close to the corresponding value for the free ion. The hyperfine field for the Tb^{3+} ion is equal to 3.14 MG.[3] In a metal and an alloy, the presence of additional contributions from neighboring magnetic ions and from polarized conduction electrons, as a rule, decreases somewhat the value of the field in comparison with the field for the free ion. The data of[1] and the present results do not contradict this fact. The reason for the more substantial difference between the results of[2] remains unclear. It must be noted that the experimental conditions in[2] and in the present paper were practically identical.

As already noted, the quadrupole interaction constant determined in the experiment contains contributions from the intrinsic 4*f* shell and from the crystal field and the conduction electrons. From experiment with atomic beams (for the ^{159}Tb isotopes) we know the quadrupole-interaction constant for the Tb^{3+} ion, namely, $P_{\parallel} = 2.55 \times 10^{-18}$ erg.[3] This yields for the ^{160}Tb isotope a constant equal to 1.14×10^{-18} erg. Comparing this value with the results of the present study (1.38×10^{-18} erg) we can conclude that the quadrupole interaction for terbium in gadolinium is determined mainly by the intrinsic 4*f* shell.

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Torsional vibrations and stimulated echo with a long memory in magnetic powders

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Two- and three-pulse echoes are studied in a number of magnetic powders at frequencies between 10 and 40 MHz in a stationary magnetic field. A long-lived (of at least several hours duration) stimulated echo is observed in powders consisting of particles with dimensions of the order of 1 μ. The results are interpreted on the basis of a model of torsional vibrations of the powder particles induced by an alternating magnetic field. The absence of damping of the stimulated echo is attributed to memory of the microcrystal orientation parameters.

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The phenomena of powder echo, first observed in magnetic materials,[1] and then in piezoelectrics[2,3] and metals,[4] are widely studied at the present time.[5–11] The experimental method is very similar to that which is used in the investigations of ordinary spin echo; upon irradiation of the powder at the instants of time $t=0$, τ, and T by pulses of an alternating electromagnetic field, signals of two-pulse (A_2) and three-pulse (A_3) echoes are observed at the instants of time $t=2\tau$ and $T+\tau$; the latter is frequently called the "stimulated" echo. The experiments are usually carried out with powders whose particles have a mean size of the order of the sound wavelength at the frequency of the alternating field, i.e., under conditions of acoustic resonance of the microcrystals. Just as in the case of magnetic resonance, the falloff of the echo signals upon change in the intervals between the sounding pulses is determined by the relations

$$A_2 \sim \exp(-2\tau/T_2), \quad A_3 \sim \exp(-2\tau/T_2 - T/T_1),$$

in which the characteristic times T_2 and T_1 appear. For most studied piezoelectrics and magnetic powders, the quantity T_2, depending on the dimensions of the crystals, the density of their packing, the viscosity of the medium in which the powders are located, and other factors, lies in the range 10^{-6}–10^{-3} sec. The time of "longitudinal relaxation" T_1 as a function of the charac-

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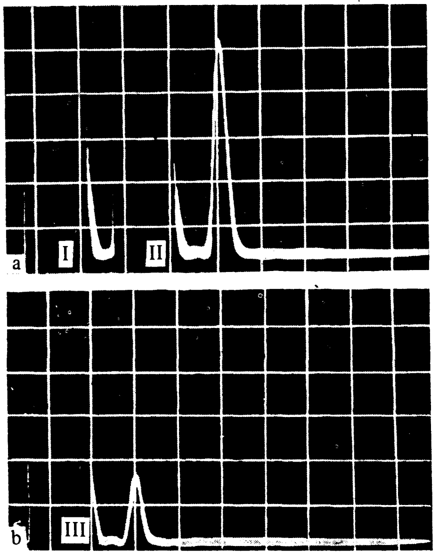

FIG. 1. Oscillograms of the two-pulse (a) and stimulated (b) echoes in finely dispersed ($d \sim 1\ \mu$) powder of $LiTbF_4$: frequency 13.4 MHz, $T = 4.2$ K, time scale 5 μsec/division; I, II, III—probing pulses.

ter of the sample reveals quite a varied behavior: in some samples, its order of magnitude is close to T_2,[1,7] in others, it can amount to minutes,[11] hours[4] and even many days.[5,6] The discovery of a stimulated echo with a long memory has caused considerable interest in connection with possible technical applications.[12]

The observation of an echo in paramagnetic powders was reported in Ref. 7; starting out from the motion of magnetostrictive excitation of sound oscillations of particles of the powder, the echo was called magnetoacoustic. In this paper, the results are given of more detailed experiments, which led to the discovery of the long-lived stimulated echo in a number of magnetic powders, and an interpretation of these results on the basis of a model in which the sound oscillations of the powder particles do not play any role.

