RESONANCE ABSORPTION OF ULTRASOUND ON NUCLEI

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Submitted to JETP editor, November 5, 1958

J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 1451-1456 (May, 1959)

An analysis is given for resonance absorption of ultrasound on paramagnetic nuclei in a simple cubic lattice under the assumption that the spin-lattice interaction is due to nuclear quadrupole forces. Absorption factors for the spin transitions characterized by $\Delta m = 1$ and $\Delta m = 2$ are obtained for arbitrary directions of propagation and polarization of the acoustic waves. A comparison between theory and experiment is made for In^{115} in InSb.

1. INTRODUCTION

WHEN acoustic waves of frequency ν are propagated in a paramagnet located in a magnetic field H_0, resonance absorption of the sound can occur if the condition $h\nu = g\beta H_0$ is satisfied, where g is the splitting factor and β is the Bohr magneton.

An analysis of this problem was first given by Al'tshuler[1] who showed that the acoustic absorption is similar to first-order spin-lattice relaxation. Al'tshuler gives the following general formula for resonance absorption of ultrasound:

$$\sigma = \frac{4\pi^2}{h^2}\frac{NhvV}{kTv\nu_{1/2}n_\nu}|\langle m, n_\nu - 1 \mid \hat{\mathcal{H}}' \mid m', n_\nu\rangle|^2, \qquad (1)$$

where N is the number of magnetic particles per unit volume, v is the acoustic velocity, V and T are the volume and temperature of the paramagnet, $\nu_{1/2}$ is the half-width of the absorption line, and $\langle m, n_\nu - 1 \mid \mathcal{H}' \mid m', n_\nu\rangle$ is the matrix element for the spin-lattice interaction for the magnetic sub-levels m and m' and the lattice states $n_\nu - 1$ and n_ν. The absorption factors have been computed by Al'tshuler for a number of materials on the basis of the relaxation mechanisms proposed by Waller and Kronig and Van Vleck.

A number of experiments[2,3] have shown that nuclear spin transitions can be induced by ultrasonic waves. Menes and Bolef[4] have also observed resonance absorption of ultrasound on In^{115} nuclei in InSb.

In the present paper we consider the nuclear quadrupole relaxation mechanism and consider resonance absorption of ultrasound on nuclei.

2. SPIN-LATTICE INTERACTION OPERATOR

The energy of the electric quadrupole interaction of the nucleus with the electric field of the crystal lattice is given by[5]

$$\hat{\mathcal{H}}' = \sum_{\alpha=-2}^{2}(-1)^\alpha \hat{Q}_\alpha \gamma \nabla E^{-\alpha}, \qquad (2)$$

where

$$\hat{Q}_0 = A[3\hat{I}_z^2 - I(I+1)],$$

$$\nabla E^0 = \frac{1}{2}\sum_j e_j r_j^{-5}(3z_j^2 - r_j^2) = \sum_j \nabla E_j^0,$$

$$\hat{Q}_{\pm 1} = \pm\frac{\sqrt{6}}{2}A[\hat{I}_\pm \hat{I}_z + \hat{I}_z\hat{I}_\pm],$$

$$\nabla E^{\pm 1} = \pm\frac{\sqrt{6}}{2}\sum_j e_j r_j^{-5} z_j(x_j \pm iy_j) = \sum_j \nabla E_j^{\pm 1},$$

$$\hat{Q}_{\pm 2} = \frac{\sqrt{6}}{2}A\hat{I}_\pm^2, \quad \nabla E^{\pm 2} = \frac{\sqrt{6}}{4}\sum_j e_j r_j^{-5}(x_j \pm iy_j)^2 = \sum_j \nabla E_j^{\pm 2},$$

$$A = eQ/2I(2I-1),$$

$$eQ = \langle I, I \left| \sum_i e_i r_i^2 \{3\hat{I}_z^2 - I(I+1)\} \right| I, I\rangle,$$

$$\hat{I}_\pm = \hat{I}_x \pm i\hat{I}_y. \qquad (3)$$

$\hat{I}$ and $\hat{Q}$ are the operators for the spin and quadrupole moment of the nucleus, e_i and r_i are the charge of an elementary volume in the nucleus and its distance from the center of the nucleus, e_j and r_j are the charge of the neighboring ion and the distance between it and the nucleus, γ is a quantity which is introduced to take account of the following three factors: the increase in the interaction due to the large quadrupole moments induced in the electron shells of the atom by the quadrupole moment of the nucleus, the reduction in the interaction because of the polarization of the atom by the crystalline field, and the increase in the interaction because of the covalent bonding. It is proposed to determine the quantity γ experimentally.

