SPIN-ACOUSTIC RESONANCE IN PARAMAGNETIC METALS

V. I. GERASIMENKO

Physico-Technical Institute, Academy of Sciences, Ukrainian S.S.R.

Submitted to JETP editor August 10, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 40, 585-589 (February, 1961)

The resonance absorption of ultrasonic energy is found for paramagnetic metals, brought about by the interaction of acoustic vibrations of the lattice with the characteristic magnetic moments of the conduction electrons.

1. In a paramagnetic metal placed in a constant magnetic field H_0 and subjected to the action of ultrasonic vibrations of frequency $\hbar\omega = g\mu H_0$, resonance transitions will take place between two spin sub-levels ($+\mu H_0$ and $-\mu H_0$) as a result of the interaction of the spin magnetic moments of the conduction electrons with the acoustic vibrations of the lattice. Such a resonance phenomenon in the rare earth metals and in paramagnetic salts was first investigated by Al'tshuler.[1] In these substances, particles possessing a characteristic magnetic moment were fixed at certain points in the crystalline lattice, and the breadth of the resonance was therefore determined only by the time of paramagnetic relaxation.

In the case of a metal, it is natural to expect a broadening of the resonance line as a consequence of the high mobility of the conduction electrons. Primary interest in this phenomenon was aroused by the existence of the effect of polarization of the nuclei which accompanies paramagnetic resonance in metals (the Overhauser effect).[2] It should be expected that in spin-acoustic resonance such polarization of nuclei will take place that it would be possible to create polarized nuclear targets of arbitrary dimensions.[3] However, as will be seen from what follows, for those conditions for which resonance exists, polarization of nuclei is absent. It is shown below under what conditions spin-acoustic resonance is possible and the order of magnitude of the resonance magnetization of the metal is estimated; the absorption coefficient of ultrasonic energy is also computed.

2. It is evident that the mechanism of interaction of the spin magnetic moments of the electrons with the sound vibrations of the lattice, which leads to resonance, is the same as the mechanism of paramagnetic relaxation which is caused by the thermal vibrations of the lattice. Investigations have shown that among relaxation mechanisms, the most important is the mechanism which takes into account the spin orbit coupling of the electrons with the field of the lattice.[2,4]

In order to estimate the magnitude of the magnetic interaction of the conduction electrons with the acoustic vibrations, it is necessary to know the wave function of the electron in the lattice with account of spin-lattice interaction. For the case of alkali metals, to which we limit our considerations, such functions were found by Yaffet:[5]

$$\psi_{1,2} = e^{i\mathbf{kr}} u_{1,2}(\mathbf{k}, \mathbf{r}),$$

$$u_{1,2} = \{u_0(r) + i\mathbf{k}\mathbf{r}\,u_1(r) \pm C_1 [\mathbf{kr}]_z u_2(r)\}\,\chi_{1/2,\,-1/2} + C_2 [\mathbf{kr}]_{x \pm iy} u_3(r)\,\chi_{-1/2,\,1/2}, \quad (1)^*$$

where the indices 1 and 2 refer to the electron whose spin is directed along and against the constant magnetic field H_0, respectively; the direction of the field is taken as the z axis; $u_1(r) \sim u_2(r) \sim u_3(r)$; $C_1 \sim C_2 \sim \Delta g$, where Δg is the difference between the g factors of the electron in the lattice and the free electron; $\mathbf{k}$ is the wave vector of the electron; $\chi_{1/2}$ and $\chi_{-1/2}$ are the eigenfunctions of the Pauli operator $\hat{\sigma}_z$; the expression for $u(\mathbf{k}, \mathbf{r})$ is reduced to a single cell.

It is clear that in turning on the interaction of the electron with the sound

$$\hat{U} = \sum_n \nabla V(\mathbf{r} - \mathbf{R}_n)\,\mathbf{A} \exp\{i\mathbf{q}\mathbf{R}_n + i\omega t\} \quad (2)$$

there will occur transitions between states 1 and 2. The state of the electron gas described by the one-particle density operator $\hat{f}$ satisfying the equation

$$\partial\hat{f}/\partial t - [\hat{\mathcal{H}}_0 + \hat{U}, \hat{f}] + (\partial\hat{f}/\partial t)_{\text{collis}} = 0,$$

can, if we use the function (1) in the transition to the matrix representation, determine the resonance magnetization of the electron gas.

We note that in place of the interaction potential

*$[\mathbf{kr}] = \mathbf{k} \times \mathbf{r}$.

