Acoustic resonance saturation as a spin "bottleneck" effect

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A new method for describing acoustic resonance (AR) is proposed, in which the main dynamic subsystem consists of resonant (with respect to mechanical vibrations of the system) phonons that are coupled to the thermostat (the remaining degrees of freedom) either directly or via another dynamic subsystem, the spin subsystem. This approach permits one to calculate in a unified manner the nonequilibrium mean values for both the spin and vibrational susbsystems. It also permits AR saturation effects to be taken into account and interpreted in a novel way. Coupled kinetic equations for the dynamic subsystems are derived by the nonequilibrium-statistical-operator technique. In the description of the spin subsystem, allowance is made for the nonequilibrium states of the dipole-dipole interaction pool. A solution of the kinetic equations is found under stationary conditions. The AR-induced saturation of the paramagnetic contribution to the speed of sound is calculated.

1. CHOICE OF MODEL

The description of resonant phenomena in condensed media is based on separating the dynamic subsystem with discrete energy levels, in which the resonance is excited, from the remaining degrees of freedom, which constitute the thermostat. To take consistent account of the external alternating field, it is necessary to introduce, in addition to the Hamiltonians of the dynamic system $\hat{\mathscr{H}}_S = \hbar\hat{E}^*(\hat{s})$ and of the thermostat $\hat{\mathscr{H}}_L = \hbar\hat{E}(\hat{q})$ also the Hamiltonian of the alternating field of the generator $\hat{\mathscr{H}}_F = \hbar\hat{K}(\hat{F})$ (here $\hat{s}$, $\hat{q}$, and $\hat{F}$ denote the degrees of freedom of the dynamic and dissipative subsystems and of the field, respectively). Figure 1a shows schematically these subsystems and the interactions between them, namely $\hbar\hat{G}(\hat{s}, \hat{q})$ and $\hbar\hat{D}(\hat{s}, \hat{F})$. If the processes that occur in the subsystems do not influence the generator, then the operators $\hat{F}$ in the interaction $\hbar\hat{D}(\hat{s}, \hat{F})$ can be replaced by their mean values F(t). Then $\hat{D}(\hat{s}, \hat{F}) \equiv \hat{D}^t(\hat{s})$, where $\hat{D}^t(\hat{s})$ contains only the operators of the dynamic subsystem and of the classical-field intensity, which depend explicitly on the time.

The universally employed acoustic-resonance (AR) treatment[1, 2] corresponds to the scheme of Fig. 1a, where the dynamic subsystem is the spin system (S system), and the alternating-field intensities are the components of the deformation field $U = U_0\cos(\mathbf{p}\cdot\mathbf{r} - \omega t)$, treated classically. In this approach, however, it becomes impossible to represent certain properties of the vibrational system, since it is assumed that the acoustic deformations are determined only by the external generator, and no account is taken of the reaction of the substance on the deformations. A more complete description makes it necessary to regard the vibrational system as a quantum system, whose operators can assume different values during the course of the experiment. To observe AR, the single crystals are processed in such a way that they become a resonant acoustic system, in which it is possible to excite intense oscillations of only definite modes, and only with a discrete set of resonant frequencies $\omega_k = k\omega_p$ (k is an integer), with the width $\Delta\omega_k$ of the resonant peaks much smaller than ω_p.[1)]

One can propose another theoretical description of AR, in which the vibrational degrees of freedom (the strains U_p connected with the set of frequencies $\omega_k = k\omega_p$) are regarded as a dynamic subsystem with a discrete spectrum (P system). All the remaining vibrational degrees of freedom constitute the thermostat. The P system is coupled, via the interaction $\mathscr{H}_{sp}$, with the spin system (the second dynamic subsystem). Both subsystems are in contact with the thermostat via the interactions $\hbar\hat{G}(\hat{p}, \hat{q})$ and $\hbar\hat{G}(\hat{s}, \hat{q})$. Finally, the external classical alternating field is the field of the elastic stresses $\sigma(\mathbf{r}, t) = \sigma_0\cos(\omega t - \mathbf{p}\cdot\mathbf{r})$ produced by the ultrasonic generator. This model corresponds to the scheme of Fig. 1b. In contrast to the universally employed approach, in this model the external alternating field is different in nature, the deformations are assumed to be quantized, and the spin degrees of freedom play the role of not the main subsystem but of the supplementary subsystem.

The AR effect is produced when the natural frequencies of the spin system coincide with $\omega \sim k\omega_p$, and energy is transferred from the generator into the spin system. Saturation of the AR sets in when the coupling $\mathscr{H}_{sp}$ is capable of transferring more energy than the coupling $\mathscr{H}_{sq}$, and the spin system deviates noticeably from equilibrium. In this sense, AR saturation can be treated as a "bottleneck," of the spin type in this case.

