Low-energy collective excitations in glasses. New relaxation mechanism for ultralow temperatures

A. L. Burin and Yu. Kagan

Russian Scientific Center "Kurchatov Institute," 123182 Moscow, Russia

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The interaction of two-level systems in dielectric glasses leads to a subsystem of spatially delocalized, collective low-energy excitations. At ultralow temperatures, when phonons are to a large extent frozen out, relaxation processes will be determined by the excitations of this subsystem. The new relaxation mechanism leads to a different picture of low-frequency sound absorption in glasses. The internal friction coefficient acquires under these conditions a linear temperature dependence instead of the classical $Q^{-1}\sim T^3$. Experiments demonstrating the change in the relaxation regime at ultralow temperatures are discussed.

1. INTRODUCTION

The tunneling model introduced in the now classical work of Anderson *et al.*[1] and Phillips[2] for a description of dielectric glasses explains, to a large extent, the universal character of the anomalous low-temperature properties of amorphous systems widely different in their nature (see, e.g., Ref. 3). Within this model, the low-temperature properties have been linked to the special properties of an ensemble of randomly located two- level (two-well) systems (TLS) with a specified parameter distribution. If the description of a TLS employed the standard pseudospin representation

$$H_0=\sum_n H_{0n};\quad H_{0n}=-\Delta_n S_n^z-\Delta_{0n}S_n^x \tag{1.1}$$

then for the distribution of the relative level shift in the wells, Δ_n, and of the tunneling amplitude, Δ_{0n}, one assumed, on a limited energy scale $\lesssim 1\,\mathrm{K}$, the representation

$$P(\Delta,\Delta_0)d\Delta\, d\Delta_0;\quad P(\Delta,\Delta_0)=P_0/\Delta_0. \tag{1.2}$$

The uniform distribution in Δ appears natural, while the uniform distribution in $\ln\Delta_0$ or in the tunneling action scale is less obvious. In this paper we take the distribution (1.2) as an experimental fact.

The total Hamiltonian contains apart from (1.1), a term describing the interaction of TLSs with the excitations of the system,

$$H'=-\sum_n \hat{V}_n S_n^z, \tag{1.3}$$

where $\hat{V}_n$ is the operator in the space of system excitation variables.

The deformation of the elementary cell due to the interaction with the excitations shifts Δ_n substantially while affecting little the value of Δ_{0n}. This familiar result (see, e.g., Ref. 3) allows retaining only terms proportional to S_n^z in (1.3).

The system (1.1) of independent TLSs having the parameter distribution (1.2) and interacting with the medium according to (1.3) explains reasonably well the anomalous temperature dependences of the thermodynamical and kinetic properties of glasses at temperatures below 1 K.

The interaction (1.3) leads, in principle, to an indirect interaction between the TLSs. In a dielectric medium, due to the virtual acoustic-phonon exchange, this indirect interaction V_{12} falls off at large distances as $1/R_{12}^3$ (in a metal there is an added indirect interaction via conduction electrons, which falls off in the same way but may be much stronger). In dielectric glasses, the inclusion of this interaction is found to be essential in the nonlinear analysis of sound or electromagnetic radiation absorption (see, e.g., Refs. 4 and 5) and in related problems.

As will be shown in the present paper, at sufficiently low temperatures the indirect interaction between TLSs becomes quite fundamental in character. For any pair of defect centers with an internal degree of freedom, this interaction leads to a coherent dynamic coupling. As a result, low-energy non-single-particle excitations appear which involve two or more TLSs in an individual transition. It turns out that the density of these excitations differs radically from that given by the distribution (1.2).

If the dissipative interaction with medium excitations does not destroy the coherent couplings, then the dynamic interaction between coherently coupled TLS pairs - taking into account the resulting parameter distribution - will inevitably lead to delocalized collective excitations at sufficiently low energy (in an interacting system of the single-particle excitations of individual TLSs, delocalized collective excitations do not appear, see below).

This strip of phonon-unrelated low-energy excitations leads to a fundamentally different relaxation mechanism at ultralow temperatures. In the case of dielectric glasses, this picture should always realize at sufficiently low temperatures, since the breakdown of coherent couplings due to the irreversible interaction with the phonons weakens as T^3 with decreasing temperature. In the case of metallic glasses, the conservation of coherent couplings occurs only in a transition to the superconducting state, when the dissipative interaction with the electrons is being suppressed exponentially as temperature is lowered.

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The change of the relaxation mechanism should be clearly manifest in the behavior of the internal friction. Instead of the standard $Q_{\text{ph}}^{-1} \sim T^3$ dependence of the phonon absorption coefficient, at ultralow temperatures the $Q^{-1} \sim T$ dependence should be observed. Such a crossover appears to have first been observed in Ref. 6 which reports low-frequency internal friction measurements ($\nu \approx 400$ Hz) on classical amorphous SiO_2 over a fairly wide low-temperature range down to $T \lesssim 1$ mK. Below, a separate analysis of these experimental results will be given.

The appearance of collective delocalized excitations is to a large extent related to the $1/R^3$ interaction between TLSs. This also occurs in lower dimensional systems, as a reflection of the fact that the indirect interaction behaves like $1/R^d$.

