Interaction of two-level systems with phonons. Application to the theory of glasses

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A criterion for the applicability of perturbation theory to the description of the dynamics of two-level systems in a phonon field is considered. It is shown that in the case of dielectric glasses perturbation theory is most likely not to be valid. This is followed by calculation of the generalized susceptibilities that characterize the response of a two-level system to external actions. In first-order perturbation theory these susceptibilities consist of the customarily employed resonant and relaxation parts, whose amplitudes are somewhat decreased, and of smooth phonon parts that correspond to single-phonon intermediate states. In addition the renormalizations of the spacings of the levels and their population differences are determined. The results are used next to analyze the low-temperature properties of glasses. It is shown that the interaction increases the density of the tunnel states, and that the amplitude of the interaction of the latter with the phonons decreases. It is found as a result that if these phenomena are not taken into account, the tunnel-state density determined from the heat-capacity data should be larger than that determined from sound absorption and thermal conductivity. It is shown next that the heat capacity receives a contribution proportional to $T^2\ln(\omega_m/T)$, which can become comparable with the Debye heat capacity at sufficiently low temperatures. Arguments are presented, according to which both results are independent of whether perturbation theory is applicable or not.

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1. INTRODUCTION

Two-level tunnel states are quite widely encountered defects in solids. They attracted particular interest after Anderson, Halperin, and Varma[1] and Phillips[2] were able in 1972 to explain with their aid the high-temperature properties of glasses. Many facts accumulated since that time indicate that the existence of such states is one of the characteristic properties of amorphous substances. At the same time, detailed experiments performed since have revealed a number of quantitative discrepancies between theory and experiment. The most important of them are the following: 1) The tunnel-state density P determined from data on sound absorption and heat conduction is found to be less than that obtained from heat capacity.[3,4] 2) The theoretically predicted logarithmic growth, with increasing measurement time, of the heat-capacity component that is linear in temperature is practically unobservable.[5] 3) The part of the heat capacity remaining after separating the contribution linear in temperature exceeds the Debye value.[5–7]

Additional "anomalous" tunnel states were introduced to explain the disparity between the values of P obtained in different experiments.[3,4] The assumption that such states exist leads in turn to a number of difficulties. The question thus remains in essence open.

At the same time, up till now the interaction of tunnel states with phonons was always considered in first order of perturbation theory. There actually no grounds whatever for this. First, as shown in Ref. 8, in the case of fused quartz, at the experimentally obtained values of the strain potentials, the level width of the tunnel state becomes comparable with its energy at $E\sim 10$–30 K. Well defined levels exist thus only at much lower energies. Next, an integral criterion of the validity of perturbation theory was obtained in Ref. 9 (hereafter cited as I) in an analysis of the interaction of a degenerate center with phonons. If the level spacing is small compared with the limiting phonon frequencies, this criterion is applicable also when the degeneracy is lifted. For a two-level system it can be written in the form

$$J=\frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2}[2N(\omega)+1]d(\omega)\ll 1, \tag{1a}$$

$$d(\omega)=\frac{\pi}{V}\sum_{k,\alpha}\frac{k^2\gamma_{k,\alpha}^2}{2\rho\omega_{k,\alpha}}[\delta(\omega-\omega_{k,\alpha})-\delta(\omega+\omega_{k,\alpha})]. \tag{1b}$$

Here $N(\omega)$ is the Planck function, $d(\omega)$ is the spectral density of the phonon field acting on the two-level system, $\alpha = l, t$ is the polarization of the phonons, $\omega_{k,\alpha}$ are their frequencies, ρ is the density, V is the volume of the system, and $\gamma_{k,\alpha}$ are the constants of the deformation potential; we use a system of units in which $\hbar = k_B = 1$. In the low-frequency limit we have for $d(\omega)$ the simple formula

$$d(\omega)=\frac{1}{4\pi\rho}\left(\frac{\gamma_l^2}{c_l^5}+2\frac{\gamma_t^2}{c_t^5}\right)\omega^3=d_0\omega^3. \tag{2}$$

Substitution of this expression in (1) leads to an integral that diverges quadratically at the upper limit. If we use for the parameters in (2) their values for fused quartz, taken from Ref. 4, we obtain for J at $T=0$

$$J=d_0\omega_m^2/2\pi=8.6\cdot 10^{-5}\omega_m^2;\qquad d_0=(43\text{ K})^{-2}, \tag{3}$$

where ω_m is the maximum phonon energy, measured in degrees. Thus, if $\omega_m = 110$ K, then $J = 1$. Nothing is known at

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present on the behavior of the constants $\gamma_{k,\alpha}$ and of the frequencies $\omega_{k,\alpha}$ at large k; in other words, we do not know where the integral in (1) is cut off at high frequencies. It is clear, however, that there are no grounds whatever for regarding J as small. It is more readily of the order of unity, and accordingly perturbation theory cannot be used to describe the interaction of phonons with tunnel states in glasses.

The purpose of the present paper is a systematic study of the corrections, in next order of perturbation theory, to the observable quantities. It is found that all physical quantities are differently renormalized. In particular, the correction to the linear contribution to the heat capacity is positive, while that to the amplitude of the resonant absorption of phonons is negative. This can be interpreted as an effective increase of the density of the tunnel states in experiments on heat capacity, and as a decrease in experiments on sound absorption and heat conduction. It turns out here that the same takes place also when the interaction is not small.

This result agrees qualitatively with experiment. It should therefore be assumed that to explain the experimental data there is no need at present to introduce anomalous tunnel states, as was done in Refs. 3 and 4. Further, the standard procedure of averaging over the distribution of the parameters of tunnel systems[3] likewise leads to a temperature-independent correction to the heat capacity, thus indicating that such a procedure is wrong at low probabilities of tunnel transitions between two states. It must be noted that the same averaging procedure leads to a logarithmic dependence of the heat-capacity component linear in T on the duration of the experiment. No such dependence was actually observed in experiment, however.[5]

Finally, it should be noted that the expression for the heat capacity acquires a term proportional to $PT^2\omega_m^{-1} \times \ln(\omega_m/T)$, which can become comparable with the Debye heat capacity C_D at temperatures of the order of tenths of a degree.

The plan of this article is the following. In Sec. 2 the corrections, which arise in first-order perturbation theory, to the spacing of the levels and to their population differences, are calculated and expressions are given for the generalized susceptibilities of two-level systems. A number of mathematical details are relegated to the Appendix. The results of this section are valid for any two-level system in a phonon field, and no details peculiar to glasses are used. The use of the results for an analysis of the low-temperature properties of glasses is contained in the third section.

2. GENERALIZED SUSCEPTIBILITIES

Following Ref. 3, we write down the Hamiltonian that describes the interaction of a two-level system with phonons in the form

$$H={}^1/_2 E\sigma_z+B_x\sigma_x+B_z\sigma_z, \tag{4}$$

where σ_α are Pauli matrices that describe the two-level system, $B_{x,z}$ are the strain-tensor components acting on the pseudospin and multiplied by the appropriate strain potential constants. Generally speaking the operators B_x and B_z are independent. A connection between them can exist only within the framework of some particular detailed model. Thus, in the so-called model of standard tunnel states considered in Refs. 3 and 4 we have

$$B_z=\frac{\Delta}{E}B, \quad B_x=\frac{\Delta_0}{E}B, \quad E=(\Delta^2+\Delta_0^2)^{1/2}, \tag{5}$$

where E is the level spacing, Δ is the parameter of the asymmetry of the potential wells, and Δ_0 is the energy of the tunnel interaction between them.

