Measurements of the Magnetic Hyperfine Fields at Hg in Fe, Co, and Ni Hosts by the e^--γ Time-Differential Perturbed-Angular-Correlation Method

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The ^{197m}Hg radioactivity was implanted into pure-Fe, -Co, and -Ni hosts with the help of an isotope separator. These ferromagnetic hosts produce strong magnetic hyperfine interactions at the nuclear site of the Hg atoms. These hyperfine interactions were studied by time-differential measurements of the 165-keV L-conversion e^-–134-keV γ angular correlation. Measurements with both nonmagnetized and magnetized foils were performed. By use of the quite accurately known g factor of the 134-keV state of ^{197}Hg, the magnetic hyperfine fields $|H_{int}| = 692 \pm 55$, 483 ± 40, and 103 ± 8 kG for the Fe, Co, and Ni lattices, respectively, were deduced.

I. INTRODUCTION

The internal fields of Hg embedded in Fe, Co, and Ni have previously been investigated by several authors.[1-6] In three cases[1-3] the integral perturbed $\gamma-\gamma$ angular-correlation method has been used with ^{198}Hg as probe. With this method systematic errors may have occurred; for example, it is impossible to determine whether all Hg nuclei occupy identical lattice sites. In addition, the g factor of the first excited 2^+ state of ^{198}Hg ($g = 0.55 \pm 0.11$) is only moderately well known.[7] In two time-differential measurements[4,5] the spin rotation of $\gamma-\gamma$ cascades in ^{199}Hg was observed. The accuracy of these measurements is limited by the short half-life of the intermediate 158-keV state of $T_{1/2} = 2.5$ nsec. However, the results of the integral and time-differential measurements do not agree very well. Because of these discrepancies and difficulties we looked for a more appropriate method for an accurate determination of the magnetic hyperfine fields of Hg in Fe, Co, and Ni. We found that the $\frac{5}{2}^-$ 134-keV state of ^{197}Hg (see Fig. 1) is an ideal level for hyperfine investigations because it has a sufficiently long half-life ($T_{1/2} = 7.0$ nsec) and because its g factor ($g = 0.380 \pm 0.025$)[8] is quite accurately known. The 23.8-h isomeric $\frac{13}{2}^+$ state of ^{197}Hg is almost exclusively depopulated by a cascade consisting of the 165-keV $M4$ transition and the 134-keV $E2$ line. The large theoretical[9] angular-correlation coefficient ($A_2^{theory} = 0.2207$) of this cascade makes it especially attractive for a time-differential investigation of hyperfine interactions. The conversion coefficients of the 165-keV $M4$ transition are very large ($\alpha_K = 70$, $\alpha_{L_{tot}} = 173$, Ref. 10) and strongly suggest observing the $e^--\gamma$ rather than the $\gamma-\gamma$ correlation. The particle parameter[11] for the 165-keV L-conversion lines is $B_2(L_{tot}) = 0.97$. Thus we expect an only slightly smaller anisotropy compared to a $\gamma-\gamma$ experiment. $e^--\gamma$ angular correlations can of course only be observed with extremely thin sources. Therefore it is not possible to alloy the radioactivity with the host metals. An ideal method is the implantation of the radioactivity by an electromagnetic isotope separator. For an implantation energy of, e.g., 70 keV, the implantation depth is less than 100 Å. For such a thin source no attenuation of the 165-keV L $e^--\gamma$ angular correlation is to be expected according to the estimates given in Ref. 12.

Our aim in this investigation was twofold: (i) We hoped to derive the internal fields of Hg embedded in Fe, Co, and Ni with high accuracy and reliability. (ii) We wanted to demonstrate that hyperfine fields can indeed be measured by observing the time-differential perturbation of $e^-\gamma$ correlations. To our knowledge $e^-\gamma$ angular correlations had been used to study attenuations due to after effects (e.g., Ref. 13) or integral spin rotations (e.g., Ref. 14) but up to recently not for time-differential investigations of internal fields. When our measurements were already in progress a publication by Kornienko *et al.*[6] appeared, in which a time-differential measurement with ^{197m}Hg in Ni was reported. In this work the value of the magnetic hyperfine field of Hg in Ni from Ref. 1 was used to redetermine the g factor of the 134-keV state in ^{197}Hg.

