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Single-Passage Resonance Studies by β Emission and the Measurement of Spin-Lattice Relaxation Times for Fe^{60} Co

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β-particle measurements of the signal produced during single- and multiple-passage nuclear magnetic resonance of oriented ^{60}Co nuclei in single crystals of iron are reported. Comparison of the signals from β^- and γ radiations shows that after single passages the changes in the orientation parameters are describable in terms of a rotation through a well-defined angle. This constitutes a known set of initial conditions for the subsequent relaxation back to equilibrium so that accurate values of the spin-lattice relaxation time T_1 can be obtained. A nonresonant method of measuring T_1 accurately is also described and is shown to give results in agreement with the single-passage technique. We obtain a value $K = 1.75 \pm 0.15$ sec K for Fe ^{60}Co. This differs from values previously reported from experiments in which the initial conditions were not well known. Comparison of the β^- and γ signals after multiple passages show, as expected, that the changes in the orientation parameters are not then describable in terms of a single angle of rotation.

I. INTRODUCTION

The first observation of NMR on oriented nuclei in ferromagnets was reported by Matthias and Holliday.[1] In this technique the resonance is detected via the partial destruction of the anisotropic distribution of radiation emitted from statically oriented radioactive nuclei. Although considerable knowledge of the hyperfine parameters has been obtained, a complete understanding of the magnitude of the fractional reduction in radiation anisotropy is lacking. The problem is complex because the large inhomogeneous broadening in ferromagnets necessitates frequency modulation.[2] Also, at the very low temperatures necessary to orient nuclei and at the relatively high frequencies used, there are additional complications with eddy-current heating and skin depth.

In a previous attempt to understand the problem, a theoretical and experimental study of single-passage NMR of ^{60}Co in Fe detected via γ radiation was reported.[3] This single-passage approach affords a considerable theoretical simplification over the continuous-modulation procedure. However, even in this simpler case, a major discrepancy was observed between the angle of rotation of the nuclear ensemble as deduced from the observed single-passage signals and the angle of rotation calculated theoretically. It was tentatively postulated that the discrepancy might be caused by the effect of the nuclear polarization back onto the electrons.

In this paper we are not concerned with the cause of this discrepancy but rather with the application

of single passages to the measurement of spin-lattice relaxation times. In Sec. II we report simultaneous measurements of the β^-- and γ-radiation anisotropy in a single-passage study of ^{60}Co in iron. This was performed to establish that the ensemble is indeed rotated through a unique angle, even though the rotation is much less than expected theoretically. As this is the case, a single-passage experiment leaves the nuclear ensemble in a known initial state so that accurate values of the spin-lattice relaxation time T_1 may then be obtained from the subsequent time dependence of the anisotropies of the radiation. In Sec. III, in a study of multiple-passage NMR, we show that the resulting changes in the initial conditions are too complex to be described by an effective single passage. Finally, in Sec. IV we describe the T_1 measurements obtained by the single-passage technique and also by a nonresonant fast-cooling method.

II. SINGLE-PASSAGE MEASUREMENTS

A. Theory

The theory of single-passage NMR on oriented nuclei has been given in an earlier paper.[3] We reproduce only the basic equations for γ-ray anisotropy and extend the description to include β^--particle anisotropy.

Our system consists of an ensemble of ^{60}Co nuclei in an iron single crystal in a static magnetic field $\vec{H}_0$ with an oscillating magnetic field $\vec{H}_1 \ll \vec{H}_0$ perpendicular to $\vec{H}_0$. The ensemble will initially have axial symmetry, being substantially aligned by hyperfine interaction, parallel to $\vec{H}_0$. During a single passage the applied rf field is swept uniformly through the resonant region, causing the resultant magnetic field in the Larmor frame to be inverted. If the motion of the nuclear ensemble faithfully follows the effective field, i.e., is adiabatic, then the ensemble will be rotated by 180°. However, in general the motion will not be adiabatic and the effect of the rf can be characterized by an adiabatic parameter[4] A such that

$$A = \omega_1^2 / \left(\frac{d\omega}{dt}\right)^{-1} = \gamma_n^2 \eta^2 H_1^2 / (2\pi \Delta f / t_p) ,$$

where the strength of the applied rf field H_1 enters through $\omega_1 = -\gamma_n \eta H_1$ with the enhancement factor[5] η being the ratio of the effective field on the nuclei to the applied field $\vec{H}_0$. The width of the frequency sweep Δf and the time of sweep t_p are the two other experimental variables. At some instant t during a single passage the nuclear ensemble will lie at an angle θ with respect to $\vec{H}_0$, and will achieve a final angle θ_f at the completion of the passage. As was shown in Ref. 3, the ensemble motion is the same as that of the magnetization, so the variation of the final angle as a function of A can be calculated[3] by solving the classical torque equation for the magnetization $\vec{m}$:

$$\frac{d\vec{m}}{dt} = \gamma_n \vec{m} \times \vec{H} .$$

For $A > 5$ the motion is thus calculated to be adiabatic while for $A < 0.001$ almost no rotation of the nuclear ensemble is expected. If the sweep time is small in comparison with the nuclear spin-lattice relaxation time then the normalized anisotropy of the β^--particle and γ-ray intensity observed in a detector at angle Θ to $\vec{H}_0$ can be described by[3]

