assuming that the positron is at rest and that the velocity of the electron is much less than the speed of light, c, then the energies of the two annihilation photons are approximately

$$E_{1,2} = m_0c^2 \pm \frac{cp_x}{2} \qquad (1)$$

where $p_x/2$ is the component of momentum of the center of mass of the two particle system which is parallel to the emission direction. Emission of the antiparallel gamma photons in all directions is equally probable. Thus, the energy distribution of the photons resulting from annihilation with electrons having momentum p is a continuous one symmetric* about $m_0c^2$ and extending from $m_0c^2$ - cp/2 to $m_0c^2$ + cp/2.

If the positron-electron pair is at rest during annihilation the two gamma photons are emitted in diametrically opposite directions. However, if only the positron is at rest then the annihilation photons will be emitted at an angle which differs from $\pi$ by the value $\theta$. For small values of $\theta$

$$\theta\ (\mathrm{rad}) \simeq \frac{p_z}{m_0c} \qquad (2)$$

where $p_z$ is the component of electron momentum perpendicular to the direction of the emitted gamma photon.

A metallic solid is frequently depicted as a periodic three dimensional array of positively charged ion cores embedded in an electron gas. The electron gas is composed of free valence electrons with an approximately parabolic energy distribution. At absolute zero temperature this ranges from the energy at the bottom of the conduction band up to the Fermi energy. The Fermi energy for many metals has a value between 4 and 10eV. In general the core electrons have higher momentum states than do the conduction electrons. Their momentum distribution is commonly approximated as a Gaussian but can be calculated in a straightforward manner (5).

In simple metals the positron annihilation parameters are similar to those that would be expected to result from annihilations in a homogeneous

*The energy distribution is not exactly symmetric about $m_0c^2$ because of the binding energy of the electron and positron to the solid. However, this is a small effect.

electron gas whose density corresponds to the conduction electron density in the metal. However, in all real metals the positron wavefunction overlaps with that of the core electrons. This results in a greater annihilation rate than would be expected from the conduction electrons alone (6). The ratio of the number of core to valence annihilations is affected by the degree of lattice perfection. This ratio is however, characteristic of each metal when it is free from defects. Due to Coulombic repulsion the probability of a positron entering the core region is low but the annihilation rate there is high. Conversely, the annihilation rate with conduction electrons is low but the probability of annihilation is high. The variability of the positron lifetime in the perfect lattice among different metals is a function of these two competing factors (7,8). The lifetime of positrons in the nondefective lattice is characteristic of each particular metal. Positron lifetimes in the perfect metal lattice range from about 100 to 400 ps (Fig. 1).

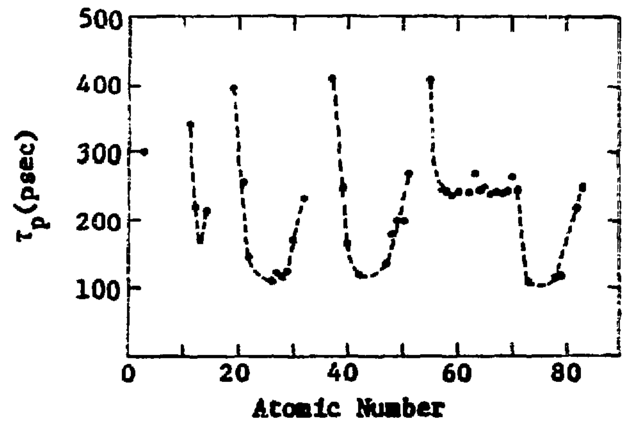



Fig. 1 - Variation of the mean positron lifetime with atomic number in the well annealed bulk lattice (8).

Positron trapping at a defect will occur when the sum of the potential and kinetic energy of the positron can be lowered by localization of the positron at that defect. The repulsive coulombic potential which a positron experiences while in a metal lattice obviously decreases with increasing distance from the positive ion cores. Open volume defects allow the positron to increase its mean distance from these positive ions (i.e. the positron wavefunction may spread out in the vicinity of these defects). This lowers the zero point energy relative to that in the perfect lattice. Even though the electron density is lower at an open volume defect than in the interstitial regions of the lattice these effects are sufficient to produce positron traps at several types of de[illegible] in many m[illegible]. Positron trapping has been reported at vacancies, voids, grain boundaries, and edge dislocations.

Due to their increased distance from the host atoms positrons which become localized at open volume defects have a reduced probability of annihilation with core electrons. The annihilation rate with conduction electrons is also reduced but the fractional reduction is less than for core electrons. Also, because the potential energy of an electron is greater in a vacancy-like region than in the interstitial region and since its total energy remains constant the momentum of the conduction electrons is reduced. These effects lead to a narrowing of the angular correlation and Doppler-broadening spectra resulting from annihilations with electrons at defects relative to that resulting from annihilations in the interstitial regions. In addition, since these positrons experience a reduced electron density their mean lifetime is increased relative to the mean lifetime of those positrons which annihilate in the non-defective lattice.

Three experimental methods are in common use for study of positron annihilations in the bulk solid. The Doppler-broadening technique, as the name implies, measures the Doppler shift of the annihilation photons. The angular correlation technique measures the angular distribution of the photons resulting from 2-$\gamma$ annihilations. The lifetime technique measures the mean lifetimes of positrons in a sample.

Energy spectroscopy measurements for Doppler-broadening experiments are performed with a Ge(Li) or intrinsic Ge solid state detector. The resulting data are stored in a multichannel analyzer in terms of counts as a function of photon energy (Fig. 2). It can be determined from equation (1) that an annihilation with a 10 eV electron results in a maximum Doppler shift of the resulting $\gamma$-photon of approximately 1.5 keV. The typical resolution of a good solid state detector is about 1.1 keV at 511 keV.

Doppler-broadening data are frequently characterized with a lineshape parameter. The most commonly used lineshape parameter, usually designated "S" (9), is defined as the ratio of the number of counts in an arbitrary symmetric central region of the photopeak to the total number of counts in the peak. Thus, the S parameter is dominated by the fraction of annihilations that take place with conduction electrons. An increase in the proportion of positrons which annihilate at vacancy-type defects results in an increase in the value of the S parameter.

Angular correlation experiments are usually performed with a two photon coincidence and narrow slit arrangement. The detectors are frequently NaI(Tl) scintillators optically coupled to photomultiplier tubes. Since

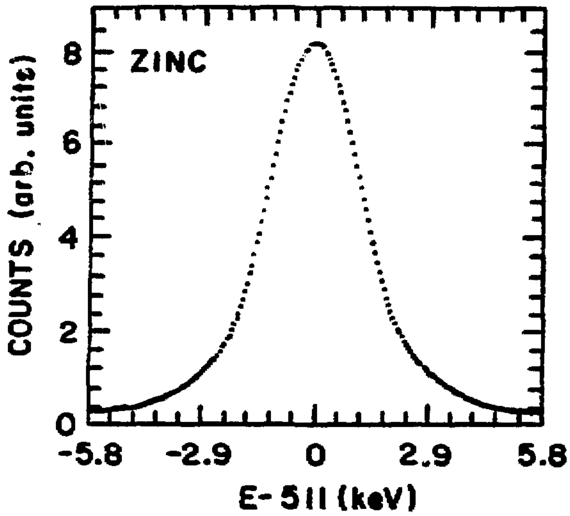



Fig. 2 - Doppler-broadening spectrum from a well annealed 99.999% Zn sample.

more than 95% of the 2-$\gamma$ annihilation photons fall within $\theta \lesssim \pm 10$ mrad (Fig. 3) the sweep angle of the detectors need not be large but high angular precision is required. Angular correlation techniques restrict data collection to one momentum range at a time (except with multiple detector (10,11) apparatus). Since it is time consuming to repeatedly scan the entire angular range, angular correlation data are sometimes collected only at one angle. Data may then be analyzed in terms of a counting rate (usually at $\theta = 0$) as a function of a variable parameter (e.g. sample temperature).

