Nonexponential Decay of Nuclear Spin Echoes in Ordered Magnetic Materials at Low Temperatures*

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We report here the first direct measurement of the nonexponential decay constant k in ordered magnetic materials. Results of our measurements of k_d and k_a, corresponding to the tetrahedral (d) and the octahedral (a) lattice sites, are presented here for Fe^{57} nuclear spin echoes in yttrium iron garnet (YIG) at 4.2 °K and below. The results can be understood on the basis of a model of fluctuations in hyperfine field caused by long-wavelength spin fluctuations. In this connection, we have extended the model of Klauder and Anderson to show that the nonexponential component of the decay of spin echoes arises from fluctuations in resonance frequency. The decay constant k can be related directly to the time-dependent correlation function $\psi(t)$ of such fluctuations. We have demonstrated this for both the Gaussian and the Lorentzian processes. On applying this model to ordered magnetic materials, we have shown that for a Heisenberg ferromagnet at low temperatures, where the long-wavelength spin fluctuations are most predominant, $k=\beta T^{5/2}$, where β involves known parameters. In deriving this relation, we have used the frequency- and wave-vector-dependent longitudinal-spin correlation calculated by Vaks *et al.* Extending this idea to a multisublattice system, we get a relation $k=\alpha+\beta T^{5/2}$ valid for each sublattice at sufficiently low temperatures. When our data are fitted with this relation, we get $\alpha=1.7\times10^4$, $\beta=6.5\times10^2$ for the a site, and $\alpha=-6.5\times10^3$, $\beta=7.9\times10^2$ for the d site. (The units of α and β are sec^{-2} and $sec^{-2}\,°K^{-5/2}$, respectively.) Both k_d and k_a are obtained taking the nonexponential decay to be Lorentzian, as is suggested by the measurements. With this model of spin fluctuations we find fairly good agreement between theory and experiment. The possible source of α has been argued to be the zero-point spin fluctuations, and our results strongly suggest this viewpoint.

I. INTRODUCTION

In spin echoes,[1] the nonexponential decay (NED) is almost as common as the more conventional exponential decay which is mainly due to relaxation effects. Since in most cases the relaxation decay is more prominent than the NED, not much attention has been paid so far to understanding it or to establishing its relation to other physical properties of interest. In liquids, the NED is usually very prominent, is well understood, and has been related to the molecular self-diffusion.[1-5] In solids, in general, this is not the case; however, the phenomenon is very difficult to observe, particularly because of the presence of a strong exponential relaxation decay and in view of the possibility of the existence of more than one relaxation rate. Only in very special cases, such as electron spin echoes in strongly inhomogeneous systems[6] where this decay is very prominent, has some effort been made to understand it.[7] Similar decay has been observed in ordered magnetic materials by different workers.[8-10] But, owing to the complexity in the nature of the decay of nuclear spin echoes in such materials, and to the difficulty in its quantitative determination, little progress has been made so far in understanding its origin. This decay is generally believed to be due to (i) spin-diffusion in Bloch walls,[8] and (ii) thermal fluctuations of Bloch walls.[10] The first is expected not to be very effective for nuclei of small moment and low abundance, such as Fe^{57} and Ni^{61}, as has been demonstrated by Weger[10] for magnetically ordered metallic systems. Other possible sources of the NED have been discussed by the present author,[11] the important ones in a magnetically ordered system being (a) fluctuations in the hyperfine field, and (b) fluctuations in the dipolar field.

In order to have a better understanding of the NED in magnetically ordered systems, we have undertaken a program to measure this decay constant for Fe^{57} in yttrium iron garnet (YIG). Fe^{57} is particularly suitable for such studies, mainly because (i) Fe^{57} is a small-moment and low-abundance nucleus; (ii) iron is present as S-state Fe^{3+} ions having a spin-only moment and occupies both the tetrahedral (d) and the octahedral (a) lattice sites in a known ratio; (iii) the Fe^{57} resonance frequences for the a and d sites are widely separated, and the signals are fairly strong over a large temperature range; and (iv) hyperfine and dipolar fields are well known for both sites. Furthermore, many investigations, both theoretical and experimental, have already been made on this material, and its magnetic properties are fairly well known.

