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ABSTRACT

Quadrupole oscillations have been observed with ^{59}Co pulsed NMR in the intermetallic compound $GdCo_2$. From these oscillations the nuclear electric quadrupole interaction (EQI) has been studied as a function of temperature in the range 4K-312K. The value measured at 4K, ν_Q = 672 ± 3 kHz, is the largest so far reported for the cobalt EQI in the RCo_2 intermetallics. The EQI decreases with increasing temperature, reaching 432 ± 10 kHz at 312 K.

The amplitude of the oscillations tends to decrease with temperature, being also dependent on the easy direction of magnetization of the compound. Thus, above 200K, as the direction of magnetization changes, large oscillations are again visible in the satellite line; the main line shows no oscillations in this range.

The observed temperature dependence of the EQI is roughly linear, as found in other transition metal systems. (Author)

Key words: NMR, $GdCo_2$, EFG, electric quadrupole interaction

1. INTRODUCTION

The presence of unresolved nuclear quadrupole interactions in an NMR spectrum gives rise to oscillations in the spin echo amplitude, as a function of pulse separation (Abe et al. [1], Degani and Kaplan [2]).

The frequency of the oscillations is related to the quadrupolar parameter by $\nu_Q = a/\pi$ where

$$a = \frac{3e^2qQ}{4I(2I-1)} \frac{1}{2} (3 \cos^2 \theta - 1) \qquad (1)$$

where eq is the electric field gradient (EFG), and θ is the angle between the principal axis of the eq tensor and the magnetic hyperfine field.

These quadrupole oscillations have been observed in the ^{59}Co resonance at 4.2K in several RCo_2 Laves phases (R ≡ Nd, Tb, Ho, Er and Tm). (Hirosawa and Nakamura [3]).

The temperature dependence of the magnetic hyperfine interaction in $GdCo_2$ was studied via ^{59}Co NMR by Cannon and others [4]; the changes in the NMR spectrum, as a function of temperature, revealed a change in the direction of easy magnetization occurring around 200K.

2. EXPERIMENTAL

The sample used in the present measurements was prepared by melting the constituents in an arc furnace (with 1.1% excess Gd); the button was homogenised at 900°C for 100 h. It was crushed under acetone, mixed with silicone oil and sealed in a sample holder. X-ray analysis revealed that the sample had the correct C15 structure.

The NMR measurements were made using a Bruker SXP spectrometer with either a helium or nitrogen bath cryostat or a flow cryostat. Pulses of length from 0.5μs to 2.0μs were used, and the separation between pulses varied in steps of 0.1μs. The sequence was repeated with a rate of ∿0.5 kHz.

3. RESULTS

The spectra of echo height versus pulse separation show at low temperature very prominent oscillations with an amplitude of the order of 15% (Fig. 1a). In an intermediate temperature range ($40K < T < 120K$) the oscillations were less evident due to a decrease in their amplitude accompanied by a shortening of T_2 (Fig. 1b).

In a higher temperature range the frequency spectrum contains two peaks [4]; only the peak at higher frequency shows oscillations (Fig. 1c). This peak corresponds to ^{59}Co nuclei that have an angle θ (Eq. 1) of about 0^o. The amplitude of the oscillations increases with temperature up to about 40% for this peak (Fig. 1c).

The frequency of the quadrupole oscillations ν_Q was extracted from the data by fitting the curves to the function

$$E(2\tau) = \exp(-2\tau/T_2)[Co + C_1 \cos(2\pi\nu_Q\tau + \delta)] \qquad (2)$$

This assumes that only one frequency was present in each curve.

The quadrupole frequencies ν_Q (Fig. 2) show an overall linear temperature dependence, with small, but significant deviations below 90K (in more than one sample) and with a slight discontinuity around 200K.

The frequency of the quadrupole oscillations was generally measured by exciting the nuclear spins with the rf frequency that produced largest echoes; in other words, by sitting at the centre of the line in the echo height vs. frequency spectrum. A small dependence of ν_Q with excitation frequency was found, similarly to the case of $^{27}A\ell$ NMR in Dy $A\ell_2$ (Bowden et al. [5]); the frequency ν_Q varied about 1,5% per MHz.

4. DISCUSSION

We have observed quadrupole oscillations in the spin echo spectra at every temperature in the range 4K - 312K. We have varied the pulse separation in time intervals of 0.1μs, five times smaller than those used by Hirosawa and Nakamura [3]; this may explain why they did not report oscillations in the NMR of $GdCo_2$.