Even though both Doppler-broadening and angular correlation methods provide the same information about the electron momentum distribution there are circumstances which may make one method preferable to the other. Energy spectroscopy with a solid state detector can yield poorer equivalent momentum resolution than the angular correlation method (typically by a factor of about 10), however, in addition to a lower background, energy spectroscopy has the advantages that a smaller positron source can be used ($\sim 10 \mu Ci$ vs. 10 mCi or more), and that data collection is much more rapid.

Positron lifetime measurements are performed using a radioactive source which emits a positron with a simultaneous gamma ray (e.g. $^{22}Na$). The prompt and annihilation gamma photons are detected with scintillation counters. The time between thermalization and annihilation of the individual positron is then approximated as the time between the detection of the prompt gamma photon and the subsequent detection of one of its annihilation photons. Since positron annihilation is a statistical process the values and corresponding intensities of one or more positron lifetimes are usually

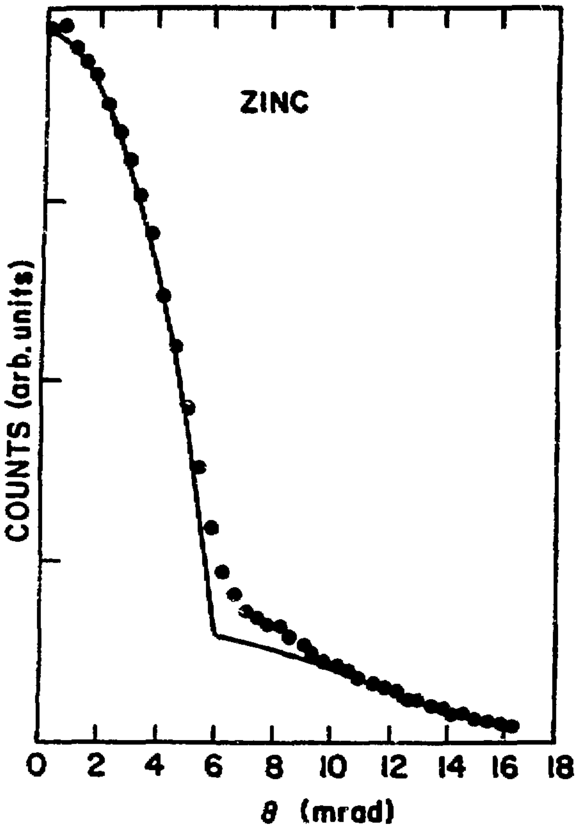



Fig. 3 - Angular correlation distribution for polycrystalline Zn [(12) and references therein].

extracted from lifetime spectra (Fig. 4) with the aid of computer programs such as Positronfit Extended (13). These programs are able to extract this information by fitting positron decay spectra to a resolution function and the sum of an appropriate number of decay terms.

Further details regarding all three techniques can be found in a number of places, including the review articles of West (12) and Doyama (14).

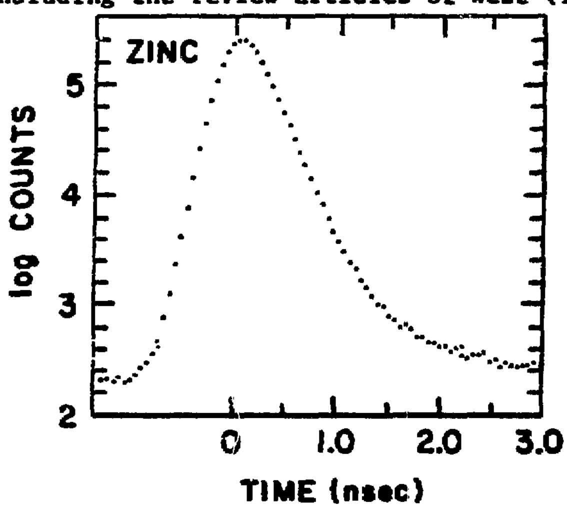



Fig. 4 - Positron lifetime spectra for well annealed 99.999% zinc single crystal

Both qualitative and quantitative information regarding the nature of defects in a sample can be obtained by measuring the angular variation of the annihilation photons, the Doppler-broadening of the annihilation photopeak and the positron lifetimes in the sample. In certain instances the same information can be deduced from all three techniques (for example vacancy formation enthalpies - see section 2). However, it should be kept in mind that to compare lifetime to either Doppler-broadening or angular correlation measurements is to compare two different physical measurements. The positron lifetime is a measure of the expectation value of the electron density of all momenta over the extent of the positron wavefunction (Bloch like in the perfect lattice). Both Doppler-broadening and angular correlation measurements, when analyzed in terms of some lineshape parameter or peak counting rate, are a measure of the momentum profile of electrons in a narrowly restricted momentum range.

Table I. Nuclear Characteristics of Some Positron Sources

| Isotope | E max (keV) | Percentage of decays giving a positron | Gamma Rays: Energy (MeV) | Gamma Rays: Percentage Coincident with Positron | Production Process | Half Life |
|---|---|---|---|---|---|---|
| $^{22}Na$ | 545 | 90 | 1.274 | 90 | $^{24}Mg(d,\alpha)$ | 2.6y |
| $^{64}Cu$ | 656 | 19 | 1.34 | 0 | $^{63}Cu(n,\gamma)$ | 12.8h |
| $^{58}Co$ | 474 | 15 | 0.810 | 15 | $^{55}Mg(\alpha,n)$ | 71 d |
| $^{68}Ge$ | 1900 | 87 | 1.078 | 1.5 | $^{66}Zn(\alpha,2n)$ | 270 d |
| $^{44}Ti$ | 1470 | 94 | 1.156 | 95 | $^{45}Sc(p,2n)$ | 48 y |

These characteristics indicate that both $^{64}Cu$ and $^{68}Ge$ are useless for positron lifetime measurements which require nuclear gamma rays in coincidence with emergent positrons since no signal is available to indicate the birth of a positron from these sources. However, $^{68}Ge$ is an excellent positron source for recent $\beta^+$-$\gamma$ coincidence lifetime techniques (26) which use the positron itself to produce the start signal. Its small nuclear gamma intensity helps keep the background low.

## 2. Vacancies

The application of the positron annihilation technique has proven to be very successful in the quantitative study of vacancies. With present methods positron trapping in vacancies is detectable at vacancy concentrations as low as 0.1 ppm. When the vacancy concentration is such that almost all positrons are trapped and annihilate in vacancies then the technique becomes insensitive to further increases in their concentration. Saturation generally occurs at vacancy concentrations of about 100 ppm. Many experimentalists have studied particle irradiation induced or thermally generated vacancies in a large number of pure metals and several alloy systems. Vacancies in alkali metals have not as yet been shown to produce changes in the positron annihilation parameters.

As previously stated, positrons which are trapped at monovacancies decay from these traps with lifetimes which are longer than those of positrons annihilating in the bulk. Also, trapped positrons produce angular correlation and Doppler-broadening parameters which are different from those generated by positrons annihilating in the perfect lattice. Therefore, the change in the annihilation component attributable to vacancy trapped positrons is a quantitative measure of the change in vacancy concentration. By determining the changes in the vacancy concentration as a function of sample temperature the vacancy formation enthalpy can be deduced. Positron annihilation has become the most widely adopted technique for measuring vacancy formation enthalpies in metals. Indeed, the positron annihilation technique is the only method that has, to date, been successful in measuring the monovacancy formation enthalpies in the refractory metals V, Nb, and Ta (15).