It should be noted that Yu and Leggett[7] introduced the hypothesis that the law $1/R^3$ for the defect-defect interaction in a medium may be responsible for *all* the spectral characteristics, as well as for the universality, of the anomalous properties of amorphous systems. This hypothesis is not yet anywhere near its full development. In the present work it is assumed that the initial spectral structure is formed by a system of independent defect centers described by Eqs. (1.1) and (1.2), and that the relatively weak interaction between the centers is responsible for the secondary spectral structure with collective delocalized excitations.

2. LOW ENERGY COLLECTIVE EXCITATIONS

In dielectric glasses at $T \ll \theta_D$, the interaction $\hat{V}$ in Eq. (1.3), as well as the inter-TLS interaction at large distances $R \gg a$ (a being the interatomic distance scale), is determined by acoustic phonons. In the general case

$$\hat{V} = \gamma_n^{ij} u_{ij}^n, \tag{2.1}$$

where $u_{ij}^n = (\partial u_i / \partial x_j)_n$ is the strain tensor and γ_n^{ij} is the deformation potential tensor. The interaction between the TLSs may be found directly in second-order perturbation theory in H', Eq. (1.3), or by canonical transformations to new equilibrium positions of the normal oscillator coordinates:

$$H_{\text{int}} = \sum_{nm} U_{nm} S_n^z S_m^z, \tag{2.2}$$

$$U_{nm} = u_{nm} / R_{nm}^3 . \tag{2.3}$$

Usually the average over angles is zero,

$$\langle u_{nm} \rangle = 0 . \tag{2.4}$$

The charge scale in the interaction (2.3) can be determined from the relation

$$\langle u_{mn}^2 \rangle = U_0^2 . \tag{2.5}$$

It is easily found that, within a constant factor,

$$U_0 \approx \gamma_0^2 / \rho c^2, \tag{2.6}$$

where γ_0^2 is a certain average value of the square of the tensor elements γ^{ij}, ρ is the mass density, and c a certain average sound speed.

The Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \tag{2.7}$$

determines the multi-particle character of excitations in an amorphous system. Consider an isolated pair of TLSs. The Hamiltonian of such a pair has the form

$$H_{12} = H_{01} + H_{02} + U_{12} S_1^z S_2^z . \tag{2.8}$$

Let us change to the representation in which the Hamiltonians H_{01} and H_{02} are diagonal. In spin space, to such a change there corresponds a rotation of quantization axes. The relation between the spin components in the old and new ($\tilde{S}^i$) coordinate system is given by

$$\begin{aligned} S_n^z &= (\Delta_n / \varepsilon_n) \tilde{S}_n^z - (\Delta_{0n} / \varepsilon_n) \tilde{S}_n^x , \\ S_n^x &= (\Delta_{0n} \varepsilon_n) \tilde{S}_n^z + (\Delta_n / \varepsilon_n) \tilde{S}_n^x . \end{aligned} \tag{2.9}$$

Then

$$\begin{aligned} H_{12} = {} & -\varepsilon_1 \tilde{S}_1^z - \varepsilon_2 \tilde{S}_2^z - U_{12} [\Delta_1 \delta_2 / (\varepsilon_1 \varepsilon_2) \tilde{S}_1^z \tilde{S}_2^z \\ & + \Delta_{01} \Delta_{02} / (\varepsilon_1 \varepsilon_2) \tilde{S}_1^x \tilde{S}_2^x - \Delta_{01} \Delta_{02} / (\varepsilon_1 \varepsilon_2) \tilde{S}_1^x \tilde{S}_2^z \\ & + \Delta_1 \Delta_{02} / (\varepsilon_1 \varepsilon_2) \tilde{S}_1^z \tilde{S}_2^x] . \end{aligned} \tag{2.10}$$

In these expressions

$$\varepsilon_n = (\Sigma_n^2 + \Delta_{0n}^2)^{1/2} \tag{2.11}$$

is the excitation energy of an isolated TLS.

In the following we will be interested in up-down transitions, when one member of a TLS pair transfers from the lower to the upper state, while the second the other way around. We shall later show that in the problem we are considering the dominant role is played by TLSs pairs with $\varepsilon_{1,2} \lesssim T$ and a distance $R_{1,2}$ such for which, *a priori*, $|U_{1,2}| \ll T$. In this case the third and second terms in brackets in (2.10) lead to a negligibly small renormalization in ε_1 and ε_2 respectively, and so may be dropped. The first term in the brackets does not change at all in the up-down transition. As a result, such transitions can be effectively described by the Hamiltonian of a two-level system (1.1) with an asymmetry energy Δ_{p} and transition amplitude Δ_{op}:

$$\Delta_p = \varepsilon_1 - \varepsilon_2 ; \quad \Delta_{0p} = U_{12} \Delta_{01} \Delta_{02} / (2 \varepsilon_1 \varepsilon_2) . \tag{2.12}$$

