Spin-lattice relaxation of ^{60}Co spins in multidomain iron*

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The spin-lattice relaxation of ^{60}Co nuclei in an iron host has been studied in various applied magnetic fields using the technique of nuclear magnetic resonance on oriented nuclei. In this method the nuclei studied are those in the bulk of the domains. The measurements show that the relaxation rate is dependent on the state of magnetization of the sample; the rate in 0.4 kOe is nearly five times faster than that measured in 6.7 kOe. This result agrees with previous measurements using conventional nuclear magnetic resonance, and demonstrates unambiguously the existence of a relaxation process in multidomain transition-metal ferromagnets that has never been adequately explained by theory. The resonance linewidths measured in the experiments show a dependence on the applied field. At high fields, when the sample is magnetically saturated, the full width at half maximum is about 0.7 MHz, while in the lowest field employed this width is only 0.2 MHz. An explanation in terms of demagnetizing effects is offered.

INTRODUCTION

Ever since the first observation of nuclear-magnetic resonance (NMR) in ferromagnetic cobalt metal,[1] nuclear spin-lattice relaxation in magnetically ordered transition metals and alloys has been studied extensively.[2,3] In unmagnetized multidomain particles, the NMR signal is dominated by the contribution from nuclei in domain walls because of the tremendous signal enhancement and the relaxation of these nuclei is understood fairly well.[4,5] By applying a magnetic field so that the sample is fully magnetized it is possible to study the "bulk" nuclei allowing the measurement of their relaxation and, again, this relaxation rate which we shall call $(1/T_1)_{\text{intrinsic}}$ has been the subject of numerous studies.[2,3,6-8]

In the unmagnetized or partially magnetized sample most of the volume is occupied by domains, not walls, and so it is of interest to study the relaxation rate of nuclear spins *within* a domain in the *multidomain* sample. Indeed there have been a number of NMR studies[3,6-8] purporting to study this relaxation rate, which we will call $(1/T_1)_{\text{multidomain}}$, and it is invariably found that the ratio $(1/T_1)_{\text{multidomain}}[(1/T_1)_{\text{intrinsic}}]^{-1}$ is greater than unity with values ranging up to 5 in some cases. This would indicate the presence of an additional relaxation process in these systems. However, the experiments are quite difficult to perform since the signal from nuclei in domain walls must be eliminated. In addition, there is some uncertainty in interpreting the measurements as has been pointed out recently.[9] For example, interplay between wall enhancement and anisotropic hyperfine interaction may easily lead to signals originating in unexpected regions of a multidomain sample. Only recently has a theoretical explanation been offered[8] to treat $(1/T_1)_{\text{multidomain}}$ and this does not adequately explain the experimental results. Because of the complications in the NMR experiments, it would appear useful to support these measurements by a study using a different technique. Consequently, we have employed the method of nuclear-magnetic resonance on oriented nuclei (NMR/ON) to determine spin-lattice relaxation times for ^{60}Co nuclei in an iron host. The usefulness of this technique in the present context lies in the fact that all radioactive nuclei in the specimen are observed with equal probability and, as mentioned above, a very high fraction of these are in domains. In fact, after the commencement of our work we learned that a similar study has been reported briefly by Kohzuki *et al.*[10] However, the lowest magnetic field applied in their experiment was 1 kOe while our measurements are performed down to 0.4 kOe. Also there are small differences between our results and theirs, and these will be discussed in a later section of this paper.

To discuss the NMR/ON technique it is necessary to review briefly the subject of nuclear orientation. For a system of radioactive nuclei, of moment μ and spin I, oriented by a hyperfine field H_n at a low temperature T, the intensity of γ rays emitted at angle θ to the quantization axis is given by[11]

$$W(\theta) = \sum_{\nu \text{ even}} A_\nu B_\nu P_\nu (\cos\theta) .$$

Here A_ν are parameters related to the radioactive decay, B_ν are functions of the Boltzmann exponent $\mu H_n/IkT$, and are measures of the degree of orientation, while P_ν are the Legendre polynomials. $W(\theta)$ is normalized to the "high" temperature ($\sim$1 K) intensity. The largest effect is observed at $\theta = 0$, and it is useful to define a γ-ray "anisotropy"

$$\alpha = 1 - W(0).$$

For a polycrystalline ferromagnet, partially

magnetized by an applied field H, the intensity measured along the field direction is given by

$$[I(0)]_H = \sum_i f_i \, W(\phi_i) ,$$

where f_i is the fraction of domains for which the magnetic axis is at angle ϕ_i to the direction of H. We can define an anisotropy, measured in field H,

$$\alpha_H = 1 - [I(0)]_H .$$

For complete magnetic saturation, the full anisotropy is observed when $\alpha_H = \alpha$, but for incomplete saturation $|\alpha_H| < |\alpha|$.

