PHONON INDUCTION AND ECHO

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Just as a coherent excited spin system emits free induction and echo electromagnetic signals, so a system of N active centers, interacting with a crystalline lattice, will generate sound pulses of power $W \propto N^2$ after narrow-pulse pumping by an external generator of arbitrary character. The excitation of the phonon analog of a light echo by sound pulses in the traveling-wave regime is considered in detail.

THE response of a system of impurity particles in a crystal to the pulsed action of a coherent sound field of high power is considered below. If the frequency of the traveling sound waves is identical with the acoustic resonance frequency,[1] then, after the passage of the exciting pulse, the system of impurity particles will be in a state in which it will generate sound of the same frequency.

The impurity particles interact with the lattice and generate sound by the same mechanism that ensures spin-lattice relaxation of paramagnetic ions in crystals (see, for example, [2, 3]). The sound generated by the system of impurities is a short, powerful pulse, the leading edge of which directly abuts the trailing edge of the exciting pulse. In what follows, we shall call this response the phonon induction signal, by analogy with the well-known phenomenon of spin induction.[4]

We assume that the first exciting pulse was introduced in the sample at the time $t = 0$. We further assume that this pulse and the phonon induction signal are not reflected from the far end of the specimen. At the time $t_1 > T_2^*$, we subject the system to a second sound-pulse excitation of the same power but of double length, introducing the sound from the same end as the first pulse. Then, at the time $t = 2t_1 < T_2$, a coherent sound signal of short duration is propagated from this end of the sample, generated by the system and growing along the path.

This response of the system to the impurity centers we call the phonon echo signal, in analogy with the well-known phenomenon of spin echo.[5] The leading edge of the phonon echo signal reaches the end of the sample after a time t_1 following passage of this end by the second exciting pulse. The time (denoted by T_2^*) of the transverse relaxation of the excited centers is determined by the inhomogeneity of the external, and the scatter of the internal, magnetic and electric fields at the impurities; T_2^* is smaller than the time T_2 of the irreversible transverse relaxation, brought about by the two-particle interaction and by the interactions of the particles with the lattice.

Figure 1 shows schematically the sequence of the exciting pulses and the responses generated by the system. For clarity, we have drawn a rather long specimen, and taken an instant of time at which the exciting pulses and the responses are localized in different parts along the sample.

Inasmuch as the isolation of the pulses passing through the sample can turn out to be very difficult, another variant of the experiment is possible, shown schematically in Fig. 2. The shaded part in it denotes the specimen with impurities, and the unshaded portion is, for example, quartz, by means of which the sound signals are transformed into

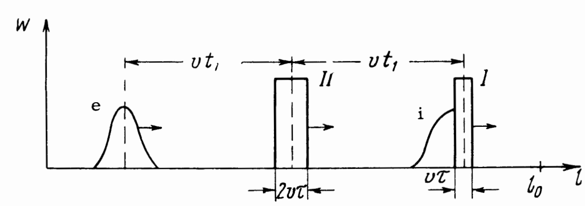

FIG. 1. Successive views along a sample in an experiment on the excitation of phonon induction (i) and echo (e). Axis 1 is directed along the axis of the cylindrical sample. The end through which the sample is excited coincides with the origin of coordinates, l_0 is the length of the sample, v is the sound speed, τ the duration of the first reflected pulse I, II is the second reflected pulse. The arrows indicate the direction of motion of the pulses, W is the amplitude of the pulses. The amplitudes (e) and (i) are not to scale and pertain to I and II, respectively.

electromagnetic ones. If $l' > v_0 t_1$ (l' is the length of the quartz, v_0 the speed of sound in it), then the traveling and reflected pulses will encounter one another outside the sample with the impurities, and the reflected signals will not distort the induction and echo signals. Phonon-phonon scattering can be neglected.[6] For characteristic parameters of nuclear acoustic resonance[7] at the resonance frequency $\nu_0 = 10^{-7}\ \sec^{-1}$ ($\tau = 10^{-5}$ sec, $t_1 = 10^{4}$ sec, and $v_0 = 5 \cdot 10^5$ cm/sec) we get $l' = 50$ cm. In the case of frequencies of the electron acoustic paramagnetic resonance,[8] $\nu_0 = 10^{10}\ \sec^{-1}$ we get $l' \geq 0.5$ cm for $\tau = 10^{-7}$ sec and $t_1 = 10^{-6}$ sec.