The coherent coupling within the pair, which determines Δ_{op}, falls of as $1/R_{12}^3$ with distance. If the magnitude of this amplitude becomes less than $\hbar / \tau_{\text{ph}}$ [where τ_{ph} is the transition time in an individual TLS due to the interaction with phonons, Eqs. (1.3) and (2.1)], then the coherent coupling in the pair is destroyed. This suggests a cutoff radius R_c for the formation region of coherently coupled pairs. A direct estimate of $\tau_{\text{ph}}(\varepsilon)$ for $\varepsilon \lesssim T$, using (1.3) and the relation between S^z and $\tilde{S}^x$, Eq. (2.9), gives

$$1/\tau_{\text{ph}} = (\Delta_0 / \varepsilon)^2 1/\tau_{\text{ph}}^{(0)} ; \quad 1/\tau_{\text{ph}}^{(0)} = (\gamma_0^2 / \rho c^5 \hbar^4) T^3 . \tag{2.13}$$

Considering thermal TLSs with $\Delta_0, \Delta \simeq T$, taking $1/\tau_{\text{ph}}^{(0)}$ to be equal to $(1/\hbar) U_0 / R_c^3$, we find, using (2.6), that

$$R_c \approx \hbar c / T \simeq \theta_D / T . \tag{2.14}$$

That we have obtained a *finite* radius for the coherent interaction region of is extremely significant.

Let us evaluate the probability w for an individual TLS to find a resonance up-down partner in the coherent region, or to form a resonance pair with $\Delta_p < \Delta_{0p}$. Let a given TLS has parameters $\Delta_1 < \Delta_{01}$ and thus $\Delta_{01}/\varepsilon \sim 1$, which is essential for the scale of coupling between the TLSs [see Eq. (2.12)]. If the second TLS is at a distance R, then the energy difference $\varepsilon_2 - \varepsilon_1$ should not exceed the value

$$\varepsilon_2 - \varepsilon_1 \approx (U_0/2R^3)(\Delta_{02}/\varepsilon_2) = \Delta_{0p}. \tag{2.14'}$$

The up-down transitions involve TLSs with energy $\varepsilon_i \sim T$. The probability w, as we will see, is accumulated over large distances, where the fractional value of the energy difference is small. Therefore, on the right-hand side of this relation ε_2 can be replaced by ε_1. Then

$$w = (1/2) \int d^3\mathbf{R} \int_0^{\varepsilon_1} (d\Delta_{02}/\Delta_{02})$$

$$\int_{\varepsilon_1}^{\varepsilon_1^+} d\varepsilon_2 \varepsilon_2 (\varepsilon_2^2 - \Delta_{02}^2)^{-1/2} P_0 \Xi (R_c - R). \tag{2.15}$$

In this expression we have changed from the variables Δ_0, Δ in the distribution (1.2) to the variables Δ_0, ε; $\Theta(x)$ is the standard, unit switching-on function.

Taking advantage of the small value of Δ_{0p}, it is directly found that

$$w \approx (\pi^2/2)(P_0 U_0) \ln(\theta_D/T). \tag{2.16}$$

For all known glasses

$$P_0 U_0 \ll 1 \tag{2.17}$$

(for example, for SiO_2 we have $P_0 U_o \approx 10^{-3}$). Generally, $w \ll 1$ even for very low temperatures. This means that at distances $R \gg R_c$ the coherent oscillations die away exponentially. Therefore under these conditions no spin-wave type delocalized states occur, in contrast to what a number of studies[8-10] have suggested. For the same reason there is no anomalous diffusion of the type described by Levitov[11] for a chaotic system of particles with $1/R^3$ interaction. Delocalization can only result from the interaction of complex excitations involving two or more TLSs simultaneously (cf. Ref. 12).

The result (2.16), (2.17) is essential for the discussion of the more complex picture of many-particle excitations since it suggests that, for $w \ll 1$, an individual TLS surely does not have two or more resonance partners.

Let us calculate the distribution in Δ_p, Δ_{0p}, Eq. (2.12), for effective two-level systems formed by TLS pairs in the coherent region. Since we are considering excitations related to up-down transitions, we must introduce statistical factors accounting for the fact that one TLSs is in the lower, and the other in the upper state. Again changing in Eq. (1.2) to the variables Δ_0 and ε, Eq. (2.11), we find

$$P^{(2)}(\Delta_p, \Delta_{0p}) = P_0^2 \int (d\Delta_{01}/\Delta_{01}) \int d\varepsilon_1 \varepsilon_1 (\varepsilon_1^2 - \Delta_{01}^2)^{-1/2} (1 + e^{-\varepsilon_1/T})^{-1}$$

$$\times \int_0^{\varepsilon_1} (d\Delta_{02}/\Delta_{02}) \int_{\varepsilon_1}^{\varepsilon_1 + \Delta_{0p}} d\varepsilon_2 \varepsilon_2 (\varepsilon_2^2 - \delta_{02}^2)^{-1/2} (1 + e^{\varepsilon_2/T})^{-1} \int d^3\mathbf{R} \Theta(R_c - R) \delta[\Delta_{0p} - (|u_{12}|/R^3) \Delta_{01} \Delta_{02}/(2\varepsilon_1 \varepsilon_2)] \delta(\Delta_p - \varepsilon_1 + \varepsilon_2). \tag{2.18}$$

