# ACOUSTIC MAGNETIC RESONANCE OF $Cr^{3+}$ IN $LiNbO_3$

B. P. Smolyakov, L. M. Meil'man, V. P. Klyuev, I. A. Shpil'ko, and U. Kh. Kopvillem
Kazan' Physico-technical Institute, USSR Academy of Sciences
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At a temperature 4.2°K and frequency $\nu_0 = 10^{10}$ Hz we observed resonant absorption of hypersound in an acoustic resonator with $Q \sim 10^6$, made of a multidomain $LiNbO_3$ single crystal. This absorption was dependent on the magnitude of the static magnetic field $H_z$ and was due to the $Cr^{3+}$ impurity ions (ion concentration ~0.05%). Insofar as we know, this is the first direct observation of electron acoustic magnetic resonance (APR) [1] in a ferroelectric. In earlier investigations of quartz [2-4], an interaction between hypersound and the spin system of impurity centers was observed indirectly by EPR saturation with sound. However, when the frequencies of the hypersound and the EPR coincided no resonant change in the EPR intensity was observed.

The employed sample had 1.6 cm length, $V = 0.7$ cm$^3$ volume, and optically flat and parallel ends (accurate to 10"). The planes of the ends were parallel to the optic axis $C_3$ of the crystal, and the sound waves propagated perpendicular to $C_3$. The hypersound was generated directly in a thin layer of the end surface, placed in the antinode of the electric field of a tuned coaxial resonator. The excitation was by means of microwave pulses of duration $\tau = 0.5 \times 10^{-6} - 10^{-6}$ sec at a conversion coefficient ~$5 \times 10^{-4}$. The sound pulses reflected from the opposite end were fed to a receiver with threshold sensitivity ~$10^{-13}$ W.

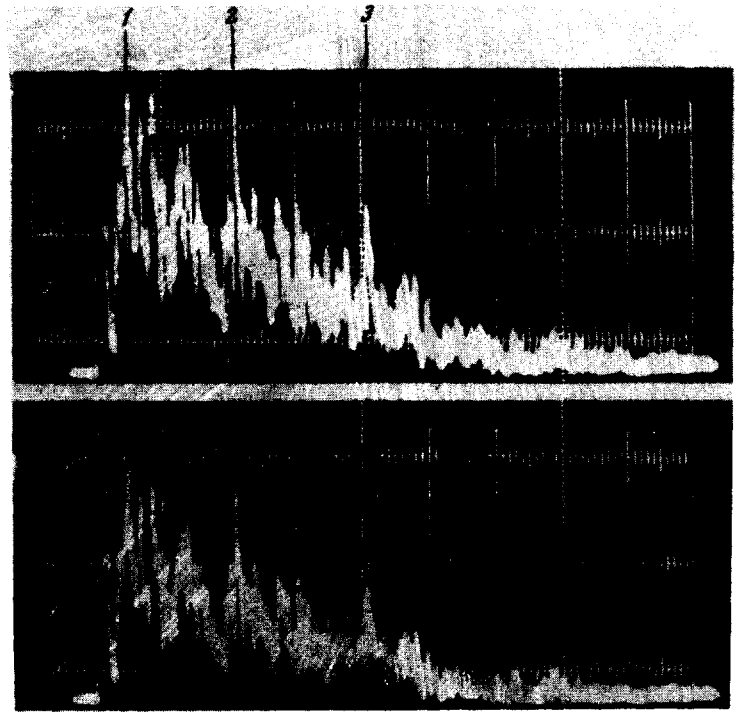


Fig. 1. Attenuation of sound pulses in $LiNbO_3$ at 4.2°K: a - far from resonance, b - at acoustic magnetic resonance in a field $H_{z\circ} = 3120$ G ($H_{z\circ} \parallel C_3$). The oscillograms were taken from the output of the video amplifier of the superheterodyne receiver. Time sweep 10 μsec/cm.

Figure 1 shows oscillograms of the damping of the sound pulses at 4.2°K (a - far from resonance and b - at acoustic magnetic resonance at the point $H_{z^\circ} = 3120$ G ($H_z \parallel C_3$) of maximum abosrption. The shape of the resonance line, shown in Fig. 2, was asymmetric with width $\Delta H = 184$ G at half-intensity. The coefficient of the additional sound absorption due to the interaction with the $Cr^{3+}$ spins was $\alpha_0 \sim 0.07$ $cm^{-1}$.

