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Nuclear Spin-Lattice Relaxation Caused by Paramagnetic Impurities*

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The behavior of the nuclear magnetization has been examined for the transient region immediately following saturation of the nuclear spin system in the case in which the nuclear relaxation is limited by spin diffusion to paramagnetic impurities. Relaxation effects in the absence of diffusion and the presence of rapid diffusion are discussed. An experiment is reported which substantiates the calculations for the case of diffusion-limited relaxation and, in addition, allows a determination of the correlation time of the magnetic moment of the paramagnetic ion and the coefficient of nuclear spin diffusion.

I. INTRODUCTION

SEVERAL authors[1–3] have considered the problem of the spin relaxation of nuclei in a crystal containing paramagnetic impurities when the transfer of spin energy to the lattice is limited by the diffusion of nuclear spin energy. It is the purpose of this paper to examine the problem of the recovery of nuclear magnetization after saturation for times very short compared to the relaxation time and to consider the problem of nuclear relaxation caused by paramagnetic impurities for two cases not discussed by the above authors. The results of an experiment are reported which show that the behavior of the nuclear relaxation follows the theoretical expression derived here.

Bloembergen[1] observed some relaxation effects which he attributed to the effect of paramagnetic impurities. He assumed that the coupling between the magnetic moment of the paramagnetic ion and the magnetic moments of the nuclei in the crystal was via terms in the dipolar Hamiltonian. Thus the transition probability of a nucleus at a distance r from the impurity is

$$P=(3/2\pi)(\gamma_p\gamma_n\hbar)^2S(S+1)r^{-6}\times\sin^2\theta\cos^2\theta\tau(1+\omega^2\tau^2)^{-1},\quad(1)$$

where θ is the angle the line joining the nucleus and the impurity makes with the magnetic field, and γ_p and γ_n are the magnetogyric ratios of the paramagnetic ion and the nucleus, respectively. The correlation time of the z component of the spin S of the paramagnetic ion is denoted by τ. In dilute crystals, where we neglect any interaction between impurities, τ becomes the spin-lattice relaxation time of the paramagnetic ion.

Bloembergen further assumed that the interaction between nuclear spins could be characterized by a characteristic time T_{ss} for spin-spin exchange of Zeeman energy. If the probability $(1/T_{ss})$ of a mutual spin flip between a given spin and one of its neighbors is independent of all previous spin flips involving either of them, this amounts to a random-walk problem of characteristic time increment T_{ss} and step distance a, the spacing between like nuclei. Thus the problem is describable in terms of the diffusion equation, provided that times much longer than T_{ss} and distances much greater than a are involved in the problem. Bloembergen derived the approximate relation for the nuclear spin diffusivity: $D\cong a^2/50T_2$, where T_2 is the transverse relaxation time of the nuclei.

If the length dimensions appearing in a problem are much greater than a, the lattice may be treated as a continuum. We define a nuclear spin magnetization density by

$$p(\mathbf{r},t)=\langle M_z(\mathbf{r},t)\rangle/a^3.\quad(2)$$

The expectation value of M_z may vary with both time and position in the crystal in a nonequilibrium situation.

Since the local magnetic field in the neighborhood of a paramagnetic ion is large, the Larmor frequency of some nuclei near the ion will be different from those in the bulk of the crystal. This has two effects: first, these nuclei do not contribute to the nuclear resonance line, and, second, these nuclei cannot undergo spin-spin transitions with the remaining nuclei in the resonance line. We may say, roughly, that all nuclei within a sphere of radius b are removed from the resonance line, where b is defined as that distance at which the magnetic field due to the ion equals the local dipolar field of the crystal. If the moment of the paramagnetic ion is static, this is given approximately by

$$b=(\mu_p/\mu_n)^{\frac{1}{3}}a,\quad \tau\gg T_2.\quad(3)$$

If the z component of the spin of the paramagnetic ion is fluctuating rapidly in a time T_2, however, the local field of the ion is motionally averaged, and only this average field is felt at a nuclear site. Thus we write

$$b=(\mu_p{}^2H/\mu_n\kappa T)^{\frac{1}{3}}a,\quad \tau\ll T_2.\quad(4)$$

We assume that there can be no spin diffusion for $r<b$, and thus we call b the diffusion barrier radius. It is possible that removal from the resonance line and quenching of diffusion do not occur at the same radius, but this would not influence the following calculations appreciably.