$$W(\Theta) = 1 + \sum_{\nu = 1,2,\ldots} B_\nu(0) U_\nu F_\nu P_\nu(\cos\theta) P_\nu(\cos\Theta) , \quad (1)$$

where $B_\nu(0)$ are the equilibrium orientation parameters which depend on $x = \hbar\omega/2kT$. U_ν and F_ν are angular momentum coupling parameters of the unobserved preceding radiation and the observed radiation, respectively. The Legendre polynomial $P_\nu(\cos\Theta)$ describes the angular dependence of the intensity of emission about $\vec{H}_0$. The time dependence of the anisotropy is given solely by the $P_\nu(\cos\theta)$, where θ is the classical rotation of the ensemble at time t. Hence after a single passage (1) shows that the final orientation parameters are given by

$$B_\nu = B_\nu(0) P_\nu(\cos\theta_f) . \quad (2)$$

If the signal S is defined as the fractional destruction of the radiation anisotropy[6] then for a single passage the β signal becomes

$$S_\beta = 1 - P_1(\cos\theta) . \quad (3)$$

For $x \leq 0.3$, the γ signal is given by

$$S_\gamma = 1 - P_2(\cos\theta) , \quad (4)$$

where parity conservation causes all the odd ν terms to be zero.

If $x \geq 0.3$, then higher-order terms must be considered. For small adiabatic parameters where little or no rotation of the ensemble occurs, the γ signal remains small; however, for $A \simeq 0.45$ a rotation of $\theta_f = 90°$ occurs and the corresponding initial signal expected is 1.5. For larger A the initial signal is smaller but will gradually increase as the nuclear ensemble relaxes back through 90°. In the earlier study of single-passage NMR[3] the observed γ signal never exceeded 0.5, suggesting the degree of rotation experienced by the nuclear ensemble is much less than predicted theoretically. This means that the description of the nuclei as simply experiencing the resultant of the hyperfine field and the applied fields, including enhancement, is incorrect for ^{60}Co in iron at low temperatures. If this description was applicable it follows from Eq. (2) that the orientation parameters immediately after a passage would be known from the

classical theory. However, in Ref. 3 it was shown that whatever the fields acting on the nuclei, the orientation parameters after a single passage should still be given by Eq. (2) but in terms of an equivalent angle of rotation which must be deduced by measurement. Although this angle is apparently quite different from that given by the simple single-passage theory it can still be determined by applying Eq. (3) or Eq. (4) to the signal immediately after a passage.

The critical assumption is thus that after a single passage the ensemble can be described in terms of a unique final angle. If this is so, then it is the first case of a resonant-nuclear-orientation experiment in which the initial conditions necessary for the accurate determination of spin-lattice relaxation times from the subsequent relaxation back to the equilibrium parameters $B_\nu(0)$ are known. By simultaneously determining the final angle of rotation by using the β^- and γ signals and Eqs. (3) and (4), this important assumption can be readily tested.

B. Experimental

Single-crystal ion specimens in the form of a 0.5-cm-diam 0.01-cm-thick disk had about 2μCi of exchange column purified ^{60}Co activity evaporated onto the highly polished face and furnaced in a hydrogen atmosphere at 830 °C for 6 min. All undiffused activity was carefully removed before use. The cobalt concentration in the iron surface would be of the order of 0.025 at.%. The samples were cooled via a copper fin system using adiabatic demagnetization of a cerium-magnesium-nitrate glycerol slurry. Using frequency modulation and $\vec{H}_0 = 0.2$ T, a strong resonance centered at 165.4 MHz was observed with a full width at half-height at 800 kHz. The β-particle detector was a Au-Si surface-barrier detector with a depletion layer of 100 μ and an active area of 25 mm^2. It was mounted in the Dewar vacuum at room temperature, coaxial with $\vec{H}_0$, about 0.1 m from the source. It was necessary to interpose two heat shields consisting of 8×10^{-6}-m aluminized Mylar between the β detector and the cooled sample. The additional heat load from this minimal shielding prevented the system from cooling below 0.012 K and it warmed to 0.03 K in about 4 h with an rf field on the sample. Counting times were restricted to the 2 h after cooling the sample. A 43-cm^3 Ge (Li) detector was used to measure the γ rays but because of the β detector housing it had to be positioned at $\Theta = 19.5° \pm 0.5°$.