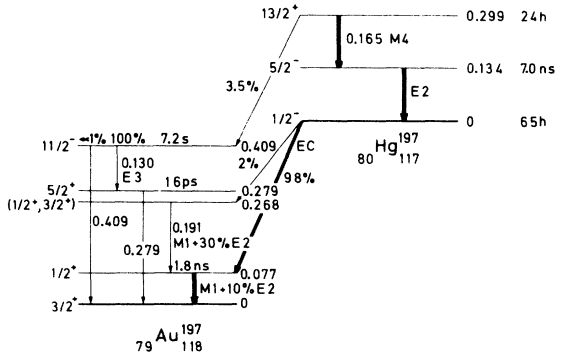

FIG. 1. Decay scheme of ^{197m}Hg.

II. EXPERIMENTAL PROCEDURES

The 23.8-h radioactivity ^{197m}Hg was produced for some runs by about 10-h irradiations of metallic Pt foils of natural abundance with 42-MeV α beams. In the ion source of the electromagnetic isotope separator of our laboratory, the Hg produced in the (α, xn) reactions was boiled out of the irradiated Pt foils. The atoms of mass number 197 (^{197m}Hg + ^{197}Hg) are implanted into the ferromagnetic hosts. For other runs naturally abundant Hg oxide was irradiated for 24 h in a thermal neutron flux of 8.10^{13} n/cm^2 sec. The Hg oxide disintegrates in the ion source of the separator and again ^{197m}Hg and ^{197}Hg are implanted into the ferromagnetic foils. From the implantation energy of 70 keV and the measured dose of Hg atoms collected in the Fe, Co, and Ni foils the maximal concentration of implanted atoms was estimated to be between less than 0.3 and about 2.0 at.% for the different sources. This dose is mainly due to the neighboring stable Hg isotopes which are not suppressed completely by the separation process. The host foils of Fe, Co, and Ni were 5–10μ thick and were bought from Goodfellow Metals, England. We used them in the isotope separator without any cleaning procedure. After the implantation these foils were used directly as sources for our $e^- - \gamma$ angular-correlation measurements without further treatment.

For the detection of the electrons we used two magnetic-lens spectrometers of the type described in Ref. 15. They are placed at an angle of 90°. The source is positioned at the intersection of the two spectrometer axes at an angle of 45° with respect to these axes in such a way that both lenses look at the irradiated side of the foil. The electrons are focused onto plastic scintillators which are coupled to *XP* 1021 photomultipliers. The energy resolutions used were about 3% at roughly 5% transmission.

A 38×26-mm plastic scintillator in connection with a RCA 8850 photomultiplier served as detector for the 134-keV γ rays. An energy gate was set on the γ spectrum such that less than 10% of the only strong transition in the decay of ^{197}Hg of 77 keV was accepted. The γ detector was moved every 2000 sec automatically between the two positions at 90° and 180° with respect to one magnetic spectrometer. Employing constant-fraction timing discriminators, we obtained time resolutions of 1 nsec full width at half-maximum (FWHM) at energies corresponding to the 165-keV L conversion and 134-keV γ lines. The time distributions obtained for both combinations of the γ detector with each electron spectrometer and the two relative angles were routed into four separate subgroups of a 4096-channel analyzer.

In two runs both a Ni and a Co foil were magnetized perpendicular to the plane of the detectors with the help of a small permanent magnet which produced about 0.5 kG. This magnet was mounted inside the vacuum chamber. It had no noticeable influence on the electron-energy spectrum or the time-resolution curve. In order to improve the degree of magnetization the Co foil was placed for a short time in a field of about 2 kG produced by a larger permanent magnet before it was attached to the pole tips of the 0.5-kG magnet.

