# Quadrupole phonon echo of a system of three-level particles in a cubic crystal field

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(Submitted May 28, 1973)
Zh. Eksp. Teor. Fiz. 65, 2297–2308 (December 1973)

We investigate theoretically the generation of spin and phonon echo under the influence of a combination of coherent pulses of an alternating electromagnetic field and traveling hypersonic waves in crystals of cubic symmetry in which the quadrupole spin-phonon interaction reaches a maximum value. The conditions are determined under which the dipole and quadrupole mechanisms of the phase memory of the elastic multipoles lead to the formation of the phonon echo. A method of generating giant acoustic pulses is proposed. The conditions of generation of superradiant acoustic pulses of maximum intensity are determined, with account taken of the concentration of the active particles, the shape of the sample, and the reaction of the coherent spontaneous emission. The theory is compared with experimental measurements of phonon echo from $Ni^{2+}$ and $Fe^{2+}$ in single-crystal MgO. It is shown that the developed phonon-echo theory describes also the echo signals from the electric quadrupole moment of the nuclei and molecules in cubic crystals, and the phonon-echo signals from nuclear spins. The prospects and advantages of this new acoustic method of investigating physical properties of crystals are explained.

## INTRODUCTION

Phonon echo, which was theoretically predicted earlier [1-3], has now been observed experimentally on $Ni^{2+}$ and $Fe^{2+}$ ions in single-crystal MgO [4,5]. In cubic-symmetry crystals (such as MgO) the spin-phonon coupling due to the interaction between the electric quadrupole moment of the ion and the dynamic gradient of the crystal electric field is particularly strong. The reason is that there are no static crystal-field gradients at the paramagnetic ions, owing to the cubic symmetry, and the dynamic gradients come clearly into play. In this case, the spin-phonon interaction operator $\mathscr{H}_{\rm sph}$ is quadratic in the spin variables, and an isotropic equidistant electron-paramagnetic-resonance spectrum is observed for a spin S = 1 in a static magnetic field $\mathbf{H}_0$. We shall call the phonon echo that appears under these conditions quadrupole phonon echo (QPE).

The present paper is devoted to the theory of QPE for S = 1 and to an interpretation of the corresponding experimental results [4,5]. The theoretically obtained formulas can also be used directly to interpret other echo phenomena described by the dynamic $SU_3$ group [6], for example excitation of echo on electric quadrupole moments from radio to $\gamma$-band frequencies. Unlike the existing papers on phonon echo, we consider here in detail, for the first time, the case $S > 1/2$ and the influence of effects of quadrupole phase memory on the phonon-echo intensity. We also take explicit account, for the first time, of the shape and dimensions of the sample and of the radiation reaction following excitation of the crystal. We explain that all these aspects of phonon-echo theory are quite important in the interpretation of the actually performed experiments.

## GENERAL THEORY

The Hamiltonian takes the form

$$\mathscr{H}=\sum_a \mathscr{H}^{(a)},\quad \mathscr{H}^{(a)}=\sum_{j=1}^{N_0}\mathscr{H}_j^{(a)},\quad \mathscr{H}_j^{(1)}=-\hbar\omega_0 S_z^j,$$

$$\mathscr{H}_j^{(2)}=-\hbar\omega_e[\gamma_j S_+^j\exp(-i\omega_0 t)+\gamma_j^* S_-^j\exp(i\omega_0 t)],$$

$$\mathscr{H}_j^{(4)}=i\hbar\omega_1[\{S_z^jS_+^j\}_+-\{S_z^jS_-^j\}_+]\cos(\omega_0 t-\mathbf{k}_{1A}\mathbf{r}_j),\quad \{AB\}_+=AB+BA,$$

$$\mathscr{H}_j^{(3)}=\hbar\omega_2(S_+^{j2}+S_-^{j2})\cos(2\omega_0 t-\mathbf{k}_{2A}\mathbf{r}_j),\quad \mathscr{H}_j^{(5)}=-\hbar\omega_{fj}S_z^j, \tag{1}$$

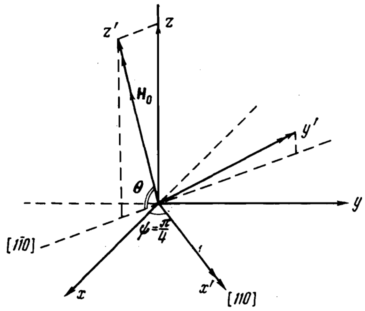
$$\mathscr{H}_j^{(6)}=-\hbar\omega_{qj}S_z^{j2},\quad \mathscr{H}^{(7)}=\sum_{j\neq l}\mathscr{H}_{jl}^{(7)}\quad (j,l=1,\ldots,N_0),$$

where $\mathscr{H}_j^{(1)}$ is the unperturbed Zeeman energy; $\mathscr{H}_j^{(2)}$, $\mathscr{H}_j^{(3)}$, and $\mathscr{H}_j^{(4)}$ are respectively the Hamiltonians of the perturbations due to the alternating magnetic field and to traveling sound waves with cyclic frequencies $2\omega_0$ and $\omega_0$; $\mathscr{H}_j^{(2)}$ and $\mathscr{H}_j^{(4)}$ give rise to transitions with change $\Delta m = \pm 1$ in the magnetic quantum number, while $\mathscr{H}_j^{(3)}$ causes transitions with $\Delta m = \pm 2$; $\mathbf{r}_j$ is the radius vector of particle j, $\mathbf{k}_\alpha$ is the wave vector, $\gamma_j = \exp(-i\mathbf{k}_e \cdot \mathbf{r}_j)$; $\mathscr{H}_j^{(5)}$ and $\mathscr{H}_j^{(6)}$ are respectively the Hamiltonians of the scatter of the local magnetic field and of the electric-field gradients; $\mathscr{H}^{(7)}$ is the operator of two-particle interactions between paramagnetic ions, and gives rise to an irreversible-transverse-relaxation time $T_{2\alpha}$ ($\alpha$ = e, 1A, 2A for transverse components of magnetic dipoles and elastic multipoles of type 1A and 2A); $S_\pm^j$ and $S_z^j$ are the components of the spin operator of particle j; $\hbar\omega_0 = g\beta_0 H_0$, $\hbar\omega_e = g\beta_0 H_1$, g is the g-factor, $\beta_0$ is the Bohr magneton, and $H_1$ is the amplitude of the alternating magnetic field with circular polarization.