In order to avoid the difficulty and the ambiguity in the measurement of the NED mentioned earlier, we have employed the method originally due to Sinha and the present author.[4,5] As will be dis-

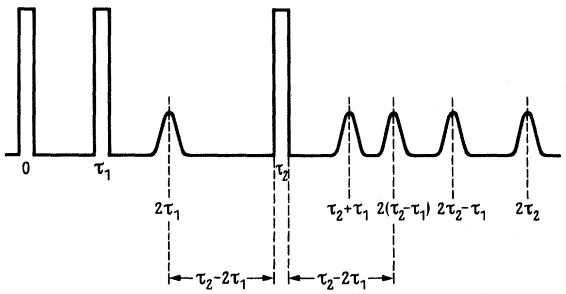

FIG. 1. Three-pulse-echo sequence. The rf pulses are applied at times 0, τ_1, and τ_2 and their durations have been assumed to be negligible in comparison to any pulse separation. The image echo at time $2(\tau_2-\tau_1)$ is shown to form owing to a "virtual" pulse at time $2\tau_1$ and the third pulse at τ_2.

cussed in what follows, this method is capable of measuring the NED even in the presence of a strong exponential decay expressible by one or more time constants. Hence, it is expected to be free of uncertainties inherent in methods used by earlier workers. For these reasons, the method is very suitable for such studies, and to our knowledge this is the first such report on directly measuring the NED in a solid, especially in a magnetically ordered solid.

In the case of liquids, it is well known that the NED arises from the fluctuations of the resonance frequency caused by the Brownian motion of molecules carrying the resonating nuclei in an inhomogeneous external magnetic field.[1–5] These fluctuations are Gaussian in character. Klauder and Anderson[7] have generalized this model to include other types of fluctuations, such as those giving rise to T_1 and T_2 in solids. Their elegant mathematical treatment also includes the Lorentzian type of diffusion. We have extended their model to show that any fluctuation in resonance frequency can give rise to an extra decay of the spin-echo signals[11] apart from their usual exponential relaxation decay. This decay is, in general, nonexponential in character and is typical of spin echoes owing to the peculiar nature of the experiment. We note that the cw line shape is determined by the stationary distribution of the frequency fluctuations, while the free-precession signal is determined by the stationary distribution as well as the transition probability of the frequency fluctuations. This may appear in contradiction to the usual belief that the free-precession signal is the Fourier transform of the cw signal.[12] It can be shown that this equivalence of the free-precession signal and the cw signal through the Fourier transform is not exactly valid because of the different experimental conditions of the two methods. A more detailed account will be reported in a future publication. We show here how this decay constant can be related directly to the correlation function of the fluctuations in the resonance frequency and how this model can be applied to understanding the origin of the NED in ordered magnetic materials. Since we use many results from the paper of Klauder and Anderson[7] in showing the relation between the decay constant and the correlation function, we give a brief sketch of their calculation of the spin-echo signals. We also include in our discussion a very simple model to illustrate the effect of fluctuations in resonance frequency. All these are included in Sec. III. A more detailed account of the origin of the NED has been given by the author elsewhere[11] and will be reported in a forthcoming publication. The experimental method is described in Sec. II. In Sec. IV we present the results of our measurements on FE^{57} in YIG at 4.2 °K and below for both the a and d sites. This section also includes discussion of our results. Finally, we conclude in Sec. V.

II. EXPERIMENTAL METHOD

As we have noted earlier, we have employed the method used by Sinha and the present author[4,5] to measure the molecular self-diffusion coefficient in liquids. In this method three radio-frequency (rf) pulses are applied in succession at times 0, τ_1, τ_2 as shown in Fig. 1. The amplitude of the "image echo" occurring at time $t=2\ (\tau_2-\tau_1)$ after the first pulse is observed as a function of τ_1, while $\tau_2-\tau_1$ is held constant. As a result of this experimental condition, the exponential part depending on $\tau_2-\tau_1$ remains constant while the nonexponential part (k-dependent term) shows time dependence, as can be seen from the expressions for the image-echo amplitudes given below. We quote below the expressions for the primary- and image-echo amplitudes for both the Gaussian and Lorentzian processes. For a Gaussian process,[4,5,7] we have

$$E(2\tau_1)\propto\exp(-2\tau_1/T_2)\exp(-\tfrac{2}{3}k\tau_1^3)\,, \tag{1a}$$

$$E(2(\tau_2-\tau_1))\propto\exp\{-2[\tau_1+(\tau_2-2\tau_1)]/T_2\}\times\exp\{-\tfrac{2}{3}k[\tau_1^3+(\tau_2-2\tau_1)^3]\}\,. \tag{1b}$$

For a Lorentzian process,[7] we have

$$E(2\tau_1)\propto\exp(-2\tau_1/T_2)\exp(-k\tau_1^2)\,, \tag{1c}$$

$$E(2(\tau_2-\tau_1))\propto\exp\{-2[\tau_1+(\tau_2-2\tau_1)]/T_2\}\times\exp\{-k[\tau_1^2+(\tau_2-2\tau_1)^2]\}\,. \tag{1d}$$

