Nuclear single-pulse echo in ferromagnets

V. I. Tsifrinovich, É. S. Mushailov, N. V. Baksheev, A. M. Bessmertnyĭ, E. A. Glozman, V. K. Mal'tsev, O. V. Novoselov, and A. E. Reĭngardt

L. V. Kirenski Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR, Krasnoyarsk
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A theoretical investigation is made of the response of an inhomogeneously broadened Hahn system to the action of a single resonance pulse. It is shown that the formation of single-pulse echo may be due to distortions of an exciting pulse. Numerous experiments on the ^{59}Co nuclei in cobalt and in FeCo and FeNiCo alloys showed that the single-pulse echo is not the result of internal interactions in a magnetic material. Deliberately induced distortions of a resonance pulse produced a sixfold amplification of the signal.

Much work has been done on the response of inhomogeneous systems to a single resonance pulse. It is well known that in many cases at a time $t = \tau$, where τ is the pulse duration and the time t is measured from the trailing edge, there is a radiation peak which is known in the literature as the edge or single-pulse echo. In the particular case of magnetic materials when the NMR line is subject to a strong inhomogeneous broadening, a nuclear single-pulse echo is observed.[1–4] In an analysis of this echo in magnetic materials it is usual to distinguish two situations: 1) a dynamic shift of the NMR frequency exceeds the resonance line width, in which case the motion of nuclear spins is described by nonlinear differential equations and the single-pulse echo is obtained within the framework of these equations[2]; 2) a dynamic shift of the NMR frequency is not observed and it is assumed that the single-pulse echo can be described using the classical Bloch equations.[1,3] We shall consider only the second situation, which was true of all our experiments.

THEORY

1. Rectangular pulse

Our theoretical task will be to analyze the response of a Hahn system to a single resonance pulse. (The Hahn system is understood to be an inhomogeneous ensemble of radiators, the motion of which is described by the classical Bloch equations.) This problem has been considered on many occasions in connection with spin echo and photon echo experiments (see, for example, Refs. 1 and 5–8).[1)] The Bloch equations in a rotating coordinate system readily yield[5] an expression describing the response of a Hahn system to a rectangular resonance pulse

$$\langle s\rangle = \int_{-\infty}^{\infty} s(\delta, t)\, g(\delta)\, d\delta, \tag{1}$$

$$s(\delta, t) = \frac{q}{\lambda}\left[\frac{\delta}{\lambda}(1-\cos\lambda\tau) + i\sin\lambda\tau\right]\exp(-i\delta t).$$

Here, $s = (M_X + iM_Y)/M$ is the dimensionless transverse magnetization (in the case of electrical systems the quantity corresponding to s is the transverse polarization); δ is the detuning between the resonance field frequency and the frequency of precession of a separate isochromat (an isochromat is a group of spins with the same precession frequency); q is the nutation frequency of a resonance ($\delta = 0$) isochromat (for nuclei in magnetic materials we have $q = \gamma\eta h$, where γ is the nuclear gyromagnetic ratio, η is the gain, and h is the pulse amplitude); $\lambda = (\delta^2 + q^2)^{1/2}$. Relaxation is ignored in the system (1) and it is assumed that the resonance field has a circular polarization and it is oriented along the X axis of a rotating coordinate system. We shall assume that the width of the distribution function $\Delta\omega$ exceeds other characteristic parameters of the problem: $\Delta\omega \gg 2/\tau, q$. In this case, g can be taken outside the integral. (We then obtain an expression known to describe excellently[7–9] the response of a Hahn system everywhere except for a narrow time interval $t \sim 1/\Delta\omega$ near the trailing edge of the pulse.) We shall now consider two possible situations.

