RESONANT ABSORPTION OF ULTRASOUND BY NUCLEI IN A SUPERCONDUCTOR IN THE INTERMEDIATE STATE

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A calculation is presented of the coefficient of absorption of ultrasound by nuclear quadrupole levels in an intercrystalline electric field in a metal that is in the intermediate state. The absorption is due to the following mechanisms: a) modulation of the critical field in the normal layers of the superconductor under the influence of ultrasonic deformations, b) motion of the nucleus under the influence of the ultrasound relative to the interface between the superconducting and normal phases $R_z(t)$, which lead to the appearance in the region of the NS transition of an alternating field

$$H(t) = R_z(t)\lambda^{-1}H_{c0}\exp(-|z_i - z_n|\lambda^{-1})$$

(λ is the thickness of the transition layer, and z_n and z_i are the coordinates of the NS boundary and of the i-th nucleus). An expression is obtained for the dependence of the absorption intensity on the temperature and on the orientation of the propagation direction of the longitudinal ultrasound relative to the direction of the external field and the plane of the boundary.

If the electric field inside a crystal has a threefold symmetry axis or a symmetry axis of higher order, then the spin levels of the nuclei with spins $I \geq 3/2$, owing to the quadrupole interaction with the field of axial symmetry, form Stark electric levels with energies equal to

$$E(m) = \frac{e^2qQ}{4I(I-1)}[3m^2 - I(I+1)], \tag{1}$$

where Q is the quadrupole moment of the nucleus and q is the gradient of the electric field[1].

It is assumed that the main mechanism causing the resonant absorption of the ultrasound on the quadrupole levels in non-conducting non-magnetic crystals is ultrasonic modulation of the gradient of the internal electric field[2,3]. In the heretofore performed experiments, the transfer of acoustic energy to the nuclear spins was effected by this mechanism. A typical value of the coefficient of absorption for dielectric crystals of the KI type is $\alpha \approx 10^{-8}\ \mathrm{cm}^{-1}$[2].

It can be stated presently with assurance that observation of resonant absorption of ultrasound on nuclear Zeeman levels in metals has been made only in tantalum, the nuclei of which have unprecedented large values of Q[4]. The difficulties in the observation of the resonant absorption in other metals are due to the small value of the coupling between the quadrupole and magnetic moments of the nucleus and the large relaxation width of the resonance line, compared with non-conducting crystals, resulting from a hyperfine interaction of the nuclei with the conduction electrons. The causes of the weak nuclear spin-phonon coupling in metals are not yet clear. It is assumed that it is due to dynamic screening of the ion-quadrupole interaction by the conduction electron.

We consider in this paper new possible mechanisms for resonant absorption of ultrasound energy by nuclear quadrupole levels in a superconductor in the intermediate state.

1. MECHANISMS OF INTERACTION OF ULTRASOUND IN NUCLEI

As is well known[5,6], at a temperature lower than the temperature of the superconducting transition T_c, a superconductor of the first kind placed in a magnetic field $H < H_c$ (H_c is the critical field) is in an intermediate state characterized by alternation of normal (N) and superconducting (S) layers with respective thicknesses d_n and d_s, $d = d_n + d_s$. The period of the intermediate structure d and the fraction of the volume occupied by the S regions are determined from the condition that the free energy be a minimum[5]. For a sample in the form of a plate of thickness l, in a magnetic field perpendicular to the plane of the plate, the fraction $\rho_s = d_s/d$ of the volume of the sample in the S state and the period d are equal to[5,6]

$$\rho_s = 1 - H/H_c, \quad d \approx 10\sqrt{l\eta},$$

where $\eta \approx 3 \times 10^{-5}$ cm.

In the regions occupied by the normal phase, the magnetic field is equal to the critical field. The condition of equilibrium between the phases is written in the form[5]

$$(\mathbf{Hn})|_{r=r_n} = 0, \quad [\mathbf{En}]|_{r=r_n} = 0, \quad [\mathbf{Hn}]|_{r=r_n} = \mathbf{H}_c, \tag{2*}$$

where n is a unit vector normal to the surface of the NS boundary at the point r_n.