All the observable physical quantities can be calculated if we know the generalized susceptibilities defined by the equations

$$\chi_{pq}(\omega)=i\int_0^\infty dt e^{i\omega t}\langle[\sigma_p(t),\sigma_q(0)]\rangle. \tag{6}$$
$$p,q=x,z,$$

To calculate these susceptibilities we shall use the method developed in I. This method is based on the Abrikosov pseudofermion formalism,[10] according to which the pseudospin operators σ_p are written in the form $a_\mu^+\sigma_p^{\mu\nu}a_\nu$, where a_μ^+ and a_ν are the creation and annihilation operators for pseudofermions with spin projections ± 1.

In I, the temperature diagrams were analytically continued to real frequencies and a diagram technique was formulated for such frequencies. This technique reduces to the following: straight lines correspond to Green's functions of pseudofermions (particles), and wavy lines to Green's functions of phonons. The phonon-field propagator is the imaginary part of the retarded Green's function, taken with a minus sign and multiplied by $\pi^{-1}N(\omega)$, where $N(\omega)$ is the Planck function, of the operators B. Integration with unity weight is carried out over all the phonon frequencies, which are chosen such that they enter in the arguments of the particle Green's functions with positive sign. Generally speaking there are three Green's functions: $D_{zz}=D_0$, $D_{zx}=D_1$, and $D_{xz}=D_2$; correspondingly there are three phonon-field propagators:

$$\Phi_i(\omega)=\pi^{-1}N(\omega)d_i(\omega), \quad d_i(\omega)=-\operatorname{Im}D_i(\omega). \tag{7}$$

In the standard tunnel model, by virtue of (5), all the d_i are expressed in terms of the quantity d corresponding to the Green's function of operators B and coinciding with (1b). Conversely, for a defect in a crystal we have $D_{zz}=0$ (the results of I for two-level systems with degeneracy correspond precisely to this case). Just as in I, we begin with a calculation of the pseudofermion Green's function. In I, owing to the degeneracy, a finite level width appeared only in second-order perturbation theory. In our case, however, where there is no degeneracy, a finite width obtains already in first order, to which we confine ourselves. Altogether there are four Green's functions ($\alpha,\beta=\pm 1$), and the diagonal ones will be designated simply g_+ and g_-.

The first-order diagrams for the self-energy parts $g_{\alpha\beta}$ are shown in Fig. 1, where the numbers 0, 1, and 2 above the phonon lines designate the propagators (7) to which they correspond.

The zero-order Green's functions are of the form

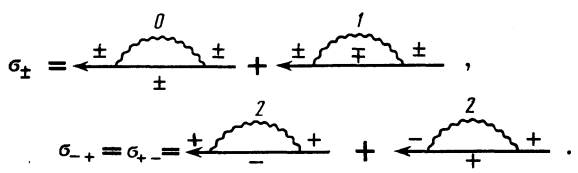

FIG. 1.

$$g_{\pm}^{(0)}=(\omega\mp E/2-\lambda+i\delta)^{-1};\quad g_{-+}^{(0)}=0, \tag{8}$$

where λ is the chemical potential taken with a minus sign. After completing all the calculations it is necessary to go to the limit $\lambda\to\infty$. As shown in I, after the analytic continuation of the temperature diagrams it is possible to shift the energy origin by λ, after which λ drops out of all the expressions. Calculations perfectly analogous to those in I lead to the following result for the diagonal Green's functions:

$$g_{\pm}(\omega)=Z_{\pm}(\omega-x_{\pm})^{-1}+g_{2\pm}(\omega), \tag{9a}$$

$$Z_{\pm}=1-\frac{1}{\pi}\int dx\frac{\Phi_0(x)}{x^2}-\frac{1}{\pi}\int dx\frac{\Phi_1(x)}{(x\pm E+i\delta)^2}, \tag{9b}$$

$$x_{\pm}=\pm\frac{E}{2}+\sigma_{\pm}\left(\pm\frac{E}{2}\right)=\pm\frac{E}{2}+\Delta_{\pm}+i\gamma_{\pm}, \tag{9c}$$

$$\gamma_{\pm}=\Phi_1(\mp E)=d_1(E)\left[N(E)+\frac{1}{2}\pm\frac{1}{2}\right], \tag{9d}$$

$$\Delta_{\pm}=\frac{1}{\pi}\int\frac{dx\,\Phi_0(x)}{x}+\frac{1}{\pi}\int\frac{dx\,\Phi_1(x)}{x\pm E}, \tag{9e}$$

$$g_{2\pm}(\omega)=\frac{1}{\pi}\int\frac{dx\,\Phi_0(x)}{x^2(x\mp E/2+\omega+i\delta)}+\frac{1}{\pi}\int\frac{dx\,\Phi_1(x)}{(x\pm E+i\delta)^2(x\pm E/2+\omega+i\delta)}. \tag{9f}$$

We see that, just as in I, the Green's functions $g_{\pm}$ have pole parts $g_{1\pm}$ and smooth background additions $g_{2\pm}$. The residue at the pole $Z_{\pm}$ is now complex because the last term of (9b) is complex. To calculate this term it must be integrated by parts, after which the real part is found to be an integral in the sense of principal value, and the imaginary part of $Z_{\pm}$ will be $\gamma'_{\pm}=\Phi'_1(\mp E)$. We note that $E\ll\omega_m$ we have $\mathrm{Re}\,Z_{\pm}=1-O(J)$, where J is the parameter (1a) of the perturbation theory. From (9c) it follows that the interaction renormalizes the distance between the levels

$$E'=E+\varepsilon=E\left(1-\frac{2}{\pi}\int\frac{dx\,\Phi_1(x)}{x^2-E^2}\right). \tag{10}$$

In the limit $E\ll\omega_m$ we have $\varepsilon=-2EO(J)$, i.e., the renormalization is again small if J is small. It is likewise easy to verify that the functions $g_{\pm}$ satisfy the sum rule

$$\pi^{-1}\int dx\,\mathrm{Im}\,g_{\pm}(x)=-1.$$

Knowledge of $g_{\pm}$ permits calculation of the numbers of pseudofermions with up and down spins, determined by the expression

$$N_{\pm}=\frac{1}{\pi}\int dx\,n(x)\,\mathrm{Im}\,g_{\pm}(x)=e^{-\lambda/T}\frac{1}{\pi}\int dx e^{-x/T}\,\mathrm{Im}\,g_{\pm}. \tag{11}$$

The quantities $N_{\pm}$ enter in the definiation of the normalizing factor $N=N_{+}+N_{-}$, which enters in the definition of the susceptibilities (see I and below) and of the average "magnetization"

$$\sigma=-\langle\sigma_z\rangle=N^{-1}(N_{-}-N_{+}). \tag{12}$$

After simple calculations we obtain

$$N=e^{E/2T}+e^{-E/2T}, \tag{13a}$$

$$\sigma=\mathrm{th}\frac{E}{2T}+\frac{2n_E}{\pi}\int\frac{dx\Phi_1'(x)}{x+E}-\frac{2(1-n_E)}{\pi}\int\frac{dx\Phi_1'(x)}{x-E}, \tag{13b}$$

where $n_E=[\exp(E/T)+1]^{-1}$ and the factor $\exp(-\lambda/T)$ is omitted from the expression for N. Here and elsewhere it is necessary to use in the terms of zeroth order in Φ for the energy E the renormalized value E' (10). It follows from (13b) that the interaction decreases the magnetization. In the case $E\ll T\ll\omega_m$ we have then

$$\sigma=\frac{E}{2T}\left[1-\frac{T}{\omega_m}O(J)\right];$$

if $T\ll E\ll\omega_m$ we have $\sigma=1-2J_1$, and finally at $E\gg\omega_m$ we have

$$\sigma=\left[1-O\left(J_1\left(\frac{\omega_m}{E}\right)^2\right)\right]\mathrm{th}\frac{E}{2T}.$$

