RESONANCE ABSORPTION OF 10^{10} cps *HYPERSOUND IN RUBY*

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Acoustic paramagnetic resonance with longitudinal 10^{10} cps hypersonic waves was studied experimentally in a ruby single crystal at liquid helium temperatures. The experimental technique is described. The resonance line characteristics, such as intensity, shape, width, dependence of the intensity on the orientation of the static magnetic field are measured for various transitions. The experimental results are compared with the theory of acoustic paramagnetic resonance.

INTRODUCTION

RESONANCE absorption of a sound wave propagating in a paramagnetic crystal arises whenever the splitting of the spin energy levels of the ions in a magnetic field becomes equal to the energy of the sound quantum. This phenomenon was first predicted theoretically by Al'tshuler[1,2] and called acoustic paramagnetic resonance (APR).

The phenomena of APR and EPR have it in common that the absorption of energy of the sound wave and the electromagnetic wave is connected with quantum transitions between levels of the spin system of the paramagnetic ions of the crystal.

In contrast with EPR, where the interaction of the external electromagnetic field with the spin system exists because of direct magnetic dipole coupling, there is no such direct coupling in APR. In this case, the sound modulates the internal crystalline field of the lattice and acts on the orbital levels of the paramagnetic ions and, because of the spin-orbital coupling, acts on the spin system.

In accord with modern representations, the single phonon process of spin-lattice relaxation is realized by means of the same mechanism. Therefore, the study of APR allows us to obtain important information on the spin-lattice interaction in paramagnetic crystals at low temperatures, where single phonon processes predominate.

The general expression for the coefficient of resonance paramagnetic absorption of sound, as a consequence of transitions between levels p and q, has the form (see [2]):

$$\alpha_{pq} = Q|\langle p, n_\omega|H_{\mathrm{sp}}|q, n_\omega - 1\rangle|^2,$$
$$Q = \frac{2\pi}{\hbar^2}\frac{N\hbar\omega V}{kTvn_\omega}g(\omega), \tag{1}$$

where H_{sp} is the spin-phonon interaction operator, N is the number of paramagnetic particles per unit volume, V is the volume of the crystal, ω the sound frequency, T the lattice temperature, n_ω the number of phonons of resonant frequency ω, $g(\omega)$ is a function of the shape of the absorption line, and v the sound speed.

To calculate α_{pq}, it is necessary to specify the form of the operator H_{sp}. The Kronig-Van Vleck model of the mechanism leads to a quadratic form of this operator (for ions with effective spin $S > 1/2$). In this case the polarizations of the sound wave and the direction in which its propagation takes place are significant. Thus, when sound propagates along the trigonal axis of the crystal, we have for longitudinal wave,

$$H_{\mathrm{sp}} = L(S_x^2 + S_y^2 - \eta S_z^2),$$

while for transverse wave H_{sp} is a combination of the operators

$$(S_xS_y + S_yS_x), \quad (S_xS_z + S_zS_x), \quad (S_yS_z + S_zS_y),$$
$$(S_y^2 - S_x^2).$$

As a result, we get the following equation from (1) for the coefficients of resonance absorption: longitudinal wave

$$\alpha^l_{pq} = PL\omega^2|\langle p|S_x^2 + S_y^2 - \eta S_z^2|q\rangle|^2, \tag{2}$$

transverse wave

$$\alpha^t_{pq} = P\omega^2|\langle p|A(S_xS_y + S_yS_x) + B(S_xS_z + S_zS_x) + C(S_yS_z + S_zS_y) + D(S_y^2 - S_x^2)|q\rangle|^2, \tag{3}$$

where $P = \pi NR^2g(\omega)/kTv^3d$, R is the equilibrium distance from the nucleus of the paramagnetic ion to the nearest diamagnetic particle, and d is the density of the crystal. The coefficients A, B, C, D, and L depend on the type of ion and are determined by the splitting of the orbital levels and the value of the spin-orbit coupling.[2]

It follows from (2) and (3) that the measurement of the absolute value of the resonance absorption of sound and of its dependence on the orientation of the static magnetic field relative to the crystalline axis makes it possible to determine the value of the energy of spin-phonon coupling, and to verify how much the standard Kronig-Van Vleck model reflects the actual character of the spin-phonon interaction.

APR has been studied experimentally in a number of researches.[3-5] In the work of Tucker[5] on the effect of sound on the intensity of the EPR line in a ruby, the angular dependence of APR was determined. In such an indirect method of investigation there act on the spin system, in addition to the longitudinal sound wave, the transverse waves generated by it in the contact layer (between the quartz transducer and the ruby). As follows from (2) and (3), the selection rules of allowed resonance transitions in the interaction with the spin system of longitudinal sound waves are essentially different from the case of transverse waves. Therefore, the experimental data on APR are obtained by an indirect method, in which an integral effect takes place, which cannot be interpreted uniquely. For example, the deviations of the curves of angular dependence of APR obtained in [5] from the calculations can be attributed to a Hamiltonian H_{sp} of non-quadratic form, and not only to absorption of the transverse sound waves by the spin system.