If the sample in the intermediate state is subjected to the homogeneous deformation with a relative change of the volume in the region r described by the function f(r) then, by virtue of the dependence of the critical field on the degree of compression and tension of the lattice,

$$H_c(r) = H_{c0}(1 + \beta f(r)),$$

the equilibrium conditions that determine the dimension in the form of the N and S regions are violated. This

*$[\mathbf{En}] \equiv \mathbf{E} \times \mathbf{n}$.

leads to a displacement of the boundaries between the N and S regions, accompanied by a phase transition $\zeta(t, rn)$.

The motion of the boundaries between the phases gives rise to eddy currents. The Joule heat released by these currents causes a change in the sample temperature, and as a result an additional change in the critical field. Thus, the critical field in the normal layers of an isotropic superconductor in a longitudinal sound wave is written in the form

$$H_c(r) = H_{c0}(1 + \beta \operatorname{div} \mathbf{u}) + \frac{\partial H_c}{\partial T} T'(r), \qquad \mathbf{u} = \mathbf{u}_0 \exp[i(\mathbf{kr} - \omega t)], \tag{3}$$

where $\mathbf{u}$ is the vector of displacement in the sound wave, and the value of β is given in[8]:

$$\beta = -\frac{\rho}{H_{c0}}\left(s_l^2 - \frac{4}{3}s_t^2\right)\left\{\left(\frac{\partial H_c}{\partial P}\right)_T - \left(\frac{\partial H}{\partial T}\right)_P \frac{T}{\rho C}\left(\frac{\partial \rho}{\partial T}\right)_P\right\},$$

where ρ is the density of the metal, s_l and s_t are the longitudinal and transverse sound velocities, C is the specific heat of the intermediate state, $C = (d_n C_n + d_s C_s)/d$. Thus, in normal layers the interaction of ultrasound with the nuclei can be the result of a new mechanism, namely the modulation of H_c by ultrasound. Obviously, resonant absorption of the ultrasound will occur when the frequency of the ultrasound modulation coincides with the frequency of the splitting between the spin levels of the nucleus in the external magnetic field H_{z0} and in the internal electric field. The energy of the splitting between the levels $\Delta_{m,\,m-1}$ usually lies in the frequency range $\nu \approx 10^6$–10^7 sec^{-1}[9,10]. The amplitude u_0 of sound oscillations of medium intensity at this frequency is 10^{-6}–10^{-7} cm[2]. According to (3), at $H_{c0} \approx 10^3$ G, div u $\approx u_0\omega/s$, sound velocity $s = 3 \times 10^6$ cm/sec, $\beta \approx 1$, the amplitude of the acoustic modulation of H_c reaches $\Delta H_c \approx 10^{-1}$ G, which is of the same order as the amplitude of the ultrasonic modulation of the energy of interaction between the quadrupole and the intercrystalline electric field in dielectric crystals[2].

There is one other possible mechanism of enhancement of the resonant absorption of ultrasound, due to the existence of intermediate NS layers. In a coordinate system with the z axis normal to the NS separation boundary, and with the x axis directed along H_c, the magnetic field inside the superconducting layer decreases like $H_c \exp(-|z_n - z|/\lambda)$, where z_n is the coordinate of the NS separation boundary. Under the influence of ultrasound, in the coordinate frame rigidly fixed in the oscillating NS boundary, the value of the z component of the coordinate of the i-th nucleus oscillates like $R_z(t) = u_z(t, z_i) - \zeta_z(t, z_n)$. As a result of the fact that the local field in the transition layer depends strongly on the coordinates, the nuclei located in the layer $z_n + R_z^0 \le z_i \le z_n + \lambda - R_z^0$, where $R_z^0 = \max|R_z(t)|$, will be acted upon in an acoustic wave by an alternating field equal to