To minimize self-absorption of the β particles, the source was mounted at ~10° to $\vec{H}_0$, with $\vec{H}_1$ in the plane of the sample. The static magnetic field $\vec{H}_0$ had a focusing effect on the β particles entering the collimated counter, and to improve statistics all but the very-lowest-energy β particles were included in the counting window. For the β^- particles of velocity v from ^{60}Co nuclei[7]

$$U_1 F_1 = -(\tfrac{2}{5})^{1/2} v/c \ .$$

In actual measurements one observes a distribution of energies (velocities) so the anisotropy is given by an integral over the velocities. In addition, to obtain absolute β^- anisotropies, scattering, solid angle, absorption, and magnetic field deflection must be taken into account. The effective equilibrium β anisotropy at $\Theta = 0$ is given by

$$W(0)_\beta = 1 - \alpha B_1 \ ,$$

where

$$\alpha = (\tfrac{2}{5})^{1/2} \iiint \frac{v}{c} \cos\Theta \, dv \, d\Theta \, d\xi \ ,$$

with $d\xi$ allowing for the scattering, absorption, focusing, etc. The reduction factor α can be determined by comparing the observed β^- anisotropy with B_1 calculated by knowing the temperature from the γ-ray anisotropy. We find $\alpha = 0.23 \pm 0.02$ over our temperature range ($1/T = 25 - 80$ K^{-1}). Providing α is temperature independent, a detailed knowledge of its value is not required since the signal is determined from a ratio.

Figure 1 shows a typical β and γ single-passage relaxation curve. The actual data were taken by choosing a frequency range to include the entire width of the resonance, then sweeping across it a number of times, leaving sufficient time between each sweep for equilibrium to be reestablished. This was repeated at various H_1 amplitudes and times of sweep. To avoid relaxation effects during a passage, all times of sweep were kept short (≤ 6 sec) compared to T_1, although, as discussed previously,[3] it is the time of passage through a spin packet which is important and this is of the order of milliseconds. The signals were obtained by least-squares fitting a single exponential to the decay curves to give the amplitudes at the end of the passage before relaxation. Table I shows the experimental results. The theoretical results are included for comparison. Both the β^- and γ measurements clearly disagree with the theoretical value calculated using the simple model of single-passage theory of Ref. 3. Apart from confirming the existence of this discrepancy, the present measurements do not shed any further light on the cause of the discrepancy which, as in Ref. 3, may possibly be due to spin-spin and frequency pulling effects as discussed by de Gennes *et al.*[8]

However, the purpose of these measurements is to confirm the existence of a unique angle of rotation for the nuclear ensemble and that this is true is shown by the good agreement between the angles of θ_f, as deduced from the β^- and γ signals immediately after the passages. This shows that the

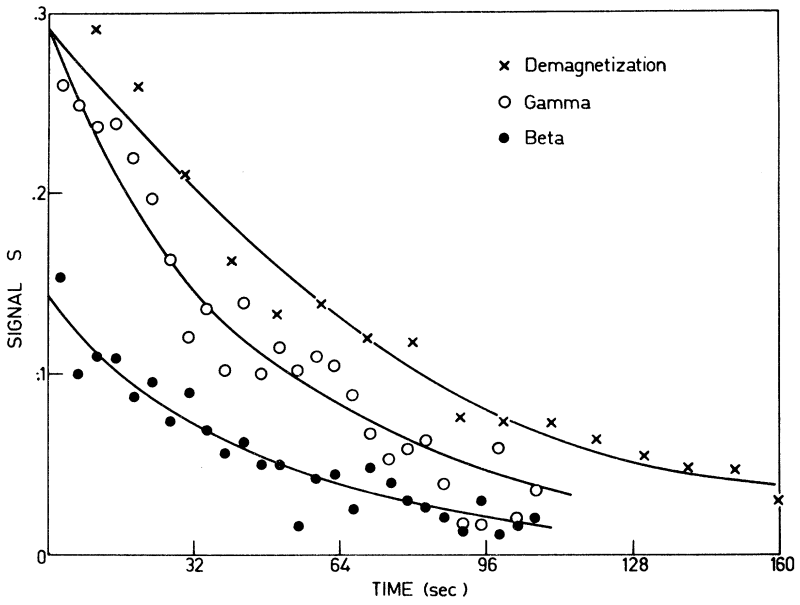

FIG. 1. Experimental relaxation curves for (×) demagnetization technique between $1/T$ of 9 to 58 K^{-1}; (○) γ-ray single passage at $1/T$ of 73 K^{-1}, (●) β-particle single passage at $1/T$ of 73 K^{-1}. The demagnetization curve has been normalized to the γ-ray curve at $t=0$. The solid lines are theoretical curves obtained from Gabriel theory.

description of the effect of such passages on the orientation parameters may be expressed in terms of a unique angle using Eq. (2). One observation concerning θ_f is that the β angle is consistently a few degrees higher than the corresponding γ angle. A possible explanation is the self-absorption of the β particles from nuclei inside the sample. Thus the β measurements would average over nuclei closer to the surface. However, this absorption is only about 6% if the nuclei were all 1μ deep. For our heat treatment, the expected mean diffusion depth of ^{60}Co is 0.8 μ[9] and since this is much less than the skin depth, the distribution of the adiabatic parameter and hence the angle θ will be quite small. Thus self-absorption of the β particles would not then be a likely explanation. Further consideration of the skin depth is given in the Appendix.

In spite of this small systematic difference, we emphasize that the experimental values of θ_f obtained from the β^- and γ measurements agree within the error limits. Therefore θ_f accurately describes the state of the ensemble immediately after a single passage.

