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Motion of nuclear magnetization under conditions of microscopic inhomogeneity of the hyperfine field

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We investigate the motion of nuclear magnetization under conditions of microscopic inhomogeneity of the hyperfine field (HFF) at low deviations from the equilibrium position. It is shown that if the HFF has a Lorentz distribution function the free-precession damping coefficient depends little on the ratio of the dynamic shift of the NMR frequency to the width of the distribution function. For a quadratic Lorentz function, which falls off more rapidly on the wings, the damping coefficient of the free precession decreases sharply with increasing dynamic frequency shift. Both the frequencies and the damping coefficients of the free precession are independent of the parameters of the pulse-exciting high-frequency field.

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Theoretical and experimental investigations¹⁻⁶ carried out during the last ten years have shown that most typical for magnets is a microscopic inhomogeneity of the hyperfine field (HFF): the correlation radius r_0 of the HFF is usually much smaller than the effective exchange-interaction radius r . In this situation, the electronic magnetization M connected with the strong exchange interacts with the resultant field of the nuclear isochromats^{5,7} and the density of the macroscopic energy of the hyperfine interaction (HFI) is expressed in the form

$$\mathcal{H} = M \int A \mu^A dA, \quad (1)$$

where A is the HFI constant, μ^A is the magnetization of the nuclear isochromat, and by isochromat is meant the set of nuclei for which the HFI constant has the same value. Naturally, under conditions of microscopic inhomogeneity of the HFF the motion of the nuclear magnetization μ^A is described by integro-differential equations, since the dynamic HFI $M_1 \int A \mu_1^A dA$, and consequently also the NMR dynamic frequency shift (DFS) due to this interaction, are "turned on" only at those time intervals when the total transverse nuclear magnetization differs from zero. A mathematical analysis of this situation is in the general case a very complicated problem. Up to recently, the theoretical calculation was carried out either for a model of a macroscopic inhomogeneity of the HFF,⁸⁻¹¹ wherein in the sample is broken up into a quasi-non-interacting sections within which the HFF is homogeneous, or for the case when the inhomogeneity of the HFF is microscopic but the DFS is small.^{6,7} In the present paper we

consider the simplest phenomena that can be analyzed without restrictions on the value of the HFF—we investigate the motion of nuclear magnetization at small deviations from the equilibrium positions.

We consider for the sake of argument a ferromagnetic sample in the form of a sphere magnetized parallel to the external constant magnetic field H . We assume that the Z axis is directed along H and that it is possible to apply to the sample a high-frequency (HF) field $h \sim \exp(i\omega t)$ polarized in the XY plane. In place of $\mu_{x,y,z}^A$ it is convenient to introduce the relative components

$$u = \mu_x^A / \mu^A, \quad v = -\mu_y^A / \mu^A, \quad m = -\mu_z^A / \mu^A, \quad (2)$$

where μ^A is the static magnetization of the isochromat and is determined at not too low temperatures by the Langevin formula

$$\mu^A = Ng(A) \frac{\gamma_n^2 \hbar^2 I(I+1)}{3kT} (AM - H). \quad (3)$$

Here N is the concentration of the magnetic nuclei, $g(A)$ is the distribution function of A , γ_n is the nuclear gyromagnetic ratio, and I is the spin of the nucleus. In a coordinate system rotating with frequency ω , the motion of the nuclear magnetization is described by the equations^{7,8}

$$\begin{aligned} \dot{u} - (\Delta + \delta)v + \Gamma_n u + L_y &= 0, \\ \dot{v} + (\Delta + \delta)u + \Gamma_n v + L_x &= -\omega_n m, \\ \dot{m} + (m-1)/T_1 + L_z &= \omega_n v. \end{aligned} \quad (4)$$

Here Δ is the difference between the frequency of the HF field and the average undisplaced NMR frequency

$\langle \omega_n \rangle$:

$$\Delta = \langle \omega_n \rangle - \omega, \quad \omega_n = \gamma_n (AM - H), \quad (5)$$