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[1] N. Bloembergen, Physica 25, 386 (1949).

[2] G. R. Khutsishvili, Proc. Inst. Phys. Acad. Sci. Georgia (U.S.S.R.) 4, 3 (1956).

[3] P-G. de Gennes, J. Phys. Chem. Solids 7, 345 (1958).

It is assumed that the concentration N of paramagnetic ions is sufficiently small that $(4\pi b^3/3)N$ is a small fraction of the volume of the crystal. Thus most of the nuclei are in local magnetic fields not perturbed by the proximity of a paramagnetic ion.

If we neglect the detailed angular dependence in (1) by averaging over θ(av $\sin^2\theta\cos^2\theta=2/15$), we may write

$$P\cong(1/5\pi)(\gamma_p\gamma_n\hbar)^2S(S+1)r^{-6}\tau(1+\omega^2\tau^2)^{-1}=Cr^{-6}, \quad (5)$$

where now C is independent of the position of a given nucleus in the crystal. The diffusion equation to be solved for the behavior of the nuclear spin system in the absence of radio-frequency excitation is then[3]

$$\partial p/\partial t=D\nabla^2p-C(p-p_0)\sum_n|\mathbf{r}-\mathbf{r}_n|^{-6}, \quad (6)$$

where $\mathbf{r}_n$ is the location of a paramagnetic ion, and p_0 is the thermal equilibrium value of p. In the region of a given paramagnetic impurity, which we take as the origin, we may write the equation approximately as

$$\partial p/\partial t=D\nabla^2p-C(p-p_0)r^{-6}. \quad (7)$$

There are several length parameters which characterize a problem of relaxation by spin diffusion to a paramagnetic ion. Of these, we have already introduced a, the internuclear spacing, and b, the barrier radius. A third is the average distance R between impurities, which is of the order of $N^{-\frac{1}{3}}$. A fourth length parameter ρ is defined by Khutsishvili[2] as

$$\rho=\frac{\pi}{2^{\frac{5}{2}}[\Gamma(5/4)]^2}(C/D)^{\frac{1}{4}}=0.68(C/D)^{\frac{1}{4}}. \quad (8)$$

This has been called the pseudopotential radius by de Gennes,[3] who developed an elegant solution to the diffusion equation using scattering theory. He has examined the solution to (6) for the case in which a and $b\ll\rho\ll R$ and has obtained the result that the return of the nuclear magnetization to its equilibrium value approaches an exponential function of time for a sufficiently long time after a disturbance of the magnetization. The characteristic time T_1 of this exponential function is given by

$$1/T_1=4\pi N\rho D=8.5NC^{\frac{1}{4}}D^{\frac{3}{4}}. \quad (9)$$

This agrees with the result of Khutsishvili, who took a much different approach to the problem and obtained (9) by solving equation (7) in the approximation that at large t, $\partial p/\partial t$ could be set to zero.

II. THEORY

A. Case of No Spin Diffusion

It will be of some interest to solve (7) for the case in which there is no spin diffusion. It is not possible to make the spin diffusion vanish, but it can be made quite small by making the internuclear spacing large. This can be accomplished by substituting a given species of nucleus dilutely into a crystal not containing that species. In such a case, it is not difficult to violate the requirement $\rho\ll R$ for de Gennes's solution to be valid.