It must be kept in mind that coherent induction and echo signals are accompanied by a background of incoherent spontaneous phonon emission. The generated background power is proportional to the number of impurity particles N and does not depend on the direction of the wave vectors of the exciting pulses, while the power of the induction and echo signals is proportional to N^2.

It is useful to emphasize that, inasmuch as the sound wavelength is approximately 10^{-5} times the length of the electromagnetic waves of the same frequency and, as a rule, is always less than the dimensions of the specimen, the phonon induction and echo are, in their coherent properties, to a great degree similar to optical rather than spin induction and echo.[9,10] The latter permits us to use the formalism developed for the description of the super-radiating states of large systems in the consideration of these phenomena.[11]

The possibility of observation of these phenomena stems from the analogy which exists between the character of particle-photon interaction and particle-phonon interaction, and also from the following general circumstances. The field of a coherent external generator at some point of space x at the time t is described by a quantum-mechanical average value $\langle \hat{H} \rangle = \text{const} \cdot \cos(\omega t - \varphi_x)$, which is possessed by the field operator $\hat{H}$ in Glauber states,[12] where the macroscopic parameters ω

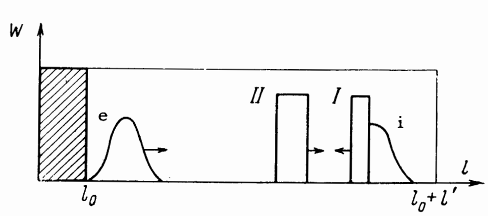

FIG. 2. Observations of induction (i) and echo (e) in a combination sample with account of reflected signals. l_0 is the sample length with impurity centers, l' the length of the additional sample without impurities ($l' > v_0 t_1$). The remaining notation is the same as in Fig. 1.

and φ_x are the angular frequency and the phase, respectively. After cessation of the interaction of matter with such a field, the material for some time manages to "remember" the values φ_x and ω (T_2 for φ_x, and T_1 for ω). If the excited system of N particles is able to generate some physical field spontaneously, then, in the course of time T_2, this field will also "recall" the value φ_x, i.e., it will be coherent. In a number of researches[4,5,13] it has been shown that, if the system excited by any stationary field can spontaneously generate an electromagnetic field of power $W \propto N$, then, for excitation pulse length $\tau \ll T_2$, this system will generate the power $W \propto N^2$ (the quantum yield certainly increases). It is natural to assume that if the system, under stationary excitation, can spontaneously generate a field of phonons with power $W_A \propto N$, then under pulsed excitation we get the sound $W_A \propto N^2$.

As a specific system, we consider a crystal with an impurity of particles j having an effective spin R^j[14] and unperturbed Hamiltonian

$$\mathcal{H}_0 = \sum_{j=1}^{N} g_z \beta H_0 R_z{}^j,$$

where g_z is the spectroscopic splitting factor,[8] H_0 the static magnetic field (directed along the z axis), and β the Bohr magneton or nuclear magneton.

The interaction Hamiltonian of the impurity j with the lattice vibrations is written in terms of the effective spin operator in the form

$$\mathcal{H}_1 = -\frac{1}{2} \sum_{\mathbf{k}_s} (\mathbf{A}_{\mathbf{k}_s} \mathbf{e} R_{\mathbf{k}_s}{}^{+} + \mathbf{A}_{\mathbf{k}_s}^{+} \mathbf{e}^* R_{\mathbf{k}_s}{}^{-}),$$

