Dephasing Rate in Dielectric Glasses at Ultralow Temperatures

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A novel mechanism for dephasing in dielectric glasses is considered. The mechanism is due to the delocalized collective excitations arising in the ensemble of the interacting two-level systems. The spectral diffusion induced by these excitations gives rise to the phonon-independent transverse relaxation. The mechanism results in the linear temperature dependence of the dephasing rate and becomes predominant at ultralow temperatures. A qualitative agreement with the experimental data is found. [S0031-9007(98)05683-X]

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The investigation of dielectric glasses has revealed the anomalous temperature behavior of their relaxation properties at ultralow temperatures. For example, the standard temperature-dependent longitudinal relaxation rate, $\tau_{1-\mathrm{ph}}^{-1} \sim T^3$, is well known to be inherent to the tunneling model [1] and is due to the relaxation driven by the one-phonon processes [2] in the two-level systems (TLS). However, the measurement of the internal friction [3] in amorphous SiO_2 and experimental investigations of the dielectric losses in different amorphous solids [4] has brought out that this dependence crosses over into $\tau_1^{-1} \sim T$ if the temperature is low enough.

On the other hand, a linear temperature dependence has also been found for the transverse relaxation rate τ_2^{-1} in the experiments concerning the echo-type investigation of the phase memory time τ_2 in Suprasil-I at temperatures $T < 30$ mK [5,6] and in the orientationally disordered systems $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ at $T < 50$ mK [7,8]. The dephasing, induced by the phonon-assisted longitudinal relaxation of the TLS, should result in just the same temperature dependence T^3. Another and more effective dephasing mechanism, which is governed by the spectral diffusion induced by the phonon-assisted transitions occurring in the surrounding TLS, leads to the dependence [9,10] $\tau_{2-\mathrm{ph}}^{-1} \sim T^2$. This mechanism is due to the $1/R^3$ interaction between the TLS. The key role of this interaction in understanding the relaxation properties of the dielectric glasses at ultralow temperatures has been demonstrated in Refs. [11,12]. It is shown in these papers that a new type of delocalized multiparticle excitation appears in the ensemble of the TLS interacting by the $1/R^3$ law. In addition, it is the relaxation of these excitations that gives rise to the anomalous relaxation properties of the glasses.

An explanation of the linear temperature dependence for τ_1^{-1} involving the relaxation of this type of excitations was given in the paper [12]. The purpose of this Letter is to show that the dephasing mechanism and the linear temperature dependence for the dephasing rate τ_2^{-1} are both closely related to the dynamics governed by these multiparticle excitations.

An isolated TLS can be described by the standard pseudospin Hamiltonian

$$h_i = -\Delta_i S_i^z - \Delta_{0i} S_i^x . \tag{1}$$

Following Ref. [1], we accept that the distribution function for the asymmetry Δ and the tunneling amplitude Δ_0 obeys the universal distribution

$$P(\Delta, \Delta_0) = \frac{\overline{P}}{\Delta_0} . \tag{2}$$

The interaction between the TLS in the dielectric glasses arises either from the strain field or from the direct electrical dipole-dipole interaction and can be described by the Hamiltonian

$$\hat{V} = \frac{1}{2} \sum_{i,j} U(R_{ij}) S_i^z S_j^z , \qquad U(R_{ij}) = \frac{U_0}{R_{ij}^3} , \tag{3}$$

where R_{ij} is the distance between two TLS, and U_0 is the characteristic coupling constant. The dimensionless parameter $\overline{P} U_0 \approx 10^{-3}$ for all the known glasses [13]. This condition indicates that the interaction between TLS is very weak.

