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Internal field measurements in Ni and Fe by the angular correlation method

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ABSTRACT

Radioactive In^{111} atoms were embedded into ferromagnetic Ni and Fe by thermal diffusion. The perturbed angular correlation of the gamma cascade in Cd^{111} was measured, giving the Larmor frequency ω due to the hf-interaction between the nuclear magnetic moment μ and the internal ferromagnetic field H. Measurements were done at 77, 300, 493 and 680 K for Ni and at 300 and 853 K for Fe. Both demagnetized and polarized samples were used. The internal fields were at room temperature -66.2 ± 1.4 kG for Ni and -349 ± 8 kG for Fe.

Introduction

Hyperfine interaction in magnetic materials has been subject to considerable interest during the last years. Several experimental techniques, some of them very recently developed, have been used and an amount of data has given an improved theoretical understanding of the interaction between electrons and nuclei. The various experimental techniques [1], among them the Mössbauer-effect, NMR and nuclear specific heat, are equally important as their ranges of particular problems do not always overlap. This is true also for the perturbed angular correlation (PAC) technique.

The study of hf interaction by PAC methods can serve two purposes. Either one can make use of more or less known hf-fields to determine nuclear g-factors or, vice versa, one can determine the hf-fields knowing the g-factor. This paper will present some details and results from PAC experiments of this second type. By time-differential technique we have measured the PAC of non-magnetic radioactive atoms which have been dissolved as impurities into the lattice of ferromagnetic materials. The Larmor frequency will then directly give the effective magnetic field at the site of the impurity nucleus. Preliminary results of these experiments have been published earlier [2].

The PAC of two gamma rays can be expressed as [3] (for notations see ref. [3])

$$W(\mathrm{k}_1\,\mathrm{k}_2\,t) = \sum_{k_1 k_2 N_1 N_2} A_{k_1}(1)\,A_{k_2}(2)\,G_{k_1 k_2}^{N_1 N_2}(t)\cdot[(2k_1+1)(2k_2+1)]^{-\frac{1}{2}} \times Y_{k_1}^{N_1 *}(\theta_1\varphi_1)\,Y_{k_2}^{N_2}(\theta_2\varphi_2), \tag{1}$$

where the influence of extranuclear fields is completely described by the perturbation factor $G(t)$. Eq. (1) represents the *time-differential* PAC. An experimental determination of eq. (1) requires that the resolving time τ_0 of the equipment is shorter than the nuclear lifetime τ_N of the intermediate level of the gamma cascade.

The details of the perturbation factor depend on the nature of the extranuclear field. E.g., in case of a classical, static magnetic field and with the detectors in a plane perpendicular to the field H, eq. (1) reduces to

$$W(\theta,t)=\sum_k A_k P_k[\cos(\theta-\omega_L t)], \tag{2}$$

where the Larmor precession frequency

$$\omega_L=g\cdot H\cdot\mu_N/\hbar. \tag{3}$$

The effect on the A.C. pattern is thus a rotation in a unique, defined sense and at a unique frequency.

The hf-fields in ferromagnetic materials are not known in such a detail that the perturbation factor can be expressed analytically with certainty. It is necessary to start with certain assumptions about the hf-fields and from there calculate the perturbation factor for a final comparison with experimental results. When dealing with the ferromagnetic materials as Fe and Ni we can assume that no electric crystal field gradients exist as we have a cubic crystal symmetry.