III. MULTIPLE-PASSAGE EXPERIMENTS

When the rf is cycled so that the nuclei experience more than one resonant passage in a time which is short compared with the relaxation times, larger γ and β^- signals are obtained. Hence it is interesting to study the state of the ensemble after multiple passages and to determine whether such experiments can also lead to known initial conditions in relaxation studies. The obvious means of analysis is to test whether, after n passages, the orientation parameters can still be written in terms of an angle θ_n such that

$$B_\nu = B_\nu(0)\, P_\nu(\cos\theta_n) \quad . \tag{5}$$

The β^- and γ signals would then be

$$S_\beta = 1 - P_1(\cos\theta_n)$$

and (6)

$$S_\gamma = 1 - P_2(\cos\theta_n)$$

for $x \leq 0.3$.

In our experiments the time for each of the constituent passages was kept to 0.5 sec. The adiabatic parameter was then $A=0.4$. In Fig. 2, values of the effective experimental angle θ_n deduced from the β^- and γ signals are plotted against n. It is seen that there is a definite trend for the angle deduced from the β^- measurements to in-

TABLE I. Experimental and theoretical β^- and γ single-passage signals and corresponding angles for various adiabatic parameters A.

		Theoretical		Experimental	
A	Radiation	Signal	Angle	Signal	Angle
0.4	γ	1.49	88°	0.17 ± 0.03	20 ± 3°
0.4	β	0.97	88°	0.10 ± 0.03	26 ± 4°
2.4	γ	0.20	167°	0.24 ± 0.02	24 ± 2°
2.4	β	1.94	167°	0.11 ± 0.02	27 ± 3°
5.6	γ	0.01	179°	0.28 ± 0.02	25 ± 2°
5.6	β	1.99	179°	0.14 ± 0.02	31 ± 2°
5.6	γ	0.01	179°	0.28 ± 0.02	25 ± 2°
5.6	β	1.99	179°	0.13 ± 0.02	29 ± 2°
5.6	γ	0.01	179°	0.27 ± 0.04	24 ± 3°
5.6	β	1.99	179°	0.09 ± 0.04	25 ± 4°

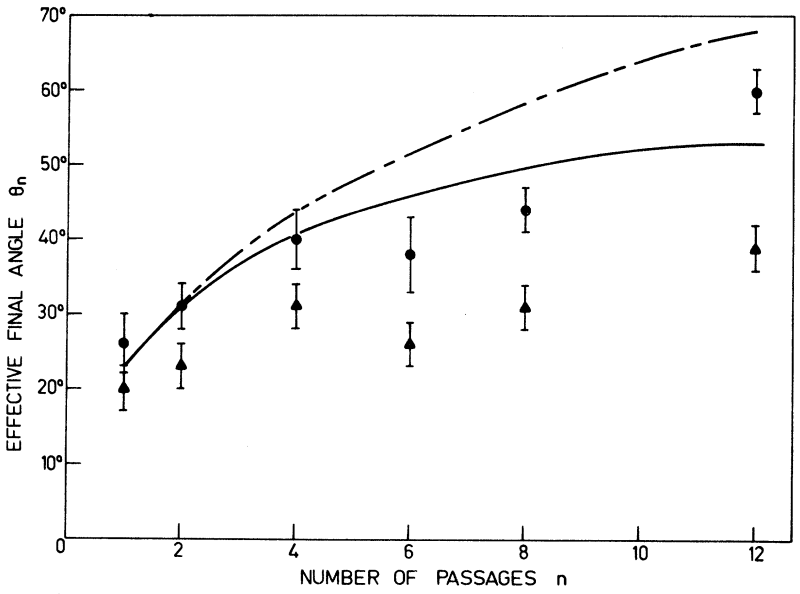

FIG. 2. Dependence of the effective final angle θ_n upon the number of passages n for γ rays (▲) and β^- particles (●). The curves were calculated by using Eq. (7) for γ rays (full) and β^- particles (dashed) determining θ_f by fitting at $n=1$.

crease more rapidly with n than does that from the γ measurements. Hence, unlike single-passage experiments, the orientation parameters after multiple passages are not expressible in terms of a single angle. This result is simple to explain. Because of the inhomogeneous broadening we must consider separately the effects of the passages on a collection of spin packets of nuclei each with a different resonant frequency. Before the first passage the nuclei in each packet are aligned parallel to $\vec{H}_0$ with no oscillatory magnetization components in the perpendicular plane. Since all packets experience the same abiabatic parameter the effect of the first passage is then the same for all of them, resulting in a unique θ_f. However each packet now has an oscillatory perpendicular component of magnetization and the effect of subsequent passages will now depend on the phase between this component and the rf field. Because of the distribution of these phases there will not be a unique θ_n but a wide distribution of θ_n values.

Allowing for this distribution of angles for $n \geq 2$, we can calculate theoretically how θ_n should differ for the β^- and γ signals. Using a simple theory in which only the applied field, hyperfine field, and enhanced rf field are considered we have shown[10] that after averaging over all phases one obtains

$$B_\nu = B_\nu(0)\,[P_\nu(\cos\theta_f)]^n \ . \qquad (7)$$

Choosing $\theta_f = 23°$ to agree with the single-passage point, the signal values were calculated from Eq. (7) and substituted in Eq. (6) to obtain effective values of θ_n which are indicated by the curves in Fig. 2. The final angles and hence signals given by this theory are larger than the observed signals. This discrepancy supports our earlier suggestion[3] that there is an additional retarding field acting on the nuclei.