The measurements, carried out at a temperature of 4.2 K with the help of a pulsed NMR relaxometer BKR-322s at frequencies 10–40 MHz, showed that the two-pulse echo in strong magnetic fields ($H_0 \sim 25$ kOe) in $LiTbF_4$ powder, with particle size $d \sim 50\ \mu$, has certain specific properties.

1. In the case of a stepwise increase in the stationary magnetic field, the echo amplitude A_2 adjusts to the new value H_0 in two stages: during 1–1.5 min, A_2 undergoes a small change and takes on a "quasi-equilibrium" value; then the amplitude increases monotonically and very slowly, not reaching saturation in periods of at least 2 hours.

2. At a fixed value of H_0 and stepwise increase in the alternating field H_1, the echo amplitude reaches a quasi-equilibrium value within 1–1.5 min, and upon a reversal of the value of H_1, the previous amplitude A_2 is established immediately.

3. Shaking up of the ampoule with the powder leads to a sharp and irrevisible change in A_2.

The singular features just enumerated, clearly testifying to processes of re-orientation of the particles of the powder under the action of the magnetic fields, prompted us to carry out the experiment under conditions which exclude the generation of elastic oscillations in the microcrystals. Figure 1 shows oscillograms obtained in the investigation at a frequency of 13.4 MHz in finely dispersed ($d \sim 1\ \mu$) $LiTbF_4$ powder. In spite of the fact that the particle sizes of the finely dispersed powder did not satisfy the condition of acoustic resonance ($d \ll \lambda$, λ is the sound wavelength), the amplitude of the two-pulse echo referred to $\tau = 0$ turns out to be 18 times larger than for powder of 50 μ; the characteristic time T_2 changed here from 34 to 14 μsec. The individual sequence of pulses at the times $t = 0$, $\tau \leq T_2$, T did not give a stimulated echo signal at $T \gg \tau$. However, the echo appeared, accumulated, and reached a maximum value $A_3(0)$ upon irradiation of the powder by a long series of pair impulses $(0, \tau)$, with the interval $\tau \approx 10$ μsec corresponding to the maximum of the accumulated echo A_3. After switching off the "accumulating" pulses $(0, \tau)$, the amplitude A_3 decreased for several minutes and approached a stationary value (Fig. 2), so that the signals of the stimulated echo could be observed subsequently over a time of several hours upon irradiation of the powder by single pulses (Fig. 1b); the time of observations was limited by the reserve of liquid helium. Shaking of the powder led to a complete disappearance of the stimulated echo. Similar effects were observed in the study of finely dispersed ($d \sim 1-2\ \mu$) powders of several garnets $Ln_3(Ga, Al)_5O_{12}$ ($Ln = Ho, Dy$) and ferrites (Fig. 2).

The qualitative picture of the formation of the echo signals in the finely dispersed magnetic powder in crossed stationary H_0 and alternating $H_1(t)$ magnetic field given below is similar to the theory of "geometric" echo in dielectric powders.[15] The most significant elements of this theory are the motions that the rotational motions of the particles of the powder are excited as a whole and that the long damping time of the stimu-

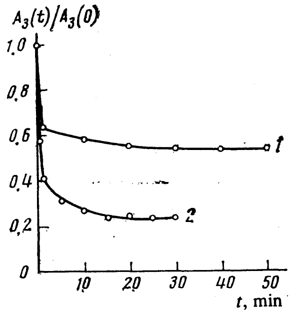

FIG. 2. Amplitude of the stimulated echo A_3 in finely dispersed magnetic powders as a function of the time elapsed from the instant of switching off the "accumulating" pulses I, II: curve 1—$LiTbF_4$, $d \sim 1\ \mu$, $T = 4.2$ K; curve 2—ferrite of type M-30-VCh, $d \sim 1\ \mu$, $T = 77$ K (measurements at a frequency of 13.4 MHz).