The temperature dependence of the quadrupole interaction parameter ν_Q measured in the present work is shown in Fig. 2. The values of ν_Q vary from 672 ± 3 kHz at 4.2K to 432 ± 10 kHz at 312K. The value at low temperature is the largest observed with ^{59}Co NMR in the RCo_2 compounds;

Hirosawa and Nakamura [3] reported values at 4.2K that range from 260 kHz ($TmCo_2$) to 420 kHz ($HoCo_2$, $NdCo_2$).

The general trend of $\nu_Q(T)$ is a linear dependence with T, apart from certain deviations below 90K. These deviations are probably related to tetragonal distortions in the compound [6]. In the range above 200K two effects may be simultaneously present: the variation of the EFG with temperature and the variation of θ described by Cannon et al. [4]. This makes the analysis of $\nu_Q(T)$ more difficult. The maximum in the amplitude of the quadrupole oscillations above 200K can be explained from the higher symmetry of the EFG tensor: when θ is near 0° the axis of the local EFG and the axis of the lattice EFG coincide.

In most metallic matrices the nuclear electric quadrupole interaction obeys the $T^{3/2}$ "law".

$$eq(T) = eq(0)(1 - BT^{3/2}) \qquad (2)$$

which applies to dilute as well to concentrated systems (Vianden [7], Verma and Rao [8]).

Our results do not allow a verification of the exponent of T. They seem, however, to be compatible with a linear dependence on T. This type of dependence has been observed in rare-earth metals and in some transition metal hosts [7], [8].

5. ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

Fig. 1 ^{59}Co spin echo height versus pulse separation in $GdCo_2$.

a) sample at 4.2°K, 62.8 MHz.

b) sample at 38°K, 61.7 MHz.

c) sample at 297°K, frequency of 47.3 MHz (first peak).

d) sample at 297°K, frequency of 52.0 MHz (second peak).

Fig. 2 Normalized electric quadrupole frequency versus temperature in $GdCo_2$.