$$\mathbf{H}(t) = \mathbf{H}_{c0}\exp[-|z_i + R_z(t) - z_n|/\lambda]. \tag{4}$$

For nuclei located in layers $z_n - R_z^0 \le z_i \le z_n + R_z^0$, the alternating field has the form of a truncated sinusoid:

$$\mathbf{H}(t) \approx \mathbf{H}\,\frac{R^0}{\lambda}\left(\frac{1}{2} - \frac{1}{\pi}\sin\frac{|z_i| - |z|}{R_z^0}\right)e^{i\omega t} \tag{5}$$

The determination of the fields acting on the nucleus inside the N layer, and of the vector $\zeta(t, z_n)$ describing the oscillations of the NS boundary reduces to finding the solution of the system of field equations and the heat-balance equation:

$$\operatorname{rot}\mathbf{H} = \frac{4\pi\sigma}{c}\left(\mathbf{E} + \frac{1}{c}[\mathbf{vH}]\right), \qquad \operatorname{rot}\mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{H}}{\partial t}, \qquad (d_n + d_s)C\frac{dT}{dt} = Q[\dot\zeta_z(z_n) - \dot\zeta_z(z_{n+1})], \tag{6}$$

where $Q = -(T/4\pi)H_c\partial H_c/\partial T$ is the heat of transition from the normal state to the superconducting state, and $\mathbf{v} = \dot{\mathbf{u}}$ is the velocity of the medium.

The velocity of the NS boundary is limited by two parameters, namely the finite relaxation time of the superconducting parameter $\Delta(r(t))$ and the rate at which the heat released in the S → N transformations is carried away. An estimate of the relaxation time of the parameter Δ, given in[11], and also data on the time constant of the cryotron[6], yield a value $\tau \lesssim 10^{-9}$ sec.

In the last relation in (6) it is assumed that the condition of homogeneity of the sample temperature

$$\omega\tau \approx (\omega d/s)(sd/v_F\Lambda) \ll 1$$

(Λ is the mean free path of the electron and v_F is the velocity of the electron with the Fermi energy) is satisfied, as can be readily verified (at $\Lambda \approx 10^{-3}$–10^{-4} cm).

The boundary conditions (2) are written as follows:

$$E_x\Big|_{z=z_n} = 0, \quad E_y + \frac{\zeta_z + u_z}{c}H_{c0}\Big|_{z=z_n} = 0, \qquad H_x - H_c\Big|_{z=z_n} = 0, \quad H_z - H_{c0}\frac{\partial(u+\zeta)}{\partial x}\Big|_{z=z_n} = 0. \tag{7}$$

It is convenient to assume that the z axis lies in the middle of the N layer.

In the stationary regime, all the quantities in (3) depend on the coordinates and on the time like $F(z)\exp(ik_x x + ik_y y - i\omega t)$. When the amplitude of the sound oscillations is small, $|ku_0| \ll 1$, it is possible to confine oneself in the solution of (2) to terms of first order in u.

The system (6) and (7) was considered by E. Lifshitz and Sharvin[7], in an approximation linear in u, in the calculation of the coefficient of the electronic sum absorption due to the motion of boundaries with a phase transition. According to[7] the alternating magnetic field $\mathbf{H}_1 = \mathbf{H} - \mathbf{H}_{c0}$ can be represented in the form

$$\mathbf{H}_1 = \frac{i}{\omega}\operatorname{rot}[\mathbf{v}H_{c0}] + H_{c0}\mathbf{h}, \tag{8}$$

where

$$h \equiv h_x = \frac{\operatorname{div}\mathbf{u}(1 + \beta - \cos^2 p)\cos lz}{\cos(l d_n/2) - il\delta D\sin(l d_n/2)} \qquad D = \frac{T\delta}{4\pi C d}\left(\frac{\partial H_c}{\partial T}\right)^2$$

$$l = (1+i)/\delta, \qquad \delta = c/\sqrt{2\pi\sigma\omega},$$

and p is the angle between the vectors $\mathbf{H}_{c0}$ and $\mathbf{k}$.