Lynn et al. (16) studied the effect of temperature from 4.2 K to 1700 K on the positron lifetime in 99.999 + wt.% polycrystalline Ni. Figure 5 is a plot of the single lifetime fit (approximately the mean positron lifetime) to the data as a function of temperature. Several metals, including Ni, exhibit a "precursor" effect in the temperature range (here 900 K to 1100 K) immediately preceding the onset of detectable vacancy trapping. A number of possible causes have been suggested ranging from extrinsic defects (17) to trapping at thermally generated transient dilations occuring in the lattice (18). At present the precursor effect is not well understood.

The sharp increase in $\tau_m$ at temperatures greater than 1100 K is unquestionably attributable to vacancy trapping of positrons. Saturation occurs in the single lifetime fit analysis of Ni beyond about 1350 K. The saturation of the lifetime at high temperatures is in part a result of shortcomings in the numerical analysis methods rather than a real physical effect. Doppler-broadening lineshape parameters, which are the result of a more simplistic analysis, do not exhibit the same saturation seen in lifetime data.

Kuribayashi et al. (19) first noted that a linear relationship exists between the so-called critical temperature, $T_c$, of a number of fcc metals

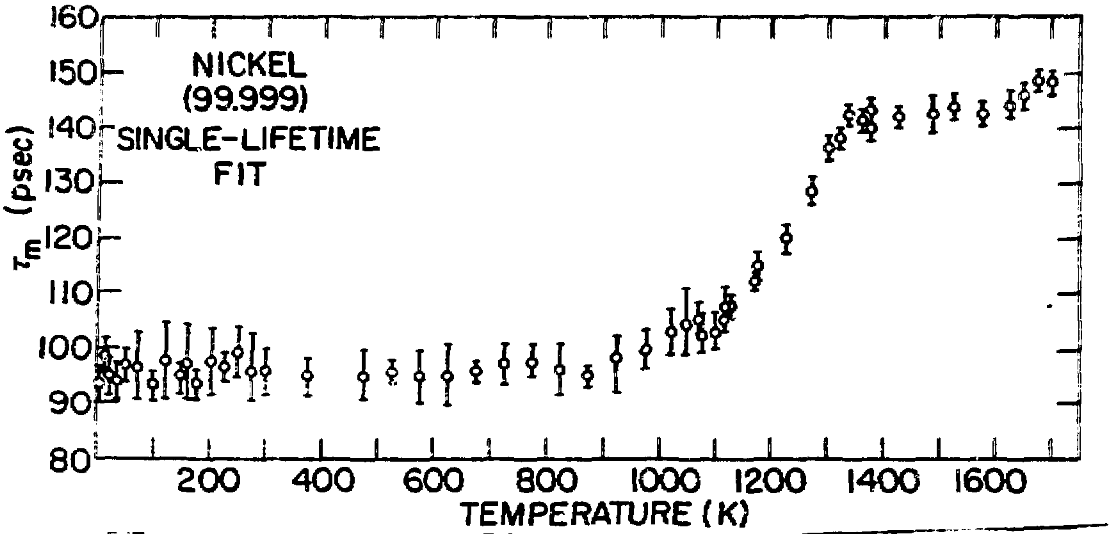



Fig. 5 - Single lifetime fit, $\tau_m$, vs temperature for well annealed Ni and alloys and their monovacancy formation enthalpies, $H^F_{IV}$. The critical temperature was defined as that temperature at which noticeable deviation occurs from the linear low temperature region. MacKenzie et al. (20) suggested an operationally simple procedure for finding the "threshold temperature", $T_t$. As with $T_c$ a linear relationship exists between $T_t$ and the monovacancy formation enthalpies of a large number of metals (practically $T_c \not\approx T_t$). $T_t$ is defined as the intersection of a linear fit to the prevacancy region with a linear extrapolation back from the early vacancy dominated region. More recently Schulte and Campbell (21) produced the equation

$$H^F_{IV} = (-0.098 \pm 0.057) + (15.2 \pm 0.7)\ T_t \times 10^{-4}\ \text{eV} \qquad (3)$$

from angular correlation and Doppler-broadening measurements. Campbell et al. (22) obtained a value of $H^F_{IV} = 1.45 \pm 0.07$ eV using a similar equation and their Doppler-broadening nickel data. By applying this procedure to their lifetime data Lynn et al. (16) obtained a value of $H^F_{IV} = 1.40^{+0.06}_{-0.01}$ eV. This value is somewhat lower than the value $H^F_{IV} = 1.54^{+0.1}_{-0.2}$ eV which was obtained by fitting the two-state trapping model (23,24,25) to the lifetime data. Lynn et al. (16) state that the generally less pronounced precursor effect exhibited by lifetime data relative to angular-correlation and Doppler-broadening data resulted in the lower value of $H^F_{IV}$ when the threshold temperature equation was applied. They therefore suggest a modified equation for lifetime data ($H^F_{IV} = 1.34\ T_t \times 10^{-3}$ eV) but warn that it is based solely on the determination of $T_t$ for their own Ni data.

Maier et al. (15) have recently measured the monovacancy formation enthalpies for a number of refractory bcc metals (W, Ta, Mo, Nb, V) and have found that the vacancy formation enthalpies for these metals do not follow the empirical relation above which is based mostly on the characteristics of close-packed metals but are better described by the equation

$$H^F_{IV} = 17k_B T_t \tag{4}$$

where $k_B$ is Boltzmann's constant.

The major advantages of using the threshold temperature method are (1) it avoids the somewhat questionable approximations inherent in the trapping model (21,27) and (2) vacancy formation enthalpies can be determined without heating the sample close to its melting point. This can be a major advantage when studying refractory metals or metals exhibiting a high vapor pressure in the solid state. This method, however, inherently assumes that the sensitivity of positrons to vacancies remains constant between metals.

The ability of the positron technique to quantitatively measure defect populations is useful in non-equilibrium as well as equilibrium situations. In contrast to resistivity, which must rely on only one parameter, positron data can be interpreted in terms of several complimentary parameters (e.g. the lifetime of free ($\tau_f$) and vacancy trapped ($\tau_t$) positrons and their associated intensities $I_f$ and $I_t$). This ability has been helpful in settling the debate over which migrating species, vacancies or interstitials, is responsible for Stage III recovery in several metals and in particular Mo (28,29,30). Eldrup et al. (29) were able to demonstrate that void formation occurred simultaneously with a decrease in the vacancy population. This was accomplished by following the intensities of the vacancy and void components in the positron annihilation spectra of electron irradiated Mo as a function of the isochronal annealing temperature. This strongly suggested that vacancies are the mobile defect.

In our own studies of 99.999 + wt.% Zn single crystals we have found that the Doppler-broadening S parameter exhibits an increase in value between the annealing temperatures 77 K and 150 K after deformation in liquid nitrogen (Fig. 6). Since this occurs in the stage III recovery temperature range for Zn (105 K to 160 K) (31) we interpret the increase as being due to the agglomeration of vacancies. Vacancy agglomeration commensurate with stage III suggests that dislocations in Zn are not efficient vacancy sinks. The knee in the curve (Fig. 6) at $\sim$200K is due to the annealing out of dislocations.

Free interstitials may combine with vacancies and so reduce the vacancy population, however, free interstitials themselves have not been observed to produce any effects on positron annihilation parameters.