Equation (2) does not take account of lattice vibrations. In order to take these vibrations into account r_j in Eq. (2) must be written in the form $R_j + u_{kj}$ where R_j is the equilibrium position of

the ion and $\mathbf{u}_{kj}$ is the relative displacement between the nucleus in question and the corresponding ion of the lattice. For simplicity we consider a cubic lattice and the six neighboring ions. Their equilibrium positions, in a coordinate system with origin at the nucleus and with axes parallel to the C_4 axis of the crystal, are at the points $\mathbf{R}_{1,2}(\pm a, 0, 0)$, $\mathbf{R}_{3,4}(0, \pm a, 0)$, $\mathbf{R}_{5,6}(0, 0, \pm a)$, where a is the lattice constant. The displacement of the ion from its equilibrium position is usually written in the form

$$u_k = \sum_{\varphi} \sum_{\delta=x,y,z} [C_{\varphi\delta} \cos(\mathbf{k}\varphi) + C'_{\varphi\delta} \sin(\mathbf{k}\varphi)]\, \mathbf{u}^0_\delta,$$

where φ is a vector normal to the wave, whose magnitude is given by $|\varphi| = 2\pi a\nu/v$; $\mathbf{k}$ is the position of the ion (components expressed in units of a); $\mathbf{u}^0_\delta$ is the normalized polarization vector.

The relative displacement of two neighboring interstices of the lattice will be:

$$\mathbf{u}_{k1} = \sum_{\varphi}\sum_{\delta} \varphi_x q_\delta(\varphi)\, \mathbf{u}^0_\delta, \qquad \mathbf{u}_{k3} = \sum_{\varphi}\sum_{\delta} \varphi_y q_\delta(\varphi)\, \mathbf{u}^0_\delta \text{ etc.}, \tag{4}$$

where $q_\delta(\varphi)$ characterizes propagation with wave number φ and polarization δ. The index k, which corresponds to the initial nucleus, will be dropped in the following. It should be recalled that $\mathbf{u}_1 = -\mathbf{u}_2$, $\mathbf{u}_3 = -\mathbf{u}_4$, $\mathbf{u}_5 = -\mathbf{u}_6$. As is well known[6]

$$\langle n_\nu - 1 \,|\, q_\delta(\varphi) \,|\, n_\nu\rangle = [n_\nu h / 4\pi M\nu]^{1/2}, \tag{5}$$

where M is the mass of the crystal.

Writing $\mathbf{r}_j = \mathbf{R}_j + \mathbf{u}_j$ we expand ΔE^α about the equilibrium distance between the ions $|\mathbf{R}_j| = a$. Limiting ourselves to first-order terms in $\mathbf{u}_j$, since we are interested only in the first-order processes:

$$\nabla E^0(\mathbf{u}) = \sum_j \nabla E^0_j(0) + \sum_j \sum_\delta [\partial(\nabla E^0_j)/\partial u_{j\delta}]_{\mathbf{r}_j=\mathbf{R}_j} u_{j\delta}, \tag{6}$$

$$[\partial(\nabla E^0_j)/\partial u_{jx}]_{\mathbf{r}_j=\mathbf{R}_j} = X_j[1 - Z_j^2 R_j^{-2}], \ \ldots\ . \tag{7}$$

Substituting (7) in (6), when $\mathbf{e}_j = e$, we have

$$\nabla E^0(\mathbf{u}) = \nabla E^0(0) + 3ea^{-4}[u_{1x} + u_{3y} - 2u_{5z}],$$

and, similarly,

$$\nabla E^{\pm1}(\mathbf{u}) = \nabla E^{\pm1}(0) \pm (\sqrt{6}/2)\, ea^{-4}[u_{5x} \pm iu_{5y} + u_{1z} \pm iu_{3z}],$$

$$\nabla E^{\pm2}(\mathbf{u}) = \nabla E^{\pm2}(0) + (\sqrt{6}/4)\, ea^{-4}[3(u_{3y} - u_{1x}) \pm 2i(u_{3x} + u_{1y})]. \tag{8}$$

