these weak shifts entails therefore an analysis of the deviations of the splittings from the quadrupole regularities.

[2)]At $I>\frac{1}{2}$ a transition to the case of the equidistant spectrum is impossible, since we have considered above an excitation which is not realizable in an equidistant spectrum, of only one transition.

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Investigation of the parametric mechanism of spin-echo formation and the dynamics of spin motion in systems with a dynamic frequency shift

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The formation of a nuclear spin-echo signal from a resonance radio-frequency pulse and a parametric-pumping pulse at double the frequency has been investigated experimentally in a system of Mn^{55} nuclei in $MnCO_3$ and $CsMnF_3$ in the case of arbitrary pulse duration and shape. Echo signals have been obtained from more complex sequences of resonance and parametric pulses. The time dependence of the oscillation amplitude of the parametrically excited nuclear spin system has been directly observed. A technique has been developed for measuring the frequency distribution of the radio-frequency radiation of a spin system excited by a resonance radio-frequency pulse. The frequency distribution of the radio-frequency radiation has been investigated in systems with a dynamic frequency shift under conditions of frequency-modulated echo formation. A theory of parametric echo formation is developed, and a theoretical interpretation of the experimental results is presented.

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1. INTRODUCTION

The known pulse techniques for investigating spin systems with inhomogeneous broadening of the resonance line consist in the excitation of these systems by a sequence of radio-frequency (RF) pulses that act in resonant fashion. There exist two mechanisms for spin-echo signal formation in such experiments: the phase mechanism (the Hahn echo)[1] and the frequency-modulation mechanism (the FM echo).[2–5]

Recently, a number of experiments on the investigation of the so-called "enhanced" echo in ferrites have been performed.[6,7] It has been shown[8] that in this case the spin echo is formed on a system of quasistationary spin waves in an inhomogeneous external magnetic field. The RF pulses excite this system parametrically. There exist, however, systems on which the pulses can act both resonantly and parametrically. To them, from among spin systems, pertain in particular, the system of Mn^{55} nuclei in antiferromagnets with the "easy-plane" type of anisotropy ($MnCO_3$ and $CsMnF_3$). In these substances the antiferromagnetic resonance frequency turns out to be so low that there arise mixed nuclear-electronic oscillations. In this case the frequency, ν_n (below it will be called the nuclear frequency), of the quasinuclear branch of the magnetic resonance shifts from the value $\nu_{n0}=\gamma H_{hf}$ by an amount, ν_{DFS}, called the dynamic frequency shift (DFS). (H_{hf} is the strength of the hyperfine field at the nuclei.) As will be shown in the Appendix, in the case of the oscillations of the quasinuclear branch the component of the resultant magnetic moment of the sample along the direction of the external magnetic field lying in the easy plane of the sample oscillates at the frequency $2\nu_n$. In view of this, a RF field of frequency $2\nu_n$ directed along the magnetic field acts parametrically on this oscillation mode.

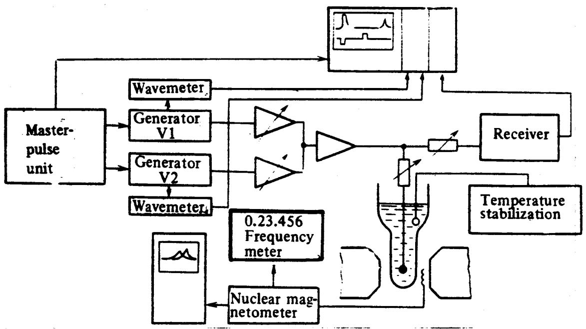

FIG. 1. Block diagram of the parametric-echo spectrometer.

We set up experiments to study the effect of parametric pumping at the frequency $2\nu_n$ on a resonantly excited nuclear spin system. For this purpose, to the system were fed a resonance RF pulse and, after a time t_2, an RF parametric-pumping pulse. An echo signal formed at the moment of time $2t_2$. The mechanism of echo-signal formation from a resonance pulse and a parametric-pumping pulse was called parametric by us. It differs significantly from the Hahn-echo and FM-echo formation mechanisms. The results of preliminary investigations of the parametric echo have already been published.[9]

In the case of the Hahn and FM echoes the amplitude of the signal is linear in the amplitude of the first RF pulse and quadratic in the amplitude of the second RF pulse (for low power of the RF pulses). In contrast, the amplitude of the parametric-echo signal is linear in the amplitudes of both RF pulses.[9] Thus, the parametric-echo formation mechanism is linear, in contrast to the earlier known echo-formation mechanisms. Because of this, it is possible to obtain parametric-echo signals from long RF pulses ($\tau_{pul} > 1/\delta\nu_n$), where $\delta\nu_n$ is the value of the inhomogeneous broadening of the NMR lines. In this case the echo signal forms primarily from the spins with frequency close to the frequency of the resonance pulse, or to half the frequency of the parametric pulse, which enables us to investigate by the pulse technique a distinct part of the spin system. This property of the parametric echo is especially useful in the study of spin systems with complex spin-motion dynamics, in particular systems with DFS, in which the spin-precession frequency depends on the angle of inclination of the spins to the equilibrium axis. The dynamics of spin motion in such systems was studied earlier with the aid of the FM echo.[3,4,10,11] However, the second resonance RF pulse changes the precession frequencies of different spins by different amounts, which, on the one hand, leads to the formation of the FM echo, but, on the other hand, makes the interpretation of the results very complicated. In the present work we have investigated with the aid of the parametric echo both the dynamics of spin motion in systems with DFS and the formation of the FM echo.

The investigations were performed on a system of Mn^{55} nuclei in the antiferromagnets $MnCO_3$ and $CsMnF_3$, which have easy planes. The magnetic properties of these substances have been studied sufficiently thoroughly.[12] The parametric excitation of nuclear spin waves and of uniform precession have been investigated.[13,14] The system of Mn^{55} nuclei in these materials undergoes a dynamic frequency shift. A detailed investigation of this system with the aid of the FM echo is presented in Ref. 3.

2. MEASUREMENT PROCEDURE

The investigations were performed on synthetic single crystals. The $MnCO_3$ samples had the form of a disk cut out parallel to the easy plane of the crystal; the $CsMnF_3$ samples had irregular shapes.[1)] The measurements were performed on an NMR pulse spectrometer (Fig. 1) in the 150–1500 MHz frequency band. For the generation of the RF pulses of resonance and double frequencies, two RF generators were used. The signals from the generators were fed to a single-turn coil, in which the sample was located. The RF parametric-echo signal was received by the same coil. The axis of the RF coil and the external magnetic field were set parallel to the easy plane of the sample at an angle of $\varphi = 55°$ to each other. It has been experimentally demonstrated[9] that, in such a geometry, the amplitude of the parametric-echo signal is maximal. This is connected with the fact that the RF field $\mathbf{h}_\perp \perp \mathbf{H}_0$ acts resonantly on the system at the frequency ν_n, while the RF field $\mathbf{h}_\parallel \parallel \mathbf{H}_0$ acts parametrically at the frequency $2\nu_n$. In its turn, the RF field of the echo signal is perpendicular to the external field. Therefore, the parametric-echo intensity

$$I \sim h_\perp h_\parallel \sin\varphi = h^2 \sin^2\varphi \cos\varphi. \quad (1)$$

The RF parametric-echo signal was fed to a superheterodyne receiver. From the receiver the signal proceeded to an oscillograph, on which its time sweep was observed. To the second ray of the oscillograph were fed signals from frequency meters, which were used to measure the frequency, and which showed the time of action of the RF pulses. On the oscillograms a signal from a frequency meter tuned to the frequency ν_n is a negative pulse, while a signal from a frequency meter adjusted to the frequency $2\nu_n$ is a positive pulse. The sensitivity of the receiver was of the order of 10^{-13} W. The echo-signal frequency was measured by fine tuning of the superheterodyne receiver, precalibrated with a signal from an external generator. The amplitude of the echo signal was measured directly on the scale of the oscillograph with the use of the entrance attenuator of the receiver. The measurements were performed in the 4.2–1.3 K temperature range. To prevent overheating, the samples were placed directly in a liquid-helium bath. The stabilization and measurement of the temperature were accomplished to within 0.03 K with the aid of a germanium thermometer.