If the oriented nuclear system is irradiated by an rf field of frequency $\nu = \mu H_n/hI$, the magnetic substate populations are altered and consequently the γ-ray intensity pattern is also changed, so that a different anisotropy $\alpha_{H,\nu}$ is measured. The first successful detection of NMR by this means was achieved by Matthias and Holliday[12] and the extension of the method to measure spin-lattice relaxation times at low temperatures was performed first by Templeton and Shirley.[13] Most NMR/ON experiments are performed using $[I(0)]_\infty$. However, the effect can of course be observed with the reduced intensity $[I(0)]_H$ and this is the basis of our experiment.

EXPERIMENTAL

The sample prepared for our experiment was an iron plate of dimensions $0.5 \times 0.5 \times 0.03$ cm into which was diffused 15 μCi of ^{60}Co which had been plated on the polished surface. The diffusion depth was estimated as 0.7 μm which is less than the skin depth for the radio-frequency fields employed (~1 μm). After diffusion the surface of the plate was etched to remove any surface activity.

This sample plate was soldered on to a copper cold finger connected to a chrome potassium alum salt pill in a demagnetization cryostat. On demagnetization from a field of 40 kOe applied at 1.1 K, specimen temperatures of about 10 mK were attained and maintained for ~50 h. The specimen could be magnetized by a superconducting coil which produced fields up to 10 kOe. A 5×5-in. NaI counter detected γ rays emitted along the field axis ($\theta = 0$ for the fully magnetized sample), this direction being in the plane of the specimen plate.

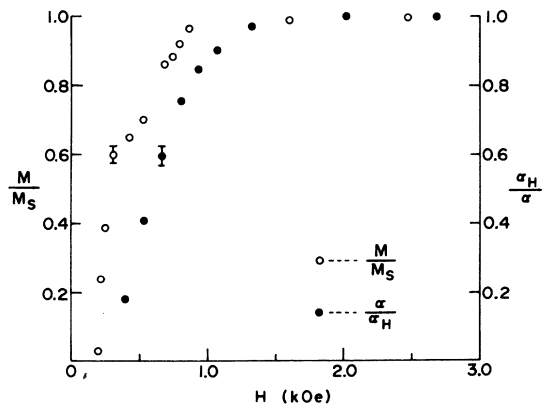

FIG. 1. Normalized values of magnetization M and γ-ray anisotropy α_H vs applied field. M_S is the saturation magnetization.

TABLE I. Values of the resonant frequency ν, partial destruction of the γ-ray anisotropy f, and FWHM linewidth observed for different applied fields. Also shown is the modulation frequency used. The error in the values of ν and FWHM is 0.05 MHz.

Applied field H(kOe)	Resonant frequency ν(MHz)	Frequency modulation (MHz)	Partial destruction f	FWHM (MHz)
6.70	162.01	0.1	0.08	0.64
2.68	164.61	0.2	0.13	0.68
2.01	164.97	0.2	0.20	0.62
0.94	165.56	0.2	0.21	0.38
0.67	165.64	0.2	0.31	0.30
0.54	165.68	0.2	0.22	0.32
0.40	165.70	0.1	0.29	0.24