Let us consider relaxation of an *excited* TLS with certain energy splitting $E = (\Delta^2 + \Delta_0^2)^{1/2}$. The simplest relaxation channel different from the phonon-assisted one is a hopping of the excitation from the excited TLS to another TLS which is initially in the *ground* state with the parameters Δ', Δ_0', E'. As a result, the first *TLS* excited before goes over into the ground state, while the second TLS proves to be in the excited state. The inverse process also takes place so that the pair of the TLS under consideration can be detected in one of the two states separated by the energy interval $|E - E'|$. In what follows, these two states of the TLS pair will be referred to as a *flip-flop configuration*. Such a TLS pair can be considered as a *new type* of the two-level system with the asymmetry $\Delta_p = |E - E'|$. In addition, as follows from Eqs. (1) and (3), the tunneling amplitude that couples two states of the flip-flop configuration of the TLS pair is given by the relation [10,12,14].

$$\Delta_{0p}(R) \approx U_0(R) \frac{\Delta_0 \Delta_0'}{E E'} . \tag{4}$$

Thus, one should describe the relaxation of the initially excited TLS in terms of the transition between the states of the flip-flop configuration of the TLS pair.

If a two-level system with the parameters Δ and Δ_0 was initially in the first state, the time-dependent probability to find the system in the second state is given by the solution of the corresponding temporal Shroedinger equation

$$W_2(t) = \frac{\Delta_0^2}{\Delta^2 + \Delta_0^2} \sin^2(\sqrt{\Delta^2 + \Delta_0^2} \cdot t). \quad (5)$$

This relation applied to the transition between the levels of the flip-flop configuration of a TLS pair means that the transition probability is noticeable provided the parameters of the TLS pair obey the condition

$$\Delta_p = |E - E'| < \Delta_{0p}(R). \quad (6)$$

Hereafter such a TLS pair is referred to as a *resonant pair* (RP). In addition, the frequency of the quantum mechanical oscillations between the states of the flip-flop configuration

$$\tau^{-1} = \sqrt{(\Delta_p^2 + \Delta_{0p}^2)} \approx \Delta_{0p}(R) \quad (7)$$

is completely determined by the parameter $\Delta_{0p}(R)$.

Let us consider an excited TLS with the parameters Δ_0, E and estimate the minimum size of a resonant pair $R_{\min}(\Delta_0, E)$ involving this TLS. For this purpose, one should calculate the average number $N[R; \Delta_0, E]$ of the TLS in the ground state with the parameters Δ_0', E' which compose a resonant pair with the examined excited TLS within the sphere of radius R. Using Eqs. (2), (6), and (4), we find

$$N[R; \Delta_0, E] = \overline{P} \int d\Delta' \int \frac{d\Delta_0'}{\Delta_0'} \int_a^R d\mathbf{r} \cdot \Theta[\Delta_{0p}(r) - \Delta_p] \approx \frac{\Delta_0}{E} \overline{P} U_0 \ln\left(\frac{R}{a}\right), \quad (8)$$

where Θ is the Heaviside function involving the resonant condition Eq. (6) and a is the minimum permissible distance between two TLS. The minimum size of a resonant pair $R_{\min}$ is a radius within which one can detect a resonant pair with a probability close to unity, i.e., $N[R; \Delta_0, E] \approx 1$. Therefore $R_{\min}(\Delta_0, E) \approx a \exp\left(\frac{E}{\Delta_0 \overline{P} U_0}\right)$. The rate of oscillations (7) for the pair of such size is given by the relation

$$\tau_{\min}^{-1} \approx \Delta_{0p}(R_{\min}) \approx \frac{U_0}{a^3} \frac{\Delta_0}{E} \exp\left\{-\frac{E}{\Delta_0 \overline{P} U_0}\right\}. \quad (9)$$

Let us note that the *relaxation* rate, at least, is smaller than the rate of the quantum mechanical frequency $\tau_{\min}^{-1}$. Bearing in mind the smallness of the parameter $\overline{P} U_0$ and the fact that Δ_0 is always less than E, we conclude that the value of the relaxation time obtained, if any, is too large to be observed. So, during such a long period, an excited TLS would relax via the standard phonon-assisted mechanism. In addition, one should also pay special attention that the estimation (9) allows us to conclude that any attempt (see Ref. [15]) to link the relaxation in the glasses with the single-quantum phononless delocalization has no real basis.