1) Case $\theta = q\tau \ll 1$, i.e., when the deviation of isochromats from an equilibrium position is small. In this case the integrand in the system (1) can be expanded as a series in powers of q. To within terms of the order of $\sim q^3$, we have

$$s(\delta, t) = \xi\{(1-\xi^2)\exp(-i\delta t) + [-1 + (3/4 + 1/2\, i\delta\tau)\xi^2] \times\exp[-i\delta(t+\tau)] + 1/4\,\xi^2\exp[-i\delta(t-\tau)]\}, \quad \xi = q/\delta. \tag{2}$$

It might seem that Eq. (2) has a singularity in the limit $\delta \to 0$, but in fact we have

$$s = i(\theta - 1/6\,\theta^3) \text{ when } \delta \to 0. \tag{3}$$

We can easily see that $\langle \mathrm{Re}\, s\rangle \equiv 0$ and it is sufficient to calculate $R = -\langle \mathrm{Im}\, s\rangle$ (the minus sign is selected for convenience of the subsequent treatment). Integrating Im s we obtain a simple expression

$$R = -2g(0)\lim_{z\to 0}\int_z^{\infty} \mathrm{Im}\, s\, d\delta = \begin{cases} (\pi/4)\, g(0)\, q^3 (t-\tau)^2, & t<\tau \\ 0, & t>\tau \end{cases}. \tag{4}$$

Hence, it is clear that the free precession signal observed after a weak rectangular pulse is proportional to q^3 and represents a monotonic decay (of the absolute value) with a characteristic time τ. [As already pointed out, Eq. (4) is valid if $t \ll 1/\Delta\omega$. For low values of t the initial part of the free

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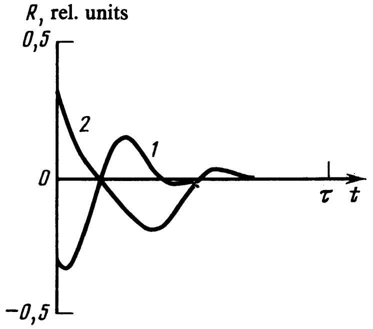

FIG. 1. Oscillations of the free precession signal at $q = \pi\Gamma$ (curve 2) and $q = (5/2)\pi\Gamma$ (curve 1).

precession signal with a characteristic time $1/\Delta\omega$ should be observed. We can demonstrate this simply by integrating Eq. (2) with the Lorentz distribution function.] It should be pointed out that although the expansion (2) contains a term proportional to $\exp[-i\delta(t-\tau)]$, the complete expression for R does not describe the single pulse echo. The absence of the free precession signal at $t > \tau$ represents a special case of the general theorem[10] which is valid for any value of θ and for an arbitrary shape of the pulse.

2) Case $\theta \gtrsim \pi$. This case has been considered in detail theoretically[7] and studied experimentally.[9] An analysis given in Ref. 7 shows that an increase of θ up to $3\pi/2$ hardly alters the profile of the free precession signal, whereas for $\theta \gtrsim 2\pi$ this signal oscillates with a period $T \approx 2\pi/q$; therefore, in the interval $(0,\tau)$ we can expect approximately $\theta/2\pi$ "peaks" of amplitude that decreases monotonically on increase in t.

Our numerical analysis shows that the free precession signal continues to oscillate even when the condition $q \ll \Delta\omega$ is no longer obeyed. By way of example, Fig. 1 shows the dependence $R(t)$ obtained by numerical integration of Eq. (1) with a Gaussian distribution function $(2\pi)^{-1/2}\Gamma^{-1}\exp(-\delta^2/2\Gamma^2)$ and the following values of the parameters: $\tau = 10/\Gamma$, $q = \pi\Gamma$, $\theta = 10\pi$ (curve 1), and $q = (5/2)\pi\Gamma$, $\theta = 25\pi$ (curve 2). We can see that if $q \gtrsim \Delta\omega$, the duration of the free precession signal decreases but the oscillations remain. Finally, as is well known, when the values of q are very high ($q \gg \Delta\omega, \tau\Delta\omega^2/q \ll 2\pi$) only the initial part of the free precession signal is retained and it represents a Fourier transform of the function $g(\delta)$.