In each of the above expressions, the first part shows the exponential decay for a single relaxation time and the second part the NED. The parts of the echo amplitudes determined by the nutational angles introduced by different rf pulses have been omitted since they are not of interest here. We have included the expressions for the primary echo, occurring at $2\tau_1$, to point out that the image echo

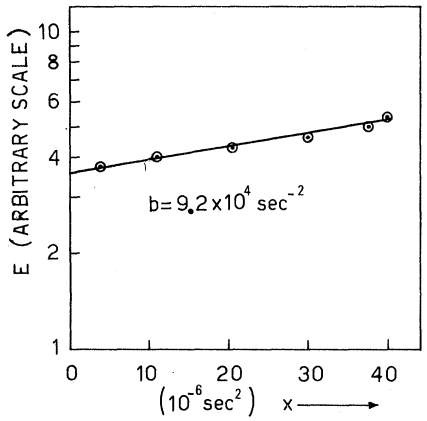

FIG. 2. Typical plot of the amplitude of the image echo E vs x ($=\theta\tau_1-\tau_1^2$, $\theta=\tau_2-\tau_1$) on a semilogarithmic scale. The straight line shows the least-squares fit to the equation $E=E_0e^{bx}$. Here $b=2k$ or $2k\theta$ in the Lorentzian or Gaussian model, respectively (see the text for details).

can be thought of as due to a "virtual" pulse at $t=2\tau_1$ and the third pulse at τ_2. The expressions have been written in a manner so as to bring out this point. Comparing the different expressions given in Eqs. (1), it becomes quite apparent that this way of looking at the image echo is valid even in the presence of nonexponential decays. Hence, it is expected that other types of nonexponential decays (besides the t^2- and t^3-dependent ones which have been considered here) will be similarly reflected in the image echo, and this method will then be very suitable for such measurements. The amplitude of the image echo is maximum for a 90°-180°-180° pulse sequence when two of the echo signals occurring after the third pulse vanish. (For more details see Refs. 4 and 5.)

The measurements were carried out with a Bruker spin-echo spectrometer (model B-KR 306) using a box-car integrator (PAR model cw-1) for the improvement of the signal-to-noise ratio. Temperatures below 4.2 °K were obtained pumping on liquid helium and the pressures were held constant with the help of a diaphragm-type manostat (fabricated at the Siemens Workshop, Munich) to maintain temperatures stable to better than 1% over the whole range studied. A constant check was maintained by a carbon resistance thermometer (Allen Bradley type 100 Ω, $\frac{1}{8}$ W) placed near the sample: The experimental data were fitted with an equation of the form $E=E_0e^{bx}$, where $x=\theta\tau_1-\tau_1^2$ and $\theta=\tau_2-\tau_1$, by the method of least squares with a desk computer (HP model 9100 A). One typical set of data is shown in Fig. 2. The quantity b, thus determined, was found to be independent of θ within experimental error, suggesting that NED was Lorentzian since in this case $b=2k$, while for a Gaussian NED $b=2k\theta$. This can easily be seen on recasting the expressions given by Eqs. (1b) and (1d) in terms of the variables x and θ. Hence, it is a simple matter to distinguish experimentally between the Gaussian and Lorentzian nonexponential decays. The NED constant k for both the a and d sites was determined using the above relation, namely $b=2k$, for temperatures 4.2 °K and below. The errors in k were on the average 15–20% as shown in Fig. 4. Errors in the individual k measurements, similar to the one shown in Fig. 2, were mostly in the range 5–15%. Error bars at the different temperatures show the root-mean-square deviation of a number of measurements of k (at least 5 and in most cases more).

III. THEORETICAL BACKGROUND

A. Simple Model for NED Spin Echoes

We present here a very simple model to illustrate how the frequency fluctuations can give rise to an additional decay of the spin echoes, apart from the usual relaxation decay. Our basic model is the same as the one used by Hahn[1] in his pioneering paper, which has been discussed so much in the literature that we refrain from giving the details here. In Fig. 3 from (A) to (E), we show the formation of the echo signal due to a 90°-180° pulse sequence applied at times 0 and τ. The different isochromatic groups of moment $\Delta M(p)$ (even the moments of these groups may be considered moments of individual spins) recluster exactly on the y axis at time 2τ as shown in (E) of Fig. 3(b), p being the index for an individual group or an individual spin. It may be pointed out here that such a description of reclustering to form the echo signal is more convenient in considering an ensemble of individual spins rather than of isochromatic groups, since with the passage of time an initially isochromatic group becomes a nonisochromatic group, although from the rigorous mathematical standpoint both these descriptions should be identical, at least for stationary processes.

