Acoustic excitation of nuclear spin echo

V. A. Kolenishchev-Kutuzov, N. K. Solovarov, and V. F. Tarasov

Kazan' Physico-technical Institute, USSR Academy of Sciences
(Submitted June 22, 1975)
Pis'ma Zh. Eksp. Teor. Fiz. **22**, No. 5, 266–270 (5 September 1975)

We report the first observation of nuclear spin echo excited by a combination of electromagnetic and acoustic pulses. It is established that the echo signal does not depend on the phase of the second pulse.

PACS numbers: 75.80.

The theoretical analysis of the excitation of spin induction and echo by resonant acoustic pulses was initiated by the researches of Koloskova, Kessel', and Kopvillem.[1] Experimental observation of these effects could furnish additional information, compared with electromagnetic excitation, concerning the dynamics of the transient processes in spin systems. This is particularly important for conducting media and superconductors. However, acoustic excitation of echo, owing to the large experimental difficulties, has been realized by now only for the electron spins of Fe^{2+}, Fe^{3+}, Mn^{2+}, and Ni^{2+} in MgO.[2]

We report here the results of the first observation of nuclear spin echo excited by a combination of electromagnetic (**M**) and acoustic (**A**) pulses. To observe the echo it is necessary to satisfy the following conditions: 1) the durations Δt_1 and Δt_2 of the exciting pulses and the distance τ between the pulses must be shorter than the longitudinal and transverse relaxation times T_1 and T_2; 2) the acoustic pulse must turn the magnetic moment through an angle on the order of $\pi/2$. The experiments were performed on the spin system of I^{127} ($I=\frac{5}{2}$) in single-crystal CsI, which has an unusually large spin-phonon coupling constant ($\sim 5\times 10^{15}$ cgs esu). A cylin-

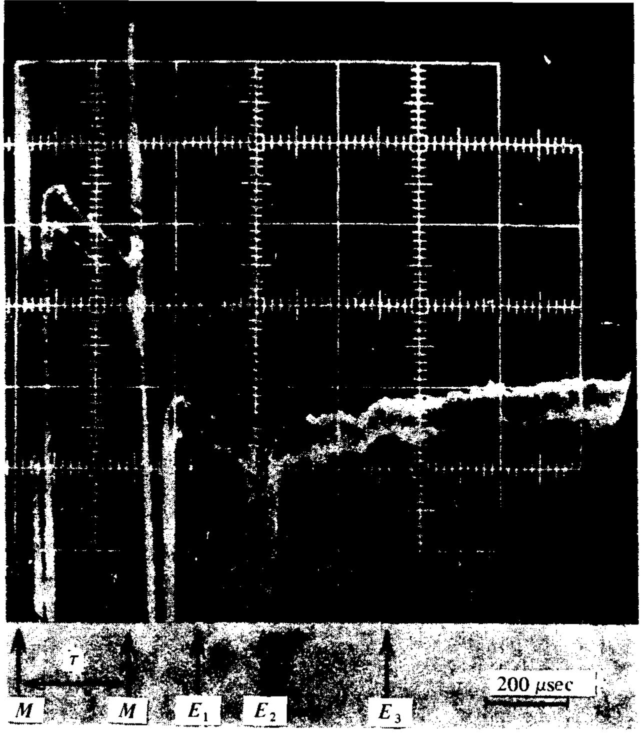

FIG. 1. Echo signals in **MM** sequence: M—electromagnetic sounding pulse, E_1, E_2, E_3—echo signals at the instants $3\tau/2$, 2τ, and 3τ, respectively, where τ is the spacing between exciting pulses.

drical sample of 8 mm diameter and $l = 20$ mm was placed in a constant magnetic field H_0 perpendicular to the cylinder axis ($l \parallel [110]$). To excite any sequence of pulses we used two transmitters, of 50 W power each, at a frequency 10 MHz (transition $\Delta m = \pm 1$) for the **M** pulses and 20 MHz (transition $\Delta m = \pm 2$) for the **A** pulses.