In what follows, in Eq. (8) we will drop the term $\nabla E^\alpha(0)$ since it causes only an unimportant shift in the levels. Using (2), (3), and (8) we write the interaction operator in the form

$$\hat{\mathcal{H}}'_0 = B[3\hat{I}_z^2 - I(I+1)][u_{1x} + u_{3y} - 2u_{5z}],$$
$$\hat{\mathcal{H}}'_{\pm1} = B[\hat{I}_\pm\hat{I}_z + \hat{I}_z\hat{I}_\pm][u_{5x} \pm iu_{5y} + u_{1z} \pm iu_{3z}],$$
$$\hat{\mathcal{H}}'_{\pm2} = {}^1/_2 B\hat{I}_\pm^2[3(u_{3y} - u_{1x}) \pm 2i(u_{1y} + u_{3x})], \tag{9}$$

where

$$B = 3e^2Q\gamma/2I(2I-1)a^4.$$

$\hat{\mathcal{H}}'_0$ does not cause transitions in the spin system and must be discarded.

3. MATRIX ELEMENTS FOR THE SPIN-LATTICE INTERACTION AND THE ABSORPTION COEFFICIENT

To find the absorption factors we consider several cases; these correspond to various orientations between the direction of propagation of the acoustic waves and the magnetic field and the various possible polarizations of the acoustic oscillations.

Perpendicular Propagation of the Acoustic Waves

The magnetic field is along the z axis and the ultrasound propagates along the x axis. In this case only $\mathbf{u}_1 = -\mathbf{u}_2$ is different from 0. Equation (9) yields

$$\hat{\mathcal{H}}'_{\pm1} = B[\hat{I}_\pm\hat{I}_z + \hat{I}_z\hat{I}_\pm]u_{1z}, \quad \hat{\mathcal{H}}'_{\pm2} = {}^1/_2 B\hat{I}_\pm^2[-3u_{1x} \pm 2iu_{1y}].$$

a) For transitions of the spin system with $\Delta m = 1$ different from 0 the matrix elements correspond to oscillations polarized along the z axis:

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm1} \,|\, m \mp 1, n_\nu\rangle = B\langle m \,|\, \hat{I}_\pm\hat{I}_z + \hat{I}_z\hat{I}_\pm \,|\, m \mp 1\rangle \langle n_\nu - 1 \,|\, u_{1z} \,|\, n_\nu\rangle.$$

From Eq. (4), for a given frequency and polarization it follows that $u_{1z} = \varphi_x q_z(\varphi)$. Then

$$\langle n_\nu - 1 \,|\, u_{1z} \,|\, n_\nu\rangle = (2\pi a\nu/v)[n_\nu h/4\pi^2 M\nu]^{1/2},$$

and the matrix element

$$\langle m \,|\, \hat{I}_\pm\hat{I}_z + \hat{I}_z\hat{I}_\pm \,|\, m \mp 1\rangle = \xi \equiv (2m-1)\sqrt{(I+m)(I-m+1)}.$$

Thus,

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm1} \,|\, m \mp 1, n_\nu\rangle = (Ba/v)\,\xi\,(n_\nu h\nu/M)^{1/2}. \tag{10}$$

Substituting (10) in (1) we find the absorption factor

$$\sigma^\perp_{12} = PD\xi^2\nu^2/T,$$

where

$$P = 9\pi^2 e^4 N_0/k = 2.05\cdot10^4,$$

$$D = Q^2\gamma^2[I^2(2I-1)^2 a^6 v^3 \nu_{1/2}\mu]^{-1}, \quad d = \frac{M}{V},$$

$N_0 = Nd/\mu$ is Avogadro's number and μ is the molecular weight.