$$A_{\mathbf{k}_s \xi} = \Big[\sum_{\alpha\beta} \frac{(-i)}{2} \Big(\frac{\hbar}{2M\omega_{\mathbf{k}_s}} \Big)^{1/2} |\mathbf{k}_s| \, d_{\mathbf{k}_s \alpha\beta} G_{\alpha\beta z \xi} H_0 \Big] a_{\mathbf{k}_s},$$

$$d_{\mathbf{k}_s \alpha\beta} = (\Phi_{s\alpha} k_{s\beta}{}^0 + k_{s\alpha}{}^0 \Phi_{s\beta}),$$

$$\mathbf{k}_s = \mathbf{k}_s{}^0 |\mathbf{k}_s|, \qquad \mathbf{e} = \mathbf{x}_0 - i\mathbf{y}_0, \qquad \xi = x, y, z;$$

$$R_{\mathbf{k}_s \pm} = \sum_{j=1}^{N} R_{\pm}{}^j \exp(\pm i \mathbf{k}_s \mathbf{r}_j), \qquad R_{\pm}{}^j = R_x{}^j \pm i R_y{}^j, \tag{1}$$

$\mathbf{x}_0$ is the unit vector along the x axis, $\Phi_{s\alpha}$ the α-component of the unit polarization vector of the s-th mode of the lattice vibrations, $\mathbf{k}_s$ the wave vector of the phonon of mode s, M the mass of the crystal, a_{ks}^{+} and a_{ks} the phonon creation and annihilation operators, ω_{ks} the angular frequency corresponding to $\mathbf{k}_s$, $G_{\alpha\beta\gamma\delta}$ the components of the spin-phonon interaction tensor,[15] and $\mathbf{r}_j$ the radius vector of the particle j.

If, at the time $t = 0$, a pulse in the form of a plane sound wave $(\sum_{\alpha\beta} \epsilon_{\alpha\beta}^{(0)}) e^{i(\omega t - \mathbf{k}_1 \cdot \mathbf{r})}$ with amplitude $\sum_{\alpha\beta} \epsilon^{(0)}_{\alpha\beta}$, frequency $\omega_0 = g_z \beta H_0 \hbar^{-1}$, and wave vector $\mathbf{k}_1$ is introduced in a sample containing the impurity j, then the acoustic induction signal is a set of traveling plane waves with wave vectors $\mathbf{k}_{0s}$. The total intensity of the induction signal and the spontaneous noise, measured at the output of the sample (directly after the passage of the trailing edge of the exciting pulse) in a unit solid angle Ω in the direction $\mathbf{k}_{0s}$, is calculated from the formula (we shall not consider the radiation reaction or emission during the action of the exciting pulse)

$$W_1(\mathbf{k}_{0s}) = W_0(\mathbf{k}_{0s}) \operatorname{Sp}\{\rho(\tau) R_{\mathbf{k}_{0s}+} R_{\mathbf{k}_{0s}-}\},$$

$$\rho(\tau) = L(\tau)\rho_0 L^{-1}(\tau), \quad L(\tau) = \exp[-i\hbar^{-1}\tau \mathcal{H}_{\mathbf{k}_1}],$$

$$\rho_0 = \left[\operatorname{Sp}\exp\left(-g_z\beta\frac{H_0R_z}{k_BT}\right)\right]^{-1}\exp\left(-g_z\beta\frac{H_0R_z}{k_BT}\right),$$

$$\mathcal{H}_{\mathbf{k}_1} = B_{\mathbf{k}_1}R_{\mathbf{k}_1+} + B_{\mathbf{k}_1}^* R_{\mathbf{k}_1-}, \quad R_\xi = \sum_{j=1}^{N} R_\xi^j,$$

$$B_{\mathbf{k}_1} = \frac{i}{2} H_0 \sum_{\alpha\beta} \varepsilon_{\alpha\beta}^{(0)} (G_{\alpha\beta zx} - iG_{\alpha\beta zy}), \tag{2}$$

where k_B is the Boltzmann constant, T the temperature of the sample before excitation, $W_0(\mathbf{k}_{0s})$ the intensity of spontaneous emission of phonons in a unit solid angle by an isolated impurity (in the case $R^j > 1/2$, one must understand by $W_0(\mathbf{k}_{0s})$ the value averaged over the transitions between different sublevels).

