2ω echo in a system with dynamic frequency shift (antiferromagnetic $MnCO_3$)

S. A. Govorkov and V. A. Tulin
Institute of Solid State Physics, Academy of Sciences of the USSR

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The response of the electron-nuclear spin system to the second harmonic of the resonant frequency is investigated using the spin-echo technique. The specimen emits the second harmonic both during the action of the resonant pulse and during the time that the echo signal exists. Additional echoes, which exist only at the second-harmonic frequency, are observed. A spin echo is observed from nuclear spin waves with $K \neq 0$ in a sequence with two parametric pulses.

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In magnetically ordered substances with strong hyperfine interaction, there exist coupled oscillations of electronic and nuclear magnetization at frequencies near the frequency of the unperturbed nuclear magnetic resonance (NMR).[1] This resonance is manifested best in antiferromagnets with an easy-surface anisotropy containing Mn^{++} ions.[2] A theoretical analysis of the motion of magnetization with these oscillations shows that longitudinal oscillations of magnetization of the specimen at the doubled frequency (relative to resonance) exist.

We posed the problem of studying this doubling by the spin echo method. The advantage of this method lies in the fact that when observing the echo signal, the signal from the generator does not enter the receiving apparatus (either at the resonant frequency or at its harmonics). The investigations showed that when resonant oscilla-

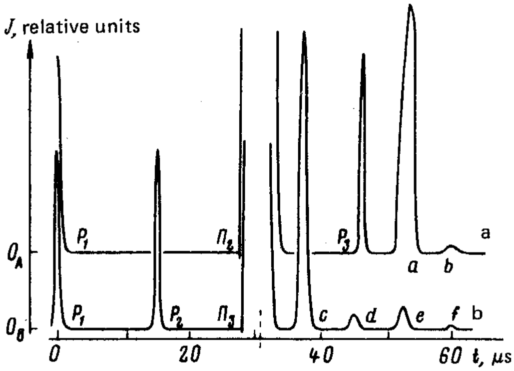

FIG. 1. Oscillograms of echo signals at frequency $2\omega/2\pi = 1160$ MHz. (a) RPR sequence. R_1R_3 are induction signals of the specimen at the time of action of the resonant pulses $\tau_p = 1\ \mu$s; P_2 is the parametric pulse $\tau_n = 2.5\ \mu$s. The dashed line corresponds to the time marking the end of the pulse. a—2ω echo $t = t_2 + t_3/2$; b—second harmonic of parametric echo $t = 2t_2$. (b) RRP sequence. $\tau_p = 1\ \mu$s, $\tau_n = 2.2\ \mu$s; c—l 2ω echo, $t = t_3 + t_2/2$, $t = 2t_3 - t_2/2$; d—f second harmonics of parametric echo $t = 2t_3 - t_2$, $t = 2t_3$, $H = 3$ kOe, $T = 1.2$ K.

tions are excited in the specimen, both during the action of the radio-frequency pulse and during the presence of the spin-echo signal in the system, the specimen emits a second harmonic. This is evident in Figs. 1 and 2. In addition, a series of spin-echo signals, which exist only at the second harmonic and never previously described, was detected. This paper is concerned with the observation of these spin-echo signals and their brief explanation.

For the object of the investigations, we used antiferromagnetic $MnCO_3$, for which the frequency of coupled electron-nuclear oscillations at temperature $T \approx 1$ K is described by the following relation:

$$\omega^2 = \omega_n^2 \left[1 - \frac{H_\Delta^2}{H(H + H_D) + H_\Delta^2} \right] \tag{1}$$

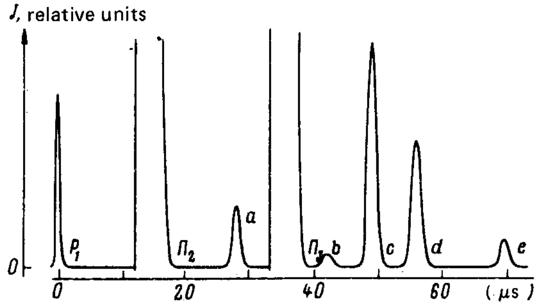

FIG. 2. Oscillogram of echo signals of the sequence RPP at frequency $2\omega = 1160$ MHz; $T = 1.2$ K, $H = 3$ kOe, $\tau_p = 1\ \mu$s, $\tau_n = 1.3\ \mu$s. c,d) 2ω echo $t = t_3 + t_2$, $t = 2t_3 - t_2$. a,b,e) second harmonics of echo $t = 2t_2$, $t = 2t_3 - 2t_2$, $t = 2t_3$.