IV. SPIN-LATTICE RELAXATION TIMES FOR ^{60}Co IN Fe

Before discussing the use of single-passage curves to determine T_1 values it is convenient to briefly review the necessary conditions required by spin-lattice-relaxation theories for the measurement of T_1. The various methods used to measure T_1 by nuclear orientation will then be considered and we show that so far only the single-passage technique and a fast demagnetizing nonresonant method fulfill the conditions necessary for the accurate determination of T_1 for ^{60}Co in Fe.

The theory of spin-lattice relaxation for any rank tensor has been developed in an elegant and general manner by Gabriel[11] and the recipe for its use at low temperatures given by Barclay and Gabriel.[12] An independent approach is given by Spanjaard and Hartmann-Boutron[13] and used by Spanjaard *et al.*,[14] while Bacon *et al.*[15] have given a simpler theoretical picture. In each case the theory shows that accurate measurements of T_1 require a knowledge of the initial and equilibrium conditions so that a multiexponential fit to the experimental anisotropy relaxation data may be made.

For a system with cylindrical symmetry, the time-dependent orientation parameter $B_\nu(t)$ can be expressed, using the notation of Barclay and Gabriel, as

$$\Delta B_\nu(t) = B_\nu(t) - B_{\nu 0} = \sum_{\nu'} G^{00}_{\nu\nu'}\,\Delta B_{\nu'} \quad (t=0) \ , \qquad (8)$$

where $G^{00}_{\nu\nu'}(t)$ are relaxation factors and $B_{\nu 0}$ is the equilibrium value. They are given by

$$G^{00}_{\nu\nu'} = \sum_\kappa A^\kappa_{\nu\nu'} e^{-\Lambda_\kappa t/T_1} \quad (\nu, \nu', \kappa = 1, 2, \cdots, 2I) \ . \qquad (9)$$

The amplitude coefficients $A^{\kappa}_{\nu\nu'}$ can be explicitly calculated from a knowledge of $x = \hbar\omega/2kT$ and the initial conditions. The coefficient Λ_κ depends only on x.[12]

The two distinct approaches to T_1 measurement on oriented nuclei in ferromagnets at temperatures below 0.1 K can then be classified as resonant and nonresonant techniques. In a nonresonant technique suggested by Turrell[16] and independently by Reid *et al.*,[17] the sample is cooled via a fin system by demagnetized paramagnetic salt and then a polarizing field of several tenths T is rapidly applied. The resulting growth in anisotropy is used to determine T_1. The original work assumed a spin temperature and a single-exponential form in the calculation of T_1. In a reevaluation of this method, Spanjaard *et al.*[14] used a multiexponential fit to the experimental data. In their analysis they do not specify the assumed initial conditions. However, Chaplin *et al.*[18] showed that when the thermal time constant for the lattice was very short this technique could result in adiabatic magnetization of the nuclei and hence exhibit no relaxation effects. Increasing the eddy-current heating or increasing the thermal time constant led to significant relaxation effects, but it was not possible to obtain accurate T_1 values because the slower response then prevents rapid establishment of an equilibrium lattice temperature. Further work in which all experimental parameters were carefully varied has shown that between these two extremes a delicate balance could be obtained in which a rapid approach of the lattice temperature to equilibrium occurred but the initial conditions for the relaxation of the nuclei were not known.[19]

A more reliable nonresonant technique of measuring T_1 is as follows. With the polarizing field applied to the sample, the demagnetizing field on the paramagnetic cooling salt is reduced almost to zero where it is held, and the sample is allowed to cool to some initial temperature T_i which can be measured by the resulting anisotropy. When equilibrium is reached, the field is rapidly reduced to zero, allowing the sample to relax down to its final temperature T_f. Since the initial and final conditions are accurately known from the anisotropy measurements, T_1 can be deduced from Eq. (8). The method assumes that the thermal cooling time of the salt–fin-system sample is very fast compared to T_1. In practice this must be monitored by a system with a very short T_1, preferably cooled via the sample of interest. Using Mn^{54} in Cu for this purpose, we cooled the lattice within 0.5 sec and observed the ^{60}Co in Fe relaxation over several minutes as shown in Fig. 1. Gabriel's multiexponential theory was applied to fit such curves and the value of T_1 deduced is shown in Fig. 3 as a function of the lattice temperature. A similar approach to monitor the lattice temperature has been employed by Chilashvili *et al.*[20] for the measurement of the much slower relaxation of $Fe^{65}Zn$. Another nonresonant technique which permitted accurate T_1 measurements to be made was the use by Stone *et al.*[21] of the 40-sec intermediate state in the decay of Cd^{109} to Ag^{109m} to obtain the initial conditions. However,