If diffusion is not important, then the equation to be solved is

$$\partial p/\partial t=-C(p-p_0)\sum_n|\mathbf{r}-\mathbf{r}_n|^{-6}, \quad (10)$$

which has the solution

$$p=p_0[1-\exp(-Ct\sum_n|\mathbf{r}-\mathbf{r}_n|^{-6})]. \quad (11)$$

If the total nuclear magnetization is measured after a time t, the value will be

$$M_z(t)=\int pd^3r, \quad (12)$$

where the volume integral is to be carried out over the entire crystal, excluding the nuclei within each barrier radius. It is impossible to perform this integral unless the details of the distribution of paramagnetic ions in the crystal are known.

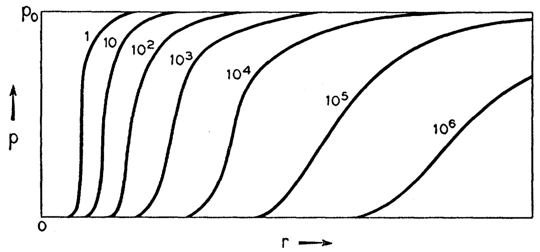

FIG. 1. Theoretical curves for the nuclear magnetization density p vs distance r from an impurity for various values of the time for the case in which there is no nuclear spin diffusion.

For a short time after the nuclear magnetization has been disturbed, any given nucleus will be influenced primarily by the nearest paramagnetic ion, and we can write an approximate solution for p as

$$p\cong p_0[1-\exp(-Ctr^{-6})], \quad \text{(small } t\text{)}, \quad (13)$$

where r is the distance from the nearest paramagnetic ion. We take the case of $p(\mathbf{r},0)=0$, i.e., initially the nuclear magnetization is saturated, and then it is possible to evaluate (12) for small t:

$$M_z(t)\cong(4\pi^{\frac{3}{2}}/3)NC^{\frac{1}{2}}t^{\frac{1}{2}}, \quad t>b^6/C. \quad (14)$$

We note that in this case the magnetization increases as the square root of time, after a time b^6/C. For $\tau<T_2$, this time is of the order of a microsecond, but in other cases it may become as large as one second.

Figure 1 shows p vs r for the region near an impurity with the time as a parameter. Note that, for each value t, p approximately defines a sphere in which the nuclei have returned to thermal equilibrium. We may define the radius r_0 of this sphere by $p(r_0,t)=1-e^{-1}$ or

$$r_0=(Ct)^{1/6}. \quad (15)$$

This solution for the case $D=0$ is of no physical importance at a long time after a disturbance of the nuclear magnetization. If it is required that the effects of diffusion be negligible, we must have $(Dt)^{\frac{1}{2}}<r_0$. As r_0 approaches R, this becomes $(DR^6C^{-1})^{\frac{1}{2}}<R$. For typical values of $N=10^{18}$ ions per cm^3 ($R=10^{-6}$ cm) and $C=10^{-40}$ cm^6/sec, this requires $D<10^{-56}$ cm^2/sec, which is absurdly small, even for very dilute nuclear systems. Thus one would gain no insight into a physical system by an evaluation of (12) with t large and using p from (11).

B. Case of Diffusion-Limited Relaxation

As Khutsishvili and de Gennes have shown independently, for a sufficiently large time $M_z(t)$ approaches $M_z(\infty)[1-\exp(-t/T_1)]$. For a short time after a disturbance of the nuclear magnetization, however, their solution does not hold. If initially the magnetization is saturated, there is no gradient of magnetization density, and, thus, diffusion cannot be of importance at the start of the relaxation process. One would therefore expect that the solution (14) would hold for a short time following saturation of the nuclear resonance line. In fact, it should be valid until $(Dt)^{\frac{1}{2}}\cong r_0$ or for $t<C^{\frac{1}{2}}D^{-\frac{3}{2}}$. For a typical physical system, this time may be as large as one second. Thus we expect $M_z(t)$ to start as $t^{\frac{1}{2}}$ and proceed asymptotically to an exponential function of time.