For the present problem, the sign of the transition amplitude in the pair does not play a role. In fact, as in Eq. (1.2), we are interested in the distribution in the modulus of Δ_{0p}. Therefore the argument of the δ function in (2.18) involves $|u_{12}|$. Integration over $d^3\mathbf{R}$ leads to the result:

$$\int d^3\mathbf{R} \Theta(R_c - R) \delta[\Delta_{0p} - (|u_{12}|/R^3) \Delta_{01} \Delta_{02}/(2\varepsilon_1 \varepsilon_2)]$$

$$\approx (2\pi/3)(\Delta_{01}\Delta_{02}/\varepsilon_1\varepsilon_2)(U_0/\Delta_{0p}^2) \Theta(\Delta_{0p} - U_0 (T/\hbar c)^3). \tag{2.19}$$

After substituting (2.19) into (2.18), the integration over $d\Delta_{0i}$ is trivial. In what follows we will be interested in the low-energy spectral properties for $\Delta_p, \Delta_{0p} \ll T$. From Eq. (2.18) it then follows that the corresponding pairs are formed from TLSs with $\varepsilon_i \approx \Delta_{0p} \sim T$. Also, it is seen that to the effective pair size there corresponds the inequality $U_0/R^3 \ll T$. Thus we find satisfied all the conditions that led to the simplification of the Hamiltonian (2.10) and thereby to the result (2.12).

Finally, for the distribution (2.18) we obtain

$$P^{(2)}(\Delta_p, \Delta_{0p}) = (\pi^3/12)(P_0 T)(P_0 U_0)(1/\delta_{0p}^2) \Theta(\Delta_{0p} - U_0 (T/\hbar c)^3). \tag{2.20}$$

The most essential point about this distribution is the sharp increase in the number of pairs with a small transition amplitude

$$P \sim 1/\Delta_{0p}^2.$$

This result is universal in character. It retains it truth in systems of arbitrary dimensionality and does not depend on the form of the parameter distribution of the initial system of TLSs.

Comparing (2.20) and (1.2), we see that for $\Delta_{0p} < T(P_0 U_0)$ the density of pair excitations becomes larger than the density of the excitations of isolated TLSs. Since the correlation volume increases with decreasing temperature as $1/T^3$, this result, as well as the sharp increase in the density of pair excitations with decreasing Δ_{0p}, will be determined, for low enough T, by pairs with a stable coherent coupling.

3. DELOCALIZATION OF COLLECTIVE EXCITATIONS

The next step is to consider the resonance interaction between the pairs. The distribution (2.20) has a remarkable feature. For any value of Δ_{0p}, in the interval

$(\Delta_{0p}-\Delta_{0p}/2,\Delta_{0p}+\Delta_{0p}/2)$ there will be one and the same number of pairs with resonance parameters $\Delta_p<\Delta)_{0p}$,

$$N_r\approx(\pi^3/12)(P_0T(P_0U_0) \tag{3.1}$$

(per unit volume). The distance between such pairs is determined by the relation

$$1/R_*^3\approx(P_0T)(P_0U_0). \tag{3.2}$$

The interaction between resonance pairs in each energy interval is characterized by the quantity

$$\Delta_*U_0/R_*^3\approx T(P_0U_0)^2. \tag{3.3}$$

For resonance pairs with $\Delta_{0p}\approx\Delta_*$, in the transition amplitude expression (2.12) the factors $\Delta_{0p}^{(2)}/\varepsilon_{0p}^{(2)}\sim 1$. Because of this, Δ_* determines the scale of the coherent coupling between such pairs for $R_c>R_*$. At the same time, this latter inequality guarantees the stability of the coherent coupling in the pairs themselves. Under these conditions, the resonance pairs with $\Delta_{0p}\approx\Delta_*$ form an infinite coherent-coupling cluster, which enables delocalized excitations to appear. For $\Delta_{0p}\gg\Delta_*$, since the interaction between the pairs remains as in Eq. (3.3), an infinite cluster does not form and the excitations turn out to be localized. For $\Delta_{0p}\ll\Delta_*$, the interaction between resonance pairs at an average distance R_* is large compared to Δ_{0p}. In this case the up-down transition amplitude is no longer given by Eq. (2.12) but has a much smaller value of

$$\Delta_{0p}^{(1)}\Delta_{0p}^{(2)}/U_{12}. \tag{3.4}$$

Thus, the resonance coupling does not form at the *average* distance. It forms between pairs at the distance $R(\Delta_{0p})$ determined by the relation

$$U_0/R^3\approx\Delta_{0p}. \tag{3.4$'$}$$

As a result, in each energy strip $(\Delta_{0p}-\Delta_{0p}/2,\Delta_{0p}+\Delta_{0p}/2)$ an infinite cluster of resonantly coupled pairs forms and in this cluster, consequently, delocalized collective excitations propagate.