III. RESULTS

Usually two or three independent measurements with different sources embedded in unmagnetized foils were performed for each host lattice. The concentration of the Hg impurities varied sometimes as much as a factor of 10 from source to source. In the individual low-statistic data no significant differences in the frequencies could be observed. On the other hand, the statistical accuracy of the data obtained in the separate runs for one detector combination was not sufficient to reliably determine whether, in addition to the main frequency, other interactions occurred. We therefore combined the data of all runs for each host lattice. In Figs. 2–4 linear plots of the asymmetry ratios

$$R(t) = 2\frac{N(180°, t) - N(90°, t)}{N(180°, t) + N(90°, t)}, \tag{1}$$

calculated from the combined data as a function of time t, are shown for the Fe, Co, and Ni host lattices, respectively. The vertical bars reflect the statistical uncertainties. The asymmetry ratio $R(t)$ can be shown to reduce approximately to $\frac{3}{2}$ of the product of the angular-correlation coefficient A_{22} and the perturbation factor $G_2(t)$ if A_{44} equals zero, which is true for the 165–134-keV cascade[9]

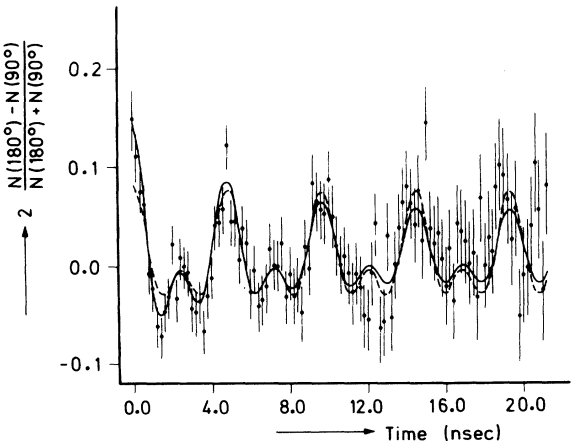

FIG. 2. Spin rotation for Hg embedded in a randomly oriented Fe host. The broken and solid lines are results of least-squares fits using the functions of Eqs. (2) and (4), respectively.

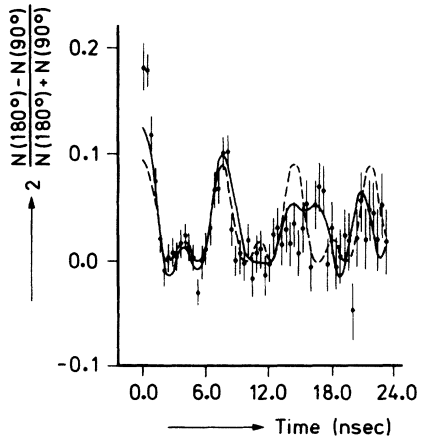

FIG. 3. Same as Fig. 2 caption for Hg in a Co Host.

in ^{197}Hg. The perturbation factor $G_2(t)$ for complete random orientation of the magnetic domains is[16]

$$G_2(t) = \tfrac{1}{5}(2\cos\omega_L t + 2\cos 2\omega_L t + 1)\,, \tag{2}$$

where

$$\omega_L = -g\mu_N B/\hbar \tag{3}$$

is the Larmor precession frequency.

Fits of this perturbation function to the experimental data, taking the shape of the time distribution for prompt coincidences into account, are indicated by broken lines in Figs. 2–4. In addition to A_{22}^{expt} and ω_L we allowed the constant term $C = 1$ in Eq. (2) to vary. The parameters derived are summarized in the first data column of Table I. The errors quoted for the frequencies are mainly due to estimated uncertainties in the time calibration. The statistical uncertainties for ω_L amount to only about 1% in all cases. The values χ^2 listed in Table I measure the quality of the fit. While the frequencies are determined quite accurately, it is obvious from Figs. 2–4 that these fits do not reproduce the experimental data well. The reason is apparently that the experimental data exhibit a damping of the spin-rotation curves. A look at Fig. 2 shows that the amplitude of the spin-rotation curve decreases rapidly within the first period but then remains essentially constant. This suggests trying to fit with the following perturbation function:

$$G_2(t) = \tfrac{1}{5}f(2\cos\omega_L t + 2\cos 2\omega_L t + 1) + [\tfrac{1}{5}(1-f)]e^{-\lambda t}(2\cos\omega_L' t + 2\cos 2\omega_L' t + 1)\,. \tag{4}$$

This perturbation function can be derived from the following model for the interactions in the lattices. A fraction f of nuclei comes to rest after the implantation on substitutional lattice sites where they experience only fields of uniform strength. The remaining nuclei occupy lattice positions where different hyperfine fields interact with them. The factor $e^{-\lambda t}$ describes approximately a frequency distribution of Lorentzian shape for this static perturbation.