The approach proposed here makes it possible to obtain in unified manner the nonequilibrium mean values of physical quantities of both spin and vibrational nature. In addition to determining the acoustic power absorbed by the spin system, it is possible, for example,

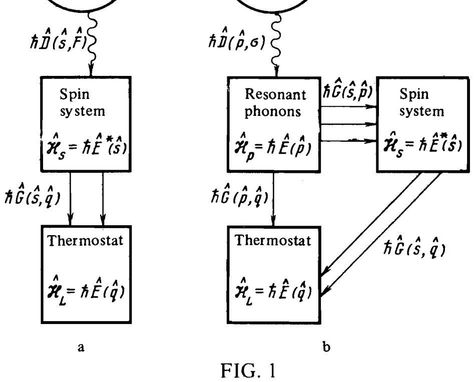

FIG. 1

to obtain the AR-induced change in the velocity and rotation of the plane of polarization of the sound introduced into the sample from the ultrasonic generator. To this end it suffices to calculate the nonequilibrium mean values of the creation and annihilation operators a_p^+ and a_p of the resonant phonons, and this is made possible by regarding the resonant oscillations as a quantum non-equilibrium system.

It should be noted that the changes of the speed of sound[3-6] and of the rotation of the polarization plane[7-8], due to spin-phonon interaction, have already been calculated earlier. The paramagnetic change of the speed of sound near AR was observed by Shiren[4]. However, the calculation method used in[5-8] has the following properties: a) it is valid under conditions of thermodynamic equilibrium, b) it pertains to thermal phonons, c) it cannot take into account the AR-induced saturation of the spin system. By using the proposed approach, we shall describe below the paramagnetic change of the speed of sound without the indicated limitations.

In accordance with the scheme of Fig. 1b, the complete Hamiltonian of the medium in which the Ar is investigated can be represented in the form

$$\hat{\mathscr{H}} = \hat{\mathscr{H}}_0 + \hat{V}^t, \qquad \hat{\mathscr{H}}_0 = \hbar\,[\hat{E}(\hat{p}) + \hat{E}^*(\hat{s}) + \hat{E}(\hat{q})],$$

$$\hat{V}^t = \hbar\,[\hat{G}(\hat{s}, \hat{q}) + \hat{G}(\hat{p}, \hat{q}) + \hat{G}(\hat{s}, \hat{p}) + \hat{D}^t(\hat{p})], \tag{1}$$

where $\hat{\mathscr{H}}_0$ is the fundamental Hamiltonian, and $\hat{V}^t$ is a small interaction. The Hamiltonians of the resonant and thermal phonons are respectively

$$\hat{\mathscr{H}}_p = \hbar\hat{E}(\hat{p}) = \hbar\omega_p\hat{a}_p{}^+\hat{a}_p,$$

$$\hat{\mathscr{H}}_L = \hbar\hat{E}(\hat{q}) = \hbar\sum_q \omega_q(\hat{a}_q{}^+\hat{a}_q + {}^1/_2); \tag{2}$$

where $\hat{a}_q^+$ and $\hat{a}_q$ are the thermal-phonon creation and annihilation operators.

Let the ultrasonic generator produce in a sample in the form of a rod of length l and cross section B the elastic stresses

$$\sigma_{ik}(\mathbf{r},\ t) = -\delta_{ix}\delta_{kx}\sigma_0\cos(\omega t - px) = \sigma_{xx}(x,\ t), \tag{3}$$

where p and ω are the wave vector and frequency of the ultrasound. In this case the operator of the energy of interaction with the external alternating field turns out to be

$$\hat{\mathscr{H}}^t = \hbar\hat{D}(\hat{p}) = B\int_0^l dx\,\hat{\mathscr{H}}^t(x), \quad \hat{\mathscr{H}}^t(x) = \hat{U}_{xx}(x)\,\sigma_{xx}(x, t), \tag{4}$$

where $U_{xx}(x)$ is a component of the strain tensor and is equal to

$$\hat{U}_{xx}(x) = i(\hbar p^2/2M\omega_p)^{1/2}(\hat{a}_p e^{ipx} - \hat{a}_p{}^+ e^{-ipx}), \tag{5}$$

and plays the role of the operator of the system, while the stress tensor $\sigma_{ik}(\mathbf{r}, t)$ is the classical external force.