It is therefore clear that a Hahn system does not produce a single-pulse echo in response to a rectangular resonance pulse. The opposite statement frequently found in the literature (see, for example, Refs. 5 and 6) is based on a qualitative analysis of the expressions for $s(\delta,t)$ and is not supported by a rigorous calculation of the relevant integrals. True, we can assume[7] that the experimentally observed single-pulse echo is simply the first peak of the oscillations of the free precession signal. However, the first oscillations resemble the single-pulse echo only in a narrow range of amplitudes $2\pi \leq \theta \leq 4\pi$ and at $\theta = 2\pi$ there should be a very smooth single-hump signal with its maximum at $t = 0.8\tau$, whereas at $\theta = 4\pi$ there should be a smooth twohump signal with its zero point at the same value of t (Ref. 7). Clearly, this situation is not characteristic of the majority of the single-pulse echo experiments: as a rule this echo is a sharp peak (with a single hump[11] or with two humps[1]) centered at $t \approx \tau$ and it is observed in a wide range of q.

2. Distorted pulse

In our opinion, the majority of the experiments can be explained in a more natural manner by postulating that a single-pulse echo forms because of the distortion of a resonance pulse. We shall support this hypothesis by an analysis of the simplest model of such a distortion in the form of a rectangular step at the leading edge of a pulse (Fig. 2a): this step has the amplitude $|q_p| \ll q$ and its duration is $t_p \ll \tau$. Solving then in turn the equations of motion, we obtain an

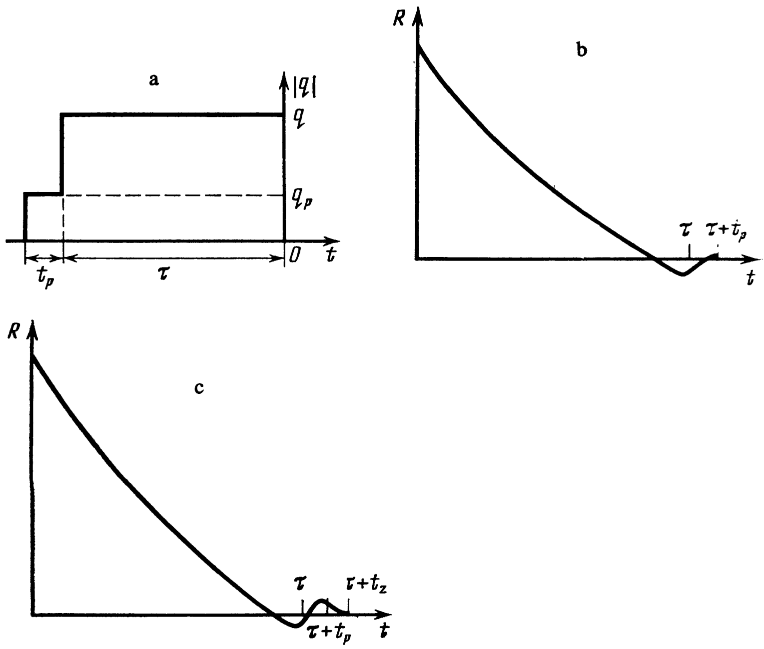

FIG. 2. a) Simplest model of a distorted pulse. b) Dependence $R(t)$ in the case when $q_p < 0$; c) same dependence in the case when $q_p < 0, q_z > 0$ and when the conditions of Eq. (10) are satisfied.

expression for $s(\delta,t)$ which we then can expand in powers of q and q_p. Next, retaining in this expansion the terms proportional to q^3 and $q_p q^2$, we integrate with respect to δ. Omitting cumbersome intermediate stages, we shall give the final expression in the time intervals of interest to us:

$$R = \frac{\pi}{2} q(0) \begin{cases} {}^1/_2 q^3(t-\tau)^2 + q_p q^2[t_p(\tau + {}^1/_2 t_p) - t_p t], & t_p < t < \tau \\ q_p q^2[t_p(\tau + {}^1/_2 t_p) - t_p t + {}^1/_2 (t-\tau)^2], & \tau < t < \tau + t_p \end{cases} \quad (5)$$