The modified perturbation function clearly improves the fits (solid lines in Figs. 2–4). The statistical accuracy of the data is not sufficient to make this particular choice of modification compel-

TABLE I. Parameters derived from fits for unmagnetized foils.

Parameter	Eq. (2)	Eq. (4)
	^{197}Hg Fe	
A_{22}	0.074 ± 0.006	0.15 ± 0.07
ω_L	1305 ± 40 MHz	1296 ± 40 MHz
C	0.33 ± 0.15	0.2 ± 0.1
f		0.34 ± 0.2
λ		(0.25 ± 0.14) × 10^9 sec^{-1}
ω_L'		$= \omega_L$ [a]
χ^2	1.03	0.98
	^{197}Hg Co	
A_{22}	0.065 ± 0.008	0.09 ± 0.02
ω_L	865 ± 30 MHz	905 ± 30 MHz
C	1.5 ± 0.3	1.1 ± 0.3
f		0.55 ± 0.15
λ		0.0[a]
ω_L'		780 ± 30 MHz
χ^2	1.68	1.35
	^{197}Hg Ni	
A_{22}	0.116 ± 0.008	0.16 ± 0.02
ω_L	182 ± 6 MHz	191 ± 6 MHz
C	0.53 ± 0.13	−0.3 ± 0.3
f		0.56 ± 0.10
λ		(0.052 ± 0.036) × 10^9 sec^{-1}
ω_L'		66 ± 26 MHz
χ^2	1.53	1.03

[a]Fixed at this value.

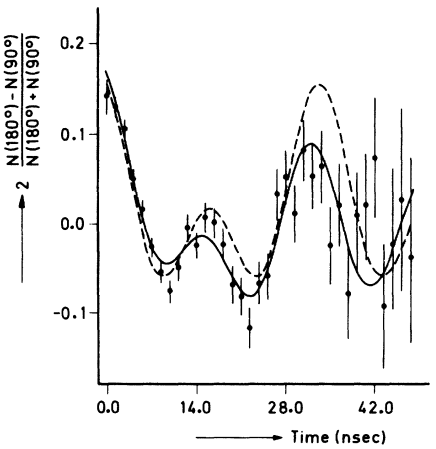

FIG. 4. Same as Fig. 2 caption for Hg in a Ni host.

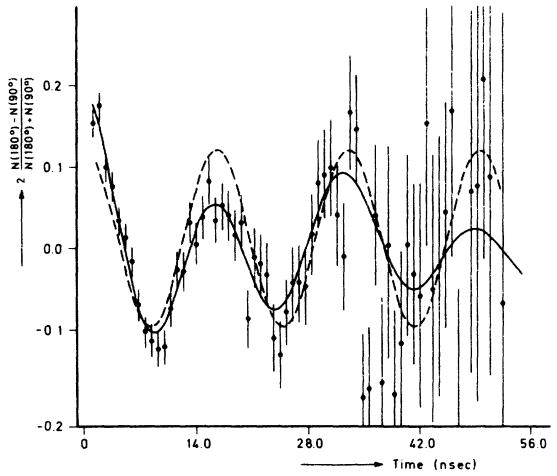

FIG. 5. Spin rotation for Hg embedded in a partially magnetized Ni host. The broken and solid lines are results of least-squares fits using the functions of Eqs. (5) and (6), respectively.

TABLE II. Parameters derived from fits for magnetized foils.