The equation for $\zeta_z(t, z_n)$ in the chosen coordinate system is obtained by using the boundary conditions (7) for E_y, where E_y is expressed by the equations in (6):

$$\zeta(\pm d_n', t) = \frac{1}{l^2}\frac{\partial h}{\partial z}\Big|_{z=\pm d_n'} \tag{8'}$$

Here $d_n' = d_n/2$. Bearing in mind that

$$C \approx \frac{p_F^2 T}{v_F \hbar^3}, \quad \frac{\partial H_c}{\partial T} \approx \frac{H_{c0}(0)}{T}\frac{T}{T_K},$$

where $H_{c0}(0) \approx T_c p_F (v_F \hbar^3)^{-1/2}$, we obtain $D \approx (\delta/d_n)(T/T_c)^2$. As will be shown below, the probabilities of the transition of nuclei between quadrupole levels as a result of modulation of the magnetic field acting on the nuclei are proportional to the square $|H_1|^2$ of the amplitude of the resonant field. For nuclei in the transition layer we have $|H_1|^2 = \text{const} \cdot (R_z^0)^2$. From relations (8) and (8′) we obtain for the square of the oscillation amplitude R_z^2

$$|R_z|^2 \approx u_{z0}^2 + \frac{\delta^4}{4}(1+\beta-\cos^2 p)^2 (ku_0)^2 \left[k_z^2 + \frac{1}{\delta^2}\left(1 - \sin\frac{d_n}{\delta}\right)\right] \quad (9)$$

when $d_n \gg \delta$, and

$$|R_z|^2 \approx u_{z0}^2 \left[1 + \delta^2 k^2 (1+\beta-\cos^2 p)\left(1+\frac{T^2}{T_c^2}\right)^{-1}\right] \text{when } d_n \ll \delta. \quad (9')$$

As follows from these formulas, when $u_z \to 0$ we have $R_z^0 \to 0$ in the case (9′) but $R_z^0 \neq 0$ in the case (9). This result can be explained qualitatively as being due to the fact that when $d_n \ll \delta$ the change of $H_c(r)$ over distances $\Delta(r) \approx d_n$ in the normal layers is small, and in the intermediate state, when ultrasound is produced and propagates along the NS boundaries, the equilibrium state of the sample is hardly violated, this leads to $\zeta(t) \to 0$.

2. CALCULATION OF THE RESONANT-ABSORPTION COEFFICIENTS

The quadrupole level with energy given by formula (1) is doubly degenerate and is described by the wave functions $|\pm m\rangle$.

In the general case, the eigenvalues of the energy of the nuclei and the wave functions $|\overline{m}\rangle$ in the internal electric and external magnetic fields are obtained by solving numerically the secular equation

$$\|\langle m|\hat{H} - E|m'\rangle\| = 0,$$

where $\hat{H} = \hat{D} + \mu\hat{I}H_{c0}$, $\hat{D}$ is the quadrupole-interaction operator, and $\mu = \mu_N$ is the nuclear Bohr magneton. In fields $H_{c0} < 10^2$ G, the energy of the magnetic interaction is smaller than the quadrupole splittings $\Delta_{m,m-1}$ ($\mu H_{c0}/\Delta_{m,m-1} \leq 1/12$), so that when calculating $|\overline{m}\rangle$ and $\overline{\Delta}_{m,m-1}$ we can use the ordinary perturbation theory. The weak anisotropy of the surface-tension energy on the boundary between the phases can cause the NS boundary to make a certain angle φ with the plane drawn through the axial symmetry axis of the electric field e_q and the vector $\mathbf{H}_{c0}$ (θ is the angle between e_q and $\mathbf{H}_{c0}$). If the anisotropy of the boundary energy depends on the symmetry of the intercrystalline field, then it is reasonable to assume that φ is equal to zero or $\pi/2$. Expressing H_{c0} and $\hat{H}_1(t)$ in a coordinate system with z' axis parallel to the quantization axis e_q, we obtain for the energy of the splitting and for the probability of the transition $|\overline{m}\rangle \to |\overline{m-1}\rangle$

