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Relaxation in glasses at low temperatures

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The interaction between tunneling system inherent in amorphous solids is established to be responsible for the univerasl behavior of their kinetics and thermodynamics properties at low temperature. In the paper we describe the relaxation mechanism induced by the intercation which falls down as R^{-3} at large distance. This interaction is either the electrostatic dipole - dipole one or is the elastic one between the point defects (tunneling system). In the last case, the intercation is due to indirect interaction induced by acoustic virtual phonon exchange. The relaxation gets significant at sufficiently low temperature when phonons are substantially frozen out. We show that in the realistic experimental situation the measuring field strongly accelerates the interaction stimulated relaxation. The characteristic temperature and field dependences of the relaxation rate are found when the rate is affected both by the interaction between tunneling systems and by the external field.

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1. Introduction

For a long time the standard model of *non-interacting* two-level-systems (TLS) [1] has served as a good background for understanding experimental data in glasses at low temperatures $T \leq 1\,\mathrm{K}$. The further investigations revealed that below 100 mK almost all measurements in dielectric glasses dealing with their relaxation properties [2 – 6] cannot be treated ignoring the TLS interaction. For this reason one can suppose that that below 100 mK TLS manifest the collective behavior induced by the interaction between them.

It is well established that in amorphous solids TLS are coupled with phonons. In dielectric glasses at $T \ll \Theta_D$ (Θ_D is the Debye temperature) coupling with acoustic phonons plays a main role. The virtual exchange of TLS by acoustic phonons results in the indirect interaction between TLS, which decays with the distance R as R^{-3}. In particular, this interaction gives rise to the flip-flop transitions between two TLS (see Fig.1). The transition amplitude also decays with the distance as R^{-3} [7, 8]. In a number of dielectric glasses, TLS possess their own dipole moment. In this case the electrostatic dipole-dipole interaction can become the dominating interaction between TLS [9]. This interaction also decreases with the distance as R^{-3}. It was experimentally discovered by Arnold and Hunklinger [10] (see also [9]) that the $1/R^3$ interaction contributes to the spectral diffusion of the TLS energy. This spectral diffusion involves the dynamic fluctuation of the TLS energy due to its interaction with neighboring TLS, making transitions between their levels [11].

Along with the interaction significance for the acoustic [10, 9] and optical [12 – 14] hole burning experiments, it also affects the decay of coherent echoes [15]. Also the interaction between TLS has been revealed in the non-equilibrium dielectric measurements [5, 16], giving rise to the reduction of the spectral density near zero energy. This is interpreted as the dipole gap formation [17]. In addition, it has been found that the transition from coherent to incoherent tunneling takes place if the typical interaction energy exceeds tunneling splitting [18]. Recently discovered anomalous low-temperature behavior of amorphous solids in the

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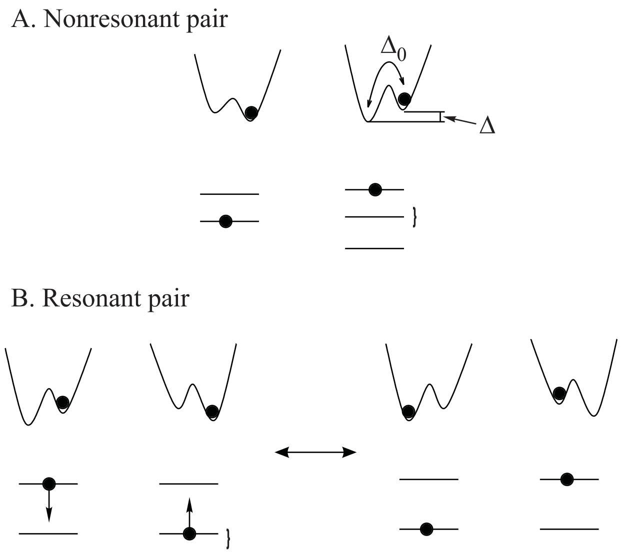

Fig.1. Energy transport between excited TLS (left) and nonexcited TLS (right). It is not efficient in the off-resonant case (A), where the enrgy level mismatch exceeds the resonant interval indicated by figure bracket. For the resonant pair (B) the intercation of TLS induces flip-flop transitions mixing up two possible quasi-dehenerated states, where either first or secon TLS is excited, while the remaining TLS is in its ground state

external magnetic field [19 – 24] can also be associated with the TLS interaction.