The spin-system Hamiltonian $\hat{\mathscr{H}}_S = \hbar\hat{E}^*(\hat{s}) = \hat{\mathscr{H}}_S^0 + \hat{\mathscr{H}}_d$ contains a term that describes its spectrum, namely $\hat{\mathscr{H}}_S^0 = \hbar\hat{E}(\hat{s})$, and a term $\hat{\mathscr{H}}_d$ that describes the adiabatic part of the magnetic dipole-dipole interactions.

As the concrete spin system we consider paramagnetic ions with effective spin S = 1 in a crystal field of axial symmetry and in a constant magnetic field H_0 parallel to this axis (z axis). In this case the operator $\hat{E}(s)$ takes the form

$$\hat{E}(\hat{s}) = \hat{E}_Q(\hat{s}) + \hat{E}_z(\hat{s}), \quad \hat{E}_Q(\hat{s}) = {}^1/_3 D\hat{Q}_0, \quad \hat{E}_z(\hat{s}) = \omega_0\hat{S}_z,$$

where

$$\hat{Q}_0 = \sum_{k=1}^N \hat{Q}_0{}^k, \quad \hat{S}_z = \sum_{k=1}^N \hat{S}_z{}^k,$$

$D = \hbar\omega_Q$ is the fine-splitting constant, $\omega_0 = g_{\|}\beta_B H_0\hbar^{-1}$, $g_{\|}$ and $g_{\perp}$ are the principal values of the g-factor, and β_B is the Bohr magneton. Q_λ^k are the irreducible tensor operators of second rank for the k-th spin[2], where $\lambda = 0, \pm 1, \pm 2$. The energy levels and the wave functions of such a paramagnetic center are shown in Fig. 2, where $|\chi_m\rangle$ is the eigenstate of the operator $\hat{S}_z$. As examples of such states we can cite the ions of trivalent vanadium and divalent iron in corundum, in which AR was already observed[9]. Slight rhombic additions in the spin Hamiltonian of these ions can be left out. We disregard below also the hyperfine splittings, which generally speaking can be clearly seen on the resonance lines of these ions.

The form of the adiabatic part of the dipole-dipole interactions $\hat{H}_d$ depends significantly on the spin-system spectrum. In our case we have

$$\hat{\mathscr{H}}_d = \sum_{j,k} A\,\{\hat{S}_z^j\hat{S}_z^k - {}^1/_2[\hat{P}_{10}^j\hat{P}_{01}^k + \hat{P}_{0,-1}^j\hat{P}_{-10}^k + \hat{P}_{10}^k\hat{P}_{01}^j + \hat{P}_{0,-1}^k\hat{P}_{-10}^j]\}, \tag{6}$$

where $A = (g^2\beta_B^2/r^3)(1 - 3\cos^2\theta)$; r, θ, and φ are the spherical coordinates of the vector joining the spins j and k. We have used here the projective operators $\hat{P}_{mn}$, defined by their matrix elements $\langle\chi_{m'}|P_{mn}|\chi_{n'}\rangle = \delta_{m'm}\delta_{nn'}$ in the basis of the eigenfunctions $|\chi_m\rangle$ of the Hamiltonian $\mathscr{H}_S^0 = \hbar\hat{E}(\hat{s})$.

The interaction of the spin system with the thermal phonons is given by[1, 2]

$$\hat{\mathscr{H}}_{sL} = \hbar\hat{G}(\hat{s}, \hat{q}) = \hbar\sum_{\lambda=-2}^{2}(-1)^\lambda\hat{F}_{-\lambda}(\hat{q})\,\hat{Q}_\lambda(\hat{s}). \tag{7}$$

The interaction of the spin system with the resonant phonons can be obtained from the operator of the dynamic spin-phonon coupling, in which, however, the strain tensor must be represented in operator form. Assume that the elastic stress produces in the medium only longitudinal acoustic waves, so that only one component of the strain tensor, U_{xx}, differs from zero. In this case, the resonant transitions between the levels $\langle\chi_{\pm 1}| \leftrightarrow |\chi_{\mp 1}\rangle$ are determined by the following terms of the dynamic spin-phonon coupling operator, (see[2], Sec. 5):

$$\hat{\mathscr{H}}_{sp} = \hbar G\sum_k[\hat{Q}_2{}^k + \hat{Q}_{-2}{}^k]\,\hat{U}_{xx}(x_k), \quad G = G_{xxxx} - G_{xxyy}, \tag{8}$$

where G_{xxxx} and G_{xxyy} are the components of the dynamic-coupling tensor.