and $R = 0$ for $t > \tau + t_p$. The first term in the upper row of the above equation describes the response of the system to an ideal rectangular pulse, whereas terms proportional to $q_p q^2$ are related to the pulse distortion. We can easily demonstrate that if $q_p > 0$, then the function $R(t)$ decreases monotonically to zero, exactly as in the absence of distortion. However, if $q_p < 0$, i.e, if the sinusoidal field of the step is shifted in phase by π relative to the field of the pulse itself, the quantity R has an extremum at

$$t_0 = \tau + t_p q_p / q. \quad (6)$$

We note that $t_0 < \tau$. The origin of this extremum is closely related to the general theorem already mentioned[10]: the signal associated with an ideal rectangular pulse vanishes at $t = \tau$. Therefore, even for small values of $|q_p|(|q_p| \ll q)$ the interval $t \gtrsim \tau$ is dominated by a distortion-induced negative signal which in turn vanishes at $t = \tau + t_p$. Clearly in this situation the function $R(t)$ "must" pass through a minimum. A graph of $R(t)$ calculated for the $q_p < 0$ case is shown schematically in Fig. 2b. A similar situation applies also in the case when the step is located near the trailing edge of a pulse.

We shall consider also a more complex case when there are steps at both the leading and trailing edges. Carrying out consecutively the necessary calculations, we obtain for this case the following correction to Eq. (5):

$$\frac{\pi}{2} g(0) q^2 q_z \begin{cases} 2\left[t_z\left(\tau + \frac{1}{2} t_z\right) - t_z t\right], & t_z < t < \tau \\ (t - \tau - t_z)^2 & \tau < t < \tau + t_z \end{cases} \quad (7)$$

which vanishes at $t > \tau + t_z$. Here, t_z and q_z denote, respectively, the amplitude and the duration of the step at the trailing edge and the time t is still measured from that edge. The derivative $\dot{R}$ vanishes at the point $t = t_a$:

$$t_a = \tau + (q_p/q) t_p + 2(q_z/q) t_z, \quad (8)$$

if $t_a < \tau$; at the point $t = t_b$, we have

$$t_b = \tau + (q_p t_p + 2 q_z t_z)/(q_p + 2 q_z), \quad (9)$$

if $0 < t_b - \tau < \min(t_p, t_z)$.

If $q_p, q_z > 0$, the function $R(t)$ decreases monotonically to zero, whereas for $q_p, q_z < 0$, this function is of the form shown Fig. 2b with an extremum at $t = t_a$. If $q_p < 0$, $q_z < 0$, we can have three situations: 1) $q_p t_p + 2 q_z t_z > 0$, in which case $R(t)$ has no extremum; 2) a situation when the following two conditions are satisfied simultaneously:

$$q_p t_p + 2 q_z t_z < 0, \quad t_p < t_z, \quad (10)$$

i.e., the amplitude of the step at the leading edge $|q_p|$ is sufficiently high in order to ensure that the first inequality is satisfied in spite of $t_p < t_z$; in this case, $R(t)$ has two extrema, one of which is a minimum at the point t_a and the other is a maximum at the point t_b (a schematic diagram of this case is shown in Fig. 2c); 3) in the third situation the first inequality of Eq. (10) is still satisfied, but not the second, so that the function $R(t)$ has only one extremum (minimum) at the point $t = t_a$. A similar result applies also if $q_p > 0, q_z < 0$. Two extrema appear if, in addition to the first inequality of Eq. (10), the second inequality is reversed; $t_z < t_p$. The appearance of two extrema can be understood quite readily from qualitative considerations. If, for example, $q_p < 0$ and $q_z < 0$ and the inequalities of Eq. (10) are satisfied, the signal representing an ideal rectangular pulse proportional to q^3 tends to zero at $t = \tau$, in the interval $\tau \lesssim t < \tau + t_p$ a negative signal originating from a shorter (but a higher) leading-edge step predominates and the corresponding term proportional to $q_p q^2$ vanishes at $t = \tau + t_p$, and finally in the interval $t \gtrsim \tau + t_p$ a positive signal proportional to $q_z q^2$ induced by the leading-edge step predominates and this signal in turn vanishes at $t = \tau + t_z$. It is quite clear that under these conditions the function $R(t)$ has a minimum near the point $t = \tau$ and a maximum near $t = \tau + t_p$.