The scenario of the phononless relaxation mechanism described above is valid only if every RP relaxes *independently* and, thus, only *two* TLS take part in the elementary relaxation act. This assumption is valid provided one can neglect the interaction between *different excited* TLS. Below we will show that, because of the availability of the *macroscopic* number of excited TLS in the system at temperature $T > 0$ and due to the $\frac{1}{R^3}$-law interaction between TLS, this assumption is not valid, and the relaxation in the ensemble of the interacting TLS is of a multiparticle origin. In other words, one should take into account the simultaneous relaxation of *two* RP and thus the *four* TLS will participate in the elementary act [11,12,14].

Generally speaking, any RP has four energy levels. Two of them correspond to the flip-flop configuration mentioned above. The two rest states of the pair correspond to the configuration when both TLS are either in the excited or in the ground state. Let us explain why a *resonant pair* [see Eq.(6)] can be considered as a new type of TLS. In fact, because of the Gibbs factor one can restrict the consideration to those TLS for which $E \approx T$. Then one should take into account that the larger the value of the tunneling amplitude $\Delta_{0p}(R)$, the greater the probability to form a resonant pair [see Eq. (6)]. Thus it directly follows from Eq. (4) that one can confine oneself to the condition $\Delta_0 \approx E$. Therefore, in what follows, one can consider only RP composed from the TLS for which $\Delta_0 \approx E \approx T$.

The states of the flip-flop configuration are separated by the energy interval $\Delta_p = |E - E'|$. In spite of the fact that $E, E' \approx T$ one can construct an RP for which $\Delta_p, \Delta_{0p} \ll T$. Then, even if the interaction $V(R)$ between these TLS is weak, the condition $\Delta_p \leq \Delta_{0p}$ can be valid. Therefore the two levels of the flip-flop configuration are strongly coupled [see Eq. (6)]. On the other hand, the rest two levels are separated from the flip-flop configuration by the energy interval of the order of the magnitude of temperature T, while the amplitude which couples them to the flip-flop configuration levels does not exceed at least $\Delta_{0p} \ll T$. For this reason, these two levels are very weakly coupled to the flip-flop configuration levels. It is this circumstance that, in the remainder of this Letter, allows us to consider only the flip-flop configurations of a RP and therefore to treat an RP as a kind of two-level system with the energy asymmetry $\Delta_p = |E - E'|$ and tunneling amplitude $\Delta_{0p}(R)$ (4).

Thus, below we investigate the relaxation of this novel RP type of the two-level system for which the distribution

function for the parameters Δ_p and Δ_{0p} is defined as

$$P^{(2)}(\Delta_p, \Delta_{0p}) = \left\langle \delta(\Delta_p - |E - E'|) \times \delta\left(\Delta_{0p} - \frac{U_0}{R^3}\frac{\Delta_0\Delta_0'}{EE'}\right)\right\rangle, \quad (10)$$

where the brackets denote the two averaging, namely, the Gibbs averaging and averaging over the distribution of the parameters of the original TLS [see (2)]. In addition, the integration over the distance R is implied, so the left-hand side of Eq. (10) can be rewritten as

$$\int d^3R \int P(\Delta, \Delta_0)\, d\Delta\, d\Delta_0 \int P(\Delta', \Delta_0')\, d\Delta'\, d\Delta_0' n(E) \times [1 - n(E')]\delta(\Delta_p - |E - E'|)\delta\left(\Delta_{0p} - \frac{U_0}{R^3}\frac{\Delta_0\Delta_0'}{EE'}\right), \quad (11)$$

with $n(E) = [1 + \exp(E/T)]^{-1}$ being the probability to find a TLS in the excited state. To estimate expression (11), it is necessary to take into account that because of the Gibbs factor the integral is determined mainly by the region of $E \approx E' \approx T$. Therefore, due to $\Delta_p \ll E \approx T$ one can omit Δ_p in the argument of the first delta function. After these comments one can easily estimate the pair distribution function within the logarithmic accuracy [12]

$$P^{(2)}(\Delta_p, \Delta_{0p}) \approx (\bar{P}T)(\bar{P}U_0)\frac{1}{\Delta_{0p}^2}. \quad (12)$$

One should pay attention that the distribution function (12) differs from (2) but also does not depend on the parameter Δ_p at all. The distribution function (12) has a stronger singularity at small Δ_{0p} than Eq. (2) at small Δ_0. For this reason, the concentration of the low energy RP excitations is larger (and correspondingly the average distance between them is smaller) than for the initial TLS. The coupling constants $U(R)$ for both TLS and RP being of the same order of magnitude, the low energy RP interact and relax stronger than the TLS.