The main point we wish to stress here is that the phases $\varphi_1(p)$ and $\varphi_2(p)$ accumulated by an individual isochromatic group p of the initial distribution in the two time intervals 1 and 2, respectively, will not, in general, cancel each other as required for the reclustering at time 2τ on the y axis. The cancellation is only possible if the resonance frequency of the group or the individual spin under consideration remains constant during the two intervals mentioned above, i.e., if $\varphi_1(p)=\varphi_2(p)$. If for some reason, $\varphi_1(p)\neq\varphi_2(p)$ then the different isochromatic groups or spins will not recluster at time 2τ. In fact, this happens in the case of fluctuations in frequency about some mean value for each frequency, where $\varphi_1(p)$ will not, in general, be equal to $\varphi_2(p)$. If the frequency

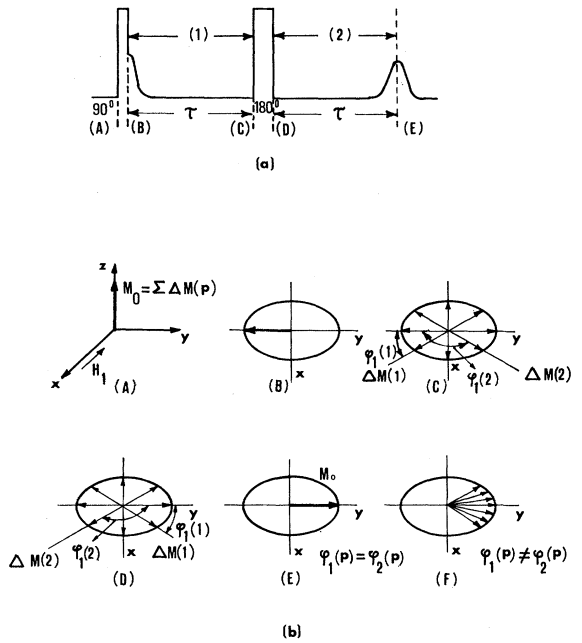

FIG. 3. Vector model showing the effects of fluctuations in resonance frequency on the amplitude of the two-pulse echo. (a) Time sequence of the two-pulse echo for a pair of 90°–180° pulses; pulse durations are assumed to be negligible in comparison to τ. The sequence of diagrams from (A) to (E) in (b), corresponding to points marked similarly in (a), shows the formation of the echo when the phases accumulated by any individual pth isochromatic group $\Delta\vec{M}(p)$, $\varphi_1(p)$ and $\varphi_2(p)$ during the intervals 1 and 2, respectively, are equal. (F) shows that the different $\Delta\vec{M}(p)$ are distributed about the y axis when $\varphi_1(p)\neq\varphi_2(p)$ owing to random fluctuations in resonance frequency.

fluctuations are of a random nature, then the φ's are also of the same nature. The different isochromatic groups in this case, instead of coming on the y axis at time 2τ, will be distributed about it as shown by (F) in Fig. 3(b). There is still a resultant along the y axis, but of reduced magnitude; the reduction will depend upon the nature and magnitude of phase fluctuations. Thus, fluctuations in resonance frequencies of different isochromatic groups introduce an extra decay term to the echo signals. In general, such a decay is nonexponential in character and can be demonstrated rigorously for simplified diffusion models.

B. Diffusion Models of Nonexponential Decay of Spin Echoes

In calculating the spin-echo signals, one needs to evaluate the quantity $\langle e^{i\eta t}\rangle$, where t is the time of occurrence of the signal, η is the precession frequency of an individual spin in the rotating frame ($\eta=\omega-\omega_0$, where ω_0 is the resonance frequency of that spin in the laboratory frame and ω is the center of the laboratory resonance line as well as the exciting frequency), and $\langle\cdots\rangle$ indicates the ensemble average over the spin system. The expression $e^{i\eta t}$ follows from the Bloch equations or from a more general argument noted by Klauder and Anderson. In the presence of fluctuations of the resonance frequency, η is no longer a constant in time but a random variable in time. This fact must be taken into account in the averaging procedure. The most convenient way to take this into account is to divide the evolution time t of the spin system into a large number of smaller time intervals Δt. If one knows the initial frequency distribution $P(\eta_0)$ and the transition or conditional probability $P[\eta;\Delta t\,|\,\eta_0]$ (where, in general, we write η_n for the precession frequency of an individual spin in the nth time interval Δt), then one can take the ensemble average quite simply in the Markoffian approximation. Of course, in this process one has to consider the effects of rf pulses, each of which introduces a phase reversal. The NED is not, in general, sensitive to the initial distribution which is mainly responsible for the shape of echo signals. We give below a short sketch of this calculation and for details we refer again to the paper of Klauder and Anderson.

In order to take account of fluctuations in the resonance frequency we have to evaluate the following expression:

$$F(t)=\langle\exp[i\int_0^t\eta(t')\,dt']\rangle\,, \tag{2}$$

where the time integration is the expression of the accumulated phase of a single spin.