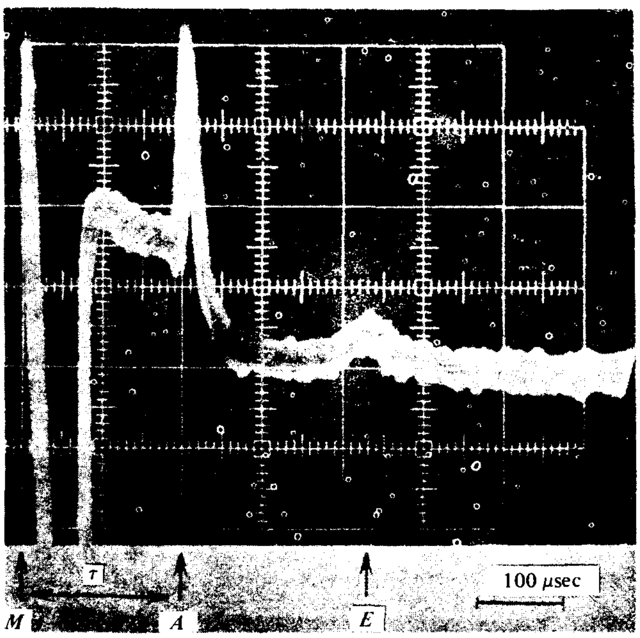

FIG. 2. Echo signal in **MA** sequence: M—electromagnetic exciting pulse, A—acoustic exciting pulse, E—echo signal, τ—spacing between exciting pulses.

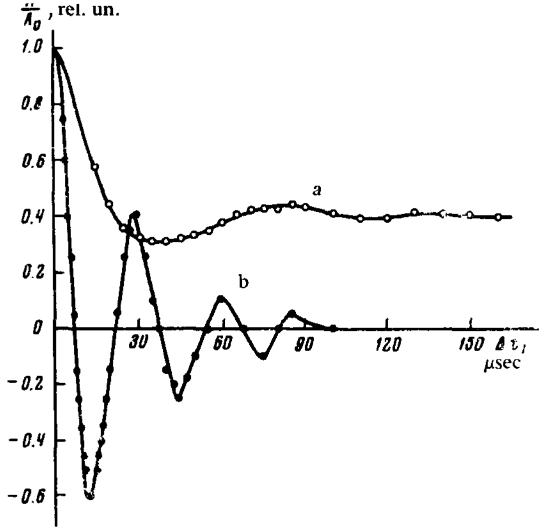

FIG. 3. Dependence of the free-induction signal amplitude, after the second pulse, on the duration of the first pulse: a—AM sequence; b—MM sequence.

The pulses of the longitudinal acoustic waves propagated along the cylinder axis. To prevent formation of standing waves, one end face of the sample was beveled and had a rough structure. In contrast to the experiments of[2], where electromagnetic and acoustic echo signals were observed, we recorded only electromagnetic induction and echo signals. The reason was the smallness of the spin-phonon interaction constant for the nuclear spins in comparison with the electron spins.

The spin system was excited by a sequence of two pulses as follows:

1) **MM** sequence. Three resolved quadrupole echoes were observed at the instants of time $3\tau/2$, 2τ, and 3τ (Fig. 1), of width 40 μsec, and a dipole echo (2τ) of width 500 μsec, in accord with the theoretical concepts.

2) **AA** sequence. No signals were registered.

3) **MA** sequence. An echo signal was observed at the instant of time 2τ (Fig. 2), with a width approximately equal to the width of the quadrupole echo in the first case. The maximum echo signal was observed at $\Delta t_2 \sim 20$ μsec and at a deformation amplitude $\epsilon \sim 4\times10^{-4}$. In the **MM** sequence, however, when the phase of the second pulse was changed by ϕ relative to the phase of the first, the phase of the echo (2τ) changed by 2ϕ, whereas in the **MA** sequence the echo phase was equal to the phase of the free-induction signal after the first pulse, and was independent of the phase of the second pulse. For the **MM** sequence, the time T_2 for the quadrupole echo (2τ) amounts to 3 msec, and for the MA sequence we have $T_2 \sim 200$ sec.

4) **AM** sequence. No echo signals are observed. The amplitude of the free induction signal following the second **M** pulse depends on the first **A** pulse (Fig. 3).

The least expected are the following experimental facts, which do not agree with the prevailing theoretical notions:

1) The anomalously large ratio of the intensities (**I**) of the echo signals in the **MA** and **MM** sequences, $I_{\mathbf{MA}}/I_{\mathbf{MM}} \sim 1$ (when referred to $\tau \to 0$). The theory has predicted $I_{\mathbf{MA}}/I_{\mathbf{MM}} \sim \lambda/2l$, [3] which under our conditions is approximately equal to 10^{-2} (λ is the acoustic wavelength).