The ensemble of RP, a new kind of the TLS, is described by the initial Hamiltonian Eqs. (1) and (3). The only, but a key, distinction from the initial model is that the distribution function (12) should be used instead of Eq. (2) at temperature $T > 0$. Again, let us remember that we are only interested in the RP for which $\Delta_p \leq \Delta_{0p}$.

To reveal the relaxation properties of the introduced RP model, let us consider an RP strip k with the transition amplitude lying within an interval $[\Delta_{0p}(k) - \Delta_{0p}(k)/2, \Delta_{0p}(k) + \Delta_{0p}(k)/2]$, with $\Delta_{0p}(k)$ being equal to any permissible value of Δ_{0p}. One can estimate the concentration of the RP N_k within this strip taking into account the distribution function (12) as

$$N_k \approx (\bar{P}T)(\bar{P}U_0) = N_*. \quad (13)$$

An important conclusion should be made. The RP concentration in the chosen strip, marked by the index number k, is completely independent of the chosen value $\Delta_{0p}(k)$. Picking out the strips corresponding to all permissible values Δ_{0p}, one can cover completely the whole ensemble with the RP. Within any strip the RP concentration is constant N_*. Because of the last circumstance the average distance between the RP within any strip does not depend on the kind of the strip and equals $R^* \approx N_*^{-1/3}$. Since the interaction between the RP is of the same origin as between the former TLS, the interaction between the RP is given by the expression U_0/R^3 [see Eq. (3)]. Therefore the typical energy of the interaction between the RP within any of the strip is

$$U(R_*) \approx U_0/R_*^3 \approx T(\bar{P}U_0)^2. \quad (14)$$

Because of this interaction the RP asymmetry energy $\Delta_p(k) \gtrsim U(R_*)$. Bearing in mind that for any RP $\Delta_p \leq \Delta_{0p}$, below one should consider only the RP strips, for which $\Delta_{0p}(k) \gtrsim U(R_*)$.

The RP being a kind of TLS, one can introduce a concept of a flip-flop configuration for two RP and deduce the expression for the transition amplitude Δ_0 between the levels of this configuration [which is similar to that of Eq. (4)]. So for the strip k one obtains that

$$\Delta_0(k) \approx U(R_*)\left(\frac{\Delta_{0p}(k)}{E_p(k)}\right)^2. \quad (15)$$

Let us investigate the relaxation of the RP due to its interaction with the RP of the same strip. Like the TLS case the relaxation of the RP, being a kind of TLS, is possible only if the RP within the strip are strongly coupled [see Eq. (6)], i.e.,

$$\Delta(k) = |E_p(k) - E_p'(k)| \leq \Delta_{0p}(k). \quad (16)$$

Let us consider the strip for which $\Delta_{0p}(k) \approx U(R_*)$. Since $E_p(k) \approx \Delta_p(k) \approx \Delta_{0p}(k) \approx U(R_*)$ in this case, it proves to be that $\Delta(k) \leq \Delta_0(k) \approx U(R_*)$. Thus the condition (16) is valid. The rate of the quantum mechanical oscillations for the RP [similar to that in Eq. (9)] is $\tau_*^{-1} \approx \Delta_0(k) \approx U(R_*)$. In addition, we simultaneously arrive at the situation which is responsible for the appearance of the infinite cluster of the strongly coupled RP. If one attempts the problem in the spirit of the general concept of the delocalization in the disordered media, developed by Anderson, we conclude that the excitations in this cluster should be delocalized and relaxation of the excitation within the strip takes place at the rate τ_*^{-1}.