To incorporate the effects of rf pulses, one can rewrite $\int_0^t\eta(t')\,dt'$ as $\int_0^t s(t')\eta(t')\,dt'$, where

$$\begin{aligned} &s(t')=1, && 0<t'<\tau_1\\ &s(t')=-1, && \tau_1<t'<\tau_2\\ &s(t')=1\,, && \tau_2<t'\,. \end{aligned} \tag{3}$$

Here 0, τ_1, and τ_2 are the times at which three rf pulses are applied.

Let $t=(N+1)\Delta t$. Then Eq. (2) can be reduced to

$$F(t)=\int\left\langle\exp\left(i\Delta t\sum_{n=0}^{N}s_n\eta_n\right)\right\rangle P(\eta_0)\,d\eta_0\,. \tag{4}$$

The above expression means that the ensemble average of Eq. (2) is equivalent to the averaging over the frequency "histories" of a particular group of spins (namely, those of frequency η_0 at $t=0$) and then subsequently averaging over η_0. Then, making use of the conditional probability concept, we can write

$$F(t)=\int\cdots\int\exp\left(i\Delta t\sum_{n=0}^{N}s_n\eta_n\right)\times P[\eta_{N+1},\ldots,\eta_1;t\,|\,\eta_0]\,P(\eta_0)\prod_{n=0}^{N}d\eta_n\,. \tag{5}$$

Under the Markoffian assumption, we have

$$P[\eta_{N+1}, \ldots, \eta_1; t|\eta_0] = \prod_{n=0}^{N} P[\eta_{n+1}; \Delta t|\eta_n]. \quad (6)$$

From a general argument valid for a Markoffian process Klauder and Anderson obtained the Fourier transform of the transition probability,

$$P[\eta; t|\eta_0] = (2\pi)^{-1} \int_{-\infty}^{\infty} \exp[iy(\eta - \eta_0) - tf(y)]\, dy\,, \quad (7)$$

where the following conditions must be satisfied:

$$\begin{aligned} f^*(-y) &= f(y) && \text{since } P \text{ is real,} \\ f(0) &= 0 && \text{since } P \text{ is normalized,} \\ \mathrm{Re} f(y) &\geq 0 && \text{since } P \text{ is nonnegative .} \end{aligned} \quad (8)$$

They have also shown that $f(y) = ky^2$ and $f(y) = k|y|$ describe, respectively, Gaussian and Lorentzian processes. In general, it is almost impossible at the present state of our knowledge to theoretically predict the physical processes that would give rise to the Gaussian or Lorentzian processes except in very special cases. The frequency distribution of a spin interacting with a small number of other spins by dipolar interaction[13] would be Lorentzian, while if the number of interacting spins is large, then this frequency distribution is expected to be Gaussian.[14] Experimentally it is a comparatively simple matter to distinguish between the two processes, as we have noted in Sec. II.

Using Eqs. (5)–(7) and the condition for the occurrence of echo signals, i.e., $\int_0^t s(t')\, dt' = 0$, it is fairly straightforward to calculate amplitudes of different echo signals. The results of such calculations have already been given in Eqs. (1) for the primary and image echo for both the Gaussian and Lorentzian processes.

C. Relation between NED Constant k and Correlation Function of Frequency Fluctuations

We know from probability theory[15] that, for any two random variables, the conditional probability is given by

$$P[\eta; t|\eta_0] = [2\pi\psi_0(1-\rho^2)]^{-1/2} \times \exp[-(\eta - \rho\eta_0)^2/2\psi_0(1-\rho^2)]\,, \quad (9)$$

where $\psi_0 = \langle\eta^2\rangle = \langle\eta_0^2\rangle$ for stationary processes. ψ_0 is the mean-square fluctuation of the random variable. $\psi(t) = \rho\psi_0 = \langle\eta(t)\eta(0)\rangle$ is the correlation function of fluctuations in η and defines the time-dependent term ρ known as the correlation coefficient. The subscript zero indicates values at $t = 0$. In terms of the Fourier transform, Eq. (9) can be written as

$$P[\eta; t|\eta_0] = (2\pi)^{-1} \int_{-\infty}^{\infty} \exp[iy(\eta - \rho\eta_0) - \tfrac{1}{2}\psi_0(1-\rho^2)]\, dy\,. \quad (10)$$