To determine $\omega_1$ and $\omega_2$ it is useful to start from the particular experimental geometry shown in the figure. Let the coordinate system x'y'z' be connected with the cubic axes of the crystal. We introduce a coordinate system xyz, in which the cubic vector of the sound ($\mathbf{k}_{1A}$ or $\mathbf{k}_{2A}$) is directed along the axis $\mathbf{x} \parallel [110]$, and the static magnetic field is directed along $\mathbf{z}$. As seen from the figure, the connection between xyz and x'y'z' is determined by a single angle $\theta$. In the system x'y'z', the spin-phonon interaction Hamiltonian is [7]

$$\mathscr{H}_{\rm s\text{-}p}^j=\sum_{\alpha\beta\gamma\delta} S_\alpha^j S_\beta^j G_{\alpha\beta\gamma\delta}e_{\gamma\delta}\quad (\alpha,\beta,\gamma,\delta=x'y'z'), \tag{2}$$

where $G_{\alpha\beta\gamma\delta}$ is the spin-phonon interaction tensor and




The relative positions of the laboratory coordinate system x'y'z' and of the system xyz connected with the cubic axes of the crystal (through an error, xyz and x'y'z' have been interchanged in the drawing).

$\epsilon_{\gamma\delta}$ is the tensor of the relative deformations produced by the acoustic excitation. For cubic-symmetry crystals, the tensor $G_{\alpha\beta\gamma\delta}$ has 12 nonzero components, of which only two are different[8]:

$$G_{22}=G_{33}=G_{11},\ G_{12}=G_{13}=G_{31}=G_{21}=G_{23}=G_{32}=-G_{11}/2,\ G_{55}=G_{66}=G_{44};\quad (3)$$

We use here the Voigt notation: x'x' = 1, etc.

In the xyz system, the crystal deformation tensor due to the longitudinal acoustic wave propagating along the x axis has only the one component $\epsilon_{xx}$. All the calculations that follow will be performed in the xyz system, in which $\mathbf{H}_0 \parallel \mathbf{z}$ and the fourth-rank tensor $G_{pqrs}$ is expressed in terms of $G_{11}$ and $G_{44}$ in accordance with the known transformation rules[9]

$$G_{pqrs}=g_{p\alpha}g_{q\beta}g_{r\gamma}g_{s\delta}G_{\alpha\beta\gamma\delta};\quad (4)$$

summation over the dummy indices $\alpha$, $\beta$, $\gamma$, $\delta$ is implied; $g_{ij}$ is the matrix of the rotation that makes the system xyz congruent with x'y'z , and is expressed in terms of the Euler angles $\pi/2-\theta$ and $\psi = 45°$.

After changing over to the xyz system, the spin-phonon interaction Hamiltonian (2) takes the form

$$\mathcal{H}_{s\text{-}p}={}^1/_4\varepsilon_{xx}\{(S_+{}^2+S_-{}^2)[{}^3/_4G_{11}+G_{44}-\sin^2\theta({}^3/_4G_{11}-G_{44})] +i\sin\theta\cos\theta({}^3/_4G_{11}-G_{44})[\{S_zS_+\}_+-\{S_zS_-\}_+]\},\quad (5)$$

where

$$G_{xxxx}={}^1/_4(G_{11}+4G_{44}),\quad G_{yyxx}={}^1/_4[G_{11}(1-3\cos^2\theta)-4G_{44}\sin^2\theta],$$
$$G_{yzxx}=\sin\theta\cos\theta(G_{44}-{}^3/_4G_{11}),$$

The term proportional to $S_z^2$ is neglected, and the index j has been omitted.

According to (5) we obtain for the frequencies $\omega_1$ and $\omega_2$ from (1), pertaining respectively to quantum transitions with magnetic-quantum number changes $\Delta m = \pm 1$ and $\Delta m = \pm 2$, the expressions

$$\omega_2=\varepsilon_{xx}[{}^3/_4G_{11}+G_{44}-({}^3/_4G_{11}-G_{44})\sin^2\theta]/4\hbar,$$
$$\omega_1=\varepsilon_{xx}\sin\theta\cos\theta({}^3/_4G_{11}-G_{44})/4\hbar.\quad (6)$$

We consider below cases in which the spin system of the crystal is acted upon in succession by two pulses differing in nature, spaced at an interval $\tau$ and of durations $\Delta t_1$ and $\Delta t_2$, described by the Hamiltonians $\mathcal{H}^{(2)}-\mathcal{H}^{(4)}$. By way of response to these perturbations, we investigate the coherent and incoherent acoustic and electromagnetic radiation of the spin system after termination of the action of each of the pulses:

$$I_{\nu w}(t)=I_{\nu w}\exp\left[-\left(\frac{t}{T_{2w}}\right)^2\right]\left\{\exp\left[-\left(\frac{t}{T_{2w}^*}\right)^2\right] +\exp\left[-\left(\frac{t-\tau_w}{T_{2w}^*}\right)^2\right]+\exp\left[-\left(\frac{t-2\tau_w}{T_{2w}^*}\right)^2\right]\right\};\quad (7)$$

here $I_{\nu w}(t)$ is the intensity of a signal of the type $\nu w$ at the instant of time t (reckoned from the instant when the first pulse is applied; $\Delta t_1$ and $\Delta t_2$ are not taken into account); $I_{\nu w}$ is the initial intensity; $T_{2w}$ is equal to the time $T_{2\alpha}$, depending on the type of the signal; $\tau_w$ and $\tau_{2w}$ are certain functions of w and $\tau$; $T_{2w}^*$ is the time of the reversible transverse relaxation for a response of type w, due to perturbations $\mathcal{H}^{(5)}$ and $\mathcal{H}^{(6)}$, while the label w denotes the method of the two-pulse excitation (w = 1, ..., 8).