Now since $R(\Delta_{0p})>R_*$, the number of pairs with a resonance coupling in such a cluster is less than N_r, Eq. (3.1), in proportion to Δ_{0p}/Δ_*. At the same time, for an arbitrary pair, the probability of finding a resonant pair in the coherent region can be calculated; this is analogous to Eq. (2.15), but with P replaced by $P^{(2)}$. It is easily shown that for $\Delta_{0p}\ll\Delta_*$ any pair belongs to an infinite resonance cluster. This means that in each energy strip actually all the N_r pairs participate in the formation of delocalized excitations.

The requirement $R_*<R_c$, from Eqs. (2.14) and (3.2), places a restriction on the temperature,

$$T<T_0,\quad T_0\approx(P_0U_0)(c^3\hbar^3/U_0)^{1/2}. \tag{3.5}$$

Physically, the temperature restriction relates to the requirement that the coherent region be sufficiently large for delocalized excitations to form. In this connection, the following remark should be made. As should be clear from the discussion above, the phenomenon of delocalization has been analyzed in terms of concepts inherent in the theory of the Anderson localization.[13] For $T\neq 0$, the interaction with phonons is conserved on a limited scale. Delocalization then implies the possibility for a local perturbation to go away to infinity in a diffusive way.

To the scale Δ_* there corresponds the characteristic time

$$\tau_*\simeq\hbar/\Delta_*. \tag{3.6}$$

In this time, for $\Delta_*\ll T\ll T_0$ practically all the pairs in the energy strip with $\Delta_{0p}\approx\Delta_*$ will change their state. The number of such pairs is N_r, Eq. (3.1). As will be seen, a change in the state of about N_r TLSs leads, statistically, to a shift in the level splitting Δ_i on the scale $\sim\Delta_*$ over the entire ensemble of the initial TLSs. This means that in a time of order τ^*, the resonance pairs with $\Delta_p\lesssim\Delta_{0p}\approx\Delta_*$ will have their resonant couplings broken and become unable to form an infinite resonance cluster. The same will occur to the clusters formed by the pairs with $\Delta_{0p}<\Delta_*$. But in these latter, the characteristic time of change of state is determined by the quantity $\hbar/\Delta_{0p}$. Therefore, in the time τ_* the probability of a real transition is $(\Delta_{0p}\tau_*)^2<1$. This allows an estimate that the total number of resonance pairs undergoing transitions in the entire interval $\Delta_{\rm op}<\Delta_*$ does not exceed N_r.

4. RELAXATION TIMES

In transitions involving a finite number of two-level systems distributed uniformly in space, the change in the strain field that results causes a change in the level shift Δ_i of individual initial TLSs. Since the parameters Δ_{0i} remain practically unchanged, it follows that, in principle, intra- and interpair resonant couplings should break down while at the same time new resonant couplings will form. Relaxation processes in the system are to a considerable extent dependent of the nature and scale of such rearrangement.

Let the density of TLSs that have undergone the transition, N, be small compared to the total density of TLSs, N_0. The change in the parameter Δ_i of an individual TLS will be written as

$$\begin{aligned} Q(\Delta'-\Delta)&=\biggl\langle\delta\biggl(\Delta'-\Delta-\sum_j{}'u_{0j}/R_{0j}^3\biggr\rangle\\ &=(1/2\pi)\int_{-\infty}^{+\infty}dte^{i(\Delta-\Delta')t}\biggl\langle\exp\\ &\quad\times\biggl\{-it\sum_j{}'u_{0j}/R_{0j}^3\biggr)\biggr\}\biggr\rangle. \end{aligned} \tag{4.1}$$

Here the prime indicates than the summation is only over the positions of those TLSs having undergone the transition. For the probability density for the change in the level shift we then have

$$J=\left\langle \exp\left\{-it{\sum_j}' u_{0j}/R_{0j}^3\right\}\right\rangle={\prod_j}'\langle\exp$$

$$\times(-itu_{0j}/R_{0j}^3)\rangle={\prod_j}'[1-x+x\langle\exp(-itu/R_{0j}^3)\rangle_u]$$

$$=\exp\left\{-x\sum_j(1-\langle\exp(-itu/R_{0j}^3)\rangle_u)\right\}. \qquad (4.1')$$

Here $\langle .... \rangle$ denotes an average over the positions of those TLSs having changed their state and, independently, over the distribution in u_{0j} (recall that $\langle u_{0j}\rangle=0$). The first distribution, assuming a statistical independence of individual TLSs, is carried out directly giving

$$J=\exp(-(2\pi^2/3)N|\bar{u}||t|). \qquad (4.2)$$

Here $x=N/N_0$ and we have used the fact that $x\ll 1$. Changing from the summation on j to the integration $\int d^3\mathbf{r}N_0$ and assuming that the distribution in u is an even function of u, it is found that

Substituting this into (4.1) we obtain

$$Q(\Delta-\Delta')=(1/\pi)\Gamma/((\Delta-\Delta')^2+\Gamma^2);$$

$$\Gamma=(2\pi^2/3)N|\bar{u}|. \qquad (4.3)$$

The average value of the modulus is close to U_0.

Thus, due to the $1/R_3$ interaction between the centers, we have a Lorentzian type distribution for level shift changes following the transition of a part of the TLSs into a new state with a width dependent on the number of such TLSs.