We shall need subsequently the off-diagonal Green's functions. In our approximation we have for them

$$g_{-+}(\omega)=g_{+-}(\omega)=g_{-}(\omega)\sigma_{-+}(\omega)g_{+}(\omega),$$

$$\sigma_{-+}(\omega)=\frac{1}{\pi}\int dx\,\Phi_2(x)\left[\left(x+\omega-\frac{E}{2}+i\delta\right)^{-1}-\left(x+\omega+\frac{E}{2}+i\delta\right)^{-1}\right]. \tag{14}$$

We proceed now to an analysis of the susceptibilities. We consider first the transverse susceptibility χ_{xx}. We calculate it with the same accuracy as the Green's function: we determine the first perturbation-theory correction to the residue of the resonant terms, and the smooth "background" part linear in the interaction, similar to that calculated in I. As always, it is most convenient in such cases to determine the circular susceptibility components $\chi_{\mp\pm}$ and $\chi_{\mp\mp}$, where $\sigma_{\pm}=1/2(\sigma_x\pm i\sigma_y)$, and then reconstruct the Cartesian components. It is necessary in this case to calculate only χ_{-+} and χ_{--}, since the following equalities are obvious:

$$\chi_{+-}(E)=\chi_{-+}(-E);\quad \chi_{++}(E)=\chi_{--}(-E). \tag{15}$$

The expression we need later for the retarded susceptibility in terms of the corresponding vertex part is given by the Ginzburg formula[I,11]:

$$\chi(\omega)=\frac{1}{2\pi iN}\int dx\,e^{-x/T}\{g(x+\omega)\Gamma(x+\omega,x)g(x)-g(x+\omega)\Gamma(x+\omega,x)\times g^*(x)-g^*(x)\Gamma(x,x-\omega)g^*(x-\omega)+g(x)\Gamma(x,x-\omega)g^*(x-\omega)\}, \tag{16}$$

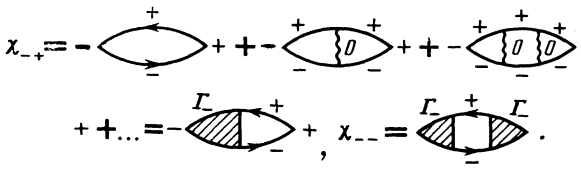

FIG. 2.

where the Green's functions to the left and to the right of the vertices Γ correspond to the incoming and outgoing lines, and in each of the terms the signs of the imaginary parts of the arguments of the vertices are the same as those of the corresponding g-functions.

To calculate χ_{-+} it suffices obviously to use the ladder approximation (see Fig. 2). In this case the integration contour for the first and third terms of (16) passes on the same side of the singularities of the Green's function, so that the first two diagrams can be used for the corresponding vertices. On the case of the two other terms, however, the integration contour is "clamped" between the singularities, and it is necessary to solve the corresponding integral equations. For the second term we have thus

$$\Gamma_-(x+\omega, x) = 1 - \frac{1}{\pi}\int dx_1\, \Phi_0(x_1-x)\, g_+(x_1+\omega)\, g_-^*(x_1)\, \Gamma_-(x_1+\omega, x_1). \tag{17}$$

This equation is solved by the method of I. The singularities of the function Φ_0 lie far from the real axis (we assume that $\omega_m, T, E \gg \gamma_\pm$). We can therefore transform the integration contour into the contour C_+ (see Fig. 3), separating the residue at the pole $g_-^*(x)$. After this the integral along C_+ must be calculated in the lowest order of perturbation term and return to the real axis from above. This yields

$$\Gamma_-(x+\omega, x) = 1 - \frac{1}{\pi}\int \frac{dx_1\, \Phi_0(x_1-x)}{(x_1-E/2+\omega+i\delta)\,(x_1+E/2+i\delta)} - \frac{2iZ_-^*\, \Phi_0(x_-^*-x)\, \Gamma_-(x_-^*+\omega, x_-^*)}{\omega-E+i\gamma}, \tag{18a}$$

$$\gamma = d_1(E)\ \mathrm{cth}\ (E/2T), \tag{18b}$$

$$\Gamma_-(x_-^*+\omega, x_-^*) = 1 - \frac{1}{\pi}\int \frac{dx\, \Phi_0(x)}{x(x-E+i\delta)}. \tag{18c}$$

We have taken it into account here that $\Phi_0(0) = 0$. Similar formulas hold also for Γ_- from the last term of (16); in this case the integral along the real axis must be transformed in to an integral along the contour C_- (Fig. 3). We note also that the first two terms of (18a) coincide with the expression for the vertex in the first term of (16), while (18b) is the customarily employed width of the resonant level $1/T_1$; in addition, it must be remembered that these expressions contain the renormalized energy (10).

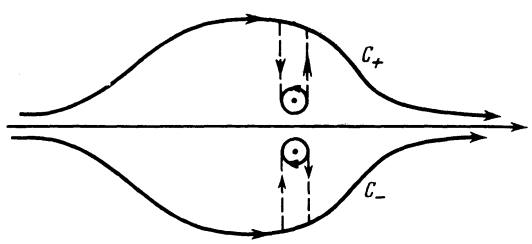

FIG. 3.

The equations obtained for the vertices must be substituted in (16), and the integration contours must be transformed in the manner described above. In those terms in which $\Gamma_- \to 1$ it is necessary, prior to returning to the real axis, to represent the function $g_\mp(x)$ in the form $g_\mp^{(0)1}(x) + g_\mp^{(0)2}(x)\sigma_\mp(x)$ (and the same for $g_\pm^*(x)$). Next, the pole parts in the terms containing the resonant denominator must be separated from the smooth background. The result is

$$\chi_{-+}(\omega) = \chi_{-+}^R(\omega) + \chi_{-+0}^F(\omega) + \chi_{-+1}^F(\omega), \tag{19a}$$

$$\chi_{-+}^R(\omega) = -\mathrm{th}\frac{E}{2T}\left\{1 - \frac{1}{\pi}\int \frac{dx\Phi_0(x)}{x^2} - \frac{1}{\pi}\int dx\, \Phi_1'(x) \times\left(\frac{1}{x+E}+\frac{1}{x-E}\right) - i\,\mathrm{cth}\frac{E}{2T}\, d_1'(E)\right\}(\omega-E+i\gamma)^{-1}, \tag{19b}$$

$$\chi_{-+0}^F = -\frac{4}{\pi}\int \frac{dx\, \Phi_0(x)}{x^2}\left(\frac{1-n_E}{x+\omega-E+i\delta}+\frac{n_E}{x-\omega+E-i\delta}\right), \tag{19c}$$

$$\chi_{-+1}^F = \frac{1}{\pi}\int \frac{dx\, d_1(x)}{(x+E+i\delta)^2(x+\omega+i\delta)}. \tag{19d}$$

Here, in (19d), the function $d_1(x)$ is the result of the property $\Phi_i(-x) = \Phi_i(x) + d_i(x)$, which is due to $d_i(x)$ being odd, while the second-order poles stem from the expansion of the Green's functions on the contours $C_\pm$ in powers of the interaction. As noted above, it is easy to get rid of such poles by integrating by parts. In particular, derivatives of Φ_1 and d_1 appeared in (19b) as a result of such an integration.

The physical meaning of the background terms is the following: an external-field quantum of frequency ω causes a transition between levels and an additional emission or absorption of a phonon. The amplitude of the resonant term must categorically be smaller than in the zeroth order; this follows from the sum rule discussed below.

