Nuclear inverse echo in ferromagnets

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The recently discovered [V. A. Ignatchenko *et al.*, JETP Lett. **37**, 520 (1983)] inverse echo that arises when the magnetization of a ferromagnet is rapidly reversed is investigated in detail. It is shown theoretically that inverse echos of most types have a "time effect": the phase of the high-frequency filling of the echo depends substantially on the time at which the magnetization is reversed (the phase changes by π when the instant of magnetic reversal is changed by a quarter of the nuclear precession period). New inverse echos are found whose amplitudes depend in qualitatively different manners on the magnetic reversal rate.

The first experimental observation of inverse nuclear echos signals in ferromagnets has been recently reported.[1] In those experiments the first high frequency resonance pulse deflects the nuclear spins from the equilibrium position, as usual, and together with the second high frequency pulse, it gives rise to the magnetization reversing video pulse, which rotates the electron magnetization, and accordingly, the hyperfine field (HFF) at the nuclei, through 180°. The idea of the dephased spins being focused as a result of the change in the direction of precession is fairly obvious and has been already noted in the literature,[2] but no theoretical and experimental study of this effect has been made. The purpose of the present work is to provide such a study.

I. THEORY

1. The time effect

The principle feature of the inverse echo can be most simply understood by considering the instantaneous reversal of the HFF. We shall use the following dimensionless components of the nuclear magnetization:

$$s=(\mu_x+i\mu_y)/\mu, \quad m=\mu_z/\mu. \tag{1}$$

We introduce the alternating magnetic reference field $h_x = h\cos\omega t$ from which the first high frequency pulse is "cut out." For convenience, we shall write all the formulas in the laboratory coordinate system.

After the first pulse, the transverse component of the nuclear isochromat is described by the expression

$$s=\sin\alpha_1 \exp[-i(\omega t+\Phi)], \quad \Phi=\delta(t-t_1'')-\pi/2. \tag{2}$$

Here and in what follows, t_j' and t_j'' are the times at which the jth high frequency pulse is turned on and off, α_j is the angle through which the nuclear magnetization $\boldsymbol{\mu}$ is rotated by the action of the jth pulse, Φ is the phase shift of μ_x with respect to the reference cosine curve h_x, and $\delta = \omega_n - \omega$, where ω_n is the precession frequency of the nuclear isochromat. For simplicity, the relaxation terms have been omitted from the formulas and is assumed that the field at the nucleus is directed along the Z axis in the initial state.

We shall assume that the reversal of the HFF begins at the time t_p' and ends at the time t_p'', the time required for the reversal being $\tau_p = t_p'' - t_p'$ (in this section $t_p'' = t_p'$). At the time t_p' the HFF rotates through 180° while the quantities s and m remain unchanged. When $t>t_p'$ the direction of precession of the nuclear spins is reversed, and accordingly

$$s=\sin\alpha_1 \exp[i(\omega t+\Phi)],$$
$$\Phi=\delta(t-t_p'-\tau_1)+\pi/2-2\omega t_p', \quad \tau_1=t_p'-t_1''. \tag{3}$$

In what follows we shall call the part of the above expression for Φ that is proportional to δ, and δ phase, and shall denote it by Ψ_δ; the remaining part of the expression we shall denote by Ψ_E. It is evident that the δ phase determines the time when the echo appears (more accurately, when it reaches its maximum), while Ψ_E is the phase of the echo, by which we mean the phase difference between the high frequency filling of the echo and the reference cosine curve. Of course the inverse echo is formed at the time $t = t_p' + \tau_1$.

We note a basic difference between the mechanisms responsible for the formation of the inverse echo and the Hahn echo, respectively. In the usual Hahn method the focusing pulse changes the phase of the isochromat while retaining the original direction of the precession, whereas here, on the other hand, the direction of the precession changes at the time $t = t_p'$ while the phase of the spins remains unchanged.

It is evident from Eq. (3) that there is a strong "time effect" in the inverse echo: the phase of the echo depends substantially on the time t_p' at which the HFF is reversed. When t_p' is changed by a quarter of the nuclear precession period ($\Delta t_p' = T/4 = \pi/2\omega$) the phase of the echo shifts by π.

The time effect can be easily understood by considering the projection of the rotating vector $\boldsymbol{\mu}_\perp$ onto the X axis without allowing for the dephasing of the spins. The direction of the precession is reversed at the time t_p' of the inversion, so the function $\mu_x(t)$ is symmetric about the point $t = t_p'$. The phase of μ_x (with respect to the phase of the reference cosine curve) at times $t > t_p'$ depends on the value of μ_x at $t = t_p'$. It is obvious, for example, that if μ_x has an extreme value ($\mu_x = \pm\mu_\perp$) at the time t_p' of the inversion, then at later times $t > t_p'$ the phase of μ_x will remain unchanged, but if $\mu_x = 0$ at t_p', the phase of μ_x after the inversion will be shifted by π.