Parameter	^{197}Hg in Co Eq. (5)	Eq. (7)
A_{22}	0.062 ± 0.005	0.17 ± 0.02
ω_L	(917 ± 30) MHz	(920 ± 30) MHz
C'	0.003 ± 0.003	−0.001 ± 0.002
f_1		0.67 ± 0.07
f_2		0.46 ± 0.07
λ		(4.2 ± 1.1) × 10^9 sec^{-1}
ω_L'		(827 ± 30) MHz
χ^2	1.23	1.04
Parameter	^{197}Hg in Ni Eq. (5)	Eq. (6)
A_{22}	0.076 ± 0.005	0.15 ± 0.01
ω_L	(190 ± 6) MHz	(190 ± 6) MHz
C'	0.013 ± 0.005	0.26 ± 0.15
f'		0.34 ± 0.05
λ		(2.2 ± 0.5) × 10^7 sec^{-1}
χ^2	1.17	0.77

ling. The parameters derived with the perturbation function of Eq. (4) are also listed in Table I.

Plots of the asymmetry ratios $R(t)$ for our measurements with the magnetized Ni and Co foils are shown in Figs. 5 and 6. The perturbation function for complete polarization is

$$G_2(t) = \cos 2\omega_L t \ . \tag{5}$$

This perturbation function plus an additive constant C' for compensation of possible experimental asymmetries was fit to the data.

The result for Hg in Ni is indicated by the broken line in Fig. 5. Again the simple function of Eq. (5) does not describe the experimental data sufficiently. In the case of the Ni host, the second maximum experimentally is lower than the first and third ones. This has to be taken as an indication that the external field was not large enough to magnetize the foil completely. This is not too surprising since we deliberately chose the field strength of the permanent magnet somewhat low in order to keep the stray fields small which influence the conversion electron trajectories. Partial polarization of the Ni foil would lead to the perturbation function

$$G_2(t) = f' \cos 2\omega_L t + \frac{(1-f')}{5}(2\cos\omega_L t + 2\cos 2\omega_L t + 1)e^{-\lambda t} \ , \tag{6}$$

if interference terms in the angular-correlation function of type G_{24} can be neglected. The exponential factor has been added in order to allow for some frequency distributions. The solid line in Fig. 5 is the result of the fit of the function of Eq. (6), again with an additive constant C' to the data.

In the case of the Co host the amplitude decreases in the first part of the curve, but for longer delay times than 20 nsec it seems to increase again. If this increase of amplitude is really significant it may be explained by the interference of two neighboring interaction frequencies. We tried to fit the data obtained with the magnetized Co foil using the perturbation function

$$G_2(t) = [f_1 \cos 2\omega_L t + (1-f_1)\cos 2\omega_L' t] \times [f_2 + (1-f_2)e^{-\lambda t}] \ . \tag{7}$$

This perturbation function reflects the following model. A fraction f_1 of the Hg nuclei occupy a different lattice site than the remaining nuclei. In both locations $(1-f_2)$ nuclei experience a frequency distribution causing a damping of the amplitude of the spin-rotation curve. The solid line in Fig. 6 is a least-squares fit of the function of Eq. (7) again

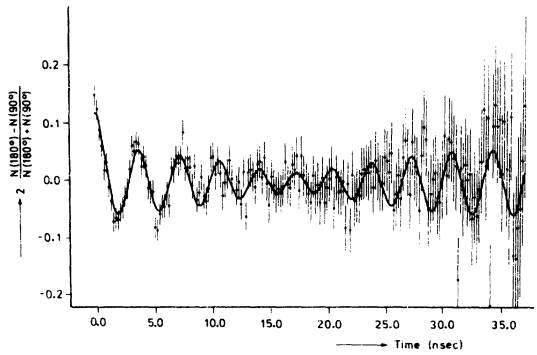

FIG. 6. Spin rotation for Hg embedded in a magnetized Co host. The solid line is a result of a least-squares fit using the function of Eq. (7).

with an additive constant C' to the data. The parameters derived from the fits with Eqs. (6) and (7), which clearly improve the quality of the fit, are summarized in Table II. Again the errors quoted for the frequencies are mainly due to uncertainties in the time calibration.