$$\overline{\Delta}_{m,m-1} = \Delta_{m,m-1} + A\cos\theta + A^2 F(I,m)\sin^2\theta,$$
$$w = \frac{1}{\hbar^2} g(\nu)\left[|\langle m|V_1|m-1\rangle|^2 - 2\sin\theta\frac{\mu H_{c0}}{\Delta_{m,m-1}} J^+(m)\,\mathrm{Re}(\langle m|V_1|m-1\rangle H_z')\right], \quad (10)$$

where

$$V_1 = \mu\hat{I}\mathbf{H}', \; J^+(m) = (I+m)(I-m+1), \; A = \mu H_{c0}(z),$$

$$F(I,m) = \sum_{j=-1}^{1} \mathrm{sign}(j)\,(1+\delta_{m,m+j}) J^+(m+j)\,(\Delta_{m+j,m+j-1})^{-1},$$

$$\begin{pmatrix} H_x' \\ H_y' \\ H_z' \end{pmatrix} = \begin{pmatrix} H_{1x} \\ H_{1y} \\ H_{1z} \end{pmatrix} \begin{pmatrix} -\sin\theta & \cos\theta\cos\varphi & -\cos\theta\sin\varphi \\ 0 & \sin\varphi & \cos\varphi \\ \cos\theta & \sin\theta\cos\varphi & \sin\theta\cos\varphi \end{pmatrix}.$$

Here $g(\nu)$ is the normalized form function of the line, and for the center of the curve it can be assumed equal to the reciprocal half-width of the curve.

The intensity of absorption is proportional to the number of nuclei taking part in the absorption and to the square of the amplitude of the resonant field. According to (9) and (9′) we have $R_z^0 \approx 10^{-6}$ cm. A rough estimate of the intensities of the absorption due to modulation of H_c by ultrasound P_N and of the absorption due to modulation of H by the motion of the nuclei in the NS walls, P_{NS}, at typical values $d_n \approx 10^{-2}$ cm, $\lambda \approx 5 \times 10^{-5}$ cm, and $k \approx 4 \times 10$ cm^{-1} yields

$$\frac{P_{NS}}{P_N} \approx \frac{\lambda}{d_n}\left(\frac{R_z^0}{\lambda}\right)^2 \frac{1}{(ku_0)^2} \approx 10^3.$$

Thus, practically all the absorption takes place in the NS walls.

The time-average energy absorbed per unit volume by nuclei as a result of oscillations of the NS walls is

$$E = \frac{N_0}{2I+1}\frac{2\lambda}{d}(n_m - n_{m-1})\hbar\omega(w_{m,m-1} + w_{-m,-m+1}), \quad (11)$$

where n_m and n_{m+1} is the relative population of the levels $\overline{m}$ and $\overline{m-1}$. Substituting in the expression for w in (10)

$$V_1 = \mu I^{\pm}(-H(z_i)\sin\theta),$$

where the values of $H(z_i)$ are determined by relations (4) and (5) we obtain for the transition probability averaged over the possible coordinates of the nucleus

$$w = \frac{1}{\hbar^2}(I+m)(I-m+1)\mu^2 H_{c0}^2 \frac{R_z^2}{2\lambda^2} g(\nu)\sin^2\theta. \quad (12)$$

Dividing expression (11) by the energy flux $s_l \rho u_0^2 \omega^2$ in the sound wave, with account taken of (12), we obtain for the sound absorption coefficient at $k_B T \gg \hbar\omega$

$$\alpha_n = \frac{N_0}{2I+1}\frac{8}{3}\frac{R_z^2}{d\lambda}\frac{\mu^2 H_{c0}^2}{k_B T}(I+m)(I-m+1)\frac{\sin^2\theta}{s_l \rho u_0^2 \gamma}, \quad (13)$$

where γ is the width of the absorption curve and θ is the angle between the crystal-field axis and the direction of the external field **H**.