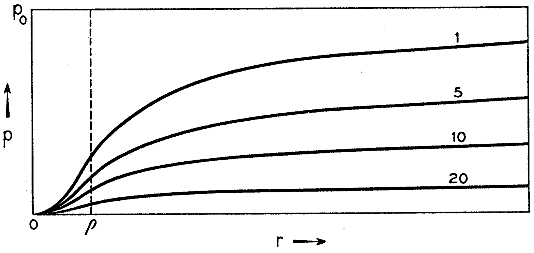

FIG. 2. Theoretical curves for the nuclear magnetization density p vs distance r from an impurity for various values of the time for the case in which the nuclear relaxation is limited by the diffusion of nuclear spin energy to the paramagnetic ion. The radius ρ is the pseudopotential radius of de Gennes.

Khutsishvili's[2] theoretical function of p vs r for various values of the time are plotted in Fig. 2. Note that, in contrast to the case with no diffusion, the sphere of radius ρ in which most of the nuclei are relaxed does not enlarge with time. This may be compared to de Gennes's approach to the problem in which he set $p(r,t)=0$ for all $r<\rho$ and solved for the behavior of the magnetization outside this sphere.

C. Case of Rapid Diffusion

Part of de Gennes's solution of the problem of diffusion-limited relaxation involved a boundary condition imposed on (6) at $r=\rho$. If $b>\rho$, however, (6) does not hold at $r=\rho$, and the method fails. Thus de Gennes's solution applies only when $\rho>b$. It is a rather common occurrence, however, that this restriction is violated. When this happens, it means, physically, that nuclear Zeeman energy can diffuse to the paramagnetic ion faster than the paramagnetic ion can transmit it to the lattice. It is clear that in the limit $\rho\ll b$ the nuclear magnetization can be redistributed among the nuclear spins fast enough to maintain an internal equilibrium while it slowly approaches its thermal equilibrium value. Thus it would be expected for this case that $M_z(t)$ would follow an exponential function of time for all values of t and that the initial region in which M_z is proportional to $t^{\frac{1}{2}}$ would be missing.

Figure 3 shows p vs r for the case of $b=5\rho$. Khutsishvili's general solution has been used, and the boundary condition that there be no diffusion across the surface of the sphere $r=b$ has been satisfied. The curves for p for any time are approximately flat, indicating that the local spin systems in all parts of the crystal are approximately at the same spin temperature as the relaxation proceeds. If this is true, we may take p to be a function of time only, and then the rate at which nuclear spin energy can be transmitted to the lattice is

$$\frac{1}{p}\frac{dp}{dt}=4\pi N\int_b^\infty (Cr^{-6})r^2dr=(4\pi/3)NCb^{-3}. \qquad (16)$$

There are two ways to distinguish between the case of diffusion-limited relaxation and the case of rapid diffusion. Most obvious is the existence of the transient region in the first case in which $M_z\sim t^{\frac{1}{2}}$. Second, there is a difference in the dependence of the relaxation rate on the magnetic field. This dependence falls into several cases according to the relative magnitudes of τ, T_2 and ω, the Larmor precessional frequency of the nuclei. We write explicitly the dependence of C and b on H as

$$\begin{aligned} &C\sim H^{-2}, && \omega>1/\tau,\\ &C\sim H^{0}, && \omega<1/\tau,\\ &b\sim H^{-1}, && \tau<T_2,\\ &b\sim H^{0}, && \tau>T_2, \end{aligned} \qquad (17)$$

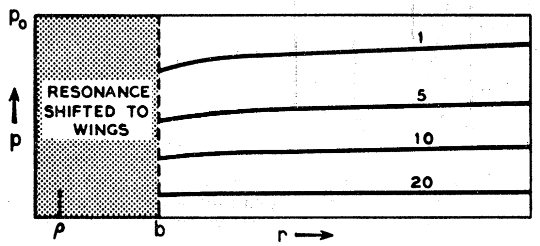

FIG. 3. Theoretical curves for the nuclear magnetization density p vs distance r from an impurity for various values of the time for the case in which the nuclear spin energy can diffuse toward the paramagnetic ion faster than the ion can transmit it to the lattice. The barrier radius b is here chosen as five times the pseudopotential radius ρ.