Comparing this expression with that of the transition probability for a Gaussian process as given by Eq. (7), it is evident that if one can express $\frac{1}{2}\psi_0(1-\rho^2)$ as equal to kt for a short time t, then the NED constant k of Eqs. (1) is obtained. Under this condition of short time t, Eq. (10) reduces to Eq. (7), since $\rho \simeq 1$. Differentiating with respect to time, we have

$$k = -\left.\frac{d\psi}{dt}\right|_{t=0}. \quad (11)$$

We have not yet been able to show rigorously that such a relation also holds good for a Lorentzian process. But comparing the expressions of transition probabilities for the two processes as given by Eq. (7), we assume a similar relation is also true for a Lorentzian process when $\frac{1}{2}\psi_0(1-\rho^2)$ can be expressed similarly as a linear function of time for a short time t. Obviously, a constant factor must be incorporated to satisfy the dimensionality. Klauder and Anderson[7] came to a similar conclusion implicitly in considering a simple model of dipolar field fluctuations.

Under our assumption, the transition probability for a Lorentzian process can be written in analogy with the corresponding expression for a Gaussian process given by Eq. (10) as

$$P[\eta; t|\eta_0] = (2\pi)^{-1} \int_{-\infty}^{\infty} \exp[iy(\eta - \rho\eta_0) - \tfrac{1}{2}C\psi_0(1-\rho^2)|y|]\, dy\,, \quad (12)$$

where C is an unknown constant. The same argument which reduces Eq. (10) to Eq. (7) also reduces Eq. (12) to Eq. (7). Then a relation similar to Eq. (11) is obtained for k with an extra constant C.

Expressions for the transition probabilities given by Eqs. (10) and (12) are superior to the corresponding expressions given by Klauder and Anderson, since they give the expected (stationary) distributions for stationary processes for $t \to \infty$. In this limit, $\rho = 0$ and Eqs. (10) and (12) reduce, respectively, to stationary distributions of the Gaussian and Lorentzian types as given below:

$$P(\eta) = (2\pi\psi_0)^{-1/2} \exp(-\eta^2/2\psi_0) \quad \text{(Gaussian)} \quad (13a)$$

$$= \frac{1}{\pi} \frac{\frac{1}{2}C\psi_0}{(\frac{1}{2}C\psi_0)^2 + \eta^2} \quad \text{(Lorentzian)}\,. \quad (13b)$$

D. Application to Ordered Magnetic Materials

It is well known that in an ordered magnetic material, with no external magnetic field and neglecting the anisotropy field, the nuclear resonance frequency is given by[16]

$$\omega_0 = A\langle S_z\rangle/\hbar\,, \quad (14)$$

where A is the hyperfine coupling constant and

$\langle S_z \rangle$ is the thermal average of the z component of the atomic spin $\vec{S}$. We consider here the simplest case where the atomic spin $\vec{S}$ and the nuclear spin $\vec{I}$ are coupled by the interaction of the form $A\vec{I}\cdot\vec{S}$. Extension to more general cases is straightforward. Considering only the spin fluctuations, one can define the instantaneous nuclear resonance frequency $\eta(t)$ as

$$\eta(t) = (A/\hbar)[S_z(t) - \langle S_z \rangle] . \quad (15)$$

Here, we have neglected any fluctuation in A, partly because of the simplicity of the theoretical treatment, and partly because of the reasonable agreement of this model with our experiment. Further, a reasonably good agreement of the temperature dependence of the experimental hyperfine field in ordered magnetic materials with the spin-wave model suggests A to be almost independent of temperature.[17]

According to our previous discussion, the NED constant is related to the time-dependent correlation function $\psi(t)$. In the present case it is given by

$$\psi(t) = (A/\hbar)^2 \langle [S_z(t) - \langle S_z \rangle][S_z(0) - \langle S_z \rangle] \rangle . \quad (16)$$

The expression inside the angular brackets is the time-dependent longitudinal spin correlation and has been calculated by Vaks *et al.*[18] for a Heisenberg ferromagnet. From their Eq. (37), one can easily derive the time-dependent part of $\psi(t)$ for a Heisenberg ferromagnet at low temperatures ($T \ll T_c$, where T_c is the Curie temperature) giving the NED constant k as

$$k = \tfrac{3}{20} C(A/\hbar)^2 D\kappa_{\max}^2 (k_B/\pi D)^{3/2} k_B \zeta(\tfrac{3}{2}) T^{5/2} . \quad (17)$$

In deriving the above expression use has been made of Eq. (11) in relating $\psi(t)$ to k, which is valid for both Gaussian and Lorentzian processes and which gives identical expressions for the two processes except for an unknown constant in the Lorentzian process. $\psi(t)$ is obtained under the following assumptions: (a) The wave vector κ is very small, (b) there is no restriction on ω,[19] and (c) $\vec{r}_1 = \vec{r}_2$, i.e., there is space autocorrelation. For low temperatures and for short times t these assumptions are expected to be valid. We have used the magnon dispersion relation $\epsilon_\kappa = D\kappa^2$; $\kappa_{\max}$ is the maximum wave vector; C is a constant and is exactly equal to unity for a Gaussian process, but is still an unknown constant for a Lorentzian process, though it is expected not to be very different from unity; k_B is the Boltzmann constant; and $\zeta(\frac{3}{2})$ is the ζ function of index $\frac{3}{2}$.