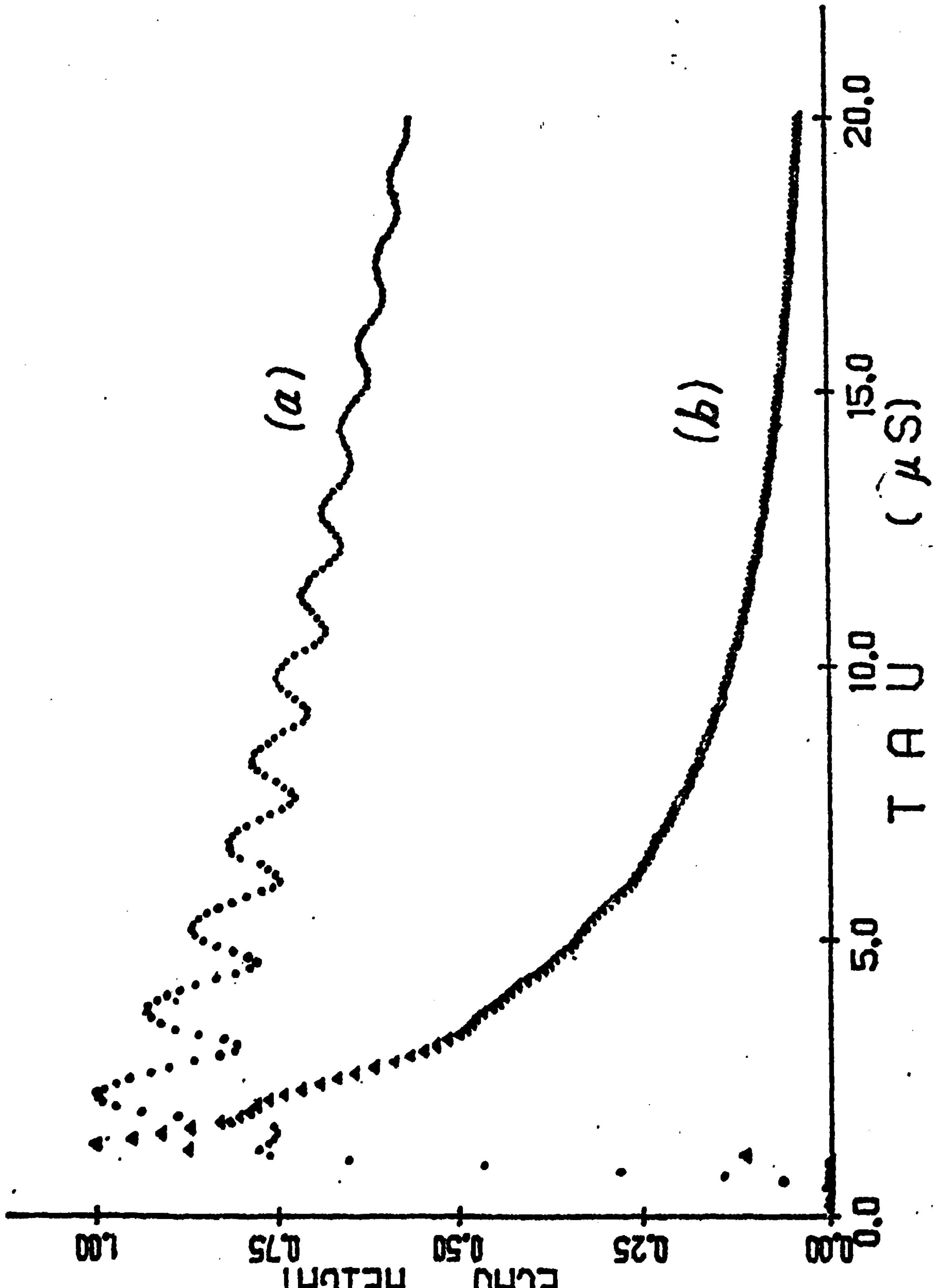

FIG.1

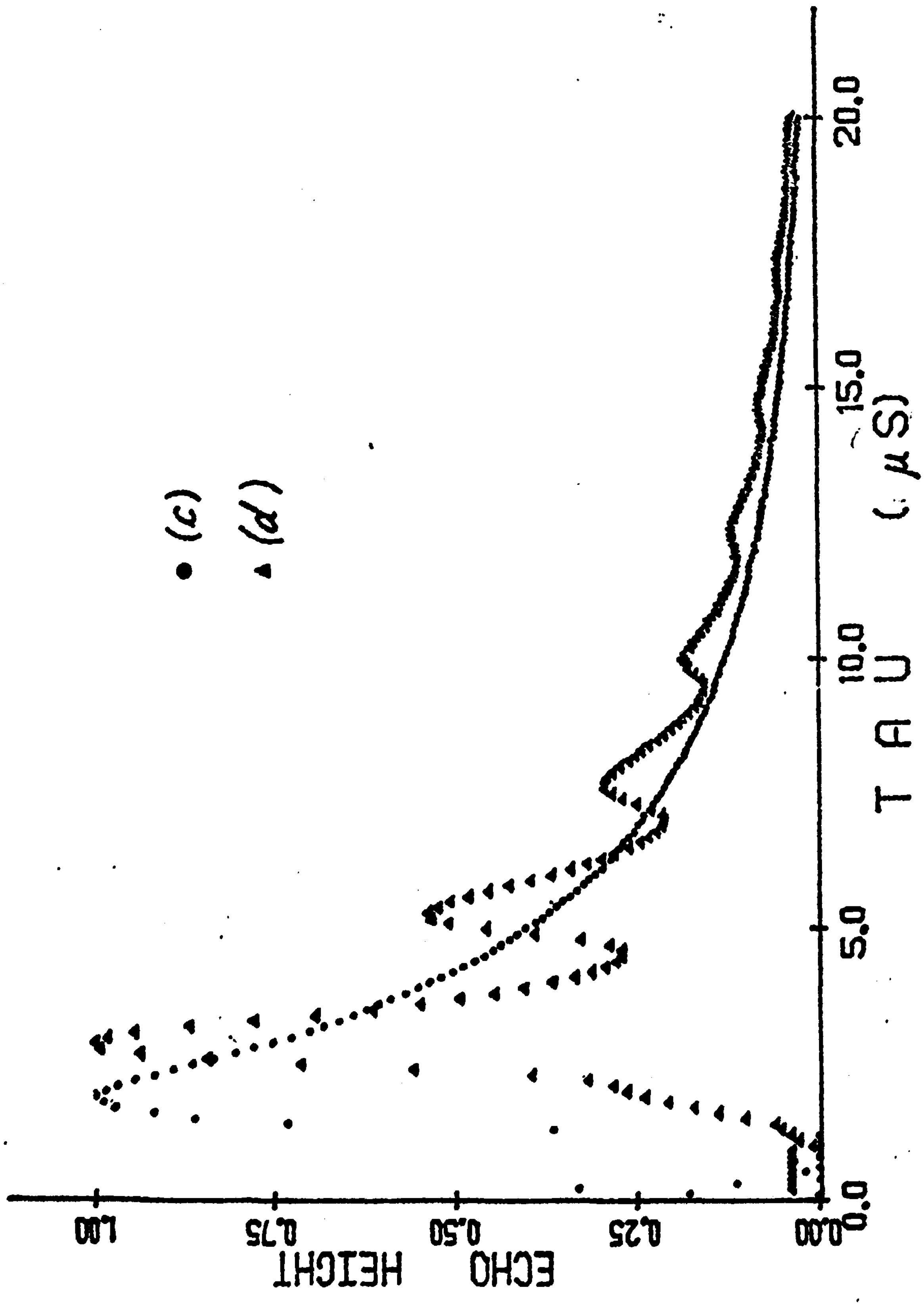

FIG.1