As a result of the time effect, the amplitude of the inverse echo depends substantially on the simultaneity of the magnetization reversal process, i.e., on whether or not the magnetizations of the various parts of the specimen are reversed simultaneously on a time scale of the order of $T/4$. Let us consider, for example, a situation in which the mag-

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netic reversal takes place by the shifting of domain walls. Even if the time Δt for complete magnetic reversal of the specimen is small as compared with the duration of the echo but still $\Delta t \gtrsim T/4$, the echo signals from different parts of the specimen, even though they were formed during the same time interval Δt, will have substantially different phases and will quench one another.

2. Magnetic reversal at a finite rate

Now let us consider how the inverse echo is modified when the finite rate of the magnetic reversal is taken into account. We shall assume that as long as $t_p' < t < t_p''$, the HFF will rotate in the XZ plane with the constant angular velocity $\Omega = \pi/\tau_p$; this corresponds to the simplest model of uniform rotation of the electron magnetization. (A similar model has been used to describe the maser effect in the nuclear magnetic system of a ferromagnet.[3]) On successively solving the equations of motion of the nuclear magnetization we obtain the following expressions for the nuclear isochromat at times $t > t_p''$:

$$s(t) = A \sin\alpha_1 \exp[i(\omega t + \Phi_A)] + B \sin\alpha_1 \exp[i(\omega t + \Phi_B)] + C \cos\alpha_1 \exp[i(\omega t + \Phi_C)], \tag{4}$$
$$m(t) = R\cos\alpha_1 + G\sin\alpha_1[\exp(i\Phi_G) + \exp(-i\Phi_G)].$$

where

$$A = \left[\frac{\Omega}{\rho}\sin\frac{\rho\tau_p}{2}\right]^2, \quad \rho = (\omega_n^2 + \Omega^2)^{1/2},$$

$$B = |\tilde{B}|, \quad \tilde{B} = -\cos\rho\tau_p - \frac{\Omega^2}{\rho^2}\sin^2\frac{\rho\tau_p}{2} - i\frac{\omega}{\rho}\sin\rho\tau_p,$$

$$C = |\tilde{C}|, \quad \tilde{C} = \frac{\Omega}{\rho}\sin\rho\tau_p + i\frac{2\omega\Omega}{\rho^2}\sin^2\frac{\rho\tau_p}{2},$$

$$G = \frac{C}{2}, \quad R = -\frac{1}{\rho^2}[\omega^2 + \Omega^2\cos\rho\tau_p], \tag{5}$$

$$\Phi_A = \pi/2 + \delta(t - t_p'' - \tau_1) - 2\omega t_0, \quad t_0 = (t_p' + t_p'')/2,$$

$$\Phi_B = \pi/2 + \delta(t - t_p'' + \tau_1) - \omega t_p + \operatorname{Arg}\tilde{B},$$

$$\Phi_C = \delta(t - t_p'') - \omega t_p'' + \operatorname{Arg}\tilde{C}, \quad \Phi_G = -\pi/2 - \delta\tau_1 - \omega t_p' - \operatorname{Arg}\tilde{C}.$$

The first term in (4), which is proportional to A, describes the inverse echo. The dependence of A on the parameter $l = 2\tau_p/T = \omega/\Omega$ is shown graphically in Fig. 1, a. The figure shows that as l increases from 0 to $\sqrt{3}$, the function $A(l)$ falls monotonically from unity to zero, reaching the value $A = 0.5$ at $l = l_0 \approx 0.8$. Generally speaking, the value of A depends on δ, but when the conditions

$$\langle|\delta|\rangle \ll \omega_n, \quad l \ll \frac{\rho}{\omega}\frac{\omega}{\langle|\delta|\rangle} \tag{6}$$

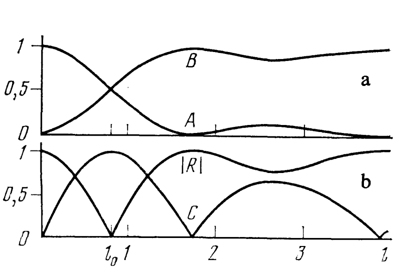

Fig. 1. The l dependences of A, B, C, and $|R|$.

are satisfied that dependence may be neglected (the same can be said about the quantities $\tilde{B}$, $\tilde{C}$, and R). In what follows we shall assume that both conditions (6) are satisfied.

