Study of the Hyperfine Magnetic Field Acting on ^{181}Ta Nuclei in Ni Matrix*

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Hyperfine interactions for Ta impurities in a polycrystalline nickel matrix produced by quenching from the melt have been studied using the perturbed-angular-correlation technique. An effective hyperfine field of -89.8 ± 1.6 kOe was measured for substitutional atoms. The fraction of the impurities occupying various lattice positions, and the respective hyperfine fields have been determined, and changes in lattice location and hyperfine field after annealing of the source have been studied.

I. INTRODUCTION

Experimental investigation of nuclear magnetic hyperfine interactions on impurities diffused or implanted into ferromagnetic metals and alloys has developed very rapidly during recent years. The interpretation of the large number of data already available is difficult not only because we do not have a good theoretical approach, but also because in many cases we do not know where the impurity atom is located in the matrix.

The technique of differential perturbed angular correlation is very powerful in the investigation of hyperfine interactions involving nuclear levels with lifetimes longer than a few nanoseconds. The well known 133–482-keV γ-γ cascade in ^{181}Ta has been used intensively to study the interaction of the level at 482 keV, $T_{1/2} = 10.5$ nsec, in different solid and liquid environments. The internal magnetic field acting on ^{181}Ta in a Ni matrix was investigated initially by Vanderleeden,[1] and more recently by Cameron *et al.*[2] and Oddou *et al.*[3] Concerning the magnitude and the sign of the field in Ni^{181}Ta, the results of these authors are in agreement despite the different methods of preparation of the alloy, but there is a striking disagreement between the amplitude of the oscillating part of the angular correlation function reported by Vanderleeden and other authors. Besides this, the authors of Ref. 2 also noticed the presence of more than one frequency in the Fourier spectrum. Considering these facts, we understook additional experimental studies of the Ni^{181}Ta system.

II. EXPERIMENTAL PROCEDURES

The measurements described in this paper were performed using the differential perturbed-angular-correlation technique. The apparatus is a conventional fast-slow coincidence system with a time-to-pulse-heighth converter (TPHC). Using NaI(Tl) crystals, the time resolution of the electronic system at the 133 and 482-keV γ energies was 2.4 nsec.

The ^{181}Hf activity was obtained by neutron irradiation of natural metallic hafnium. The alloy was prepared by depositing 25 mg of active ^{181}Hf in a pot made of 1000 mg of 99.999%-pure nickel. The pot was sealed under vacuum with a tapered pin and then melted in an argon atmosphere, using an induction furnace and a levitating water-cooled-silver boat. In the next step the sample was successively attacked with acid showing decreases of activity proportional to mass decreases, thus ensuring uniform distribution of activity in the Ni matrix. The final sample was then cold worked into the form of a cylinder with 6-mm height and 5-mm diameter.

The alloy containing 1 at.% of ^{181}Hf impurity was placed between the pole tips of a minimagnet[4] and a field saturation curve was run for the source. The magnet was subsequently operated at the knee of the saturation curve, corresponding to an external aligning field of 1700 Oe. The measurements were performed at room temperature.

The first series of measurements was carried out with no annealing of the source. In a second series of measurements the source produced by the above procedure was annealed for four hours at 800 °C in a hydrogen atmosphere and subsequently attacked with acid to remove any activity which might have migrated to the surface. Less than 5% of the activity was lost during this step. The experimental results of the measurements are described in the Sec. III.

