Intrinsic dynamics of two-level systems in glasses and the virial expansion for the Ising model in a transverse field

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The mechanism of the onset of intrinsic dynamics in dielectric and structured glasses, due to "transverse" *xx* interaction of two-level systems (TLS) with one another, is considered. It is shown that in dielectric glasses this time becomes shorter than the phonon-relaxation time already at a temperature ~0.1 K. The conditions under which a TLS loses its individuality are examined.

1. INTRODUCTION

The low-temperature properties of dielectric glasses are described at present by starting with the phenomenological two-level-system (TLS) model proposed by Anderson, Halperin, and Varma[1] and by Phillips.[2] This model is based on the premise that low-energy two-level excitations exist in glasses. Owing to the low density of these excitations, their interaction is neglected and it is assumed that thermal equilibrium is produced by their interaction with acoustic phonons. At the same time, to explain nonlinear absorption of sound and phonon-echo damping in glasses it became necessary to take TLS interaction into account.[3] A detailed theoretical analysis of these phenomena is contained in Refs. 4–6, where the corresponding experiments are cited. It was assumed in this case that both the nonlinear absorption of sound and the damping of the phonon echo are due mainly to the phase memory of two-level systems and not to direct transitions between their levels. The authors of Refs. 4–6 therefore confined themselves to "longitudinal" TLS interaction, which contains terms $\sigma_z^i\sigma_z^i$, where σ_z^i is the Pauli matrix for the pseudospin corresponding to the *i*th system. Since the longitudinal *zz* interaction conserves the spin projections of the individual TLS, it was proposed that the transitions between the levels are effected as before by direct emission and absorption of phonons. It is known,[1–4] however, that the phonon-relaxation time is proportional to T^{-3} and therefore increases rapidly with decrease of temperature. It is clear therefore that at sufficiently low temperatures the thermal equilibrium in a TLS should be due not to the phonons but to their interaction with one another. In this case thermal equilibrium is first produced in the TLS (a "spin temperature" is established), and then equilibrium sets in between the TLS and the lattice as a result of emission and absorption of phonons. To describe the establishment of equilibrium in the TLS it is necessary to take into account the interactions that conserve the spin projections of neither the individual TLS, nor the entire TLS subset. These interactions can be transverse ($\sigma_x^i\sigma_x^j$) and mixed ($\sigma_x^i\sigma_z^j$).

Unfortunately, it is impossible to solve the complete problem that encompasses all three interactions (*xx*, *xz*, *zz*). We confine ourselves therefore in the present paper to the transverse *xx* interaction, for which the nonconservation of the TLS spin projections is strongest. It is just this interaction which is expected to play the principal role in the dynamic phenomena in the TLS. In this formulation, the TLS reduces to the Ising model in a random transverse field.

It should be noted here that the problem considered has also a bearing on the so-called structured glasses, which contains molecular constructs with internal degrees of freedom, if the spacing of the levels of an individual construct is a quantity determined by the random atomic surround of the construct.

As shown in Ref. 3, TLS interaction is the result of a virtual phonon exchange and decreases as the cube of the distance. Owing to this decrease, TLS interaction cannot be treated by perturbation theory, for the principal role is played here by interaction of close systems with interatomic spacing, a situation with negligible probability. It is therefore necessary to solve first the problem of the interaction between TLS with specified spacing, and then average over the spacing. Such a program is executed in the present paper for the transverse susceptibility of two TLS: the transverse susceptibility of two TLS is determined first, followed by averaging both over the distances between them and over their energies. The result is the first term of the virial expansion of the transverse susceptibility of a TLS in powers of their density. We determine *in passim* the characteristic time of energy transfer from one TLS to another, meaning in fact the time to establish the spin temperature

$$\tau_{xx}^{-1}\sim T[(P_0\gamma^2/\rho s^2)\ln(E_m/T)]^2; \tag{1}$$

where P_0 is the density of states of the TLS at zero energy, γ the deformation potential, ρ the density of the medium, s the speed of sound, and E_m the maximum possible TLS energy. This time becomes shorter than the phonon time under the condition

$$T<T_0=P_0\gamma\left(\frac{s}{\rho}\right)^{1/2}\ln\left[\frac{E_m}{P_0\gamma}\left(\frac{\rho}{s}\right)^{1/2}\right]. \tag{2}$$

In other words, an intrinsic dynamics sets in at $T<T_0$ in the TLS. An accurate estimate, carried out in Sec. 4 for fused quatrz, yields1) $T_0\sim0.1$ K. The temperature T_0 is in the range in which f contemporary experiments are performed, a fact that must be taken into account in the interpretation of the experiments. It must be noted, however, that for a complete theoretical description of TLS at $T<T_0$ it is necessary to take into account, besides the pair interaction, also more complicated intereactions, i.e., to sum the entire virial series, but this is far beyond the scope of the present article.

In addition to the problem of glass dynamics, the problem of the onset of intrinsic dynamics in TLS with equal

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energies ("resonant" TLS) is in itself of interest. The approach developed by us permits analysis of this problem, too. In Secs. 2 and 3 we carry out all the calculations in a form suitable for the analysis of the glass problem and of the problem of TLS with equal energies. Section 4 deals with dynamics of glasses and Sec. 5 with the problem of identical TLS.