The amplitude A_E and phase Ψ_E of the inverse echo are given by the expressions

$$A_E = A\sin\alpha_1, \quad \Psi_E = \pi/2 - 2\omega t_0. \tag{7}$$

How the time effect is modified when the finite rate of the magnetic reversal is taken into account is evident from the formula for Ψ_E: the phase of the echo is now determined by the averaged "instant" of the magnetic reversal.

3. Inverse echos of other types

It is evident from Eqs. (4) and (5) that if the transverse component s has the δ phase $-\delta\tau_1$ at the time $t = t_p'$, then after the magnetic reversal of the specimen ($t = t_p''$) it will have a component proportional to A with the original δ phase $-\delta\tau_1$, a component proportional to B with the opposite δ phase $+\delta\tau_1$, and in addition, it will have an admixture proportional to C of the longitudinal component, which is independent of δ. For convenience we shall denote all these components by formulas[4] that make it possible to trace their origin. For the first, second, and third components, respectively, we have

$$1U{-}2_p, \quad 1U{-}2_pF, \quad 2_pU. \tag{8}$$

The symbol U denotes the "mixing" of the longitudinal component with the transverse component by the action of the corresponding pulse, and the symbol F indicates the change in the sign of the δ phase of the transverse component.

Thus, the formula $1U{-}2_p$, for example, means that the first high frequency pulse "produced" a transverse component from the constant (i.e., independent of δ) longitudinal component, and later, on the interval τ_1, this component "collects" the phase $-\delta\tau_1$; the second, magnetic reversing, pulse did not change the δ phase (but of course it changed the direction of the precession), so after the 2_p pulse the δ phase is given by the expression $-\delta\tau_1 + \delta(t - t_p'')$, which describes the inverse echo. Further, we introduce the symbols D_+ and D_- to denote the mixing of the transverse component with the longitudinal one. (Since the component m is real, in the case of such mixing there appears not only a component having the initial δ phase, but also a component having the opposite δ phase, i.e., the component m gets modulated.) In this notation, after the magnetic reversal the component m will not only have the term proportional to R, which is independent of δ, but will also have the terms $1U{-}2_pD_+$ and $1U{-}2_pD_-$, which are proportional to G.

Of the six terms in Eqs. (4), only one describes the echo signal. The basic idea of this section is to use an additional high frequency pulse to "refocus" the components proportional to B, C, and G. In this manner, on solving the equations of motion we obtain inverse echos of new types; these differ substantially in their characteristics from the previously discussed $1U{-}2_p$ signal, which we shall denote by E_1. (By inverse echos we now mean signals whose formation is associated with the magnetic reversal of the specimen.)

We shall denote the time interval between the third high frequency pulse and the 2_p pulse by τ_2 ($\tau_2 = t_3' - t_p''$) and

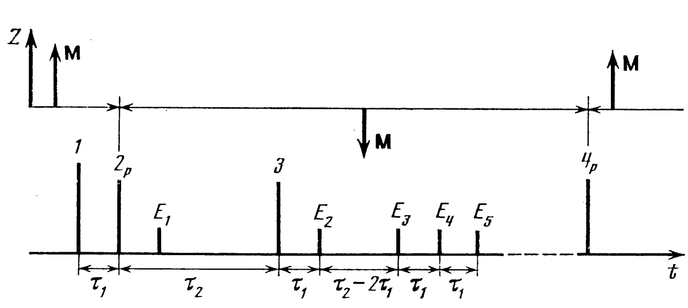

Fig. 2. Time sequence of pulses and signals: 1 and 3—high frequency pulses, 2_p—pulse initiating rapid magnetic reversal, 4_p—auxiliary magnetization-reversing pulse, E_1–E_5—echo signals.

shall assume for definiteness that $\tau_2 > \tau_1$ (see Fig. 2).

Now let us write the formulas and the corresponding expressions for the new inverse echos.

1) The echo E_3. This signal is proportional to $A(l)$ and its formula is $1U$–2_p–$3F$. The first high frequency pulse produces a transverse component that collects the δ phase $-\delta\tau_1$ on the interval τ_1. The δ phase does not change on magnetic reversal. The δ phase $+\delta\tau_2$ is gathered on the interval τ_2. The third high frequency pulse changes the sign of the δ phase, so at $t = t_3''$ we have $\Psi_\delta = \delta(\tau_1 - \tau_2)$. The phase advance continues after the third pulse has ended: $\Psi_\delta = \delta(t - t_3'' + \tau_1 - \tau_2)$. The expression obtained describes the formation of the echo at $t = t_3'' + \tau_2 - \tau_1$. Its amplitude A_E and phase Ψ_E are given by the formulas

$$A_E = A \sin\alpha_1 \sin^2(\alpha_3/2), \qquad \Psi_E = -\pi/2 + 2\omega t_0 + 2\varphi_3, \tag{9}$$

where φ_3 is the phase of the high frequency pulse 3 (see Fig. 2).