On the other hand, if the material is properly demagnetized, we have the existence of ferromagnetic domains, each one having its own internal field direction randomly oriented in space. As pointed out by Caspari [4] and by Matthias *et al.* [5] the randomly oriented field directions do not make a PAC-experiment impossible. With the assumption that the internal fields in ferromagnetic materials are purely static we can state that eq. (1) is valid for each domain separately. For a macroscopically demagnetized sample one can integrate eq. (1) over the various domain field directions, giving as the result [5] (W^0 stands for the demagnetized sample)

$$W^\circ(\theta,t)=\sum_k A_k G_k(t) P_k(\cos\theta), \tag{4}$$

where

$$\left.\begin{aligned} G_k(t)&=\frac{1}{2k+1}\sum_{N=-k}^{k}\cos(N\omega_L t),\\ \omega_L&=g\cdot \mathrm{H}_{\mathrm{INT}}\cdot\mu_N/\hbar. \end{aligned}\right\} \tag{5}$$

If the sample is polarized, i.e. all domain field directions are oriented by means of an external field, we are coming back to eq. (2), with the difference that

$$\omega_L=g(\mathrm{H}_{\mathrm{INT}}+\mathrm{H}_{\mathrm{EXT}})\cdot\mu_N/\hbar. \tag{6}$$

We denote the polarized AC-function as $W^{\uparrow}(\theta,t)$.

Equipment

The present work has been carried out with the time-differential coincidence technique. The main difference between time-differential and time-integral PAC-methods is the fact that in the first method it is possible to distinguish directly between static and dynamic components in the perturbing fields [6].

Photomultipliers (XP1020) and NaI-crystals were used as detectors. In order to obtain a fast response in combination with a low dark current from the photomultipliers a special arrangement [7] with two tunnel-diodes in the detector circuit was used. The first (low-biased) tunnel-diode was triggered by the "first" photoelectron, thus defining the time of the event. The second tunnel-diode (high-biased) was triggered at a level when the dark current pulses were negligible. The outpulses from the diodes were then, after proper time adjustment, put to a coincidence circuit, which finally defined the detector pulse. The time-pulse height converter (TPHC) was of the start-stop type using the charging of a condenser (designed by Hansén [7]). The total time-range could be varied from 1 μs to 10 ms, although in this experiment only the shortest range was used. The electronic time resolution was of the order of 50 ps. The overall time resolution (τ_0) for 511 keV was 1.4 ns.

For measurements at source temperatures higher than room temperature a bifilar tungsten wire was wound around an open end quartz tube. The wire itself was in vacuum in an evacuated bulb. The temperature inside the quartz tube was measured by a Pt–Pt 10 %Rh thermo-couple. The temperature was extremely constant and the error in the absolute value was estimated to be less than two degrees. At the 77 K measurement the sample (in an evacuated quartz tube) was simply dipped into liquid nitrogen in a conventional dewar. The absorption of the gamma rays in both heating bulb and dewar was negligible as checked by anisotropy measurements.

Source preparation

The non-magnetic "probe" atom used in this experiment was radioactive In^{111} decaying to Cd^{111}. The production and chemical separation of the radioactivity followed standard procedure [6].

The radioactive indium was electro-plated onto Ni resp. Fe metals. The diffusion was carried out in a high temperature oven under a low pressure argon atmosphere. The proper diffusion procedure was somewhat difficult to find, partly due to the low melting point of indium. The evaporation of the indium to the surrounding quartz tube wall increased with the temperature but seemed to be independent of diffusion time (3–12 hrs). The diffusion depth as measured by a microsond technique was proportional to both temperature and diffusion time. A diffusion depth of 120 μ was detected for 10 hrs and 1000°C. To start the procedure at room temperature or to put the metal directly into the hot oven did not seem to have any influence. It seemed as if the electroplating process itself could influence the result.

The optimal conditions for indium in Ni turned out to be 12 hrs diffusion at 1000°C followed by an immediate "quenching" of the process by cold water. The solubility of indium in Fe is much poorer than In in Ni. In order to get any appreciable activity diffused in Fe it was necessary to use a relatively large area of Fe-sheet of thickness 0.1 mm. After the diffusion the surface layer of the samples were solved in concentrated HCl in order to remove the activity which had not diffused into proper sites.