2. FORMULATION OF MODEL AND EXACT SOLUTION OF THE TLS-PAIR PROBLEM

We use the model of standard tunnel states,[1–4] according to which the initial Hamiltonian of an ensemble of low-energy excitations in glasses is

$$H=H_{ph}+H_{TLS}+H_I, \tag{3a}$$

$$H_{TLS}=\frac{1}{2}\sum_j E_j\sigma_z^j,\qquad E_j=(\Delta_j^2+\Delta_{0j}^2)^{1/2}, \tag{3b}$$

$$H_I=\sum_j\left(\frac{\Delta_{0j}}{E_j}\sigma_x^j+\frac{\Delta_j}{E_j}\sigma_z^j\right)B_j, \tag{3c}$$

$$B_j=\gamma_{\mu\nu}^j\varepsilon_{\mu\nu}(\mathbf{r}_j), \tag{3d}$$

where H_{ph} is the phonon Hamiltonian, $H_{\rm TLS}$ the Hamiltonian of the TLS produced as a result of tunneling in an asymmetric potential double well, Δ_j the asymmetry of the potential minima, Δ_{0j} the tunnel parameter, the subscript j the number of the tunnel system, and H_I the Hamiltonian of the TLS interaction with phonons. In espression (3d), $\gamma^j_{\mu\nu}$ is the strain potential tensor and $\varepsilon_{\mu\nu}$ the strain potential. It is assumed that the TLS are uniformly distributed in space, and the parameters Δ_j and Δ_{0j} are random quantities over which averaging is carried out with the aid of the equation

$$\overline{A(E,\Delta,\Delta_0)}=P_0\int_0^{E_m}dE\,E\int_{\Delta_{0\min}}^{E}\frac{d\Delta_0}{\Delta_0(E^2-\Delta_0^2)^{1/2}}A(E,\Delta,\Delta_0), \tag{4}$$

where P_0 is their density of states at $E\ll E_m$ and $\Delta_{0\,\min}$ is the minimum possible value of the tunnel parameter.

The TLS properties are described by their Green's functions, defined by the equation

$$G_{\mu\nu jk}(\omega)=-i\int_0^\infty dt\,e^{i\omega t}\langle[\sigma_\mu^j(t),\sigma_\nu^k(0)]\rangle. \tag{5}$$

It is known that in the lowest approximation in the interaction with phonons the Green's functions for individual TLS are given by

$$G_{-+}(\omega)=\text{th}(E/2T)(\omega-E+i\Gamma_{ph})^{-1}, \tag{6a}$$

$$G_{zz}(\omega)=2i\Gamma_{ph}[T\,\text{ch}^2(E/2T)(\omega+2i\Gamma_{ph})]^{-1}, \tag{6b}$$

$$\Gamma_{ph}=(\Delta_0/E)^2\tau_{ph}^{-1},\qquad \tau_{ph}^{-1}=(E^3/2\pi\rho)[(\gamma_l^2/s_l^5)+(2\gamma_t^2/s_t^5)]\text{cth}(E/2T)=(E^3/E_0^2)\text{cth}(E/2T), \tag{6c}$$

where G_{-+} are the Green's function of the operators $\sigma_\pm=(\sigma_x\pm i\sigma_y)/2$, $s_{l,t}$, the longitudinal and transverse sound velocities and $\gamma_{l,t}$ the corresponding strain potentials (their connection with the components of the tensor $\gamma_{\mu\nu}$ in (3d) is given in Ref. 5); the right-hand side of (6c) determines the characteristic energy E_0. The influence of the corrections of next order in the interaction with the phonons was considered in Ref. 7 (we shall not need these corrections).

Virtual exchange of phonons leads to interaction of the TLS[3,4]:

$$H_{int}={}^1/_2\sum_{j,k}J_{ik}[(\Delta_j\Delta_k/E_jE_k)\sigma_z^j\sigma_z^k+(\Delta_j\Delta_{0k}/E_jE_k)\sigma_z^i\sigma_x^k+(\Delta_{0j}\Delta_k/E_jE_k)\sigma_x^j\sigma_z^k+(\Delta_{0j}\Delta_{0k}/E_jE_k)\sigma_x^j\sigma_x^k]=H_{zz}+H_{xz}+H_{xx}, \tag{7}$$

where the "exchange" integral is of the form

$$J_{jk}=-\gamma_{\mu\nu}^j\gamma_{\rho\varphi}^k\nabla_{jk}^\nu\nabla_{jk}^\varphi D_{\mu\rho}^{(0)}(\mathbf{r}_{jk})=g_{jk}r_{jk}^{-3},$$
$$D_{\mu\rho}^{(0)}(\mathbf{r})=-(8\pi\rho r)^{-1}[s_l^{-2}(\delta_{\mu\rho}-r_\mu r_\rho r^{-2})+s_t^{-2}(\delta_{\mu\rho}+r_\mu r_\rho r^{-2})]. \tag{8}$$