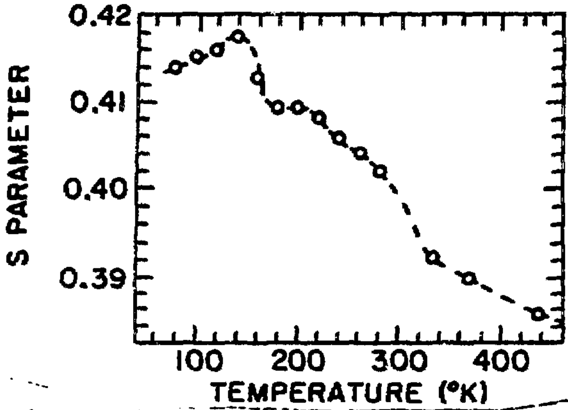



Fig. 6 - Doppler-broadening S parameter as a function of isochronal annealing temperature for Zn single crystals after deformation (10%) at 77K. All data points were taken at a sample temperature of 77K.

3. Voids

The production of voids in metals by particle irradiation is of great concern in the design of critical reactor components because of the swelling which accompanies void growth. In 1972 Mogensen et al. (32) and Cotterill et al. (33) established positron trapping at voids in Mo. Positron trapping has since been observed at voids in other metals. The positron technique is sensitive to voids of sizes well below that at which they become detectable with transmission electron microscopy. This sensitivity permits much shorter sample irradiation times. In addition, since the positron annihilation technique is non-destructive there is a possibility that it could be used to continuously monitor the void concentration in nuclear reactor components (33).

Positrons which annihilate in monovacancies do so with lifetimes which are typically 20% to 50% longer than those of positrons annihilating in the bulk. The lifetime of positrons annihilating in annealed Mo is 118 ± 2 ps (29), that of positrons annihilating in monovacancies in Mo is 193 ± 5 ps (34), and that of positrons annihilating in large voids (> 10 Å diameter) in Mo is 474 ± 10 ps (35); approximately 300% longer than that for free annihilations. Similarly, the narrowing of the angular correlation distribution and the reduction in the width of the Doppler-broadened photopeak which results from positron annihilation at voids in Mo is greater than that produced by the same fraction of positrons annihilating at vacancies.

This seems to be generally the case for positrons annihilating at voids in metals; for example in Ni (36) and in Al (37).

Positronium (electron-positron bound state) formation in voids was first considered as a possible cause for these very long lifetime and narrow Doppler-broadening distributions. However, room temperature magnetic quenching experiments performed on Mo samples containing voids (32,33) did not alter the resulting spectra. Magnetic quenching, associated with Doppler-broadening or angular correlation, is a sensitive test for positronium (Ps) only if it causes Ps transitions from a state with a broad momentum spectrum to a state with a narrow one. Since the nature of the supposed Ps state in a void is unknown, the evidence against Ps formation is not very strong.

Two models have been proposed to describe the state of positrons at a void. In the model of Hautojärvi et al. (38) the positron enters the void with its screening cloud. In the model of Hodges and Stott (39) the positron is trapped at the surface of the void. In either case both models predict that the positron lifetime should be very sensitive to changes in void dimensions for voids with diameters below about 10 Å (Fig. 7). Grynszpan et al. (40) state that the surface trap model is probably superior for larger voids whereas the model of Hautojärvi et al. (38) is probably a better representation of the physical situation in small voids.

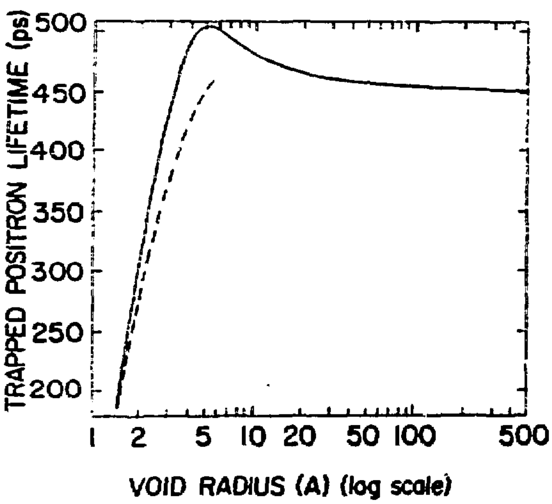



Fig. 7 - Predicted positron lifetime as a function of void radius. (---) Hautojärvi et al. (38) (—) Grynszpan et al. (40)

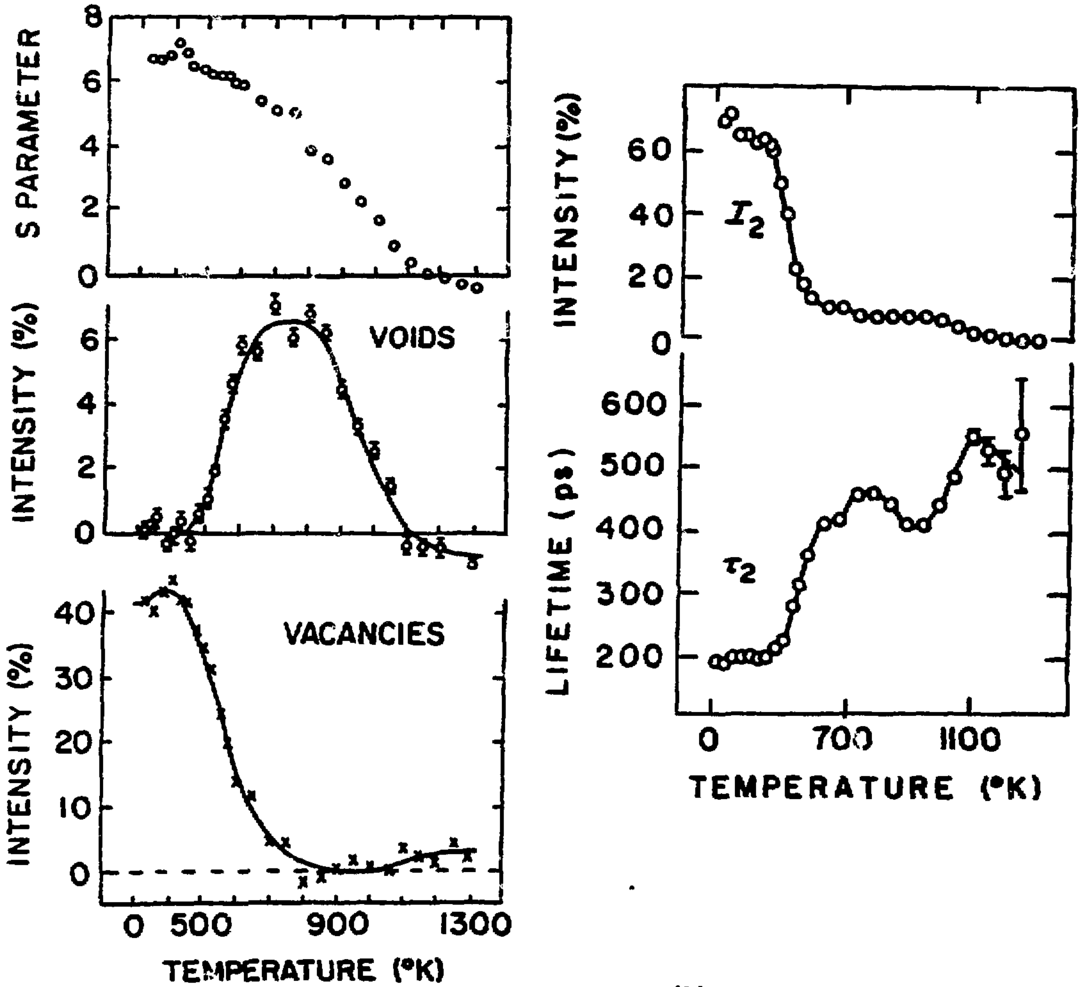
In any real sample there is a distribution of void sizes as well as a significant number of dislocation loops. This will result in an associated distribution of positron lifetimes. However, because of the inherent difficulties in resolving many similar lifetime components, an experimentally determined void lifetime is actually a weighted average of many lifetimes.