We also carried out calculations for the case when the amplitudes of the steps q_p and q_z are comparable with q (it is necessary to include all the cubic terms: $q_p^2 q$, $q q_z^2$, $q_p q_z^2$, etc.), and, moreover, we considered a situation when a rectangular edge is replaced by a triangle. However, the resultant solutions produced no qualitatively new results, and, therefore, they are not given here. Thus, even an analysis of the simplest model shows that weak distortions of a resonance pulse may induce a single-pulse echo. The amplitude and profile (one or two humps) of this echo are governed entirely by the nature of the distortion of the real pulse. We recall that our calculations are valid only for small deviations of θ from the equilibrium position; a single-pulse echo then represents simply a weak peak at the end of the free precession signal. However, we are of the opinion that even at large angles θ a distortion of a pulse may induce the observed single-pulse echo.

EXPERIMENTS

1. Free precession signal and single-pulse echo

The first experimental investigations of the single-pulse echo were made by us on single-crystal films of fcc cobalt, grown by epitaxy.[12] The film thickness ranged from 50 to 120 nm and the anisotropy field H_k from 800 to 1300 Oe. Observations were made at room, liquid nitrogen, and liquid helium temperatures. Figure 3 shows characteristic oscillograms obtained employing a conventional spectrometer when the amplitude h of a high-frequency resonance pulse was gradually increased. It is clear from this figure that at low values of h only the free precession signal decreasing to zero in the limit $t \to \tau$ was observed. On increase in h the amplitude of this signal first rose and then gradually fell. Even before the amplitude of the free precession signal reached its maximum, we observed a single-pulse echo at $t = \tau$ (second oscillogram from the top). At still higher values of h the single-pulse echo amplitude first rose and then began to fall. On increase in τ the amplitude of the single-

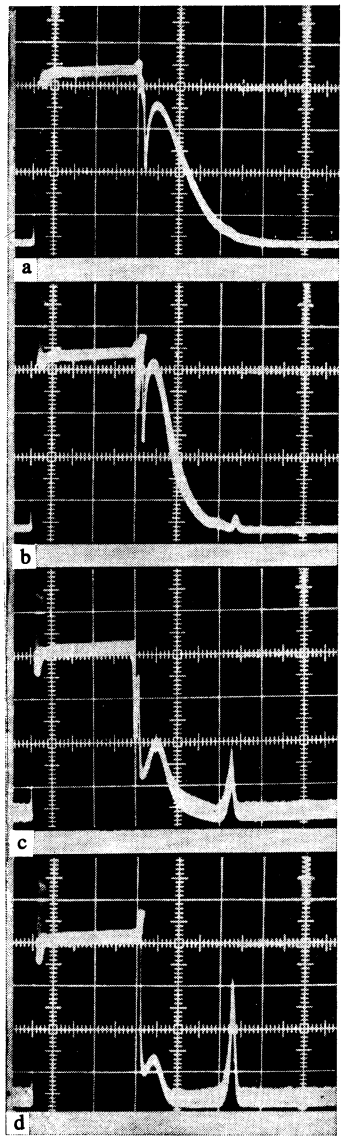

FIG. 3. Oscillograms of the signals generated in a single-crystal fcc cobalt film at helium temperature. NMR line width $\Delta\omega/2\pi \approx 1$ MHz, pulse duration $\tau = 47\ \mu$sec, the amplitude of h increases from top to bottom.

pulse echo fell exponentially with a characteristic time approximately equal to $0.7T_2$, where T_2 is the nuclear transverse relaxation time measured by the two-pulse echo method. At liquid helium temperature ($T_2 = 87\ \mu$sec) a clear single-pulse echo signal was observed for τ from 0.6 to 120 μsec.