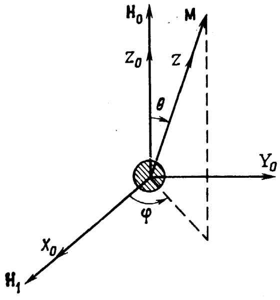

FIG. 3. Illustrating the torsional oscillations of the particles of a powder with magnetic moment **M** in crossed magnetic fields.

lated echo is a consequence of the storage of the orientation parameters of the particles.

We write out the energy of a spherical particle with radius r of a uniaxial magnet (the conditions of sphericity and uniaxiality are not necessary for formation of echo signals, but they materially simplify the analysis) in the form (see Fig. 3)

$$E=E_{\mathrm{rot}}+E'-H_0M\cos\theta-H_1(t)M\sin\theta\cos\varphi, \tag{1}$$

where E_{rot} is the energy of rotation, E' is the energy of interaction of the given particle with all the others, and determines the frictional force in their relative motion, and **M** is the magnetic moment of the particle (in the case of a paramagnetic powder, we set $g_{\parallel}\beta H_0>kT$ for simplicity). The field H_0 orients the microcrystals, and we shall be interested in the motion only of those for which $\theta\sim 0$. The equations of motion have the form

$$\frac{d^2\theta}{dt^2}+2\Gamma\frac{d\theta}{dt}+\omega_0^2\theta=\omega_0^2\frac{H_1(t)}{H_0}\cos\varphi+\left(\frac{d\varphi}{dt}\right)^2\theta+O\left(\theta^3\omega_0^2,\ \omega_0^2\theta^2\frac{H_1}{H_0}\right), \tag{2}$$

$$\frac{d}{dt}\left(\theta^2\frac{d\varphi}{dt}\right)+2\theta^2\Gamma'(\theta)\frac{d\varphi}{dt}=-\omega_0^2\frac{H_1(t)}{H_0}\theta\sin\varphi, \tag{3}$$

where $\omega_0\approx(MH_0/I)^{1/2}\sim 1/r$ is the frequency of the characteristic torsional oscillations of a particle with moment of inertia I in the field H_0 (for example, for the paramagnetic powder $LiTbF_4$ with particle size $d\sim 1\ \mu$ in a field $H_0=10$ kOe and at liquid helium temperatures, $\omega_0/2\pi\sim 8$ MHz), Γ and Γ' are the constants of the damping due to friction. The components that are nonlinear in θ and that are determined by the frictional forces are not written out explicitly.

In the case of excitation of the motion of the powder particles by pulses of an alternating field of duration Δt_i at the times $t_i=0$, τ, and T, the signal induced in the receiving coil is determined by the projection of the total magnetic moment of the microcrystals in the direction of the alternating field:

$$A\sim\sum M\theta\cos\varphi, \tag{4}$$

where the sum extends over particles with different initial orientations and eigenfrequencies ω_0. If the frequency of the alternating field ω is close to the frequency of the characteristic torsional oscillations ($|\omega_0^2-\omega^2|\ll\omega\Gamma\ll(\Gamma')^2$), then the nonlinear system of equations (2) and (3) has a solution with $\varphi=\varphi_0+\Delta\varphi(t)$, $\Delta\varphi(t)\ll\varphi_0$, $d\varphi/dt\ll\omega_0$ ($\Gamma\ll\Gamma'$), i.e., in this case, we can neglect in first approximation the change of the angle φ, as well as the second component in the right side of Eq. (2); this equation determines the amplitudes of the stimulated oscillations at $H_1\neq 0$ and, with account taken of the nonlinear components—the two-pulse echo signal. After each pulse of the alternating field the linearized Eq. (2) has the solution

$$\theta_i(t>t_i)=a_i\cos\varphi\exp[(i\omega_0-\Gamma)(t-t_i)]+\text{c.c.} \tag{5}$$

The explicit expressions for the amplitudes a_i as functions of the pulse length Δt_i can easily be written down by using the results of Refs. 10, 13, and 14; however, the absence of stability in the results of the measurement of the amplitude of the signal echoes, due to the gradual orientation of the powder in the stationary field, does not permit us to carry out a quantitative analysis of the theoretical dependences. The nonlinear response of the powder after the second pulse can be estimated by replacing, for example, θ^3 in Eq. (2) by $(\theta_1+\theta_2)^3$. The induced oscillations with