3. THE TWO-PULSE PARAMETRIC ECHO

To generate the parametric-echo signal, to the spin system were fed a RF pulse at the resonance frequency and, after a time t_2, a RF pulse at double the frequency. An induction signal was observed immediately after the resonance pulse. The induction signal did not appear during the time of action of the parametric pulse. At the moment of time $2t_2$ a nuclear-spin echo signal ap-

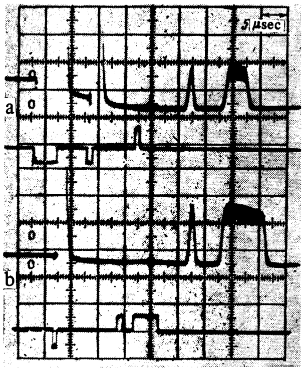

FIG. 2. Oscillogram of the parametric-echo signal in the case when the resonance (a) and parametric (b) pulses have complex shapes (the location of a resonance pulse is indicated as a negative signal on the lower ray, while the location of a parametric pulse is indicated as a positive signal).

peared at the NMR frequency.

The parametric-echo method allows the investigation of the NMR signal in the case of large DFS values. We were able to observe parametric-echo signals at frequencies of 150–656 MHz, which correspond to DFS values of 516–10 MHz, whereas by the standard NMR pulse technique we observed echo signals at DFS values of not more than 150 MHz. The amplitude of the parametric-echo signal is proportional to the RF-field amplitudes of both pulses,[9] which allows the observation of the parametric-echo signal at lower RF-pulse powers than in the case of the FM echo.

When the parametric-echo signal is generated by short RF pulses (τ_r, $\tau_p \ll 1/\delta\nu_n$, where τ_r is the duration of the resonance, and τ_p that of the parametric pulse), the echo-signal length is, as in the case of the normal echo, equal to the reciprocal halfwidth, $1/\delta\nu_n$, of the resonance line. However, in contrast to the normal echo, the parametric-echo signal also forms from long RF pulses (τ_r, $\tau_p \gg 1/\delta\nu_n$). The investigations of the echo signal formed from long RF pulses were performed on $CsMnF_3$ at the frequency $\nu_n = 591$ MHz ($\nu_{DFS} = 75$ MHz) at a temperature of 2 K. The halfwidth of the NMR line at this point is $\delta\nu_n = 1.5$ MHz (see Fig. 11). In the investigations the amplitude of the resonance RF pulse satisfied the condition $\nu_{DFS}(\gamma\eta_\perp h_\perp)^2 \ll 1/\tau_r^3$, where $\eta_\perp = H_{hf}/H_0$ is the enhancement ratio for the RF field $h_\perp$. In this case, as was demonstrated in Ref. 15, we can neglect the effects connected with the variation of the spin-precession frequency during the time of action of the resonance pulse.

In the case of a long rectangular resonance pulse of length τ_r and a short parametric pulse, an echo signal of duration τ_r and frontal width $\sim 1/\delta\nu_n$ is observed in the interval of time from $2t_2-\tau_r$ to $2t_2$. The echo-signal amplitude does not depend on the resonance-pulse length. The echo-signal frequency corresponds to the resonance-pulse frequency.[2)] When a resonance pulse of complex shape is applied, the echo signal duplicates the pulse shape (with allowance for the relaxation process) in the reversed-time direction (Fig. 2a).

In the case of a short resonance pulse and a long rectangular parametric pulse of length τ_p, an echo signal of duration $2\tau_p$ and frontal width $\sim 1/\delta\nu_n$ is observed in the time interval from $2t_2$ to $2t_2 + 2\tau_p$. The echo-signal amplitude does not depend on the parametric pulse length. The echo-signal frequency is equal to half the frequency of the parametric pulse (see footnote 2). When a parametric pulse of complex shape is supplied, the echo signal duplicates the shape of this pulse (if the relaxation process is taken into consideration) with the time scale increased by a factor of two (Fig. 2c).

In the case when both RF pulses are long, the parametric-echo signal has a complex shape. We can then distinguish three regions in the signal: a central region and two side regions that behave differently. Let us consider in detail the situation when $\tau_r < \tau_p/2$. In this case the side regions of the echo signal lie in the time intervals from $2t_2 - \tau_r$ to $2t_2$ and from $2t_2 + 2\tau_p - \tau_r$ to $2t_2 + 2\tau_p$, while the central region lies in the interval from $2t_2$ to $2t_2 + 2\tau_p - \tau_r$ (see Fig. 3).

If the frequency of the parametric pulse is equal to double the frequency of the resonance pulse, the parametric-echo signal has the shape of a trapezium with a flat central part (Fig. 3a). When the resonance-pulse frequency is varied, the central region of the echo signal remains flat. In this case the intensity of the echo signal in this region decreases to zero (Fig. 3b) and then begins to oscillate, attaining its maximum values at $(\nu_r - \nu_p/2)\tau_r = n + \frac{3}{2}$ (Fig. 3, c–d). The dependence of the echo-signal amplitude in the central region on the detuning $\nu_r - \nu_p/2$ for $\tau_r = 4$ μsec is depicted in Fig. 4 by the small circles. The echo-signal frequency in the central region is equal to $\nu_p/2$. In the case of maladjustment there arise in the side regions of the echo signal spikes, the number of which is equal to the number of oscillations in the central region (Fig. 3, a–d).

In the case when $\tau_r > \tau_p/2$, the experimental picture is completely similar to the preceding picture if we interchange in it τ_r and $2\tau_p$, as well as ν_r and $\nu_p/2$. The dependence of the echo-signal amplitude in the central region on the detuning $\nu_p/2 - \nu_r$ for $\tau_p = 2$ μsec in this case is depicted in Fig. 4 by the crosses. The continuous curve in Fig. 4 represents the Fourier transform of a

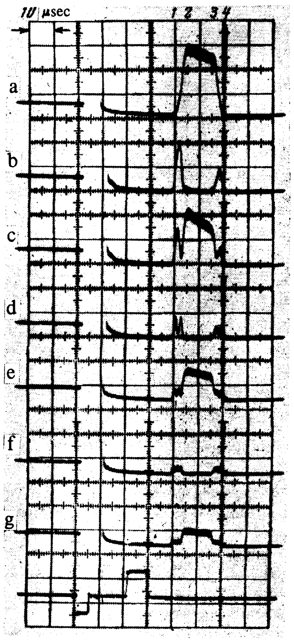

FIG. 3. Oscillograms of the parametric-echo signal upon the variation of the resonance-pulse frequency: $(\nu_r - \nu_p/2)\tau_r = 0$ (a), 1 (b), $\frac{3}{2}$ (c), 2 (d), $\frac{5}{2}$ (e), 3 (f), $\frac{7}{2}$ (g); $t = 2t_2 + \tau_r$ (1), $2t_2$ (2), $2t_2 + 2\tau_p - \tau_r$ (3), $2t_2 + 2\tau_p$ (4).

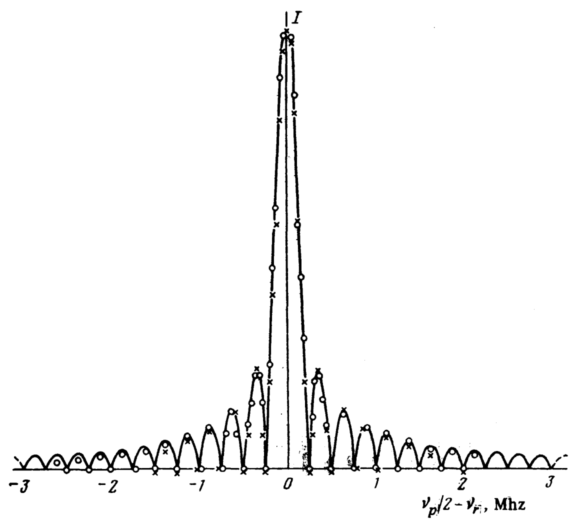

FIG. 4. Dependence of the amplitude of the parametric-echo signal on $\nu_r - \nu_p/2$: O) $\tau_r = 4$ μsec, $\tau_p = 10$ μsec, and ν_r is varied; ×) $\tau_r = 10$ μsec, $\tau_p = 2$ μsec, ν_p is varied; the continuous curve is the theoretical curve–the Fourier transform of a rectangular pulse of duration 4 μsec.

rectangular pulse of duration 4 μsec. As will be shown later, the amplitude of the parametric-echo signal in the central region is proportional to the amplitude of the spin precession with eigenfrequency $\nu_p/2$ after the RF pulse of frequency ν_r and duration τ_r (in the case when $\tau_r < \tau_p/2$), the latter amplitude being, for small angles of inclination of the spins, proportional to the Fourier transform of the form of the resonance pulse. In the case when $\tau_r > \tau_p/2$, the amplitude of the parametric-echo signal is proportional to the change in the amplitude of the spin precession with eigenfrequency ν_r under the action of the parametric pulse, a change which is proportional to the Fourier transform of the form of the parametric pulse with a doubled time scale.