The intensity of the induction and echo signals in the **k** direction, following a two-pulse excitation of the spin system, is calculated with the formula

$$I_{\nu w}=I_{\nu 0}(\mathbf{k})\left[\sum_{j,l}(\mathrm{Sp}\,\rho_jQ_{\mathbf{k}j}^{(+)}(t))(\mathrm{Sp}\,\rho_lQ_{\mathbf{k}l}^{(-)}(t))\right]_w,$$
$$Q_{\mathbf{k}m}^{(\pm)}(t)=L^+Q_m^{(\pm)}L\exp(\mp i\mathbf{k}\mathbf{r}_m),\quad (8)$$
$$\rho_m=\exp(-\mathcal{H}_0{}^m/k_BT)[\mathrm{Sp}\exp(-\mathcal{H}_0{}^m/k_BT)]^{-1},$$

where the subscript w denotes that all the expressions in this bracket pertain to the case w; $Q_m^{(\pm)}$ is the spin operator of the observed quantity, corresponding to the quadrupole ($\Delta m = \pm 1, \pm 2$) and dipole ($\Delta m = \pm 1$) transitions; $\mathcal{H}_0$ is the unperturbed Hamiltonian of the quantum system and specifies an equidistant Zeeman spectrum $\mathcal{H}_0 = \mathcal{H}^{(1)}$; $k_B$ is the Boltzmann constant; T is the temperature of the spin system in thermal equilibrium with the crystal; L is the operator of evolution of spin particles under the influence of pulsed excitations: $L = L_4L_3L_2L_1$. The operator $L_{1(3)}$, which is the operator for the evolution of a spin system under the influence of perturbations caused by an alternating magnetic field or by a generator of an acoustic wave with circular frequency $\omega_0$ or $2\omega_0$ in the time interval $\Delta t_1$ ($\Delta t_2$), takes the following form:

(a) in the case of an electromagnetic-wave generator

$$L_{1(3)}=\exp(i\omega_0S_z\Delta t_{1(2)})\exp[i\omega_e(\gamma S_++\gamma^*S_-)\Delta t_{1(2)}],\quad (9a)$$

b) in the case of acoustic excitation at frequency $\omega_0$

$$L_{1(3)}=\exp(i\omega_0S_z\Delta t_{1(2)})\exp[\omega_1(\beta\{S_zS_+\}_+-\beta^*\{S_zS_-\}_+)\Delta t_{1(2)}],\quad (9b)$$

c) for an acoustic generator of frequency $2\omega_0$

$$L_{1(3)}=\exp(2i\omega_0S_z\Delta t_{1(2)})\exp[-i\omega_2(\alpha S_+{}^2+\alpha^*S_-{}^2)\Delta t_{1(2)}].\quad (9c)$$

The operator $L_2$ describes the reversible relaxation of the excited spin system due to the scatter of the local magnetic fields $\mathcal{H}^{(5)}$ and the electric-field gradients $\mathcal{H}^{(6)}$ in the interval $\tau$ between pulses, when the spin angular momenta become dephased:

$$L_2(\tau)=\exp\{i[(\omega_0+\omega_f)S_z+\omega_qS_z^2]\tau\}.\quad (9d)$$

The operator $L_4$ corresponds to reversible relaxation, like $L_2$, but in the interval $t-\tau$ (angular-momentum phasing time). $L_4(t-\tau)$ is similar to (9d) with $\tau$ replaced by $t-\tau$.

The intensity $I_{\nu 0}$ per unit solid angle of the phonons emitted spontaneously by an isolated particle, and corresponding to a relative deformation $\epsilon_{\alpha\beta}^{(0)}$ due to zero-point oscillations, is calculated from the formula[10]

$$I_{\nu 0}=\hbar\omega\frac{2\pi}{4\hbar}\sum_{\gamma>\delta}|\langle|S_\gamma S_\delta+S_\delta S_\gamma|\rangle|^2G_{\gamma\delta\alpha\beta}(\varepsilon_{\alpha\beta}^{(0)})^2g(\alpha\beta),\quad (10)$$

where $g(\alpha\beta)$ determines the density of the final phonon states for $\epsilon_{\alpha\beta}^{(0)}$. For the Debye model of the crystal we have

$$g(\alpha\beta)=\frac{3\omega^2V}{2\pi^2v^3},\quad(\varepsilon_{\alpha\beta}^{(0)})^2=\frac{\hbar\omega}{Mv^2}.$$

We then obtain from (10) an expression for the phonon emission in the quadrupole transition $\Delta m = 1$:

$$I_{\nu 0}(\mathbf{k})=\frac{3\omega_0^4}{4\pi\rho v^5}\frac{(\hbar\omega_1)^2}{(\varepsilon_{xx}^{(0)})^2}|\langle|\{S_zS_+\}_+-\{S_zS_-\}_+|\rangle|^2 \quad (\nu=1A) \qquad (11a)$$

and in the case of a transition with $\Delta m = 2$

$$I_{\nu 0}(\mathbf{k})=\frac{3(2\omega_0)^4}{4\pi\rho v^5}\frac{(\hbar\omega_2)^2}{(\varepsilon_{xx}^{(0)})^2}|\langle|S_+^2+S_-^2|\rangle|^2 \quad (\nu=2A) \qquad (11b)$$

In the derivation of the general formulas we disregarded the fact that during the time that the exciting pulse acts on the spin system the latter begins immediately to radiate an acoustic and an electromagnetic response. As a result, the external action increases the angle $\omega_\xi \Delta t_\xi$ ($\xi$ = 1, 2, 3), and the radiation of the system tends to decrease this angle. Consequently, it is far from always possible to attain the value $\psi^{(0)} = \omega_\xi \Delta t_\xi \geq \pi/2$, i.e., it is possible to obtain in the limit a value $\psi \leq \psi^{(0)}$ at which the rate of change of the angle $\omega_\xi \Delta t_\xi$ is equal to the rate of the backward rotation due to the radiation of the system.

