SINGLE-PULSE SPIN ECHO IN NUCLEAR SYSTEMS WITH LARGE DYNAMIC FREQUENCY SHIFTS

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We consider theoretically the formation of an echo of one pulse in a system with dynamic frequency shift. A single-pulse echo was obtained experimentally from Mn^{55} nuclei in $MnCO_3$. The experimental conditions for echo production and the echo characteristics are given.

Investigations of a two-pulse echo in nuclear spin systems with large dynamic frequency shifts [1 — 3] have shown that echo signals can be produced in such systems, besides by the known mechanism, also by the frequency-modulation mechanism proposed by Gold [4]. We shall show that the Gold mechanism can lead to production of echo signals of not only several pulses but even of a single pulse. We consider here one of the possible situations that could be realized relatively simply in experiments.

The main point is that in systems with dynamic frequency shifts the precession of the nuclear magnetization $\vec{m}$ depends on the value of its projection m_z on the equilibrium direction

$$\Omega = \omega_n - \omega_p \frac{m_z}{m} = \omega_n - \omega_p \cos\theta, \tag{1}$$

where ω_n is the unshifted NMR frequency, ω_p is the dynamic frequency shift, and θ is the angle through which m is deflected from its equilibrium direction under the influence of the high-frequency (hf) field. We consider the case when the nuclear system is excited by an hf field of frequency ω that differs from the NMR frequency by an amount

$$|\delta\omega| = |\omega_n - \omega_p - \omega| \gg \omega_p(\omega_1/\omega_p)^{2/3}, \tag{2}$$

where ω_1/γ is the amplitude of the hf field with allowance for the amplification effect, and γ is the nuclear gyromagnetic ratio. The quantity $\omega_p(\omega_1/\omega_p)^{2/3}$, as shown in [5], corresponds to the average change of the dynamic shift when $\vec{m}$ moves under the influence of the hf field. This change can be neglected in the first order under the condition (2), and then the problem reduces to the motion of free spins in the presence of a large detuning $\delta\omega$ of the hf field. In a coordinate system rotating with frequency ω, this motion represents precession with a frequency equal to $\delta\omega$ in first-order approximation. Then the value of $\cos\theta$ at the instant of termination of a pulse of duration t_1 is given by[1)]

$$1 - \cos\theta = (\omega_1/\delta\omega)^2 (1 - \cos\delta\omega t_1). \tag{3}$$

After the pulse is turned off, the spins precess at a frequency Ω determined by the given angle θ. The precession frequency $\delta\Omega$ relative to the rotating coordinate system is obtained by substituting (3) in (1)

$$\delta\Omega = \Omega - \omega = \delta\omega + \omega_p(\omega_1/\delta\omega)^2(1 - \cos\delta\omega t_1). \tag{4}$$

According to [4], the appearance of a modulating term with $\cos\delta\omega t_1$ is sufficient to cause the decay of the induction signal to take the form of a pulsation. Indeed, let us assume that the detuning $\delta\omega$ has in the interval Δ a scatter that is determined by the inhomogeneous broadening, and let us consider a group of spins with $\delta\omega = \delta\omega_j$. The corresponding nuclear magnetic moment m_j at the time t after the pulse, precessing about the equilibrium direction, will experience a phase change amounting to

$$\phi_j = \phi_j^0 - \delta\phi_j, \tag{5}$$

where

$$\phi_j^0 = t(\delta\omega_j - \omega_p(\omega_1/\delta\omega_j)^2) \approx t\,\delta\omega_j, \tag{6}$$

$$\delta\phi_j = t\omega_p(\omega_1/\delta\omega_j)^2\cos\delta\omega_j t. \tag{7}$$

Let now $t = t_1$, with $t_1\Delta \gg 1$, so that the phases of the initial spin groups become fully intermixed during a time on the order of t_1. Then the additional phase increment for those groups of spins for which $\pi/2 \le \phi_j^0 \le 3\pi/2$ is $\delta\phi_j < 0$, and for those groups of spins with $-\pi/2 \le \phi_j^0 \le \pi/2$ we have $\delta\phi_j > 0$ (Fig. 1). Thus, as follows from Fig. 1, at the instant $t = t_1$ there exists a resultant transverse magnetization that causes the high-frequency signal. At $t \ne t_1$ there is no correlation between ϕ_j and the sign of $\delta\phi_j$, and the signal is equal to zero. We call this phenomenon a single-pulse echo[2)]. We can show analogously that similar signal can arise also at the instants of time $t = 2t_1$, $3t_1$, etc.