Here $D^{(0)}_{\mu\rho}(\mathbf{r})$ is the Green's function of the atomic displacements at zero frequency. An explicit expression for g_{jk} is contained in Refs. 4 and 5. We do not need it here. Starting from the definition of g_{jk}, it is easy to verify that the following holds for arbitrary $\gamma^{i,k}_{\mu\nu}$

$$\int d\Omega_{jk}\,g_{jk}(\mathbf{r}_{jk})=0. \tag{9}$$

We consider now two interacting TLS labeled below by the subscripts 1 and 2. We take into account in (7) only the transverse interaction H_{xx}. By using the equations-of-motion method we easily obtain equations for the transverse Green's functions of this pair of TLS. We get two sets of four equations each. The first set relates the Green's functions $G_1=G_{-+11}$, $F_1=G_{++11}$ and the functions

$$L_1^{(\mp)}(\omega)=-i\int_0^\infty dt\,e^{i\omega t}\langle[\sigma_\mp^{(2)}(t)\sigma_z^{(1)}(t),\sigma_+^{(1)}(0)]\rangle. \tag{10}$$

It takes the form

$$\begin{aligned}&(-\omega+E_1)G_1-\lambda(L_1^{(-)}+L_1^{(+)})=\langle\sigma_z^{(1)}\rangle,\\&(-\omega-E_1)F_1+\lambda(L_1^{(-)}+L_1^{(+)})=0,\\&(-\omega+E_2)L_1^{(-)}-\lambda(G_1-F_1)=-K_{+-},\\&(-\omega-E_2)L_1^{(+)}-\lambda(G_1-F_1)=-Q_{++},\end{aligned} \tag{11}$$

where

$$\lambda=(\Delta_{01}\Delta_{02}/E_1E_2)J_{12},$$
$$K_{+-}=2\langle\sigma_+^{(1)}\sigma_-^{(2)}\rangle,\qquad Q_{++}=2\langle\sigma_+^{(1)}\sigma_+^{(2)}\rangle. \tag{12}$$

The second set of equations relates the Green's functions $G_{12}=G_{-+12}$, $F_{12}=G_{++12}$, and

$$L_{12}^{(\mp)}=-i\int_0^\infty dt\,e^{i\omega t}\langle[\sigma_\mp^{(2)}(t)\sigma_z^{(1)}(t),\sigma_+^{(2)}(0)]\rangle. \tag{13}$$

This set takes the form

$$\begin{aligned}&(-\omega+E_1)G_{12}-\lambda(L_{12}^{(-)}+L_{12}^{(+)})=0,\\&(-\omega-E_1)F_{12}+\lambda(L_{12}^{(-)}+L_{12}^{(+)})=0,\\&(-\omega+E_2)L_{12}^{(-)}-\lambda(G_{12}-F_{12})=Q_0,\\&(-\omega-E_2)L_{12}^{(+)}-\lambda(G_{12}-F_{12})=0,\end{aligned} \tag{14}$$

where $Q_0=\langle\sigma_z^{(1)}\sigma_z^{(2)}\rangle$.

These sets are easily solved, and the result is

$$G_1=-\{\langle\sigma_z{}^{(1)}\rangle[(\omega+E_1)(\omega^2-E_2{}^2)-2\lambda^2\omega]$$
$$+\lambda(\omega+E_1)[(\omega-E_2)Q_{++}+K_{+-}(\omega+E_2)]\}D^{-1}, \qquad (15a)$$

$$F_1=\{2\lambda^2\omega\langle\sigma_z{}^{(1)}\rangle+\lambda(\omega-E_1)[(\omega+E_2)K_{+-}-(\omega-E_2)Q_{++}]\}D^{-1}, \qquad (15b)$$

$$L_1{}^{(-)}=[2\lambda^2\omega(Q_{++}-K_{+-})+(\omega^2-E_1{}^2)(\omega+E_2)K_{+-}$$
$$+\lambda(\omega+E_1)(\omega+E_2)\langle\sigma_z{}^{(1)}\rangle]D^{-1}, \qquad (15c)$$
$$L_1{}^{(+)}=[-2\lambda^2\omega(Q_{++}-K_{+-})+(\omega^2-E_1{}^2)(\omega-E_2)Q_{\ +}$$
$$+\lambda(\omega+E_1)(\omega-E_2)\langle\sigma_z{}^{(1)}\rangle]D^{-1}, \qquad (15d)$$

$$G_{12}=Q_0\lambda(\omega+E_1)(\omega+E_2)D^{-1}, \qquad (15e)$$

$$F_{12}=-Q_0\lambda(\omega-E_1)(\omega+E_2)D^{-1}, \qquad (15f)$$

$$L_{12}^{(-}=Q_0[2\lambda^2\omega-(\omega^2-E_1{}^2)(\omega+E_2)]D^{-1}, \qquad (15g)$$

$$L_{12}^{(+)}=-2Q_0\omega\lambda^2D^{-1}. \qquad (15h)$$

The common determinant of systems (11) and (13) is here

$$D=(\omega^2-E_1{}^2)(\omega^2-E_2{}^2)-4\lambda^2\omega^2, \qquad (16)$$

and can be written in the form

$$D=(\omega^2-\omega_+{}^2)(\omega^2-\omega_-{}^2),$$

$$\omega_\pm{}^2=\{E_1{}^2+E_2{}^2+4\lambda^2\pm[(E_1{}^2+E_2{}^2+4\lambda^2)^2-4E_1{}^2E_2{}^2]^{1/2}\}/2. \qquad (17)$$

Obviously, the $\omega^2_\pm$ are always real. For a complete description of a TLS we need eight more functions $G_2=G_{-+21}$, $G_{21}=G_{-+21}$, etc. They are obtained from (15) by interchanging the subscripts 1 and 2, since K_{+-} is real.