We have found earlier that in the energy strip of delocalized excitations, in the time τ_*, Eq. (3.6), about a number N_r, Eq. (3.1), of TLSs will change their state. Noting that $N_rU_0=\Delta_*$, Eq. (4.3) implies that in the time τ_* the characteristic scale for the change in Δ_i in individual TLSs will exceed Δ_*. Since there is no change either between or within the pairs forming the infinite resonant cluster, this change in Δ_i leads to a breakdown of such a cluster in a time of order τ_*. However, due to (4.3) and the uniform distribution of the pairs in Δ_p, Eq. (2.20), and also because of the uniform distribution of the initial TLSs in Δ, it follows that during the same time, an infinite resonance cluster consisting now of *other* pairs will form. As a result a self-consistent picture arises which leads to a continuous growth of the number of TLSs, N, that have undergone a change of state. The kinetics of the process is described by the simplest equation

$$dN/dt=2N_r\Delta_*/\hbar. \qquad (4.4)$$

As we have seen in the derivation of the expression (2.20) for the function $P^{(2)}$, pairs with Δ_p, $\Delta_{0p}\ll T$ are formed primarily by two-level systems with $\Delta,\Delta_0\sim T$. The number of such TLSs is, from (1.2), P_0T. As the self-consistent time evolution proceeds, all of these TLSs will change their state after passing through the stage of their staying in the infinite delocalized cluster.

In order to see this, let us assume first that, having left the resonance strip Δ_*, pairs do not ever come back; neither do individual TLSs, these latter forming resonance pairs with their likes. Then the relaxation time τ_0 for the system of P_0T thermal TLSs is found, from (4.4) and (3.1), to be

$$\tau_0^{-1}\approx 2N_r\Delta_*/P_0T\approx 10(T/\hbar)(P_0U_0)^3. \qquad (4.5)$$

In this time, the characteristic distribution width (4.3) reaches a value of

$$\varphi\approx(2\pi^2/3)\Delta_*(\tau_0/\tau_*)\sim P_0U_0T. \qquad (4.6)$$

As the above value corresponds to the interaction of thermal TLSs at their average separation, it determines the limiting scale of Γ. Let us estimate the probability for an individual pair to return back to a resonance state. Let us divide the time $t-t'$ (as measured from the instant t' at which the pair moved out of the resonance) into intervals τ_*. At an arbitrary instant $m\tau_*$, in the time τ_*, an individual resonance pair may return from the nearest energy interval $\Gamma_0\approx(2\pi^2/3)\Delta_*$ with a probability close to $(\Delta_*/n\Gamma 0)\times(1/m)$ [cf. (4.3); estimates for an individual TLS and for a pair are in fact identical]. The factor of $1/m$ comes from the inclusion of the linear growth of Γ with t, this growth leading to a decrease in the probability distribution. The total reentering probability by the time t is

$$w_b\approx(\Delta_*/\pi\Gamma_0)\ln\{(t-t')/\tau_*\},$$

or, on averaging over the time t',

$$w_b(t)\approx(\Delta_*/\pi\Gamma_0)\ln(t/\tau_*e). \qquad (4.7)$$

For $t\approx\tau_0$, the argument of the logarithm is of order $1/(P_0U_0)$. Even if we take for the parameter P_0U_0 the SiO_2 value of 10^{-3}, $w_b(\tau_0)<1$. This means that, as an approximation, we may ignore pair reentering. Even for extremely small P_0U_0, such that reentering starts to contribute at times less than τ_0, a self-consistent estimate shows $w_b(t)$ to increase with time no slower that $t/\ln(t/\tau_*)$. This means that the conclusions to be reached in the following would not change much.

Now let us estimate the number of pairs formed by an individual TLS with $\Delta_0,\Delta\sim T$ with initial two-level systems with similar parameters under the conditions that $|\Delta'-\Delta|\lesssim P_0U_0T$ and that we have localization in the interval $R_*\pm R_*/6$. The former condition secures entering during the time τ_0 in the resonance interval Δ_*, whereas the latter means that the given TLS forms a resonance pair with $\Delta_{0p}\sim\Delta_*$ and thus can belong to an infinite resonance cluster. Using Eq. (1.2) for this estimate we find [cf. Eq. (3.2)]

$$P_0(P_0U_0T)R_*^3\sim 1.$$

Thus one can argue that once having left the infinite resonance cluster, a TLS will not come back in time τ_0. This confirms the validity of the assumptions that led to Eq. (4.5) and proves that τ_0 given by that expression does indeed characterize the relaxation time of the thermal resonance TLSs.

In analyzing the relaxation caused by the delocalization of collective excitations in a system of TLSs, it is implicitly assumed that the temperature is sufficiently low that we may ignore the relaxation due to the interaction with phonons. For this it suffices that the temperature be less than the value

T' as determined from the relation $\tau_{\mathrm{ph}}^{(0)}(T)\approx\tau(T)$. From the expressions (2.13) and (4.5) and the definitions (2.6) and (3.5) we find

$$T'\approx 3T_0(P_0U_0)^{1/2}. \tag{4.8}$$

The requirement $T<T'$ automatically implies $T\ll T_0$. Thus the linear dimension of the coherent region, R_c, will be much greater than R_*, and the problem turns out to be completely self-consistent.

