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Dynamics of Ultrasonic Propagation in Metals with Paramagnetic Impurities

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A theory of acoustic magnetic resonance on paramagnetic impurities in metals is developed. Spin relaxation in an instantaneous local field is taken into account in the equations of motion. Expressions are obtained for the magneto-elastic dynamic susceptibility, which describes the appearance of the magnetic moment, and also for the elastic-elastic dynamic susceptibility, which describes the appearance of an induced elastic moment under the action of sound deformation, and which determines the sound absorption coefficient. Various mechanisms of spin-phonon interaction are discussed: modulation of the impurity g factor, electric quadrupole interaction between the ions and lattice, and modulation of two-particle interactions between the impurities. Preliminary estimates show that the energy absorption coefficient of the acoustic deformation field should be large for a system of paramagnetic impurities in metals.

THE increased interest in the investigation of electron paramagnetic resonance (EPR) on paramagnetic impurities in metals is due to the fact that it gives information on the exchange interaction of the paramagnetic impurities with the conduction electrons. Since the sound deformations modulate the constant of exchange interactions, one can expect that the spectra of acoustic EPR contain more information on the exchange than ordinary EPR.

In spite of the fact that attempts have been reported in the literature to analyze spin-acoustic resonance on conduction electrons in paramagnetic metals,[1] the problem of acoustic resonance on paramagnetic impurities has not been touched on.

In the present work, we develop a quantum mechanical theory of acoustic paramagnetic resonance on impurities in metals, including the spin relaxation in the instantaneous local field in the equations of motion of the spins.[2,3] In such an approach, the induced magnetic moment relaxes not to the equilibrium direction of the magnetic moment, which is determined by the external static magnetic field, but to some new direction of the effective field, which rotates with the frequency of the external alternating field. The direction of this effective field is determined as the vector sum of the external static magnetic field, the molecular field, which is defined by exchange with the conduction electrons, and the field induced at each moment of time by the external field of the static sound deformations. Exact account of relaxation in an instantaneous local field leads to the result that the coefficient of resonance absorption of the sound by a quantum system with a decrete spectrum consists of two terms—the usually considered absorption term, which is maximal at exact resonance and describes the sound absorption due to resonance transitions in the quantum system, and the "dispersion" term, which has not been studied to date, but which can be essential for a certain shift of the sound frequency from resonance.

In the first part of the work, we shall discuss the possible mechanisms of the energy transfer of the field of acoustic deformations of the impurity paramagnetic system; in the second part we briefly describe the method of investigation of the dynamics of a spin system consisting of paramagnetic ions and conduction electrons. We present the results of calculations of the dynamic susceptibility for all the mechanisms of excitation of the paramagnetic system analyzed above. In the last part of the work, concrete estimates are made of the sound absorption coefficient. For the calculations, we use a system of units in which $\hbar = \mu_B = k = 1$, where $\hbar$ is Planck's constant, μ_B the Bohr magneton, and k Boltzmann's constant.

1. EXCITATION OF THE SPIN SYSTEM

We now consider the most effective mechanisms of sound interaction with the paramagnetic impurities, and also with the conduction electrons. The latter is necessary because the interaction of the sound with the conduction electrons by means of exchange indirectly affect the state of the paramagnetic impurities.

Investigations of the spin-phonon interaction of the conduction electrons[4] show that it is due essentially to spin-orbit coupling. The corresponding Hamiltonian has the form:

$$\mathscr{H}_{e-ph} = \sum_{ijkl} T_{ijkl}\sigma_i H_j \epsilon_{kl}(t), \tag{1}$$

where T_{ijkl} is the electron-phonon interaction tensor, H_j the external magnetic field, σ_i the Pauli matrices, $\epsilon_{kl}(t)$ the relative deformation tensor. For simple metals with a spherical Fermi surface, the tensor T_{ijkl} from (1) is described by two constants T_{xxxx} and T_{yzyz}.[5]

In accord with existing experimental data, the following mechanisms of spin-phonon interactions in metals are important for paramagnetic impurities. The first of these is sound modulation of the g factor of the impurity. Its phenomenological form is well known:[5]

$$\mathscr{H}^{\alpha}_{s-ph} = \sum_{ijkl} G_{ijkl} S_i^{\alpha} H_j \epsilon_{kl}(t), \tag{2}$$

where S_i^{α} are the components of the spin operator of the α-th localized magnetic moment and G_{ijkl} the spin-phonon interaction tensor.

The second is the mechanism due to electron quadrupole interaction of the ions with the lattice. In this case, as is well known,[5] the Hamiltonian of the spin-phonon interaction is represented in the form

$$\mathscr{H}_{s-ph}^{\alpha}=\sum_{ijkl}G_{ijkl}S_i^{\alpha}S_j^{\alpha}\varepsilon_{kl}(t). \qquad (3)$$

The next of the possible mechanisms describes the two-particle interaction:

$$\mathscr{H}_{s-ph}^{ij}=\sum_{\alpha\beta\gamma\delta}\mathscr{P}_{\alpha\beta\gamma\delta}^{ij}S_{\alpha}{}^{i}S_{\beta}{}^{j}\varepsilon_{\gamma\delta}(t). \qquad (4)$$

In contrast with dielectrics, in which the operator (4) describes the Waller-Al'tshuler mechanism (the modulation of dipole-dipole and exchange interactions by sound), a unique mechanism is effective in metals, namely the super-exchange of ions by means of a system of conduction electrons. This mechanism acts at great distances. If there is a substitution atom in the matrix of the metal, with a valence different from the valence of the matrix atoms, then the conduction electrons naturally experience excitation in the vicinity of the impurity atom. The appearance of a screening charge around the impurity is accompanied by a phase shift η_l in the l-th spherical components of the electron wave functions. If the conditions here are such that a localized magnetic moment can develop,[6] then the phase shifts are seen to be different for electrons with opposite spins, which leads to the generation of oscillations of the spin density in the host matrix of the metal, in addition to the oscillations of the charge density. As a consequence, magnetic interaction between the impurities becomes possible at large distances.