Equation (15) contains four quantities $\langle\sigma_z^{(1)}\rangle$, K_{+-}, Q_{++} and Q_0. They are obtained by the following procedure. We introduce temperature Green's functions defined as T-products, taken with a minus sign, of imaginary-time operators.[8] Using the commutation rules for Pauli matrices, it is easy then to obtain the equations

$$\langle\sigma_z{}^{(1)}\rangle=-[2G_1(\tau=-\delta)+1],$$
$$K_{+-}=2L_1{}^{(-)}(\tau=-\delta),\quad Q_{++}=-2F_{12}(\tau=-\delta),$$
$$Q_0+\langle\sigma_z{}^{(1)}\rangle=-2L_{12}(\tau=-\delta). \qquad (18)$$

To determine the right-hand sides of these equations we must use the easily verified equality

$$R(\tau=-\delta)=\frac{1}{\pi}\int_{-\infty}^{\infty}d\omega\,N(\omega)\,\mathrm{Im}\,R(\omega), \qquad (19)$$

where $N(\omega)$ is the Planck function and $R(\omega)$ one of the retarded Green's functions defined by (15). The imaginary parts are calculated from the rule

$$\mathrm{Im}(\omega-\omega_1)^{-1}=-\pi\delta(\omega-\omega_1). \qquad (20)$$

Using (15)–(20) we get

$$Q_0=\mathrm{th}\frac{\omega_+}{2T}\,\mathrm{th}\frac{\omega_-}{2T},$$
$$\langle\sigma_z^{(1)}\rangle=\frac{E_2}{\omega_+{}^2-\omega_-{}^2}\left[\frac{E_1{}^2-\omega_+{}^2}{\omega_+}\mathrm{th}\frac{\omega_-}{2T}-\frac{E_1{}^2-\omega_-{}^2}{\omega_-}\mathrm{th}\frac{\omega_+}{2T}\right],$$
$$K_{+-}=\lambda\left(\mathrm{th}\frac{\omega_-}{2T}-\mathrm{th}\frac{\omega_+}{2T}\right)(\omega_+-\omega_-)^{-1},$$
$$Q_{++}=-\lambda\left(\mathrm{th}\frac{\omega_-}{2T}+\mathrm{th}\frac{\omega_+}{2T}\right)(\omega_++\omega_-)^{-1}, \qquad (21)$$

where $\omega_\pm$ are determined from (17). If $E_1=E_2$, Eqs. (15) and (21) coincide with the results of Ref. 9. The complete system of Green's functions includes also the functions G_{+-}, G_{--}, etc. It is easy to verify that they are obtained from (15) by simply replacing ω by $-\omega$. As a result we have

$$G_{xx11}=G_{xx1}=G_1(\omega)+G_1(-\omega)+F_1(\omega)+F_1(-\omega). \qquad (22)$$

At $T=0$, Eqs. (21) are greatly simplified and yeild

$$\langle\sigma_z{}^{(1)}\rangle=-(E_1+E_2)/(\omega_++\omega_-),$$
$$K_{+-}=0,\quad Q_{++}=-2\lambda/(\omega_++\omega_-). \qquad (23)$$

It follows from (23) that since

$$\omega_++\omega_-=[(E_1+E_2)^2+4\lambda^2]^{1/2},$$

we get $|\langle\sigma_z^{(1,2)}\rangle|<1$ even at $T=0$.

3. CONCENTRATION CORRECTION TO TRANSVERSE SUSCEPTIBILITY. AVERAGING OVER THE DISTANCES

We have thus determined the transverse Green's functions G_{xx1} and G_{xx12} of two TLS. The first of them describes the behavior of one TLS in the presence of the other, and the second the transfer of excitation from one TLS to the other. The linear response of TLS to an external interaction is described by the Green's function

$$G_{xx}(\mathbf{q},\omega)=N^{-1}\left[\sum_j G_{xxjj}(\omega)+\sum_{j\neq k}G_{xxjk}(\omega)\exp(i\mathbf{q}\mathbf{R}_{jk})\right], \qquad (24)$$

where N is the total number of TLS. In particular, it's just this Green's function which enters in the polarization operator Π of the phonon Green's function D:

$$D_{l,+}^{-1}(\mathbf{q},\omega)=\omega^2-\omega_{l,t}^2(\mathbf{q})-\Pi_{l,t}(\mathbf{q},\omega), \qquad (25)$$

$$\Pi_{l,t}(\mathbf{q},\omega)=P_0\gamma_{l,t}^2q^2\rho^{-1}\langle G(\mathbf{q},\omega)\rangle_n, \qquad (26)$$

where the angle brackets denote averaging over a TLS ensemble, and the subscript n indicates that the irreducible part of G must be used. This part is defined by the condition that it contain no contributions representing products of Green's functions from nonoverlapping aggregates of TLS connected by only one interaction line λ_{pq}. The irreducibility is analyzed in greater detail below, with two TLS as the example. In the present article we are interested mainly not in the actual forms of G_{xx} and Π, but in the determination of the conditions under which the transverse interaction of TLS becomes substantial. The point is that if these conditions are met it is necessary to sum the entire virial series in powers of P_0. This has not yet been accomplished. We confine ourselves therefore to the correction of first order in P_0 for the Green's function (24), and since the concentration P_0 is assumed small, we calculate the most singular terms whose divergence can compensate for the smallnes of P_0.