δ is the difference between the undisplaced frequency of the individual isochromat and the average frequency $\langle \omega_n \rangle$:

$$\delta = \gamma_n (A - \langle A \rangle) M, \quad (6)$$

$\Gamma_n = 1/T_2$ is the reciprocal transverse nuclear relaxation time, $\omega_N = \gamma_n \eta h$ is the nutation frequency, $\eta = A_{\chi_e}^0$ is the gain of the HF field, χ_e^0 is the static electronic susceptibility, and the nonlinear terms $L_{x,y,z}$ describe the DFS of the NMR:

$$L_x = -DmK_u, \quad L_y = DmK_v, \quad L_z = D(vK_u - uK_v), \quad (7)$$

where

$$D = \gamma_n \langle A \rangle \eta \mu, \quad K_u = \int_{-\infty}^{\infty} u g(A) dA, \quad K_v = \int_{-\infty}^{\infty} v g(A) dA, \quad (8)$$

$\mu = \int \mu^A dA$ is the total static mechanization of the nuclear system and, in addition, it is assumed that $\langle \Delta A \rangle \ll \langle A \rangle$.

The equations of motion (4), strictly speaking were written out for the case when complete magnetic symmetry exists in the XY plane, i.e., the electronic magnetization M precesses along a circle. Nonetheless, in analogy with the procedure in Ref. 8, it is easy to show that if we neglect the inessential rapidly oscillating terms, then Eqs. (4) are valid also in the case when the M -precession trajectory is elliptic. These equations remain valid also for antiferromagnets of the "easy plane" type for each magnetic sublattice. If the deviations of the nuclear magnetization from the equilibrium position are small, then the equations (4) can be linearized, i.e., we can put $m = 1$. We shall henceforth use δ in place of the variable A . We introduce the complex converse amplitude

$$s(\delta, t) = u + iv \quad (9)$$

and write down the linearized equation in the form

$$\dot{s} + i(\Delta + \delta)s + \Gamma_n s - iDR = -i\omega_n, \quad (10)$$

$$R = R(t) = \int_{-\infty}^t s g(\delta) d\delta.$$

The real and imaginary parts of $R(t)$ determine the transverse components of the summary nuclear magnetization:

$$\mu_x = \mu \operatorname{Re}(R), \quad \mu_y = \mu \operatorname{Im}(R). \quad (11)$$

For the sake of argument we assume a Lorentz distribution function g :

$$g(\delta) = \frac{\Gamma}{\pi} \frac{1}{\delta^2 + \Gamma^2}. \quad (12)$$

To facilitate the discussion, we shall initially disregard the dependence of the gain η on δ , i.e., we put $\eta = \langle \eta \rangle$ and accordingly

$$D = D_0 = \gamma_n \langle A \rangle^2 \chi_e^0 \mu. \quad (13)$$

We consider first the free precession of the nuclear isochromats in the simplest case, when all the iso-

chromats have at the initial instant of time $t = 0$ the same relative amplitude $s(\delta) \equiv s_0$. To obtain the solution of Eq. (10), we employ the following procedure. We carry out a Laplace transformation and express the transform of the function $s(\delta, t)$ in terms of the transform of the function $R(t)$:

$$\hat{s}(\delta) = \frac{iD_0 \hat{R} + s_0}{p + \Gamma_n + i\delta + i\langle \omega_n \rangle}. \quad (14)$$

Here \hat{s} and \hat{R} are respectively the transform of the functions s and R . We now multiply both sides of the equation by $g(\delta)$ and integrate with respect to δ . As a result we obtain an algebraic equation for \hat{R} , from which we get

$$\hat{R} = \frac{s_0}{p + i\Omega + \Gamma_n^*}, \quad (15)$$

$$\Omega = \langle \omega_n \rangle - D_0, \quad \Gamma_n^* = \Gamma + \Gamma_n.$$