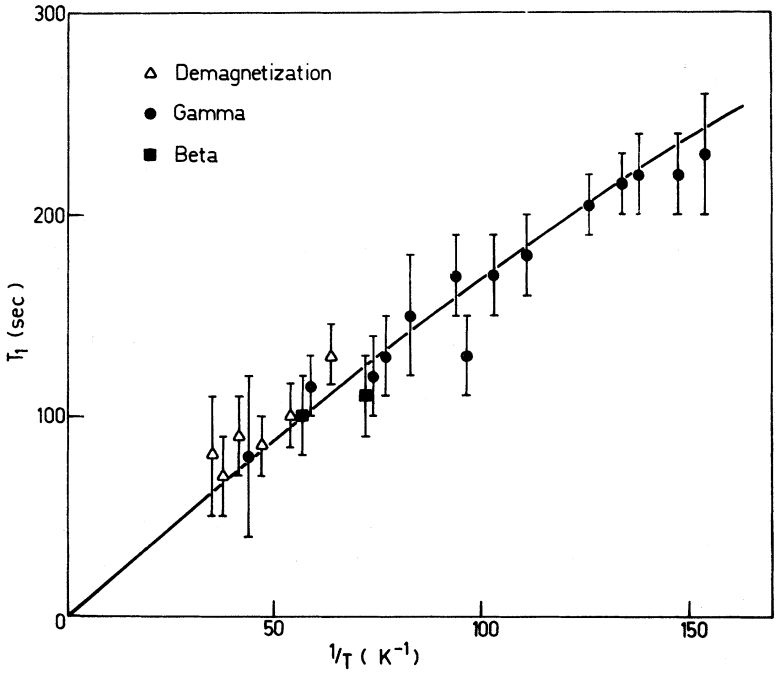

FIG. 3. Variation of T_1 with $1/T$ for both single-passage and demagnetization technique. The curve represents a Korringa constant of 1.75 sec K.

this method is limited to very special nuclear decay systems, i.e., $t_{1/2} \simeq T_1$.

The first measurement of T_1 for $Fe^{60}Co$ by a resonance technique was by Templeton and Shirley,[2] followed by a more detailed study by Brewer *et al.* [22] A single-exponential fit was made to the observed relaxation curves when the rf power modulation was removed, and the resulting relaxation time T_1 for ^{60}Co in Fe showed a marked deviation from the Korringa relation $T_1T = \text{const}$ at a temperature ~ 0.013 K. Attempts to fit the data with a multiexponential theory gave a wide scatter of points mainly because the theory is sensitive to the initial conditions which are not yet well defined during modulation. Recently, Bacon *et al.* [15] have proposed the introduction of a new "magnetic spin-lattice relaxation time" T_μ which avoids the need for known initial conditions. T_μ is measured by using the tail of the decay curve where the rapidly decaying terms of the multiexponential fit will have vanished leaving only a single-exponential term. However, this method requires extremely accurate data in order to test whether or not the decay curve is a single exponential.

As discussed in Sec. II, after a single passage the initial condition of the nuclei before relaxation can be accurately determined from the observed signal. Therefore, we have used the single-passage decay curves to determine T_1 for ^{60}Co in Fe. For a given signal, Gabriel's theory was used to determine the theoretical relaxation curve as a function of t/T_1. This theoretical curve, shown as a solid line in Fig. 1, was then compared to the experimental curve and the value of T_1 which produced the best fit was chosen. Checks were made of the effect of changing the initial choice of signal, the lattice temperature, or the initial starting time. The most sensitive quantity is the temperature, while changes of the assumed initial angle by several degrees still keep the deduced value of T_1 well within the indicated error bars. The choice of initial starting time is least sensitive. The condition used in our analysis was the end of the sweep time, when the signal was deduced by a least-squares fit of a single exponential to the experimental points. However, if an earlier time, say the midpoint of the passage, is chosen as the start of the decay then the corresponding signal is in-