The absorption line width calculated by the usual method of moments is, in frequency units,

$$\gamma = \frac{1}{\hbar\pi} A\cos\theta \left\{\left(\frac{1}{12} + \frac{1}{\pi}\right)\frac{A}{\Delta} + \frac{2}{\pi}\frac{A^2}{\Delta}\left[2F(I,m)\sin^2\theta - \frac{1}{\Delta}\cos^2\theta\right]\right\}^{-1/2}.$$

At $H_c = 10^2$ G we have $\gamma \approx 5 \times 10^4 \cos\theta$ (Hz).

Among the superconductors of the first kind, the most convenient for experimental observation of the absorption under consideration are In, Ga, and α-Hg^{201} with nuclear angular momenta I equal to 9/2 and 3/2, and with critical fields $H_{c0}(0)$ equal to 283, 51, and 412 G. Quadrupole splitting by the method of the magnetic nuclear quadrupole resonance in In and Ga was investigated in[9,10].

In Ga, the quadrupole splitting of the levels $3/2 \leftrightarrow 1/2$ is 1.1×10^7 Hz; in In, the quadrupole splitting between the levels $\pm 1/2, \ldots, \pm 9/2$ is respectively 1.88, 3.77, 5.65, and 7.53 MHz.

In (9′) we have $u_{z0} = u_0 \sin p \sin \varphi_1$, where φ_1 is the angle between the plane of the NS boundary and the plane passing through the vectors **k** and H_{c0}. The average thickness of the transition layer is $\lambda = 1.89\, \xi(T)$, $\xi(T) = 0.74(1 - T/T_c)^{-1/2} \xi_0$, $\xi_0 \approx 3 \times 10^{-5}$ cm[6]. Substituting the expression for λ in $H_c(T)$ in (13), we obtain for the dependence of α_n on the temperature, frequency, and the angles at $d_n \gg \delta$:

$$\alpha_n = \mathrm{const} \cdot \sin^2\theta \frac{1}{T}\left(1 - \frac{T}{T_c}\right)\left(1 + \frac{T}{T_c}\right)^{1/2} \left[\sin^2 p \sin^2 \varphi_1 + \frac{c^2}{8\pi\sigma} \frac{\omega}{s_l^2} \cdot \left(1 - \sin\frac{d_n}{\delta}\right)\right]. \quad (14)$$

In numerical estimates of the absorption coefficient for a plate of In 0.5 cm thick we use the following constants in (11): $N_0 = 3 \times 10^{22}$ cm^{-3}, $\delta = 10^{-2}$ cm, $s_l = 2.7 \times 10^5$ cm/sec, $\rho = 7.3$, $\gamma = 2 \times 10^4$ sec^{-1}, $T = 1.5^\circ$K, $H_c = 250$ G, $p = \varphi = \pi/2$, $\lambda = 5 \times 10^{-5}$ cm, $d = 5 \times 10^{-2}$ cm, $I = 9/2$, and $m = 3/2$. We obtain for the absorption coefficient $\alpha_n \approx 0.6 \times 10^{-7}$ cm^{-1}.

In the case of a polycrystalline sample we have in (14) $\sin^2\theta \rightarrow 1/3$. The line width, which depends on H_c is determined in this case by second-order effects[1] and is smaller than the line width γ_0 due to the relaxation effects.

The absorption coefficient $\alpha_n(N)$ in N layers, due to modulation of the magnetic field by ultrasound, will obviously be equal in order of magnitude to the experimentally measured values $\alpha_n \approx 10^{-9}$ cm^{-1} for superconductors with $H_c = 10^3$ G.

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