TABLE I. Magnetic field dependence of relaxation rate

Restrictions[a]	Diffusion-limited case	Rapid diffusion case
$\tau<1/\omega$, $\tau<T_2$	H^0	H^{-1}
$\tau>1/\omega$, $\tau>T_2$	$H^{-\frac{1}{2}}$	H^{-3}
$\tau>1/\omega$, $\tau>T_2$	$H^{-\frac{1}{2}}$	H^{-2}

[a] It is assumed that τ is independent of H.

provided τ is independent of H. If the dependence of τ on H is known, it can easily be included in (17). Combining these dependences on magnetic field, we obtain from (9) and (16) the resulting dependence for $1/T_1$ shown in Table I. From an examination of Table I, we may, for example, conclude that the experiment of Uehling and Bjorkstam,[4] who found that the relaxation times of H^1 and P^{31} nuclei in certain crystals were linearly dependent on the magnetic field, falls into the case of rapid diffusion with $\tau<1/\omega$ if, for their impurity, τ was independent of H.

III. EXPERIMENT

Since the transient region in which $M_z\sim t^{\frac{1}{2}}$ had not been reported in the literature, an experiment was undertaken to search for this phenomenon. The nuclei chosen were protons in NH_4HSO_4. This compound was chosen because of its high density of protons and its long intrinsic relaxation time at room temperature (29 sec). This compound was doped with $(NH_4)_2CrO_4$, which dissolves readily in fused NH_4HSO_4. Polycrystalline samples were made in relative concentrations of 1, 2, 4, 8, and 16 by successive dilution. The most dilute sample contained 5.3×10^{17} Cr ions per cm^3. It is assumed that the Cr ions exist in the crystal in the Cr^{+++} $(S=\frac{3}{2})$ state.

A pulsed nuclear resonance experiment[5] was carried out on each of these samples and an undoped sample at a magnetic field of 3500 gauss (12-Mc/sec Larmor frequency for the protons). The apparatus was programmed to operate in the following manner. The radio-frequency transmitter was triggered to give a series of ten $\pi/2$ pulses[6] spaced 50 microseconds apart. Since the T_2 of this compound is approximately 35 microseconds, there was no appreciable coherent effect between pulses, and thus the ten pulses saturated the nuclear spin system. At a time t later, the transmitter was triggered to give another $\pi/2$ pulse to measure the nuclear magnetization. The detected free precession signal resulting from the last $\pi/2$ pulse was displayed directly on an oscilloscope. The signal-to-noise ratio was sufficiently large (>100) to allow measurements to be taken directly from the face of the oscilloscope.

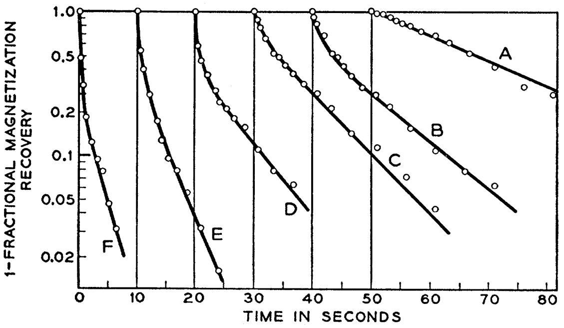

FIG. 4. Nuclear magnetization recovery vs time for the pure sample (A) and each of the doped samples (B–F). The abscissa has been shifted 10 sec for successive samples to separate the curves.