It is evident that the l dependence of the amplitude of E_3 accurately repeats the corresponding dependence for the previously discussed E_1 signal.

2) The echo E_5. This signal is proportional to $B(l)$ and has the formula $1U$–2_pF–$3F$. On the basis of similar considerations, it is not difficult to reach the conclusion that the E_5 signal is formed at $t = t_3'' + \tau_1 + \tau_2$ and that the expressions for its amplitude and phase are

$$A_E = B \sin\alpha_1 \sin^2(\alpha_3/2), \qquad \Psi_E = \pi/2 + \omega\tau_p - \operatorname{Arg}\tilde{B} + 2\varphi_3. \tag{10}$$

Figure 1, a shows the l dependence of B. It is evident that the graph of the function $B(l)$ differs substantially from that of $A(l)$. The value of B increases from zero to unity as l increases from 0 to $\sqrt{3}$, and it reaches the value 0.5 at $l = l_0$.

The E_5 signal differs from all the other inverse echos in that it does not have a time effect. As is evident from (10), the phase Ψ_E depends only on $\tau_p = 2\pi/\Omega$, i.e., on the magnetic reversal rate Ω. It is evident from this that E_5 can be observed even when the magnetic reversal is far from synchronous, e.g., when the magnetic reversal is due to motion of domain walls.

3) The echo E_4. This signal is proportional to $C(l)$, has the formula 2_pU–$3F$, and is formed at the time $t = t_3'' + \tau_2$. Its amplitude and phase have the formulas

$$A_E = C \cos\alpha_1 \sin^2(\alpha_3/2), \qquad \Psi_E = \omega t_p'' - \operatorname{Arg}\tilde{C} + 2\varphi_3. \tag{11}$$

The function $C(l)$ is shown graphically in Fig. 1, b; it reaches its maximum value $C = 1$ at $l = l_0$.

4) The echo E_2. This signal is proportional to G, has the formula $1U$–2_pD_+–$3U$, is an analog of the stimulated echo, is formed at the time $t = t_3'' + \tau_1$, and has the following amplitude and phase:

$$A_E = G \sin\alpha_1 \sin\alpha_3 \qquad \Psi_E = -\omega t_p' - \operatorname{Arg}\tilde{C} + \varphi_3. \tag{12}$$

Since $G = C/2$, the graph of $G(l)$ is similar to that of $C(l)$.

Thus, the additional high frequency pulse forms echo signals having substantially different amplitude and phase characteristics. The dependence of the amplitude of the echo on τ_p is easily understood from qualitative considerations. For short magnetic reversal times ($\tau_p \ll T$) the nuclear spins are unable to "budge." Then the longitudinal and transverse components of μ do not change and the quantities A and R are accordingly close to unity while all the other coefficients are close to zero. When the magnetic reversal is slow ($\tau_p \gg T$) μ is fine-tuned by the varying HFF, so the phase of the transverse component and the sign of the longitudinal one are reversed, and accordingly $B \approx 1$ and $R \approx -1$, while the remaining coefficients are close to zero. Finally, at the intermediate value $\tau_p \approx 0.4T$ the nuclear magnetization μ lags the HFF by $\pi/2$, so the longitudinal component becomes the transverse one and vice versa, while the quantities C and G reach their respective maxima.

II. EXPERIMENT

The inverse echos were investigated at room temperature in polycrystalline cobalt films having the following characteristics: induced anisotropy field $H_k \approx 30$ Oe, coercive force $H_c \approx 10$ Oe, and NMR frequency $\omega_n/2\pi = 218$ MHz with the spectral width $\Delta\omega_n/2\pi \approx 10$ MHz.

1. Apparatus

Figure 3 is a block diagram of the apparatus. The apparatus is based on an NMR pulse spectrometer,[5] which consists of the programmer P, the high-frequency pulse generator G_3, the collecting and amplifying channel A, the oscilloscope O, and the transducer T. The last not only contains the high frequency circuit with the specimen, but also contains two systems, associated with the pulse generators

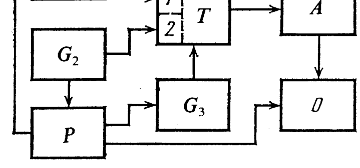

Fig. 3. Block diagram of the experimental setup.