In the present work we have investigated APR in a ruby by a direct method, by using pure longitudinal 10^{10} cps hypersonic waves and helium temperatures.

EXPERIMENTAL TECHNIQUES

APR was studied in an optically homogeneous single-crystal ruby with a Cr^{3+} concentration 0.05%; the ruby had the shape of a rod of diameter 2.6 mm and length 18 mm, the geometric axis of which was directed along the optic axis of the crystal. The end faces of the rod were optically flat and parallel to within 10″.

Longitudinal hypersonic waves with frequency 10^{10} cps were excited in the ruby rod by an X-cut quartz transducer, placed in the electric field of a toroidal cavity (Fig. 1) which was placed in liquid helium.

The acoustic resistances and the coefficients of linear splitting of the quartz and the ruby are significantly different. In this connection, the transmission of the hypersound from one crystal to the other proved to be a very difficult problem, the more so at liquid helium temperatures. A number of adhesive materials were tested, including carbinol and epoxy resins. However, satisfactory results were obtained with vacuum grease and ceresin (an oil refining byproduct). These materials made it possible to make an acoustic hypersonic contact between the quartz and the ruby, which was not disturbed during the numerous changes of temperature from room to liquid helium values. Special attention was turned to obtaining a uniform thickness of the contact layer over the entire surface of the ruby and the quartz, since even a small inclination of this layer leads to a transformation of longitudinal hypersonic waves to transverse, because of the large difference in the acoustic characteristics of quartz and ruby.

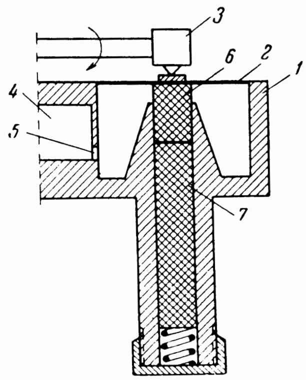

FIG. 1. Construction of the cavity: 1 – Cavity body, 2 – diaphragm for tuning the cavity frequency, 3 – mechanism for moving the diaphragm, 4 – waveguide, 5 – aperture for coupling the cavity with the guide, 6 – piezoquartz, 7 – ruby rod.

A block diagram of the arrangement (APR spectrometer) for excitation of the hypersound and investigation of the line of acoustic paramagnetic resonance is shown in Fig. 2. A pulsed mag-

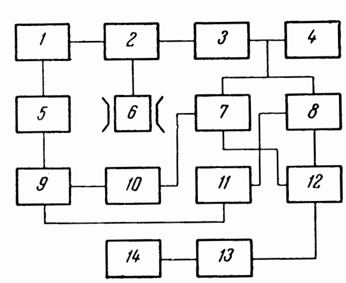

FIG. 2. Block diagram of the APR spectrometer. 1 – Pulsed magnetron, 2 – hybrid ring, 3 – receiver for the 3-cm band, 4 – oscilloscope, 5 – modulator, 6 – cavity with specimen in a magnetic field, 7 – first time gate, 8 – second time gate, 9 – driving pulse generator, 10 – first delay line, 11 – second delay line, 12 – integrator, 13 – dc amplifier, 14 – electronic potentiometer ÉPP-09.

netron was used as the source of high frequency field for excitation of the hypersound. An attenuator and a hybrid ring were placed in the waveguide that connects the magnetron and the cavity, to feed to the detector the pulse reflected from the cavity and the echo pulses corresponding to hypersonic waves excited in the quartz and ruby rod.

The microwave power fed to the cavity amounted to several tens of watts in the pulse, while the power of the echo signals in the cavity, corresponding to hypersonic waves in the ruby, amounted to $10^{-11} - 10^{-10}$ watt; the ratio of signal to noise at the input of the detector, which has a sensitivity of 10^{-12} W, was 5–10 (Fig. 3). The small ratio of the power of the echo signals to the power fed to the cavity is associated with the large losses in the double conversion of the microwave electromagnetic oscillations and the hypersonic waves, and also with the transmission of the hypersound from the quartz to the ruby.

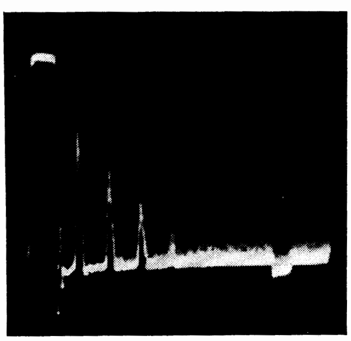

FIG. 3. Echo pulses corresponding to hypersonic waves in the ruby.

The method of amplitude comparison of the echo pulses with different acoustic path lengths in the ruby was used to handle such a weak echo signal and to detect the resonance absorption line against the background of various interferences due to the instability of the apparatus. As calculation shows, the difference in amplitudes of these pulses is insensitive to the interferences indicated, and its variation is determined only by the resonance absorption of the sound.