Taking the inverse Laplace transform, we obtain the solution of Eq. (10):

$$s(\delta, t) = s_0 \exp(-\Gamma_n t) \frac{(\Gamma - i\delta) \exp(-i\omega_n t) - iD_0 \exp(-i\Omega t - \Gamma t)}{\Gamma - i(D + \delta)} \quad (16)$$

and also an expression for R :

$$R(t) = s_0 \exp(-i\Omega t - \Gamma_n^* t). \quad (17)$$

From the expression for $s(\delta, t)$ it is seen that the character of the motion of each isochromat is determined by a sum of two terms. The first term describes the independent precession of each isochromat with undisplaced frequency ω_n , while the second term is connected with the DFS and describes the coherent motion of the nuclear isochromats with common frequency Ω . As to the summary nuclear magnetization μ_x , which is registered in the experiments, it is seen from (17) that μ_x precesses at a frequency Ω and a damping coefficient Γ_n^* , and this result does not depend on the relation between Γ and D_0 .

We consider now the motion of the summary nuclear magnetization μ under the influence of a rectangular HF-field pulse of arbitrary duration τ . Using the same procedure as before, we obtain

$$\hat{R} = -i\omega_n (1 - e^{-p\tau}) / p(p + \Gamma_n^* + i\bar{\Delta}), \quad (18)$$

where $\bar{\Delta}$ is the detuning between the frequency of the HF field and the shifted NMR frequency $\bar{\Delta} = (\Omega - \omega)$. Taking the inverse Laplace transform, we get

$$R(t) = \frac{\omega_n}{\bar{\Delta} - i\Gamma_n^*} [\exp(-i\bar{\Delta}t - \Gamma_n^* t) - 1], \quad t < \tau;$$

$$R(t) = \frac{\omega_n [1 - \exp(i\bar{\Delta}\tau + \Gamma_n^* \tau)]}{\bar{\Delta} - i\Gamma_n^*} \exp(-i\bar{\Delta}t - \Gamma_n^* t), \quad t > \tau. \quad (19)$$

From the expression (19) at $t \gg 1/\Gamma_n^*$ we obtain the stationary solution

$$R(t) = -\omega_n / (\bar{\Delta} - i\Gamma_n^*). \quad (20)$$

This solution describes a stationary NMR line with center on the displaced frequency Ω and with half-width Γ_n^* . The second term of (19) describes the decrease of the free precession (DFP) after turning on the HF field. It is seen from this expression that regardless of the duration τ and the carrier frequency of the exciting pulse, the DFP frequency is always determined by the central frequency Ω . This result is

valid for any value of D_0 , including $D_0=0$, and is naturally common to all systems of interacting or non-interacting oscillators. In the literature, however, this effect is not considered specially, and we shall therefore dwell on it in greater detail. At first glance, the result is paradoxical. In fact, one should expect, if the frequency ω corresponds, for example, to the left edge of the absorption line ($\omega < \Omega$), and the durations of the pulses are long enough ($\tau \gg 1/\Gamma_n^*$), then by the instant the pulse is turned-on the oscillators on the left edge of the line will be excited more strongly than on the right edge, and consequently the DFP frequency should be displaced to the left relative to the central frequency Ω . The cause of this effect lies in the phase relations in the oscillator system.

Figure 1 shows the dependence of the phase φ of the oscillators (relative to the phase of the HF field) on the oscillator frequency ω_0 at $t = \tau \gg 1/\Gamma_n^*$. It is clear from this figure that although the resonant oscillators on the left edge of the line are more strongly excited at the instant when the pulse is turned off than on the right edge, they are much more strongly dephased relative to each other than the nonresonant oscillators on the right edge of the line. As a result, the resonant and nonresonant oscillators make equal contributions to the total amplitude, and the DFP frequency coincides with the central frequency Ω . For the same reason, the damping of the DFP signal is determined by the parameter Γ_n^* .