Last in our consideration is the mechanism due to sound modulation of hyperfine and superhyperfine interactions. The Hamiltonian here has the following form:

$$\mathscr{H}_{s-ph}^{ij}=\sum_{\alpha\beta\gamma\delta}\mathscr{F}_{\alpha\beta\gamma\delta}^{ij}S_{\alpha}{}^{i}I_{\beta}{}^{j}\varepsilon_{\gamma\delta}(t); \qquad (5)$$

where I_{β}^{j} are the spin operators of the j-th nucleus.

If we choose the z axis, defined by the external static magnetic field H_{03}, as our axis of quantization, then, as easily seen from the symmetry properties of the Hamiltonian (1), the transitions can be brought about only by the deformations $\epsilon_5(t)$ and $\epsilon_4(4)$ (in the Vogt notation), which imposes definite limitations on (2)–(5).

2. THE MAGNETO-ELASTIC SUSCEPTIBILITY OF THE SPIN SYSTEM

The dynamics of a spin system excited by sound can be described by means of two dynamic susceptibilities, magneto-elastic and elastic-elastic. The first of these determines the value of the magnetic-moment components due to the sound deformations. The second determines elastic-moment components due to the sound deformations, and also the absorption coefficient of the energy of the field of acoustic deformations by the spin system. In the present portion of the paper, we shall briefly explain the method of investigation of the dynamics of the spin system using as an example the calculation of the magneto-elastic susceptibility and of the transverse components of the magnetic moment.

We consider some metal in whose crystal lattice there are randomly distributed paramagnetic impurities with a concentration c = n/N, where n is the number of impurities per unit volume and N the number of lattice sites in the matrix of the metal lattice in this same volume. We assume that the sample is placed in a magnetic field. Then the Hamiltonian of the system of conduction electrons and localized spins of the magnetic impurities can be represented in the following way:

$$\begin{gathered}\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1,\\ \mathscr{H}_0=\sum_j\omega_sS_j{}^z+\frac{1}{2}\omega_e\sigma_0{}^z+\sum_{p,\lambda}(\varepsilon_{p,\lambda}-\varepsilon_F)\overset{+}{a}_{p,\lambda}a_{p,\lambda},\\ \mathscr{H}_1=-\frac{J}{N}\sum_{q,j}e^{-i\mathbf{qR}_j}\mathbf{S}_j\boldsymbol{\sigma}_q,\end{gathered} \qquad (6)$$

where $\mathbf{S}_j$ is the spin vector of the localized impurity: σ_q the spin vector of the conduction electron with wave vector $\mathbf{q}$; the index λ is equal either to +(↑) for conduction electrons whose spins are oriented with the field, or −(↓) for conduction electrons with opposite orientation of spin; J is a constant characterizing the exchange interaction of the conduction electrons with the paramagnetic impurity. Further, $\omega_s = -g_sH_{03}$, $\omega_e = -g_eH_{03}$; g_s and g_e are the g factors of the paramagnetic impurities and the conduction electrons, respectively.

We further introduce the operator **M** of the mean localized spin and the operator m for the conduction electron in the following way:

$$\langle\delta S^{\alpha}(t)\rangle_{ext}=-i\int_{-\infty}^{t}dt'\langle[S^{\alpha}(t),\mathscr{H}_{ext}(t')]\rangle, \qquad (7)$$

We also introduce the canonical mean values $R = \langle S_j^z\rangle$ and $\xi = \langle\sigma_0^2\rangle/2N$.

Then, following Kubo,[7] and assuming that the external excitation is applied adiabatically at the instant of time t = −∞ (in our case, the external acoustic field of the deformation), the change in the mean spin component is determined in the linear approximation by the formula

$$M^{\alpha}=\frac{g_s}{N}\sum_jS_j{}^{\alpha},\ m^{\alpha}=\frac{g_e}{2N}\sigma_0{}^{\alpha}. \qquad (8)$$

where $S^{\alpha}(t)$ is the projection of the spin operator on the α axis at the time t, and $\mathscr{H}_{ext}(t')$ is the Hamiltonian of the external excitation at the time t′.