Finally, the interaction of the resonant and thermal phonons is the result of the crystal-oscillation anharmonicity due to three-phonon processes, and takes the form

$$\hat{\mathscr{H}}_{pL} = \hat{a}_p{}^+\hat{H}_p{}^+ + \hat{a}_p\hat{H}_p, \quad \hat{H}_p = 3\sum_{\mathbf{k}_1,\,\mathbf{k}_2 \neq \mathbf{p}} V(\mathbf{p}, \mathbf{k}_1, \mathbf{k}_2)\,\hat{A}_{\mathbf{k}_1}\hat{A}_{\mathbf{k}_2},$$

$$\hat{A}_{k_i} = \hat{a}_{k_i} + \hat{a}_{-k_i}^+, \quad \hat{A}_{k_i}^+ = \hat{a}_{-k_i} + \hat{a}_{k_i}^+, \tag{9}$$

where $V(\mathbf{p}, \mathbf{k}_1, \mathbf{k}_2)$ is the matrix element of the three-phonon interaction[10].

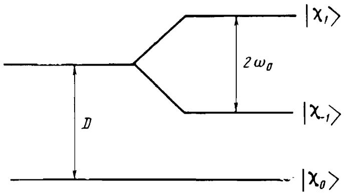

FIG 2

2. KINETIC EQUATIONS FOR VIBRATIONAL SUBSYSTEMS INTERACTING WITH A SPIN SUBSYSTEM AND SITUATED IN AN ELASTIC STRESS FIELD

We employ the Zubarev nonequilibrium-statistical-operator method[11] to obtain the kinetic equations for the AR with allowance for the nonequilibrium states of the resonant vibrational subsystem. We consider only excitations at the fundamental mechanical-resonance frequency ($\omega_k = \omega_p$).

We choose first dynamic variables that describe the nonequilibrium states of the system. It is known that in the presence of a strong interaction $\hat{H}_d$ a cancellation takes place in the description of the spin system, and the transverse magnetization components cease to describe the system for times $t > t_d = \hbar(\mathrm{Tr}\hat{\mathscr{H}}_d^2)^{-1/2}$. During these times, the system is described by the nonequilibrium averaged operators $\hat{\mathscr{H}}_s^0 = \hbar\hat{E}(\hat{s})$ and $\hat{\mathscr{H}}_d$[12]. In the vibrational subsystem, there is no reason for such a cancellation. Therefore, to describe the nonequilibrium behavior of the considered system during times $t > t_d$ we choose the following operators:

$$\hat{a}_p,\ \hat{a}_p^+,\ \hat{n}_p=\hat{a}_p^+\hat{a}_p,\quad \hat{\mathscr{H}}_d,\ \hat{S}_z=\sum_{k=1}^{N}\hat{S}^z{}_k,\quad \hat{Q}_0=\sum_{k=1}^{N}\hat{Q}_0{}^k. \tag{10}$$

The entropy operator $\hat{S}_e(t, 0)$ is equal in this case to[11]

$$\hat{S}_e(t,0)=\Phi(t)+\beta_z(t)\hat{\mathscr{H}}_z+\beta_d(t)\hat{\mathscr{H}}_d+\beta_Q(t)\hat{\mathscr{H}}_Q+\beta_p(t)\hat{\mathscr{H}}_p+\beta_p^+(t)\hat{a}_p^++\beta_p^-(t)\hat{a}_p+\beta_L\hat{\mathscr{H}}_L, \tag{11}$$

where $\Phi(t)$ is the Massieu-Planck functional, $\beta_n(t)$ are the dynamic parameters that are conjugate to the operators (10), and β_L is the reciprocal lattice temperature. Following Zubarev[11], we construct a nonequilibrium statistical operator corresponding to the entropy operator (11), and obtain in the high-temperature approximation kinetic equations for the operator (10):