The universality of the observed effects was checked by carrying out similar experiments on polycrystalline films, a thin foil, and a polycrystalline cobalt powder (the signal generated by the powder was due to nuclei in domain walls). Moreover, investigations were made on the ^{59}Co nuclei in FeCo alloys. Samples were prepared by diffusion from thin cobalt films into a single-crystal iron base in which the concentration of the cobalt nuclei reached 0.2–3%. The NMR line width $\Delta\omega/2\pi$ ranged from 0.8 to 80 MHz from sample to sample (the inequality $q \ll \Delta\omega$ was always obeyed) and the time T_2 ranged from 20 to 1000 μsec; however, the principal features observed for fcc cobalt were exhibited by all samples without any exception: the maximum amplitude of the single-pulse echo was always approximately 10% of the maximum amplitude of the two-pulse echo. Hence, the formation of a single-pulse echo was not related to the characteristic features of the structure or properties of any specific sample.

2. Amplification of single-pulse echo

The problem of the nature of the single-pulse echo was analyzed by us deliberately using the experimental results on polycrystalline uniaxial $Fe_9Ni_{21}Co_{70}$ films (thickness 200 nm, NMR frequency $\omega_n^0/2\pi = 208$ MHz, $H_k = 25$ Oe, $\Delta\omega/2\pi = 10$ MHz).

The experiments were carried out at $T = 77$ K using a coherent NMR spectrometer. Continuous oscillations were generated by a G4-119A oscillator; a resonance pulse was selected by an electronic switch from a continuous sinusoid and was applied to a circuit with a sample.

It is clear from the preceding section that the formation of the single-pulse echo cannot be due to internal interactions in the sample.

The dynamic hyperfine interaction in ferromagnetic films can change within wide limits when the NMR is combined with the FMR (Ref. 13). We carried out an additional direct experiment proving that the formation of the single-pulse echo was not related to the dynamic hyperfine interaction. The NMR and FMR frequencies were made to coincide by the application of a static field H ($H > H_k$) at right-angles to the anisotropy axis. As the NMR and FMR frequencies approached ($H \to H_k$), the amplitudes of the single-pulse and double-pulse echos rose considerably because of an increase in the gain. However, the ratio of the amplitudes of the single-pulse (A_0) and double-pulse (A_d) echos remained unchanged.

The formation of the single-pulse echo could not be related either to the electrodynamic interaction of a sample with the intrinsic field of the radiation, since the ratio A_0/A_d remained constant when the space factor of the measuring coil was altered. We determined whether the single-pulse echo could be due to a deviation of the frequency of the hf pulse ω from the central NMR frequency ω_n^0 by investigating the dependence of the amplitude A_0 on ω (in this case, as in

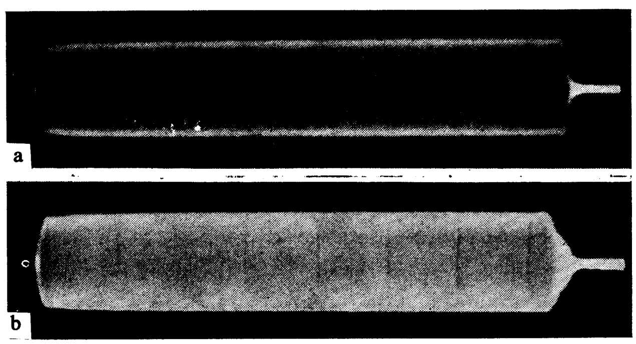

FIG. 4. Oscillogram of a resonance pulse ($\omega/2\pi = 208$ MHz) recorded directly from a measuring circuit using an S1-75 oscilloscope (pass band 250 MHz); b) same oscillogram on application of a control voltage $V(t)$ to a variable capacitor.