We begin with G_{xx1}. In the zeroth approximation we get from (6) and (22)

$$G_{xx1}^{(0)}=2E_1\,\mathrm{th}(E_1/2T)[(\omega+i\Gamma_{ph})^2-E_1{}^2]^{-1}. \qquad (27)$$

An exact expression for G_{xx1} is easily obtained from (15), (21), and (23). At $\lambda=0$, i.e., when the distance r_{12} is infinite, this exact equation must obviously coincide with $G^{(0)}_{xx1}$. It is clear therefore that to calculate the corrections to G_{xx1} it is necessary to consider the difference $\Delta G_{xx1} = G_{xx1} - G^{(0)}_{x_{x1}}$. It is convenient to express it in the form

$$\Delta G_{xx1}=\Delta G^{(1)}_{xx1}+\Delta G^{(2)}_{xx1}+\Delta G^{(3)}_{xx1}, \tag{28a}$$

$$\Delta G^{(1)}_{xx1}=-8E_1\omega^2\lambda^2\langle\sigma_z^{(1)}\rangle(\omega^2-E_1^2)^{-1}D^{-1}, \tag{28b}$$

$$\Delta G^{(2)}_{xx1}=-2E_1[\langle\sigma_z^{(1)}\rangle+\operatorname{th}(E_1/2T)](\omega^2-E_1^2)^{-1}, \tag{28c}$$

$$\Delta G^{(3)}_{xx1}=-4\lambda(\omega^2Q_{++}+K_{+-}E_1E_2)D^{-1}. \tag{28d}$$

We have neglected here the phonon damping. We shall explain below when this is permissible.

To calculate the concentration increment to G_{xx1} we must first average over all the distances between the two TLS. To this end we must multiply Eqs. (28) by P_0 and integrate over all the $\mathbf{r}_{1,2}$. As a result, taking into account (8), (12), and the fact that G_{xx1} depends only on λ^2, we get

$$(\Delta G_{xx1})_{\mathbf{r}}=\frac{1}{3}P_0\overline{|g_{12}|}\,\frac{\Delta_{01}\Delta_{02}}{E_1E_2}\int\frac{d\lambda}{\lambda^2}\Delta G_{xx1},$$

$$\overline{|g_{12}|}=\int d\Omega_{12}\langle|g_{12}(\mathbf{r}_{12}/r_{12})|\rangle, \tag{29}$$

where the angle brackets denote averaging over the components of the tensor $\gamma^{(2)}_{\mu\nu}$.

It is now easy to calculate the averages, over $\mathbf{r}_{12}$, of all three $\Delta G^{(a)}_{xx1}$ defined by Eqs. (28). We begin with $\Delta G^{(1)}_{xx1}$. We are interested in a region close to resonance, when $\omega^2\to E_1^2$. It is then obvious that the major role in the integrand is played by small $\lambda^2\sim\omega^2-E_1^2$ and therefore $\langle\sigma_z^{(1)}\rangle = -\tanh(E_1/2T)$. The integral with respect to λ is easily evaluated, but it must be remembered that ω always has a positive increment $i\delta$, so that at large $|\omega|$ the sign of the imaginary part of the denominator of the integrand coincides with the sign of ω. As a result we get

$$(\Delta G^{(1)}_{xx1})_r=-\frac{1}{3}P_0\overline{|g_{12}|}\,\frac{\Delta_{01}\Delta_{02}}{E_1E_2}\,\frac{2\pi i\omega E_1\operatorname{th}(E_1/2T)}{(\omega^2-E_1^2)^{3/2}(\omega^2-E_2^2)^{1/2}}. \tag{30}$$

The radicands are taken here to be positive if $\omega^2>E^2_{1,2}$. As $\omega^2\to E^2_{1,2}$ the calculated correction is more singular than the free function (27) is at $\Gamma_{ph}=0$.

It is very complicated to average $\Delta G^{(2)}_{xx1}$ over $\mathbf{r}_{12}$ for arbitrary temperatures. It will be seen, however, that it suffices to average at $T=0$. In this case, using (28c) and (23), we obtain

$$(\Delta G^{(2)}_{xx1})_r=-\frac{1}{3}P_0\overline{|g_{12}|}\,\frac{\Delta_{01}\Delta_{02}}{E_1E_2}\,\frac{4E_1}{(\omega^2-E_1^2)(E_1+E_2)}. \tag{31}$$

It is easy next to verify, by considering $T=0$, that the contribution from $\Delta G^{(3)}_{xx1}$ near resonance is proportional to

$$\omega(E_1+E_2)^{-1}[(\omega^2-E_1^2)(\omega^2-E_2^2)]^{-1/2},$$

i.e., it is small compared with (30) and can therefore be neglected.

