Parametric nuclear spin echo

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A spin-echo signal of Mn^{55} nuclei in $CsMnF_3$, from a radio-frequency pulse at NMR frequency and a parametric-pumping pulse at double the frequency, was obtained and was investigated experimentally. The formation of the parametric-echo signal is analyzed theoretically.

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In antiferromagnets with easy plane anisotropy ($MnCO_3$, $CsMnF_3$), at liquid helium temperature, the coupling between the system of the Mn^{55} nuclei and the electron spin system becomes so large that it is necessary to consider mixed nuclear-electronic oscillations, subdividing the resonance modes by frequencies into quasinuclear and quasielectronic. In the quasinuclear spin system, collective effects appear, described by the Suhl-Nakamura interaction. In[2,3], by the method of parallel parametric pumping at double frequency, it was possible to excite nuclear spin waves. Our investigation of the effect of a parametric-pump pulse on a nuclear spin system excited by a ratio frequency (RF) pulse at the NMR frequency has led to observation of a new echo-formation mechanism, which we call "parametric."

The investigation was carried out on single-crystal $CsMnF_3$, using the spectrometer described in[1]. The external magnetic field and the radio frequency fields were in the basal plane of the sample at an angle ϕ to each other. The radio-frequency fields were excited by a loop at the end of the coaxial line. The RF signal from the sample was received by the same loop. The parametric-echo signal was excited in the NMR-frequency band $\nu_{NMR} = 500–600$ MHz. The investigations were carried out at $\nu_{NMR} = 500$ MHz, which corresponds to a dynamic frequency shift (DFS) $\nu_p = 166$ MHz. At the instant of time t_1, a short RF pulse at the NMR frequency was applied to the system. After time $t = t_{12}$, an RF pulse at double the frequency (parametric pulse) was applied. At the instant $t = 2t_{12}$, an echo signal was produced at the NMR frequency (Fig. 1a). The echo signal was observed only in the region of the homogeneous precession of the nuclear spin system $|\nu_{NMR} - \nu_2/2| < \delta\nu_{NMR}$, where $\delta\nu_{NMR} \approx 1$ MHz is the inhomogeneously broadened NMR line, and ν_1 and ν_2 are the frequencies of the first and second RF pulses.

We plotted the signal intensity against the angle between the RF and the constant magnetic fields (Fig. 2). At the minimum possible delay times ($\sim 5\ \mu$sec), we plotted the dependence of the intensity of the echo signal on the RF pulse power (Fig. 3). The power of one of the pulses was varied, and the power of the other pulse remained constant at 100 MW. The echo-signal intensity (in power units), is proportional to the power of the RF pulses. The deviation from the linear law at large powers of the first pulse is connected with the nonlinearity of the trajectories of the motion of the nuclear spins in systems with dynamic frequency shift.[4] For comparison, we show the dependence of the intensity of

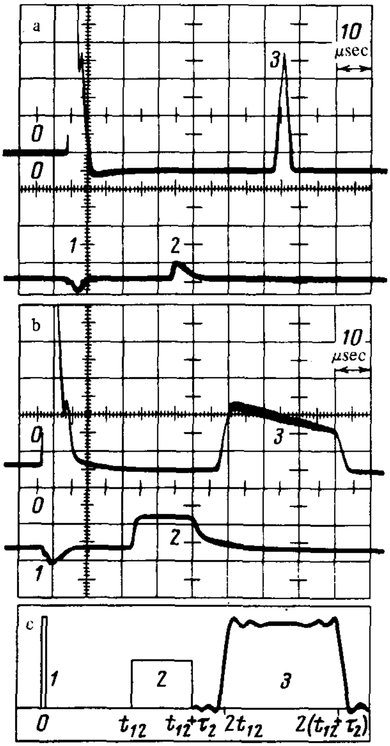

FIG. 1. a, b) Oscillograms of parametric-echo signal. Lower trace—signals from frequency meters, showing the location and duration of the RF pulses. c) Shape of parametric-echo signal in accordance with formula (4): 1) signal from frequency meter $\nu = 500$ MHz; 2) signal from frequency meter $\nu = 1$ GHz; 3) parametric-echo signal.

the ordinary frequency two-pulse echo on the power of two identical RF pulses at the same system parameters ($J \sim p^3$).

At a parametric-pulse duration $\tau_2 > 1/\delta\nu_{NMR}$, the echo signal increases with increasing τ_2. At $\tau_2 > 1/\delta\nu_{NMR}$, the echo-signal intensity ceases to depend on τ_2. The echo signal was observed then in the time interval from $2t_{12}$ to $2(t_{12} + \tau_2)$, equal to double the duration of the parametric pulse (Fig. 1b). The echo-signal intensity at the instant of time $t = 2t_{12} + 2\tau$ at $0 < \tau < \tau_2$ is determined only by power of the RF pulse at the instant of time $t_{12} + \tau \pm (1/\delta\nu_{NMR})$, and does

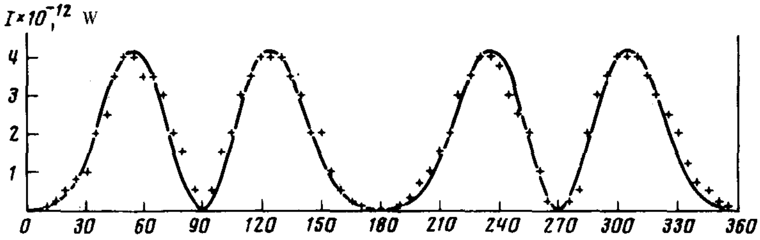

FIG. 2. Dependence of the echo-signal intensity on the angle between the RF and the constant magnetic fields. The theoretical dependence is $J \sim \sin^2 2\phi \sin^2\phi$.