Let $\Delta_{0p}(k) \gg U(R_*)$. Then $E_p(k) \gg U(R_*)$, and consequently $\Delta(k) \gg U(R_*)$. On the other hand, as follows from Eq. (15), at any case $\Delta_0(k) \leq U(R_*)$. Thus the criterion (16) is not fulfilled and relaxation of the RP within the strip of such kind does not take place.

Thus only the RP within the strip characterized by the tunneling amplitude $\Delta_{0p}(k) \approx U(R_*)$ do relax, and the relaxation rate is equal to

$$\tau_*^{-1} \approx U(R_*) \approx T(\bar{P}U_0)^2. \quad (17)$$

The relaxation of a RP is nothing else but a relaxation of two TLS. For this reason, the relaxation of the RP

combined with the $1/R^3$ law for the interaction between the TLS gives rise to the spectral diffusion meaning the dephasing in the system.

Let us consider the time-dependent fluctuation of the phase at an arbitrary probed TLS $\delta\Phi(t)$, induced by the energy-splitting fluctuation $\delta E(t)$. The proper dephasing time τ_2 should be estimated from the relation

$$\delta\Phi(\tau_2) = \tau_2 \delta E(\tau_2) \approx 1 . \tag{18}$$

First, let us calculate the energy-splitting fluctuation $\delta E(t)$ within the time interval $t = \tau_*$. The overall number of the RP and therefore approximately the number of the TLS undergoing the transition during the interval $t = \tau_*$ is of the order of the magnitude of N_*. Bearing in mind the fact that every RP experiencing the transition at the distance r from the probed TLS contributes a value $U(r)$ [see Eq. (3)] into the energy-splitting fluctuation for the probed TLS, one can estimate

$$\delta E(\tau_*) = \sum_r U(r) \approx N_* U_0 \approx U(R_*) .$$

Then, taking into account Eq. (17), one can find that $\tau_*^{-1} \delta E(\tau_*) \approx 1$. The comparison of this relation with Eq. (18) allows us to link τ_* with τ_2 and therefore

$$\tau_2^{-1} \approx U(R_*) \approx \frac{T}{\hbar} (\bar{P} U_0)^2, \tag{19}$$

where Planck's constant $\hbar$ is introduced to restore the correct dimensionality. This dephasing rate decreases linearly with the temperature. Compared with the phonon-induced channel providing a T^2 dependence (see [9]), the dephasing governed by the mechanism concerned predominates at sufficiently low temperatures.

It was found in [12] that, for Suprasil-I at temperature $T' \approx 30$ mK, the longitudinal relaxation governed by the mechanism suggested crosses over into that driven by the traditional phonon-induced mechanism. The crossover point for the transverse relaxation is determined from the relation $\tau_{2-\mathrm{ph}}(T'') \approx \tau_2(T'')$. The calculation of T'' shows that it has the same value as T'. Thus, the transition to the relaxation regime driven by the phononless mechanism occurs for $\tau_1(T)$ and $\tau_2(T)$ simultaneously.

A number of experimental data is available for the direct measurement of $\tau_2(T)$ at ultralow temperatures [5,6]. A linear temperature dependence for the dephasing rate $\tau_2^{-1}(T)$ at $T < 30$ mK has been reported in these papers. At higher temperature the phonon-induced spectral diffusion dominates, and τ_2^{-1} follows a T^2 dependence (see [16]). In addition, recently the measurement of the transverse relaxation rate has been made on a quite different material $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ which is known to exhibit glassy properties [8]. Here again the linear temperature dependence for τ_2^{-1} was revealed for $x = 0.08$ at $T < 50$ mK. These experiments are in agreement with the result obtained in this Letter.

Thus, our approach for the dephasing rate in the amorphous systems indicates the decisive role of the long-range interaction between the tunneling centers and strongly supports the existence of the novel relaxation mechanism in the glasses considered in [11,12].

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