Since the quantity $\langle\delta S^{\alpha}(t)\rangle_{ext}$ represents the difference between the mean value $\langle S^{\alpha}(t)\rangle_{ext}$ at the time t with account of the effect of $\mathscr{H}_{ext}(t')$ and the mean value $\langle S^{\alpha}(t)\rangle$ without account of the effect of $\mathscr{H}_{ext}(t')$, we have

$$\langle\delta S^{\alpha}(t)\rangle_{ext}=\langle S^{\alpha}(t)\rangle_{ext}-\langle S^{\alpha}(t)\rangle.$$

In the absence of energy dissipation, the dynamics of the spin system are described by the Heisenberg equations of motion

$$i\frac{\partial}{\partial t}S_j{}^{\alpha}(t)=[S_j{}^{\alpha}(t),\mathscr{H}],\quad i\frac{\partial}{\partial t}\sigma_q{}^{\alpha}(t)=[\sigma_q{}^{\alpha}(t),\mathscr{H}]. \qquad (9)$$

If we now include the damping of the α-th component of the spin operators, we then obtain the following two equations:

$$i\frac{\partial}{\partial t}M^\alpha=[M^\alpha,\mathscr{H}]-i\Delta_s{}^\alpha\{M^\alpha-\chi_{sM}(H_{03}\delta_{\alpha3}+\lambda m^\alpha)$$
$$-\chi_{s3}[\varepsilon^5(t)\delta_{\alpha+4,5}+\varepsilon^4(t)\delta_{\alpha+2,4}]\},$$
$$i\frac{\partial}{\partial t}m^\alpha=[m^\alpha,\mathscr{H}]-i\Delta_e{}^\alpha\{m^\alpha-\chi_{eM}(H_{03}\delta_{\alpha3}+\lambda M^\alpha)$$
$$-\chi_{e3}[\varepsilon^5(t)\delta_{\alpha+4,5}+\varepsilon^4(t)\delta_{\alpha+2,4}]\},\tag{10}$$

where Δ_s^{-1} and Δ_e^{-1} are the damping times of the spin components of the paramagnetic impurities and the conduction electrons, respectively, χ_{sM} and χ_{eM} are the magnetic static susceptibilities, χ_{s3} and χ_{e3} the magnetic-deformation static susceptibilities and $\lambda = 2J(g_e g_s)^{-1}$ the constant of the molecular field.[8]

Then, if we limit ourselves to the molecular field approximation, corresponding to the decoupling

$$\langle\sigma_q{}^zS_j{}^-\rangle\approx\langle\sigma_q{}^z\rangle\langle S_j{}^-\rangle,\quad\langle\sigma_q{}^-S_j{}^z\rangle\approx\langle\sigma_q{}^-\rangle\langle S_j{}^z\rangle,\tag{11}$$

and recognize that in the linear approximation we have for the stationary case ($t \gg \Delta^{-1}$)

$$\chi_{sM}=-\frac{g_s{}^2cR}{\omega_s{}'},\quad\chi_{eM}=-\frac{g_e{}^2\xi}{\omega_e{}'},$$
$$\omega_s{}'=\omega_s-2J\xi,\quad\omega_e{}'=\omega_e-2cJR,\tag{12}$$

it is easy to obtain from (10)

$$\langle M^-(\omega)\rangle=i\varepsilon^-(\omega)\left\{(\omega-\omega_e{}'+i\Delta_e)\Delta_s\chi_{s3}+2\Delta_e\chi_{e3}\frac{g_s}{g_e}Jc\tilde{R}\right\}D^{-1}(\omega),$$
$$\langle m^-(\omega)\rangle=i\varepsilon^-(\omega)\left\{(\omega-\omega_s{}'+i\Delta_s)\Delta_e\chi_{e3}+2\Delta_s\chi_{s3}\frac{g_e}{g_s}J\tilde{\xi}\right\}D^{-1}(\omega),$$
$$D(\omega)=(\omega-\omega_s{}'+i\Delta_s)(\omega-\omega_e{}'+i\Delta_e)-4J^2c\tilde{\xi}\tilde{R},\tag{13}$$
$$\tilde{\xi}=\xi\frac{\omega_e{}'-i\Delta_e}{\omega_e{}'},\quad\tilde{R}=R\frac{\omega_s{}'-i\Delta_s}{\omega_s{}'};$$

here, cubic symmetry of the crystal and a spherical Fermi surface are assumed, and $\epsilon^-(t) = \epsilon_5(t) - i\epsilon_4(t)$.

The shifts in the g factors and the widths of the lines, computed in second order of decoupling in the constant of exchange interaction,[9] do not depend on the method of excitation of the spin system, and are determined only by their internal characteristics (the value of J, the electron state density, width of the conduction band); we shall limit ourselves therefore to the first order in J.

We consider now the second half of the Kubo equation. For this purpose we need the Hamiltonian of interaction of the spin system with the field of acoustic deformations. Limiting ourselves to cubic symmetry and using (1) and (2), we get

$$\mathscr{H}_{ext}=\frac{N}{2}\frac{H_{03}}{g_eg_s}\{[g_eG_{44}M^++g_sT_{44}m^+]\varepsilon^-(t)+\text{h.c.}\}.\tag{14}$$

Further, substituting this Hamiltonian in (8), we get the result that the magnetization of the localized spins and the spins of the conduction electrons will be

$$\langle M^-(\omega)\rangle_{ext}=\langle M^-(\omega)\rangle+\frac{\omega_s}{2N}[G_{44}F_R(\omega)+T_{44}B_R(\omega)]\varepsilon^-(\omega),$$
$$\langle m^-(\omega)\rangle_{ext}=\langle m^-(\omega)\rangle+\frac{\omega_e}{2N}[T_{44}K_R(\omega)+G_{44}\tilde{B}_R(\omega)]\varepsilon^-(\omega).\tag{15}$$