Methods of measurements

The 173–247 keV cascade in Cd^{111} was used for the PAC-measurement. The intermediate level has a lifetime of $\tau_N = 123$ ns and a g-factor of -0.318 ± 0.007 [8]. Both

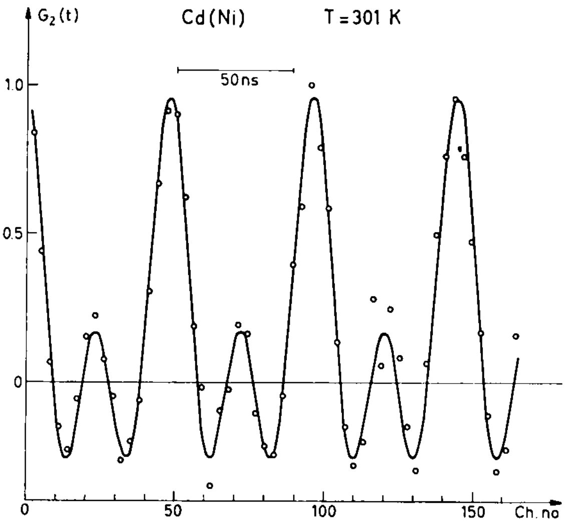

Fig. 1. Experimental $G_2(t)$ at room temperature for Cd(Ni). The full line is the L.Sq.F.

demagnetized samples giving $W^0(\theta, t)$, and polarized samples giving $W^{\uparrow}(\theta, t)$ were used. The A_4-term in the AC function of this Cd-cascade is very small and can be neglected.

Eq. (2) can therefore be expressed as

$$W^{\uparrow}(\theta, t) = 1 + A_2 P_2[\cos(\theta - \omega_L t)], \tag{7}$$

eq. (4) as

$$W^0(\theta, t) = 1 + A_2 G_2(t) P_2(\cos\theta), \tag{8}$$

and eq. (5) as

$$G_2(t) = \tfrac{1}{5}[1 + 2\cos\omega_L t + 2\cos 2\omega_L t]. \tag{9}$$

Out of these equations we can make some remarks as to a comparison between the demagnetized and the polarized methods. The experimentally recorded coincidence rate is given by

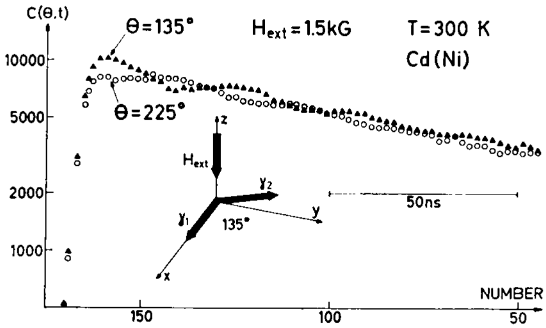

Fig. 2. Cd(Ni), room temperature, sign-of-field-experiment. $H_{ext} = 1.5$ kG (ex. of eq. (11): $F_A = -1$, $F_C = +1$ $(\theta = 135°)$, $F_S = +1$ $(\theta = 135°)$, $F_g = -1$, $F_H = -1$, thus $F_{INT} = -1$).

Table 1

Host lattice	T (K)	$B(T)$ (kG)	T/T_c
Ni	77.4	-68.0 ± 1.6	0.123
($T_c = 631$ K)	300	-66.2 ± 1.4	0.473
	493	-51.4 ± 1.3	0.782
	680	0	1.078
Fe	300	-349 ± 8	0.288
($T_c = 1043$ K)	853	-282 ± 6	0.818

$$C(\theta, t) = \text{const} \cdot e^{-t/\tau_N} \cdot W(\theta, t), \tag{10}$$

i.e. a normal decay curve modulated by the AC function. For W^0 this modulation has two frequences, ω_L and $2\omega_L$, resulting in large amplitudes at frequency ω_L. For $W^{\uparrow}$ there is only *one* frequency, but due to the quadratic terms in the Legendre polynomial P_2 the apparent frequency is $2\omega_L$. The very strong internal fields that usually exist in ferromagnetic materials may therefore result in such high frequencies that the time resolution is not sufficient for the equipment to resolve $2\omega_L$-modulations, while it is sufficient for ω_L-modulations. It may thus be necessary to use demagnetized samples only. On the other hand one cannot obtain any information about the sign of the internal field without using a polarization of the domains.