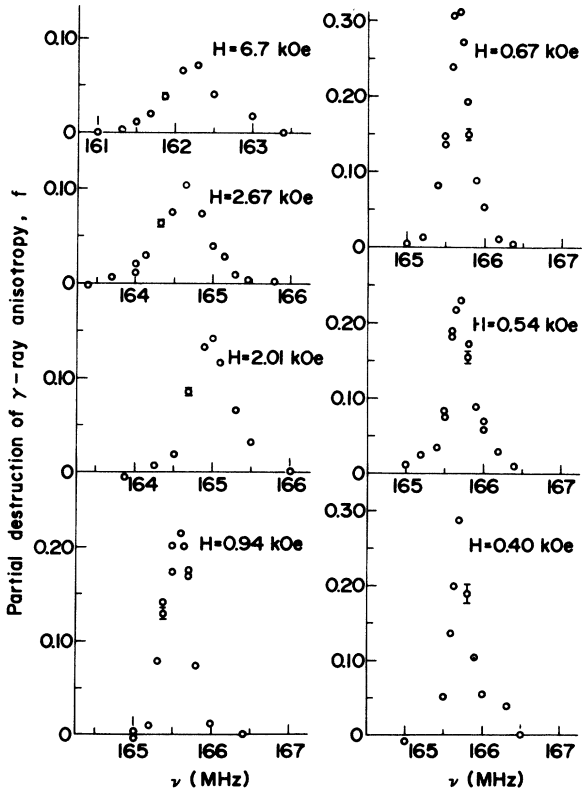

FIG. 2. Resonance lines observed in different applied fields H. The parameter f is a measure of the partial destruction of the γ-ray anisotropy. Note the decrease in linewidth with decreasing field.

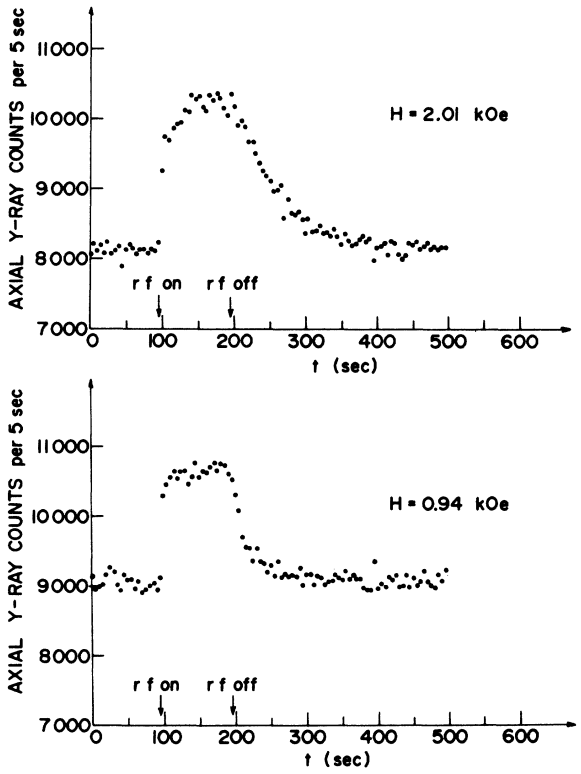

FIG. 3. Typical relaxation measurement for two fields. Six such measurements were made for each value of H and the data averaged.

The rf field was produced by a Helmholtz pair of one turn each which produced a field parallel to the surface of the specimen plate and perpendicular to the applied polarizing field.

After demagnetization the specimen temperature could be estimated from the ^{60}Co γ-ray anisotropy[14] α, since A_ν, μ, I, and H_n are all known. A preliminary measurement of $W(0)$ versus applied field was taken and this could be compared with a magnetic saturation curve previously obtained by measuring the inductance of a coil wound around the specimen plate. These results are shown in Fig. 1. That these curves may not correspond exactly has been demonstrated recently.[15]

The rf field amplitude used in the experiments was 0.005 Oe, a value selected by compromising between a power high enough to effect a substantial change in $I(0)$ and one low enough to avoid excess eddy current heating in the specimen cold finger. It should be noted that the domain nuclei feel a field enhanced by a factor of approximately H_n/H or H_n/H_a, whichever is the smaller, where H_a is the anisotropy field.

The resonance was found by sweeping the frequency of the rf field, which was also frequency modulated, until partial destruction of the γ-ray anisotropy α_H was observed. This partial destruction, which represents the signal amplitude can be described quantitatively by a parameter

$$f=(\alpha_H-\alpha_{H,r})/\alpha_H.$$

The resonance frequency ν and the partial destruction f measured for different fields H are shown in Table I. The resonance frequency extrapolated to $H=0$ was 165.8 ± 0.1 MHz in agreement with the accepted value.[16] The full width at half-maximum (FWHM) of the line in the highest applied field, $H=6.8$ kOe, was 0.7 MHz, but this width was observed to depend strongly on H. Thus in Fig. 2 are shown lines observed in various applied fields. The FWHM at the lowest field used, $H=0.4$ kOe, was 0.2 MHz, much smaller than the values normally measured. At fields lower than 0.4 kOe, the value of α_H was too small to be useful. A compilation of FWHM values for different fields is also included in Table I.