Here $F_R(\omega)$, $B_R(\omega)$, $\tilde{B}_R(\omega)$ and $K_R(\omega)$ denote four propagators of the following sort:

$$G_{AB}{}^r(t)=i\theta(t)\langle[A(t),B(0)]\rangle\equiv\langle\langle A(t)|B\rangle\rangle,$$

$$F_R(\omega)=\sum_{ij}\langle\langle S_i{}^-|S_j{}^+\rangle\rangle_\omega,\quad B_R(\omega)=\frac{1}{2}\sum_i\langle\langle S_i{}^-|\sigma_0{}^+\rangle\rangle_\omega,$$

$$\tilde{B}_R(\omega)=\frac{1}{2}\sum_j\langle\langle\sigma_0{}^-|S_j{}^+\rangle\rangle_\omega,\quad K_R(\omega)=\frac{1}{4}\langle\langle\sigma_0{}^-|\sigma_0{}^+\rangle\rangle_\omega.\tag{16}$$

Consideration of the equations of motion for these propagators and the use of the decoupling procedure (11) lead to the following set of equations:

$$\begin{aligned}(\omega-\omega_s{}'+i\Delta_s)F_R(\omega)&=2cNR+2cJ\tilde{R}\tilde{B}_R(\omega),\\(\omega-\omega_e{}'+i\Delta_e)\tilde{B}_R(\omega)&=2J\xi F_R(\omega),\\(\omega-\omega_s{}'+i\Delta_s)B_R(\omega)&=2cJ\tilde{R}K_R(\omega),\\(\omega-\omega_e{}'+i\Delta_e)K_R(\omega)&=2N\xi+2J\tilde{\xi}B_R(\omega).\end{aligned}\tag{17}$$

The system breaks up into two independent subsystems, solving which we get

$$\begin{aligned}F_R(\omega)&=2cNR(\omega-\omega_e{}'+i\Delta_e)D^{-1}(\omega),\\K_R(\omega)&=2N\xi(\omega-\omega_s{}'+i\Delta_s)D^{-1}(\omega),\\B_R(\omega)&=4JcN\xi\tilde{R}D^{-1}(\omega),\\\tilde{B}_R(\omega)&=4JcN\tilde{\xi}RD^{-1}(\omega).\end{aligned}\tag{18}$$

We now introduce the notation $a_s = \omega - \omega_s' + i\Delta_s$, $a_e = \omega - \omega_e' + i\Delta_e$ and define the transverse dynamic magneto-elastic susceptibility in the form

$$\langle M^-(\omega)\rangle_{ext}=\chi_s{}^-(\omega)\varepsilon^-(\omega),\quad\langle m^-(\omega)\rangle_{ext}=\chi_e{}^-(\omega)\varepsilon^-(\omega);$$

we then get

$$\chi_s{}^-(\omega)=[2Jc\tilde{R}g_sg_e{}^{-1}(T_{44}\xi\omega_e+i\Delta_e\chi_{e3})+a_e(G_{44}cR\omega_s+i\Delta_s\chi_{s3})]D^{-1}(\omega),$$
$$\chi_e{}^-(\omega)=[a_s(T_{44}\xi\omega_e+i\Delta_e\chi_{e3})+2\tilde{J}\xi g_eg_s{}^{-1}(G_{44}cR\omega_s+i\Delta_s\chi_{s3})]D^{-1}(\omega).\tag{19}$$

We now consider a Hamiltonian of the type (3). Experiments on nuclear acoustic resonance in metals have shown that the mechanism of interaction of the crystalline field with the quadrupole moment is effective both in dielectrics and in metals; therefore, there is a basis for supposing that this mechanism is also effective for paramagnetic ions. The Hamiltonian which takes the deformations $\epsilon_4(t)$ and $\epsilon_5(t)$ and the transitions $\Delta m = \pm 1$ into account has the form

$$\mathscr{H}_{s-ph}=\frac{1}{4}G_{44}\sum_j\{[S_j{}^-,S_j{}^z]_+\varepsilon^+(t)+\text{э. с.}\};\tag{20}$$

here $[\,,]_+$ denotes the anticommutator. Making use of (7) and (8) we obtain in place of (15)

$$\langle M^-(\omega)\rangle_{ext}=\langle M^-(\omega)\rangle+\frac{\omega_s}{2N}\left\{T_{44}B_R(\omega)+\frac{G_{44}}{2H_{03}}\left[F_R(\omega)\right.\right.$$
$$\left.\left.+2\sum_{k,j}\langle\langle S_k{}^-|S_j{}^+S_j{}^z\rangle\rangle_\omega\right]\right\}\varepsilon^-(\omega),$$
$$\langle m^-(\omega)\rangle_{ext}=\langle m^-(\omega)\rangle+\frac{\omega_e}{2N}\left\{T_{44}K_R(\omega)+\frac{G_{44}}{2H_{03}}\left[\tilde{B}_R(\omega)\right.\right.$$
$$\left.\left.+\sum_j\langle\langle\sigma_0{}^-|S_j{}^+S_j{}^z\rangle\rangle_\omega\right]\right\}\varepsilon^-(\omega).\tag{21}$$