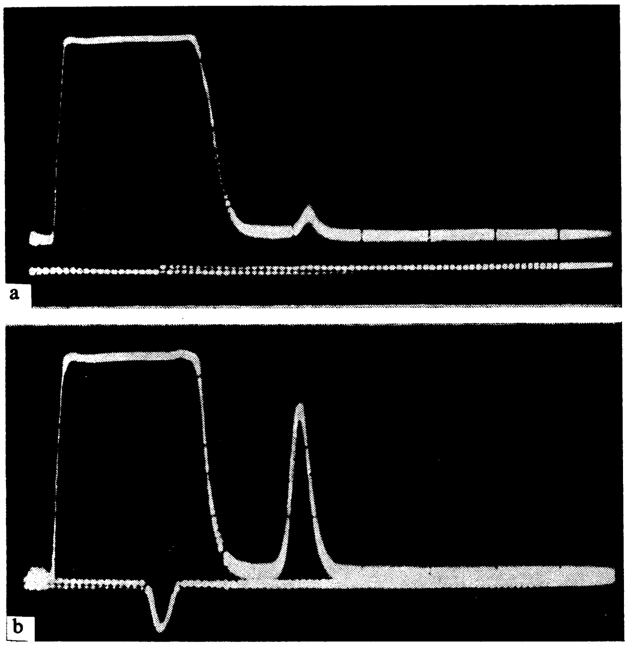

FIG. 5. Oscillogram of a single-pulse echo: a) before introduction of additional distortions; b) after application of a control voltage $V(t)$. The lower trace shows the voltage across the variable capacitor.

the usual NMR measurements, the detector frequency was tuned to the oscillator frequency ω). It was found that the dependence $A_0(\omega)$ repeated completely the NMR spectrum recorded by the double-pulse echo method. The maximum single-pulse echo was observed at $\omega = \omega_n^0$. When the oscillator frequency was detuned from the detector frequency, the amplitude A_0 fell monotonically. All this was evidence of the resonance nature of the excitation of the single-pulse echo.

In order to demonstrate experimentally that the observed single-pulse echo was associated with the effects of the distortions, which are shown for a real rf pulse in Fig. 4a, it was sufficient to prove that the echo depended strongly on the nature of these distortions. The simplest way of controlling the distortions was to change the slopes of the leading and trailing edges of a pulse. This was done by introducing an RC circuit into the power supply of an electronic switch and this altered the slope of the leading edge of the modulation pulses. In the experiment the slope of the leading edge of a resonance pulse decreased by a factor of 3 (while there were practically no distortions to the trailing edge because these were governed by the characteristics of the circuit). The amplitude A_0 also decreased monotonically by a factor of about 3. Weakening of the single-pulse echo signal on reduction of the slope of the leading edge showed that such distortions caused deterioration in the conditions favoring the formation of the echo signal.

On approach to the optimal conditions for the excitation of the single-pulse echo it is obviously possible to use a different method of introducing distortions. In our experiment we employed the following method. A variable capacitor was introduced into the oscillatory circuit of an hf oscillator. This made it possible to control the oscillation frequency by an external voltage $V(t)$. The strongest effect was observed when a $V(t)$ pulse was applied at a time $t \approx 0$, i.e., at the trailing edge of the exciting pulse. The interaction with the oscillatory circuit resulted in the application of an hf pulse with a distorted trailing edge to a sample (Fig. 4b). We were able to select a $V(t)$ pulse of such amplitude and shape that the value of A_0 increased sixfold (Fig. 5). The amplitude of this single-pulse echo amounted to 60% of the maximum amplitude of the double-pulse echo.

Therefore, both a theoretical analysis and our experiments indicated that the formation of a single-pulse echo was associated with distortions of a resonance pulse. Clearly, the amplification of the single-pulse echo could find practical applications.

In our opinion, the multiple echo signals observed on interaction of two extended resonance pulses[14,15] also formed because of the distortions of the pulses near the edges. Introducing these distortions deliberately, as described above, we were able to enhance greatly (by a factor of 5-6) any of the multiple echo signals.

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