The spin-lattice relaxation of the ^{60}Co spins was observed by switching off the rf field at resonance and monitoring the γ-ray intensity at $\theta=0$ with a multichannel analyzer. This was set to count in the multiscaling mode with a dwell time of 5 sec. Typical measurements for two different polarizing fields are shown in Fig. 3. For a given value of H, six such measurements were made and the data averaged.

Streever and Caplan[6] have measured a transverse relaxation time of about 5 msec for ^{59}Co nuclei in a 1-at.%-Co–Fe alloy. However, it is not clear whether the relaxation is dominated by spin-spin interactions or spin-lattice interactions. In any event their measurement allows us to place a *lower* limit of approximately 1 sec for the spin-

TABLE II. Values of the Korringa constant C computed for various values of the magnetizing field H under the two assumptions (a) and (b), where T_s is spin temperature. Also shown are the χ^2 values measuring the "goodness" of the fits.

H (kOe)	Number of data points fitted	(a) T_s assumed C(sec K)	χ^2	(b) T_s not assumed C (sec K)	χ^2
6.70	36	2.58	31	3.06	44
6.70	36	2.23	40	2.64	60
2.68	30	2.15	145	2.60	236
2.68	36	2.14	140	2.48	293
2.68	36	2.21	94	2.55	201
2.01	30	1.91	136	2.28	242
1.34	18	1.13	14	1.30	31
1.21	15	1.01	10	1.16	17
1.07	9	0.75	20	0.88	25
0.94	9	0.60	21	0.69	8
0.81	9	0.58	70	0.65	43
0.67	9	0.50	42	0.58	40
0.54	9	0.52	10	0.60	5
0.40	9	0.76	53	0.87	46

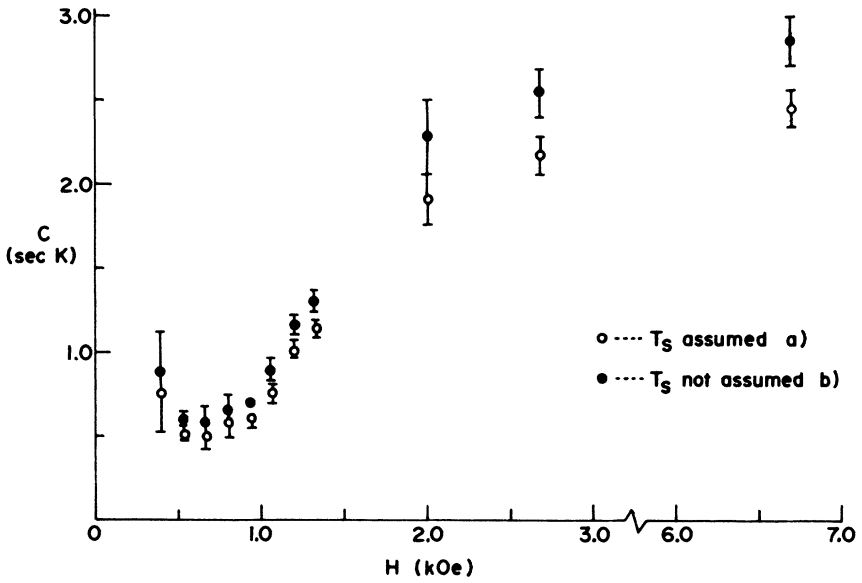

FIG. 4. Korringa constant C vs applied field H. The two sets of values for C correspond to fitting the relaxation data using the two theoretical models (a) and (b). Note break in H axis. The error bars on each value of C are statistical and represent the standard error of the mean.

spin relaxation time T_2 for the ^{60}Co nuclei in our sample. Since $T_1 \sim 250$ sec at 10 mK it is uncertain whether a common spin temperature can be assumed for the ^{60}Co spins in their relaxation. Consequently, the relaxation data has been analyzed in two ways: (a) assuming a spin temperature existed and (b) assuming it did not. A discussion of the low-temperature theory appropriate for these two cases has been given by Bacon *et al.*[17] The analysis consists of fitting to the data a curve calculated with two adjustable parameters, the initial population distribution and the effective "Korringa constant" C defined by

$$C = T_1\left(\frac{h\nu}{2k}\right)\left[\tanh\left(\frac{h\nu}{2kT}\right)\right]^{-1},$$

where relaxation via the conduction electrons is assumed. In the high-temperature limit $kT \gg h\nu$, the expression for the Korringa constant reduces to $C = T_1T$. The initial population distribution in the resonance experiments is determined by the γ-ray anisotropy $\alpha_{H,r}$, and in these experiments the best fit was obtained by assuming that *all* the ^{60}Co spins in the sample were at the same initial spin temperature.