Calculations by means of Eqs. (2) lead to the result

$$W_1(\mathbf{k}_{0s}) = W_0(\mathbf{k}_{0s})\Big\{N \operatorname{Sp}\rho^j R_+^j R_-^j + \sum_{j\neq l}(\operatorname{Sp}\rho^j R^j_{\mathbf{k}_{0s}+}) \times (\operatorname{Sp}\rho^l R^l_{\mathbf{k}_{0s}-})\Big\},$$

$$\rho^j = \left\{\operatorname{Sp}\exp\left(-\frac{g_z\beta H_0}{k_BT}R_z^j\right)\right\}^{-1}\left\{\exp\left[-\frac{g_z\beta H_0}{k_BT}\times\left\{R_z^j\cos\theta_1 - \frac{i}{2}(R^j_{\mathbf{k}_1+} - R^j_{\mathbf{k}_1-})\sin\theta_1\right\}\right]\right\},$$

$$R^j_{\mathbf{k}_\xi\pm} = R_\pm^j \exp(\pm i\mathbf{k}_\xi \mathbf{r}_j), \quad \theta_1 = 2\hbar^{-1}\tau|B_{\mathbf{k}_1}|. \tag{3}$$

After taking the traces, we get

$$W_1(\mathbf{k}_{0s}) = W_0(\mathbf{k}_{0s})\cdot {}^2/_3 R^j(R^j+1)N\Big\{1 - \gamma\cos\theta_1 + {}^2/_3R^j(R^j + 1)\gamma^2\sin^2\theta_1 \cdot N^{-1}\sum_{j\neq l}\exp[i(\mathbf{k}_1 - \mathbf{k}_{0s})(\mathbf{r}_j - \mathbf{r}_l)]\Big\}. \tag{4}$$

In this expression, the term containing the sum $N^{-1}\sum_{j\neq l}$ describes the power of the acoustic induction signal $W_{ai}(\mathbf{k}_{0s})$.

In the case $R^j = 1/2$ we have $j = \tanh(\hbar\omega_0/2k_BT)$. In the approximation of the Debye model of the lattice, we have

$$W_0(\mathbf{k}_{0s}) = \frac{3\omega_0^4 H_0^2}{32\pi m_0 v^5}\left|\sum_{\alpha\beta}(G_{\alpha xz\beta} + iG_{\alpha yz\beta})d_{\mathbf{k}_{0s}\alpha\beta}\right|^2, \tag{5}$$

where m_0 is the density of the crystal. When $R^j > 1/2$ and at higher temperatures we have $j = g_z\beta H_0/2k_BT$.

It is seen from (4) that phonons with $\mathbf{k}_{0s} = \mathbf{k}_1$ are emitted in the induction signal of maximum intensity. For sample dimensions much smaller than the wavelength $(\mathbf{k}_1\cdot(\mathbf{r}_j - \mathbf{r}_l), \mathbf{k}_{0s}\cdot(\mathbf{r}_j - \mathbf{r}_l) \ll 1)$, the sum is

$$N^{-1}\sum_{j\neq l}\exp[i(\mathbf{k}_1 - \mathbf{k}_{0s})(\mathbf{r}_j - \mathbf{r}_l)] = N - 1.$$