With demagnetized samples it is most convenient to measure at $\theta = 180°$ and $90°$ and then calculate the experimental $G_2(t)$ as shown in ref. [6]. The determination of ω_L is then carried out by a least square fit to a theoretical $G_2(t)$, taking the finite resolving time τ_0 into consideration.

For sign determination the polarization of the sample is achieved by applying a relatively weak external field H_{ext} (a few kG) perpendicular to the plane of the

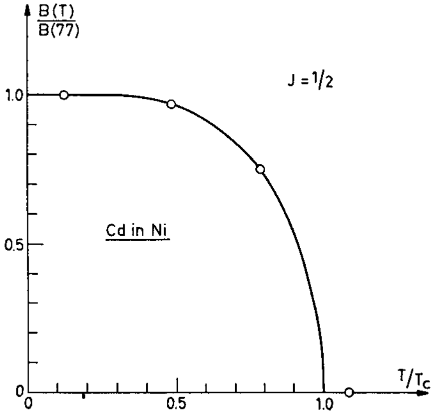

Fig. 3. Temperature dependence of the internal field at Cd in Ni. The full line is the Weiss-theory for Ni (J = ½).

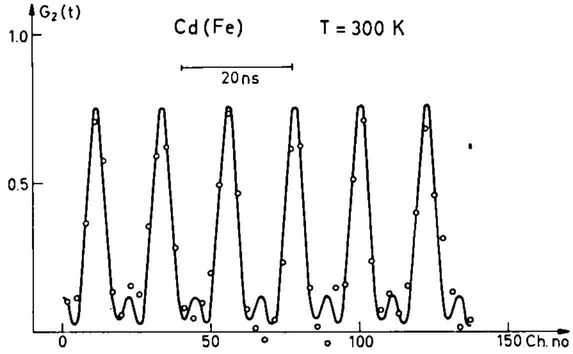

Fig. 4. Experimental $G_2(t)$ at room temperature for Cd(Fe). The full line is the L.Sq.F. The asymmetry of the pattern is due to a slight asymmetry in the time resolution curve.

detectors. In principle one measurement is sufficient at $\theta = 135°$ but a more unambiguous result is obtained by reversing the external field direction or by taking also $\theta = 225°$.

A simple rule of thumb for the sign determination can be given as follows. The various details and physical parameters for the actual experimental arrangement can be represented by symbolic factors F_i taking values $+1$ or -1.

F_A $= +1$ (-1), if the anisotropy is $A > 0$ $(A < 0)$.
F_{int} $= +1$ (-1), if the internal field is parallell (antiparallell) to the axis of quantization, i.e. positive z-direction.
F_C $= +1$ (-1), if the modulation of the coincidence rate at time $= 0$ starts to increase (decrease).
F_S $= +1$ (-1), if the coordinate system defined by the directions of γ_1, γ_2 and the positive z-direction is positive (negative).
F_g $= +1$ (-1), if the g-factor is positive (negative).
F_H $= +1$ (-1), if the external field is antiparallell (parallell) to the positive z-direction (it is assumed that the magnitude of the internal field is larger than the external field).

These factors are related through the eq. (11):

$$F_{\text{int}} = F_A \cdot F_C \cdot F_S \cdot F_g \cdot F_H. \tag{11}$$

According to the definition the internal field is then positive (negative) if $F_{\text{int}} = +1$ (-1) (see Fig. 2). It can sometimes be difficult to determine F_C properly. This will be commented on in connection with Fig. 5.