4. MECHANISM OF PARAMETRIC-ECHO SIGNAL FORMATION

To construct a theory of parametric-echo formation, we propose a model in which the spin system is regarded as consisting of spins whose motion in an external magnetic field ($\mathbf{H} \parallel z$) is described by the following expressions:

$$m_x(t) = m_0\alpha_i \cos\omega_i t, \quad m_y(t) = m_0\alpha_i \sin\omega_i t, \qquad m_z(t) = m_0 k\alpha_i^2 \sin 2\omega_i t, \tag{2}$$

where α_i is the angle of inclination of the spins with precession frequency ω_i to the equilibrium axis ($\alpha_i \ll 1$). Here and below $\omega \equiv 2\pi\nu$.

The variation of the precession amplitude of these spins under the action of a RF field of double frequency, polarized along $\mathbf{H}_0$, is described by the equation

$$\frac{d\alpha_i}{dt} = \alpha_i \gamma k h_\parallel \sin(2\varphi_i - \varphi_{\rm RF}), \tag{3}$$

where $2\varphi_i - \varphi_{\rm RF}$ is the difference between the phases of the oscillation of the i-th spin and the RF field. This equation is equivalent to Eq. (41) (see below), obtained for the experimentally investigated system, if under k we understand the amplification factor, $\eta_\parallel$, for the longitudinal RF field.

Let the spin system be deflected through an angle α by a short ($\tau_r \ll 1/\delta\nu_n$) resonance pulse. Then the component of the magnetic moment of the i-th spin along the y axis will be

$$m_{yi} = m_0\alpha \sin\omega_i t. \tag{4}$$

The total magnetization

$$M_y(t) = \int_0^\infty \Phi(\omega) m_0\alpha \sin\omega t \, d\omega \tag{5}$$

($\Phi(\omega)$ is the density of the spins precessing with frequency ω) induces in the receiving system an induction signal that dies down during a time $t \sim 1/\delta\nu_n$. If at the moment of time t_2 a RF pulse of double the frequency (a parametric pulse) and of duration $\tau_p \ll 1/\delta\nu_n$ is supplied, then the oscillation amplitude of the i-th spin will change, and will be equal to (Eq. (3) is solved in the approximation $\gamma\eta_\parallel h_\parallel \tau_p \ll 1$)

$$\begin{aligned} m_{yi} &= m_0(\alpha + \alpha\gamma\eta_\parallel h_\parallel \tau_p \sin(2\omega_i t_2 - \psi)) \sin\omega_i t = m_0\alpha \sin\omega_i t \\ &+ {}^1/{}_2 m_0\alpha\gamma\eta_\parallel h_\parallel \tau_p \cos(2\omega_i t_2 - \psi + \omega_i t) + {}^1/{}_2 m_0\alpha\gamma\eta_\parallel h_\parallel \tau_p \cos(2\omega_i t_2 - \psi - \omega_i t), \end{aligned} \tag{6}$$

where ψ is the phase of the RF field of the parametric pulse at the moment of time t_2. Here it is assumed that $(2\nu_i - \nu_p)\tau_p \ll 1$; consequently, the phase difference between the i-th oscillation and the RF field does not change during the time of action of the pulse. The total magnetization from the first two terms in (6) for $t \gg 1/\delta\nu_n$ is equal to zero, and therefore

$$M_y(t) = {}^1/{}_2 m_0\alpha\gamma\eta_\parallel h_\parallel \tau_p \int_0^\infty \Phi(\omega) \cos(\omega(2t_2 - t) - \psi) \, d\omega. \tag{7}$$

At the moment of time $t = 2t_2$ there arises a total magnetization whose time dependence is the Fourier transform of the line shape. This magnetization induces a RF echo signal in the intake.

In analyzing the echo-signal formation from long rectangular RF pulses (τ_r, $\tau_p \gg 1/\delta\nu_n$), we shall consider $\Phi(\omega)$ to be a constant ($\Phi(\omega) \equiv m$). The first resonance RF pulse will excite the spins in accordance with the Fourier transform of the pulse shape:

$$F(\nu_i - \nu_r, \tau_r) = \gamma\eta_\perp h_\perp \frac{\sin[\pi\tau_r(\nu_i - \nu_r)]}{\pi(\nu_i - \nu_r)}. \tag{8}$$

(Generally speaking, this is true only for linear systems. However, if the angle of displacement of the spins under the action of the RF field $\alpha_i \ll 1$, then the excitation of the spin system can be considered in the linear approximation. The condition $\alpha_i \ll 1$ is weaker than the condition $\nu_{\rm DFS}(\gamma\eta_\perp h_\perp)^2 \ll 1/\tau_r^3$, which was satisfied in the experiments.)

Thus, the amplitude of the oscillation of the i-th spin after the resonance pulse has the form

$$m_{yi}=m_0F(\nu_i-\nu_r,\ \tau_r)\sin(\omega_i t+\varphi(\omega_i)). \tag{9}$$

The effect of the second parametric pulse on the amplitude of the i-th oscillation should be integrated over the time of action of the pulse. As a result, the total magnetization after the parametric pulse

$$M_y(t)=m_0\int_0^\infty\Big[F(\nu-\nu_r,\tau_r)+F(\nu-\nu_r,\tau_r)\gamma\eta_\parallel h_\parallel\cdot\times\int_0^{\tau_{\rm II}}\sin(2\omega(t_2+\tau)-\omega_p\tau+\varphi(\omega))d\tau\Big]\sin\omega t\,d\omega. \tag{10}$$

(Here ψ has been set equal to zero, since its difference from zero leads only to a shift of the dependence $M_y(t)$ by the amount $\psi/\nu_n<2$ nsec.) The evaluation of this integral in the case when $t>t_2$ leads to the following result:

$$M_y(t)=\frac{2m\gamma^2\eta_\parallel\eta_\perp h_\parallel h_\perp}{2\omega_r-\omega_p}\{[\theta(t-2t_2+\tau_r)-\theta(t-2t_2)]\cos\omega_r(t-2t_2-2\tau_r)$$
$$-[\theta(t-2t_2-2\tau_p+\tau_r)-\theta(t-2t_2-2\tau_p)]\cos[\omega_r(t-2t_2-2\tau_r)+(\omega_p-2\omega_r)\tau_p]$$
$$-[\theta(t-2t_2+\tau_r)-\theta(t-2t_2-2\tau_p+\tau_r)]\cos[(t-2t_2-2\tau_r)\omega_p/2]$$
$$+[\theta(t-2t_2)-\theta(t-2t_2-2\tau_p)]\cos[(t-2t_2-2\tau_r)\omega_p/2+(\omega_p-2\omega_r)\tau_r/2]\}, \tag{11}$$

where

$$\theta(x)=0 \text{ for } x<0 \text{ and } \theta(x)=1 \text{ for } x>0.$$

In the case when $\tau_r<\tau_p/2$ there occur in the time intervals from $2t_2-\tau_r$ to $2t_2$ and from $2t_2+2\tau_p-\tau_r$ to $2t_2+2\tau_p$ in the signals of frequencies ν_r and $\nu_p/2$ beats that are easily observable experimentally (Fig. 3, b–d). From $2t_2$ to $2t_2+2\tau_p-\tau_r$ in the central region

$$M_y(t)=mF(\nu_p/2-\nu_r,\ \tau_r)\sin(\omega_p t/2). \tag{12}$$

Thus, under the assumed limitations, the amplitude of the parametric-echo signal in the central region is proportional to the amplitude of the spin excitation with eigenfrequency $\nu_p/2$ excited by the resonance RF pulse, which is in good agreement with the experimental data.

Let us consider the behavior of the echo signal in the case when the excitation of the spin system by the resonance pulse is not linear. In this case the frequency distribution of the RF emission of the spin system after the resonance RF pulse can be represented in the form

$$A(\nu_i)=\Gamma(\nu_i-\nu_r)\Phi^*(\nu_i),$$

where $\Gamma(\nu_1-\nu_r)$ is the amplitude of the excitation of the spins having, after the RF pulse, a precession frequency of ν_1, while $\Phi^*(\nu_i)$ is the density of the spins precessing with frequency ν_i after the RF pulse. Then after the action of the weak parametric pulse, which action we shall describe in the linear approximation, the term in $M_y(t)$ that produces the parametric-echo signal is equal to

$$M_y(t)=m\int_0^\infty A(\nu_i)F(\nu_i-\nu_p/2,2\tau_p)d\nu_i. \tag{13}$$

If $2\tau_p\gg\tau_r$, then the function $A(\nu_i)$, which has a period of $1/\tau_r$, varies little in a frequency interval $\sim 1/\tau_p$, while the function $F(\nu_i-\nu_p/2,\ 2\tau_p)$ attenuates rapidly when $|\nu_i-\nu_p/2|>1/\tau_p$. Therefore, the echo-signal intensity

$$I\approx A(\nu_p/2). \tag{14}$$

As a result, the parametric-echo method enables us to investigate the frequency distribution of the RF emission of an excited spin system. Let us call this experiment frequency scanning of the excited resonance line. With the aid of this experiment we investigated the form of the function $A(\nu_i)$ in systems with DFS and under conditions of FM-echo formation (Secs. 7 and 8).