Results of the experiment for $t>0.2$ second are presented in Fig. 4. The ordinate of Fig. 4 is $1-M_z(t)/M_z(\infty)$ plotted on a logarithmic scale. The abscissa has been shifted 10 seconds for successive samples to separate the curves. It is noted that the effects of the transition region are quite apparent for all the samples containing Cr. Eventually, however, all these curves approach an exponential time dependence. Thus it is concluded that the relaxation is diffusion-limited. The pure sample is observed to have an exponential dependence for all time. We may say that its relaxation, if caused by paramagnetic impurities, is not diffusion-limited. The final rate W of each of these exponentials may be expressed as

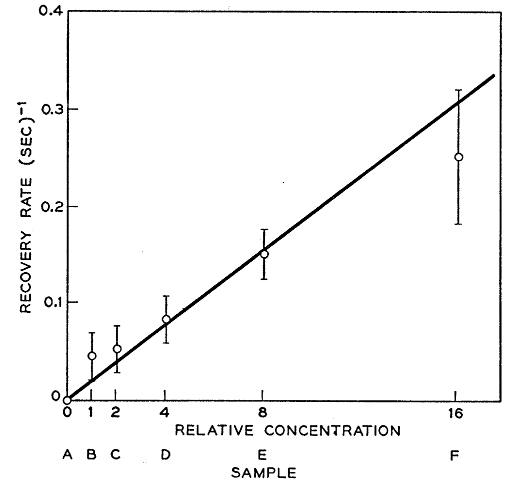

FIG. 5. Asymptotic rate of relaxation due to paramagnetic impurities vs concentration. The relative concentration 1 corresponds to 5.3×10^{17} Cr ions per cm^3.

$$W=1/T_{1\ \text{intrinsic}}+1/T_{1\ \text{paramagnetic}}. \qquad (18)$$

De Gennes[3] has shown that, as long as the intrinsic relaxation rate is not large, the rates due to it and the

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[4] E. A. Uehling and J. L. Bjorkstam, Bull. Am. Phys. Soc. **3**, 318 (1958).

[5] The pulsed nuclear resonance apparatus used has been described in detail by E. G. Wikner, thesis, University of California, 1959 (unpublished).

[6] E. L. Hahn, Phys. Rev. **80**, 580 (1950).

effects of impurities are additive. From (18) and the final slopes of the curves of Fig. 4, the relaxation rate due to the impurities was calculated and is shown in Fig. 5. The errors of the points, especially that of sample F, are rather large, but one can say that there is an approximate linear dependence of the impurity relaxation rate on concentration as would be indicated by (9) or (16).

Figure 6 shows the experimental points taken for $t \leqq 0.2$ second. The ordinate is $M_z(t)/M_z(\infty)$ and is plotted against the square root of time. The ordinate of curves for successive samples has been shifted upward by 0.02 to prevent overlap of the experimental points. It is observed that, for each of the samples doped with Cr, the signal rises proportional to $t^{\frac{1}{2}}$, but for the pure sample no signal at all was observed in the first 0.2 second. This would indicate that Eq. (14) holds even for the smallest time shown, 10^{-3} sec. The slopes of these curves should be proportional to the concentration as is indicated in (14). The slopes of these curves are plotted against concentration in Fig. 7. An approximate linear dependence is observed.