So far, to facilitate the exposition, we have written down mathematical expressions obtained without allowance for the dependence of the gain η on A , and consequently we have neglected the resultant scatter of the DFS. Solving Eqs. (10) with allowance for the total expression for D .

$$D = D_0(1 + \delta/\langle \omega_n \rangle), \quad (21)$$

we find that all the formulas obtained above for $R(t)$ remain in force if we make the substitution

$$\Gamma \rightarrow \Gamma(1 - D_0/\langle \omega_n \rangle). \quad (22)$$

It is seen from this that the scatter of the values of D , due to the scatter of A , leads to a narrowing of NMR line. The reason is that larger values of ω_n (i.e., larger values of A) correspond also to larger values of D . Therefore the right edge of the NMR line is displaced more strongly than the left edge, and as a result the NMR line becomes narrower (Fig. 2). The $\eta(A)$ dependence causes also a scatter of the nutation frequency ω_N . It is easy to verify, however, that the scatter of ω_N leads only to an inessential renormalization of the amplitude of HF field by an amount $\sim \hbar\Gamma/\langle \omega_n \rangle$.

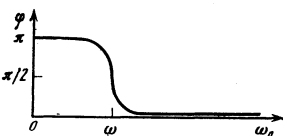


FIG. 1. Dependence of the phase φ of the oscillator (relative to the phase of the HF field) on the oscillator frequency ω_0 .

It follows from the foregoing analysis that the damping coefficient of the DFP depends very little on the ratio of D_0 and Γ . On the other hand, analysis^{11,12} shows that under conditions of strong DFS ($D_0 \gg \Gamma$) the interaction of the transverse components of the nuclear spins should lead to suppression of the microscopic inhomogeneity of the HFF, i.e., the free precession damping coefficient should decrease sharply. In the course of a discussion of the present paper, M. I. Kurkin advanced the opinion that the obtained disparity is connected with the slow decrease of the Lorentz function on the wings, and that for more rapidly decreasing distribution functions the microinhomogeneity of the HFF should be suppressed.

We shall show now that such an effect is indeed realized even for a quadratic Lorentz function. We put

$$g(\delta) = \frac{2}{\pi} \frac{\Gamma^2}{(\delta^2 + \Gamma^2)^2}, \quad (23)$$

$s(\delta) \equiv s_0$ at $t=0$, and neglect the inessential $\eta(\delta)$ dependence. Using the same method as before, we obtain

$$\hat{R}(p) = s_0 \frac{p + \Gamma_n + 2\Gamma}{(p + \Gamma_n + \Gamma + i\langle \omega_n \rangle)^2 - iD_0(p + \Gamma_n + 2\Gamma) + D_0\langle \omega_n \rangle} \quad (24)$$

and correspondingly

$$R(t) = s_0 \exp(-i\langle \omega_n \rangle - \Gamma_n)t \frac{(p_1 + 2\Gamma) \exp(p_1 t) - (p_2 + 2\Gamma) \exp(p_2 t)}{p_1 - p_2}. \quad (25)$$

The points

$$p = p_{1,2} + i\langle \omega_n \rangle - \Gamma_n \quad (26)$$

represent here the poles of the function $\hat{R}(p)$, while p_1 and p_2 are given by the expression

$$p_{1,2} = \Gamma [(-1 \pm (2\xi a)^{1/2}) + i(2\xi \pm (2\xi/a)^{1/2})], \quad (27)$$

$$a = (\xi^2 + 1)^{1/2} - \xi, \quad \xi = D_0/4\Gamma.$$

At $\xi=0$ expression (25) describes the free precession with frequency $\langle \omega_n \rangle$ and with a damping coefficient Γ_n^* :

$$R(t) = s_0(1 + \Gamma t) \exp(-\Gamma_n^* t). \quad (28)$$

At $\xi \neq 0$ the motion of the nuclear magnetization is described by the sum of two terms. The first term in (25) corresponds to precession with a frequency close to the displaced NMR frequency Ω , while the second term to precession with frequency close to $\langle \omega_n \rangle$. With increasing ξ , the relative amplitude of the second term in (25) decreases rapidly, therefore the character of the motion is determined by the first term, for which the damping coefficient decreases monotonically from Γ (at $\xi=0$) to zero (at $\xi \rightarrow \infty$). For $D_0 \gg \Gamma$ we get, accurate to terms $\sim \Gamma^3$,