Results

By diffusing radioactive In, which decays to Cd, in the ferromagnetic material it is thus the effective field at the site of a Cd-nucleus we measure. The half-life of the upper level of the gamma-cascade is 0.12 ± 0.03 ns [9] and the Cd atoms is in a metallic

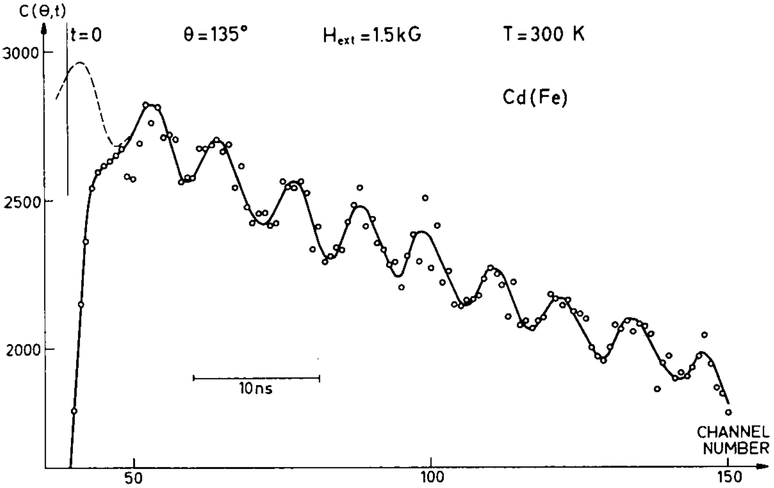

Fig. 5. Cd(Fe), room temperature, sign-of-field-experiment. In order to determine F_C in eq. (11) it was necessary to extrapolate the modulation back to time = 0, which was determined from a prompt curve.

environment. It can therefore be assumed that no after-effects of the K-capture would be observed in the experiment.

(*a*) Cd *in* Ni. The internal fields were determined at four temperatures, from 77.4 K to 680 K. The Curie temperature of Ni is $T_C = 631$ K. Fig. 1 shows the experimental $G_2(t)$ at room-temperature together with the theoretical curve. As can be seen there is no damping of the amplitude of $G_2(t)$. In our earlier publication [2] we published a corresponding curve which did show a damping effect. We interpreted this damping as a possible existence of a time-dependent perturbation. However, in the experiment by Matthias *et al.* [5] no such damping was found. We therefore repeated our experiment with special care to the temperature constancy and our new result is shown in Fig. 1. We believe now that our earlier results were due to temperature variation of the sample.

With a polarized sample in an external field of 1.5 kG the sign of the field in Ni was determined to be negative according to Fig. 2.

In Table 1 is shown the temperature dependence of the internal fields. Corrections for Lorentz fields and demagnetizing fields have not been done.

In Fig. 3 the temperature dependence is compared with the saturation magnetization for Ni according to the Weiss-theory. The proportionality seems to be valid over the whole temperature range.

(*b*) Cd *in* Fe. The effective internal fields at room temperature (Fig. 4) and 853 K were determined for demagnetized samples. The Curie temperature of Fe is $T_C = 1043$ K.

The sign determination experiment, giving a negative Fe-field, is represented in Fig. 5. As mentioned earlier the determination of F_C is in this case somewhat difficult due to the fact that the precession period and the time resolution are of the same order of magnitude. In the figure we have sketched the procedure in which the modulation pattern is extrapolated back to time = 0 in order to see what really happens. The results are given in Table 1.