$$\begin{aligned}
&\frac{\partial\langle\hat{S}_z\rangle}{\partial t}=-\frac{\langle\hat{S}_z\rangle-\langle\hat{S}_z\rangle^0}{T_z}-W_{sp}\langle\hat{S}_z\rangle\langle\hat{n}_p\rangle,\\
&\frac{\partial\langle\hat{n}_p\rangle}{\partial t}=\mathscr{F}\langle\hat{V}_p+\hat{V}_p^+\rangle-\frac{\langle\hat{n}_p\rangle-\langle\hat{n}_p\rangle^0}{T_p}+\frac{1}{2}W_{sp}\langle\hat{S}_z\rangle\langle\hat{n}_p\rangle,\\
&\frac{\partial\langle\hat{V}_p\rangle}{\partial t}=-i\Delta\omega\langle\hat{V}_p\rangle-\frac{\langle\hat{V}_p\rangle}{2T_p}+\frac{1}{4}W_{sp}\langle\hat{S}_z\rangle\langle\hat{V}_p\rangle+\mathscr{F},\\
&\frac{\partial\langle\hat{V}_p^+\rangle}{\partial t}=i\Delta\omega\langle\hat{V}_p^+\rangle-\frac{\langle\hat{V}_p^+\rangle}{2T_p}+\frac{1}{4}W_{sp}\langle\hat{S}_z\rangle\langle\hat{V}_p^+\rangle+\mathscr{F},\\
&\frac{\partial\langle\hat{\mathscr{H}}_d\rangle}{\partial t}=-\frac{\langle\hat{\mathscr{H}}_d\rangle-\langle\hat{\mathscr{H}}_d\rangle^0}{T_d}-W_{dp}\langle\hat{\mathscr{H}}_d\rangle\langle\hat{n}_p\rangle,\\
&\frac{\partial\langle Q_0\rangle}{\partial t}=-\frac{\langle\hat{Q}_0\rangle-\langle\hat{Q}_0\rangle^0}{T_Q};
\end{aligned} \tag{12}$$

Here

$$\Delta\omega=\omega_p-\omega,\quad \hat{V}_p=\hat{a}_pe^{i\omega t},\quad \Omega=2\Delta\omega T_p,$$

$$\mathscr{F}=\frac{B}{\hbar}\left(\frac{\hbar p^2}{2M\omega_p}\right)^{1/2}\int_0^l\sigma_{xx}(x)e^{-ipx}\,dx=\frac{Bl}{2\hbar}\left(\frac{\hbar p^2}{2M\omega_p}\right)^{1/2}\sigma_0.$$

For the relaxation times that enter in (12) we obtain the expressions

$$\frac{1}{T_Q}=\frac{1}{2\pi}\int d\omega\,\varphi_1(\omega)\left[f_{1,0}(\omega_0+\omega_Q-\omega)+f_{-1,0}(\omega_0-\omega_Q-\omega)\right],$$

$$\frac{1}{T_z}=\frac{1}{T_Q}+\frac{16}{2\pi}\int d\omega\,\varphi_2(\omega)f_{1,-1}(2\omega_0-\omega),$$

$$\frac{1}{T_d}=\frac{12}{2\pi}\int d\omega\,\varphi_1(\omega)\left[f_{1,0}(\omega_0+\omega_Q-\omega)+f_{-1,0}(\omega_0-\omega_Q-\omega)\right]=12\frac{1}{T_Q}, \tag{13}$$

where

$$\varphi_\lambda(\omega)=\int e^{i\omega t}\langle F_\lambda(t,q)F_{-\lambda}(q)\rangle dt,$$

$$f_{mn}(\omega_{mn}-\omega)=\int\exp\{i(\omega_{mn}-\omega)t\}\langle P_{mn}(t)P_{nm}\rangle dt; \tag{14}$$

$f_{mn}(\omega_{mn}-\omega)$ is a function of the shape of the resonance line at the frequency ω_{mn} and is determined by the broadening interaction $\hat{\mathscr{H}}_d$. Direct calculation of the relaxation times by formula (13) in the case of phonon mechanisms that couple the spins with the lattice can be found in[13,14]. The phonon-lattice relaxation time is calculated in[10]. The quantities

$$W_{sp}=32\pi G^2\frac{\hbar p^2}{2M\omega_p}f_{1,-1}(2\omega_0-\omega)=\frac{1}{2}W_{dp} \tag{15}$$

have the meaning of probabilities of energy exchange between the dynamic subsystems as a result of the interaction $\hat{\mathscr{H}}_{sp}$.

From the kinetic equations we obtain an expression for the power absorbed under stationary conditions

$$\begin{aligned}
\mathscr{P}&=\left.\frac{d\langle\hat{\mathscr{H}}_z\rangle}{dt}\right|_{T_1}+\left.\frac{d\langle\hat{\mathscr{H}}_p\rangle}{dt}\right|_{T_1}+\left.\frac{d\langle\hat{\mathscr{H}}_d\rangle}{dt}\right|_{T_1}\\
&=\hbar\omega_p\mathscr{F}\langle\hat{V}_p+\hat{V}_p^+\rangle+\langle\hat{n}_p\rangle\left[\hbar\left(\frac{\omega_p}{2}-\omega_0\right)W_{sp}\langle\hat{S}_z\rangle-W_{dp}\langle\hat{\mathscr{H}}_d\rangle\right]\\
&=\frac{m_p[\hbar\omega_p+\hbar\omega_0Z_p-W_{dp}T_p\langle\hat{\mathscr{H}}_d\rangle]}{[(1+Z_p)^2+\Omega^2]T_p},
\end{aligned} \tag{16}$$

where the symbol $|_{T_1}$ indicates that no account is taken of the changes connected with the relaxation processes, and $m_p = (2T_p\mathscr{F})^2$.