On a straightforward extension of this model to two- or more-sublattice systems, one expects that an individual sublattice would behave like a single-sublattice Heisenberg ferromagnet. This expectation, of course, comes mainly from the different

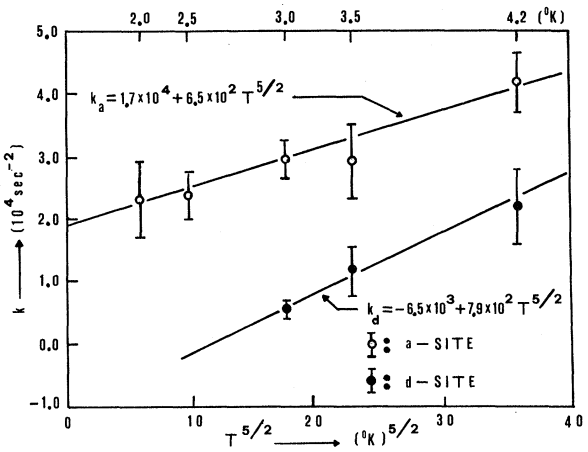

FIG. 4. Plot of the nonexponential decay constant k vs $T^{5/2}$ for Fe^{57} nuclear spin echoes in YIG. The subscripts a and d stand for the corresponding lattice sites. The straight lines show the least-squares fit to Eq. (18). The top scale shows the temperature itself.

experimental and theoretical studies on sublattice magnetization.[20] In such studies it has also been observed that each individual sublattice would show up some deviation from its actual spin value S at absolute zero owing to zero-point motion.[20,21] An additional contribution to k for individual sublattices due to zero-point spin fluctuations is then expected. With this consideration, for an individual sublattice in a multisublattice system, where each sublattice can be approximated by a Heisenberg ferromagnet, k is expected to follow a relation like

$$k = \alpha + \beta T^{5/2} . \quad (18)$$

Here, the first term is due to any temperature-independent process, such as zero-point spin fluctuations, and the second term is due to the temperature-dependent process of the long-wavelength spin fluctuations. From Eq. (17), β is given by

$$\beta = \tfrac{3}{20} C(A/\hbar)^2 D\kappa_{\max}^2 (k_B/\pi D)^{3/2} k_B \zeta(\tfrac{3}{2}) . \quad (19)$$

IV. RESULTS AND DISCUSSION

Experimental variation of the parameter θ in the image-echo experiment implicitly showed the Lorentzian process to be the active process in determining the NED in the present case. We then fitted our data for k with an equation of the form given by Eq. (18) using the method of least squares. This is shown in Fig. 4. From such a fit, we obtain $\alpha = 1.7\times10^4$, $\beta = 6.5\times10^2$ for the a site, and $\alpha = -6.5\times10^3$, $\beta = 7.9\times10^2$ for the d site, where the units of α and β are sec^{-2} and $\text{sec}^{-2}\,^\circ\text{K}^{-5/2}$, respectively.

For Fe^{57} in YIG, $AS/h \simeq 70$ MHz, $D \simeq 10^{-28}$ erg cm^2, and $\kappa_{\max} \simeq 10^7\ \text{cm}^{-1}$. Using these values

in the expression for β given by Eq. (19), we get $\beta \simeq 10^3$, which is in excellent agreement with the experimental results in view of the approximations we made. In the absence of any calculation of the time-dependent longitudinal spin correlation for zero-point spin fluctuations, it has not been possible to estimate the magnitude of α for such a process. But it seems reasonable to expect the ratio of the α's for the two sites to be equal to the square of the ratio of spin deviations at the zero-point for such a process. Our result for this ratio is of magnitude 2.6, to be compared with 2.0 when we consider the exact ratio of hyperfine fields at the two sites. This agrees well with the theoretically predicted value of 2.25 of Walker.[21] Further, the opposite signs of the α's for the two sites can be qualitatively understood when we consider that the Walker theory predicts the sum of the spin deviations for YIG at the zero point to be zero. The spin deviations are of opposite signs. Hence, we might expect the correlation functions of zero-point spin fluctuations for the two sites also to be of opposite signs. Reasonable agreement between experiment and theory for both α and β suggests that Eq. (18) gives a fair description of the NED in YIG for the temperature range of our study. However, as has been discussed below, a smaller and much weaker temperature-dependent contribution in k cannot be excluded from our measurements. Further observations extending the measurements to a much lower and wider temperature range than those reported here are needed to clarify this point of a weaker temperature-dependent contribution in k and will be made in a future publication. A physically disturbing feature of our results is that k_d becomes negative below about 2 °K. Owing to the limitations of our spectrometer, it has not been possible for us to extend the measurements below 3 °K for the d site. Our results suggest that some other contribution in k of weaker temperature dependence than the one due to the process we have considered here becomes more effective in this temperature range. The most probable source of such a weaker temperature-dependent term is the fluctuations in A; we have neglected them in our theory, and this may not be completely justified. The weaker temperature dependence of A in YIG cannot be ruled out from temperature dependence of the hyperfine field, as has been pointed out by Litster and Benedeck.[22] In the absence of an exact theory of fluctuations in A or its temperature dependence in ordered magnetic materials, it is not possible to estimate the contribution of fluctuations in A in k. However, from the above considerations, the apparent negative values of k_d for about 2 °K and below do not seem unreasonable.