Thus, for $T<T'$ a radical change of the relaxation regime occurs: the classical law $\tau^{-1}\sim T^3$ changes into the linear $\tau^{-1}\sim T$ law.

The relaxation we have discussed so far is actually the fastest in the system. Really there exist a whole ensemble of non-resonant TLSs with $\Delta_0/\varepsilon<1(\varepsilon<T)$ each of which will also relax, but more slowly. This is readily seen by considering the possibility for a given TLS, with fixed Δ_0 and ε, to form an up-down pair with another TLS, with Δ_0' and ε', under the condition

$$|\varepsilon-\varepsilon'|\lesssim(P_0U_0)T.$$

Under this conditions, we have seen earlier that in time τ_0, Eq. (4.5), such a pair will inevitably find itself in the energy strip

$$|\varepsilon-\varepsilon'|\lesssim\Delta_*. \tag{4.10}$$

The interaction with the collective excitations in an infinite cluster provides, in this case, the possibility of a real up-down energy-conserving transition in the pair. The probability of such a transition is

$$(\Delta_{0p}\tau_*/\hbar)^2, \tag{4.10}$$

where [cf. Eq. (2.12)]

$$\Delta_{0p}\approx(U_0/R^3)(\Delta_0\Delta_0'/\varepsilon\varepsilon') \tag{4.10'}$$

($\Delta_{0p}<\Delta_*$, see below). The effective relaxation time will be determined by the pair with the maximum value of Δ_{0p}.

Let us evaluate the pair distribution function by keeping the parameters of one of the TLSs fixed. To this end we use the expression (2.18), removing the integration over $d\Delta_{01}$ and $d\varepsilon_1$ together with the statistical factor (specifically, let the TLS we have fixed be in the lower state $\varepsilon<T$). Then

$$P^{(2)}(\Delta_{0p},\Delta_p)=(\pi^2/3)(\Delta_0/\varepsilon)(P_0U_0/\Delta_{0p}^2). \tag{4.11}$$

Integrating this over Δ_p within $(P_0U_0)T$, for the total number of pairs with transition amplitude of order Δ_{0p} we find

$$(\pi^2/3)(\delta_0/\varepsilon)(P_0U_0)^2T/\Delta_{0p}. \tag{4.11'}$$

Equating this to unity we find the maximum value of Δ_{0p}, for which the corresponding pair forms with probability about unity,

$$\Delta_{0p}\approx\Delta_*(\Delta_0/\varepsilon). \tag{4.12}$$

Thus Δ_{0p} is indeed less than Δ_*. The structure of the expression (4.12) corresponds to the fact that the optimum size of the pair is R_*, the second TLS being a resonant one. If the TLS under study is resonant ($\Delta_*\sim\varepsilon$), in particular thermal ($\Delta_0\sim\varepsilon\sim T$), then the characteristic transition amplitude equals Δ_*, as it must.

Reentering the interval (4.9) in the course of the evolution process occurs in time of order τ_0. Consequently the inverse relaxation time for a non-resonant TLS is determined by

$$\tau^{-1}(\Delta_0,\varepsilon)\approx\tau_0^{-1}(\Delta_{0p}\tau_*)^2\approx\tau_0^{-1}(\Delta_0/\varepsilon)^2. \tag{4.13}$$

Since the size of the pairs responsible for the transition is R_*, the condition for the realization of coherent coupling, $R_*<R_{\mathrm{c}}$, and hence the restrictions (3.5), also hold for (4.13). The identical dependence of τ_{ph}, Eq. (2.13), and τ, Eq. (4.13), on the ratio (Δ_0/ε) has a consequence that the temperature T', Eq. (4.8), of relaxation regime crossover is common for the resonant and for the slower nonresonant TLSs. Thus, on going over to the relaxation regime due to collective excitations in the infinite resonance cluster, we have the spectrum of relaxation times cut off from below by

$$\tau_{\min}\approx\tau_0. \tag{4.14}$$

5. ANOMALOUS SOUND ABSORPTION MECHANISM

A radical change of the relaxation mechanism at $T<T'$ [see Eq. (4.8)] should alter the nature of internal friction at a frequency

$$\omega<\Delta_*. \tag{5.1}$$

Let us employ the standard formula for internal friction in an amorphous medium (see, e.g., Refs. 3, 4, and 6)

$$Q^{-1}=P_0(\gamma_0^2/\rho c^2T)\int d\Delta\int(d\Delta_0/\delta_0)(\omega\tau(\varepsilon,\Delta_0)/(1+(\omega\tau(\varepsilon,\Delta_0))^2)\mathrm{ch}^{-2}(\varepsilon/2T). \tag{5.2}$$

Let

$$\omega\tau_0\gg 1. \tag{5.3}$$

Then, from Eqs. (4.14), (4.13), and (2.11) it is found that

$$Q^{-1}\approx P_0(\gamma_0^2/\rho c^2)(1/\omega\tau_0). \tag{5.4}$$

Thus, for $T<T'$, the internal friction due to collective excitations in the system of TLSs leads to the temperature dependence of the form [see Eq. (4.5)]

$$Q^{-1}\sim T. \tag{5.5}$$

If the condition (5.3) (with the replacement $\tau_0\Rightarrow\tau_{\mathrm{ph}}^{(0)}$) holds also for $T>T'$, then upon decreasing temperature the internal friction due to phonons, with

$$Q_{\mathrm{ph}}^{-1}\sim T^3 \tag{5.6}$$

will go over to a phononless absorption regime with a linear temperature dependence.