Petersen et al. (30) studied the effect of increasing temperature on 99.99% Mo single crystals which had been irradiated with fast neutrons ($E > 0.1$ MeV) to a fluence of $1.5 \times 10^{18}$ n/cm$^2$ at 333 K. A similar experiment was performed on 10 MeV electron irradiated low carbon polycrystalline Mo by Eldrup et al. (29). The two studies yield results which are in qualitative agreement. Both experiments demonstrate that it is possible to correlate positron annihilation behavior with the detailed mechanisms leading to void production. They also demonstrate that positron annihilation techniques can be used to complement the results of both electron microscopy and resistivity studies. The work of Eldrup et al. (29) has shown that the positron annihilation technique can be particularly useful in complimenting resistivity studies both because of its multiparameter nature and because positron annihilation, unlike resistivity, is insensitive to isolated interstitials.

Angular correlation and Doppler-broadening data for defect studies are almost always described in terms of peak counting rates or lineshape parameters. When the full angular distribution or Doppler-broadened photopeak is analyzed in this way it is possible to overlook much of the information contained in the spectra. Computer programs such as PAACFIT (41), PARAFIT (42), or ANNIH (43) should be used whenever possible. With such programs the contributions of different types of defects can be extracted from the angular correlation or Doppler-broadening data in a manner that is analogous to that in which multiple lifetime components may be deduced from a lifetime spectrum. When the changes occurring to two types of defects simultaneously produce opposite effects on the momentum distribution a lineshape parameter or peak counting rate may exhibit only a slight change and the effect may appear small. However, when the data are analyzed with a program such as PAACFIT (41) large changes in the defect concentration and type may become evident. This was the case in the experiment on electron irradiated Mo by Eldrup et al. (29) (Fig. 8a)

In both the neutron and electron irradiation studies of Mo, $\tau_2$ values (the lifetime component associated with voids) changed rapidly in two stages (Figure 8b). During the first stage $\tau_2$ increased and seemed to saturate at roughly 460 ps at a temperature of about 700 K. This increase is associated with void formation. $\tau_2$ began its second increase starting at approximately 973 K. This second increase was attributed to thermal coarsening of voids; larger voids growing at the expense of smaller ones. A later study by Thrane et al. (44) showed that, in agreement with theory, the long lifetime component in high purity Mo is independent of average void size for void radii in the range $r = 9$ Å to $r = 45$ Å. Further study

(35) associated the increase in $\tau_2$ above $\sim$470 ps with the migration of O, C or N impurities to void surfaces at high temperatures. This does not invalidate the basic annealing processes deduced from the irradiation damage annealing studies, however, it does demonstrate the need for more sophisticated analysis of the data. As noted by Thrane and Evans (45): "Provided such effects are (properly) identified they could add more power to the effectiveness of positrons annihilation."




(a) (b)

Fig. 8 – Results of isochronal annealing studies of electron irradiated Mo. (Eldrup et al. (29)).

A recent paper by Schultz et al. (46) suggests thermal detrapping of positrons from void surfaces and concommitant Ps formation inside voids in Mo at high sample temperatures. The voids were characterized as having a mean diameter of $\sim$37 Å. In addition to thermal detrapping of positrons from surfaces. slow positron studies (47) indicate that the presence of certain impurities may either reduce or increase the surface trap depth. Coupling these slow positron surface results with void studies should produce a quantitative description of positron trapping at voids.

4. Dislocations

The most common techniques used to study the effects of plastic deformation on the production of dislocations are transmission electron microscopy (TEM), X-ray topography, and etch pitting at emergent dislocations. TEM is destructive since the samples must be thinned to several hundred angstroms. In addition, because of thinning, relaxation of the dislocation substructure may occur. X-ray topography is usually limited to dislocation densities below about $10^6$ cm/cm$^3$. However, a typical laboratory grown metal crystal usually contains $10^4$ to $10^6$ cm/cm$^3$ of dislocation line. Deformation can increase this concentration several orders of magnitude. Chemical etch pitting techniques frequently rely on critical temperature regulation, etching times, and crystal orientations. In addition, chemical etching effects the surface under study. By comparison, the positron annihilation technique can be used on bulk crystals (typically several millimeters thick) and in many instances critical temperature regulation and crystal alignment are not required. It is thought that the dilated side of an edge dislocation should provide a trapping site for positrons. However, deformation produces other types of defects in addition to increasing the dislocation density. Jogs in dislocation lines can be produced when moving dislocations cut other dislocations. The non-conservative motion of jogs can produce strings of vacancies or interstitials. The difficulty then lies in distinguishing the positron annihilation signal that originates in dislocations from those that originate in defects associated with the dislocation. Since screw dislocations are not generally believed to be capable of trapping positrons the ideal situation would be the production of a high concentration of long straight edge dislocations in an otherwise perfect single crystal. In this way the annihilation parameters associated with a dislocation could be established (48). Failing this, what is usually done is to deform a single or polycrystalline metal sample and then take advantage of the fact that point defects can generally be annealed out of a sample at lower temperatures than can dislocations.

Hautojärvi et al. (49) performed an experiment of this type on 99.998 wt.% and commercial grade polycrystalline iron. In this experiment the samples were rolled to 40% of their initial thickness. Deformation to this extent produces large numbers of dislocations. After several days (presumably at room temperature) a series of isochronal anneals were performed at increasing temperatures. The lifetime and Doppler-broadening data which were taken resulted in the curves of Figure 9. Vacancies in high purity iron are known to become mobile at 220 K (50) and are therefore not expected to seriously effect the results. A sharp recovery is observed between 673 K and 873K. Recrystallization is known to occur in this temperature range and therefore the recovery is ascribed to dislocation movement to grain boundaries. Polygonization is the suggested cause of the slight recovery beginning at 573 K in the high purity sample.

In an attempt to produce dislocations without the associated debris resulting from deformation Cotterill et al. (51) quenched 99.9999% polycrystalline Al samples from high temperatures to freeze in vacancies and then annealed at 353 K to produce dislocation loops.

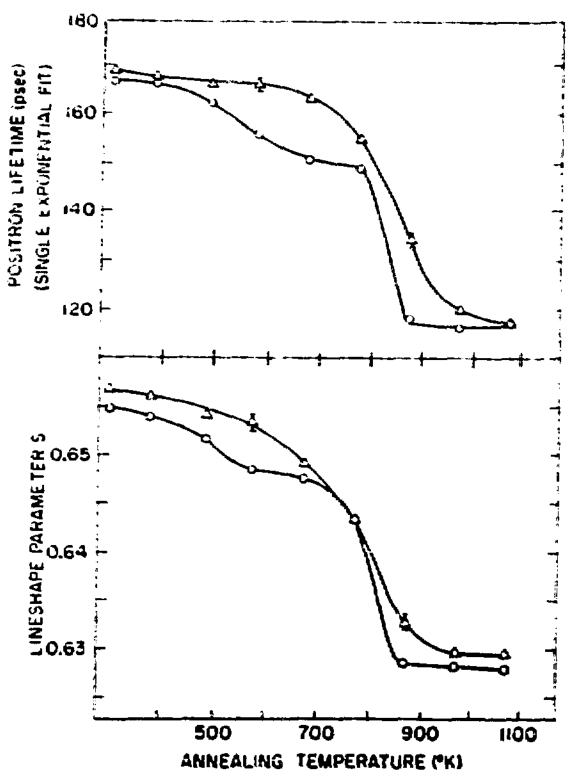



Fig. 9 - Positron lifetime and Doppler-broadening recovery curves produced by isochronal annealing after deformation of polycrystalline iron (49).
△ Fe 99.86% ○ Fe 99.998%

The lifetime of positrons annihilating at vacancies and dislocation loops was found to be 246 ± 4 ps and 250 ± 30 ps, respectively. A free positron lifetime of 166 ± 2 ps was measured. The similarity of positron lifetimes at vacancies and dislocations exists in a number of metals. This has lead several investigators to suggest that positrons may be trapped by dislocations but ultimately annihilate at vacancies which are themselves trapped by the dislocation (52) or at jogs with vacancy-like character (53,58). Hjelmroth et al. (54) have noted that quenching and subsequent annealing of vacancies in Al is known to produce a mixture of voids and dislocation loops (55). The presence of voids could have contributed to the results of this experiment since voids in Al are known to be deep traps for positrons (37,56,57).