The relaxation in a disordered system can be due to delocalized excitations. The single particle energy delocalization is not efficient for the ensemble of interacting TLS because the static energy disordering is very strong there. Therefore, the Anderson localization [25] of all excitations takes place. In other words, the localization in the disordered TLS system is due to a large energy level mismatch for a typical pair of TLS (Fig.1A) compared with the flip-flop transition amplitude for this pair. Suppose that a TLS excitation energy can experience large time-dependent fluctuations. These fluctuations can reduce the energy mismatch value, stimulating level crossing and supporting flip-flop transitions (see the resonant pair in Fig.1B). Such fluctuations can be induced by either the external alternating (measuring) field or they can be due to the spectral diffusion, caused by transitions of neighboring TLS. Thus, such fluctuations can promote the delocalization in the system. In the absence of the external field this delocalization can be treated as a self-consistent process in which the spectral diffusion induces TLS transitions. In its turn, TLS transitions cause the spectral diffusion due to TLS interaction.

This paper is a comprehensive presentation of our results concerning the effect of R^{-3} interaction on the dynamic properties of amorphous solids. It is organized as follows. In Sect.2 we address the single particle localization problem for zero temperature and zero external alternating field. In Sect.3 the effect of an external alternating field on the single particle localization is studied. We demonstrate that the field having a sufficiently large amplitude and a small frequency can break the Anderson localization. In Sect.4 we study the finite temperature many-body delocalization of a TLS energy within the TLS ensemble induced by the self- consistent dynamics of resonant pairs (see Fig.1B). Sect.5 is devoted to the most complicated, but experimentally relevant regime where both external field and interaction of TLS must be taken into account simultaneously. In Summary we present the brief discussion of the application of our theory to the experimental studies.

2. Anderson localization of the TLS energy. The relaxation of TLS is mainly due to *resonant* TLS for which bias asymmetries Δ and tunneling amplitudes Δ_0 are of the same order of magnitude. The energy $E = \left(\Delta^2 + \Delta_0^2\right)^{1/2}$ is supposed to be uniformly distributed within a wide energy interval, with P being the distribution constant. The pseudospin 1/2 Hamiltonian of the interacting *resonant* TLS reads [8, 26, 27]

$$H = -\sum_{\iota} E_i S_i^z - \sum_{ij} U_{ij} S_i^x S_j^x, \quad U(R_{ij}) = \frac{U}{R_{ij}^3}, \quad (1)$$

U_{ij} is the coupling amplitude, and the amplitude constant $\langle U \rangle = 0$. The phononless relaxation induced by the interaction is due to TLS pair obeying the resonant condition

$$\mid E - E' \mid < U(R_{ij}). \quad (2)$$

Hereafter such a TLS pair is referred to as a *resonant pair* (RP).

The Anderson delocalization and/or localization are explicitly associated with a TLS capability to form resonant pairs. If the typical TLS has on average the large number of resonant neighbors, one would expect the delocalization. In fact, each resonance shares the energy between two TLS. One can use a percolation analogy, connecting all resonant pairs into a cluster. When the infinite cluster can be formed, excitations become delocalized (cf. Ref.[28]). The average number N_0 of TLS, forming a resonant pair with a certain given TLS reads

$$N_0 \approx \chi \ln\left(\frac{L}{a}\right), \chi = 4\pi PU. \quad (3)$$

L is the size of the whole system and a is the minimum distance between two TLS. Since in all glasses the parameter $\chi \propto PU$ is very small (see e.g. [29, 30])

$$\chi = 4\pi PU \leq 10^{-3} - 10^{-2}, \quad (4)$$

the number of resonant neighbors is much less than unity for any reasonable sample size L. Therefore, only a small fraction of two level systems belongs to resonant pairs, while majority of the others are immobile. This proves the full Anderson localization of all excitations in this system.

3. Delocalization of Floquet states. Consider the effect of an external alternating field on the Anderson localization within the TLS ensemble at *zero* temperatures. An approximate zero temperature condition means that one can neglect the interaction between excited TLS because their total number is negligibly small in this case. In particular, there is no spectral diffusion. Due to the field, the energy E_i acquires the oscillating part $a_i \cos(\omega t)$. We assume that the field varies sufficiently slow and the amplitude a_i is small compared to the typical scale of the excitation energy

$$\omega \ll a_i \ll E_i. \tag{5}$$

Let b_i be the amplitude of the excited TLS state (pseudospin projection up) at the site i. An evolution of a single-particle excitation can be described by the Schrödinger equation with the external field involved

$$i\dot{b}_i = (E_i - a_i \cos(\omega t))\, b_i + \sum_j U_{ij} b_j. \tag{6}$$

Equation (6) can be rewritten in terms of the Floquet state amplitude d_{in} [31]

$$\begin{gathered} \varepsilon d_{in} = (E_i - n\omega)\, d_{in} + \sum_{jp} T_{in;jp} d_{jp}, \\ T_{in;jp} = U_{ij} J_{p-n}\left(\frac{a_j - a_i}{\omega}\right) \end{gathered} \tag{7}$$

with ε being an eigenvalue of a Floquet eigenstate quasienergy. Here $J_p(x)$ is the Bessel function of order p.