III. EXPERIMENTAL RESULTS

The angular correlation function in the presence of a magnetic field perpendicular to the plane of the detectors can be expressed as

$$W(\theta, H, t) = \sum_{k\ \text{even}} b_k \cos[k(\theta - \omega t)] e^{-t/\tau_N}. \quad (1)$$

The angular correlation function for an unpolarized ferromagnetic material consisting of randomly oriented domains, on the other hand, can be written as

$$W(\theta, H = 0, t) = \sum_{k\ \text{even}} A_k G_k(t) P_k(\cos\theta) e^{-t/\tau_N}. \quad (2)$$

In the case of interest only $k = 0$ and 2 contribute,

and we have

$$A_0G_0(t)=1,\ A_2G_2(t)=A_2(\tfrac{1}{5}+\tfrac{2}{5}\cos\omega t+\tfrac{2}{5}\cos 2\omega t) \tag{3}$$

with the Larmor precession frequency ω being given by

$$\omega=-\mu_N g H/\hbar. \tag{4}$$

The angular correlation coefficients A_2 and b_2, in our case, are related by

$$b_2\cong\tfrac{3}{4}A_2=-0.18. \tag{5}$$

Three different types of measurements have been made in the experiments reported here. In the first, the angle between the two detectors was held fixed and coincidences were measured for the external polarizing field up ($+H$) and down ($-H$). These results are analyzed in terms of the ratio

$$R(t)=2\,\frac{W(\theta,+H,t)-W(\theta,-H,t)}{W(\theta,+H,t)+W(\theta,-H,t)}\ , \tag{6}$$

which, using $b_4\ll b_2$ and $\theta=225°$ in (1), reduces to

$$R(t)\cong-2b_2\sin(2\omega t). \tag{7}$$

In the second arrangement the polarizing external field was maintained fixed perpendicular to the plane of the detectors and the coincidence time spectra were measured at angles of 90° and 180°. The data were analyzed in terms of the ratio

$$\epsilon(t)=2\,\frac{W(\pi,H,t)-W(\pi/2,H,t)}{W(\pi,H,t)+W(\pi/2,H,t)}\ . \tag{8}$$

Using (1) this ratio becomes

$$\epsilon(t)\cong+2b_2\cos(2\omega t). \tag{9}$$

Finally, measurements were made on an unpolarized source accumulating time spectra at angles of 90° and 180°. Using (2) and the approximation $A_4\ll A_2$, the data were analyzed by extracting the ratio

$$A_2G_2(t)=2\,\frac{W(\pi,t)-W(\pi/2,t)}{W(\pi,t)+2W(\pi/2,t)}\ , \tag{10}$$

where $G_2(t)$ is given by (3) above.

In principle, if only a single magnetic interaction were present, the above experiments would all determine the same information, the Larmor precession frequency ω due to the hyperfine field. The measurement of $R(t)$ permits the determination of the sign of the field as well as the magnitude. However, in more complicated situations where some of the ions in the source may feel a hyperfine field which is independent of the polarizing field—such as strong quadrupole interactions or magnetic interactions in unpolarized domains—the situation is different, since while such interactions appear in $\epsilon(t)$ and $A_2G_2(t)$ they will not appear in $R(t)$. Thus in cases where the impurities occupy inequivalent positions in the host matrix, measurements with different geometries can provide important additional information concerning the nature of the interactions involved.

In the present study all of the three ratios $\epsilon(t)$, $R(t)$, and $A_2G_2(t)$ were measured during different phases. In each case after formation of the appropriate ratio the data were treated both by Fourier analysis and nonlinear-least-squares-fitting techniques. For all measured spectra it was possible to fit the data assuming at the most three distinct frequencies. Reasonable agreement was obtained between the Fourier and nonlinear-least-squares results both for the amplitudes and the frequencies of the various Fourier components. Using Eqs. (3), (7), and (9), it was possible to estimate the percentage of the atoms that are in a lattice site corresponding to a certain hyperfine field from the measured amplitude of the respective frequency component. In order to derive this fraction the measured amplitudes were corrected for finite solid angle and for the finite time resolution of the system according to Eq. (3) of Ref. 5. The corrected values were compared to $2b_2=-0.36$ for $R(t)$ and $\epsilon(t)$ and $\frac{2}{5}A_2=-0.096$ for $A_2G_2(t)$ (except for $\omega=0$). The resulting ratio is interpreted as the fractional part of the atoms interacting through the respective frequency components.