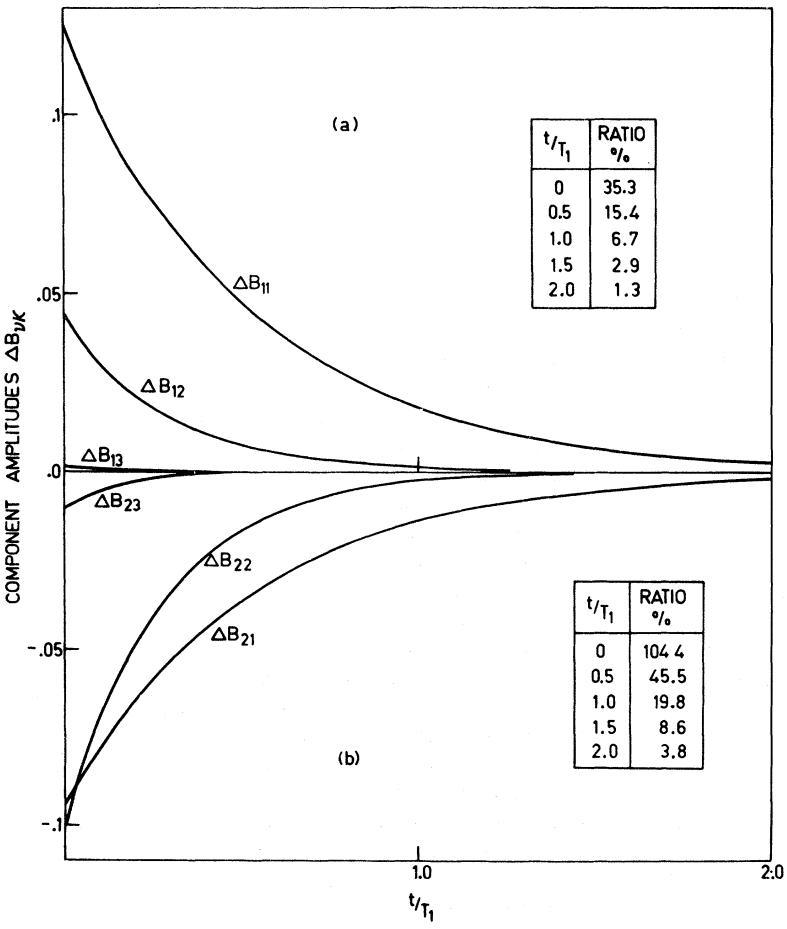

FIG. 4. Component exponentials as a function of t/T_1 for the theoretical (a) β^- single-passage decay and (b) γ single-passage decay shown in Fig. 1. The inserts indicate the ratio expressed as a percentage, of (a) ΔB_{11} to ΔB_{12} and (b) ΔB_{21} to ΔB_{22} as a function of t/T_1.

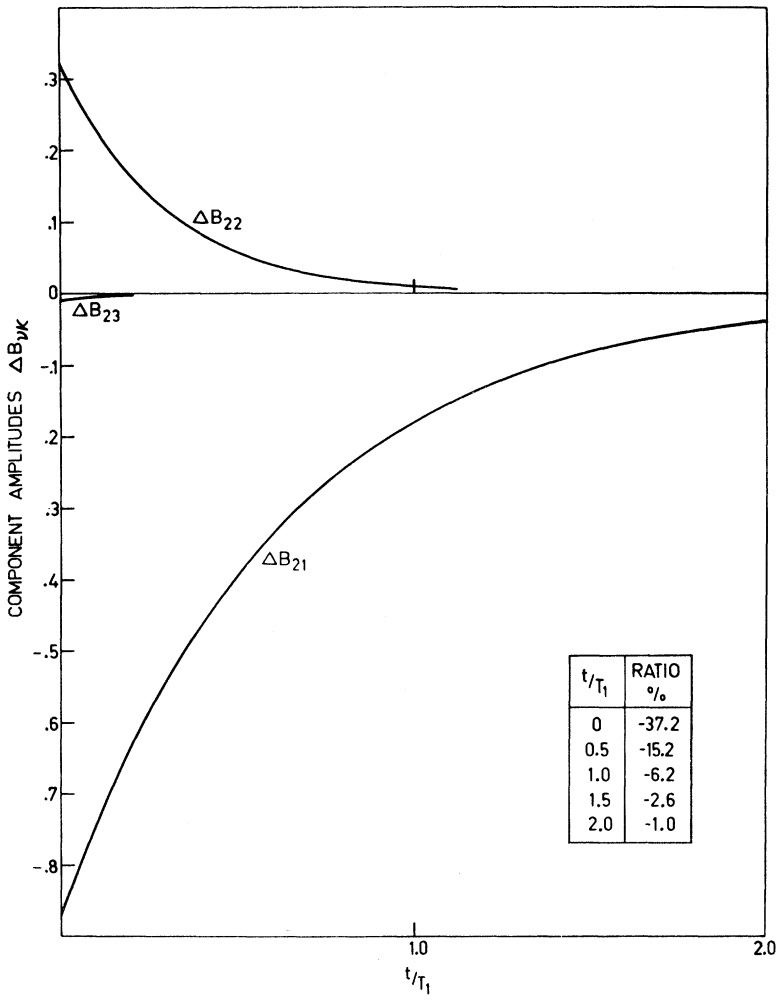

FIG. 5. Component exponentials as a function of t/T_1 for the theoretical demagnetization decay shown in Fig. 1. The insert indicates the ratio, expressed as a percentage, of ΔB_{21} to ΔB_{22} as a function of t/T_1.

creased slightly. The errors in the deduced T_1 value due to this increase in the signal and to the earlier time origin tend to cancel each other.

Figure 1 compares the experimental relaxation curves obtained using the nonresonant demagnetizing method with the curves of single-passage decay. Clearly the nonresonant method results in a longer apparent decay time, even though the lattice temperature is higher and therefore T_1 is shorter than for the single-passage curves. The explanation for the different apparent decay times is illustrated by considering the amplitude of the component exponentials of the theoretical curves in Fig. 1. From Eqs. (8) and (9) we can write

$$\begin{aligned}\Delta B_\nu &= \sum_\kappa \Big(\sum_{\nu'} A^\kappa_{\nu\nu'} \Delta B_{\nu'}\Big) e^{-\Lambda_\kappa t/T_1} \\ &= \sum_\kappa \beta_{\nu\kappa}\, \epsilon^{-\Lambda_\kappa t/T_1} = \sum_\kappa \Delta B_{\nu\kappa} \\ &= \Delta B_{\nu_1} + \Delta B_{\nu_2} + \Delta B_{\nu_3} + \cdots . \end{aligned} \tag{10}$$