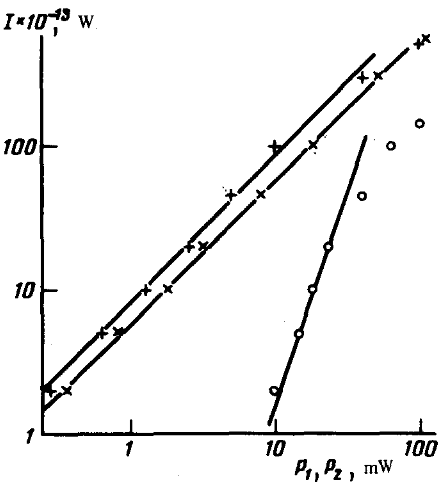

FIG. 3. Dependence of the echo-signal intensity on the RF pulse-power: +) $P_2 = 100$ MW, P_1 is varied; ×) $P_1 = 100$ MW, P_2 is varied; O) frequency mechanism of echo formation $\nu_1 = \nu_2$, $P_1 = P_2$. The straight lines correspond to the theoretical relations at small P_1 and P_2.

not depend on whether parametric pumping was produced at other times. When a parametric-pumping pulse of complicated waveform is applied, the echo signal duplicates the waveform of the pulse. The echo-signal intensity decreases with increasing $t_{12} + \tau$ in analogy with the frequency two-pulse echo.

It is shown in[5] that in antiferromagnets with anistropy of the easy plane type there exists a nonlinear connection between the high-frequency and low-frequency AFMR oscillation modes. As a result, the summary electronic magnetic moment, precessing about the external magnetic field with frequency ν, executes oscillations along the field with frequency 2ν. Because of this, it became possible in these systems to excite parametrically both spin waves and homogeneous precession. Owing to the coupling between the electronic and nuclear spin systems, it was possible to excite parametrically also the quasinuclear spin system.

Experiments performed by Prozorova and Smirnov[6] have shown that when the phase of the RF parametric pumping changes the electronic RF wave can both emit and absorb RF energy, depending on the relation between the phases of the RF pump and the sum of the time phases of the parametrically excited pair of spin waves. One can expect this property to persist in a quasinuclear spin system, since all the collective properties in a quasinuclear system are governed by the strong coupling with the electronic spin system.

Assume that at the initial instant of time a homogeneous precession of the nuclear spin system is excited by a short RF pulse at resonant frequency.[1)] Let the perpendicular magnetization of the spin i be $m_{xi} = m_0 \alpha \sin \omega_i t$, where α is the angle of inclination of the spins to the equilibrium axis and ω_i is the frequency of the oscillation of the spin i. At the instant $t = t_2$, a short parametric parallel-pumping pulse is applied. According to the solution of the Mathieu equation, which describes the parametric processes, the amplitude of the ith oscillation changes by an amount $\alpha \gamma \eta h \tau \sin(2\phi_i - \phi_0)$, where $(2\phi_i - \phi_0)$ is the difference between the phase of the RF field of the parametric pulse and double the homogeneous-precession phase, h is the intensity of the parallel-pumping

RF field, τ is the duration of the RF pulse, and η is a coefficient to be determined subsequently. (It is assumed that $\tau \ll 1/\delta\nu_{\mathrm{NMR}}$ and $a \gg \gamma\eta h\tau$.) As a result, after the parametric pulse we obtain

$$m_{xi} = m_0 (a + a\gamma\eta h\tau \sin(2\omega_i t_{12} - \phi_0)) \sin\omega_i t = m_0 a \sin(\omega_i t) - \frac{m_0 a\gamma\eta h\tau}{2} \cos(\omega_i (2t_{12} + t) - \phi_0) + \frac{m_0 a\gamma\eta h\tau}{2} \cos(\omega_i (2t_{12} - t) - \phi_0). \tag{1}$$

At the instant $t = 2t_{12}$, the phases of all the oscillations contained in the third term of (1) turn out to be equal. A macroscopic perpendicular magnetization $M_x = \Sigma_i m_{xi}$ is therefore produced; it will precess with frequency ν_{NMR} and will induce in the receiving system an echo signal with duration $\approx 1/\nu_{\mathrm{NMR}}$. If we introduce in formula (1) the relaxation frequency $\tilde{\eta}$ and consider the regime of parametric excitation of the spin waves, it turns out that the threshold excitation field is equal to $h_c = 2\tilde{\eta}/\gamma\eta$. Yakubovskiĭ[2] obtained theoretically and confirmed experimentally a formula for the parallel-pumping threshold field, $h_c = 2(\tilde{\eta}/\gamma)(H_0/H_{hf})(\nu_{\mathrm{NMR}}/\nu_p)$ (at $k=0$ and $H_D = 0$). Here H_{hf} is the hyperfine field at the nucleus. Therefore $\eta = (H_{hf}/H_0)(\nu_p/\nu_{\mathrm{NMR}}) = \eta_{\parallel}$ is the gain at $h \parallel H$, which in our case is equal to 80. The gain for $h \perp H$ is $\eta_{\perp} = H_{hf}/H_0 = 300$.