The set of diagrams for χ_{--} is also shown in Fig. 2. It is easy to verify here, using (18a), that there is no need to consider the renormalization of the vertices Γ_-. As a result we obtain after calculation perfectly similar to those described above

$$\chi_{--}(\omega) = \chi_{--}^R(\omega) + \chi_{--}^F(\omega), \tag{20a}$$

$$\chi_{--}^R(\omega) = \mathrm{th}\frac{E}{2T}\left(\frac{1}{\omega+E+i\gamma}-\frac{1}{\omega-E+i\gamma}\right)\frac{1}{\pi}\int \frac{dx\Phi_1(x)}{(x+E)(x-E)} - \frac{id_1(E)}{2E}\left(\frac{1}{\omega+E+i\gamma}+\frac{1}{\omega-E+i\gamma}\right), \tag{20b}$$

$$\chi_{--}^F(\omega) = -\frac{1}{\pi}\int \frac{dx\, d_1(x)}{(x-E+i\delta)(x+E+i\delta)(x+\omega+i\delta)}. \tag{20c}$$

Equations (19), (20), and (15) enable us to calculate the Cartesian components of the transverse susceptibility. In our approximation we have $\chi_{--}=\chi_{++}$, and therefore $\chi_{xy}=-\chi_{yx}$, as usual. However, $\chi_{xx}\neq\chi_{yy}$. Simple calculations yield

$$\chi_{xx}^{R}=-Z_R \operatorname{th}\frac{E}{2T}\left(\frac{1}{\omega-E+i\gamma}-\frac{1}{\omega+E+i\gamma}\right)$$
$$+i\left(d_1'(E)-\frac{d_1(E)}{E}\right)\left(\frac{1}{\omega-E+i\gamma}+\frac{1}{\omega+E+i\gamma}\right), \quad (21a)$$

$$Z_R=1-\frac{4}{\pi}\int\frac{dx\,\Phi_0(x)}{x^2}-\frac{1}{\pi}\oint dx\,\Phi'(x)$$
$$\times\left(\frac{1}{x+E}+\frac{1}{x-E}\right)+\frac{2}{\pi}\oint\frac{dx\,\Phi_1(x)}{(x+E)(x-E)}\approx 1-4J_0, \quad (21b)$$

$$\chi_{xx0}^{F}=-\frac{4}{\pi}\int\frac{dx\,\Phi_0(x)}{x^2}\Big[(1-n_E)$$
$$\times\left(\frac{1}{x-E+\omega+i\delta}+\frac{1}{x-E-\omega-i\delta}\right)$$
$$+n_E\left(\frac{1}{x+E+\omega+i\delta}+\frac{1}{x+E-\omega-i\delta}\right)\Big], \quad (21c)$$

$$\chi_{xx1}^{F}=\frac{1}{\pi}\int\frac{dx\,d_1(x)}{(x+\omega+i\delta)}\left[\frac{1}{(x+E+i\delta)^2}\right.$$
$$\left.+\frac{1}{(x-E+i\delta)^2}-\frac{2}{(x+E+i\delta)(x-E-i\delta)}\right]. \quad (21d)$$

We shall not write out the remaining transverse components, which we do not need. We note only that χ_{yy} is obtained from χ_{xx} by reversing the signs of the last terms in the expressions for Z_R and χ_{xx1}^{F}. The approximate equality in the right-hand side of (21b) takes place at $\omega_m \gg E$. It is also obvious that at $\omega=\pm E$ the susceptibility χ_{dd1}^{F} is finite, since all the singularities of the integrand lie on one side of the integration contour.

In what follows we shall need the imaginary part of χ_{xx}. It must be calculated with the following taken into account. $\operatorname{Im}\chi_{xx}^{R}$ contains terms proportional to $(\omega\pm E)[(\omega\pm E)^2+\gamma^2]^{-1}$, which we shall understand hereafter to be the principal values of the quantities $(\omega\pm E)^{-1}$. The quantity $\operatorname{Im}\chi_{xx1}^{F}$ is finite at $\omega=\pm E$, but it is convenient to separate in it the terms that cancel out the aforementioned principal values, after which the remainder as $\omega\to\pm E$ becomes infinite and must now be understood in the sense of the principal value. As a result we have for $\operatorname{Im}\chi_{xx}$

$$\operatorname{Im}\chi_{xx}^{R}=Z_R \operatorname{th}\frac{E}{2T}\left[\frac{\gamma}{(\omega-E)^2+\gamma^2}-\frac{\gamma}{(\omega+E)^2+\gamma^2}\right], \quad (22a)$$

$$\operatorname{Im}\chi_{xx0}^{F}=4\frac{(1-n_E)\Phi_0(E-\omega)-n_E\Phi_0(-E+\omega)}{(\omega-E)^2}$$
$$-4\frac{(1-n_E)\Phi_0(E+\omega)-n_E\Phi_0(-E-\omega)}{(\omega+E)^2}, \quad (22b)$$

$$\operatorname{Im}\chi_{xx1}^{F}=\frac{d_1(\omega)-d_1(E)}{(\omega-E)^2}+\frac{d_1(\omega)+d_1(E)}{(\omega+E)^2}-\frac{2d_1(\omega)}{(\omega-E)(\omega+E)}. \quad (22c)$$

Obviously, $\operatorname{Im}\chi_{xx}$ is an odd function of ω, as it should, and decreases rapidly at $\omega\gg\omega_m, E$.

It is well known that the generalized susceptibilities satisfy the sum rule

$$\langle AB\rangle=-\frac{1}{2\pi i}\int d\omega\, N(-\omega)[\chi_{AB}(\omega+i\delta)-\chi_{AB}(\omega-i\delta)]. \quad (23)$$

In our case $AB=\sigma_x^2=1$, and it can be easily verified, by using (22), that the sum rule does indeed hold.

We proceed now to discuss the longitudinal susceptibility χ_{zz}. Unfortunately the simple ladder approximation is insufficient for this purpose. The point is that in expression (16) for χ_{zz} the contributions from Γ_z in the second and last terms cancel out in lowest order, in contrast to the case χ_{-+}. Therefore the principal term is formally of the order of the correction linear in Φ (it is of the order of unity only at $\omega\sim 2i\gamma$), and it is consequently necessary to calculate the vertices in the next order in Φ. It is simultaneously necessary to take into account the next-order corrections also for the imaginary parts of the poles of the Green's functions $g_{\pm}$; the corresponding diagrams are shown in Fig. 4. Generally speaking, it is necessary here to take into account not only the diagonal parts D_0 and D_1 of the interaction, but also the off-diagonal part D_2. It can be shown, however, that allowance for this interaction influences only the two-phonon contribution to the longitudinal relaxation (the "Raman" processes). We therefore disregard it. The corresponding rather cumbersome calculations are given in the Appendix, but the final result is quite simple:

$$\chi_{zz}=\chi_{zz}^{rel}+\chi_{zz}^{F}, \quad (24a)$$

$$\chi_{zz}^{rel}=\frac{1}{T\operatorname{ch}^2(E/2T)}\frac{2i\gamma_{\parallel}}{\omega+2i\gamma_{\parallel}}$$
$$\times\left[1-\frac{2}{\pi}\oint dx\,\Phi_1'(x)\left(\frac{1}{x+E}+\frac{1}{x-E}\right)\right], \quad (24b)$$