We proceed now to an analysis of G_{xx12}. Using (15e), (15f), (21), and (22) we get

$$G_{xx12}=4\lambda E_1E_2D^{-1}\operatorname{th}\frac{\omega_+}{2T}\operatorname{th}\frac{\omega_-}{2T},$$

$$G^{(0)}_{xx12}=4\lambda E_1E_2[(\omega^2-E_1^2)(\omega^2-E_2)]^{-1}\operatorname{th}\frac{E_1}{2T}\operatorname{th}\frac{E_2}{2T}. \tag{32}$$

We have to integrate G_{xx12} with respect to $\mathbf{r}_{12}$. Since $\lambda\sim r_{12}^{-3}$, the corresponding integral must be additionally defined. We consider $G^{(0)}_{xx12}$ for this purpose. In the corresponding expression, λ stems from the phonon propagator at $\omega=0$. Assuming $\omega\neq 0$ for the phonon, $G^{(0)}_{xx12}$ describes the transition of a phonon from one resonance center to another. Just this quantity, averaged over the parameters of the centers, yields the square of the contribution, linear in P_0, to the phonon Green's function. This contribution is taken into account by introducing a polarization operator linear in P_0 and therefore needs not be considered. It is thus necessary to average the difference $\Delta G_{xx12}=G_{xx12}-G^{(0)}_{xx12}$. This is what was meant by separation of the irreducible part, referred to above. Using (8), (12), (15e), (15f), and (21) for the corresponding mean value, we have

$$(\Delta G_{xx1,2})_r=\frac{4}{3}P_0\frac{\Delta_{01}\Delta_{02}}{E_1E_2}E_1E_2\int d\Omega_{12}\,g(\mathbf{r}_{12}/r_{12})\int\frac{d\lambda}{\lambda}$$
$$\times\left[\frac{\operatorname{th}(\omega_+/2T)\operatorname{th}(\omega_-/2T)}{D(\omega)}-\frac{\operatorname{th}(E_1/2T)\operatorname{th}(E_2/2T)}{(\omega^2-E_1^2)(\omega^2-E_2^2)}\right]$$
$$\times\exp[i\mathbf{q}\mathbf{r}_{12}(\lambda)],$$

$$\mathbf{r}_{12}(\lambda)=(\mathbf{r}_{12}/r_{12})\left(\frac{\Delta_{01}\Delta_{02}}{E_1E_2\lambda}|g_{12}|\right)^{1/3}. \tag{33}$$

For small λ this expression converges well, but for

$$\lambda^2>\lambda_\omega^2=(\omega^2-E_1^2)(\omega^2-E_2^2)(4\omega^2)^{-1}$$

it has a logarithmic divergence cut off by the condition $\lambda<|g_{12}|r_{\min}^{-3}$, where $r_{\min}$ is the interatomic distance. As a result, expression (33) for $T=0$ can be written in the form

$$(\Delta G_{xx1,2})_r=\frac{4}{3}P_0\frac{E_1E_2}{(\omega^2-E_1^2)(\omega^2-E_2^2)}$$
$$\times\frac{\Delta_{01}\Delta_{02}}{E_1E_2}\int d\Omega_{12}\,g_{12}[\Phi(q\rho)-\ln|g_{12}|],$$

$$\rho=\left\{\frac{\Delta_{01}\Delta_{02}}{E_1E_2}|g_{12}|\left[\frac{4\omega^2}{(\omega^2-E_1^2)(\omega^2-E_2^2)}\right]^{1/2}\right\}^{1/3}, \tag{34}$$

where $\Phi(x)\sim x^2$ at $x\ll 1$ and decreases at large x; in addition, in view of condition (9) it is not important how $|g_{12}|$ in the second term is made nondimensional.

4. AVERAGING OVER E_1 AND E_2, AND THE DYNAMICS OF GLASSES

The results of the preceding section must be further analyzed differently for glasses and for systems containing TLS of equal energy. The present section is devoted to glasses, and it is therefore important for our purposes that $E_1\neq E_2$.

Near resonance, expression (30) is the principal correction to the free Green's function (27). Averaging (30) in accordance with (4) over the parameters of the second system as $\omega\to E_1$, we obtain

$$G_{xx1}^{(res)}=\frac{\operatorname{th}(E_1/2T)}{\omega-E_1}\left[1-\frac{\pi^2}{6}P_0\overline{|g_{12}|}\frac{\Delta_{01}}{E_1}\left[\frac{E_1}{2(\omega-E_1)}\right]^{1/2}\times\ln\left(\frac{2E_m}{\omega}+\frac{i\pi}{2}\right)\right]. \qquad (35)$$

A theoretical analyis in Ref. 10 yields for all glasses the estimate $E_m \sim 1$–10 K. Obviously, the second term in (35) must be taken into account if it is of the order of or larger than unity at $|\omega-E_1|\sim\Gamma_{ph}$, where Γ_{ph} is the phonon width (6c). It is convenient to rewrite this condition in this form

$$E_1[\operatorname{cth}(E_1/2T)]^{1/2}\leqslant P_0\overline{|g_{12}|}E_0\ln\frac{E_m}{E_1}. \qquad (36)$$

For thermal TLC with $E_1\sim T$ the condition that the phonon damping be small takes the form

$$T\ll T_0=P_0\overline{|g_{12}|}E_0\ln(E_m/E_0P_0\overline{|g_{12}|}). \qquad (37)$$

Inequalities (36) and (37) determine the region of TLS energies and temperatures in which the xx interaction is stronger than the interaction with phonons, and the phonon dynamics is replaced in the TLS by a regime which we call below intrinsic dynamics.