Another experiment which attempted to isolate the response from positrons trapped at dislocations was performed by Dannefaer et al. (48). In this experiment Ni, Co and a series of Ni-Co alloys were deformed and then subjected to a series of isochronal anneals at increasing temperatures. In most of the samples the lowest trapped positron lifetime observed after annealing was 140 ps. The fact that this same value is obtained from materials with widely differing stacking fault energies and hence different dislocation core configurations implies that the positrons are not annihilating in the dislocations. Since this lifetime is close to the 142 ps Ni monovacancy lifetime (16) it is suggested that the annihilations are taking place at vacancies or at jogs with vacancy-like character.

Hjelmroth et al. (54) proposed a model consisting of two types of traps in close physical proximity [nominally (A) dislocations and (B) jogs]. In this model either defect may trap positrons but once trapped a positron may not escape into the bulk. However, positrons trapped at the A-type traps may escape or diffuse to the deeper B-type traps. There is no detrapping from B-type traps.

Smedskjaer et al. (53) have produced a similar model. In this theory the positron binding energy to a perfect edge dislocation is small ($\sim$0.1 eV). The positron signal from the dislocation is not easily resolved from the bulk signal. The presence of dislocations results in changes in the positron lifetime primarily because of the trapping of positrons by point defects associated with dislocations (e.g. jogs). Positron binding to these point-like defects is presumed to be high (a few eV). Figure 10 (53) shows a schematic representation of the trapping model. This version also takes into account detrapping from dislocations with a rate $\delta$. The model predicts the possibility of a temperature dependent trapping rate to shallow traps (i.e. dislocations). It also predicts, for positrons already localized at a dislocation, a high probability of trapping into deep traps along the dislocation line when the deep trap "density" is $\gtrsim 10^{-4}$ Å$^{-1}$. The authors suggest that the low temperature effects that have been observed by several experimentalists (17,59,60) might be explained by their model.

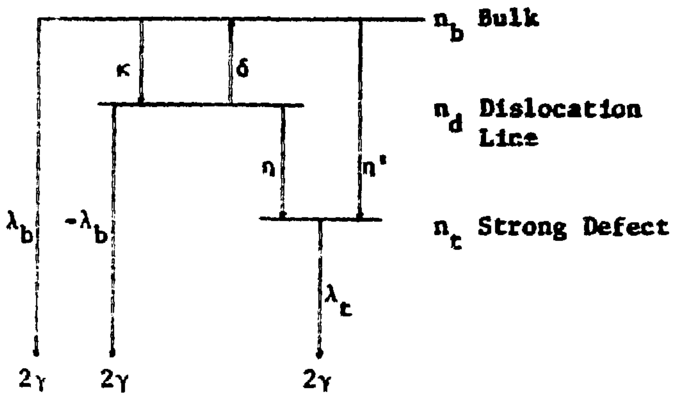



Fig. 10 - Schematic representation of the trapping model (53). κ, η, and η' are trapping rates to the states shown. δ is the detrapping rate from dislocations. $\lambda_b$ and $\lambda_t$ are annihilation rates.

Although the exact nature of positron trapping at dislocations is clearly not well understood at the present time this need not preclude the "applied" use of positrons for dislocation related defect studies. Lynn et al. (61) have demonstrated that positron annihilation techniques can be used to complement X-ray particle size studies during fatigue in Ni and Ni-Co alloys. During this experiment the mean positron lifetime was observed to increase monotonically (Fig. 11) and saturate at ~7% of the fatigue life. The changes in mean positron lifetime saturated earlier than did changes in X-ray particle size (Fig. 12) and thus the mean positron lifetime is a more sensitive indicator of early fatigue damage.

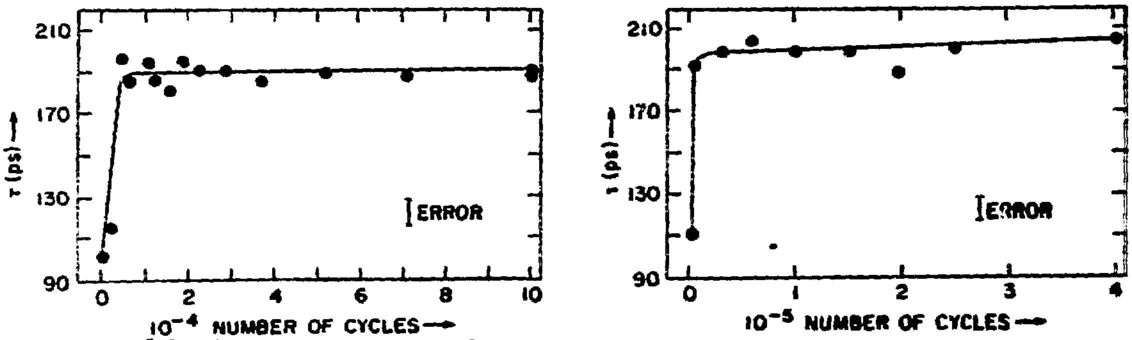



Fig. 11 - (a) The mean positron lifetime in pure Ni as a function of cyclic deformation. (b) in Ni - 66.5% Co (61).

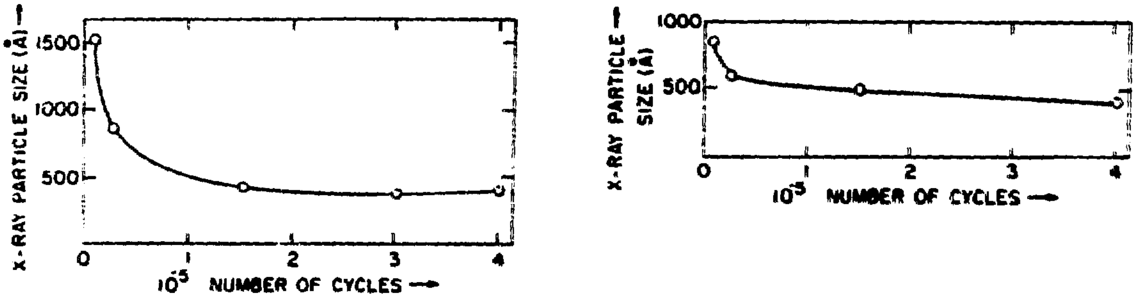



Fig. 12 - (a) X-ray particle size in pure Ni as a function of cyclic deformation. (b) in Ni-66.5% Co (61)

5. Grain Boundaries

Several experiments have been performed to measure positron annihilation characteristics as a function of grain size (62,63,64). Positron trapping at grain boundaries is expected because of the lower atomic density there relative to the bulk lattice. As noted by McKee et al. (62) it is probable, as with dislocations, that a spectrum of lifetimes favoring the deepest traps are associated with annihilations at grain boundaries.