where ω_n is the nuclear resonance frequency in the absence of the dynamic shift ($\omega_n/2\pi = 640$ MHz), H_Δ^2 is a quantity proportional to the magnetization of the nuclear system ($H_\Delta^2 = 5.8/T$ kOe^2), $H_D = 4.4$ kOe is the effective field of the Dzyaloshinskii interaction, and H is the external constant magnetic field, parallel to the plane of easy magnetization of the crystal. A sequence of radio-frequency (rf) pulses was directed onto the specimen: resonant pulses (R) whose duty cycle corresponds to (1), and parametric pulses (P) at the doubled frequency. The alternating magnetic fields $h_\omega \perp H$, $h_{2\omega} \| H$.

An oscillogram of the echo signals of the sequence RPR is shown in Fig. 1a. The signals at the second harmonic of the induction of the spin system during the action of the R pulses with duration 1 μs (R_1, R_3) can be seen at times $t_1 = 0, t_3$. Observation of the echo at the frequency ω^3 showed that the sequence RPR contains echo signals at times $t_n = t_3 + n(t_3 - 2t_2)$; $n = -2, -1, 0, 1$. The 2ω echo signals with intensities $I_{2\omega} \sim I_\omega^2$ must correspond to these signals of the ω echo [the echo b in Fig. 1a at $t = 2t_2$ is the second harmonic of the parametric echo ($n = 1$)]. Echo a, $t = t_2 + t_3/2$, exists only at frequency 2ω.

We shall analyze the induction signal of the second harmonic after the exciting pulses act on the spin system. We shall use the model in Ref. 3, retaining the same notations. After the first pulse (R_1) at $t = 0$, the system of spins tilts by an angle $\alpha \ll 1$. After the second pulse (P_2), $t = t_2$, the tilt angle of the ith spin will be

$$\alpha + \alpha\beta \sin(2\omega_i t_2 - \phi_2) \cdot \tag{2}$$

After the third pulse (R_3), $t = t_3$, we obtain the tilt angle

$$\theta = \alpha + \alpha\beta \sin(2\omega_i t_2 - \phi_2) + \gamma \sin(\omega_i t_3 - \phi_3), \tag{3}$$

Then the $z(t)$ component of magnetization of the ith spin will be $m_{z_i} = \theta^2 \sin 2\omega_i t$, while the induction signal at frequency 2ω will be

$$M_z(t) \sim \sum_i \theta^2 \sin 2\omega_i t \rightarrow \int_0^\infty n(\omega)\theta^2 \sin 2\omega t \, d\omega, \tag{4}$$

where $n(\omega)$ is the number of spins precessing with frequency ω. The integral in (4) reduces to a sum of integrals of the form

$$A_k \int_0^\infty n(\omega) \cos[2\omega(\tau_k - t - \psi_k)] d\omega \, , \tag{5}$$

It follows that at $t = \tau_k$ we obtain a nonzero signal at frequency 2ω. At $t = 0$ and $t = t_3$, we obtain the signal $\sim\alpha^2$ and γ^2 at the time of action of the resonant pulses (R_1, R_3). The R pulses were short $\tau = 1$ μs and the induction signal at the doubled frequency did not have time to excite the resonator ($\omega_n/2\pi = 1160$ MHz, $Q \approx 1500$, $\tau = 1.3$ μs), which effectively decreased the induction signals of the R pulses.

At the time $t = 2t_2$ (echo b), we obtain the signal $\sim\alpha^2\beta^2$ (second harmonic of parametric echo) and at time $t = t_2 + t_3/2$, we obtain the signal $\sim\alpha\beta\gamma$. For small α, β, γ, the amplitude of the a echo at $t = t_2 + t_3/2$ turned out to be proportional to the amplitudes of all three pulses.

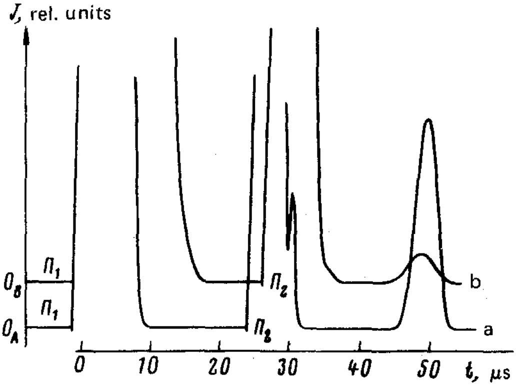

FIG. 3. Oscillograms of echo with parametric excitation of NSW. $\omega_n/2\pi = 1160$ MHz: (a) $k = 0$, $\tau_{n_1} = 4$ μs, $\tau_{n_2} = 1.3$ μs, $H = 3$ kOe, $T = 1.2$ K. (b) $k = 5.6\times 10^5$ cm^{-1}, $\tau_{n_1} = 7$ μs, $\tau_{n_2} = 1.3$ μs, $H = 1.65$ kOe, and $T = 1.2$ K.