By considering the commutation relations of the spin operators $[S_j^z, S_k^+]_- = S_j^+\delta_{jk}$, it is easy to obtain

$$\sum_{kj}\langle\langle S_k{}^-|S_j{}^+S_j{}^z\rangle\rangle_\omega=D^{-1}(\omega)cN[3\langle S_z{}^2\rangle-\langle S_z\rangle-S(S+1)]a_e,$$
$$\frac{1}{2}\sum_j\langle\langle\sigma_0{}^-|S_j{}^+S_j{}^z\rangle\rangle_\omega=D^{-1}(\omega)2J\tilde{\xi}cN[3\langle S_z{}^2\rangle-\langle S_z\rangle-S(S+1)].\tag{22}$$

For the magneto-elastic susceptibility, we get from (21) and (22):

$$\begin{aligned}\chi_s{}^-(\omega)&=D^{-1}(\omega)\{2Jc\tilde{R}g_sg_e{}^{-1}(T_{44}\xi\omega_e+i\Delta_e\chi_{e3})\\&\quad-a_e[{}^1\!/_2g_sG_{44}c(3\langle S_z{}^2\rangle-S(S+1))-i\Delta_s\chi_{s3}]\},\\\chi_e{}^-(\omega)&=D^{-1}(\omega)\{a_s(T_{44}\xi\omega_e+i\Delta_e\chi_{e3})-\\&\quad-2J\tilde{\xi}g_eg_s{}^{-1}[{}^1\!/_2g_sG_{44}c(3\langle S_z{}^2\rangle-S(S+1))-i\Delta_s\chi_{s3}]\}.\end{aligned}\tag{23}$$

If we assume that each impurity ion interacts only with a single neighbor j, then only the single term P_{44}^{ij} is important in the Hamiltonian (4), and we obtain

$$\chi_s^-(\omega) = D^{-1}(\omega)\{2Jc\tilde{R}g_s g_e^{-1}(T_{44}\xi\omega_e + i\Delta_e\chi_{e3}) - a_e(\mathscr{P}_{44}c^2R^2g_s - i\Delta_s\chi_{s3})\},$$

$$\chi_e^-(\omega) = D^{-1}(\omega)\{a_s(T_{44}\omega_e\xi + i\Delta_e\chi_{e3}) - 2J\tilde{\xi}g_e g_s^{-1}(2\mathscr{P}_{44}c^2R^2g_s - i\Delta_s\chi_{s3})\}. \tag{24}$$

For the interaction described by the Hamiltonian (5), by assuming

$$A_i = \sum_j \mathscr{F}_{44}^{ij}\langle I_z^j\rangle = \text{const},$$

we obtain

$$\chi_s^-(\omega) = D^{-1}(\omega)\{2Jc\tilde{R}g_s g_e^{-1}(T_{44}\xi\omega_e + i\Delta_e\chi_{e3}) - a_e(Ag_s cR - i\Delta_s\chi_{s3})\},$$

$$\chi_e^-(\omega) = D^{-1}(\omega)\{a_s(T_{44}\xi\omega_e + i\Delta_e\chi_{e3}) - 2J\tilde{\xi}g_e g_s^{-1}(Ag_s cR - i\Delta_s\chi_{s3})\}. \tag{25}$$

The calculated dynamic magnetoelastic susceptibilities $\chi_S^-(\omega)$ and $\chi_e^-(\omega)$ consist of two parts—imaginary and real—which describe respectively the in-phase transverse magnetization and that shifted by $\pi/2$ relative to the external field. To find the total susceptibilities χ_S and χ_e, we need to construct $\chi_e^+(\omega)$ and $\chi_S^+(\omega)$, after which, in accord with[10], and assuming that $\chi = \chi' + i\chi'$, we obtain $\chi'' = \frac{1}{2}(\chi''_- - \chi''_+)$. But the resultant expression will differ from (10) and (23)—(25) only in that it will be even relative to ω'_e and ω'_S, since the absorption effect is even in the field H_{03}. Inasmuch as the terms corresponding to frequencies will disappear in a negative magnetic field for $\omega \gg T_e^{-1}$, T_S^{-1}, it is quite natural to disregard them.

In spite of the fact that (19) and (23)—(25) are corrections in first order in J, in order to reveal the characteristic features of the behavior of the absorption curve we neglect terms that are proportional to J^2; then (19) goes over into

$$\chi_s''(\omega) = \frac{\pi}{2}G_{44}H_{03}^2\frac{\chi_{sM}(1+\lambda\chi_{eM})}{1-\lambda^2\chi_{sM}\chi_{eM}}g_s(\omega) - \frac{\pi}{2}\chi_{s3}(\omega_s' - \omega)g_s(\omega)$$
$$+ \frac{\pi^2}{2}\frac{\omega_e}{2}\Big\{T_{44}\frac{\omega_s}{2J}[T_e(\omega_e' - \omega)\omega + T_s(\omega_s' - \omega)\omega_s' + T_s^{-1}]$$
$$+ \frac{g_s}{g_e\omega_s'}\chi_{e3}[(\omega_e' - \omega)T_s^{-1} - \omega T_e^{-1} + \omega_s'(\omega_s' - \omega)(\omega_e' - \omega)T_s]\Big\} \tag{26}$$
$$\times g_s(\omega)g_e(\omega),$$

$$g_a(\omega) = \frac{T_a}{\pi}\frac{1}{1 + T_a^2(\omega_a' - \omega)^2} \quad (a = e, s).$$