(2) we can use another potential $\hat{U}'$, which is so defined that the matrix elements of the commutator $[\hat{U}', \hat{f}]$ in the plane wave representation ($\psi_{1,2} = e^{i\mathbf{k}\cdot\mathbf{r}}\chi_{1/2,-1/2}$) are identical with the matrix elements of the previous representation. It is not difficult to show that

$$\hat{U}' = 4C_2 A\hat{\sigma}_n k_l q_i e_j \lambda_{mij}\varepsilon_{lmn}\exp\{i\mathbf{qr} + i\omega t\},$$

$$\lambda_{mij} = \int d\mathbf{r}\, u_0(r)\, u_1(r)\, r_m \sum_n (\mathbf{R}_n - \mathbf{r})_i \nabla_j V(\mathbf{R}_n - \mathbf{r}), \quad (3)$$

where ϵ_{lmn} is the asymmetric unit tensor, A the amplitude of the sound wave, e the polarization vector of the sound, $q = \omega/s$ the wave vector of the sound, s the sound velocity, and λ a quantity of the order of the product of the lattice constant by the mean energy of the electron in the lattice; the integral is taken over the volume of the elementary cell. It is clear from (3) that the contribution to resonance will be made both by longitudinal and transverse components of the sound wave. As will be shown from what follows, inasmuch as the resonance will exist only in weak fields H_0, when one can neglect the quantization of the electron orbits for the determination of the resonance magnetization of the metal, one can use the density operator which is quantized only in the spin of the electrons.

3. The kinetic equation which $\hat{f}$ satisfies is then written in the form[6]

$$\partial\hat{f}/\partial t + \mathbf{v}\nabla_r\hat{f} + \dot{\mathbf{p}}\nabla_p\hat{f} + [\mu\hat{\boldsymbol{\sigma}}(\mathbf{H}_0 + \mathbf{H}), \hat{f}] + (\partial\hat{f}/\partial t)_{\text{collis}} = 0, \quad (4)$$

where the effective magnetic field $\mathbf{H}$ is introduced with the components determined from (3) (since we can write formally $\hat{U}' = \mu\hat{\boldsymbol{\sigma}}\cdot\mathbf{H}$):

$$H_n(\mathbf{r}, t) = \alpha_n A\exp\{i\mathbf{qr} + i\omega t\},$$
$$\alpha_n = 4\mu^{-1}C_2 k_l q_i \lambda_{mij} e_j \varepsilon_{lmn}. \quad (5)$$

The collision integral, as in reference 6, will be assumed to be equal to $(\hat{f} - \bar{\hat{f}})/\tau + (\bar{\hat{f}} - f_0)/T_{ff}$, where τ is the time of free flight of the electron in the lattice, T_{ff} is the paramagnetic relaxation time, $\bar{\hat{f}}$ is the average of the operator $\hat{f}$ over the bounding Fermi surface and

$$\hat{f}_0 = \begin{pmatrix} f_0^+ & 0 \\ 0 & f_0^- \end{pmatrix}, \qquad f_0^{\pm} = \left[\exp\left\{\frac{\varepsilon \pm \mu H_0 - \varepsilon_0}{T}\right\} + 1\right]^{-1}.$$

The magnetization is determined by the expression

$$\mathbf{M} = \mu(2\pi\hbar)^{-3}\int d\mathbf{p}\,\mathrm{Sp}(\hat{\boldsymbol{\sigma}}\hat{f}).$$

Decomposing $\hat{f}$ over the complete set of matrices consisting of the matrices $\hat{\boldsymbol{\sigma}}$ and the unit matrix $\hat{I}$ ($\hat{f} = \hat{f}_I\hat{I} + \hat{\mathbf{f}}\cdot\hat{\boldsymbol{\sigma}}$) and making the substitution $\mathbf{f} = -\mu H_0 f_0'(\epsilon)(\mathbf{n} - \mathbf{w})$, we get for $\mathbf{w}$ the equation

$$\dot{\mathbf{w}} + \mathbf{v}\nabla_r\mathbf{w} + (e/c)[\mathbf{v}\mathbf{H}_0]\nabla_p\mathbf{w} + \mathbf{w}/\tau^* = \bar{\mathbf{w}}/\tau + [(\mathbf{n} - \mathbf{w})(\Omega_0 + \Omega)],$$
$$\mathbf{M} = \chi H_0(\mathbf{n} - \bar{\mathbf{w}}). \quad (6)$$

Here $1/\tau^* = 1/\tau + 1/T_{ff}$, $\mathbf{n} = \mathbf{H}_0/H_0$, $\Omega_0 = \hbar^{-1}g\mu\mathbf{H}_0$, $\Omega = \hbar^{-1}g\mu\mathbf{H}(\mathbf{r}, t)$, and χ is the paramagnetic susceptibility of the metal.