The oscillogram of the echo pulses of the sequence (RRP) is shown in Fig. 1b. Echo c at $t = t_3 + t_2/2$ with amplitude $\sim\alpha\beta\gamma$ corresponds to echo a in Fig. 1a of the sequence RPR. Both of these echoes are described by the same general equation $t = t_n + t_p/2$ and transform into each other as the RPR sequence changes into the RRP sequence. The RRP sequence includes one more echo at frequency 2ω (echo e) $t = 2t_3 - t_2/2$ with amplitude $\sim\alpha\beta\gamma^2(\gamma \sim h_n\tau_n)$. It is interesting to note that the angle to which the spins are tilted by the P pulse can be obtained from the ratio of the intensities of the c and e echoes. The echoes d and f at $t = 2t_3$ and $t = 2t_3 - t_2$ with amplitudes $\sim\alpha^2\gamma^2$ and $\sim\beta^2\gamma^2$ are the second harmonics of the parametric echo.[3]

Two echoes at frequency 2ω were observed in the sequence RPP (see Fig. 2): echo c at $t = t_3 + t_2$ with amplitude $\sim\alpha^2\beta\beta_1$ and echo d at $t = 2t_3 - t_2$, $\sim\alpha\beta\beta_1^2$. Echoes a, b, and e at $t = 2t_2, 2t_3 - 2t_2, 2t_3$ with amplitudes $\sim\alpha^2\beta^2, \alpha^2\beta^2\beta_1^2, \alpha^2\beta_1^2$ are the second harmonics of the signals of the ω echo.[3] In addition, echo signals were observed in the sequence RRP at $t = \frac{3}{2}t_2$ and in the sequence RPP at $t = 3t_3 - 2t_2$ ($P \sim 10^{-11}$–10^{-12} W), which are not obtained in the calculation.

The formation of the echo signal only at the second harmonic corresponds to a configuration of spins collected into two bunches and shifted in phase by π (the spins are tilted by an angle α and $-\alpha$ from the equilibrium position). Such a spin configuration will not emit at a frequency $\omega(M_\perp = 0)$.

It was noted that the echo d in the sequence RPP (Fig. 2) remains when the R pulse is switched off. For this reason, the echo in the sequence PP was investigated (Fig. 3).

Before the arrival of the first P pulse, the spin system is in an equilibrium state. The P pulse parametrically excites nuclear spin waves (NSW) with $k \neq 0$. The second pulse forms the echo signal at time $2t_2$ (Fig. 3a). For $H < H_{\rm res}$, where $H_{\rm res}$ is the NMR field at frequency $\omega_n/2$, the R_1 pulse excites a NSW with wave vector $k \neq 0$. The oscillogram of the echo with excitation of NSW with $k = 5.6\times 10^5$ cm^{-1} ($H_{\rm res} = 3$ kOe, $H = 1.65$ kOe) is shown in Fig. 3b.

In our opinion, the most interesting result is the observation of the echo accompanying excitation of nuclear spin waves with $k \neq 0$. Many papers have appeared recently on the investigation of relaxation of spin waves with $k \neq 0$ using the method of parametric excitation of these quasiparticles. In so doing, the numerical value of the relaxation rate is calculated starting from the value of the threshold pumping field, whose absolute magnitude can be determined with a low accuracy. In investigations with the help of the echo method, the magnitude of the lifetime of the excited spin waves is measured directly. This circumstance and the comparatively uncomplicated measurement technique (at least in the case of nuclear spin waves) demonstrate the possibility of the broad application of the described echo method for studying relaxation of spin waves in magnetic substances.

[1] P. G. Gennes, P. Pincus, F. Hartmann-Boutron, and J. M. Wintor, Phys. Rev. **129**, 1105 (1963).
[2] A. S. Borovik-Romanov and V. A. Tulin, Pis'ma Zh. Eksp. Teor. Fiz. **1**, 18 (1965) [JETP Lett. **1**, 134 (1965)].
[3] Yu. M. Bun'kov and S. O. Gladkov, Zh. Eksp. Teor. Fiz. **73**, 2181 (1977) [Sov. Phys. JETP **46**, 1141 (1977)].

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