If condition (36) is met we get from (35) the following estimate for the TLS line width due to its interaction with other TLS:

$$\Gamma_{xx}\sim\left(P_0\overline{|g_{12}|}\frac{\Delta_{01}}{E_1}\right)^2E_1\ln^2\frac{E_m}{E_1}=\left(\frac{\Delta_{01}}{E_1}\right)^2\tau_{xx}{}^{-1}. \qquad (38)$$

In addition, it follows from (35) that the line should be very asymmetric and certainly not Lorentzian. We note also that for $P_0|\overline{g_{12}}|\sim P_0\gamma^2/\rho s^2\sim 1$ the width Γ_{xx} becomes $\sim E_1$ and the TLS lose all their individuality.

We discuss now the physical reason for the onset of the dampling Γ_{xx}. It follows from (16) and (17) that at a fixed energy E_1 any frequency ω close to E_1 is at resonance with an entire ensemble of remote TLS whose parameters E_2 and λ are such that ω_- or ω_+ is equal to ω. In other words, the external action on a selected TLS causes, albeit with low probability, transitions in the entire ensemble of the TLS. Transitions in these TLS are accompanied in turn by transitions in other TLS. ensembles. As a result, the energy of the selected TLS is redistributed in small batches among all the TLS and the quantity τ_{xx} defines the characteristic time of this process. It must be emphasized here once more that the xx interaction does not conserve the z projections of the individual TLS and of their ensemble as a whole, and leads therefore to a real mixing of the states in the system. At the same time, the zz interaction considered so far causes only inhomogeneous broadening of the spectra of individual TLS and relaxation of their phase.[4,5,11] It cannot lead to a mixing of their levels. Therefore, particularly in discussions of the echo in a set of TLS in glasses (see Refs. 4 and 5), the time that sets the upper bound of the echo signal was taken to be τ_{ph} [see (6c)]. If, however, the conditions (36) and (37) are met, this time should be τ_{xx} defined by Eq. (38).

The values of $P_0|\overline{g_{12}}|$ and T_0 for fused quartz can be determined by using the data of Refs. 4 and 12. Thus, according to Ref. 4, $\pi^{-1}|\overline{g_{12}}|\leqslant 1.6\cdot 10^{-35}$ erg·cm³ and[2)] $P_0=2.2\cdot 10^{21}$ (erg·cm³)$^{-1}$. Furthermore, according to Ref. 12, $\rho=2.2$ g/cm³, $s_l=3.8\cdot 10^5$ cm/s, $s_t=5.1\cdot 10^5$ cm/s, $\gamma_l=1.5$ eV and $\gamma_t^2=\gamma_l^2/2$. As a result, taking (6c) into account, we obtain $E_0=40$ K (Ref. 7). Less certain is the value of E_m in the logarithm. We assume in accordance with Ref. 10 that $E_m\approx 5$ K. We obtain then

$$P_0\overline{|g_{12}|}\leqslant 1.1\cdot 10^{-3},\quad T_0\leqslant 0.22\text{ K}. \qquad (39)$$

We see thus that the temperature T_0 of the transition to the intrinsic dynamics of a TLS in fused quartz lies in the region in which experiments are being performed at present. This must be taken into account without fail in the analysis of those experiments in which an important role is played by the time to establish equilibrium, such as in echo experiments. It should be noted that Γ_{xx}, just as Γ_{ph}, is proportional to $(\Delta_0/E)^2$. It is just this factor which leads to the existence of a broad spectrum of relaxation times for TLS in glasses.[1,2,4] The xx interaction introduced with the aid of (7) has therefore no influence on the existence of such a spectrum. The response of a set of TLS to an external action is obtained after averaging g_{xx1} also over the parameters of the first system. We obtain then from (27) for a free TLS

$$\langle G_{xx1}^{(0)}\rangle=-2\{\ln[E_m/(\max|\omega|,T)]+(i\pi/2)\operatorname{th}(\omega/2T)\}. \qquad (40)$$

Substitution of this expression in (25) yields the well known results for damping of sound and for renormalization of its velocity[1–4]:

$$\Delta\omega_\lambda/(s_\lambda q)=(\omega_\lambda-s_\lambda q)/s_\lambda q=-\alpha_\lambda\{\ln[E_m/(\max|\omega|,T)]+(i\pi/2)\operatorname{th}(\omega/2T)\}\approx-\alpha_\lambda[\ln(E_m/\omega)+i\pi/2], \qquad (41)$$

where $\lambda=l, t$, $\alpha_\lambda=P_0\gamma_\lambda^2/(\rho s_\lambda^2)$; the approximate equality on the right side takes place at $\omega\gg T$, and the logarithm is defined to be real at $\omega>0$. We find now the corrections of order P_0^2 to this expression. The main contribution comes from averaging Eq. (28c) for $\Delta G_{xx1}^{(2)}$. Using (31) and (4) with $T=0$, we get

$$\Delta\omega_\lambda/(s_\lambda q)=-\alpha_\lambda[\ln(E_m/\omega)+i\pi/2]\times\{1-(\pi^3/24)P_0\overline{|g_{12}|}[\ln(E_m/\omega)+i\pi/2]\}. \qquad (42)$$

In the derivation of this expression we took it into account that the xx interaction of a phonon with a TLS is proportional, in accord with (3c), to Δ_0/E, so that the considered correction for $\Pi_\lambda(\mathbf{q})$ contains $(\Delta_{01}/E_1)^3$. In addition, we have assumed that $|\overline{g_{12}}|$ does not depend on the components of the tensor $\gamma_{\mu\nu}^{(1)}$. Equation (42) is valid with logarithmic accuracy. A correction proportional to $P_0^2\ln(E_m/\omega)$ is due to the averaging of Eq. (28b) for $\Delta G_{xx1}^{(1)}$. We shall not need it hereafter. If $\omega\ll T$, it is necessary to replace, in the logarithm of (42), ω by T and $i\pi/2$ by a quantity porportional to $i\omega/T$.