For the excitation of the echo, we introduce a pulse of plane sound waves of the same frequency, of doubled length, and with a wave vector $\mathbf{k}_2$ at the time $t = t_1$. The total intensity of the echo and spontaneous noise at the time $t = 2t_1$ is computed from the formula

$$W_2(\mathbf{k}_{0s}) = W_0(\mathbf{k}_{0s})\cdot{}^2/_3R^j(R^j+1)N\Big\{1 - \gamma\cos\theta_1\cos\theta_2 + {}^1/_6R^j(R^j+1)\gamma^2\sin^2\theta_1(1-\cos\theta_2)^2N^{-1}\sum_{j\neq l}\exp[i(\mathbf{k}_{0s} + \mathbf{k}_1 - 2\mathbf{k}_2)(\mathbf{r}_j - \mathbf{r}_l)]\Big\}, \tag{6}$$

where $\theta_2 = 2\theta_1$ and the term containing the sum $N^1\sum_{j\neq l}$ describes the power of the phonon echo signal $W_{ae}(\mathbf{k}_{0s})$. As is seen from (6), the maximum power of the echo signal will be emitted in a direction $\mathbf{k}_{0s}$ satisfying the equation $\mathbf{k}_{0s} = 2\mathbf{k}_2 - \mathbf{k}_1$.

Equation (6) is obtained from (2) upon substitution of

$$L(2t_1) = \exp\left[-i\sum_j \Delta\omega^j(t - t_1)R_z^j\right]\exp[-2i\hbar^{-1}\tau\mathcal{H}_{\mathbf{k}_2}] \times \exp\left[-i\sum_j\Delta\omega^j t_1 R_z^j\right]L(\tau),$$

for L(t), where $\Delta\omega^j = \omega_0^j \sim \omega_0$ is the spread of frequencies brought about by the inhomogeneity of the external and internal fields, ω_0^j the resonance frequency for the particle j, ω_0 the frequency of of the exciting sound $\langle(\Delta\omega^j)^2\rangle_{av} \propto (T_2^*)^{-1}$, $\langle\ \rangle_{av}$ is the average over all j (see also Eqs. (A1)–(A18) in [10]). If $T_2^* < \tau$, then the number N in (4) and (6) must be replaced by $T_2^*\tau^{-1}N$.

The total induction and echo signal power is then

$$W_{A\xi} = \int d\Omega W_{A\xi}(\mathbf{k}_{0s}), \quad \xi = \text{i, e} \tag{7}$$

The form of the phonon induction signal at the time $t \leq l_0 v^{-1}$ is described by the formula

$$W_{ai}(t) = W_{ai}^{(1)} f_\zeta(t), \quad f_1(t) = \operatorname{sech}^2(\eta t),$$

$$f_2(t) = \exp[-(t/T_2^*)^2], \qquad \eta = l_0(vtN\hbar\omega_0)^{-1}W_{ai}^{(1)}, \tag{8}$$

where $W_{ai}^{(1)}$ is computed from (7) and (4) with the replacement of N by $l_0^{-1}vtN$. The quantity $f_1(t)$ describes the case in which the form of the signal is determined by the coherent phonon radiation decay ($\eta^{-1} = T_{2A}^W < T_2^*$), and $f_2(t)$ the case in which the decay is brought about by a transverse relaxation ($T_{2A}^W > T_2^*$).

Inasmuch as the induction and echo signals have sharp power maxima corresponding to the directions of $\mathbf{k}_1$ and $2\mathbf{k}_2 - \mathbf{k}_1$ respectively, the integration in the calculation of the total power from Eq. (7) must be carried out from $\Omega = 0$ to $\Omega = 4\pi$.[16] For $\mathbf{k}_1 \parallel \mathbf{l}$ or $\mathbf{k}_2 \parallel \mathbf{k}_1 \parallel \mathbf{l}$, we get

$$W_{A\xi} = \left(\frac{\pi N}{4|\mathbf{k}_0|}\right)^2 S_0^{-1} W_0(\mathbf{k}_0)\, \mathrm{th}^2\left(\frac{\hbar\omega_0}{2k_B T}\right) f_\xi,$$

$$f_i = \sin^2\theta_1, \quad f_e = {}^1\!/_4 \sin^2\theta_1 (1 - \cos\theta_2)^2,$$

$$|\mathbf{k}_0| = \omega_0/v, \quad R^j = {}^1\!/_2. \tag{9}$$

where S_0 is the area of the end face of the sample.