TABLE I

	I	$Q\cdot10^{26}$ cm^2	$a\cdot10^8$ cm	$v\cdot10^{-5}$ cm/sec	$10^{-4}\gamma^2$	ξ^2	η^2	$D\cdot10^{33}$
Br^{79} in LiBr	3/2	28	2.745	3.9	3	12	12	1.2
Br^{79} in KBr	3/2	28	3.29	3.39	3.4	12	12	0.51
Br^{79} in AgBr	3/2	28	2.88	3	20	12	12	6.1
I^{127} in KI	5/2	—46	3.52	2.9	25	80	40	0.69
In^{115} in InSb	9/2	117	2.23			576	144	

b) For transitions of the spin system with $\Delta m = 2$ different from 0 we have the following matrix elements: for x-polarization (cf. also reference 7)

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm 2} \,|\, m \mp 2, n_\nu \rangle = -(3Ba/2v)(n_\nu h\nu/M)^{1/2},$$

for y-polarization

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm 2} \,|\, m \mp 2, n_\nu \rangle = \pm (iBa/v)[n_\nu h\nu/M]^{1/2}\eta,$$

where $\eta^2 = (I+m)(I+m-1)(I-m+1)(I-m+2)$. In this case the absorption factors are

$$\sigma_{2x}^{\perp} = {}^9/_4 PD\eta^2\nu^2/T, \qquad \sigma_{2y}^{\perp} = PD\eta^2\nu^2/T.$$

Parallel Propagation of the Acoustic Waves

The magnetic field and the direction of propagation of the acoustic waves are along the z axis. Only u_5 is different from 0. Equation (9) now yields

$$\hat{\mathcal{H}}'_{\pm 1} = B[\hat{I}_\pm \hat{I}_z + \hat{I}_z \hat{I}_\pm][u_{5x} \pm iu_{5y}], \qquad \hat{\mathcal{H}}'_{\pm 2} \equiv 0.$$

Thus only transitions with $\Delta m = 1$ are possible. For x-polarization

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm 1} \,|\, m \mp 1, n_\nu \rangle = (Ba/v)(n_\nu h\nu/M)^{1/2}\xi,$$

for y-polarization

$$\langle m, n_\nu - 1 \,|\, \hat{\mathcal{H}}'_{\pm 1} \,|\, m \mp 1, n_\nu \rangle = \pm (iBa/v)(n_\nu h\nu/M)^{1/2}\xi.$$

The corresponding absorption factors are

$$\sigma_{1x}^{\parallel} = PD\xi^2\nu^2/T, \qquad \sigma_{1y}^{\parallel} = PD\xi^2\nu^2/T.$$

From these general formulas it is possible to find the numerical values of σ for a number of materials whose pertinent parameters are known. These parameters and the quantities ξ^2, η^2 and D are shown in Table I. The quantities Q and I are taken from reference 8; a is taken from reference 9; v is obtained from the elasticity constant; (c_{11}) given in reference 10; the mean line width for the nuclear resonance is taken as 10^4 sec^{-1}.[11] The quantity γ^2 is taken from the experimental data of Wikner and Das,[12] who computed the γ_T^2 due to the quadrupole moment of the electronic shell of the atom. However, the theoretical relaxation times computed by these authors, T_T, do not coincide with the experimental values τ_E. According to the general theory given by Kranendonk[5] the spin-lattice relaxation time τ is proportional to γ^{-2}. It is reasonable to assume that γ_E^2, which takes account of all effects and agrees with experiment, will be given by $\gamma_E^2 = \gamma_T^2\tau_T/\tau_E$.

In Table II are shown values of σ for transitions from the lower magnetic level with m = I.