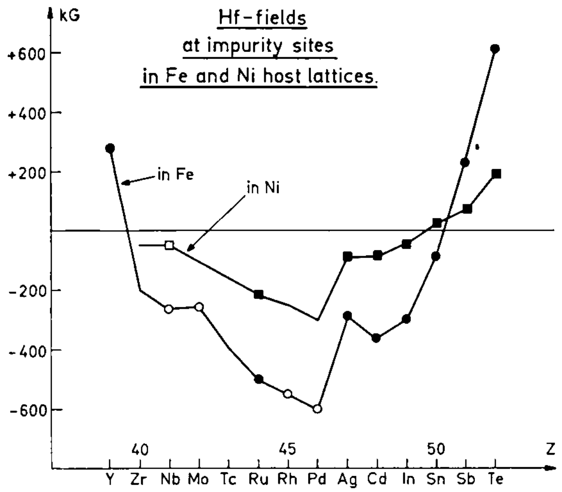

Fig. 6. Internal magnetic fields at the sites of nuclei $Z = 39$–52 in Fe and Ni host lattices. Data from Shirley [11]. Unfilled points represent cases for which the sign of the field has not been determined experimentally

Discussion

The results obtained for Ni are in good agreement with those of Matthias *et al.* [5]. The Fe-fields at room-temperature agrees also with results by Matthias [10], whereas we have not found in the literature measurements at other temperatures or sign determination for Fe.

The origin of the induced hyperfine-fields at non-magnetic nuclei in ferromagnetic materials are not fully understood quantitatively. A qualitative picture has been given in a review article by Shirley [11] as follows. In an external field the $3d$-electrons in the transition metals are polarized. By exchange polarization these $3d$-electrons polarize the "inner" $2\ s$ core-electrons and the "outer" $4\ s$ conduction-electrons. This may be regarded as a distortion of the radial wave functions of these s-electrons in such a way that the s-electrons with their spin-substates parallell to the $3d$-spins are "attracted" towards the $3d$ wave-function and vice versa for the antiparallell case. This results in a net spin density at the nucleus which for $2\ s$-electrons will be positive and for $4\ s$-electrons will be negative. The resulting field due to the Fermi contact interaction is then negative resp. positive. The relative strength between the two components gives the sign and magnitude of the total field acting. Other contributions beside the Fermi contact terms are negligible for $3d$-elements.

In the recent years quite a few internal fields have been determined by various methods. The fields vary in a systematic way with the atomic number. Fig. 6 shows some results in the region of interest for this investigation. The values are taken from the review article by Shirley [11].

The merits of the PAC-method for the determination of hf-fields are several. The accuracy is comparable with that of the Mössbauer technique and NMR. The main advantage of the PAC-method is, however, that it is a complementary method giving results where no other methods work. Compared with the Mössbauer method one deals with quite different impurity atoms. Compared with the NMR method we can accept a much lower concentration of the impurity atoms in the host lattice.

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REFERENCES

1. Freeman, A. J., and Watson, R. E., *in* Magnetism (Rado-Suhl), II A, ch. 4. Academic Press, New York, 1965.
2. Cisneros, J. I., Guimaraes, A. P., Liljegren, G., Lindqvist, T., and Lopez-Garcia, A., Physics Letters *21*, 245 (1966).
3. Steffen, R. M., and Frauenfelder, H., *in* Perturbed Angular Correlations (E. Karlsson, E. Matthias and K. Siegbahn), ch. 1. North-Holland Publ. Co., Amsterdam, 1964.
4. Caspari, M. E., Frankel, S., and Wood, G. R., Phys. Rev. *127*, 1519 (1962).
5. Matthias, E., Rosenblum, S. S., and Shirley, D. A., Phys. Rev. Letters *14*, 46 (1965).
6. Salomon, M., Boström, L., Lindqvist, T., Matthias, E., and Perez, M., Nucl. Phys. *46*, 377 (1963).
7. Hansén, J., private communication.
8. Matthias, E., Boström, L., Maciel, Alice, Salomon, M., and Lindqvist, T., Nucl. Phys. *40*, 656 (1963).
9. Sparrman, P., Sundström, T., and Lindström, J. O., Arkiv för Fysik *26*, 479 (1964).
10. Matthias, E., private communication.
11. Shirley, D. A., Ann. Rev. Nucl. Sci. *16*, 89 (1966).

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