All the results of the theory for the $\tau_r<2\tau_p$ case turn out to be the same as in the case when $\tau_r>2\tau_p$ if we interchange τ_r and $2\tau_p$, as well as ν_r and $\nu_p/2$. In particular, the echo-signal amplitude is proportional, in the linear approximation, to $F(\nu_p/2-\nu_r,\ 2\tau_p)$, which is in good agreement with the experimental data (Fig. 4). An elucidation of the experimental data in the case when one of the pulses is short can be obtained from the formula (11) in the limit $\tau\to 0$ for $\gamma\eta h\tau=\text{const}$. Indeed, in the case when $\tau_p\to 0$,

$$M_y(t)=m\gamma^2\eta_\parallel h_\parallel\tau_p h_\perp\eta_\perp[\theta(t-2t_2+\tau_r)-\theta(t-2t_2)]\sin[\omega_r(t-2t_2-2\tau_r)], \tag{15}$$

i.e., the parametric-echo signal should have the frequency ν_r, should be observed in the interval of time from $t=2t_2-\tau_r$ to $t=2t_2$, and its amplitude should be proportional to the resonance-pulse amplitude.

In the case when $\tau_r\to 0$,

$$M_y(t)=m\gamma^2\eta_\perp h_\perp\tau_r h_\parallel\eta_\parallel[\theta(t-2t_2)-\theta(t-2t_2-2\tau_p)]\sin[(t-2t_2)\omega_p/2], \tag{16}$$

i.e., the parametric-echo signal should have the frequency $\nu_p/2$, should occur in the interval of time from $t=2t_2$ to $t=2t_2+2\tau_p$, and its amplitude should be proportional to the amplitude of the parametric pulse.

In conclusion, let us consider the effect of DFS on the formation of a parametric-echo signal. In systems with DFS the spin-precession frequency depends on the angle of inclination of the spins to the equilibrium axis according to the law

$$\nu_n(\alpha_i)=\nu_n(0)-\nu_{\rm DFS}(1-\alpha_i^2),\qquad \alpha_i\ll 1.$$

After the action on the system of short resonance and parametric pulses, the precession of the i-th spin is described by the formulas

$$m_{yi}=m_0(\alpha+\alpha\beta\sin 2\omega_i t_2)\sin[\omega_i t+2\omega_{\rm DFS}\alpha^2\beta\cdot\times(t-t_2)\sin 2\omega_i t_2+\omega_{\rm DFS}\alpha^2\beta^2(t-t_2)\sin^2 2\omega_i t_2],$$
$$m_{xi}=m_0(\alpha+\alpha\beta\sin 2\omega_i t_2)\cos[\omega_i t+2\omega_{\rm DFS}\alpha^2\cdot\times\beta(t-t_2)\sin 2\omega_i t_2+\omega_{\rm DFS}\alpha^2\beta^2(t-t_2)\sin^2(2\omega_i t_2)], \tag{17}$$

where

$$\alpha=\gamma\eta_\perp h_\perp\tau_r,\quad \beta=\gamma\eta_\parallel h_\parallel\tau_p,\quad \omega_i=\omega_{0i}-\omega_{\rm DFS}(1-\alpha^2),$$
$$\beta<\alpha\ll 1,\quad \tau_r,\tau_p\ll 1/\delta\nu_n.$$

Here we have neglected the variation of spin-precession

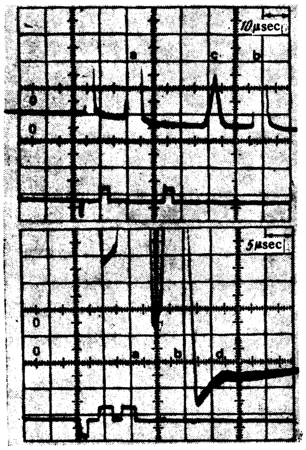

FIG. 5. Oscillograms of the echo signals in the case of the RPP pulse sequence: a) parametric-echo signal at $t = 2t_2$; b) parametric-echo signal at $t = 2t_3$; c) secondary-echo signal at $t = 2t_3 - 2t_2$; d) secondary-echo signal at $t = 2t_3 + 2t_2$.

frequencies during the time of action of the RF pulses, which can be done when the following conditions are fulfilled: $\nu_{DFS}\gamma\eta_\perp h_\perp \tau_r^2 \ll 1$ for the resonance pulse[15] and $\nu_{DFS}\gamma\eta_\| h_\| \tau_p^2 \alpha \ll 1$ for the parametric pulse.

To compute the spin-echo signal amplitude at the moment of time t, we multiply the magnitude of the magnetic moment, $m_i(t)$, by the frequency distribution of the spin density, $\Phi(\nu_i)$, and integrate over ν_i, the integration being carried out in much the same way as the one performed in Ref. 4 for the case of the ordinary FM echo. Then at the moment of time $t = 2t_2$, for the echo-signal intensity, we obtain

$$I \approx [\alpha^2\beta^2(J_0(z) - J_1(z)/z)^2 + \alpha^2 J_1^2(z)]^{1/2}, \tag{18}$$

where $z = 2\omega_{DFS}\alpha^2\beta t_2$ and $J_0(z)$ and $J_1(z)$ are Bessel functions. The linear term $\alpha\beta(J_0(z) - J_1(z)/z)$ gives, in the absence of DFS ($z = 0$), the parametric-echo signal. A DFS decreases the magnitude of this term. The term $\alpha J_1(z)$ arises as a result of the redistribution of the spins over frequency (the frequency mechanism of echo formation). Thus, when $\alpha^2\beta \sim 1/\omega_{DFS}t_2$, the frequency mechanism of echo formation makes a greater contribution to the echo signal than the linear formation mechanism. When the RF-pulse duration is increased, the contribution of the nonlinear frequency mechanism of echo formation decreases.

Unfortunately, in the antiferromagnets being studied, when the power of the resonance RF pulse is increased, the rate of the observable spin-spin relaxation increases sharply, which does not allow a direct observation of the dependence $I(\alpha, \beta)$ for a fixed delay time t_2 in the region of deviation angles corresponding to the FM-echo peak. At the moment of time $t = 4t_2$, the echo-signal intensity

$$I \approx [\alpha^2\beta^2 J_1^2(z) + 4\alpha^2 J_2^2(z)]^{1/2}, \quad z = 6\omega_p\alpha^2\beta t_2, \tag{19}$$

i.e., there appears an echo signal similar to the FM echo in the case of excitation by two resonance RF pulses. Furthermore, FM echo signals should appear at the moments of time $t = 2nt_2$. These signals could not be detected experimentally because of their small intensity ($I_{4t_2} \sim \alpha^3\beta^2$ for small α and β).

5. FORMATION OF ECHO SIGNALS FROM THREE RF PULSES

The parametric-echo formation mechanism allows the observation of echo signals from diverse sequences of resonance and parametric pulses. Some of the resulting echo signals are unusual for the traditional echo-formation mechanisms. We investigated the action on the spin system of a sequence of three RF pulses, two of which excited the spin system in accordance with the FM- or parametric-echo formation mechanisms, while secondary echo signals were generated with the aid of the third pulse. The experiments were performed on $CsMnF_3$ at $\nu_n = 500$ MHz and at a temperature of 1.3 K.

a) A RPP *(resonance, parametric, parametric pulse) sequence.* Let us consider the action on the oscillation of the i-th spin of a sequence of one resonance pulse and two parametric pulses supplied at the moments of time t_2 and t_3. In analogy to (6), we find that after the second parametric pulse (τ_{p2})

$$\begin{aligned} m_{yi} = \ldots &+ {}^1/_2 m_0\alpha\beta\cos[\omega_i(t-2t_2)] + {}^1/_2 m_0\alpha\delta\cos[\omega_i(t-2t_3)] \\ &+ {}^1/_4 m_0\alpha\beta\delta\sin[\omega_i(t+2t_2-2t_3)] + {}^1/_4 m_0\alpha\beta\delta\sin[\omega_i(t-2t_2-2t_3)], \\ &\beta = \gamma\eta_\| h_\| \tau_{p1}, \quad \delta = \gamma\eta_\| h_\| \tau_{p2}. \end{aligned} \tag{20}$$

Here we have retained only the terms whose phases vanish at $t > 0$.

The spins with different precession frequencies will become phased-in, and will give echo signals at the moments of time $2t_2$ and $2t_3$ respectively (normal parametric echo) and at the moments of time $2t_3 - 2t_2$ and $2t_3 + 2t_2$ (secondary echo signals).