The parameters obtained in our fits need some discussion. The values for the constant term in the perturbation functions scatter between -0.3 and $+1.5$ for the unmagnetized foils, while values of 1.0 are expected. There are two possible reasons for the observed deviations:

(i) If the magnetic domains do not have completely random orientations the interaction frequencies remain unchanged but the relative amplitudes of the cosine functions and the constant term vary.

(ii) The finite-source dimensions (3-mm diameter) and slight misadjustments of the sources relative to the electron spectrometers are also expected to produce effects of the observed magnitude.

Corrections for the geometrical effects seem very difficult and unreliable at present and have not been applied. We therefore can not decide which is the correct interpretation of this phenomenon.

The experimental A_{22} values obtained with the fits using the functions of Eqs. (2) and (5) are a factor of 2 below the theoretical prediction. The A_{22} parameters derived with the refined functions (4), (6), and (7) are consistently around 0.16, except for the measurement with unmagnetized Co foils where it is obvious, from Fig. 3 and the large χ^2 value, that this fit is the worst of all and particularly bad for data points around $t=0$. Application of the correction for the finite solid angles of both the γ and e^- detectors and possibly small amounts of backscattered electrons result in perfect agreement of those larger experimental A_{22} coefficients with theory.

The fraction f of nuclei that occupy regular sites with no additional perturbation is only about 50% in all cases. This value seems quite reasonable since perturbations are expected because of the high-impurity dose produced by the ion implantation. Furthermore, due to the low implantation energy of 70 keV an appreciable part of the nuclei is expected to come to rest in the surface layers.

The second frequency ω_L', which was fitted for the unmagnetized Ni foil, but which apparently did not appear in the magnetized Ni foil, is probably not significant. On the other hand, we think that the interference phenomena which we observed for both the randomly oriented and magnetized Co foils definitely exist. They may be produced by two different but well-defined positions of the Hg atoms in the Co lattice.

It should be mentioned, however, that as was shown by Boström *et al.*,[17] the superposition of an additional electric quadrupole interaction may produce spin rotation curves very similar to ours even if the electric interaction frequency is a factor of 20–30 smaller than the magnetic frequency ω_L [see, e.g., Figs. 4(b), 4(c), and 5 from Ref. 17]. An electric interaction is expected because for Co metal below 400 °C the hexagonal-close-packed (hcp) structure is the stable configuration.[18] In the following we shall estimate the order of magnitude one could expect for the quadrupole frequency.

From the lattice constants of the hexagonal phase at room temperature

$$c/a = 1.6228 \quad \text{and} \quad a = 2.5071 \text{ Å},$$

one calculates the lattice contribution to the effective electric field gradient according to the formula of Das and Pomerantz[19]:

$$V_{zz\,\mathrm{eff}}^{\mathrm{lat}} = -(28\times 10^{15}) Z \text{ V/cm}^2 .$$

This value includes the Sternheimer correction factor $(1-\gamma_\infty)$ with $\gamma_\infty(\mathrm{Hg}^{2+}) = -60.20$.[20] Inserting for the effective ionic charge of the Co atoms in Co metal

$$Z = 4 ,$$

and assuming an enhancement of the field gradient by the conduction electrons by a factor of 2–3 [21] we obtain an effective-field gradient on the order of

$$V_{zz}^{\mathrm{eff}} \approx 2.5\times 10^{17} \text{ V/cm}^2 .$$

TABLE III. Hyperfine fields of Hg in ferromagnetic hosts in kG.

Nucleus Method	^{198}Hg IPAC	^{198}Hg IPAC	^{198}Hg IPAC	^{199}Hg DPAC	^{199}Hg DPAC	^{197}Hg DPAC	^{197}Hg DPAC
Field in Fe	-440 ± 105	-490 ± 125	-980[a]	670 ± 65			692 ± 55
Field in Co	-370 ± 78						483 ± 40
Field in Ni	-86 ± 22				-124 ± 12	$(-)101\pm7$[b]	103 ± 8
Reference Source	1 molten	2 molten annealed	3 annealed	5 implanted	4 molten	6 diffused	This work implanted

[a]Revised by authors of Refs. 1 and 3 according to private communication to Ref. 5.