It is seen from (26) that the first term in $\chi_S''(\omega)$ is due to the absorption of the energy of the field of acoustic deformations by the paramagnetic ions as a result of modulation of the g factor in some effective magnetic field

$$H_{\text{eff}} = H_{03}\sqrt{\frac{1+\lambda\chi_{eM}}{1-\lambda^2\chi_{eM}\chi_{sM}}},$$

the second term is the admixture of "dispersion" absorption due to relaxation in the instantaneous local field, and the third represents exchange with the conduction electrons (the picture is reversed in the case $\chi_e''(\omega)$). It is then completely clear that only the first term in $\chi_S''(\omega)$ from (26) will depend on the mechanism of the transfer of the energy of the acoustic deformation field to the system of paramagnetic impurity ions, i.e., we obtain in place of (23),

$$\chi_s''(\omega) = \frac{\pi}{2}cg_s[3\langle S_z^2\rangle - S(S+1)]G_{44}g_s(\omega) - \ldots \tag{27}$$

Eq. (24) goes over into

$$\chi_s''(\omega) = \frac{\pi}{2}\mathscr{P}_{44}\chi_{sM}^2H_{03}^2\frac{(1+\lambda\chi_{eM})^2}{g_s(1-\lambda^2\chi_{sM}\chi_{eM})^2}g_s(\omega) - \ldots, \tag{28}$$

and Eq. (25) becomes

$$\chi_s''(\omega) = \frac{\pi}{2}A\chi_{sM}H_{03}^2\frac{1+\lambda\chi_{eM}}{1-\lambda^2\chi_{sM}\chi_{eM}}g_s(\omega) - \ldots \tag{29}$$

The appearance of transverse magnetization of the sample, due to the action of the external variable field of sound deformations and described by Eqs. (19) and (23)—(25), can be revealed by the signal of the stimulated Bloch induction generated by the transverse components of the magnetization of the impurity ions in the outer layer of the sample. Furthermore, in the expressions for $\langle M_\pm(\omega)\rangle_{\text{ext}}$, there appear factors of the type $\sum \exp(i\mathbf{k}\cdot\mathbf{r}_j)$, where $\mathbf{k}$ is the sound wave vector and $\mathbf{r}_j$ $\mathbf{r}_j$ is the radius vector of the paramagnetic impurity. Factors of such a type vanish for an even number of half waves. Therefore, a contribution to the magnetization can be made only by a layer of the dimensions of a half wave, which also makes it more difficult to record the effect. However, in our opinion, several methods of eliminating these difficulties exist. The first is to prepare a layered sample: if $\zeta < \Lambda$ the metal layers should have a thickness ζ and should be spaced half a wave length of sound apart with the intervals between them filled with a dielectric; if $\zeta > \Lambda$, all the thicknesses and the interval are equal to Λ, where ζ is the skin depth. The use of the Abrikosov vortex lattice presents a similar possibility of eliminating regions with an even number of half waves. Further, the problem of the half wavelength layer will completely be eliminated if stimulated sound oscillations are excited in the sample at the resonance frequency, but with $k_{\text{eff}} \sim 0$.

3. ABSORPTION COEFFICIENT

As has been pointed out earlier, the coefficient for the absorption of the energy of the field of acoustic deformations by the spin system is determined by the elastic-elastic susceptibility, which causes the appearance of components of the elastic moment. Therefore, for our further calculations, we need $\langle P_\pm(\omega)\rangle_{\text{ext}}$ and $\langle p_\pm(\omega)\rangle_{\text{ext}}$, where $P_\pm(\omega)$ and $p_\pm(\omega)$ are the "elastic magnetic" moments, defined in the following way for the ions and conduction electrons, respectively:

$$P_\pm = \frac{G_{44}H_{03}}{N}\sum_j S_j^+, \qquad p_\pm = \frac{T_{44}H_{03}}{2N}\sigma_0^\pm.$$

Calculation in accord with the procedure described above leads to the result

$$\chi_s^-(\omega) = D^{-1}(\omega)\{-a_e(G_{44}^2H_{03}^2cR - i\Delta_s\chi_{s3}) - 2cJ\tilde{R}G_{44}T_{44}^{-1}(T_{44}^2H_{03}^2\xi - i\Delta_e\chi_{e3})\},$$

$$\chi_e^-(\omega) = D^{-1}(\omega)\{-2J\tilde{\xi}T_{44}G_{44}^{-1}(G_{44}^2H_{03}^2cR - i\Delta_s\chi_{s3}) - a_s(T_{44}^2H_{03}^2\xi - i\Delta_e\chi_{e3})\}. \tag{30}$$

We assume that the EPR lines for ions and conduction electrons are resolved (this is satisfied for $|\omega'_S - \omega'_e| > 2(T_e^{-1} + T_S^{-1})/\sqrt{3}$). Then we can neglect the term containing $g_S(\omega)g_e(\omega)$. This leads to the relation

$$\chi_s''(\omega) = \frac{\pi G_{44}^2H_{03}^2cN\omega_s'}{6kT}S(S+1)\left[1 + \frac{\omega_s' - \omega}{\omega_s'}\right]g_s(\omega). \tag{31}$$

Here we have taken it into account that χ_{sM}

$= cNg_S^2 S(S+1)/3T$ and $\chi_{S3} = cNG_{44}^2 H_{03}^2 S(S+1)/3T$. The sound absorption coefficient is equal to

$$\sigma_{s\mu} = \frac{\omega}{\rho v^3}\chi_s''(\omega) = \frac{\pi c N G_{44}{}^2 H_{03}{}^2 \omega^2}{6kT\rho v^3} S(S+1)\left[1 + \frac{\omega_s' - \omega}{\omega_s'}\right] g_s(\omega), \tag{32}$$

where ρ is the density of the crystal and v the speed of sound. It is seen from (32) that the "dispersion" absorption due to relaxation in an instantaneous local field in the resonance region is small (if $(\omega_S' - \omega)/\omega_S' \ll 1$). At a certain deviation from resonance, this term will cause asymmetry of the absorption curve.