We examine the action of a given parametric pulse ($\tau_2 > 1/\delta\nu_{\mathrm{NMR}}$). During the time of action of the pulse, the difference between double the phase of the ith oscillation and the RF-field phase changes with time. Therefore,

$$m_{xi} = m_0 \left(a + a\gamma\eta_{\parallel} h \int_0^{\tau_2} \sin(2\omega_i (t_{12} + \tau) - \omega_2 \tau)\, d\tau\right) \sin\omega_i t. \tag{2}$$

Retaining only the terms that are in phase, we have

$$m_{xi} = m_0 a\gamma\eta_{\parallel} h \frac{\sin(\omega_i (t_1 - 2t_{12})) - \sin(\omega_i (t - 2t_{12} - 2\tau_2) + \omega_2 \tau_2)}{2\omega_i - \omega_2}. \tag{3}$$

Assuming for simplicity a rectangular form of the inhomogeneous broadening and $\nu_2 = 2\langle\nu_{\mathrm{NMR}}\rangle = 2\nu_0$, we have

$$M_x = \int_{\nu_0 - \frac{\delta\nu}{2}}^{\nu_0 + \frac{\delta\nu}{2}} m_{xi}\, d\nu_i = \frac{m_0 a\gamma\eta_{\parallel} h}{2\delta\nu} \left[\mathrm{si}\left(\frac{\delta\nu}{2}(2t_{12} - t)\right) - \mathrm{si}\left(\frac{\delta\nu}{2}(2t_{12} + 2\tau_2 - t)\right)\right] \times \cos(\omega_0 t + \phi). \tag{4}$$

The echo-signal envelope calculated from formula (4) is shown in Fig. 1c. The echo signal should be observed from the instant of time $t = 2t_{12}$ to $t = 2(t_{12} + \tau)$, which agrees well with experiment. The change of the echo-signal intensity, which is seen on the oscillogram (Fig. 1b), is connected with the relaxation process.

The calculated parametric-echo signal amplitude is expressed by the formula $M_x = \gamma^2 \eta_{\perp} \eta_{\parallel} h_{\perp} h_{\parallel} \tau_1 M_0 / 2\delta\nu$ at small $h_{\perp}$, τ_1, and $\tau_2 > 1/\delta\nu$. In our experiment $h_{\perp} = h \sin\phi$ and $h_{\parallel} = h \cos\phi$. In addition, the sensitivity of the receiving loop is

$\sim \sin\phi$, since $M_x \perp H$. Consequently the echo signal intensity (in power units) is $J \sim \sin^2 2\phi \sin^2\phi$. All the experimental data agree well with the developed model of formation of the parametric-echo signal.

Experiment with an analogous sequence of pulses, acting on the electronic spin system in a ferrite, was described in[7]. However, in view of the fact that the power of the second pulse in[7] was larger by several orders of magnitude than the power of the first pulse, the echo-formation mechanism proposed by us could not manifest itself in these experiments.

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[1)] The nuclear spin system is assumed to be inhomogeneously broadened, i.e., consisting of a set of noninteracting oscillators with a frequency spread $|\nu_i - \nu_0| \lesssim \delta\nu$.

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[1] Yu. M. Bun'kov and B.S. Dumesh, Zh. Eksp. Teor. Fiz. **68**, 1161 (1975) [Sov. Phys.-JETP **41**, 576 (1975)].
[2] A.Yu. Yakubovskiĭ, Zh. Eksp. Teor. Fiz. 67, 1539 (1974) [Sov. Phys.-JETP **40**, 766 (1974)].
[3] B.T. Adams, L.W. Hinderks, and P.M. Richards, J. Appl. Phys. **41**, 931 (1970).
[4] M.I. Kurkin and V.V. Nikolaev, Fiz. Metal. Metalloved. **38**, 957 (1974).
[5] V.I. Ozhogin, Zh. Eksp. Teor. Fiz. **58**, 2079 (1970) [Sov. Phys.-JETP **31**, 1121 (1970)].
[6] L.A. Prozorova and A.I. Smirnov, Zh. Eksp. Teor. Fiz. **67**, 1952 (1974) [Sov. Phys.-JETP **40**, 970 (1974)].
[7] V.V. Danilov, V.I. Sudakov, and A.V. Tychinskii, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 520 (1975) [JETP Lett. **15**, 368 (1975)]; Izv. vyssh. uch. zav. Fizika, No. 3, 37 (1973).