3. STATIONARY SOLUTION OF KINETIC EQUATIONS

Under stationary conditions, the system (12) makes it possible to express all the mean values in terms of $\langle\hat{n}_p\rangle$:

$$\langle\hat{S}_z\rangle=\frac{\langle S_z\rangle^0}{1+Z_s},\quad \langle\hat{\mathscr{H}}_d\rangle=\frac{\langle\hat{\mathscr{H}}_d\rangle^0}{1+Z_d},\quad \langle\hat{V}_p\rangle=\frac{m_p^{1/2}[1+Z_p-i\Omega]}{(1+Z_p)^2+\Omega^2}. \tag{17}$$

Here $m_p = (2T_p\mathscr{F})^2$, $\Omega = 2T_p\Delta\omega$, and we have introduced the nonequilibrium saturation factors:

$$Z_s=W_{sp}T_z\langle\hat{n}_p\rangle,\ Z_d=W_{dp}T_d\langle\hat{n}_p\rangle,$$

$$Z_p=-{}^1/{}_2W_{sp}T_p\langle\hat{S}_z\rangle=-{}^1/{}_2W_{sp}T_p\langle\hat{S}_z\rangle^0(1+Z_s)^{-1}.$$

In particular, the average strain can be obtained by substituting solutions of the type $\langle\hat{V}_p\rangle = \langle\hat{a}_p\rangle e^{i\omega t}$ in expression (5). After substitutions we have

$$\begin{aligned}
&U_{xx}(x,t)=[\chi_u'(\omega)\cos(\omega t-px)+\chi_u''(\omega)\sin(\omega t-px)]\sigma_0,\\
&\chi_u'(\omega)=\chi_u{}^0\frac{\Omega}{(1+Z_p)^2+\Omega^2},\quad \chi_u''(\omega)=\chi_u{}^0\frac{1+Z_p}{(1+Z_p)^2+\Omega^2},\\
&Z_p=\frac{Z_p{}^0}{1+Z_s},\quad \chi_u{}^0=\frac{T_pp^2}{\rho\omega_p}.
\end{aligned} \tag{18}$$

The average number of resonant phonons is determined from the equation

$$\langle\hat{n}_p\rangle=\frac{\langle\hat{n}_p\rangle^0}{1+Z_p}+\frac{m_p}{(1+Z_p)^2+\Omega^2}. \tag{19}$$

We see therefore that m_p is the number of coherent resonant phonons that are excited by the acoustic generator in the sample in which there is no spin-phonon coupling. One can speak of induced acoustic oscillations only if $m_p \gg \langle\hat{n}_p\rangle^0$. Under this condition, the equation can be rewritten in the form

$$Z_s^3+\frac{2(1+Z_p^0+\Omega^2)-Z_s^0}{1+\Omega^2}Z_s^2+\frac{(1+Z_p^0)^2+\Omega^2-2Z_s^0}{1+\Omega^2}Z_s=\frac{Z_s^0}{1+\Omega^2}, \quad (20)$$

where we have introduced the equilibrium saturation factors

$$Z_s^0=W_{sp}T_zm_p, \quad Z_p^0={}^1/{}_2W_{sp}T_pm_s$$

and the equilibrium population difference between the levels E_1 and E_{-1}

$$m_s=-\langle\hat{S}_z\rangle^0=2\hbar\omega_0N/3kT.$$

The real positive solutions of Eq. (20), together with relations (17), determine all the nonequilibrium average characteristics of the system at a given instant of time. The analytic form of the solutions of the equations is complicated. The dependence of $Z_s(\equiv x)$ on $Z_s^0(\equiv y)$ at several values of the parameter Z_p^0 and at $\Omega = 0$ is shown in Fig. 3. We note also that at the limiting values of the amplitudes of the elastic stresses we have

$$\lim_{\sigma_0\to 0} Z_s\to 0, \quad \lim_{\sigma_0\to\infty} Z_s=\frac{Z_s^0}{1+\Omega^2}$$

We now consider several particular cases.