These components $\Delta B_{\nu\kappa}$ are shown in Fig. 4 for the β- ($\nu = 1$) and γ-ray ($\nu = 2$) cases. As can be seen from this figure the single-passage relaxation curves cannot be considered as a single exponential until quite large values of t/T_1. Figure 5 shows the component exponentials for the nonresonant demagnetizing method; in this case the decay curve becomes a single exponential faster. The amplitude of the experimental signal and the apparent decay times may be found by summing $\Delta B_{\nu\kappa}$ over κ, adding B_ν (equilibrium), and multiplying by $U_\nu F_\nu$. The longer apparent decay time of the nonresonant method is caused by the opposite sign of ΔB_{22} which tends to reduce ΔB_{21} initially. On the other hand, in Fig. 4 the single-passage-component curves add and thus tend to increase the apparent decay constant. The explanation of the different signs is straightforward. For the nonresonant method the initial conditions $\Delta B_\nu(t=0)$ are the difference between the B_ν at two equilibrium temperatures while for the single-passage method $\Delta B_\nu(t=0) = B_\nu(0)\,[P_\nu(\cos\theta_f) - 1]$. Thus, for the nonresonant method, $\Delta B_1 > -\Delta B_2 > \Delta B_3$, while for single passages, $\Delta B_1 < -\Delta B_2 > \Delta B_3$.

TABLE II. Nuclear-spin-lattice-relaxation Korringa constants for Fe ^{60}Co by various authors. The method of analysis is also shown.

Author(s)	Korringa constant (sec K)	Method	Ref.
Turrell	1.0 ± 0.3	Nonresonant single exp	16
Reid *et al.*	1.0 ± 0.1	Nonresonant single exp	17
Spanjaard *et al.*	2.6	Nonresonant multiexp	14
Templeton and Shirley	0.89 ± 0.04	Resonant single exp	2
Brewer *et al.*	None	Resonant single exp	22
Bacon *et al.*	2.5	Resonant T_μ	15
Bacon *et al.*	1.76	Resonant initial slope	15
This paper	1.75 ± 0.15	Demagnetization multiexp	
This paper	1.75 ± 0.15	Single-passage multiexp	

These initial conditions clearly indicate that an initial spin temperature can be assumed for the fast-cooling method described here but cannot be assumed for the single-passage method. In neither case should a spin temperature be assumed during relaxation of dilute alloys of Fe ^{60}Co.[12] The results of β^- and γ single-passage T_1 measurements are shown in Fig. 3 along with the demagnetization-method results. The good agreement between all three methods with such different initial conditions leads us to the conclusion that these methods are correct. The data are fitted with[12,13]

$$T_1 = (2kK/\gamma hH)\tanh x$$

to obtain a value of the Korringa constant $K = 1.75 \pm 0.15$ sec K. Published results for ^{60}Co in Fe are collected in Table II.

V. CONCLUSION

Our β measurements have confirmed the much smaller rotation of the ^{60}Co nuclei in the ferromagnetic host than expected on simple theoretical grounds. The results give no further explanation of this discrepancy; however, comparison of the β^- and γ signals has shown that the effect of a single passage on the orientation parameters is describable in terms of a single angle of rotation, whereas this is not so for multiple passages. Single-passage experiments and nonresonant fast-cooling experiments both permit relaxation measurements with known initial conditions. For Fe ^{60}Co we obtain $K = 1.75 \pm 0.15$ sec K.

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APPENDIX

The usual definition of the skin depth δ in SI units is[23]

$$\delta = (\tfrac{1}{2}\omega\mu\mu_0\sigma)^{-1/2},$$

where ω is the frequency of the rf, σ is the conductivity, μ is the relative permeability, and μ_0 is the permeability of free space. In calculating the skin depth one must use the rf permeability μ for a polarized Fe specimen. Johnson and Rado[24] give the real and imaginary parts of μ at 200 MHz to be ~1 and ~0, respectively, above 0.2 T applied field. Using the resistivity of 99.98 at.% Fe = 10^{-7} Ωm[25] one finds at room temperature $\delta f^{1/2} = 0.159$, with f in hertz and δ in meters. This value differs from that given in the *American Institute of Physics Handbook*[26] because they use the initial incremental permeability rather than the fully magnetized rf permeability. Assuming a resistivity temperature ratio of 10 for our samples, $\delta = 3.88$ μ at 165 MHz. This number has been indirectly experimentally verified as follows. We have observed that with full modulation over the entire resonance line the maximum signal obtainable is 0.62 ± 0.02[10] in a 1-μ polycrystalline Fe^{60}Co foil as well as in a single-crystal source with the mean diffusion depth of 0.8 μ. However, using a 8.5-μ polycrystalline foil we obtain a saturation signal of 0.35 using full modulation. We can use this to estimate an experimental δ by evaluating

$$s(8.5\mu)/s(\max) = 0.35/0.62 = \int_0^{8.5} e^{-x/\delta}dx \Big/ \int_0^{8.5} dx.$$

From this we obtain $\delta \approx 7$ μ, which suggests that our assumed resistivity ratio of 10 is an upper limit. This also means our ^{60}Co nuclei are well within the skin depth and therefore experience a relatively unique $\vec{H}_1$.

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