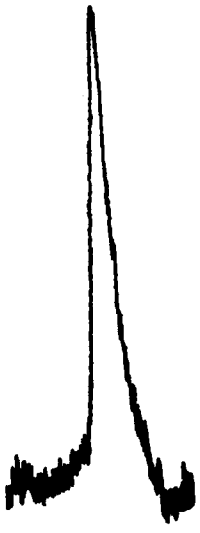

Fig. 2. Line shape of acoustic magnetic resonance as a function of $Hz^\circ \parallel C_3$ at 4.2°K. $Hz^\circ$ increases to the right, one division corresponding to 530 G. The curve was obtained by separating the first of the echo pulses shown in Fig. 1 and by further storing it and recording with an EPP-09M2 automatic plotter. The storing caused also an increase in the signal/noise ratio compared with the ratio on oscillograms a and b.

According to the data of [5,6], the EPR spectrum of the $Cr^{3+}$ ion in $LiNbO_3$ is described by an axial spin Hamiltonian $H_s(S = 3/2)$ with a crystal-field constant $D = 0.45 \pm 0.05$ $cm^{-1}$ and spectroscopic-splitting factors $g_{\parallel} = g = 1.929$ and

$$g_{\perp} = 2g\left[1 - \frac{3}{16}(h\nu_0/2D)^2\right] \qquad (h\nu_0 \ll 2D).$$

In our samples at room temperature we observed in the interval $0 \leq H_z \leq 5$ kG one asymmetric EPR line with width $\Delta H_e = 360$ G between inflection points, at $H_{z^\circ} = 3180$ G, corresponding to the central transition $|1/2\rangle \leftrightarrow |-1/2\rangle$. Comparison of the variation of the resonant value of $H_{z^\circ}$ for EPR and APR as a function of the direction of $H_{z^\circ}$ relative to $C_3$ shows that the APR is also due to the transition $|1/2\rangle \leftrightarrow |-1/2\rangle$.

The correlation observed by us between the EPR frequency of the $Cr^{3+}$ ion and the frequency of resonant absorption of hypersound in $LiNbO_3$ shows that the resonant change of the coefficient $\eta$ for the conversion of hypersound energy into electromagnetic-field energy (which is actually measured by the receiver) is connected with quantum transitions of the $Cr^{3+}$ spins. Since the hypersound is excited directly in the surface layer of the end face of the $LiNbO_3$ crystal containing $Cr^{3+}$ impurities, the change of $\eta$ might be due to the EPR as a result of the penetration of the electromagnetic field into the sample.

We note, first, that variation of $\eta$ as a result of the EPR would cause the oscillogram

b to be derivable from a by decreasing the amplitudes of all the echo signals by the same factor. It is seen from Fig. 1 that this is not the case. In particular, the peaks marked 1, 2, and 3 are respectively related as 1.24, 1.29, and 1.39, i.e., the absorption increases with increasing path covered by the sound wave, as is typical of APR. In $LiNbO_3$, the picture of the reflected echo pulses is distorted with increasing path length, owing to interference between the pulses. Therefore the absorption curve was plotted by us on the basis of the change in the amplitude of the first reflected pulse.

In addition, we performed the following control experiments, which confirmed that EPR makes no contribution to the shape of the sound-absorption line: 1. The $LiNbO_3$ sample was taken out completely from the electromagnetic resonator. The sound was excited, on the other hand, in a quartz cylinder 25 mm long and was transmitted in the usual manner to the $LiNbO_3$. 2. The diameter of the aperture coupling the electromagnetic resonator with the sample was decreased to 3 mm, the diameter of the sample being 8 mm; this should have led to an appreciable attenuation of the magnetic component of the field in the sample. No change of the coefficient of sound absorption in $LiNbO_3$ was noted in these experiments.

The APR results can be described with the aid of the formulas

$$\alpha_0 = (\zeta v_{\parallel}^3 \hbar)^{-1} \pi \Delta n \nu_0 g(\nu_0) G^2 |\langle 1/2 | H_0 | -1/2 \rangle|^2, \; H_{ph} = \epsilon_{\parallel} G H_0, \qquad (1)$$

where $H_{ph}$ is the effective operator of the energy of spin-phonon interaction of $Cr^{3+}$ in $LiNbO_3$, $\epsilon_{\parallel}$ the relative deformation due to the longitudinal sound oscillations, G the spin-phonon interaction constant, $H_0$ the dimensionless operator part, $\zeta$ the crystal density, $v_{\parallel}$ the velocity of propagation of the longitudinal oscillations perpendicular to $C_3$, $\Delta n$ the difference in the populations, $g(\nu_0)$ the form factor of the homogeneously broadened line, and $\langle 1/2|H_0|-1/2\rangle$ the matrix element.