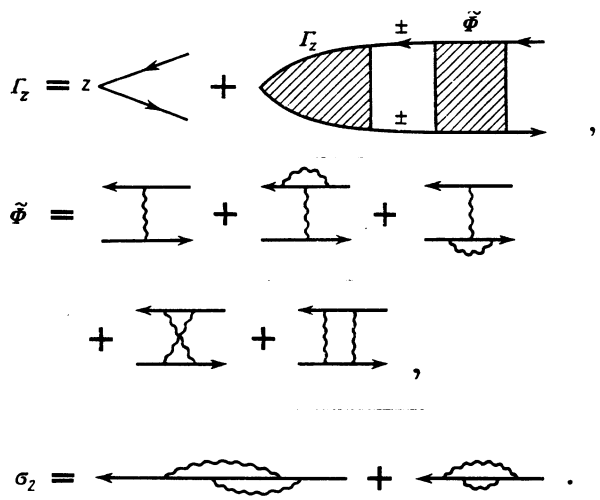

FIG. 4.

$$\chi_{zz}^{F}=-\frac{4}{\pi}\int dx\,\Phi_1(x)$$
$$\times\Big\{(1-n_E)\Big[\frac{1}{(x-E+i\delta)^2(x-E+\omega+i\delta)}$$
$$+\frac{1}{(x-E-i\delta)^2(x-E-\omega-i\delta)}\Big]$$
$$+n_E\Big[\frac{1}{(x+E+i\delta)^2(x+E+\omega+i\delta)}$$
$$+\frac{1}{(x+E-i\delta)^2(x+E-\omega-i\delta)}\Big]\Big\}. \quad (24c)$$

Here $2\gamma_\parallel = 1/T^2$ is the reciprocal time of the longitudinal relaxation. At our accuracy it coincides with the expression for 2γ, where γ is given by Eq. (18b). In the Appendix, however, $2\gamma_\parallel$ is calculated with higher accuracy, with account taken of both the renormalization of γ, which is of the order of J, and the two-phonon Raman contribution, which is of the order of T^5 at $E \ll T$. Allowance for this last contribution makes it possible to go to the limit $E = 0$. This is impossible in expression (21a) for χ_{xx}^{R}, since the Raman terms are not taken into account, and it therefore vanishes at $E = 0$.[1)]

The imaginary part of the longitudinal susceptibility can be reprsented in the following form:

$$\mathrm{Im}\,\chi_{zz}^{rel}=\frac{1}{T\,\mathrm{ch}^2(E/2T)}\frac{2\gamma\omega}{\omega^2+(2\gamma)^2}$$
$$\times\Big[1-\frac{2}{\pi}\int dx\Phi_1'(x)\Big(\frac{1}{x+E}+\frac{1}{x-E}\Big)\Big], \quad (25a)$$

$$\mathrm{Im}\,\chi_{zz}^{F}=\frac{4}{\omega^2}[(1-n_E)\Phi_1(E-\omega)-n_E\Phi_1(-E+\omega)]$$
$$+\frac{4}{\omega^2}[n_E\Phi_1(-E-\omega)-(1-n_E)\Phi_1(E+\omega)]-\frac{2\gamma}{T\omega\,\mathrm{ch}^2(E/2T)}. \quad (25b)$$

The last term in $\mathrm{Im}\,\chi_{zz}^{F}$ plays a double role: it ensures $\mathrm{Im}\,\chi_{zz}(0)=0$, and at $\omega \gg E, \omega_m$ it cancels out the first term in $\mathrm{Im}\,\chi_{zz}^{rel}$, ensuring thereby a good decrease of $\mathrm{Im}\,\chi_{zz}$ as $\omega\to\infty$. (Allowance for the second term in $\mathrm{Im}\,\chi_{zz}^{rel}$ at such ω is an exaggeration of the accuracy.) It is easy to verify also the satisfaction of the sum rule (23), whose left-hand side now contains $1-\sigma^2$, where σ is given by (13b).

It remains now to discuss the off-diagonal part of the susceptibility, which we calculate in the lowest order in the interaction Φ^2. As before, it is convenient to calculate in lieu of χ_{xz} and χ_{zx} the quantities $\chi_{\mp z}$ and $\chi_{z\pm}$, and obviously $\chi_{+z}(E)=\chi_{-z}(-E)$. Diagrams for the function χ_{-z} are shown in Fig. 5, where the lines with the point in the last two diagrams show the off-diagonal Green's functions g_{-+} (see (14)), while $\Gamma_z^{(\pm)}$ are the longitudinal vertices corresponding to incoming lines with spin projections ± 1. It is easy to verify that the equality $\chi_{-z}=\chi_{z+}$ holds also in the considered order, so that to calculate χ_{xz} and χ_{zx} it suffices to know χ_{-z}. The corresponding calculations are completely analogous to the calculations of χ_{zz}. It turns out here, first, that the resonant part of χ_{-z} is equal to zero, and second, that

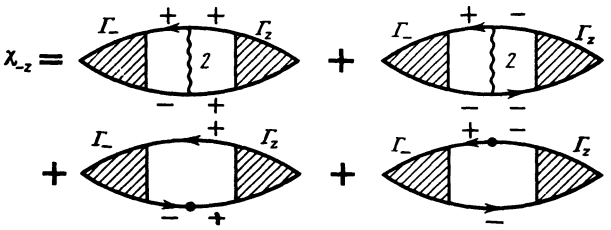

FIG. 5.

the relaxation part is of the same order as the correction term in (24b); we do not need its exact form. Finally, the background is of the form

$$\chi_{-z}^{F}=\frac{4}{\pi}\int\frac{dx\,\Phi_2(x)}{x}$$
$$\times\Big[\frac{1-n_E}{(x-E+i\delta)(x-E+\omega+\delta i)}+\frac{n_E}{(x+E-i\delta)(x+E-\omega-i\delta)}\Big]. \quad (26)$$

For the quality of subsequent interest $\chi_{xz}^{F}+\chi_{zx}^{F}$ we obtain

$$\chi_{xz}^{F}+\chi_{zx}^{F}=\frac{8}{\pi}\int\frac{dx\Phi_2(x)}{x}$$
$$\times\Big\{(1-n_E)\Big[\frac{1}{(x-E+i\delta)(x-E+\omega+i\delta)}$$
$$+\frac{1}{(x-E-i\delta)(x-E-\omega-i\delta)}\Big]$$
$$+n_E\Big[\frac{1}{(x+E-i\delta)(x+E-\omega-i\delta)}$$
$$+\frac{1}{(x+E+i\delta)(x+E+\omega+i\delta)}\Big]\Big\}, \quad (27a)$$

$$\mathrm{Im}(\chi_{xz}^{F}+\chi_{zx}^{F})=8\frac{(1-n_E)\Phi_2(E-\omega)-n_E\Phi_2(-E+\omega)}{\omega(E-\omega)}$$
$$+8\frac{(1-n_E)\Phi_2(E+\omega)-n_E\Phi_2(-E-\omega)}{\omega(E+\omega)}. \quad (27b)$$

These equations for χ_{pq} do not depend on the actual form of the operators $B_{x,z}$ in (4). The criteria for the validity of perturbaton theory, however, are different, depending on the forms of the functions $d_i(\omega)$. In the case of interest to us, that of interaction with phonons, the values of J_i are determined by a frequency region of the order of ω_m. In other cases, for example in interactions with phonons, in quasi-two-dimensional and quasi-one-dimensional cases (see I), or else in interaction with spin waves in amorphous ferromagnets, an important role is assumed by the convergence of the integrals as $\omega\to 0$. We, however, shall not discuss these questions here in greater detail.