$$\Delta\theta_2\sim a_1^*a_2^2\exp[i\omega_0(t-2\tau)]+\text{c.c.}, \tag{6}$$

which are dephased because of the scatter of the eigenfrequencies ω_0, become phased-in and give a signal echo at the instant of time $t=2\tau$. We note that the nonlinear components, considered in Ref. 15, of the Hamiltonian of the torsional oscillations, are quadratic in the alternating field, and are effective only when the moment of the particle (in our case, the magnetic moment) reacts instantaneously to the alternating field, i.e., if $\omega\ll T_1^{-1}$, where T_1 is the spin-lattice relaxation time. According to existing data on the spin-lattice relaxation in paramagnets, at liquid helium temperatures $T_1^{-1}\ll 10^7\ \text{sec}^{-1}$, i.e., the terms of the Hamiltonian that are quadratic in the alternating field are ineffective in our case.

For observation of the two-pulse echo, the condition $\Gamma\tau\lesssim 1$ must be satisfied: the torsional powder-particle oscillations induced by the first pulse should exist at the instant of action of the second pulse. The stimulated echo can be observed upon total extinction of the oscillations of the microcrystals ($T\gg 1/\Gamma$), since it is the result of the interference of the oscillations induced in the microcrystals by the third pulse and rotated around H_0 during the second pulse; the fixed rotations through the angles $\Delta\varphi(\tau,\Delta t_2)$ also contain the "memory" of the interval τ between the first and second pulses. The rotations of the particles of the powder about H_0 are described by the nonlinear equation (3). We shall be interested in the monotonic solution of this equation, which, after replacement of $\theta_1(t)$ by θ takes at the time of action of the second pulse the form

$$\frac{d^2\varphi}{dt^2}+2\Gamma'\frac{d\varphi}{dt}=-\frac{1}{2}\omega_0^2\frac{H_1}{H_0}\frac{\sin\varphi_0}{|\theta|^2}e^{-\Gamma t}\cos\varphi_0\cdot(a_1\exp\{i[\omega\tau+(\omega_0-\omega)t]\}+\text{c.c.}) \tag{7}$$

and is identical in form with Eq. (4) of Ref. 10, which forms the basis of the mechanism of formation of the long-lived stimulated echo in piezoelectric powders. The solution of Eq. (7) can be written in the form

$$\Delta\varphi(t)\sim(be^{i\omega_0\tau-\Gamma\tau}+\text{c.c.})\sin\varphi_0\cos\varphi_0; \quad (8)$$

the amplitude of the rotation angle b is a function of the intensity of the alternating field of the first and second pulses. The response of the powder to the third pulse

$$A\sim\sum M\theta_3(t>T)\cos(\varphi_0+\Delta\varphi)$$

contains the signal of the stimulated echo

$$A_3\sim\sum Ma_3b^*(\sin\varphi_0\cos\varphi_0)^2e^{i\omega_0(t-T-\tau)}+\text{c.c.} \quad (9)$$

as a result of the phasing-in of the transverse components of the magnetic moments of the particles in the direction of H_1 at the instant of time $t=T+\tau$. As a consequence of the gradual orientation of the magnetic moments of the particles along the constant field H_0, the value of the stimulated echo A_3 can accumulate if the powder is irradiated by a sequence of pairs of pulses $(0, \tau)$. Because of the nonlinear dependence of the amplitude of the rotation angle b on the interval τ, the function $A_3=A_3(\tau)$ changes non-monotonically—its maximum can be expected at $\tau\sim 1/\Gamma$.

The absence of a stimulated echo with long memory in coarse powder and the presence of it in finely dispersed powder can be understood if we assume that, in contrast to the coarse particles, the fine ones are bound to one another by strong coupling forces, which prevent the disruption of the fixed-rotation structure formed as a result of the action of the pulses $(0, \tau)$. The fact that the fine particles easily adhere to one another, forming clumps, testifies to the validity of this assumption, since the coarse particles exhibit no such tendency toward adhesion and remain individual particles.

In conclusion, it should be noted that the study of the powder echo does not yield significant physical information, because the individual properties of the solids are lost in this effect, and only the characteristics of the powder remain mainly essential. Important information on the spin-phonon and spin-spin interactions in concentrated paramagnetic crystals could be given by the magneto-acoustic echo—the analog of the electroacoustic or phonon echo, studied in piezo- and ferroelectric crystals.[16–18] However, the magneto-acoustic echo in single crystals has not been observed to date and there remains only the hope that the search for it at frequencies of 10^9–10^{10} Hz will ultimately meet with success.

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