The results of the measurements and the respective least squares fits are shown in Fig. 1. Figure 1(a) shows the measurement of $R(t)$ for the unannealed source, while Fig. 1(b) shows the result for $\epsilon(t)$ on the same source. Fig. 1(c) gives the results for the measurement of $A_2G_2(t)$ after annealing and before applying a polarizing field, while Fig. 1(d) shows $R(t)$ measured for the same sample.

The results of the nonlinear-least-squares fit are shown in Table 1. In each case the measured frequency components and respective fractions of full amplitude calculated as discussed above are given. The last column shows the sum of the amplitudes for all frequency components. [Note that in the measurement of $A_2G_2(t)$, f_1 and f_2 represent the same frequency component; the average of the two amplitudes was used in the final sum.] For each component the effective magnetic field acting on the respective nuclei was determined from (4). Hyperfine fields were then determined from the relation

$$H_{\rm hyp}=H_{\rm eff}-H_{\rm ext}-\tfrac{4}{3}\pi M_s+4\pi D M_s, \tag{11}$$

where $H_{\rm ext}$ is the external polarizing field, $4\pi M_s$ = 6900 Oe, the saturation magnetization of nickel, and D the demagnetization factor.

After these corrections, our results show that before annealing there are two different local magnetic hyperfine fields acting on Hf in a Ni host.