A summary of the results of our analysis is shown in Table II. It is evident that with assumptions (a) and (b) values of C that are a little different from each other are obtained. A χ^2 analysis shows that the former assumption yields a slightly better fit to the data. The values of C obtained with assumptions (a) and (b) for different applied fields H are shown in Fig. 4.

DISCUSSION

The results of experiments to determine the field dependence of γ^2T_1T, where γ is the gyromagnetic ratio, for various cobalt nuclei in an iron host are summarized in Table III. It can be seen that there is good agreement between our values and those measured in the NMR experiments.[6,8] It should be noted that in the NMR experiment on a powder sample[6] much higher fields were required for magnetic saturation. Our high field value of 2.45 ± 0.11 sec K, using assumption (a), also agrees well with the previously published values of 2.4 sec K[17] and 2.6 sec K[18] for ^{60}Co in iron obtained by the NMR/ON technique. The results of Kohzuki *et al.*[10] appear less accurate than our values. Their value of $C = 1.9 \pm 0.5$ sec K measured at 9 kOe is somewhat lower than our high-field value, but just within the experimental

TABLE III. Values of γ^2C, where γ is the gyromagnetic ratio and C is the Korringa constant, obtained in various experiments. The suffices a and b refer to the assumptions (a) and (b) made in deducing the value of C from the data.

Specimen	Method	γ^2C (10^6K sec^{-1} Oe^{-2}) Low field	High field	Ref.
^{59}Co 1 at.% in Fe powder	NMR	9	41	6
^{59}Co 1 at.% in Fe foil	NMR	13	37	8
^{60}Co ≪1 at.% in Fe foil	NMR/ON	9	26	10
^{60}Co <1 at.% in Fe foil	NMR/ON	7[a]	32[a]	This work
		8[b]	37[b]	

error. Their value of $C = 0.67 \pm 0.06$ sec K measured at the lowest field employed (1 kOe) agrees very well with our value of 0.65 ± 0.05 sec K measured at the same field of 1 kOe. If a discrepancy exists at higher fields, this could be due to an incomplete penetration of the rf field into their 5-μm-thick specimen foil, since the skin depth is ~ 1 μm and the rf field enhancement factor is $\sim H_n/H$. Alternatively, the origin of any discrepancy might lie in their method of analysis which is not mentioned in their paper. Thus the use of a single exponential function to fit the data would lead to too small a value for C.

The high-field value of $C = 2.4$ sec K is in reasonable agreement with the value of 2.0 sec K calculated by Kontani *et al.*[8] assuming that the dominant interaction for the relaxation process is that between the nuclear spins and the orbital moment.[19,20] (Here we have converted the value calculated for ^{59}Co to that for ^{60}Co.)

It is evident that, for low fields, the relaxation rate is indeed dependent on the field strength and hence on the sample magnetization. Kontani *et al.*[8] suggest that an interaction between the nuclear spins and the conduction electrons via spin waves, a process first suggested by Weger,[4] would be field dependent. However, they show that such an interaction would have the form

$$T_1 T = D(1 + H/B),$$

where B and D are constants, the former having a value of approximately 20 kOe. Even if a lower value of B were assumed, the form of the C-vs-H curve in Fig. 4 is not in agreement with a linear dependence on H over the whole range of H values. Thus, although the proposed interaction may partly contribute to the relaxation process, a complete explanation of $(1/T_1)_{\text{multidomain}}$ is still lacking. Other suggested processes involving a direct interaction between the nuclear spins and the spin waves[8] would appear to be too weak to account for the observed relaxation rates. (It is important to note here that for the low-temperature experiments, even if $T_2 \sim 1$ sec, spin-lattice relaxation by spin diffusion to the rapidly relaxing spins in the domain wall is *not* a significant contribution to the total process. We estimate using a random-walk spin diffusion model[21] that spin-lattice relaxation by this process is several orders of magnitude slower than the observed rate.)