The foregoing can be explained with the aid of formulas pertaining to spin S = 1/2, excitation with an electromagnetic field, and radiation of electromagnetic energy. Taking only coherent radiation into account, we obtain

$$\operatorname{tg}\frac{\psi}{2}=\omega_H\left[\omega_{0\psi}-(\omega_H^2-\omega_{0\psi}^2)^{1/2}\operatorname{ctg}\frac{t(\omega_H^2+\omega_{0\psi}^2)^{1/2}}{2}\right]^{-1} \quad (\omega_H^2>\omega_{0\psi}^2), \qquad (12)$$

where $\psi$ is the angle between the magnetic field $\mathbf{H}_0$ and the macroscopic spin angular momentum, $\omega_H$ is the rate of rotation of the angular momentum under the influence of the excitation generator, $\omega_{0\psi}\sin\psi dt$ is the angle of rotation of the spin as a result of the loss of energy in the form of radiation, and $\omega_{0\psi}\sin\psi$ is the rate of change of $\psi$ in this process, while t is the time during which a rotation through an angle $\psi$ is produced by the external perturbation. At $\psi = 0$ there is no superradiance, and when $|\psi|$ increases the superradiance power increases. Since the direction of the rotation due to the superradiance is taken to be positive, the rotation of $\psi$ under the influence of the exciting pulse is negative in sign. At $\omega_{0\psi}^2 = 0$ we obtain the usual formula, where the radiation reaction is disregarded. At $\omega_H^2 \gg \omega_{0\psi}^2$ it follows from (12) that

$$\operatorname{tg}(\psi/2)=[\omega_{0\psi}/\omega_H-\operatorname{ctg}(\omega_H t/2)]^{-1}, \qquad (13)$$

from which we see immediately that the reaction of the radiation leads to a decrease of the maximum possible rotation angle due to an external perturbation of given power.

Another case is possible, in which the system already radiates at $\psi = 0$ at a power exceeding the pump power ($\omega_H^2 < \omega_{0\psi}^2$):

$$\operatorname{tg}\frac{\psi}{2}=\frac{(\omega_{0\psi}^2-\omega_H^2)^{1/2}\operatorname{ch}y+(\omega_{0\psi}-\omega_H)\operatorname{sh}y}{(\omega_{0\psi}^2-\omega_H^2)^{1/2}\operatorname{ch}y-(\omega_{0\psi}-\omega_H)\operatorname{sh}y}, \quad y=\frac{t(\omega_{0\psi}^2-\omega_H^2)}{2}. \qquad (14)$$

In this case, in spite of the pumping, the superradiance intensity decreases and the change of $\psi$ with time is positive. In the limiting case $\omega_H = 0$ we obtain from (14) $\tan(\psi/2) = \exp(\omega_{0\psi}t)$ or

$$\sin\psi=\operatorname{sech}(\omega_{0\psi}t). \qquad (15)$$

The square of $\sin\psi$ determines the spin-system radiation power in the absence of pumping. It is seen from (15) that the radiation power decreases strongly with increasing time, i.e., radiative damping takes place.

Formulas (12)—(14) show that just as giant pulses are generated in optics, acoustic pulses can be shortened and their power increased with the aid of superradiant processes. To this end it is necessary to "spoil" the Q of the acoustic resonator during the time of action of the exciting sound pulse, and to produce a maximal acoustic superradiant state. It is then necessary to introduce in pulsed fashion an acoustic Q that leads to the appearance of a giant acoustic pulse. For example, at an active-particle concentration $N_c = 10^{22}$ cm$^{-3}$, at a constant $G \sim 10^{-12}$ erg/def. unit, and at a sample thickness $l = 10^{-2}$ cm it is possible to obtain an acoustic pulse at a frequency $4 \times 10^{10}$ sec$^{-1}$, power $W \sim 10^{12}$ erg/sec-cm$^2$, and duration $\Delta t = 10^{-8}$ sec. At $l = 10^{-4}$ cm, the limiting values are $\Delta t = 10^{-10}$ and $W \sim 10^8$ erg/sec-cm$^2$.

Using formulas (7), (8), and (10) for the calculations, it is appropriate to present the conditions for the onset of superradiance:

$$L/v\ll T_2^*, T_2, \Gamma^{-1}, \Gamma_{\text{coh}}^{-1}, \quad L/v\ll\omega_1^{-1}, \omega_2^{-1}, \omega_e^{-1}, \qquad (16)$$

where L/v is the time during which all $N_0$ paramagnetic particles take part in the superradiance, L is the length of the active layer of the sample with $N_0$ impurities, v is the velocity of the acoustic wave passing through the sample, $T_2^*$ is the time of reversible phase relaxation due to the inhomogeneous broadening, $\omega_1$, $\omega_2$, and $\omega_e$ determine the rate of rotation of the spin angular momenta under the influence of the acoustic and electromagnetic excitations, $\Gamma$ is the width of the emission line of the isolated atom, $T_2$ is the time of irreversible transverse relaxation, and $\Gamma_{\text{coh}}$ is the coherent-radiation line width.

For an intense superradiance signal to be produced, it is necessary that an appreciable number of active impurities take part in this process, i.e.,

$$N_c\lambda^3\gg 1. \qquad (17)$$

It is then possible to change over in (8) from summation over the particles to integration over the volume. It should be noted that in the case when the condition $L/v \ll T_2^*$ is violated, but the remaining conditions (16) remain in force, the number of active impurities decreases to $N_{\text{eff}} = N_0T_2^*L'/T_2L$, where L is the length of the sample and L′ is the length of the effective layer containing $N_{\text{eff}}$ particles, i.e., in this case it is necessary to substitute $N_{\text{eff}}$ for $N_0$ ($N_{\text{eff}} \ll N_0$) in the formulas for the superradiance intensity.