We can neglect the term $(e/c)[\mathbf{v}\times\mathbf{H}_0]\nabla_p\mathbf{w}$ in Eq. (6) since in weak fields H_0 (when the radius of the Larmor orbit is much greater than the mean free path of the electron) the character of the motion of the electron is determined only by the collisions. Transforming to the combinations $w = w_x + iw_y$ and $\Omega = \Omega_x + i\Omega_y$, we get from (6)

$$\dot{w} + \mathbf{v}\nabla w + w/\tau^* - i\Omega_0 w = \bar{w}/\tau + i\Omega(1 - w_z), \quad (7)$$

$$\dot{w}_z + \mathbf{v}\nabla w_z + w_z/\tau^* = \bar{w}_z/\tau - \mathrm{Im}(w^*\Omega). \quad (8)$$

If the resonance is far from saturation, then $w_Z \ll 1$, and, consequently, we can neglect the value of w_Z in Eq. (7). The solution of Eq. (7) is sought in the form $w(\mathbf{r}, t) = w\exp\{i\mathbf{q}\cdot\mathbf{r} + i\omega t\}$. Here w_Z is obviously independent of $\mathbf{r}$ and t. We then get from (7) and (8)

$$w = \left\{\overline{\frac{i\Omega\tau_1}{1 + i\mathbf{qv}\tau_1}}\left(\frac{\tau}{\tau_1} - \overline{\frac{1}{1 + i\mathbf{qv}\tau_1}}\right)^{-1} + i\Omega\tau_1\right\}\frac{1}{1 + i\mathbf{qv}\tau_1}, \quad (9)$$

$$\bar{w} = \overline{\frac{i\Omega\tau}{1 + i\mathbf{qv}\tau_1}}\left(\frac{\tau}{\tau_1} - \overline{\frac{1}{1 + i\mathbf{qv}\tau_1}}\right)^{-1}, \quad (10)$$

$$\bar{w}_z = T_{ff}\,\mathrm{Im}(\overline{w^*\Omega}), \qquad 1/\tau_1 = 1/\tau + 1/T_{ff} + i(\omega - \Omega_0). \quad (11)$$

The bar denotes averaging over the bounding Fermi surface.

It is obvious that the resonance will exist only under the condition $\mathbf{q}\cdot\mathbf{v}\tau \ll 1$ (we note that $\tau_1 \approx \tau$, since $\Delta\omega\tau \ll 1$). Here

$$\overline{(1 + i\mathbf{qv}\tau_1)^{-1}} \approx 1 - \tfrac{1}{2}\overline{(\mathbf{qv}\tau)^2} \text{ and } \overline{i\Omega\tau(1 + i\mathbf{qv}\tau_1)^{-1}} \approx \overline{\Omega\tau(\mathbf{qv}\tau)}$$

since $\bar{\Omega} = M0$ by virtue of the linear dependence of Ω on the momentum of the electron). Thus, by neglecting the nonresonant term in (9), we get

$$w = [\overline{\Omega\mathbf{qv}}\tau/(i\Delta\omega + \Gamma)][1 + i\mathbf{qv}\tau]^{-1}, \quad (12)$$

$$\bar{w} = \overline{\Omega\mathbf{qv}}\tau/(i\Delta\omega + \Gamma), \quad (13)$$

$$\bar{w}_z = |\overline{\Omega\mathbf{qv}}|^2 T_{ff}\tau^2\Gamma/(\Delta\omega^2 + \Gamma^2). \quad (14)$$

Here $\Gamma = 1/T_{ff} + \tfrac{1}{2}\overline{(\mathbf{q}\cdot\mathbf{v})^2}\tau$ = width of the resonance line.

The resonance will be significant if $\Gamma \ll \omega$ or $1/\omega T_{ff} + \omega(v/s)^2\tau \ll 1$ (since $\overline{(\mathbf{q}\cdot\mathbf{v})^2}\tau \sim \omega^2(v/s)^2\tau$),

i.e., the resonant frequencies should satisfy the equation

$$T_{ff}^{-1} \ll \omega \ll \tau^{-1} (s/v)^2.$$

It then follows that we need as large a T_{ff} as possible and as small a τ as possible for the existence of resonance. For example, in lithium and beryllium, where T_{ff} depends only on the presence of impurities, for proper purity of the metal one can obtain $T_{ff} \gtrsim 10^{-7}$ sec. At higher temperatures (when $\tau \sim 10^{-13} - 10^{-14}$ sec) a resonance in these metals will be observed in a region of frequencies $\omega \sim 10^8 - 10^9$ sec^{-1}.

From what has been said, the impossibility of polarization of nuclei in spin acoustic resonance immediately follows, inasmuch as strong magnetic fields and low temperatures are required for the existence of polarization, which contradicts the conditions of resonance. Moreover, as is easily established by numerical estimation, $w_Z \ll 1$ always, while it is necessary that $w_S \sim 1$ to achieve appreciable polarization.