The difference of the two echo signals was separated by a two-channel system with time gating. Pulses which are gated in correspondence with their time length are fed to the time gate from the driving pulse generator.

The starts of the time intervals during which the first and second gates are open, are selected by means of variable delay lines such that they are identical with the instants of the arrival of the first and second comparison pulses.

The echo pulses which are separated in this fashion are compared, integrated separately, and substracted from one another. As a result, a signal is produced which is proportional to the difference of the echo pulses and which, after amplification, is recorded as a function of the static magnetic field, which changes linearly with the time, and in which a cavity is placed with a paramagnetic specimen.

The two-channel system of time gating allows us to make separate studies of the resonance absorption of the longitudinal and transverse sound waves which are excited simultaneously in the crystal. From the entire series of echo pulses we choose the signals which correspond to any particular sound wave and which, as a consequence of a different velocity of propagation in the crystal, enter the receiver at different instants of time.

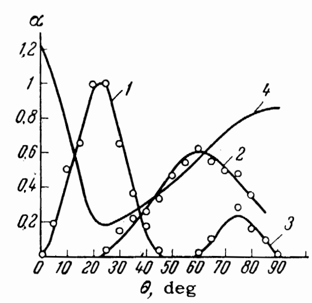

FIG. 4. Angular dependence of the resonance absorption of hypersound in a ruby. Absorption of the longitudinal wave: 1 – For the transition $3/2 \leftrightarrow 1/2$; 2 – for the transition $1/2 \leftrightarrow 1/2$; 3 – for the transition $-3/2 \leftrightarrow -1/2$; 4 – absorption of the transverse wave for the transition $3/2 \leftrightarrow 1/2$. Solid curve – theoretical, o – experimental data. T = 1.8°K.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 4 shows the dependence of the resonance absorption coefficient of longitudinal and transverse hypersonic waves on the angle θ between the trigonal axis of the ruby and the magnetic field, for different transitions, calculated on the basis of Eqs. (2) and (3), and the data on the characteristic functions for the spin levels of the ruby, contained in [6]. In this same drawing, experimental results are plotted for the resonance absorption of the longitudinal waves. The excellent agreement between computation and experiment confirms the validity, at least for ruby, of the quadratic form of the Hamiltonian of the spin-lattice interaction. The curve given in the drawing for the angular dependence of the absorption of transverse waves shows that it can materially exceed the absorption of the longitudinal wave (for example, at $\theta = 0$). This is connected with the difference pointed out above between the selection rules for absorption of longitudinal and transverse waves, and with the fact that $\alpha_{pq} \sim v^{-3}$ while the velocity of the transverse wave is much less than the longitudinal. Therefore, even a small admixture of transverse wave in the sound wave propagating in the ruby can appreciably distort the data on the sound absorption obtained with the help of the indirect method. The maximum measured value of the resonance absorption of the longitudinal wave takes place for $\theta = 20°$ for the transition $^3/_2 \longleftrightarrow {}^1/_2$, and amounts to 0.07 cm^{-1}. This value is one third to one fourth of that calculated on the basis of (2).

This difference can be connected with the insufficient accuracy in the determination of the quantities used for the calculation, for example, of the concentration of the paramagnetic particles, etc. The measured temperature dependence of the resonance absorption also agrees well with the theoretical value determined by the relation $\alpha_{pq} \sim T^{-1}$.

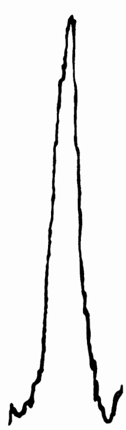

FIG. 5. APR absorption line.

The shape of the APR line (Fig. 5) is close to Gaussian both for the transition $^3/_2 \longleftrightarrow {}^1/_2$ and for the transition $^1/_2 \longleftrightarrow -{}^1/_2$. The width of the line corresponds to 40 and 35 Oe for the first and second transitions, respectively. According to theory,[7] the width and shape of the APR line are determined by the same factors as the EPR line. A quantitative difference can appear as the result of a difference in the operators corresponding to transitions between the spin levels.

The calculations[8] carried out on the basis of the theory of dipole-dipole interaction under very simple assumptions give the same value for the width of the APR and EPR lines. However, the reduced widths of the APR lines exceed the widths of the corresponding EPR lines. For an explanation of this difference, one must go to the effect of exchange interactions which, according to theory, narrow the EPR line and broaden the APR line. However, it is uncertain whether such an explanation is valid, since the concentration of the paramagnetic ions in the ruby (0.05%) is not so large that the exchange interaction can materially affect the width of the APR line.

Evidently, the width of the APR line, and also that of EPR, is determined principally not by the dipole-dipole or exchange interactions, which are small at the given concentrations, but by the local inhomogeneities of the electric field in the lattice,[9] and by the scatter of the crystallographic directions in the microblocks existing in the ruby.

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