An oscillogram of the secondary-echo signal at the moment of time $2t_3 - 2t_2$ is shown in Fig. 5a. The echo-signal amplitude increases linearly with increasing amplitude of the second parametric pulse. As can be seen from the formula (15), the secondary-echo signal amplitude is smaller by a factor of δ than the first-parametric-echo signal amplitude. Thus, we can measure the quantity $\eta_\| h_\|$, but this quantity is measured more accurately in experiments on the direct observation of the parametric excitation of the spin system (see Sec. 6). An oscillogram of the secondary-echo signal at the moment of time $2t_2 + 2t_3$ is shown in Fig. 5b. The amplitude of the echo signal is small, which, apparently, is connected with the increased effect of the relaxation processes on the generation of this signal.

b) RPR *sequence.* A more complex situation arises when a resonance RF pulse acts on a spin system that has been excited by a resonance, and a parametric, pulse. Let us consider the oscillation amplitude of the i-th spin in this case. After the first (resonance) pulse,

$$m_{yi} = m_0\alpha\sin\omega_i t, \quad m_{xi} = m_0\alpha\cos\omega_i t. \tag{21}$$

After the second (parametric) pulse,

$$\begin{aligned} m_{yi} &= m_0(\alpha + \alpha\beta\sin 2\omega_i t_2)\sin\omega_i t, \\ m_{xi} &= m_0(\alpha + \alpha\beta\sin 2\omega_i t_2)\cos\omega_i t. \end{aligned} \tag{22}$$

Let the third resonance pulse deflect the entire spin

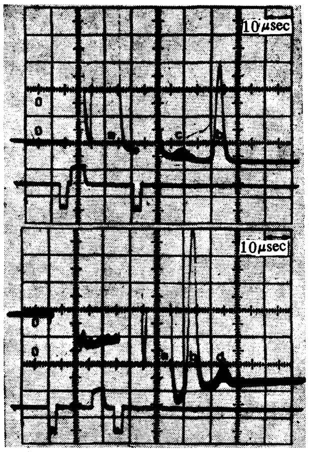

FIG. 6. Oscillograms of the echo signals in the case of the RPR pulse sequence: a) parametric-echo signal, $t=2t_2$; b) FM-echo signal, $t=2t_3$; c) secondary-echo signal, $t=t_3+(t_3-2t_2)$; d) secondary-echo signal, $t=t_3-2(t_3-2t_2)$.

system through an angle $\delta=\gamma\eta_\perp h_\perp\tau_{r2}$. Then the amplitude of the oscillation of the i-th spin

$$|m_{\perp i}|=m_0[(\alpha+\alpha\beta\sin 2\omega_i t_2)^2+\delta^2+(\alpha+\alpha\beta\sin\omega_i t_2)\delta\sin\omega_i t_3]^{1/2}$$
$$=[\alpha^2+\delta^2+2\alpha^2\beta\sin 2\omega_i t_2+\alpha\delta\sin\omega_i t_3+{}^1/_2\alpha\beta\delta\cos(\omega_i(2t_2-t_3)) -{}^1/_2\alpha\beta\delta\cos(\omega_i(2t_2+t_3))]^{1/2}. \quad (23)$$

It can be seen from this that the oscillation amplitude of the spin system is modulated with periods t_3, t_3-2t_2, and t_3+2t_2. In view of the fact that the spin system has a dynamic frequency shift, the echo signals due to the FM mechanism of echo formation should appear at moments of time that are multiples of t_3 (the FM echo), t_3-2t_2, and t_3+2t_2 after the third pulse. An oscillogram of the echo at the moments of time $t_3+n(t_3-2t_2)$ is shown in Fig. 6.

We were able to produce echo signals for $n=1$ (c) and $n=-2$ (d). The echo signals for $n=0$ and $n=1$ correspond to a FM echo (b) and a parametric echo (a) (Fig. 6). We could not produce echo signals at the moments of time $t_3+n(t_3+2t_2)$.

c) RRP *sequence*. New experimental possibilities of investigating the FM mechanism of echo formation arise when a parametric pulse is applied after two resonance RF pulses. After the pair of resonance RF pulses the spin-excitation amplitude is modulated with period t_2. In the presence of DFS this leads to FM-echo signal formation at the moments of time nt_2. A short parametric RF pulse generates from each oscillation with phase φ a secondary oscillation with phase $-\varphi$, which is equivalent to changing the sign of time (see Sec. 4). As a result, the oscillations, having been phased-in before the parametric pulse, and having produced FM-echo signals, become phased-in again in the reverse sequence, generating echo signals at the moments of time $2t_3-nt_2$.

An oscillogram of echo signals in the case of the RRP sequence of pulses is shown in Fig. 7. It is worth noting that, with the aid of a parametric pulse, it is possible to phase-in the oscillations with $n<0$ (the echo e), i.e., those oscillations that would give a FM-echo signal before the action on the system of RF pulses (an analog of the virtual image in optics). It is to be expected that the ratio of the intensity of the parametric-echo signals (the echo c) to the secondary-echo signals (the echoes d and e) will be equal to the ratio of the induction signal after the resonance pulse to the FM-echo signal (the echo a). Usually, the amplitude of the induction signal cannot be measured because of the dead time of the receiving system. For this same reason, it is not possible to investigate the FM-echo signal in the case of short delay times t_2. The third parametric pulse allows us to perform these investigations.

6. DIRECT OBSERVATION OF THE PARAMETRIC EXCITATION OF A SPIN SYSTEM

The experiment on the direct observation of the parametric excitation of the spin system of Mn^{55} nuclei in $MnCO_3$ and $CsMnF_3$ was conducted as follows. To the system were fed a short resonance RF pulse and a long parametric pulse of duration τ_p. As a result, a parametric-echo signal of duration $2\tau_p$ was produced at the frequency $\nu_p/2$. At the moment of formation of the echo signal a second parametric pulse of duration τ_{p2} was fed to the system. The effect of the parametric pulse on the echo signal can be seen in the oscillogram (Fig. 8). During the time of action of the pulse, the echo signal grows or attenuates, depending on the relation between the phases of the parametric pulse and double the phase of the oscillation generating the echo signal. For coherent pulses, this relation depends on the delay times t_2 and t_3, which we could not stabilize with the necessary precision (better than $1/\nu_n$). Therefore, in the oscillogram we show a superposition of many echo signals with a random phase relation. After the parametric pump has been switched off, the echo signal is regenerated. The rate of recovery of the intensity of the echo signal depends on the relation between the times $t_3+\tau_{p2}$ and $2t_3-2t_2$. If $2t_3-2t_2>t_3+\tau_{p2}$, then the echo signal does not change in the interval between these moments of time. In the oscillogram shown in Fig. 8, $t_3+\tau_{p2}=2t_3-2t_2$. After the moment of time $t=2t_3-2t_2$, the echo signal begins to be regenerated at a rate lower by a factor of two than the rate of parametric excitation.

The theoretical analysis of the mechanism of the formation of a parametrically excited signal amounts to the analysis of the integrals discussed in Sec. 4. Let the first short resonance pulse deflect the entire system of nuclei through an angle α. After the long second RF (parametric) pulse, the magnetization of the i-th spin

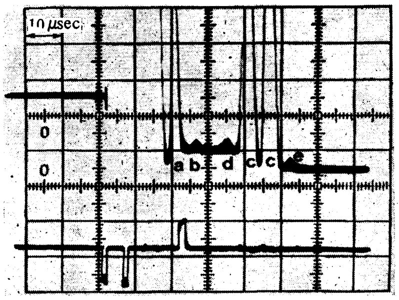

FIG. 7. Oscillogram of the echo signals in the case of the RRP pulse sequence: a) FM-echo signal, $t=2t_2$; b) secondary FM-echo signal, $t=3t_2$; c) parametric-echo signal, $t=2t_3$ and $t=2t_3-t_2$; d) secondary-echo signal, $t=2t_3-2t_2$; e) secondary-echo signal, $t=2t_3+t_2$.