4. The coefficient of absorption of ultrasonic energy in spin acoustic resonance is determined in the usual way:

$$\gamma = |T\dot{S}|_{ff} / sE_{so},$$

where T is the temperature of the specimen; $E_{SO} = \frac{1}{4}\rho\omega^2 A$ is the energy of the sound wave;

$$S = (2\pi\hbar)^{-3} \int dp \, \mathrm{Sp} \{(1 - \hat{f}) \ln (1 - \hat{f}) + \hat{f} \ln \hat{f}\}$$

is the entropy of the electron gas. In order to carry out the operation Sp, we transform to the diagonal representation of the matrix of the operator f:

$$\hat{f} = \begin{pmatrix} f_{11} & f_{12} \\ f_{21} & f_{22} \end{pmatrix} \to \begin{pmatrix} f_1 & 0 \\ 0 & f_2 \end{pmatrix},$$

$$f_{1,2} = f_{11} \mp \left\{ \frac{f_{11} - f_{22}}{2} + \left[\left(\frac{f_{11} - f_{22}}{2} \right)^2 + |f_{12}|^2 \right]^{1/2} \right\}.$$

Noting $f = f_I \hat{I} + \mathbf{f} \cdot \boldsymbol{\sigma}$, we obtain the result that $f_{11} + f_{22} = 2f_I$, $f_{11} - f_{22} = 2f_Z$ and $f_{12} = f_X + if_Y$. Therefore, $f_{1,2} = f_I \mp f$, where $f = |\mathbf{f}|$. Solution of the equation for f_I is usually sought in the form $f_0 + f'$, where f' is the addition to the equilibrium distribution function. Taking this fact into account, we get

$$T\dot{S} = T (2\pi\hbar)^{-3} \int dp \left[(\dot{f} - \dot{f}') \ln \frac{1 - f_I + f}{f_I - f} + (\dot{f} + \dot{f}') \ln \frac{f_I + f}{1 - f_I - f} \right].$$

Thence, after expansion in f and f', we have for the spin part of the dissipation function

$$(T\dot{S})_{ff} = 2T (2\pi\hbar)^{-3} \int dp \, f\dot{f} \left[\frac{\ln (1 - f_0)}{1 - f_0} + \frac{\ln f_0}{f_0} \right].$$

Close to the bounding Fermi surface, $\ln f_0 \approx \ln (1 - f_0)$. Noting that $T^{-1}(1 - f_0) f_0 = \partial f_0 / \partial \epsilon$, we get [after substitution $f = -\mu H_0 f_0' (\mathbf{n} - \mathbf{w})$],

$$(T\dot{S})_{ff} = 2 \ln 2 \cdot \chi H_0^2 \left(\frac{\overline{w}_z - \overline{|w|^2}}{T_{cn}} + \frac{\overline{|w|^2} - \overline{|w|^2}}{\tau} \right),$$

where $\overline{w}_Z$, w and $\overline{w}$ are the solutions of Eqs. (7) and (8).

Making use of Eqs. (12) — (14), we obtain the following expression for the coefficient of absorption of ultrasonic energy at spin acoustic resonance:

$$\gamma = 12 \ln 2 \cdot \frac{\Omega_0^2}{\omega^2} \frac{\chi \tau^3 \overline{(qv)^2}}{\rho s} \frac{\overline{|\alpha q v|^2}}{\Delta\omega^2 + \Gamma^2},$$

where α is determined by Eq. (5). In order of magnitude, $\alpha q \cdot v \sim 4\mu^{-1} \Delta g q^2 k a \epsilon v$ (a is the lattice constant, ϵ and v are the energy and velocity of the electron on the bounding Fermi surface, $\Delta g \sim 10^{-3} - 10^{-4}$). Numerical estimates under conditions in which there is resonance lead to values $\gamma \lesssim 10^{-5}$ cm^{-1} close to resonance.

[1] S. A. Al'tshuler, JETP **28**, 38 and 49 (1955), Soviet Phys. JETP **1**, 29 and 37 (1955).

[2] A. Overhauser, Phys. Rev. **92**, 411 (1953); **89**, 689 (1953).

[3] A. Kastler, J. phys. radium **15**, 300 (1954).

[4] R. J. Elliott, Phys. Rev. **96**, 266 (1954).

[5] Y. Yaffet, Phys. Rev. **85**, 478 (1952).

[6] Azbel', Gerasimenko, and Lifshitz, JETP **32**, 1212 (1957), Soviet Phys. JETP **5**, 986 (1957).

Translated by R. T. Beyer
89