2) Independence of the intensity and phase of the spin-echo signal excited by the **MA** sequence of the phase of the second pulse. In all preceding experiments on spin-echo excitation by **MM** sequences there was such a dependence, it was explained theoretically, and led to the need for producing coherent spin-echo relaxometers.

The theoretical explanation proposed by us for the foregoing effect is based the standard theory of echo in systems with discrete spectra, without resorting to any additional assumptions. [4]

The density matrix of the j-th spin after the action of the **MA** sequence of pulses, in the interaction representation, is determined by the expression

$$\hat{\rho}^j(t) = (\hat{L}^j)^{-1}\,\hat{\rho}^j(0)\,\hat{L}^j$$

$$\hat{L}^j = \exp\{i\hbar^{-1}\Delta t_1\hat{\mathcal{H}}^j_M\}\exp\{i\hbar^{-1}\tau\hat{\mathcal{H}}^j_{01}\}\exp\{i\hbar^{-1}\Delta t_2\hat{\mathcal{H}}^j_A\}$$
$$\exp\{i\hbar^{-1}(t-\tau)\hat{\mathcal{H}}^j_{01}\},$$

$$\hat{\mathcal{H}}^j_M = a(\hat{I}^j_+ + \hat{I}^j_-),\quad \hat{\mathcal{H}}^j_A = b[(\hat{I}^j_+)^2\exp\{i(\mathbf{k}\mathbf{r}_j+\phi)\} + (\hat{I}^j_-)^2\exp\{-i(\mathbf{k}\mathbf{r}_j+\phi)\}],$$
$$\hat{\mathcal{H}}^j_{01} = \hbar\Delta\omega^j\hat{I}^j_z + \hbar\Delta\phi^j[(\hat{I}^j_z)^2 - \tfrac{1}{3}I(I+1)],$$

$$\rho(0) = \exp\{-\hat{\mathcal{H}}_0/kT\}/\mathrm{Sp}\exp\{-\hat{\mathcal{H}}_0/kT\},\quad \hat{\mathcal{H}}_0 = -\gamma\hbar H_0\hat{I}_z,$$

where a and b are the interaction constants, ϕ is the initial phase of the second pulse, k is Boltzmann's constant, T is the temperature in °K, $\mathbf{k}$ is the wave vector of the acoustic wave, $\mathbf{r}_j$ is the radius vector of the j-th spin, and $\Delta\omega^j$ and $\Delta\phi^j$ characterize respectively the dipole and quadrupole inhomogeneous broadening of the levels. The observed quantity is the transverse magnetization component M_x and is proportional to $\langle\hat{I}_x(t)\rangle = \Sigma_j\langle\hat{I}^j_x(t)\rangle = \Sigma_j \mathrm{Tr}\rho^I(t)(I_+e^{i\omega t} + I_0e^{-i\omega t})/2$ (expressed in the interaction representation). We are therefore interested only in the $\rho_{i,i+1}(t)$ density-matrix elements. The structure of the matrices $A^{\pm} = \exp\{\pm ik^{-1}\Delta t_2\hat{\mathcal{H}}_A\}$ is such that the matrix elements $\rho_{i,i+1}(t)$ following the action of the second acoustic pulse include terms that do not contain dependences on the phase of the second pulse $(\exp\{+i(\mathbf{k}\cdot\mathbf{r}_j+\phi)\})$. It is precisely the absence of this dependence that explains the two singularities noted above. It is important that such terms lead to the appearance of an echo signal at the instant 2τ only in the quadrupole mechanism of inhomogeneous broadening.

The difference between the values of T_2 in the **MM** and **MA** sequences is apparently due to the existence of different relaxation times for each transition.

The authors thank A. R. Kessel' for useful discussions.

[1]A. R. Kessel', Fiz. Tverd. Tela **2**, 1943 (1960) [Sov. Phys.-Solid State **2**, 1751 (1961)]; N. G. Goloskova and U. Kh. Kopvillem, Fiz. Met. Metallurg. **10**, 818 (1960).
[2]N. S. Shiren, T. G. Kazyaka, Phys. Rev. Lett. **28**, 1304 (1972); D. R. Taylor and I. G. Bartlett, *ibid.* **30**, 96 (1973).
[3]A. R. Kessel'. Yadernyĭ akusticheskiĭ rezonans (Nuclear Acoustic Resonance), Nauka, 1969.
[4]N. K. Solovarov, VINITI Dep. No. 1131-69. V. R. Nagibarov and N. K. Solovarov, Opt. Spektr. **28**, 993 (1970).