When conditions (16) and (17) are satisfied, averaging over all the wave-vector directions and integration over the volume yields, taking the diffraction effect into account, the following results[11]:

a) For small Fresnel numbers ($R^2 \gg \lambda L$)

$$I_{\nu w}\sim\lambda/L \qquad (18a)$$

for all particles contained in the sample but not on its end faces;

b) for large Fresnel numbers ($R^2 \ll \lambda L$)

$$I_{\nu w}\sim\lambda^2/R^2. \qquad (18b)$$

Equations (18a) and (18b) were obtained for a cylindrical sample of length L and radius R in which an acoustic wave propagates along the axis.

Let us discuss the feasibility of satisfying conditions (16) and (17). Usually the length of the acoustic wave (see[4,5]) is $\lambda \sim 10^{-5}$ cm (hypersound), i.e., $N_0 \geq 10^{15}$. Under the conditions of the acoustic experiments[4,5] we have $N_0 \sim 10^{18}$—$10^{19}$, i.e., (17) is satisfied. In the case of terasound of frequency $\sim 10^{12}$ Hz, when the length of the sound wave is comparable with the interatomic distance, Eq. (17) can be violated while all the remaining superradiance conditions are fulfilled.

The conditions (16) are well satisfied in solids: at a length L = 0.01 cm and a sound velocity v = $10^6$ cm/sec we have in MgO $L/v \sim 10^{-8}$ sec and the shortest characteristic time is $T_2^* \sim 10^{-7}$ sec. In acoustic experiments[4,5] we usually have $T_2^* < T_2$ and $T_2^* < \Gamma^{-1}, \Gamma_{coh}^{-1}$, while the amplitudes $\omega_1$, $\omega_2$, and $\omega_e$ of the electromagnetic excitations can always be chosen such as to satisfy the inequality (16). The diffraction effects that yield a dependence of the type $L^{-1}\lambda$ play an important role in samples in the form of filamentary crystals. Thus, for hypersound ($\lambda \sim 10^{-5}$ cm) at a crystal length L ~ 0.1 cm the thickness of the filaments should be much less than $10^{-3}$ cm, and in order to satisfy a relation of the type $(\lambda/R)^2$ in (18b) it suffices to use a cylindrical sample with length L ~ 0.1 cm and thickness $2R \gg 10^{-3}$ cm.

Taking all the foregoing into account, we have calculated all the possible variants of combined acoustic and electromagnetic two-pulse excitations and the intensities of coherent responses in the form of spin induction and echo. The results of the calculations are presented below—namely the amplitudes of the coherent responses, the time of appearance of the maximum signal, and the value of its effective wave vector. The following cases are considered: 1) when only the scatter of the local magnetic fields ($\omega_q = 0$) contributes to the mechanism of the reversible phase relaxation, the so-called dipole reversible relaxation (DRR), and 2) when, in addition to the inhomogeneity of the external magnetic field, scatter of the electric-field gradients is also present ($\omega_q \neq 0$; $\omega_f \neq 0$)—the so-called quadrupole reversible relaxation (QRR). The initial intensity $I_{\nu w}$ for a $\nu$-coherent response in the w-method of excitation of the quantum system is equal to the product of the amplitude of the type-$\nu$ response by the intensity (11) of the spontaneous emission and by the quantity $\lambda^2/R^2$ for a sample satisfying the condition (18b).

An important role is played by the instants of time t at which the responses appeared, and by the requirements with respect to the spatial coherence of the responses, requirements described by their effective wave vector. For example, two acoustic pulses of type 1A and 1A, causing $\Delta m = \pm 1$ transitions, produce four electromagnetic responses (ER) ($\nu$ = 1 ER) at the frequency of the sound pulses. At the instant $t = 3\tau$, a signal of amplitude

$$\Phi_c \sin^4 2\omega_1\Delta t_1 \sin^2 2\omega_1\Delta t_2 \sin^4 \omega_1\Delta t_2$$

is annihilated by the QRR mechanism, but in the absence of this mechanism the signal is formed as a result of the DRR. This calls for satisfaction of the equality $\mathbf{k}_{ER} = 3\mathbf{k}_{1A}^{(2)} - 2\mathbf{k}_{1A}^{(1)}$, but inasmuch as $|\mathbf{k}_{ER}| \ll |\mathbf{k}_{1A}^{(2)}|$, $|\mathbf{k}_{1A}^{(1)}|$ this equality cannot be satisfied, and a response is observed in practice only in a plate of half-wave thickness $\lambda_{1A}/2$ or in a "supercrystal" (a layered film structure consisting of alternating active and inactive layers of thickness $\lambda_{1A}/2$). For the same reason it is difficult to observe the remaining three signals at the instants $t = 2\tau, \tau, 0$.

Unlike the electromagnetic responses, the acoustic responses in transitions with $\Delta m = \pm 1$ (1AR) and $\Delta m = \pm 2$ (2AR) are observable, since the spatial-coherence requirements that their wave vectors must satisfy are fulfilled. The acoustic coherent response in a transition with $\Delta m = \pm 1$ (1AR) at the instant $t = 2\tau$ (echo) has the amplitudes

$$4\Phi_s \sin^2 2\omega_1\Delta t_1 \sin^4 \omega_1\Delta t_2, \quad \Phi_s \sin^2 2\omega_1\Delta t_1 \sin^4 2\omega_1\Delta t_2$$

respectively for DRR and in the presence of DDR and QRR; the wave vector is $\mathbf{k}_{1AR} = 2\mathbf{k}_{1A}^{(2)} - \mathbf{k}_{1A}^{(1)}$. At the instant $t = \tau$, the 1 AR signal coincides in direction with the second pulse $\mathbf{k}_{1AR} = \mathbf{k}_{1A}^{(2)}$ and its amplitude is

$$4\Phi_s \sin^2 2\omega_1\Delta t_2 \cos^2 2\omega_1\Delta t_1.$$

At the instant t = 0 we have $\mathbf{k}_{1AR} = \mathbf{k}_{1A}^{(1)}$ and the signal amplitudes are equal to

$$4\Phi_s \sin 2\omega_1\Delta t_1 \cos^4 \omega_1\Delta t_2$$

for DRR and

$$4\Phi_s \sin^2 2\omega_1\Delta t_1 \cos^2 2\omega_1\Delta t_2 \cos^4 \omega_1\Delta t_2$$

for DRR and QRR. The temperature coefficients $\Phi_c$ and $\Phi_s$ are given by

$$\Phi_c = (\mathrm{ch}\,\zeta - 1/2\mathrm{ch}\,\zeta + 1)^2, \quad \Phi_s = (\mathrm{sh}\,\zeta/2\mathrm{ch}\,\zeta + 1)^2, \quad \zeta = \hbar\omega_0/k_B T.$$