4. CONCLUSION

Because of the lack of data, a comparison of theory with experiment is possible only for In^{115} in InSb.[4] The authors propagated ultrasound at a frequency $\nu = 9.976$ Mcs in a InSb single crystal located in magnetic fields $H_0 = 10.69 \times 10^3$ gauss and $H_0 = 5.35 \times 10^3$ gauss for the $\Delta m = 1$ and $\Delta m = 2$ transitions respectively. These authors observed clearly defined resonance absorption peaks and also investigated the dependence of these peaks on the angle θ between the magnetic field and the direction of propagation of the acoustic wave. The values $\theta = 0$ and π correspond to maxima for the $\Delta m = 1$ transitions and minima for the $\Delta m = 2$ transitions; the values $\theta = \pi/2$ and $3\pi/2$ correspond to maxima for $\Delta m = 2$ transitions and minima for $\Delta m = 1$ transitions. The resonance peaks are given in relative units. Minimum absorption corresponds to one relative unit σ_0, maximum corresponds to 7. The ratio

TABLE II

	$\sigma_{1z}^{\perp}\frac{T}{\nu^2}\cdot10^{17}$	$\sigma_{2x}^{\perp}\frac{T}{\nu^2}\cdot10^{17}$	$\sigma_{2y}^{\perp}\frac{T}{\nu^2}\cdot10^{17}$	$\sigma_{1x}^{\parallel}\frac{T}{\nu^2}\cdot10^{17}$	$\sigma_{1y}^{\parallel}\frac{T}{\nu^2}\cdot10^{17}$
Br^{79} in LiBr	0.295	0.64	0.295	0.295	0.295
Br^{79} in KBr	0.13	0.28	0.13	0.13	0.13
Br^{79} in AgBr	1.5	3.4	1.5	1.5	1.5
I^{127} in KI	11.3	12.7	5.7	11.3	11.3

of the absorption maxima for the transitions characterized by $\Delta m = 2$ and $\Delta m = 1$ is 0.66 ± 0.8.

To make a comparison with experiment we find the dependence of the absorption factor on the angle θ. Let the magnetic field be along the z axis and let the acoustic wave be propagated in the xz plane at an angle θ with respect to the z axis. Since σ is proportional to φ^2 all the $\sigma^{\perp}$ must be multiplied by $\sin^2\theta$ and the $\sigma^{\parallel}$ by $\cos^2\theta$. If we assume that waves of all three polarizations propagate, in the notation of Sec. 3

$$\sigma_1 = \sigma_{1x}^{\perp} + \sigma_{1y}^{\perp} + \sigma_{1z}^{\perp} + \sigma_{1x}^{\parallel} + \sigma_{1y}^{\parallel} + \sigma_{1z}^{\parallel}$$

$$= PD\xi^2(2\cos^2\theta + \sin^2\theta), \quad \sigma_2 = \frac{13}{4} PD\eta^2 \sin^2\theta. \qquad (11)$$

Since P and D are positive, σ_1 has maxima for $\theta = 0$ and π and minima for $\theta = \pi/2$, $3\pi/2$; σ_2 has maxima for $\theta = \pi/2$, $3\pi/2$ and minima for $\theta = 0$, π, in accordance with experiment.

The theoretical ratio of the maximum values of σ_2 and σ_1 can be obtained taking σ_1 at $\theta = 0$ and σ_2 at $\theta = \pi/2$ in Eq. (11):

$$(\max \sigma_2 / \max \sigma_1)_T = 13\eta^2 / 4 \cdot 2 \cdot \xi^2 = 0.41.$$

It is noteworthy that of all the materials considered in Sec. 3 this ratio is less than unity only for InSb. The estimate was made out under the assumption that the linewidth is the same for $\Delta m = 1$ and $\Delta m = 2$. However, as is noted by the authors, the linewidth for the $\Delta m = 2$ signal is somewhat smaller. Consequently, with an exact calculation of the widths $(\max \sigma_2/\max \sigma_1)_T$ would increase.

At those values of θ for which there is theoretically no absorption, $\sigma = \sigma_0$.[4] If σ_0 refers to usual nonresonance absorption then it, and $\sigma_{1E} = 6\sigma_0$, can be estimated numerically from the dimensions of the crystal and the nonresonance acoustic absorption factor σ_{nonres}. Unfortunately the data on σ_{nonres} and on the velocity of sound and γ^2 required for a numerical calculation are not available in the literature. A rough estimate indicates that for agreement of $\max \sigma_{1E}$ and $\max \sigma_{1T}$ it is necessary to take $\gamma^2 > 2 \times 10^6$ at room temperature.

In conclusion we wish to express our gratitude to Prof. S. A. Al'tshuler for his valuable advice and interest in this work.

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