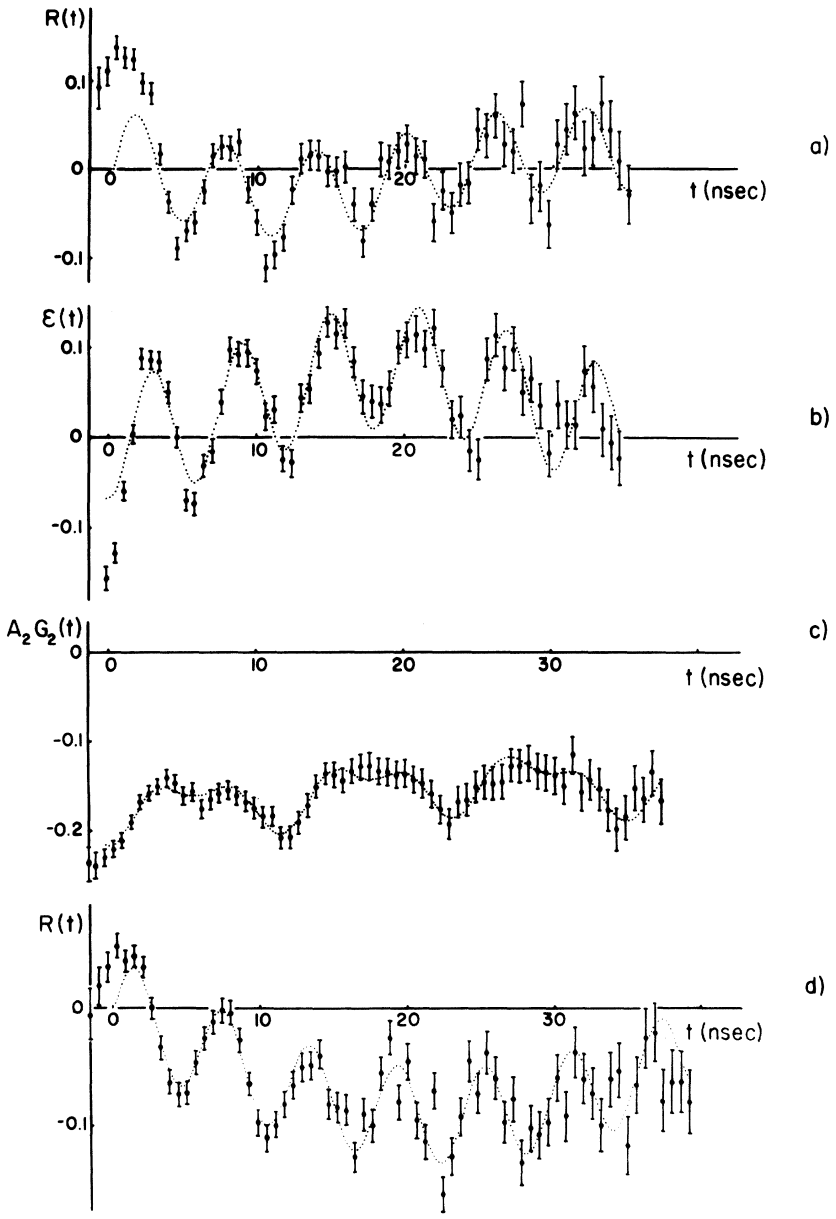

FIG. 1. Experimental time spectra for the four different measurements on Ta in Ni studied in this work. (a) and (b) show $R(t)$ and $\epsilon(t)$, respectively, for the unannealed source. (c) and (d) show $A_2G_2(t)$ and $R(t)$, respectively, after annealing. The dotted curves are nonlinear least-squares fits to the data, as described in the text.

One large and negative of -87.8 ± 1.6 kOe, and the other small and positive of 9.6 ± 1.0 kOe. After annealing the positive field disappears while the measured negative field remains unchanged.

IV. DISCUSSION

Previous experiments on Ta in nickel have been performed using polycrystalline sources produced by fusion and annealing,[1] and by implantation,[2] as well as both fused and implanted single-crystal sources.[3] Direct comparison of the various results is difficult because of the varying methods of source preparation, although in all experiments the components corresponding to a hyperfine field of about -90 kOe have been seen.

With the unique exception of the single-crystal source made by fusion[3] a common characteristic of previous measurements has been the fact that the observed amplitudes of $R(t)$ were much lower than should be expected theoretically. These experiments identified in general only one hyperfine field, although in the measurements of $\epsilon(t)$ by Cameron *et al.*[2] a second frequency not found in $R(t)$ was observed. According to the discussion above this frequency would have to be assigned to a nonmagnetic interaction of some type. It was not observed in the single-crystal experiments of Oddou *et al.*[3] nor in our experiments.

TABLE I. Summary of experimental results: column 1 shows the frequencies observed for each experiment; column 2, the derived hyperfine fields according to Eq. (11) of the text; column 3, the fraction of full amplitude for each observed frequency component calculated as described in the text; column 4, the fraction of full amplitude for each experiment, summing all observed frequency components. [The average of f_1 and f_2 was used to compute the total amplitude in column 4 for $A_2G_2(t)$.]

Measurement		1 Observed frequency (MHz)	2 Corrected hyperfine field (kOe)	3 Fraction of full amplitude	4 Total fraction of full amplitude for all components
Before annealing	$R(t)$ f_1	163.5	-87.1 ± 1.6	0.25	0.33
	f_2	27.6	$+10.0\pm1.0$	0.08	
	$\epsilon(t)$ f_1	166.3	$(-)88.6\pm1.6$	0.37	0.49
	f_2	26.1	$(+)9.3\pm1.0$	0.12	
After annealing	$A_2G_2(t)$ f_1	167.6	$(-)85.2\pm1.6$	0.29	1.11
	f_2	85.8	$(-)87.2\pm1.6$	0.35	
	f_3	18.5	$(\pm)9.4\pm1.0$[a]	0.19[a]	
	f_4	0	0	0.60	
	$R(t)$ f_1	168.6	-89.7 ± 1.6	0.22	0.51
	f_2	10.9	$+1.6\pm0.6$	0.29	

[a] f_3 assumed to correspond to 2ω component of Eq. (3), with the ω component unobserved.

The principal results of our measurements are summarized in Table I. For each set of experimental conditions the observed frequency components, fraction of full amplitude derived as discussed above, and the corrected hyperfine field are given, as well as the summed amplitudes for all components.