The subscripts w = 1A, 2A, and 1E describe the types of exciting pulses: acoustic on transitions with $\Delta m = \pm 1$, $\pm 2$, and electromagnetic on a transition with $\Delta m = 1$ with corresponding wave vectors $\mathbf{k}_{1A}$, $\mathbf{k}_{2A}$, $\mathbf{k}_{1E}$; the subscripts $\nu$ = 1 AR, 2 AR, and 1 ER denote the corresponding responses.

As follows from the calculations, the quadrupole relaxation has no influence on the formation of type-2A responses at the instants of time $t = n\tau$ (n = 0, 1, 2), but it does cancel out the signals in the cases when $t = m\tau/2$ (m = 1, 3). The signals of maximum intensity are produced at the instant $t = 2\tau$ with respective wave vector and amplitude

$$\mathbf{k}_{2AR} = 4\mathbf{k}_{1A}^{(2)} - 2\mathbf{k}_{1A}^{(1)}, \quad \Phi_c \sin^4 2\omega_1\Delta t_1 \sin^8 \omega_1\Delta t_2,$$

at the instant $t = \tau$ with

$$\mathbf{k}_{2AR} = 2\mathbf{k}_{1A}^{(2)}, \quad {}^1/_4\Phi_c \sin^4 2\omega_1\Delta t_2 (1 - 3\cos^2 2\omega_1\Delta t_1)^2$$

and at the instant t = 0 with

$$\mathbf{k}_{2AR} = 2\mathbf{k}_{1A}^{(1)}, \quad \Phi_c \sin^4 2\omega_1\Delta t_1 \cos^8 \omega_1\Delta t_2.$$

When the crystal is excited by two acoustic pulses of type 2A and 2A, only responses of type 2A are possible, with the maximum intensity at the instant $t = 2\tau$, and with wave vector and amplitude

$$\mathbf{k}_{2AR} = 2\mathbf{k}_{2A}^{(2)} - \mathbf{k}_{2A}^{(1)}, \quad 4\Phi_s \sin^2 2\omega_2\Delta t_1 \sin^4 \omega_2\Delta t_2,$$

at the instant $t = \tau$ with

$$\mathbf{k}_{2AR} = 2\mathbf{k}_{2A}^{(2)}, \quad 4\Phi_s \cos^2 2\omega_2\Delta t_1 \sin^2 2\omega_2\Delta t_2,$$

and at the instant t = 0 with

$$\mathbf{k}_{2AR} = \mathbf{k}_{2A}^{(1)}, \quad 4\Phi_s \sin^2 2\omega_2\Delta t_1 \cos^4 \omega_2\Delta t_2.$$

In the case of $(\Delta m = \pm 1) - (\Delta m = \pm 1)$ excitation, the QRR mechanism annihilates the 1 AR and 1 ER signals at the instant $t = 3\tau$, changes the amplitudes of the responses at the instants $t = 2\tau$ and t = 0, and does not affect the two-pulse induction signals at the instant $t = \tau$.

When pulsed excitations of the type $(\Delta m = \pm 1) - (\Delta m = \pm 2)$ or $(\Delta m = \pm 2) - (\Delta m = \pm 1)$ are applied, there is no acoustic or electromagnetic echo on the transition with $\Delta m = \pm 1$, owing to the QRR. In the case of a pulse sequence 1A − 2A and in the presence of only DRR at the instant of time $t = 2\tau$, a 1 ER signal can appear, provided the condition $\mathbf{k}_{ER} = \mathbf{k}_{2A}^{(2)} - \mathbf{k}_{1A}^{(1)}$ is satisfied, and this is possible only if the pulse of type 1A is an acoustic wave with transverse polarization and 2A has longitudinal polarizations (see[12]), inasmuch as the velocity of the longitudinal acoustic wave is double the velocity of the transverse acoustic wave in a number of crystals (InSb and other crystals). For MgO, the ratio of these velocities is $\approx 3/2$, and consequently the condition of spatial coherence for $\mathbf{k}_{ER}$ at the instant $2\tau$ is not satisfied, i.e., there is no signal.

The situation is different for the reversed sequence of the pulses: 2A − 1A. In this case the signal 1 ER is produced at the instant $t = 3\tau$ with wave vector $|\mathbf{k}_{ER}| = |3\mathbf{k}_{1A}^{(2)} - \mathbf{k}_{2A}^{(1)}|$ (i.e., the velocity of the 1A wave should exceed the velocity of the 2A wave by a factor 3/2; to this end it suffices for the 2A wave in MgO to be transverse and for 1A to be longitudinal), and with amplitude

$$4\Phi_s \sin^2 2\omega_2\Delta t_1 \sin^2 2\omega_1\Delta t_2 \sin^4 \omega_1\Delta t_2.$$