We now consider in more detail the case in which the interaction of the paramagnetic impurities in the metal with the field of acoustic deformations is brought about by the modulation of the internal crystalline field, and the Hamiltonian of this interaction is quadratic in the spin. In this case, taking into account transitions with $\Delta m = \pm 1$, we need to allow for the appearance of the following quadrupole moments, viz.,

$$D_- = \frac{G_{44}}{2N}\sum_j [S_j^z, S_j^-]_+,$$

and also the "elastic magnetic" moment $p_- = T_{44}H_{03}\sigma_0^-/2N$.

Taking (8) and (16) into consideration, we can at once obtain

$$\langle p_-(\omega)\rangle_{ext} = \langle p_-(\omega)\rangle - \frac{1}{2N}\Big\{T_{44}{}^2 H_{03}{}^2 K_R(\omega) + \frac{1}{2}G_{44}T_{44}H_{03}\Big[\tilde{B}_R(\omega) + \sum_j \langle\langle \sigma_0^- | S_j^+ S_j^z\rangle\rangle_\omega\Big]\Big\}\varepsilon^-(\omega). \tag{33}$$

So far as the quadrupole moment is concerned,

$$\begin{aligned}\langle D_-(\omega)\rangle_{ext} = \langle D_-(\omega)\rangle - \frac{1}{2N}\Big\{&\frac{1}{2}G_{44}T_{44}H_{03}B_R(\omega) + \frac{1}{4}G_{44}{}^2F_R(\omega)\\ &+ \frac{1}{2}G_{44}{}^2\sum_{i,l}\langle\langle S_i^- | S_l^+ S_l^z\rangle\rangle_\omega + \frac{1}{2}G_{44}T_{44}H_{03}\sum_j \langle\langle S_j^z S_j^- | \sigma_0^+\rangle\rangle_\omega\\ &+ \frac{1}{2}G_{44}{}^2\sum_{j,l}\left(\langle\langle S_j^z S_j^- | S_l^+\rangle\rangle_\omega + 2\langle\langle S_j^z S_j^- | S_l^+ S_l^z\rangle\rangle_\omega\right)\Big\}\varepsilon^-(\omega).\end{aligned} \tag{34}$$

The propagators appearing in (34), with account of (22) are rather simple to calculate:

$$\begin{aligned}&\frac{1}{2}\sum_j \langle\langle S_j^z S_j^- | \sigma_0^+\rangle\rangle_\omega = 2N\xi Jc(D - \tilde{R}'')D^{-1}(\omega),\\ &\sum_{j,l}\langle\langle S_j^z S_j^- | S_l^+\rangle\rangle_\omega = 4J^2c^2NR\tilde{\xi}(D - \tilde{R}'')\{a_s D(\omega)\}^{-1} + \frac{cN}{a_s}(R - D),\\ &\sum_{j,l}\langle\langle S_j^z S_j^- | S_l^+ S_l^z\rangle\rangle_\omega = \frac{cN}{a_s}[D - 2\langle S_z^3\rangle + R(2S(S+1) - 1)] +\\ &\qquad + 2J^2c^2N\tilde{\xi}(D - \tilde{R}'')(D - R)\{a_s D(\omega)\}^{-1},\\ &D = 3\langle S_z^2\rangle - S(S+1), \qquad \tilde{R}'' = R(\omega_s' + i\Delta_s)/\omega_s'.\end{aligned} \tag{35}$$

We now discuss the equations for the inhomogeneous term in the case of quadrupole moments. We introduce here two new "quadrupole" moments:

$$D_1^- = \frac{G_{44}}{2N}\sum_j S_j^z S_j^- \text{ и } D_2^- = \frac{G_{44}}{2N}\sum_j S_j^- S_j^z,$$

then

$$\begin{aligned}&i\frac{\partial}{\partial t}D_- = [D_-, \mathscr{H}] - i\Delta_s\Big\{D_- - \chi_{s3}\varepsilon^-(t) - \chi_{sM}\frac{\lambda g_e G_{44}}{2g_s T_{44}H_{03}}p_-\Big\},\\ &i\frac{\partial}{\partial t}p_- = [p_-, \mathscr{H}] - i\Delta_e\Big\{p_- - \chi_{e3}\varepsilon^-(t) - \chi_{eM}\frac{2\lambda g_s T_{44}H_{03}}{g_e G_{44}}(D_2^- - D_1^-)\Big\}\\ &D_1^- + D_2^- = D_-.\end{aligned} \tag{36}$$

In writing out (36), we have assumed an isotropic exchange between the conduction electrons and the unfilled shells of the paramagnetic impurity, i.e., the effect of exchange on D_1^- and D_2^- is the same; moreover, the static acoustic deformation induces both D_1^- and D_2^- with equal probability.