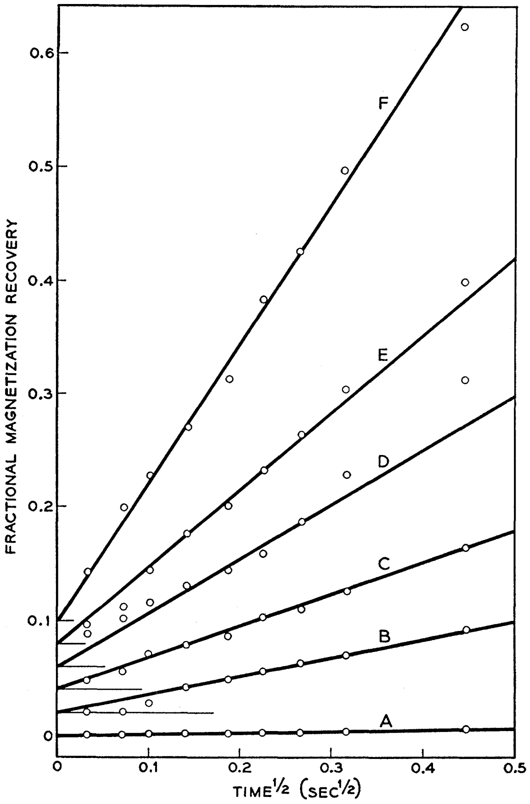

FIG. 6. Nuclear magnetization recovery in the first 0.2 seconds after saturation for various samples (A–F). The abscissa is the square root of the time. Successive curves have been displaced vertically 0.02 to prevent overlapping of the experimental points.

From (14) it is seen that the slope of the line in Fig. 7 should be $(4\pi^{\frac{3}{2}}/3)C^{\frac{1}{2}}$. Since this factor involves τ for the paramagnetic ion spin as the only unknown, it may be used to obtain τ. In this case, one obtains $\tau = 10^{-8}$ sec. This value is not inconsistent with the observed paramagnetic resonance[7] line width of the most concentrated sample and the observed saturation behavior of the resonance line. The total line width, undoubtedly inhomogeneously broadened due to the polycrystalline nature of the samples, was 700 gauss so that $\tau > 2 \times 10^{-10}$ sec. With a microwave field H_1 of approximately 0.3 gauss, the system was only partially saturated, indicating $\gamma_p H_1 \tau < 1$ or $\tau < 2 \times 10^{-7}$ sec.

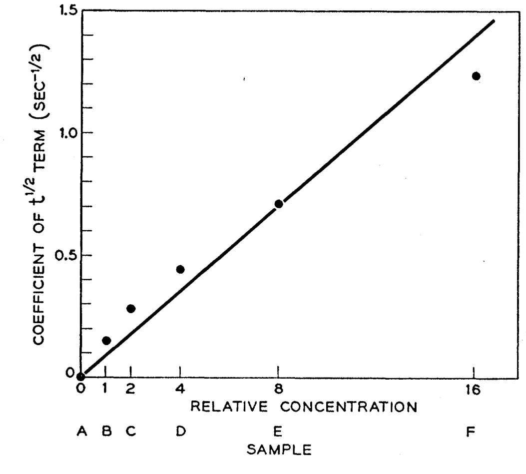

FIG. 7. Slope of the lines of Fig. 6 (coefficient of the $t^{\frac{1}{2}}$ term) vs concentration of Cr ions.

Using the value of C obtained from Fig. 7, it is now possible to calculate the coefficient of diffusion of the nuclear spin system from the slope of the line in Fig. 5. This slope, from Eq. (9), should be $8.5C^{\frac{1}{4}}D^{\frac{3}{4}}$, which gives $D = 10^{-14}$ cm²/sec. Unfortunately the protons in NH_4HSO_4 are not arranged on a simple lattice, and it is not feasible to compute a theoretical value for D. This value of D is somewhat smaller than the value expected by Bloembergen[1] for a cubic lattice, 10^{-12} cm²/sec. This might be expected if the protons in NH_4HSO_4 lie in groups of five with relatively weak interaction between groups.