[b]Recalculated from ω_L given in Ref. 6 using g factor of Ref. 8.

The quadrupole moment of the $\frac{5}{2}^-$ state of the ^{197}Hg is not yet known. If we assume a value of

$$|Q| = 0.5 \text{ b},$$

which seems a reasonable value in comparison to the known quadrupole moments in the Hg region,[22] we obtain an estimated interaction frequency of approximately

$$\omega_0 \approx 30 \text{ MHz}.$$

This frequency might be large enough to produce a modulation of the angular-correlation pattern similar to the behavior which we observed. Investigation of the electric quadrupole interaction of ^{197}Hg in nonmagnetic hexagonal metallic lattices are in progress in order to check this hypothesis.

The magnetic interaction frequencies ω_L do not change significantly with different assumptions for the perturbation function and from experiment to experiment where the impurity dose is often quite different. For this reason, despite the irregular perturbation, the main magnetic hyperfine interaction frequencies are determined quite accurately and reliably. Using the measured[8] g factor $g = 0.380 \pm 0.025$ of the 134-keV level in ^{197}Hg, we calculate the hyperfine fields listed in the last column of Table III.

IV. DISCUSSION

In Table III we list all previously published data of hyperfine fields of Hg in ferromagnetic hosts together with our results.

The integral perturbed-angular-correlation (IPAC) measurements[1-3] for the ^{198}Hg in Fe consistently yielded fields of about 465 kG (see Ref. 5 for a revision of the value reported in Ref. 3) while the differential-perturbed-angular-correlation (DPAC) results (Ref. 5 and this work) for ^{199}Hg and ^{197}Hg implanted in Fe agree on fields around 680 kG. This discrepancy already led the authors of Ref. 5 to propose that the g factor measured for the first excited 2^+ state in ^{198}Hg may be in error. A crucial point in this argument was that both the implantation of Tl into Fe and the diffusion or melting process in order to embed the ^{198}Au radioactivity in Fe lead to a very high percentage of Hg atoms at substitutional sites. Channeling[23] and other experiments[24,25] performed by different authors had to be considered to prove this point.

The fact that our DPAC measurements revealed several irregular perturbations should not influence the reliability of the IPAC results for the first 2^+ state of ^{198}Hg because of its very short half-life. Therefore comparing all measurements with ^{198}Hg as probe and all measurements with ^{197}Hg or ^{199}Hg there now seems to be stronger support for the proposal that the g factor of the first excited state in ^{198}Hg is not correct, since the fields derived with the ^{198}Hg probe are systematically lower.

It can, however, not be excluded that a few percent of the Hg nuclei in the IPAC measurements[1-3] might have occupied grain boundary sites where they would not experience the full hyperfine field. Therefore, for a definite decision on this question, a new measurement of the g factor of the 2^+ state of ^{198}Hg would be very valuable.

Several authors[26-28] calculated the magnetic hyperfine fields of impurities in ferromagnetic host lattices. In general, systematic trends but not actual values in specific cases could be predicted accurately. Comparing our data to the phenomenological formula of Balabanov and Delyagin[29] we find good agreement for the hyperfine field of Hg in Fe but discrepancies for fields of Hg in Co and Ni hosts. The reason is that for Hg the experimental data are not proportional to the magnetic moments of the host lattices ($\mu/\mu_B = 2.2$, 1.7, and 0.6 for Fe, Co, and Ni, respectively). Often in other cases proportionality within 10–20% is observed.[29] We note that very similar deviations are observed[30] for the isoelectric Cd at room temperature. The hyperfine field of Cd in Ni has been measured[31] as a function of temperature. No significant deviations from the Curie law have been found. It may be interesting to perform such experiments for Hg in Fe, Co, and Ni hosts as well as for Cd in Fe. Possibly these investigations would indicate reasons for the observed discrepancies between hyperfine fields and magnetic moments.

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