The narrowness of the resonance lines observed in our experiment merits some discussion. For ^{60}Co in iron, $\nu \simeq 166$ MHz, so that the rf skin depth is about 1 μm. The active layer of the specimen used in our experiments was 0.7 μm, and, typically, in all NMR/ON experiments, specimens have a foil thickness or active-layer thickness of about 1 μm. Now even after polishing, surface irregularities might be $\gtrsim 0.1$ μm, so that in the conventional method using an applied field large enough to produce magnetic saturation, i.e., $\alpha_H = \alpha$, there might be a distribution of demagnetizing fields over the sample. It is usually assumed that the demagnetization factor has the value $D = 0$, but let us assume that, in fact, there is a variation ΔD. The linewidth due to this effect would be

$$\Delta\nu \simeq \gamma(\Delta D)M,$$

where M is the magnetization. For $\Delta D = 0.3\pi$, the resulting linewidth is about 1 MHz. On reduction of the field, the demagnetizing energy would be reduced by the formation of domains so that this contribution to the linewidth would be much reduced, as is observed.

CONCLUSIONS

Our experiment supports the results of NMR experiments which show that there is a field-dependent spin-lattice relaxation mechanism in 3*d* ferromagnets, presumably associated with the multidomain regime. Since this process has not been explained satisfactorily, we feel that a systematic study of $(1/T_1)_{\text{multidomain}}$ in samples with known domain geometry would be rewarding. Our NMR/ON experiments also show that a significant narrowing of the resonance line occurs on reducing the applied field. This suggests that the high-field linewidth may be dominated by the contribution due to a spread in demagnetizing fields over the sample.

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[1] A. M. Portis and A. C. Gossard, J. Appl. Phys. 31, 205S (1960).

[2] For example, M. Weger, E. L. Hahn, and A. M. Portis, J. Appl. Phys. 32, 124S (1961); M. Weger, Phys. Rev. 128, 1505 (1962); T. Moriya, J. Phys. Soc. Jap. 19, 681 (1964); R. L. Streever, Phys. Rev. 134, A1612 (1964); N. Kaplan, V. Jaccarino, and J. H. Wernick, Phys. Rev. Lett. 16, 1142 (1966).

[3] M. B. Salamon, J. Phys. Soc. Jap. 21, 2746 (1966); V. Jaccarino, N. Kaplan, R. E. Waldstedt, and J. H. Wernick, Phys. Lett. 23, 514 (1966).

[4] J. M. Winter, Phys. Rev. 124, 452 (1961).

[5] M. Weger, Phys. Rev. 128, 1505 (1962); M. B. Stearns, Phys. Rev. 162, 496 (1967).

[6] R. L. Streever and P. J. Caplan, Phys. Lett. A 38, 439 (1972).

[7] M. H. Bancroft, Phys. Rev. B 2, 182 (1970).

[8] M. Kontani, T. Hioki, and Y. Masuda, J. Phys. Soc. Jap. 32, 416 (1972).

[9] M. A. Butler, J. Phys. Soc. Jap. 34, 1419 (1973);

M. A. Butler, Int. J. Magn. 4, 131 (1973).
[10]S. Kohzuki, Y. Aoki, N. Yamashita, and J. Itoh, J. Phys. Soc. Jap. 32, 1678 (1972).
[11]R. J. Blin-Stoyle and M. A. Grace, Handbook Phys. 42, 555 (1957).
[12]E. Matthias and R. J. Holliday, Phys. Rev. Lett. 17, 897 (1966).
[13]J. E. Templeton and D. A. Shirley, Phys. Rev. Lett. 18, 240 (1967).
[14]N. J. Stone and B. G. Turrell, Phys. Lett. 1, 39 (1962).
[15]K. S. Krane, B. T. Murdoch, and W. A. Steyert, Phys. Rev. Lett. 30, 321 (1973).
[16]T. A. Koster and D. A. Shirley, in *Hyperfine Interactions in Excited Nuclei*, edited by G. Goldring and R. Kalish (Gordon and Breach, New York, 1971), Vol. 4, p. 1239.
[17]F. Bacon, J. A. Barclay, W. D. Brewer, D. A. Shirley, and J. E. Templeton, Phys. Rev. B 5, 2397 (1972).
[18]D. Spanjaard, R. A. Fox, I. R. Williams, and N. J. Stone, in Ref. 16, Vol. 1, p. 345.
[19]Y. Obata, J. Phys. Soc. Jap. 18, 1020 (1963).
[20]T. Moriya, J. Phys. Soc. Jap. 19, 681 (1964).
[21]A. Abragam, *Nuclear Magnetism* (Oxford U. P., Oxford, England, 1961), Chaps. 5 and 9.