In their internal friction study at $\nu=400$ Hz in amorphous SiO$_2$, Esquinazi *et al.*[6] were the first to observe that at low enough temperature a regime crossover occurs, manifesting itself in the change from the dependence (5.6) to approximately (5.5). It is of interest to compare theoretical estimates with experimental results.

If one writes τ_{ph} in the form $\tau_{\mathrm{ph}}^{-1}=AT^3$ then, as shown in Ref. 6, $A\approx 8\cdot 10^7$.1) Analysis of the body of experimental data suggests the conclusion that the dimensionless parameter P_0U_0 has a value close to $3 \cdot 10^{-3}$. Equating $\hbar/\tau_{\rm ph}$ to Δ_* as given by Eq. (3.3) for the temperature T_0 from Eq. (3.5), we have

$$T_0 \approx (2k_B/\hbar A)^{1/2}(P_0U_0).$$

Using the above-quoted values of A and P_0U_0, an estimate for T_0 is $T_0 \approx 0.2$ K. Then, according to Eq. (4.8),

$$T' = 0.03 \text{ K}.$$

This value is close to the temperature of the crossover to the linear regime predicted in Ref. 6.

At sufficiently low T the condition (5.1) should break down. The characteristic width of the collective excitation quasiband is of order Δ_*. The corresponding temperature bound may be determined from the relation

$$T_b \approx 2 \cdot 10^{-8}/(P_0U_0)^2 (K).$$

This estimate is in reasonable agreement with the experimental value of $T_{\rm b} \approx 2$ mK.

Under the condition opposite to (5.3), integration in (5.2) yields the familiar expression

$$Q^{-1} = (\pi/2)P_0(\gamma_0^2/\rho c^2). \tag{5.7}$$

Experimentally, we here have a plateau in the temperature dependence of the internal friction coefficient. The ratio of the internal friction (5.4) for $T = T'$ to the expression (5.7) is $(2/\pi\omega\tau_0)$. Using the expression for τ_0, Eq. (4.5), and the quoted value for P_0U_0, the ratio is found to be 0.27. The predicted value again turns out to be close to the experimental one. Thus we ascertain a reasonable agreement between theoretical and experimental results.

There is one point to note, however. Throughout the paper, and entirely for the sake of simplicity, we have used a single effective value for each the sound speed and the quantity γ_0. However, when comparing absolute absorptions, we must remember that experimentally one measures the absorption of the longitudinal sound, whereas the indirect interaction between two- level systems is to a large extent determined by transverse phonons. Consequently the ratio $\gamma_0^2/\rho c^2$ entering Eqs. (5.4) and (5.7) turns out to be much less than U_0, Eq. (2.6). This is presumably the reason for the discrepancy between the above cited value of P_0U_0 and the effective value of its analog for the longitudinal sound, the latter being deducible from the value of internal friction in the plateau region.

Upon a considerable increase in ω we enter, in a natural way, a regime which corresponds to Eq. (5.7). In this case, the internal friction does not depend on the relaxation time at all, and does not differ from the phonon absorption case. Therefore, under these conditions it is impossible to trace the relaxation mechanism crossover.

However, the crossover to a new regime manifests itself, under these conditions, in the renormalization of the sound speed. In fact, for $\omega\tau_{\rm min} < 1$, the relaxational renormalization of the sound speed is determined by (see, e.g., Refs. 3 and 6)

$$(\Delta c/c)_{rel} \approx -(P_0\gamma^2/2\rho c^2)\ln(1/\omega\tau_0). \tag{5.8}$$

The crossover of the relaxation regime at $T \sim T'$ is accompanied by a decrease in the coefficient of ln(T) by a factor of three. In all cases there simultaneously occurs a resonant sound renormalization associated with the interaction with the initial TLS structure,

$$(\Delta c/c)_{\rm res} \approx +(P_0\gamma^2/2\rho c^2)\ln(T/\Delta_{0\max}). \tag{5.9}$$

As a result, the total sound speed renormalization, rather than changing the sign of its temperature derivative on achieving the condition $\omega\tau_{\rm min} \approx 1$ at $T > T'$, undergoes a sign-conserving change of the derivative in the region $T < T'$. The behavior of the dependence of $\Delta c/c$ on ln(T) in the latter case is reminiscent of sound speed renormalization in a metal.[3]

Recently, the $Q^{-1} \sim T$ behavior has also been found in superconducting polycrystalline Nb in the temperature range $T \lesssim 100$ mK (Ref. 14). That the crossover temperature T' is higher than in $\rm SiO_2$ may result from the inter-TLS interaction being stronger due to the indirect exchange by virtual electron-hole pairs.

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