The coefficients of the first and second powers of the logarithm in (42) are of opposite sign. This indicates apparently that in the limit as $T\to 0$ no phase transition takes place in the TLS, and the speed of sound changes by a finite amount. For ordinary glasses, in view of the smallness of $P_0|\overline{g_{12}}|$, this question is only academic. In structured glasses where the TLS is due to the presence of different equilibrium positions of individual molecular complexes, $P_0|\overline{g_{12}}|$ can be arbitrary and the xx interaction influences the speed of sound in a wide temperature range (see, e.g., Ref. 13).

We consider now the q-dependent transport part of $\Delta G_{xx\,12}$. We confine ourselves to the case $T=0$. It follows from (34) that the corresponding correction is small near resonance compared with the $\Delta G_{xx_1}^{(1)}$ contribution taken into account in (35). This is natural, for at $E_1\neq E_2$ the transfer of the "spin" deviation from one TLS to another is difficult. Using (33) and (34) for the corresponding contribution to the phonon energy, averaged over the parameters of both TLS, we get

$$(\Delta\dot{\omega}_\lambda/s_\lambda q)_t \sim (P_0\gamma^2/\rho s^2)^2 \varphi[q(i|g_{12}|/\omega_\lambda)^{1/3}], \tag{43}$$

where the dimensionless function $\varphi(x) \sim x^2$ for $x \ll 1$ and $\varphi^{(\infty)} = 0$. The imaginary unity in (43) ensures that the phonon propagator is real on the positive imaginary axis in the ω plane.[14] Comparing (43) with (42), we see that the q-dependent "transport" part makes a small contribution to the phonon energy. In addition, the second term of (34) leads to a change of the coefficient in the term quadratic in P_0 in (42). The integral $\int d\Omega_{12} g_{12} \ln |g_{12}|$ cannot be calculated, however, without specifying the angular dependence of the interaction. Since the integrand is of alternating sign, the integral is apparently numerically small compared with $\overline{|g_{12}|}$.

5. RESONANT TLS

By resonant TLS we mean here TLS having equal energies, $E_1 = E_2$. This problem arises if the substance contains a small concentration of identical TLS of one type or another, such as Jahn-Teller centers or molecular complexes with internal degree of freedom. In this case, obviously, the coefficients $\Delta_{01,2}/E_{1,2}$ in all the equations must be regarded as constants and included in g_{12}, while the averaging should be only over the configurations, i.e., over the relative placement of the TLS. In this case we get near resonance, using (27) and (30),

$$G_{xx1} = \frac{\mathrm{th}(E/2T)}{\omega - E}\left[1 - \frac{i\pi\overline{|g_{12}|}n_0}{6(\omega - E)}\right], \tag{44}$$

where n_0 is the concentration of the resonant TLS. It follows from this equation that the characteristic width of the resonant TLS, due to their interaction, is of the form

$$\Gamma_r \sim \pi n_0 \overline{|g_{12}|}/6. \tag{45}$$

Unlike Eq. (38) for Γ_x, this width does not depend on the TLS energy. This means that if $E < \Gamma_r$ the individuality of the TLS is lost, and this occurs at any concentration n_0.

We proceed now to the q-dependent "transport" part of G_{xx}, described by Eqs. (33) and (34). In this expression we have now $\omega_\pm = (E^2 + \lambda^2)^{1/2} \pm \lambda$. We consider the resonance region, as before. Recognizing that the small λ in the integrand are significant, we obtain from (33) the q-dependent correction to G_{xx} in the form

$$\Delta G_{xx}(q) = \frac{n_0\,\mathrm{th}^2(E/2T)}{3(\omega - E)}\overline{|g_{12}|}\psi\left(\frac{q}{q_r}\right),$$

$$q_r = [\overline{|g_{12}|}(\omega - E)^{-1}]^{1/3}, \tag{46}$$

where the dimensionless function $\psi(x) \sim x^2$ if $x \ll 1$, is of the order of unity at $x = 1$, and decreases at large x. If $\omega - E \lesssim \Gamma_r$, where Γ_r is defined by Eq. (45), $\omega - E$ in (46) must be replaced by Γ_r. It turns out as a result that $q_r \sim n_0^{1/3}$, and that at $q \sim q_r$ expression (46) becomes of the order of the second term in (44). Thus, in contrast to random TLS, the transfer of excitation from one resonant TLS to another is no less important than the redistribution of the energy among the different TLS. We note also that the part of (33) which diverges at large λ alters the coefficient in the second term of (44). With regard to this change, the same statements can be made as at the end of the preceding section.

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[1] It is shown in Sec. 4 that the times τ_{xx} and the phonon relaxation times obey the same distribution law.

[2] Different values of P_0 were obtained in different experiments.[12] The value of P_0 in Ref. 4 is the smallest and apparently the most reliable.

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