The trapping rate to a defect may be either transition limited or diffusion limited. In the transition limited case the trapping rate is solely determined by the defect density and the positron transition time between the free and trapped states. In the diffusion limited case the trapping rate is determined by the positron's ability to get to the defect. (See McKee et al. (62) and references therein.) McKee et al. (62) find a monotonically decreasing mean lifetime and Doppler-broadening S parameter with increasing grain size (0.38μm to 1.9 μm) in fine grain Zn-22 wt.% Al alloy. Using the diffusion limited model they measured a positron diffusion coefficient in Zn-22 wt.% Al at 293°K of $D_{e+} = 0.6\ cm^2\ sec^{-1}$; a value of both theoretical and practical importance. Slow positron measurements on high purity Al at room temperature yield a value of $D_{e+} = 0.4\ cm^2\ sec^{-1}$ (see section 8).

6. Alloys

The positron annihilation technique is often used in the study of lattice defect mechanisms in pure metals, thus avoiding many of the complications inherent in the study of alloys. However, because of their technological importance lattice defect mechanisms in alloys are worthy of similar consideration. Al-Cu and Fe-C form the basis for a large number of technologically important alloys and are therefore interesting candidates for positron annihilation studies.

The positron annihilation technique has recently been used to study the formation of precipitates in high purity Al samples which had been alloyed with 0.5, 2.0, and 4.0 wt.% Cu (65). The eventual formation of the Cu rich incoherent tetragonal θ phase precipitate occurs via a sequence of metastable phases GP1 → GP2 → θ' → θ. During the initial stage of precipitation coherent precipitate particles known as Gunier-Preston (GP) zones are formed. The GP1 precipitate is a single atomic layer of Cu atoms formed in the Al (100) planes. GP2 zones are ordered Al-Cu platelettes up to about 25 layers thick. The θ' phase forms as a partially incoherent precipitate which, upon further growth, finally results in the fully incoherent tetragonal θ phase precipitate.*

Gauster and Wampler (65) homogenized their alloyed samples at 823 K and then quenched to 200 K. A series of isochronal anneals were then performed at successively increasing temperatures. A 99.995% Al sample underwent similar treatment for comparison. It was quenched to 200 K from 773 K. All of the positron measurements were taken at 85 K. They interpret the results (Fig. 13) as follows: The large value of the S parameter for the as-quenched samples (relative to the annealed state) is attributable to positron trapping at quenched-in thermally generated vacancies. The increase in the value of the S parameter immediately after the 200 K anneal is ascribed to vacancy agglomeration. Close association of vacancies to Cu allows for less agglomeration with increasing Cu content. The drop in the value of the S parameter between 230 K and 300 K is related to a decrease in the total number of vacancies. Electron microscopy provides evidence of GP zone formation by 300 K. S parameter analysis shows that positrons are not sensitive to the further growth (GP1→GP2) of the precipitate between 300 K and 450 K. In this temperature range the S parameter for the 2.0 and 4.0 wt.% Cu alloy is lower than that for pure annealed Al. This indicates that the copper rich GP zones are being preferentially sampled by positrons.** Vacancies are thought to be associated with these zones for temperatures up to 450 K. Positron trapping at vacancies associated with the precipitate would provide a mechanism for preferential sampling of the Cu rich regions.

For annealing temperatures below ~450 K the GP1 and GP2 zones remain coherent with the Al lattice, however, growth beyond this stage results in lattice strain too great to be accommodated coherently. The θ' phase is formed as an ordered tetragonal precipitate of composition near $CuAl_2$. It grows in the form of thin platelettes whose faces are coherent with the Al lattice but whose edges have misfits which are accommodated with dislocations. In addition, vacancy clusters may be associated with these interphase boundaries. Positron trapping at defects associated with the θ' precipitates would result in preferential sampling of the Cu rich areas.

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*Gauster and Wampler (65) cite a number of references on θ phase precipitation.

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**Gauster and Wampler (65) find that the S parameter in their analysis is 19% larger for pure annealed Al than for pure annealed Cu.

Gauster and Wampler (65) suggest that the creation of the lattice defects associated with the θ' precipitate formation would produce positron trapping which results in the sharp reduction in the S parameter immediately after the 450 K anneal for the 2% and 4% Cu samples. Annealing at temperatures above ~650 K produces the fully incoherent θ phase. Since these precipitates are large and widely separated the probability of positron trapping is reduced to the precipitate volume fraction.

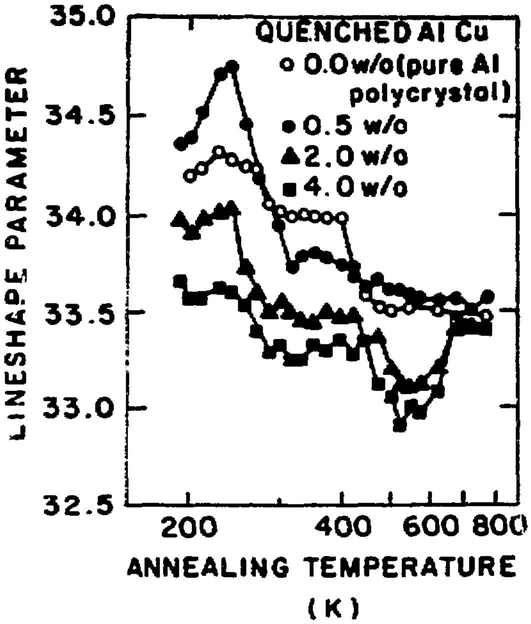


Fig. 13 – Doppler-broadening lineshape parameter S (x100) as a function of isochronal annealing temperature (log scale) (65).

The usual situation in lattice defect studies is that positron trapping results in an increase in both the S parameter and the mean lifetime. In many cases the longer second lifetime and its associated intensity can be deduced from the data. The results of this study, however, suggest that lattice defect trapping of positrons results in a decrease in the S parameter due to preferential sampling of one component of the alloy. Gauster and Wampler (65) did not report any lifetime data. It might be interesting to see if two positron lifetimes could be resolved. One could, however, imagine running into prohibitive numerical problems since, assuming that vacancy and dislocation trapped positron lifetimes are approximately equal, the positron lifetime in well annealed Cu and Al differ by only about 45 ps (120 ps Cu vs. 165 ps Al) and the lifetimes of positrons annihilating in bulk Al and Cu vacancies (~180 ps) are nearly the same.

Carbon in Fe is one of the oldest, most frequently studied, and most technologically important alloys. Yet, a number of questions remain unanswered with regard to defect mechanisms in α-Fe (66). Specifically, the mechanism responsible for stage III recovery in α-Fe has been the subject of much controversy ([illegible],66). Recently Hautojarvi et al. (50) have used the positron annihilation technique to study the interaction of monovacancies and interstitial carbon impurities in electron-irradiated α-Fe. In this experiment pure Fe samples and samples doped with 50 and 750 ppm carbon were annealed at 1023 K, quenched to 273 K, and electron irradiated at 20 K with 3 MeV electrons to produce isolated vacancies. Positron lifetime measurements were then performed after a series of isochronal anneals at increasing temperatures up to 600 K. The results of the study are interpreted by the authors to indicate that vacancy carbon pair formation occurs at 220 K resulting from vacancy capture by immobile carbon atoms. The pair still exhibits positron trapping with a lifetime of 160 ps; only slightly shorter than the positron lifetime in vacancies (175 ps) for pure Fe. This suggests that, in agreement with theoretical calculations (67), carbon is located rather far off center of the vacancy. Above 350 K interstitial carbon becomes mobile further decorating the vacancies and inhibiting positron trapping.