3. PHYSICAL CONSEQUENCES

All the physical consequences, such as the corrections to the heat capacity, to the sound absorption, to the thermal conductivity, and others can be determined if one knows the phonon Green's function, which can be represented in the following manner:

$$D_{k\alpha}^{-1}(\omega)=\omega^2-\omega_{k\alpha}{}^2+\frac{\gamma_{k\alpha}{}^2k^2}{\rho}\sum_{pq}\left\langle\int_0^{E_m}dE\,P_0M_pM_q\chi_{pq}(E,\omega)\right\rangle, \tag{28}$$

where M_p are the dimensionless constants of the phonon interaction with σ_x and σ_z, and the angle brackets denote averaging over all types of tunnel states with a distribution function P_0. The integration is over all possible unrenormalized distances between the levels E, while E_m is the maximum possible distance. We shall find it convenient to use a normalization of the constants M_p in the form $\langle M_x^2P_0\rangle=\overline{P}_0$, where $\overline{P}_0$ is the average density of the tunnel states (per unit volume and in a non-unity energy interval). With this choice of the interaction we have $\Phi_0=M_z^2\Phi$, $\Phi_1=M_x^2\Phi$, and $\Phi_2=M_xM_z\Phi$. In the standard tunnel model, according to (5), $M_z=\Delta_0/E$, $M_x=\Delta_0/E$, and the averaging procedure, which is discussed in detail with Refs. 3 and 4, is conveniently written in the form

$$\bar{A}=\bar{P}_0\int_0^{E_m}dE\,E\int_{\Delta_{0\,min}}^{E}\frac{d\Delta_0}{\Delta_0(E^2-\Delta_0^2)^{1/2}}A(E,\Delta_0), \tag{29}$$

where $\Delta_{0\,\min}$ is the minimum value of the tunnel parameter Δ_0. A characteristic feature of this procedure is the logarithmic dependence of the mean values on $\Delta_{0\,\min}$ if $A(E,0)\neq0$. We recall also that for tunnel defects in crystals there is no integration with respect to E or averaging, and in addition only diagonal terms with $p=q=x,z$ are contained in the sum over p and q.

We proceed now to the effects connected with sound propagation. The principal result is obtained here if account is taken of two facts: the decrease of the amplitude of the resonant scattering because of the appearance of the factor Z_R in (21a) and (22a), and the renormalization of the spectrum (10) of the tunnel states. The renormalization causes the mean free path $l_\alpha(\omega)$ of the phonons to take the form

$$\frac{1}{l_\alpha(\omega)}=Z_{ph}\frac{1}{l_\alpha^{(0)}(\omega)}, \tag{30a}$$

$$Z_{ph}=\left\langle Z_R\left(1-\frac{d\varepsilon}{dE}\right)_{E=0}\right\rangle=1-\langle 4M_x{}^2M_z{}^2-2M_z{}^4\rangle J, \tag{30b}$$

$$\frac{1}{l_\alpha^{(0)}(\omega)}=\frac{\pi\omega}{\rho c_\alpha{}^3}\bar{P}_0\,\mathrm{th}\frac{\omega}{2T}. \tag{30c}$$

Here $l_\alpha^{(0)}$ is the standard expression for the unrenormalized mean free path.[3] An expression for Z_{ph} in terms of $M_{x,z}$ exists in the limit $E\ll\omega_m$. In the standard tunnel model, by virtue of (5) and (29), we have $Z_{ph}=1$, i.e., there is no renormalization in the first-order perturbation theory considered by us. Such a renormalization does exist in any other model. It follows also from (30) that the phonon thermal conductivity is also renormalized by the factor Z_{ph}

$$\varkappa=Z_{ph}^{-1}\varkappa_0,\quad \varkappa_0=\frac{\rho}{6\pi}\left(\sum_\alpha\frac{c_\alpha}{\bar{P}_0\gamma_\alpha{}^2}\right)T^2, \tag{31}$$

where $\varkappa_0$ is the thermal conductivity without allowance for the interaction effects (see, e.g., Ref. 4).

We note also that the influence of the background on the sound absorption was recently investigated in Ref. 8. It was shown there that it must be taken into account only if the resonant absorption is suppressed by a sufficiently strong sound signal.

We proceed now to discuss the influence of the interaction on the heat capacity. We recall first an expression from the standard tunnel model[3,4]

$$C=\frac{\pi^2}{6}\bar{P}_0T\left(\frac{1}{2}\ln\frac{4\tau_{max}}{T_{1\,min}}\right), \tag{32}$$

where $T_{1\min}$ is the minimum time of transverse relaxation at the temperature T and corresponds to $\Delta_0^2=E^2=4T^2$, while $\tau_{\max}$ is equal to the duration t of the experiment if it is shorter than $T_{1\max}=T_{1\min}(T/\Delta_{0\,\min})^2$ and to $T_{1\max}$ in the opposite limiting case. As already noted in the Introduction, no such dependence on the duration of t of the experiment was observed in experiment.[5] We shall therefore use below in place of (32) the formula $C=(1/6)\pi^2\overline{P}_0T$, where $\overline{P}_0$ is the density of states, which we assume to be independent of energy. Our purpose is to determine $\overline{P}$ in the lowest order of perturbation theory. This is simplest to do by using expression (10) for the renormalization of the energy E. It turns out then that

$$\bar{P}=\bar{P}_0\left\langle 1-\frac{d\varepsilon}{dE}\Big|_{E=0}\right\rangle=P_0(1+2J). \tag{33}$$

(We recall that we have defined M_x such that $\langle M_x^2\rangle=1$.)

Thus, by softening the spectrum in first order, the interaction increases the density of the tunnel states that enter in the expression for that part of the heat capacity which is linear in T. Therefore, if we forget the effects of the interaction, the tunnel-state density determined from acoustic experiments should in this approximation be less than the same density determined from heat-capacity data. This conclusion should remain in force in the general case. The point is that both (30) and (33) contain the factor $1-d\varepsilon/dE$ that renormalizes the density of states. In addition, Z_{ph} (30) contains the quantity Z_R, which is always less than unity. In fact, when account is taken of higher orders of perturbation theory, the background part of χ_{xx} should increase, for it acquires a multiphonon contribution on top of the single-phonon one. Since χ_{xx} is normalized by the sum rule (23), an increase of the background should lead in this case to a decrease of the resonant paper, i.e., of Z_R. It follows from the foregoing that, in our opinion, the difference in the values of P determined from different experiments is natural. It seems to us therefore that to explain this difference there is at present no need for introducing anomalous tunnel states, as was done in Refs. 3 and 4.

We proceed now to a more detailed analysis of the corrections to the heat capacity. We use for this purpose a known formula for the correction to the free energy, in the form of an integral with respect to the interaction,[13] introducing formally in place of the constants $\gamma_{k\alpha}$ the quantity[2)] $g\gamma_{k\alpha}$. After changing from discrete to real frequencies (see I) and using (28), we have in the approximation linear in P_0

$$\Delta F=\int_0^1 dgg$$

$$\times\sum_{k\alpha}\frac{k^2\gamma_{k\alpha}}{\rho\pi}\int_{-\infty}^{\infty}dxN(x)\int_0^{E_m}dE\left\langle P_0M_pM_q\,\mathrm{Im}\left[\frac{\chi_{pq}(E,x)}{x^2-\omega_{k\alpha}{}^2}\right]\right\rangle. \tag{34}$$

With the aid of (1b) we obtain from (34)

$$\Delta F=\int_0^1 dgg\frac{1}{\pi^2}\int_{-\infty}^{\infty}dxdx'd(x')\frac{N(x)-N(x')}{x-x'}$$

$$\times\int_0^{E_m}dE\langle P_0M_pM_q\,\mathrm{Im}\,\chi_{pq}(E,x)\rangle. \tag{35}$$

It is convenient to transform this equation by using the sum rule (23). It is necessary for this purpose to separate x'^{-1} from $(x-x')^{-1}$. The corresponding term is easily transformed, with the aid of (23) and of the fact that Im χ is an odd function of x, to the following form:

$$\Delta F_1=-\left[\frac{1}{2}+\int_0^1 dgg\int_0^{E_m}dE\langle P_0(\sigma_z+\sigma)^2M_z{}^2\rangle\right]\frac{2}{\pi}\int_0^{\infty}\frac{dxd(x)}{x}, \tag{36}$$

where σ is the "magnetization" (13b). In the remaining term of the numerator there appears a factor x, thanks to which the relaxation parts of Im χ_{zz} and Im $(\chi_{xz}+\chi_{zx})$ make a negligibly small contribution. It is therefore necessary to take into account only the resonant part of Im χ_{xx} and the background terms (22b), (22c), (25b), and (27b).