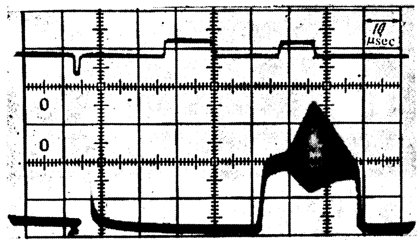

FIG. 8. Oscillogram of the direct observation of the process of parametric excitation of a nuclear spin system.

along the y axis has the form

$$m_{yi}(t)=m_0\left[\alpha+\alpha\gamma\eta_\parallel h_\parallel\int_0^{\tau_p}\sin(2\omega_i(t_2+\tau)-\omega_p\tau)\,d\tau\right]\sin\omega_i t=m_0\alpha\beta(\omega_i)\sin\omega_i t; \quad (24)$$

during the time of action of the third RF pulse

$$m_{yi}(t)=m_0\left[\alpha\beta(\omega_i)+\alpha\beta(\omega_i)\gamma\eta_\parallel h_\parallel\int_0^{t-t_3}\sin(2\omega_i(t_3+\tau)-\omega_p\tau)\,d\tau\right]\sin\omega_i t, \quad (25)$$

where t_3 is the moment of time at which the third pulse begins to act. After the third RF pulse the upper integration limit, $t-t_3$, in the dependence $m_{yi}(t)$ is replaced by τ_{p2}. Then, if $2t_2 < t_3 < 2t_2+2\tau_p$ and $\gamma\eta_\parallel h_\parallel\tau_{p2} \ll 1$, the echo signal has the form

$$\begin{aligned}I(t)=I_0\{&\theta(t-2t_2)-\theta(t-2t_2-2\tau_p)+{}^1/_2\gamma\eta_\parallel h_\parallel\sin\omega_p t_2\sin\omega_p t_3\cdot\\&[(t-t_3)(\theta(t-t_3)-\theta(t-t_3-\tau_{p2}))+\tau_{p2}(\theta(t-t_3-\tau_{p2})-\theta(t+2t_2-2t_3))\\&+(\tau_{p2}+t_3-t_2-t/2)(\theta(t+2t_2-2t_3)-\theta(t+2t_2-2t_3-2t_{p2}))]\},\end{aligned} \quad (26)$$

where I_0 is the amplitude of the parametric-echo signal in the absence of a third pulse.

Thus, during the time of action of the parametric pulse (from t_3 to $t_3+\tau_{p2}$) the parametric excitation of the spin system and, consequently, the growth of the echo-signal amplitude proceed at the rate of $\frac{1}{2}I_0\gamma\eta_\parallel h_\parallel\cos\Delta\varphi$, where $\Delta\varphi$ is the phase difference between the oscillations of the spin system along the external magnetic field and the RF field. After the parametric pulse has been switched off, the echo-signal amplitude remains constant up to the moment of time $t=2t_3-2t_2$, and is then restored at the rate of $\frac{1}{4}I_0\gamma\eta_\parallel h_\parallel\cos\Delta\varphi$ to the initial value. If $2t_3-2t_2<t_3+\tau_{p2}$, then after the instant $2t_3-2t_2$ the echo-signal amplitude increases at the rate of $\frac{1}{4}I_0\gamma\eta_\parallel h_\parallel \times\cos\Delta\varphi$, and is then restored at the same rate.

Such behavior of the echo signal was also observed experimentally. We succeeded in setting up an experiment in which the process of parametric excitation of the spin system by a longitudinal RF field can be observed directly. This experiment allows the direct measurement of the quantity $\gamma\eta_\parallel h_\parallel$. Unfortunately, the strength of the RF field $h_\parallel$ is known with low accuracy, and changes when the frequency of the parametric pulse is changed. Therefore, it is not possible to measure the quantity $\eta_\parallel$ accurately. The dependence of $\eta_\parallel$ on the magnitude of the DFS corresponds qualitatively to the theoretical dependence (see the formula (42)).

The enhancement ratios for $MnCO_3$ and $CsMnF_3$ were compared at a frequency of 500 MHz at $T=2$ K and for the same RF-field strength. It turned out that $\eta_\parallel(MnCO_3)/\eta_\parallel(CsMnF_3)=1.25$, which agrees well with the theoretical value of 1.4. (In $CsMnF_3$, $H_\Delta^2=6.2/T$[kOe^2/deg], $H_D=0$[16]; in $MnCO_3$, $H_\Delta^2=5.8/T$[kOe^2/deg], $H_D=4.4$ kOe.[17])

7. INVESTIGATION OF THE DYNAMICS OF SPIN MOTION IN A SYSTEM WITH DFS

As was theoretically shown by de Gennes,[18] in systems with DFS the spin-precession frequency depends on the angle, α, of inclination of the spins to the equilibrium position:

$$\nu_n=\nu_{n0}-\nu_{DFS}(H,T)\cos\alpha. \quad (27)$$

The angle α can be changed without changing the modulus of the nuclear magnetization vector by the action on the system of a short resonance RF pulse. In this case the frequency of the induction signal should change. Such an experiment was performed,[3] and it qualitatively confirmed the theoretical dependence (27). However, the experimental difficulties that arise in investigations with induction signals did not allow a quantitative comparison of theory and experiment to be carried out.

The theory of spin motion in systems with DFS was developed further in Ref. 2. In it a system of differential equations is obtained which describes the motion, under the action of a RF field, of the spins possessing DFS (Eqs. (23) and (24) in Ref. 2). A numerical integration, carried out by us on a computer, of these equations allowed us to compute the excitation amplitude and the change in the spin-precession frequency under the action of a rectangular RF pulse of given intensity. From these data we can compute the frequency dependence of the amplitude of the RF signal emitted by the excited spin system, $A(\nu_i)$, if we assume $\Phi(\nu_i)=$ const before the RF pulse. In Fig. 9 the dot-dash and dashed curves depict the form of the function $A(\nu_i)$ for a RF magnetic field amplitude, $\eta_\perp h_\perp$, equal to 0.4 and 0.6 Oe, a RF-pulse duration of 4 μsec, and a $\nu_{DFS}=75$ MHz. The continuous curve is a plot of the function $F(\nu_i-\nu_r$, 4

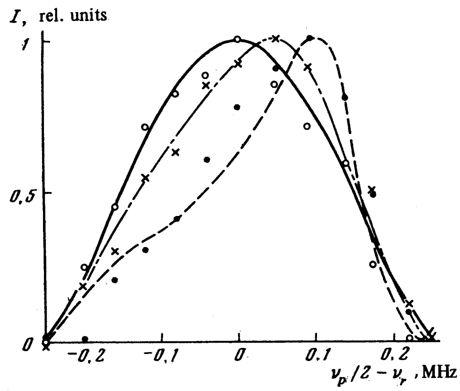

FIG. 9. Dependence of the amplitude of the parametric-echo signal on the detuning $\nu_p/2-\nu_r$ in the case when $\tau_r=4$ μsec, $\tau_p=8$ μsec: o) $\eta_\perp h_\perp=0.1$ Oe; ×) $\eta_\perp h_\perp=0.4$ Oe; •) $\eta_\perp h_\perp=0.6$ Oe; the continuous curve depicts the Fourier transform of a RF pulse of duration 4 μsec; the dot-dash, the dependence $A(\nu_i)$ for $\eta_\perp h_\perp=0.4$ Oe, $\tau_r=4$ μsec; the dashed curve, $A(\nu_i)$ for $\eta_\perp h_\perp=0.6$ Oe, $\tau_r=4$ μsec.

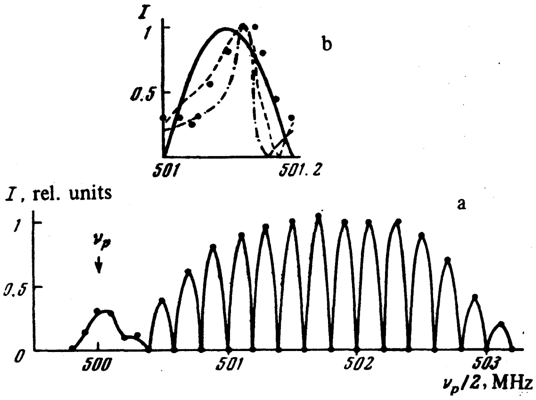

FIG. 10. Dependence of the amplitude of the parametric-echo signal on $\nu_p/2$ for RF-field intensities $\eta_\perp h_\perp = 0.1$ Oe (a) and 0.4 Oe (b) under the conditions of single-pulse formation. The dashed curve depicts the shape of the function $A(\nu_i)$ for $\eta_\perp h_\perp = 0.4$ Oe, $\tau_r = 5$ μsec; the dot-dash curve, the shape of the function $A(\nu_i)$ for $\eta_\perp h_\perp = 0.6$ Oe, $\tau_r = 5$ μsec.

μsec), i.e., of the amplitude of the radiation emitted by the excited system in the linear approximation (see Sec. 4).