4. SYSTEM FAR FROM ACOUSTIC RESONANCE ($\omega_p \neq \omega_{mn}$)

A function such as $f_{1,-1}(2\omega_0-\omega)$ has a sharp maximum at $\omega_p - 2\omega_0 = 0$, and vanishes far from resonance. Therefore, in the case of a consideration, the probabilities W_{sp} and W_{dp} tend to zero, and the equations break up into two uncoupled systems describing noninteracting dynamic subsystems. All the spin averages turn out to be equal to the equilibrium values at the lattice temperature, and for the vibrational mean values we obtain

$$\langle\hat{n}_p\rangle=\langle\hat{n}_p\rangle^0+\frac{m_p}{1+(2T_p\Delta\omega)^2}, \quad m_p=(2\mathscr{F}T_p)^2, \quad (21)$$

$$\langle\hat{a}_p\rangle=2\mathscr{F}T_pe^{i\omega t}\frac{1-2iT_p\Delta\omega}{1+(2T_p\Delta\omega)^2}=\langle\hat{a}_p^+\rangle^*. \quad (22)$$

Expressions (21) and (22) describe the stationary oscillations at the frequency ω in a sample with mechanical-resonance frequency ω_p and damping T_p. In particular, for the power absorbed by the crystal from an existing generator in the absence of a contact with the spin system, we obtain

$$\mathscr{P}_p=\frac{\omega}{2}\chi_{\mathscr{P}}''(\omega)\sigma_0^2, \quad \chi_{\mathscr{P}}''(\omega)=\frac{p^2T_p}{\rho\omega}\frac{1}{1+\Omega^2}, \quad (23)$$

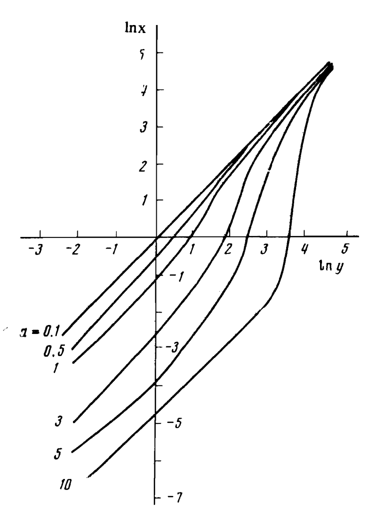

FIG. 3

and the acoustic deformation is equal to

$$U_{xx}(x,t)=[\chi_u'(\omega)\cos(\omega t-px)+\chi_u''(\omega)\sin(\omega t-px)]\sigma_0,$$

$$\chi_u''(\omega)=\frac{\omega}{\omega_p}\chi_{\mathscr{P}}''(\omega), \quad \chi_u'(\omega)=2T_p\Delta\omega\chi_u''(\omega), \quad (24)$$

where ρ is the density of the sample. The quantities $\chi_{u,\mathscr{P}}$ are the vibrational susceptibilities. Accurate to a factor ω/ω_p (which is equal to unity near mechanical resonance), the susceptibilities χ_u'' and $\chi_{\mathscr{P}}''$ are equal.

The results of (23) and (24) correspond to a treatment of the resonant oscillations as a dynamic subsystem according to the scheme of Fig. 1a.

5. ACOUSTIC RESONANCE FAR FROM SATURATION

Let us find an expression for the power absorbed in the AR under the following conditions: a) the acoustic generator excites the resonant mechanical oscillations ($\omega = \omega_p$); b) there is no saturation (dynamic ordering of saturation factors of the dynamic subsystems are Z_s, $Z_p \ll 1$); c) the change in the energy of the dipole-dipole interactions under the influence of the ultrasound can be neglected. Taking these conditions into account we substitute the relations (17) into the right-hand side of (16). After eliminating from $\mathscr{P}$ the power that flows out of the P system directly into the thermostat, we obtain the power $\mathscr{P}_s^0$ that goes to the thermostat via the spin system:

$$\mathscr{P}_s^0=-\hbar\omega_0m_p\langle\hat{S}_z\rangle^0W_{sp}(1+\Omega^2)^{-1}. \quad (25)$$

Recognizing that $\langle\hat{S}_z\rangle^0 = -2\hbar\omega_0N/3kT \equiv -m_s$, where m_s is the population difference between the levels E_1 and E_{-1}, and also the fact that under the considered conditions we have $\chi_u''(\omega_p)\sigma_0 = \langle U\rangle_0$, we obtain in the case of exact mechanical resonance ($\omega = \omega_p$) the usual expression for the absorbed power:

$$\mathscr{P}_s^0=2\hbar\omega_0m_sW, \quad W=8\pi G^2(\langle U\rangle_0)^2f_{1,-1}(2\omega_0-\omega), \quad (26)$$

where W is the probability of the transition between the levels E_1 and E_{-1} under the influence of the interaction (4), in which U_{xx} is regarded as a classical force: $U_{xx} = \langle U\rangle_0\cos(\omega t - px)$.