Comparing the results for $\epsilon(t)$ and $R(t)$ for the unannealed source one sees that the amplitudes in the latter case are about 30% lower than in the former. This would seem to suggest that about this fraction of the atoms were not polarized. This explanation is supported by the existence of a peak in the Fourier spectrum of $\epsilon(t)$ at half the principal frequency with about this amplitude, as would be expected for an unpolarized sample. Unfortunately the statistics of the measurement do not permit a quantitative estimate of this component of the spectrum.

Taking into account the various components one finds for the unannealed source about 40% of the atoms in the high-field site, 10% in the low-field site, and about 50% unaccounted for. About 20% of each group of atoms were not polarized by the external field. Observation of the time spectra of $R(t)$ and $\epsilon(t)$ suggest that the unobserved fraction of the atoms contribute to the strongly damped peak occurring near $t=0$ in both spectra. Such a behavior of the time spectra can be understood as arising from a wide distribution of frequencies centered around zero.

Analysis of the results after annealing shows a considerably different structure. Frequencies corresponding to the -90-kOe component are observed both in $R(t)$ and $A_2G_2(t)$. However both the strongly damped initial peak and the component corresponding to a positive 10-kOe field are now absent, although a new low-frequency component has appeared in $A_2G_2(t)$ which is unobserved in $R(t)$. (No explanation of the nature of this component was found.) The measurement of $A_2G_2(t)$ further shows that more than half the atoms feel no hyperfine field at all within the accuracy of the measurement. More detailed analysis of the respective Fourier spectra also shows a significant distribution of frequencies around -90 kOe which was not observed before annealing.

On the basis of the results a phenomenological explanation of the origin of the various hyperfine interactions observed can be attempted. We imagine that as a result of melting followed by rapid cooling about 40% of the atoms are left on regular lattice sites corresponding to the -90-kOe field. Another 10% of the atoms are frozen into a second distinct lattice position—perhaps interstitial—while the remaining half are segregated out onto grain boundaries, voids, or other types of imperfections. It is interesting to note that there was no significant precipitation of hafnium into clusters in the material since this should give rise to a clearly observable quadrupole spectrum.

As a result of the annealing the atoms located on the site corresponding to the small positive field precipitate out onto grain boundaries along with the 50% of the atoms in random positions. Few of the substitutional atoms precipitate out. However, their tendency to act as traps for vacancies is demonstrated by the fact that after annealing a distribution of frequencies around the -90 kOe value appears.

Some support for the assignment of the -90 kOe field to substitutional atoms as well as further information concerning lattice locations of hafnium in nickel has been obtained from preliminary channeling experiments carried out at Bell Telephone Laboratories on hafnium implanted into a single crystal of nickel at room temperature.[6] These experiments showed that at room temperature approximately half of the implanted atoms were located at regular lattice sites and that no distinct lattice position could be assigned to the rest of the atoms. A systematic study of the effects of annealing was also carried out. It was found that at 600 °C most of the substitutional atoms had migrated to the surface of the crystal. This result, in contrast to ours, possibly results from a strong enhancement of diffusion due to the high density of imperfections caused by the implantation.

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[1]J. C. Vanderleeden, in *Hyperfine Structure and Nuclear Radiations*, edited by E. Matthias and D. Shirley (North-Holland, Amsterdam, 1968), p. 495.

[2]J. A. Cameron, P. R. Gardner, W. V. Prestwich, Z. Zamori, and D. C. Santry, Can. J. Phys. **48**, 2725 (1970).

[3]J. L. Oddou, J. Berthier, P. Peretto, and M. Robin, Phys. Status Solidi B **45**, K139 (1971).

[4]J. D. Bowman, H. E. Henrikson, and F. C. Zawislak, Nucl. Instrum. Methods **84**, 77 (1970).

[5]R. Béraud, I. Berkes, J. Danière, G. Marest, and R. Rougny, Nucl. Instrum. Methods **69**, 41 (1969).

[6]E. N. Kaufmann and J. M. Poate (private communication).