G_1 and G_2, respectively, of which the first ensures rapid magnetic reversal and the second restores the magnetization of the specimen to its initial state. For G_2 we used a standard G5-15 pulse generator, while G_1 is a controlled high voltage discharger connected in series with a peaking device[6] to regulate the leading edge of the magnetization-reversing pulse. The first system (associated with G_1) ensures a working range for the rate of rise $\dot{H}$ of the magnetization reversing field of from 4–6 to 100–150 Oe/nsec. The amplitude of the pulse was 120–200 Oe, while its duration was 0.5–100 nsec. The magnetic reversal of the specimen takes place at the leading edge of the pulse at a rate approximately proportional to $\dot{H}^{1/2}$.[6]

2. Experimental technique

The experiment was conducted in accordance with the scheme illustrated in Fig. 2. In the initial state the specimen is magnetized along the anisotropy axis. The high frequency pulse 1 is applied first. At at a time τ_1 later the magnetization reversing pulse 2_p, which rotates the electron magnetization **M**, and accordingly the HFF, through 180°, is applied. The high frequency pulse 3 is applied at the time τ_2 after the 2_p pulse. After the entire program is finished the auxiliary magnetization reversing pulse 4_p is applied; this pulse returns **M** to its initial state. The program is repeated every 4–5 msec; this time is considerably longer than the longitudinal nuclear relaxation time $T_1 \approx 300\,\mu$sec.

The value of l is determined experimentally, using the maser effect.[3,7] The Hahn echo program is applied to the specimen at a time τ_0 after the rapid magnetic reversal. In the limit $\tau_0 \to 0$, the amplitude of the Hahn echo is proportional to $\mu_z/\mu = R(l)$, and l is determined from that.

We note that the actual conditions of the experiment differ substantially from the simple model discussed in the theoretical section. The two most important differences are as follows: 1) The condition $\omega_N \gg \Delta\omega_n$, where ω_N is the nutation frequency under the action of the high frequency pulse, is not satisfied because of the comparatively broad (10 MHz) NMR line of the investigated specimens. In this case the action of the pulse cannot be described by a single rotation angle α. 2) The videopulse 2_p not only reverses the magnetization of the specimen (the magnetization reversal takes place at the leading edge of the pulse) but also acts within the program for the formation of echo signals as an additional pulse of the magnetic field H_p. One result of such action is a change in the magnetic structure and a corresponding nonuniform shift of the nuclear-spin precession frequency, which substantially weakens the echo signal.[8]

3. Experimental results

In the experiment we reliably observed all five of the echo signals marked in Fig. 2. The amplitudes of the high

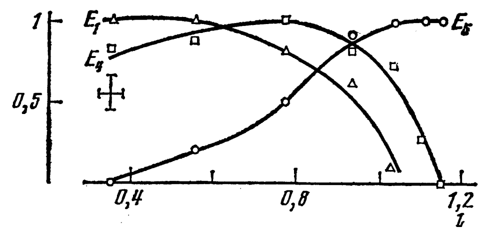

Fig. 4. Echo amplitudes vs l.

frequency pulses were chosen for optimal excitation of all the signals except E_4, whose value decreases monotonically with increasing amplitude of the first high frequency pulse.

Figure 4 shows the l dependence of A_E for the signals E_1, E_4, and E_5 in one of the investigated specimens. Generally speaking, the graphs $A_E(l)$ of the echo amplitudes may differ somewhat from specimen to specimen, but the qualitative trends of the curves are retained.

As was to be expected, the E_1 and E_5 signals have substantially different l dependences. The l dependence of E_3 is virtually the same as that of E_1, the only difference being in the absolute magnitudes of the signals ($E_1 \gg E_3$), so the $E_3(l)$ curve is not given in the figure. The cessation of the growth of E_1 and E_3 at small values of l is evidently associated with the enhancement of the action of the magnetic field H_p, which was discussed in the preceding section.

The l difference of E_2 is also very similar to that of E_4, the only difference being that as l decreases, E_2 appears somewhat earlier than E_4. We note that, in accordance with the theory, the maxima of the E_2 and E_4 signals occur at medium values corresponding to magnetic reversal of the specimen at the rate $\Omega \approx 0.8\,\omega_n$.

Thus, there is good qualitative agreement between the theoretical and experimental results.

In concluding, the authors thank V. A. Ignatchenko for valuable advice.

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