6. ACOUSTIC SATURATION OF SPIN SYSTEM. SPIN "BOTTLENECK"

Let us estimate the ratios of the equilibrium saturation factors that enter in (20). Putting $T_p = Q/\nu_p$ and $m_p = 10^7 I/\hbar\omega_p v$, we have

$$\eta\equiv Z_p^0/Z_s^0\approx 2\cdot 10^{-34}N_0\nu_p/TT_zI,$$

where N_0 is the number of spins per unit volume, I is the intensity of sound in W/cm^2, and we use for the Q factor of the sample and for the speed of sound v the respective values 10^5 and 4.2×10^5 cm/sec. At nuclear resonance ($N_0 \sim 10^{22}$ cm^{-3}, $\nu_p \sim 3 \times 10^7$ Hz) at room temperature, typical values are $T_Z \sim 10^{-4}$ sec and $\eta = 2 \times 10^{-3}/I$, while at helium temperature $T_Z > 10$ sec and $\eta < 1.5 \times 10^{-6}/I$. Consequently, even at slight sound intensities we have $Z_p^0 \ll Z_s^0$. For electron resonance at room temperature, a typical value is $T_Z \sim 10^{-8}$ sec and $\eta \sim 1/I$. At liquid-helium temperature we have $T_Z \sim 10^{-4}$ sec, and $\eta \sim 10^{-2}/I$. Thus, a condition $Z_p^0 \ll Z_s^0$ is easily reached also in EPR, although the inverse situation can be realized at room temperature, since T_Z and I are small. In this case, however, there will be no saturation. We consider below the easily-realized and therefore important particular case $Z_p \ll 1$ and $Z_s \gg 1$. When considering two interacting subsystems in accordance with the scheme corresponding to Fig. 1b, the case $Z_s \gg 1$ can be called "bottleneck" (spin bottleneck in the present problem).

At $Z_p \ll 1$ it makes sense to calculate all the mean values in the first nonvanishing approximation in Z_p. In the zeroth approximation we have for the vibrational subsystem the result (21) and (22), while the spin main values are

$$\langle\hat{S}_z\rangle=\langle\hat{S}_z\rangle^0/(1+Z_s^0),\quad \hat{\mathscr{H}}_d=\langle\hat{\mathscr{H}}_d\rangle^0/(1+Z_d^0),$$

$$Z_d^0=W_{dp}T_d m_p.$$

The power absorbed through the spin system in this approximation is equal to

$$\mathscr{P}_s=\mathscr{P}_s^0[1/(1+Z_s^0)+2\omega_d(1+Z_d^0)/\omega_0],$$

where $\mathscr{P}_s^0$ is the power absorbed in the absence of saturation and $\omega_d = \mathrm{Sp}\hat{\mathscr{H}}_d^2/\mathrm{Sp}(\hbar\hat{S}_z)^2$.

Unlike the previously known expressions for $\mathscr{P}_s$, such as (26), the result (27) describes the saturation connected with the AR when the excitation of the vibrational subsystem deviates from resonance, and also takes into account the nonequilibrium states of the dipole dipole pool DDP. We note that the role of the DDP in the saturation, under AR conditions, was investigated earlier by the Shutilov group[15], but their analysis pertained to a spin system with equidistant spectra.

Measurement of the speed of sound as a result of the spin-phonon interaction arises in the first approximation in Z_p. Using the relation $v = -\sigma_0/\rho\xi$ (ξ is the time derivative of the amplitude of the vibrational displacements in the traveling wave[16]) and determining ξ from the strain (18), we obtain the sound velocity with allowance for the spin-phonon coupling:

$$v_s=\frac{1}{pT_p}[(1+Z_p)^2+\Omega^2]^{1/2}.$$

This yields for small Z_p

$$\frac{v_s^2-v^2}{v^2}\approx\frac{2Z_p}{1+\Omega^2}=\frac{v_s^2-v^2}{v^2}\bigg|_{\text{я-с}}\frac{1}{(1+Z_p)(1+\Omega^2)}. \tag{28}$$

In the case of mechanical resonance $\Omega = 0$ and in the absence of saturation $Z_p = 0$, and also under the assumption $T_Z g(\omega_p - 2\omega_0)$, $(\omega_p - 2\omega_0)^{-2}$, expression (28) goes over into the results of Jacobson and Stevens[3], $[(v_S^2 - v^2)/v^2]_{J-S}$.

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[1)] The oscillations of the sample cease to be critical to high acoustic frequencies $\omega \sim 10^{10}$ rad/sec. The theory developed below includes also this case, which corresponds simply to the condition of exact mechanical resonance.

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