In the case of a type 2AR signal and an excitation sequence 1A − 2A we obtain $\mathbf{k}_{2AR} = 2(\mathbf{k}_{2A}^{(2)} - \mathbf{k}_{1A}^{(1)})$ at the instant $t = 2\tau$ (the echo signal coincides in direction with the second pulse), and the signal amplitude is

$$\Phi_c \sin^4 2\omega_1\Delta t_1 \sin^4 \omega_2\Delta t_2.$$

Let us consider further a series of pulses 1A − 1E, which excites from among the electromagnetic responses only two-pulse induction at the instant $t = \tau$ ($\mathbf{k}_{1E} = \mathbf{k}_E^{(2)}$) with amplitude

$$4\Phi_s \sin^2 2\omega_E\Delta t_2 \cos^2 2\omega_1\Delta t_1.$$

Electromagnetic echo and single-pulse induction cannot appear, in view of the failure to satisfy the spatial-coherence condition for the wave vectors of the corresponding responses. The acoustic response 1 AR at the instant $t = 3\tau$ is possible only in the case of the DRR mechanism and in crystals in which the velocities of the longitudinal and transverse waves differ by a factor of 2; the echo signal at the instant $t = 2\tau$, with wave vector $\mathbf{k}_{1AR} = -\mathbf{k}_{1A}^{(1)}$, propagates in the direction opposite to the first acoustic pulse, with amplitude

$$4\Phi_s \sin^2 2\omega_1\Delta t_1 \sin^4 \omega_E\Delta t_2 (1-4\cos^2\omega_E\Delta t_2)^2$$

in the case of DRR, and with amplitude

$$\Phi_s \sin^2 2\omega_1\Delta t_1 \sin^4 2\omega_E\Delta t_2$$

in the presence of the two reversible-relaxation mechanisms DRR and QRR. Analogously, the 2 AR echo signal at the instant $t = 2\tau$ propagates in a direction opposite to the first acoustic pulse, with amplitude

$$\Phi_c \sin^4 2\omega_1\Delta t_1 \sin^8 \omega_E\Delta t_2;$$

the signals at the instants $t = 3\tau/2$ and $\tau/2$ cannot be observed, inasmuch as the equality $2v_{2AR} = v_{1A}$ is not satisfied in MgO ($v_W$ are the velocities of the acoustic waves).

The pulse series 1E − 2A does not excite an electromagnetic echo signal, and only a single-pulse induction signal (t = 0) with $\mathbf{k}_{1ER} = \mathbf{k}_{1E}$ and with amplitude

$$4\Phi_s \sin^2 2\omega_E\Delta t_1 \cos^2 \omega_2\Delta t_2.$$

In MgO it is possible in this case to observe only the acoustic signals on the transition with $\Delta m = \pm 2$. The echo ($t = 2\tau$) propagates in the same direction as the 2A pulse, with amplitude

$$\Phi_c \sin^4 2\omega_E\Delta t_1 \sin^4 \omega_2\Delta t_2.$$

The pulse series 1E − 1A excites from among the electromagnetic signals only the single-pulse induction signal at the instant t = 0, with vector and amplitude

$$\mathbf{k}_{1ER} = \mathbf{k}_{1E},\quad 4\Phi_s \sin^2 2\omega_E\Delta t_1 \cos^2 2\omega_1\Delta t_2 \cos^4 \omega_1\Delta t_2,$$

and from the acoustic signals in MgO it excites a type 1 AR two-pulse induction signal with

$$\mathbf{k}_{1AR} = \mathbf{k}_{1A},\quad 4\Phi_s \sin^2 2\omega_1\Delta t_2 \cos^2 2\omega_E\Delta t_1$$

and two signals on the $\Delta m = \pm 2$ transition: at the instant $t = 3\tau/2$ with amplitude

$$4\Phi_s \sin^2 2\omega_E\Delta t_1 \sin^2 2\omega_1\Delta t_2 \sin^4 \omega_1\Delta t_2$$

for only the DRR, and at the instant $t = \tau$ with the amplitude

$$\tfrac{1}{4}\Phi_c \sin^4 2\omega_1\Delta t_2 (1-3\cos^2 2\omega_E\Delta t_1)^2.$$

According to (8), the intensity of the response consists of an incoherent noise part proportional to $N_0$ and a coherent pulse that assumes the role of a useful signal and is proportional to $N_0^2$. Let us estimate these parts for acoustic excitation of a crystal by 2A − 2A pulses. The 2 AR superradiance signal is of the form

$$\begin{aligned} I_{\nu 2} = 4I_{\nu 0}(\mathbf{k}) \Big\{ & N_0 [\Phi_{ch}^{1/2} - \Phi_s^{1/2}(\cos 2f_1 \cos 2f_2 + \sin 2f_2 \,\mathrm{Re}\,[h(t-\tau)\alpha_j\beta_j^*])] \\ & + \Phi_s \sum_{j\neq l} \delta_j\delta_l^* [\alpha_j^*\alpha_l \cos^2 2f_1 \sin^2 2f_2 h(t-\tau) + \beta_j^*\beta_l \cos^4 f_2 \sin^2 2f_1 h(t) \quad (19) \\ & + (\alpha_j^*\alpha_l)^2(\beta_j\beta_l^*)\sin^4 f_2 \sin^2 2f_1 h(t-2\tau)] \Big\}, \end{aligned}$$

where $\nu = 2A$, $f_i = \omega_2\Delta t_i$,

$$\Phi_{ch}^{1/2} = \frac{\mathrm{ch}\zeta}{2\mathrm{ch}\zeta+1},\quad h(t-n\tau) = \exp[-2i\omega_j(t-n\tau)],$$
$$\delta_j = \exp(-i\mathbf{k}_{2AR}\mathbf{r}_j),\quad \alpha_j = \exp(-i\mathbf{k}_{2A}\mathbf{r}_j) = \beta_j$$

and the initial intensity $I_{\nu 0}(\mathbf{k})$ is given by (11).

To compare the intensities of the coherent and incoherent parts, we note that superradiance takes place in the direction of the applied pulses and is contained in the solid angle $\Delta w \approx \lambda^2/S$, where $\lambda$ is the wavelength of the applied coherent sound and S is the cross-sectional area of the sample through which the acoustic wave passes. Consequently, in the course of the estimate it is necessary to multiply the term with $N_0$ in (19) by $\Delta\omega/4\pi$—the part of the incoherent radiation that enters together with the coherent signal into the acoustic-response detector.