The experiment on the scanning of the excited spin system by a parametric pulse, the theory of which is set forth in Sec. 4, allowed us to experimentally obtain the form of the function $A(\nu_i)$. According to the theory of the formation of a parametric-echo signal from a long parametric pulse, the amplitude of the parametric-echo signal

$$I \approx A(\nu_p/2) = \Gamma(\nu_p/2 - \nu_r)\Phi^*(\nu_p/2)$$

(see the formula (14)). The connection between the spin density before and after the RF pulse can be represented in the form of a function $C(\nu_p/2 - \nu_r)$, so that

$$\Phi^*(\nu_p/2) = C(\nu_p/2 - \nu_r)\Phi(\nu_p/2). \quad (28)$$

As a result, the echo-signal intensity

$$I \approx \Gamma(\nu_p/2 - \nu_r) C(\nu_p/2 - \nu_r)\Phi(\nu_p/2). \quad (29)$$

Thus, in the resonance-line scanning experiment, we can obtain both the form of the function $A(\nu_i)$ by varying the frequency of the parametric pulse and the form of the function $\Gamma(\nu_i - \nu_r)C(\nu_i - \nu_r)$ by varying the frequency of the resonance RF pulse (i.e., $A(\nu_i)$ under the condition $\Phi(\nu_i) = \text{const}$).

The resonance-line scanning experiment was conducted in $CsMnF_3$ at a frequency of 591 MHz at a temperature of $T = 2$ K and with RF pulses of durations $\tau_r = 4$ μsec and $\tau_p = 8$ μsec. The change in the amplitude of the spin deviation under the action of the parametric pulse was an order of magnitude less than the magnitude of the spin deviation by the resonance pulse. Therefore, the parametric pulse changed slightly the frequency distribution of the spins. The frequency dependence of the amplitude of the parametric echo for RF-pulse amplitudes $\eta_\perp h_\perp = 0.1$; 0.4; and 0.6 Oe is shown in Fig. 9, and is in satisfactory agreement with the theoretical dependence. Thus, we have obtained a quantitative corroboration of the theory of spin motion in a system with DFS.

8. INVESTIGATION OF THE FREQUENCY MECHANISM OF ECHO-SIGNAL FORMATION

In a spin system with DFS, the spin-echo signals arising under the action on the system of two short resonance RF pulses form on account of the FM mechanism of echo formation, i.e., because of the redistribution of the spin density over frequency. As is shown in Ref. 19, the FM mechanism of echo formation allows us to obtain single-pulse echo signals, i.e., echoes arising at the moments of time $n\tau_r$ after a RF pulse of duration $\tau_r \gg 1/\delta\nu_n$.

It is convenient to consider the one-pulse echo formation mechanism, using the Fourier-transform technique. Let us consider the excitation of the spin system in the linear approximation. Then the amplitude of the RF radiation of the spin system after the resonance pulse has the form

$$\bar{A}(\nu_i) = F(\nu_i - \nu_r, \tau_r)\Phi(\nu_i). \quad (30)$$

To determine the time dependence of the RF emission of the excited spin system, we should take the Fourier transform of the frequency distribution of the RF emission. Then, if we assume $\Phi(\nu_i)$ is a slowly varying function in comparison with $F(\nu_i - \nu_r, \tau_r)$, then $I(t) \approx h_\perp(t)$, and the echo signal does not form. However, in the presence of DFS, there occurs under the action of a RF pulse a redistribution of the spin density over frequency (see the formula (28)). As a result, the function $A(\nu_i)$ has a more complicated form, but retains its period $1/\tau_r$. The shape of one period of the function $A(\nu_i)$, computed from the equations of spin motion in systems with DFS[2] for $\tau_r = 5$ μsec, $\eta_\perp h_\perp = 0.4$ and 0.6 Oe, and $\nu_{DFS} = 166$ MHz, is depicted by the dashed and dot-dash curves in Fig. 10b. The form of the function $F(\nu_i - \nu_r, \tau_r)$ is depicted there by the continuous curve. The deviation of the shape of the function $A(\nu_i)$ from that of $F(\nu_i - \nu_r, \tau_r)$ leads to the result that the Fourier transform of $A(\nu_i)$ is not equal to zero at the moments of time $n\tau_r$, i.e., one-pulse echo signals are generated.

We carried out an experimental investigation of the frequency distribution of the RF emission of the spin system under conditions of single-pulse echo formation by the method of resonance-line scanning (see Secs. 4 and 7). The experiment was performed in $CsMnF_3$ at the frequency $\nu_n = 500$ MHz with $\tau_r = 5$ μsec and $\tau_p = 10$ μsec. The frequency ν_r was lower than the frequency of the NMR-line center by 1.5 MHz, which corresponded to conditions of single-pulse echo formation. In Fig. 10a, we show the dependence of the intensity of the parametric-echo signal on $\nu_p/2$. According to the theory of parametric-echo signal formation, the echo-signal intensity in this case is proportional to $A(\nu_p/2)$ (see (14)). In the case of a small amplitude of the resonance RF pulse, each peak of the dependence $I(\nu_p/2)$ is nearly sinusoidal in shape. As the intensity of the RF field is increased, the shape of the peaks become distorted and less sinusoidal. In Fig. 10b the points indicate the

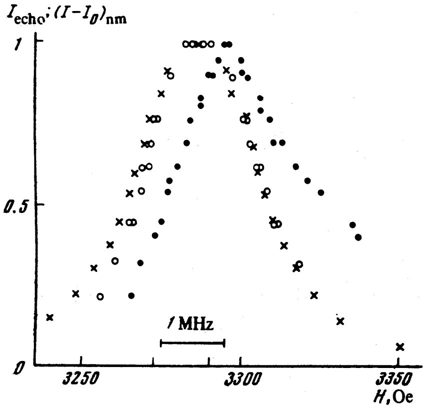

FIG. 11. The NMR-line shape in $CsMnF_3$, measured at a frequency of 591 MHz and a temperature of 2 K by the FM-echo method (●), the parametric-echo method (○), and the continuous technique (×).

parametric-echo amplitude for $\eta_\perp h_\perp = 0.4$ Oe. The satisfactory agreement between the theoretical and experimental data demonstrates the existence of the frequency mechanism of single-pulse echo signal formation.

9. THE NMR-LINE SHAPE

We carried out a comparative investigation of the shape of the resonance NMR line by the FM- and parametric-echo techniques and by the continuous method. The investigations were carried out simultaneously on one and the same setup. In the pulse methods, we measured the dependence of the echo-signal amplitude on the intensity of the constant magnetic field; in the continuous method, the dependence of the reflected-RF-signal level during the time of action of the RF pulse on the intensity of the constant magnetic field. The measurements were carried out at a frequency of 591 MHz and a temperature of 2 K with an RF field of amplitude $\eta_\perp h_\perp = 0.1$ Oe and a pulse duration of 1 μsec (in the continuous method 10 μsec). It can be inferred from the experimental data (Fig. 11) that the parametric-echo technique and the continuous method yield the real shape of the distribution, $\Phi(\nu_i)$, of the spins over frequency. The line-shape distortion arising in the case of the FM echo is evidently connected with the fact that the optimal conditions for the formation of the FM echo arise when the frequency of the RF pulses lies below the NMR frequency, i.e., when $H > H_{NMR}$.

CONCLUSION

We have succeeded in discovering and investigating a new mechanism of echo formation, called the "parametric" mechanism. It follows from the theoretical analysis that the parametric echo can be generated in all resonance systems (including those that are linear in the excitation) with inhomogeneous resonance-line broadening in which both resonance and parametric excitations are possible.

An interesting object in which it is likely we shall be able to observe the parametric echo is superfluid He^3. In He^3 there exist two coupled vibrational branches, one of which is excited by a transverse, and the second by a longitudinal, RF field.[20] Owing to the coupling between the modes, by parametrically influencing one of them, we can, we believe, excite the other mode, which will enable us to observe the parametric echo in He^3.

In contrast to the earlier known echo-formation mechanisms, a review of which is given in Ref. 21, the parametric-echo is linear in the amplitudes of the exciting RF pulses. Because of this, the parametric-echo technique provides new experimental potentialities for investigating resonance systems with complex dynamics. The parametric-echo technique enables us to obtain "long" echo signals that duplicate the shape of any of the exciting RF pulses.

Usually, the parametric excitation of spin systems is investigated with the aid of the threshold-absorption effect, which sets in only when the RF energy being fed into the system exceeds the energy being consumed on account of the relaxation processes.[13,14,22,23] The parametric-echo method allows the investigation of the process of parametric excitation of a spin system in a RF field of virtually any intensity.

In conclusion, the authors express their profound gratitude to A. S. Borovik-Romanov for direction and constant interest in the work, and to L. A. Prozorova, M. P. Petrov, M. I. Kaganov, M. I. Kurkin, and V. P. Chekmarev for fruitful discussions.

APPENDIX

Let us show how a homogeneous nuclear precession is parametrically excited in an antiferromagnet with an easy-plane type of anisotropy in the case when there is a strong dynamic coupling between the electronic and nuclear spin systems. Theoretically, this question has been investigated in papers on the parametric excitation of spin waves.[22] However, here it is presented in a form suitable for the description of the mechanism of parametric-echo formation.