The single-pulse echo signal was observed on Mn^{55} nuclei in the easy-plane antiferromagnet $MnCO_3$ (Fig. 2). The signal was observed at helium temperatures (4.2 - 1.7°K). The unshifted NMR frequency was ω_n = 640 MHz. Under our conditions the dynamic shift was $\omega_p \simeq$ 30 MHz.

The echo signal was observed only when the NMR frequency did not coincide with the hf field frequency. The minimal detuning at which the single-pulse echo signal could be observed was 0.5 MHz, whereas the two-pulse echo is maximal in the absence of detuning.

The echo-signal frequency coincides with the NMR frequency. Figure 3 shows the dependence of the echo-signal frequency on the

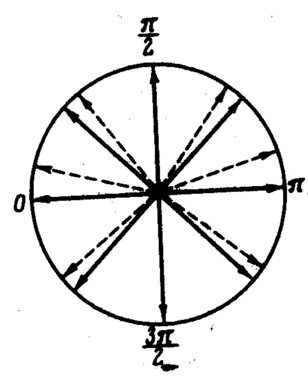

Fig. 1. Phase diagram of the distribution of the transverse moments of the groups of spins at a time $t = t_1$ following an hf pulse. Solid arrows — without allowance for modulation, dashed — with allowance for modulation.

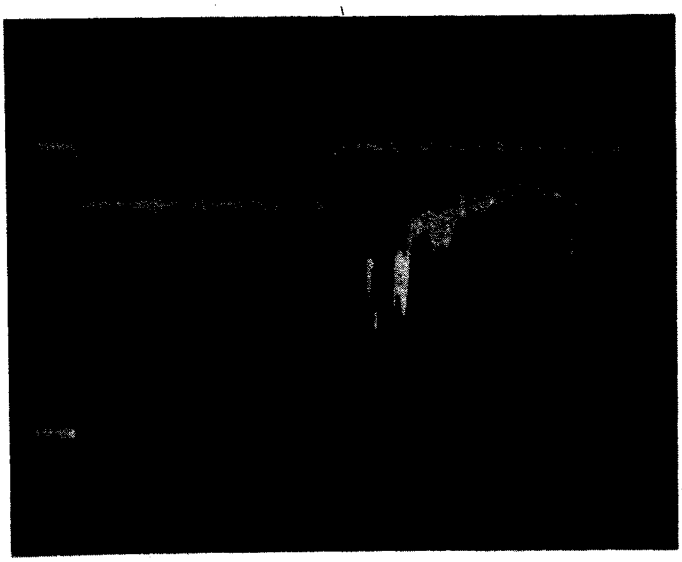

Fig. 2

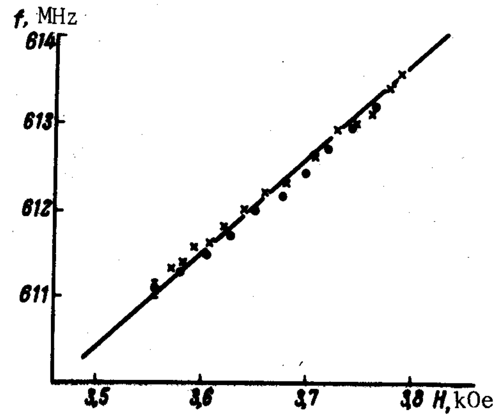

Fig. 3

Fig. 2. Oscillogram of single-pulse echo signal. Horizontal scale — 20 μsec/division. Upper trace — frequency-meter signal, lower trace — induction and echo signal.

Fig. 3. Spectrum of single-pulse echo at constant hf field frequency. Points: ω = 610.4 MHz, crosses — ω = 616 MHz. Solid line — NMR spectrum.

magnetic field. The hf pulse frequency is constant at 610.4 MHz (points) or 616 MHz (crosses). The continuous line represents the NMR spectrum obtained with the same sample from experiments on two-pulse echoes.

We observed experimentally three peaks located at t_1, $2t_1$, and $3t_1$ past the hf pulse, respectively. The first echo peak was observed with t_1 from 30 to 150 μsec. The second and third peaks, which were weaker by one order of magnitude than the first, could be resolved only at small $t_1 \lesssim 30$ μsec.

The echo-signal amplitude depends strongly on the rise time of the pulse front. When the rise time is increased from 0.3 to 1 μsec, the amplitude of the signal decreases by one order of magnitude.

With decreasing pulse duration, the echo-signal decreases exponentially, with a time constant close to the spin-spin relaxation time measured with the aid of a two-pulse echo.

When the detuning is varied, the intensity of the echo signal oscillates and the distance between the maxima is 0.05 MHz. The accompanying change of the echo signal is 20%.

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1) Formula (4) does not take relaxation processes into account.

2) The signal width is determined by the inhomogeneous broadening. A single-pulse echo due to another mechanism has been considered in [6].

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