We would like to point out here that none of the other sources of fluctuations in the resonance frequency noted earlier offer a satisfactory explanation of our results. From thermal fluctuations of Bloch walls, we expect $k_d \simeq k_a$ at all temperatures, the magnitudes depending on the properties of the walls. Obviously, this is not the case. The spin diffusion as a possible source can be ruled out also on the argument of the small moment and low abundance of Fe^{57}. Further, we expect k to be proportional to $1/T_2^2$ for the Lorentzian process. In order to check this point we also measured T_2 at different temperatures for both sites and found no such relation to hold true, confirming the conclusions reached earlier by Weger.[10] Preliminary data of our T_1 and T_2 measurements suggest that these results also can be understood on the model of spin fluctuations. This will be reported in a separate publication.[23] Dipolar field fluctuations as a possible source of k can be ruled out easily in this case, since we expect $k_a \gg k_d$ at all temperatures owing to the presence of a large dipolar field at the a site but none at the d site.[24]

V. CONCLUSIONS

We have shown here that the NED constant k can be related to the time-dependent correlation function of fluctuations in the resonance frequency $\psi(t)$ for both Gaussian and Lorentzian processes. For a Gaussian process, k can be exactly related to $\psi(t)$. An additional assumption and an unknown constant are involved in k for a Lorentzian process. We have applied this model of fluctuations in resonance frequency as the origin of k to ordered magnetic materials and have shown that the spin fluctuation is the most important contribution to k at low temperatures. We have also found that for a multisublattice system the NED constant of an individual sublattice follows a relation $k = \alpha + \beta T^{5/2}$. Such a relation is borne out by our experimental observations on Fe^{57} in YIG. The spin fluctuations can be approximated to a Lorentzian process in this case. The coefficient of the temperature-dependent term in k could be related to long-wavelength spin fluctuations. Our experimental results strongly suggest that the origin of the temperature-independent term is the zero-point spin fluctuations. The extrapolation of our data of the d-site to temperatures lower than about 2 °K also suggests a weaker temperature-dependent contribution, more effective on k_d due to its smaller value, from fluctuations in A which we have neglected in our theory. As we have noted earlier, a weak temperature dependence of A, and hence a weak temperature dependence of the time-dependent correlation of fluctuations in A, cannot be excluded from the studies reported so far.

Further, we would like to point out that there are other possible sources of fluctuation in the hyper-

fine field which we have not considered here. The important ones may be magnon-magnon, magnon-phonon, and electron-phonon interactions. None of these is expected to be very effective for YIG in the temperature range of our experiment. A more comprehensive discussion of these different processes will be included in a future publication.

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Theory of Acoustic Paramagnetic Resonance in Dense Magnetic Insulators*

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Several mechanisms which describe the interaction between spins and phonons in a dense paramagnetic insulator are theoretically explored in order to estimate their resonant contribution to the ultrasonic attenuation at high temperatures. In particular, one mechanism provides a quasiresonant contribution to the attenuation which is roughly proportional to the square root of the difference of the frequency and certain multiples of the Larmor frequency. Other mechanisms may lead to the same type of line shape.

I. INTRODUCTION

In this paper we shall theoretically examine the question of acoustic paramagnetic resonance (APR) in dense magnetic insulators. The basic model used for the spin system is a Heisenberg paramagnet with an isotropic exchange energy much greater than the Zeeman energy due to an applied magnetic field or any anisotropic spin-dependent forces. For the most part, only temperatures much greater than the magnetic transition temperature of the spins are considered. For our purposes the "resonant" contribution to the acoustic attenuation will be taken to mean that contribution which depends on an externally applied magnetic field.

Electron paramagnetic resonance (EPR) experi-