In the lowest order in the interaction the resonant part yields expression (33). Next there appear in the heat capacity terms of the order of

$$\bar P_0Td_0T^2\left[A\ln\frac{\omega_m}{T}+B\ln\frac{E_m}{T}\right],$$

where $A\sim B\sim 1$ stem from the logarithmic renormalization of the speed of sound on account of the resonant interaction and the irregular correction to the density of states. The large logarithm notwithstanding, the smallness of $\bar P_0$ makes these terms very small compared with the Debye heat capacity C_D and therefore of no interest. Finally, the contribution from the background contains terms proportional to T and T^3. The former are next-order corrections to expression (33) and are therefore of no interest to us here, while the latter are also small because $\bar P_0$ is small. It remains to consider expression (36). It describes the contribution to the energy from the fluctuations of the "spin." If we replace σ in it by the first term of (13b) we obtain a term linear in T that makes no contribution to the heat capacity. This, however, is not true in the case of the "standard tunnel state" model, for which the corresponding term is proportional to $(-T)\ln(T/\Delta_{0\,\min})$. As a result, the contribution to the heat capacity is independent of temperature at $T>\Delta_{0\,\min}$, in strong contradiction to experiment. This indicates once more that this model does not correspond to reality.

At $T=0$ the expression for the interaction-induced corrections to σ takes the form $a+bE+cE^2\ln(\omega_m/E)$. If we substitute this expansion in (36), the first term makes no contribution to the heat capacity, the second makes a linear contribution, and the third yields finally, after accurate calculations

$$C_2=G\bar P_0T\left(Td_0\int_0^{\infty}\frac{dxd(x)}{x}\right)\langle M_z{}^2M_x{}^2\rangle\ln\frac{\omega_m}{T},$$

$$G=\frac{9}{\pi^2}\left[6\zeta(3)+\int_0^{\infty}\frac{dxx^2}{(e^x+1)^2}\right]. \tag{37}$$

This is the heat capacity of the magnetization fluctuations and is due to the coupling of two-level systems with phonons.

We see thus that the interaction gives rise to a heat-capacity correction proportional to $\bar P_0T^2\ln(\omega_m/T)$. This correction is small compared with the linear part of the heat capacity, but at temperatures on the order of tenths of a degree it can be comparable with the Debye heat capacity C_D. We note also that the terms proportional to $E^2\ln(\omega_m/E)$ appeared in the expressions for σ and ε because of the single-phonon intermediate state. It is easy to verify that intermediate states with a large number of phonons contain in the expansion in E the quantity $\ln(\omega_m/E)$ multiplied by a higher power of E (the two-phonon $E^4\ln(\omega_m/E)$). Therefore an expression of the type (37), but with a renormalized amplitude, should take place also in the case when perturbation theory does not hold. In the general case, therefore, the low-temperature heat capacity of glasses should be of the form

$$C=\frac{\pi^2}{6}\bar PT+a\bar PT\frac{T}{\omega_m}\ln\frac{\omega_m}{T}+C_D;\qquad a\sim 1. \tag{38}$$

Thus, the customarily employed approximation of the heat capacity, $C=C_1T+C_3T^3$ or $C=C_1'T^{1+\alpha}$, should not fit well the experimental data. It should be noted that indications of the presence of a heat-capacity contribution proportional to T^2 are contained in Refs. 5 and 12. In addition, the theoretical data of Ref. 13 (Fig. 9 of Ref. 13) are also in qualitative agreement with (38).

In conclusion, the author thanks Yu. N. Skryabin for a large number of discussion of the questions touched upon in this article. It is also a pleasure to thank V. L. Gurevich and D. A. Parshin for an opportunity to read Ref. 8 prior to publication and for interesting discussions, and to E. M. Pavlenko for much help in preparing the paper.

APPENDIX

We demonstrate now how to calculate the longitudinal susceptibility. As explained in the main text, the simple ladder approximation is insufficient for the calculation of the vertex part Γ_z and it is necessary to take into account the diagrams shown in Fig. 4. We consider this question in greater detail. First, since σ_z has two nonzero matrix elements, the product $gg\Gamma$ in (16) must be regarded as the difference $g_+\Gamma_z^{(+)}g_+-g_-\Gamma_z^{(-)}g_-$. Next, if the second and third terms of (16) are transformed as described above, using the contours of $C_\pm$ (Fig. 3), and expression (9) is used for the Green's functions, the following terms are separated:

$$-\frac{|Z_+|^2[e^{-x_+^*/T}\Gamma_z^{(+)}-e^{-x_+/T}\tilde{\Gamma}_z^{(+)}]}{N(\omega+2i\gamma_+)}$$

$$+\frac{|Z_-|^2(e^{-x_-^*/T}\Gamma_z^{(-)}-e^{-x_-/T}\tilde{\Gamma}_z^{(-)})}{N(\omega+2i\gamma_-)}, \quad \text{(A1a)}$$

$$\Gamma_z^{(\pm)}=\Gamma_z^{(\pm)}(x_\pm^*+\omega, x_\pm^*), \quad \tilde{\Gamma}_z^{\pm}=\Gamma_z^{(\pm)}(x_\pm, x_\pm-\omega). \quad \text{(A1b)}$$

If we neglect the difference between $x_\pm$ and $x_\pm^*$, i.e., the damping $\gamma_\pm$, and calculate the expression for $\Gamma_z^{(\pm)}$ in the simple ladder approximation, it turns out that (A1a) is equal to zero. It is therefore necessary to calculate Γ_z more accurately. The gist of our approximation is to determine Γ_z both at $\omega \gg \gamma_\pm$ and at $\omega \sim \gamma_\pm$ with account taken of the terms linear in the interaction. This leads formally to the need of calculating Γ_z with allowance for terms of order Φ^2. But then these terms are important only if $\omega \sim \gamma_\pm$. We consider for the sake of argument the vertex $\Gamma_z^{(+)}(x+\omega, x)$ which enters in the second term of (16). The equation for it is

$$\Gamma_z^{(+)}(x+\omega, x)=1+\frac{1}{\pi}\int dx_1\tilde{\Phi}_{++}(x_1, x)g_+(x_1+\omega)g_+^*(x_1)$$
$$\times\Gamma_z^{(+)}(x_1+\omega, x_1)$$
$$+\frac{1}{\pi}\int dx_1\tilde{\Phi}_{-+}(x_1, x)g_-(x_1+\omega)g_-^*(x_1)\Gamma_z^{(-}(x_1+\omega, x_1). \quad \text{(A2)}$$