Positron trapping is not generally believed to occur at isolated substitutional impurities (68,69), however, the results of recent experiments (70,71) suggest the contrary for Al-Mg alloys. In these experiments 99.9999% Al was alloyed from the melt with 100 ppm and 1000 ppm Mg. The samples were annealed in vacuum and surface impurities were chemically polished away. Results of the analysis of lifetime data (70) were shown to be inconsistent with positron trapping at nonequilibrium defects (such as Mg-vacancy pairs). The results seem to suggest positron trapping at isolated substitutional Mg. Angular correlation studies on the same samples (71) support the previous lifetime studies. As noted by the authors these results are important because, in addition to showing unexpected trapping at substitutional impurities, the possibility of positron trapping by substitutional elements may alter the interpretation of positron studies of the Fermi surfaces of alloys.

Bernardin and Dupasquier (71) state only that their Al-Mg samples were alloyed in the melt and annealed in vacuum. Experiments performed by Alam (72) have demonstrated that positrons are sensitive to MgO which is produced in the bulk of an Al-500 ppm Mg sample if it is annealed in the presence of even a small amount of $O_2$ ($N_2$ containing 500 vol. ppm $O_2$). Also, muon depolarization measurements (73) have suggested the possible existence of small clusters of Mg or even MgO in annealed Al-1000 ppm Mg alloy. Thus, an alternative interpretation, and one more consistent with the general concepts of positron trapping, may be possible depending upon the specific sample preparation conditions.

## 7. Fatigue

From a technological point of view detailed knowledge of the defect mechanisms associated with the fatigue of metals is obviously highly desirable. However, fatigue is a complex process involving point, line, and surface defects. The ability of the positron technique to distinguish both defect type and concentration should be of use when applied to the study of this process.

Since the positron technique is nondestructive and since the gamma ray detectors need not be in physical contact with the sample, dynamic fatigue experiments can be performed involving temperature, ultimate stress and strain or even composition of the ambient atmosphere as variable parameters. Dynamic fatigue measurements are probably not amenable to the usual sample-source-sample sandwich arrangement. In certain instances "massless" sources (74) may be applicable or corrections for annihilations outside the sample may be necessary when the positron source cannot be sandwiched between two samples.

By using isotopes which emit positrons at different energies and through the use of an energy tuneable positron beam (see section 8) a variable depth of penetration may be attained. This would allow the sample to be studied in layers without the need to polish away surface material. To date, this method has not been applied to fatigued samples. However, it has been used in a study of Al clad aluminum alloy (75) (Fig. 14).

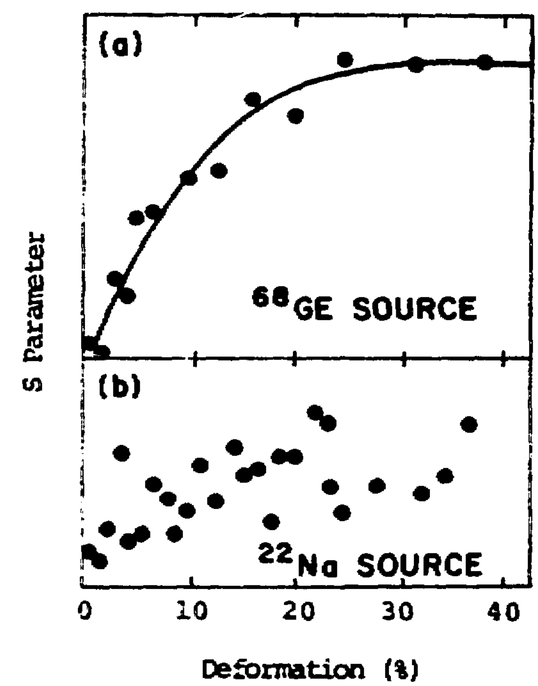


Fig. 14 - Variations of the Doppler-broadening annihilation line width with deformation in rolled Al clad 2024 samples irradiated with (a) $^{68}$Ge positrons (b) $^{22}$Na positrons (J. Eady et al. unpublished).

Despite these possibilities very few positron studies of fatigued metals have been performed, possibly owing to supposed difficulties in analysis and interpretation of data. In one of the few positron studies of fatigued metals (76) the mean positron lifetime in fatigued 304 stainless steel attained, early on in the fatigue process, a value which was ~7% greater than that in a heavily deformed sample of the same material. This result indicates the presence of vacancy agglomerates which are apparently produced during cyclic deformation but not during monotonic deformation.

In another study (77) the difference in the S parameter, ΔS, between a fatigued and annealed sample of a low carbon steel was found to have a square dependence on the maximal applied stress for a fixed number of cycles. If the ΔS value is assumed to be proportional to the defect density then this dependence will reflect the relation between defect density and applied stress.

Hjelmroth et al. (54) studied the effects on positron annihilation of fatigue in 99.9999% Al single crystals. Only the free positron lifetime was found for fatigue below 60 cycles. During a later stage (100-350 cycles) in addition to the free lifetime a 215 ± 15 ps lifetime was also measured. Still later (>350 cycles) a lifetime of 230 ± 15 ps increasing with the number of fatigue cycles to 260 ± 15 ps was measured. Under the experimental conditions a relatively untangled dislocation population should be produced during the initial stage of fatigue (<60 cycles) and subsequently rearrange (~100 to 350 cycles) to form subgrain boundaries whose walls are composed of dense ($10^9$ -$10^{11}$ cm/cm$^3$) heavily jogged dislocations. This implies that untangled dislocations are not efficient positron traps. Fatigue for ~100 to ~350 cycles produces heavily jogged dislocation tangles. During this period a 215 ps lifetime is measured and ascribed to positron trapping and subsequent annihilation at jogs. Fatigue beyond ~350 cycles produces a rearrangement of dislocations which precipitates the production of a large number of point defects. During this "third stage" a third lifetime component (230 to 260 ps) is detected and assigned to voids/vacancy dipoles.

## 8. Future Prospects and Present Limitations on the Positron Annihilation Technique

The positron annihilation technique provides the researcher with one or more annihilation components each of which may be associated with annihilations in the bulk or at vacancy-like defects. The combination of insensitivity to interstitials and a unique "signal" resulting from annihilations at each type of vacancy-like defect promises to make positron annihilation useful in the study of complex recovery or recrystallization mechanisms. This is particularly so under conditions where several types of defects may exist simultaneously. By following the intensity and character of the annihilation components, defect types, mechanisms, and kinetics may be determined. However, with presently available experimental techniques it is usually impossible to resolve more than two or at best three annihilation components. Further complications can arise when the data contain annihilations from defects such as grain boundaries and dislocations. Annihilations at these defects probably result in a range of very similar annihilation components which are very difficult to resolve numerically.

The large mean energy of positrons (50 keV-MeV range) obtained from radioactive isotopes has not allowed positrons to be used in the study of surface or near surface defects. Typically less than 1% of the particles injected into a solid diffuse back to the surface before annihilation. This is clearly an advantage for bulk studies but makes surface studies practically impossible. Recently, however, it has become possible to greatly increase this percentage by using an energy tuneable (1 eV to 10 keV) monoenergetic positron beam (78). The variable energy beam allows the initial implantation depth to be varied and therefore permits surface as well as near-surface positron annihilation studies. In addition to conventional Doppler-broadening measurements the condition of the surface of a sample may be studied by measuring the fraction, energy, and direction of reemitted positrons and the fraction of positrons which are reemitted as positronium. Very recently it has become possible, by pulsing the slow positron beam (79), to perform lifetime measurements as well.

The positron annihilation technique is a useful laboratory tool for, among other things,* basic research in the field of crystal lattice defect mechanisms. In many instances its greatest value, to date, has been when used in conjunction with other techniques such as resistivity or electron microscopy. While the positron annihilation technique is not yet mature enough to be used for nondestructive evaluation of samples in the field, there is certainly the promise of many practical applications in applied as well as basic research settings; particularly in regard to slow positron studies of defects at metal-semiconductor and semiconductor-semiconductor oxide interfaces.





*The positron annihilation technique has also been used to study the electronic structure of [illegible].

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