IV. CONCLUSION

We have seen that of the three possible cases of nuclear relaxation by paramagnetic impurities, (1) relaxation without spin diffusion, (2) diffusion-limited relaxation, and (3) relaxation with rapid diffusion, only the latter two are of physical importance, but a solution for the first case suggests a behavior for the second for a very short time after a disturbance of the nuclear magnetization. We have seen that the latter

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[7] The author is indebted to D. T. Teaney, who allowed his equipment to be used for this measurement. The equipment used has been described by A. M. Portis and Dale Teaney, J. Appl. Phys. 29, 1692 (1958).

two cases can be distinguished in two ways: by the existence or nonexistence of a transient region of $M_z(t) \sim t^{\frac{1}{2}}$ and by the magnetic field dependence of $M_z(t)$ for large t. In either case, it is possible to compute C and thus determine τ for the paramagnetic ion independent of a paramagnetic resonance experiment. In the case of diffusion-limited relaxation, it is also possible to calculate the coefficient of spin diffusion from the asymptotic behavior of $M_z(t)$ for large t.

ACKNOWLEDGMENTS

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Gyromagnetic Ratios of Fe and Ni

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It has been shown that the previously observed decrease in the values of g' for weakly magnetized specimens of Fe and Ni was caused by a systematic error in the measurement of magnetic moment. Recent experiments on these two metals indicate g' values of 1.919 ± 0.002 for Fe and 1.835 ± 0.002 for Ni.

INTRODUCTION

A NEW laboratory has recently been built for the purpose of making measurements of the mechanical inertia effects associated with magnetism.[1] This laboratory was designed with the objective of obtaining a working space in which magnetic fields could be reduced to the order of 10^{-5} to 10^{-6} oersted. All ferromagnetic materials were eliminated from the building construction.

Much of the equipment used is similar in design to that previously reported,[2-4] however, many refinements have been made. The variometers for following changes in the earth's magnetic field are considerably altered and the changes are now followed by photoelectric servo systems.

Refinements have also been made in the previously used procedures[3,4] for measuring angular momentum and magnetic moment changes. Electronic timers are now used to supply current reversals automatically to the torsional pendulum system at the proper intervals to obtain resonance. Also as a precautionary measure the magnetic moment changes of the sample are monitored throughout the experiment. This is accomplished by a pair of pickup coils surrounding the instrument in which the ferromagnetic sample is resonating. Reversal of the magnetic moment induces a current pulse in these pickup coils. This pulse is led through the secondary of a mutual inductance to a ballistic galvanometer. Current in the primary of the mutual inductance is reversed simultaneously with that flowing in the magnetizing winding of the sample. After adjusting for a null the primary current is determined by measuring its drop across a standard resistor. This monitoring system is calibrated by the torsional comparator previously used.[3,4]

RESULTS

It was found that the previously observed change in the values of the gyromagnetic ratios for weakly magnetized specimens of Fe, Ni, and the FeNi alloys,[4-6] was caused by a systematic error in the determination of magnetic moments.

In the nonlinear initial region of the B-H curve the induced magnetization is a function of the rate at which H is applied. In our old equipment a conducting ring surrounded the specimen in the torsional comparator. The additional circuit damping introduced by this ring slowed the rate at which H changed. This resulted in a decrease in the magnetic moment change when weakly magnetized specimens of these metals were reversed. Since this ring was not used when the corresponding angular momentum changes were measured, an error resulted. This effect became insignificant for the larger values of current used in the older work. Hence all of the high intensity values in references 4–6 are reliable.

A new series of experiments on a different sample of

TABLE I. Analysis of ellipsoidal Fe sample.

Iron	99.89%
Nickel	0.05%
Silicon	0.01%
Oxygen	0.008%
Cobalt	0.005%

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[1] Constructed by the Charles F. Kettering Foundation.

[2] G. G. Scott, Rev. Sci. Instr. **28**, 270 (1957).

[3] G. G. Scott, Phys. Rev. **82**, 542 (1951).

[4] G. G. Scott, Phys. Rev. **99**, 1241 (1955).

[5] G. G. Scott, Phys. Rev. **99**, 1824 (1955).

[6] G. G. Scott, Phys. Rev. **103**, 561 (1956).