The solution of (36) yields

$$\begin{aligned}&\langle p^-(\omega)\rangle = \frac{i\varepsilon^-(\omega)a_s\Delta_e\chi_{e3}}{a_s a_e - 4J^2cR\tilde{\xi}},\\ &\langle D_-(\omega)\rangle = D^{-1}(\omega)\,i\Big\{a_e\Delta_s\chi_{s3} - \frac{\Delta_s}{a_s}\chi_{s3}4J^2cR\tilde{\xi}\\ &\qquad + \frac{\Delta_e\chi_{e3}JcG_{44}}{T_{44}H_{03}\omega_s'}[iR\Delta_s + \omega_s' D]\Big\}\varepsilon^-(\omega).\end{aligned} \tag{37}$$

We shall not write out the complete expression for the susceptibility in view of its complexity, but only calculate the absorption coefficient for S = 1, since $\langle S_Z^3\rangle = \langle S_Z\rangle$, when the lines are well resolved. Then, recognizing that $D = -(1/3)N(\omega_S'/T)^2$, we obtain

$$\chi_s''(\omega) = \frac{\pi c N G_{44}{}^2\omega_s'}{3T}\left[\frac{1}{2} + \frac{\omega_s'}{4T} + \frac{\omega_s' - \omega}{\omega_s'}\right] g_s(\omega),$$

and the absorption coefficient is

$$\sigma_{sq} = \frac{\pi c N G_{44}{}^2\omega^2}{3\rho v^3 kT}\left[\frac{1}{2} + \frac{\hbar\omega_s'}{4kT} + \frac{\omega_s' - \omega}{\omega_s'}\right] g_s(\omega). \tag{38}$$

If we compare (38) with the expressions for the absorption coefficient of transverse sound waves (see[11]), we then see that they differ in two terms: the first depends on the square of the reciprocal temperature and the second is connected with the relaxation in the instantaneous local field. This latter process can take place under the action of thermal spin-phonon interactions, as well as a result of two-particle spin-spin interactions. Furthermore, at sufficiently low temperatures and high transverse components of the magnetic and elastic multipoles brought about by the action of the variable fields, the relaxation in the instantaneous local field may be determined by the coherent spontaneous transitions of the electromagnetic and elastic multipoles (i.e., as the result emission of a coherent elastic or electromagnetic field by them).

We note that the theory of acoustic resonance which we have developed can be used without special changes for the description of the resonance excitation of spin systems and systems of electric dipoles by an alternating field.

Electron paramagnetic resonance on impurity ions in metals has been observed on Gd ions in Pd,[8] Mn in Au, Cu, and Ag.[12] In the case of Gd, the temperature dependence of the line width shows that $T_1 \sim 10^{-9}$ sec at helium temperatures. In the second case evidently, the spin-lattice relaxation time is much greater and $T_1 \gtrsim 10^{-8}$ sec. A comparison of these times with the corresponding values for ions in dielectric crystals shows that in metals the impurity ions interact strongly with the lattice vibrations. Therefore, for an estimate of the absorption coefficient, it is reasonable to set the constant of spin-phonon interaction at $G \gtrsim 10^{-14}$–10^{-16} erg per deformation unit. Then, assuming that $c = 10^{-2}$, $N \sim 10^{22}$, $H_{03} \sim 10^4$ G, $T = 4.2$°K, $\rho = 12$ g/cm³, $g_S(\omega) \sim 10^{-10}$ sec, and $v \sim 2.2 \times 10^5$ cm/sec, we obtain

$$\sigma_{sq} \approx (10^{-20} - 10^{-24})\omega^2 \text{cm}^{-1}.$$

Naturally, these estimates are very approximate, but undoubtedly demonstrate the presence of a high absorption coefficient, which is quite natural.

4. CONCLUSION

In the present work, we have not gone into detail regarding the possibility of acoustic magnetic resonance on paramagnetic metals (Gd, Cu, Ce, etc). Experiments on EPR in these metals show that the mechanism of modulation of the crystalline field and of the hyperfine interactions by sound in them are also effective for excitation of acoustic magnetic resonance on the host ions of these materials. Estimates show that at hypersonic frequencies of the order of 10^{11} sec^{-1}, the acoustic paramagnetic resonance can be observed in them at helium temperatures.

When sound passes through a metal, generation of a variable magnetic field takes place. This field, acting directly on the electron spins, produces acoustic magnetic resonance. This phenomenon is well known in nuclear acoustic magnetic resonance in metals (for example, Al[13]). The formulas obtained in these researches also describe acoustic electron resonance in metals without any appreciable changes. The corresponding quantum transitions have a magnetic dipole character and a different orientation dependence on the static magnetic field than those considered in the present paper.

Finally, the feasibility of observing acoustic electron resonance on impurities in liquids is of interest. This applies to the case in which the same mechanisms of spin-phonon interaction are effective, but are modulated by the Brownian motion. The observation of acoustic magnetic resonance in them is possible if the Brownian motion does not completely disrupt the coherence of the sound excitation.

Inasmuch as spin-phonon interaction in metals is extremely strong, acoustic paramagnetic resonance can be detected in them by means of the change in the intensity of optical transitions (the acoustical analog of the optical detection by the Kastler method[14]), the Mössbauer effect, and $\gamma - \gamma$ angular correlation. In the last two cases, the hyperfine interaction on radioactive nuclei is modulated by the electron acoustic resonance.

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133