Transforming the integrals along the real axis into integrals along the contour C_+ (Fig. 3) we obtain

$$\Gamma_z^{(+)}(x+\omega, x)=1+\frac{1}{\pi}\int_{C_+} dx_1\tilde{\Phi}_{++}(x_1, x)g_+(x_1+\omega)g_+^*(x_1)$$
$$\times\Gamma_z^{(+)}(x_1+\omega, x_1)+\frac{1}{\pi}\int_{C_+} dx_1\tilde{\Phi}_{-+}(x_1, x)g_-(x_1+\omega)g_-^*(x_1+\omega)$$
$$\times\Gamma_z^{(-)}(x_1+\omega, x_1)+2i\tilde{\Phi}_{++}(x_+^*, x)|Z_+|^2\frac{\Gamma_z^{(+)}}{\omega+2i\gamma_+}$$
$$+2i\tilde{\Phi}_{-+}(x_-^*x)|Z_-|^2\frac{\Gamma_z^{(-)}}{\omega+2i\gamma_-}, \quad \text{(A3)}$$

where $\Gamma_z^{(\pm)}$ are defined by Eqs. (A1b). In these formulas, $\gamma_\pm = -\operatorname{Im} x_\pm$ must be considered with allowance for the second-order perturbation-theory diagrams shown in Fig. 4. The corresponding result is

$$-\operatorname{Im} x_\pm=\gamma_\pm^{(1)}=\gamma_\pm^{(0)}\bar{Z}+(\varphi_\pm^{(0)}+\gamma_{\pm R}^{(0)})|Z_\pm|^2, \quad \text{(A4a)}$$

$$\bar{Z}=|Z_+Z_-|-\frac{2}{\pi}\int dx\left[\frac{\Phi_0(x)}{x^2}-\frac{\Phi_1(x)}{x^2-E^2}\right], \quad \text{(A4b)}$$

$$\varphi_\pm^{(0}=\frac{1}{\pi}\int dx\left\{\frac{[\Phi_1(x)\Phi_1(-x)]'}{x\pm E}-\frac{\Phi_1(x)\Phi_1(-x)}{x^2-E^2}\right\}, \quad \text{(A4c)}$$

$$\gamma_{\pm R}^{(0)}=\frac{4}{\pi}\int\frac{dx\Phi_0(x)\Phi_1(-x\mp E)}{x^2}, \quad \text{(A4d)}$$

where the $\gamma_\pm^{(0)}$ are defined by Eq. (9d), $\bar{Z}$ renormalizes the single-phonon contribution to the damping, and the last two terms in (A4a) give the two-phonon contribution.

The integral terms in (A3) are transformed in the following manner. It is necessary again to substitute in them expressions for $\Gamma_z(x+\omega, x)$. This separates single integrals with respect to C_+, in which $\Gamma_z^\pm$ is replaced by ± 1, and double integrals in which the second integration is along the real axis. It is convenient to transform these last integrals into integrals with respect to C_+, separating again the contribution from the residue. This contribution, just as in I, takes into account the nonsingular part of the second iteration of the ladder diagram and should be included in $\tilde{\Phi}$, as shown in Fig. 4. In the double integral, however, we can now replace $\Gamma_z^{(\pm)}$ by ± 1 and put $\omega = 0$. It is now easy to obtain the following equations for $\Gamma_z^{(\pm)}$:

$$\Gamma_z^{(\pm)}=\pm(1+\Delta_\pm)+\frac{2i\varphi_\pm\Gamma_z^{(\pm)}}{\omega+2i\gamma_\pm}+\frac{2i\tilde{\gamma}_\pm\Gamma_z^{(\mp)}}{\omega+2i\gamma_\mp}, \quad \text{(A5a)}$$

$$\varphi_\pm=|Z_\pm|^2\{\varphi_\pm^{(0)}\mp i(\gamma_+'\gamma_--\gamma_-'\gamma_+)\};$$
$$\gamma_\pm'=\frac{d\Phi_1(x)}{dx}\Big|_{x=\pm E}, \quad \text{(A5b)}$$

$$\tilde{\gamma}_\pm=\gamma_\pm^{(0)}\bar{Z}|Z_\pm Z_\mp^{-1}|\pm i|Z_\pm|^2\gamma_\pm'(\gamma_--\gamma_+)+\gamma_{\pm R}^{(1)}|Z_\pm|^2, \quad \text{(A5c)}$$

$$\gamma_{\pm R}^{(1)}=\frac{2}{\pi}\int\frac{dx}{x^2}[\Phi_1(x)\Phi_0(-x\mp E)+\Phi_0(x)\Phi_1(-x\mp E)]. \quad \text{(A5d)}$$

The values of $\Delta_\pm$ in the lowest order in the interaction depend on ω and are given by

$$\Delta_\pm=-\frac{1}{\pi}\int\frac{dx\Phi_0(x)}{x(x+\omega+i\delta)}-\frac{1}{\pi}\int\frac{dx\Phi_1(x)}{(x\pm E+i\delta)(x\pm E+\omega+i\delta)}. \quad \text{(A6)}$$

It is also easy to verify that $\tilde{\Gamma}_z^{(\pm)}(\omega)=\Gamma_z^{(\pm)*}(-\omega)$. To calculate the relaxation part of χ_{zz} in the lowest order in the interaction it is necessary to take into account the $\Gamma_z^\pm$ obtained in the simple ladder approximation, and the increment that distinguishes Γ_z from $\tilde{\Gamma}_z$, which is obtained if only the imaginary parts are taken into account in the expressions for $\psi_\pm$ and $\tilde{\gamma}_\pm$.

Using the foregoing formulas, we can easily obtain the following expression for the determinant of Eqs. (A5a):

$$D=\omega(\omega+2i\gamma_\parallel), \quad \text{(A7a)}$$

$$\gamma_\parallel=\bar{Z}(\gamma_+^{(0)}+\gamma_-^{(0)})+\gamma_{R+}^{(1)}+\gamma_{R-}^{(1)}. \quad \text{(A7b)}$$

Here $2[\gamma_+^{(0)}+\gamma_-^{(0)}]\bar{Z}$ is the reciprocal time $1/T_2$, renormalized by the interaction, of the longitudinal single-phonon relaxation, and γ_R is the two-phonon Raman increment. At $T \ll E$ it is of the order of $d_0^2E^5$ and is consequently significant if $d_0E^2 \gtrsim 1$. This is again the same parameter whose existence was pointed out in Ref. 6. As noted there, if $d_0E^2 < 1$, it is necessary to consider processes with an arbitrary number of phonons, i.e., analyze the entire perturbation-theory series. If, however $T \gg E$, then $\gamma_R \sim d_0^2T^5$, and again perturbation theory is applicable only if $d_0T^2 < 1$. We note also that one of the roots is equal to zero, but the entire expression for χ_{zz} is so formulated that there is no infinity at all at $\omega = 0$. Substitution of the solutions that follow from

(A5) for $\Gamma_z^{(\pm)}$ in expression (16) corresponding to χ_{zz} leads after quite prolonged and cumbersome calculations to Eqs. (24).

[1)]We use the opportunity to correct an error in Eq. (25) of I. The correct expression is

$$\chi_P(\omega) = \bar{P}^2 \left\{ \frac{2i\Gamma_P Z_\varphi}{\omega + 2i\Gamma_P} + \frac{2\varphi_P f^2}{\pi\theta^2} \int \frac{dx\, x\Lambda}{x+\omega+i\delta} \right\},$$

$$Z_\varphi = 1 - \frac{2\varphi_P f^2}{\pi\theta^2} \int dx\, xN(x)\Lambda.$$

Expressions (24) go over into this formula with $\psi_P = 2$ and $\bar{P}^2 = 1$ in the limit $E = 0$ if expression (A7c) is used for $\gamma_\parallel$.

[2)]Since the number of tunnel centers is constant, we are dealing with the free energy and not with the potential Ω.

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