For numerical estimates we use data given in[4,13] for the $Ni^{2+}$ ion in the MgO crystal:

$$\nu_0 = \omega_0/2\pi = 10^{10}\,\text{Hz},\quad G_{11} = 1.13\cdot 10^{-14}\,\text{erg/def. unit},$$
$$G_{44} = 0.99\cdot 10^{-14}\,\text{erg/def. unit},\quad v_{ac} = 10^6\,\text{cm/sec},\quad \rho = 3.6\,\text{g/cm}^3.$$

We choose the sample in the form of a cylinder with cross section area $S = 1\ \text{cm}^2$ and length 1 cm, and with a paramagnetic-particle concentration $N_c = 10^{-4}$, i.e., we assume a number of impurities $N_0 = 10^{19}\ \text{ion/cm}^3$, a temperature 2°K, and times $T_2^* = 2\times 10^{-8}$ sec and $T_2 = 1.2\times 10^{-6}$ sec; the acoustic pulses are of $\Delta t = 5\times 10^{-7}$ sec duration and produce a relative crystal deformation $\epsilon_{xx} = 10^{-8}$ def. unit.

For comparison we choose the maximal part of the incoherent radiation and the echo signal in formula (19). Since $T_2^* < L/v_{ac}$ (see (16)), we choose for the effective number of particles participating in the radiation the value $N_{eff} = N_0 T_2^* L'/T_2 L$. We put $L' = L$, and then the intensity (19) can be estimated from the formula

$$\begin{aligned} I_{2A,2} &= 4I_{0.2A}(\mathbf{k})\left[N_0 \frac{T_2^*}{T_2}\frac{\Delta w}{4\pi}\Phi_{ch}^{1/2} + \Phi_s N_0 \left(\frac{T_2^*}{T_2}\right)^2 \frac{\lambda^2}{S}\sin^4 f_2 \sin^2 2f_1\right] \\ &= 4I_{0.2A}(\mathbf{k})\{2.64\cdot 10^5 + 5.46\cdot 10^{14}\}, \quad (20) \\ I_{0.2A} &= 0.48\cdot 10^{-15}. \end{aligned}$$

It is seen from (20) that the acoustic-echo signals are

$10^9$ times more intense than the noise and produce a relative deformation

$$\varepsilon=(2I^2_{2A,2}/\rho v_{ac}^3 S)^{1/2}\approx 10^{-9} \text{ def. unit}$$

The foregoing calculations of the phonon echo and of the electromagnetic responses are also valid for nuclear spins and quadrupole nuclear electric moments in a cubic crystal. Let us estimate the superradiance signals relative to the incoherent part (spontaneous noise) in the system of nuclear spins of $D_2$ with I = 1, having a quadrupole moment, in the single-crystal ferroelectric $KD_2PO_4$.

According to [16], the crystal has a symmetry $D_{2d}$ and the spins, owing to the weak quadrupole coupling (case of strong magnetic fields) have three nonequidistant levels. It is possible, however, to choose the orientation of the external magnetic field $\mathbf{H}_0$ such that there is no shift of the spectral line due to the quadrupole interaction. To this end the field $\mathbf{H}_0$ is oriented at an angle $\theta = 55°$ [14] to the axis in the xy plane, perpendicular to the fourfold inversion-rotation axis—the c axis (xyz is the crystallographic coordinate system, x(b) and y(b) are twofold axes, and z is the c axis). As a result, the level will be equidistant with a splitting $\hbar\omega_0$.

Since $L/v_{ac} < T_2^*$ for $KD_2PO_4$, the number of spin particles taking part in the superradiance is $N_{eff} = N_0 = 10^{22}$ particles/cm$^3$, the sample is chosen with length L = 1 cm and area S = 1 cm$^2$; $v_{ac} = 3.7 \times 10^5$ cm/sec [14], $T_2^* = 10^{-4}$, $\epsilon_{xx} = 10^{-5}$ def. un., $\nu = \omega_0/2\pi = 10^7$ Hz, pulse duration $\Delta t = 10^{-4}$ sec. The coherent signal then predominates over the noise at T = 0.1°K, i.e., in the case when the spins are polarized, if $|G| \sim 5 \times 10^{-22}$ erg/def. un. [15], and its intensity is $I_{coh} \sim 10.79$ erg/sec, corresponding to the relative deformation $\epsilon_{xx} \sim 10^{-8}$ def. un. In the cubic-symmetry single crystal $RbMnF_3$, the superradiance effect on the $Mn^{55}$ nuclei with a spin-phonon interaction constant $|G| \sim 10^{-6}$ erg/def. un. is comparable with the effect on the electron spins.

The numerical estimates show that the correct choice of the shape of the sample, of the impurity concentration, and of the times $T_2^*$ and $T_2$ is determined by the maximum signal/noise ratio and by the possibility of phonon-echo detection. It is interesting that when the concentration of the active impurities increases, the shortening of the time of the coherent acoustic radiative damping can result not in the expected increase of the echo-signal intensity, but in the impossibility of its observation. At $T_2^* \ll T_2$, the use of samples that are too large is also detrimental to the conditions for the observation of phonon echo, since under these conditions only part of the sample can produce a coherent acoustic signal, which is masked by the incoherent acoustic noise from the remaining part of the crystal.

## CONCLUSION

The foregoing investigation illustrates the great diversity of the new coherent signals that can be obtained simultaneously with the usual spin-echo signals. The high sensitivity of the measurements and the clear-cut phonon-echo signals obtained in the very first experiments give grounds for hoping that a transition from stationary to coherent pulsed methods in quantum acoustics will lead to the same appreciable progress in this research as occurred in the field of magnetic microwave spectroscopy on the transition from stationary magnetic resonance to research by means of spin echo. Also highly promising is the detection of phonon echo by the methods of neutron diffraction, the Mössbauer effect, $\gamma$-$\gamma$, $\gamma$-$\beta$, and other methods of perturbed angular correlations of nuclear radiation.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Translated by J. G. Adashko
239