Let the external magnetic field be directed along the x axis, and the difficult axis along the z axis. Then the Hamiltonian of the magnetic system has the form

$$\mathcal{H}=\mathcal{H}_0+{}^1/_4B(M^2-L^2)+{}^1/_2bL_z^2+{}^1/_2A(\mathbf{Mm}+\mathbf{Ll})-(\mathbf{M}+\mathbf{m})(\mathbf{H}+\mathbf{h}), \quad (31)$$

where $\mathcal{H}_0$ does not depend on the directions of **M**, **L**, **m**, and **l**, the ferromagnetism and antiferromagnetism vectors for the electronic and nuclear spin systems.[24]

Let us linearize the Hamiltonian of the system with respect to the deviations μ, λ, η, and ξ. Let us assume

$$\begin{aligned} &\mathbf{M}=(M_0+\mu_x,\ \mu_y,\ \mu_z), \quad \mathbf{L}=(\lambda_x,\ L_0+\lambda_y,\ \lambda_z), \\ &\mathbf{m}=(m_0+\eta_x,\ \eta_y,\ \eta_z), \quad \mathbf{l}=(\xi_x,\ l_0+\xi_y,\ \xi_z). \end{aligned} \quad (32)$$

For this choice of variables, the equations of motion separate in the linear approximation. For the low-frequency AFMR-oscillation mode and the NMR-oscillation mode connected with it, the equations of motion have the form

$$\dot{\mu}_y=(H+{}^1/_2Am_0)\mu_z+{}^1/_2AM_0\eta_z+O(\alpha^2),$$

$$\dot{\mu}_z=-(H+{}^1/_2Am_0)\mu_y-{}^1/_2Al_0\lambda_x-{}^1/_2AM_0\eta_y+O(\alpha^2),$$

$$\dot{\lambda}_x=-(BL_0-{}^1/_2Al_0)\mu_z-{}^1/_2AL_0\eta_z+O(\alpha^2),$$

$$\dot{\eta}_y={}^1/_2\gamma Am_0\mu_z+\gamma(H-{}^1/_2AM_0)\eta_z+O(\alpha^2), \quad (33)$$

$$\dot{\eta}_z=-{}^1/_2\gamma Am_0\mu_y+{}^1/_2\gamma Al_0\lambda_x-\gamma(H-{}^1/_2AM_0)\eta_y-{}^1/_2\gamma AL_0\xi_x+O(\alpha^2),$$

$$\dot{\xi}_x=-{}^1/_2\gamma Al_0\mu_z+{}^1/_2\gamma AL_0\eta_z+O(\alpha^2).$$

Here the $O(\alpha^2)$ denote the terms quadratic in the displacement from the equilibrium position; $\gamma_e=1$.

Taking into account the fact that, in equilibrium, $M_0=H/B$, we find the eigenfrequencies of the coupled nuclear-electronic vibrations:

$$\omega_e^2=H^2\left(1-\frac{ABl_0L_0}{2H^2}\right)=H^2+H_\Delta^2 \quad \text{(quasi AFMP)},$$
$$\omega_n^2=\frac{1}{4}\gamma^2A^2L_0^2\left(1-\frac{ABl_0L_0}{2H_2}\right)^{-1}=\gamma^2H_{\rm hf}^2\left(1+\frac{H_\Delta^2}{H^2}\right)^{-1} \quad \text{(quasi NMR)}, \quad (34)$$

where H_Δ is the effective anisotropy field, as seen by the nuclei, which leads to a gap in the AFMR spectrum. For the frequency ω_n, the set of eigenvector components for the system has, up to the $O(\gamma)$ terms, the form

$$\mu_y=\alpha l_0\frac{AL_0}{2H}\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-1}\sin\omega_nt,$$

$$\mu_z=\alpha l_0\frac{A}{2B}\left(1+\frac{ABL_0l_0}{2H^2}-\gamma\frac{ABL_0^2}{2H^2}\right)\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-1}\cos\omega_nt,$$

$$\lambda_x=-\alpha l_0\frac{ABL_0^2}{2H^2}\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-1}\sin\omega_nt, \quad (35)$$

$$\eta_y=\alpha l_0\frac{2H}{AL_0}\left(\frac{A}{2B}-1-\frac{ABL_0l_0}{2H^2}\right)\sin\omega_nt,$$

$$\eta_z=-\alpha l_0(1+ABl_0L_0/2H^2)^{1/2}\cos\omega_nt, \quad \xi_x=\alpha l_0\sin\omega_nt.$$

It can be seen from this that each nuclear spin rotates on an ellipse with a ratio of the axes equal to $(1+ABL_0l_0/2H^2)^{1/2}$, while in the electronic system the oscillations in M_z are much smaller than in M_y and L_x.

The possibility of a parametric excitation of the quasinuclear oscillation mode is connected with the fact that, during the time of precession of the nuclear-electronic spin system at the frequency ω_n, the spin system-magnetic field coupling energy $((M_0+\mu_x+m_0+\eta_x)H)$ oscillates with frequency $2\omega_n$. As a result, by modulating the external magnetic field strength with the frequency $2\omega_n$, we can parametrically excite the quasinuclear oscillation mode. Let us find the oscillation amplitude, μ_x, from the equations of motion for the high-frequency AFMR oscillation mode, regarding the quadratic terms, which couple this mode to the low-frequency AFMR mode, and the NMR as the exciting force:

$$\dot{\mu}_x=(-bL_0+{}^1/_2AL_0)\lambda_z-{}^1/_2A\eta_z\mu_y+{}^1/_2A\eta_y\mu_z+O(\alpha^2),$$

$$\dot{\lambda}_y=\left(\frac{bH}{B}-\frac{AHl_0}{2BL_0}\right)\lambda_z+B\lambda_x\mu_z-\frac{A}{2}\xi_x\mu_z+\frac{A}{2}\lambda_x\eta_z+O(\alpha^2), \quad (36)$$

$$\dot{\lambda}_z=\left(BL_0-\frac{A}{2}l_0\right)\mu_x+\frac{AHl_0}{2BL_0}\lambda_y-B\lambda_x\mu_y+\frac{A}{2}\xi_x\mu_y-\frac{A}{2}\eta_y\lambda_x+O(\alpha^2).$$

Substituting the oscillation amplitudes, μ_y, μ_z, λ_x, η_y, η_z, and ξ_x, found above, we obtain

$$\mu_x=\frac{A^2BL_0^2l_0^2}{16H^3}\alpha^2\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-2}\sin 2\omega_nt. \quad (37)$$

Similar oscillations are executed also by η_x, but the amplitude of these oscillations is several orders of magnitude smaller. The energy entering the system under the action of the RF field $h_\parallel$ in one period is equal to

$$\varepsilon=\int_0^T\mu_xh_x\,dt=\frac{h_\parallel A^2BL_0^2l_0^2}{32H^3}\alpha^2\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-2}\cos(\varphi_{\mu x}-\varphi_{\rm RF}). \quad (38)$$

The energy of the quasinuclear oscillation mode is equal to

$$E={}^1/_8AL_0\alpha^2(1+ABL_0l_0/2H^2)^{1/2}. \quad (39)$$

The variation in time of the energy of the system is equal to

$$\frac{dE}{dt}=2\omega_n\varepsilon, \quad (40)$$

from which we find the equation of motion for α:

$$\frac{d\alpha}{dt}=\alpha\gamma h_\parallel\frac{A^2L_0^2Bl_0}{8H^3}\left(1-\frac{ABL_0l_0}{2H^2}\right)^{-2}$$
$$\times\left(1+\frac{ABL_0l_0}{2H^2}\right)^{-1/2}\cos(\varphi_{\mu x}-\varphi_{\rm RF})=\alpha\gamma\eta_\parallel h_\parallel. \quad (41)$$

The quantity attached to $h_\parallel$ as a coefficient is the enhancement ratio for the longitudinal RF field, $\eta_\parallel$, which, in the effective-field notation, is equal to:

$$\eta_\parallel=\frac{H_\Delta^2HH_{\rm hf}}{2(H^2+H_\Delta^2)^2}\frac{\nu_{n0}}{\nu_n}.$$

For low DFS values

$$\eta_\parallel\approx\frac{H_{\rm hf}}{H}\frac{\nu_{\rm DFS}}{\nu_n}.$$

The qualitative picture of the process remains entirely the same in the presence of weak ferromagnetism. In this case

$$\eta_\parallel=\frac{H_\Delta^2(H_D+2H)H_{\rm hf}}{4(H(H+H_D)+H_\Delta^2)^2}\frac{\nu_{n0}}{\nu_n}. \quad (42)$$

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2) To verify this conclusion, we separately varied the frequencies of the resonance and parametric pulses and determined the echo-signal frequency by the method of fine tuning of the heterodyne receiver.

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