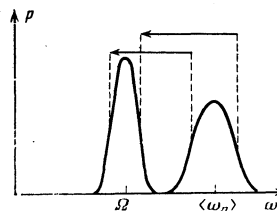


FIG. 2. Narrowing of the NMR line on account of the scatter of D . Here P is the absorption of the HF field energy.

$$R(t) \approx s_0 \exp(-i\Omega - 2\Gamma^2/D_0^2)t. \quad (29)$$

We see therefore that at $\Gamma \ll D_0$ the damping coefficient is determined by the expression $2\Gamma^2/D_0^2 \ll \Gamma$. Thus, the effect of suppression of the microscopic inhomogeneity of the HFF^{11,12} is realized even for a quadratic Lorentz function.

If the system is acted upon by a rectangular HF-field pulse, then the transform of the function $R(t)$ is given by

$$\hat{R} = \frac{-i\omega_s[1 - \exp(-p\tau)](p + i\Delta + \Gamma_n + 2\Gamma)}{p[(p + i\Delta + \Gamma_n + \Gamma)^2 - iD_0(p + i\Delta + \Gamma_n + 2\Gamma)]}, \quad (30)$$

where τ is the pulse duration. The pole at the point $p=0$ corresponds to a stationary solution as $\tau \rightarrow \infty$. The two other poles describe the frequency and the damping coefficients in the transient processes. These poles are obtained from an algebraic equation for (24) only in the substitution $\langle \omega_n \rangle \rightarrow \Delta$. It is therefore clear that at $t > \tau$ the frequencies and the damping coefficients of the DFP are determined by expressions (26) and (27) and do not depend on the duration and carrier frequency of the exciting pulse.

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Absorption of sound in dilute magnetic alloys

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The electronic absorption of sound in metals with magnetic impurities is considered. An expression is obtained for the absorption coefficient of transverse sound α_t , with account of effects connected with the motion of impurities in the field of the sound wave. This expression is connected with the electron self-energy part $\Sigma(\omega)$. Two types of dilute magnetic alloys are analyzed. In alloys with disordered magnetic impurities, the characteristic temperature dependence of the absorption coefficient is obtained. In spin glasses there is an anomalous frequency dependence of α_t . At frequencies at which $\alpha_t \propto \omega^{-3}$ in metals without magnetic impurities the relation of spin glasses is $\alpha_t \propto \omega^{-1}$. The latter result is obtained without recourse to any specific model of the spin glass and is the result of the impurity spin's being "frozen in". The proposed method can also be employed in calculating the absorption of longitudinal sound.

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1. INTRODUCTION

A number of interesting effects, which are due to the interaction of the conduction electrons with the magnetic impurities, have been observed in dilute magnetic alloys (see, for example, Refs. 1 and 2). The specific character of this interaction leads to a characteristic dependence of the lifetime of the electronic excitations on the energy and temperature, and also to rearrangement of the electron spectrum in the case in which any of the types of magnetic ordering of the impurities (ferromagnetic, antiferromagnetic, spin glass, see,

for example, Refs. 3 and 4) is established. Inasmuch as the basic contribution to the sound absorption at low temperatures is made by the interaction of the conduction electrons with the incident sound wave, it is natural to expect that the characteristic features of the dilute magnetic alloys are essentially manifested in the sound absorption.

The problem of the sound absorption has been posed and solved in many researches. There exist several approaches to its solution. One of them—the phenomenological—is based on the use of the kinetic equation of Boltzmann with a single relaxation time τ .^{5,6} This ap-