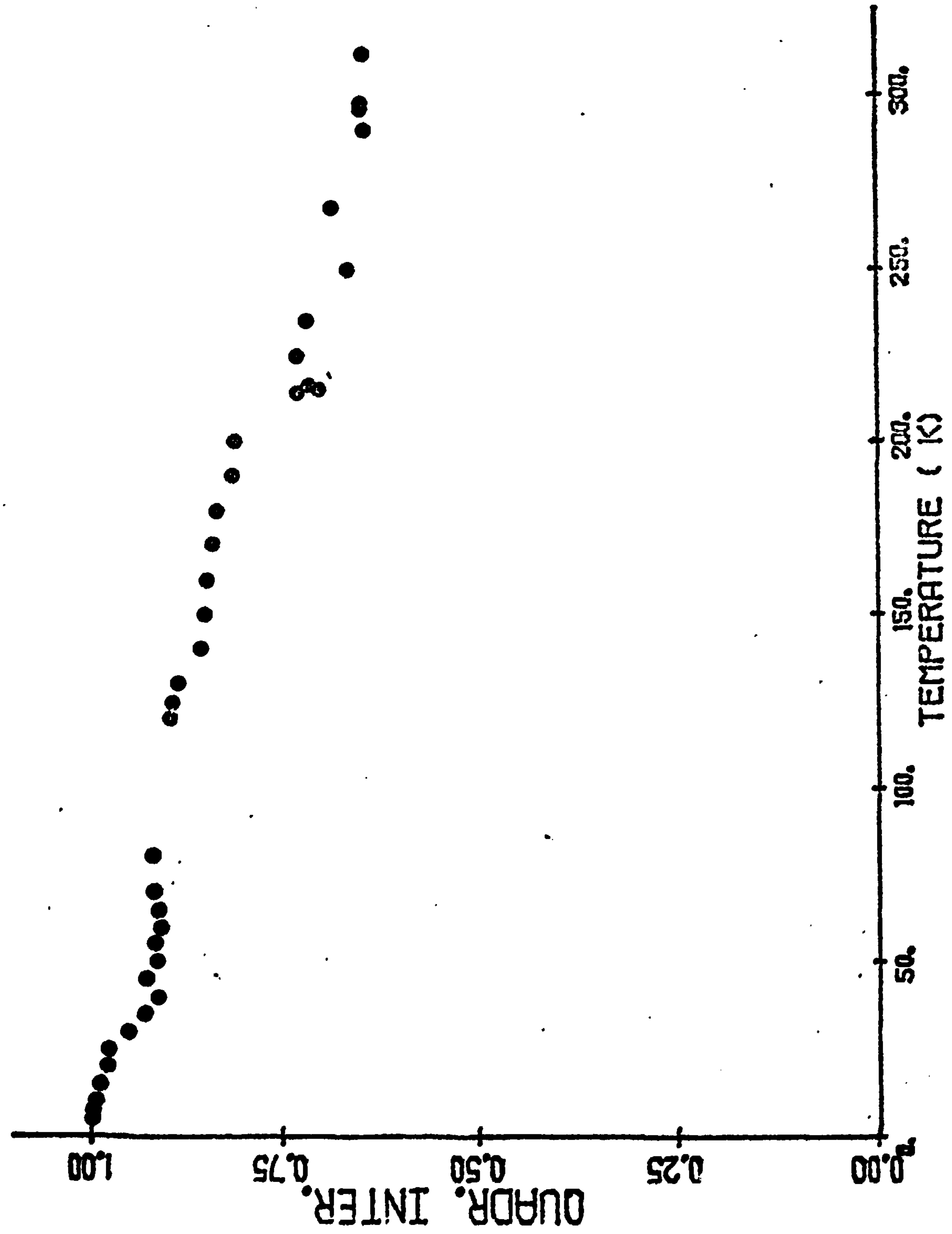
1.00
0.75
0.50
0.25
0.00
QUADR. INTER.
0.
50.
100.
150.
200.
250.
300.
TEMPERATURE (K)

FIG. 2