Thus, for Co^{2+} in MgO,[15] for $\mathbf{l}$ parallel to one of the crystallographic axes in a field $H_0 \sim 10^4$ Oe, directed at an angle of 45° to $\mathbf{l}$, T = 4°K, $S_0 = 1\ \mathrm{cm}^2$, $N = 10^{20}$, $\theta_1 = \pi/2$, $\theta_2 = \pi$.

$$W_{A\xi} \sim 10^{-4}[N\omega_0 H_0 (G_{xxyy} - G_{xxxx})]^2 [(2\pi)^2 m_0 S_0 v^3]^{-1}.$$

Using the data from [15], we find that $W_{A\xi} \sim 10^{11}$ erg/sec for induction and echo pulses of duration 10^{-7} sec.

In the excitation of the phonon induction and echo by sound pulses, the duration and amplitude of the deformations of the excited pulses should satisfy the condition:

$$\varepsilon_0\tau = \begin{cases} \pi b/2 & \text{for } 90° \text{ pulse} \quad (\theta_1 = 90°), \\ \pi b & \text{for } 180° \text{ pulse} \quad (\theta_2 = 180°), \end{cases}$$
$$b = 4\hbar H_0^{-1} |G_{xxxx} - G_{xxyy}|^{-1} \sim 4\cdot 10^{-13}\,\mathrm{sec},$$

i.e., for $\tau \sim 10^{-7}$ sec, $\epsilon_0 \sim 4 \cdot 10^{-6}$.

We consider an Fe^{2+} impurity in MgO,[17] spin $R^j = 1$. It is easy to see that the calculations in this case can lead to the same formulas upon replacement of $H_0 G_{\alpha\beta\gamma\delta}$ (with accuracy to within an order of magnitude) by the spin-phonon interaction constant for a particle with spin $R^j > 1/2$. In our specific case, we obtain $W_{ai} \sim 10^{15}$ erg/sec for a pulse duration of $\sim 10^{-7}$ sec, and $b \sim 10^{-14}$ sec.

The phenomena of phonon induction and echo can be used for the pulse generation of sound. One of the most appropriate objects for such a type of experiment are crystals of KCl with OH^- electric dipoles. The constant of dipole-phonon interaction in this case $\sim 10^{-11}$ erg. If an external electromagnetic pulse in such a sample excites a signal of electromagnetic induction or echo, then, as follows from (4) and (6), sound pulses will be generated with $W \sim 10^{19}$ erg/sec, for duration of the process of 10^{-8} sec.

In conducting media, phonon induction and echo can be excited effectively by helicons.[20] The observed acoustic signals can be excited by electromagnetic pulses and in paramagnetic crystals. Thus, if spin induction or echo is excited in MgO with Fe^{2+} impurity, then sound will be generated with a power $\sim 10^8$ ergs/sec (for a sample length of ~ 1 cm). By such a method, sound can be effectively generated in fiber crystals (whiskers) which should possess a high quality factor for longitudinal sound. Thus, in the excitation by an electromagnetic wave of whiskers of thickness $|k_0|^{-1} \sim 10^{-5} - 10^{-4}$ cm, length ~ 1 cm, for a concentration of impurities $\sim 10^{19}\ \mathrm{cm}^{-3}$ and $H_0 G_{\alpha\beta\gamma\delta} \sim 10^{-15}$ erg, the generated power $\sim 10^8$ erg/sec. For sound excitation of spin echo,[19] the spontaneous generation of coherent sound reduces to a situation in which the spin echo signals will be damped out not within a time T_2 but within the time T_{2A}^W which is less than T_2 in this case. This effect appears especially strong for sound excitation of nuclear spin echo. Thus, setting $\omega_0 = 2\pi \cdot 10^7\ \mathrm{sec}^{-1}$, the spin-phonon interaction constant $\sim 10^{-17}$ erg, sample length ~ 10 cm, $v \sim 10^5$ cm/sec, $\hbar\omega_0/k_B T \sim 10^{-3}$, we get $T_{2A}^W \sim 10^{-7}$ sec $\ll T_2 \sim 10^{-3}$ sec.

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