Substituting the values $v_{\parallel} = 1.1 \times 10^6$ cm/sec, $\zeta = 4.64$ g/cm$^3$, $\alpha_0 = 0.07$ cm$^{-1}$, $\Delta n = (h\nu_0/kT) N_0 V \Delta H_1 (\Delta H)^{-1}$, $g(\nu_0) = 10^{-7}$ sec, and $N_0 = 2 \times 10^{22}$ cm$^{-3}$, we obtain $G \sim 10^{-15} \times |\langle 1/2 | H_0 | -1/2 \rangle|^2$ erg/deformation unit. Here $N_0$ is the number of $LiNbO_3$ molecules per unit volume and $\Delta H_1 (\Delta H)^{-1} \sim 10^{-1}$ describes the fraction of the spins which participate simultaneously in the resonance as a result of the inhomogeneous broadening, and $\Delta H_1$ corresponds to the homogeneous line width. When the angle $\theta \leq \pi/2$ between $H_z$ and $C_3$ is increased, the widths of the EPR and APR lines increase. For $\theta = \pi/2$ we have $\Delta H \sim 300$ G and $\alpha_0 \sim 0.035$ cm$^{-1}$.

To calculate $|\langle 1/2|H_0|-1/2\rangle|^2 = b$ it is necessary to know the mechanism of the spin-phonon interaction. If the sound interacts with the $Cr^{3+}$ via the Van-Vleck mechanism [7], then $H_0$ is quadratic in the spin. If $H_z \parallel C_3$ and the Hamiltonian $H_s$ contains no rhombic terms, then b = 0. The character of the EPR and APR line shape indicates that $H_s$ may contain terms of the type $E(S_x^2 - S_y^2)$, $E \sim (0.05 - 0.1)$ cm$^{-1}$. Then $b \sim (E/D)^2$ and $G \sim 10^{-14}$ erg/deformation unit, which is some 10 times larger than the value of G for $Cr^{3+}$ in $Al_2O_3$ [8]. Since it follows from the EPR and APR line shape that large internal-field gradients are present in the sample, it is possible that the sound causes the $Cr^{3+}$ ions to move between regions where they are subject to the action of different internal fields [9]. In this case

$b \sim 1$ and $G \sim 10^{-15}$ erg/deformation unit. Such a spin-phonon interaction mechanism was observed in quartz [2-4], and the dependence of $\alpha_0$ on $\theta$ obtained by us recalls the dependence of $\alpha_0$ on $\theta$ for paramagnetic centers in quartz.

Comparison of G for $Cr^{3+}$ in $Al_2O_3$ and $LiNbO_3$ shows that the presence of electric domains strongly affects the character of the spin-phonon interaction.


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## RADIATION OF AN ATOM (MOLECULE) IN AN ABSORBING MEDIUM


E. L. Feinberg
P. N. Lebedev Physics Institute, USSR Academy of Sciences


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1. We shall estimate the possible role played by one simple effect when a source emits in an absorbing medium (an effect to which apparently no attention has been paid before) - the absorption of radiation and its conversion into heat in the non-wave zone of the emitter. This is essentially a collective interaction between the emitter and a large number of particles that fall into this zone, when the dipole oscillations are inevitably accompanied by quasistationary currents. This effect takes place if $[(4\pi/3)(N/k^3)] \gg 1$, where $\lambda = \lambda_0/\sqrt{|\epsilon|} = 2\pi/(k_0\sqrt{|\epsilon|}) = 2\pi/|k|$ is the wavelength in the medium, $\epsilon = \epsilon' + i\epsilon''$ is the complex dielectric constant, N is the number of absorbing particles per unit volume. The effect has a classical nature and is estimated classically.

The energy absorbed per unit time, dW/dt, referred to the initial emitter energy $\hbar\omega = \hbar ck_0$, gives the following line width (without allowance for the microscopic structure of the medium, i.e., without the impact and Stark widths, and also without the Doppler width):

$$\gamma = \frac{1}{\hbar\omega}\frac{dW}{dt} = \frac{1}{\hbar\omega}\int \sigma |\mathbf{E}|^2 dV = \frac{1}{\hbar\omega}\frac{\omega\epsilon''(\infty)}{4\pi}\int_{(V_0)} |\mathbf{E}|^2 dV \qquad (1)$$

(the notation is standard), where the integration over the volume must be carried out outside a volume $V_0$ of radius $R_0$ such that on the average the volume $V_0$ contains one particle of the medium (the binary broadening theory is valid inside the volume):

$$\frac{4}{3}\pi R_0^3 N = 1. \qquad (2)$$

Since $|\vec{E}| \sim R^{-3}$, the integral depends strongly on $R_0$ when $P < \lambda$ and therefore we cannot obtain an exact estimate.