If the hopping term $T_{in;jp}$ is neglected, the eigenstates of the system in the external field correspond to localized single-site excitations of a TLS coherently dressed by n "quanta" of the frequency ω, named dressed excitation. The resonant condition for this excitations, similar to (2), reads

$$|E_i - n\omega - E_j| \le |T_{i0;jn}| = \left|U_{ij} J_n\left(\frac{a_j - a_i}{\omega}\right)\right|, \tag{8}$$

This criterion is similar to multiphoton resonances in the nonlinear optics.

Let us denote the average of a site-fluctuating difference $|a_i - a_j|$ by a. The argument of the Bessel function in Eq. (7) is a large parameter of order of $a/\omega \gg 1$ (see Eq. (5)). If $|n| > a/\omega$, the magnitude of the corresponding Bessel function is exponentially small, entailing a negligible probability of resonant coupling. The opposite condition $a >| n | \omega$ means that, in order to have resonant coupling, the energy difference should be at least less than the field amplitude a:

$$| n |< a/\omega \to \quad |E_i - E_j| < a. \tag{9}$$

On the classical language this condition means that the interaction of TLS can be efficient only when the external field stimulates their real level crossing.

Under condition (9) and $a/\omega \gg 1$ the Bessel function in (7) can be approximately replaced by its asymptotic value. Omitting the standard oscillating prefactor which plays no role in a random discrete problem, one can estimate the coupling amplitude for dressed excitations (DE) Eq. (7) as

$$|T_{i0;jn}| \approx \frac{U}{R_{ij}^3}\sqrt{\frac{\omega}{a}}, \tag{10}$$

Consider the delocalization of an excitation due to the alternating field within the framework of the concept of resonant coupling. Two sites i and j are in the resonance when the condition Eq. (8) is satisfied at least for some $n < a/\omega$ (see Eq. (9) and Fig.2). Since the energy splitting between two subsequent levels is equal

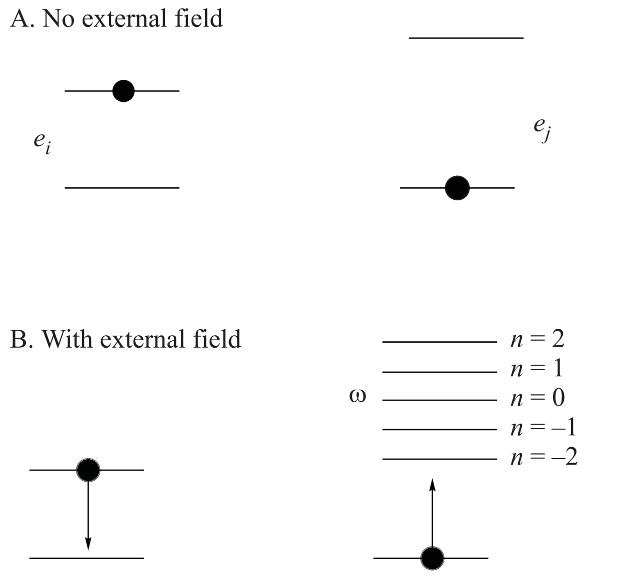

Fig.2. The formation of resonant pair from the initially non-resonant one (A) in the external alternating field. The coherent excitation dressing (B) enables transitions with the energy change given by the integer number n of field frequencies. The case $n = -2$ satisfies the resonance conditions (see Eq. (8))

to the field frequency ω, the resonance *always* occurs if the hopping amplitude $T_{i0;jn}$ exceeds ω and condition

(9) is fulfilled. As directly follows from Eq. (10), this happens when sites i and j are separated by a sufficiently small distance R_{ij}

$$R_{ij} < r_* = \left(U/\sqrt{a\omega}\right)^{1/3}. \quad (11)$$

Thus all TLS j located closer than the crossover distance r_* to TLS i under consideration with $|E_i - E_j| < a$ are resonantly coupled with each other. Taking into account that the number of such centers in a unit volume is equal to Pa, we find that the total number of the resonant neighbors within the sphere of a radius r_* is given by

$$W(r_*) = \frac{1}{3}\chi\sqrt{\frac{a}{\omega}}. \quad (12)$$

At larger distances $R > r_*$ the total number of resonances in each layer $r_* < r_1 < R < r_2$

$$W(r_1, r_2) \approx P\int_{r_1}^{r_2} d^3R\left(\frac{U}{R^3}\sqrt{\frac{\omega}{a}\frac{a}{\omega}}\right) = \chi_* \ln\frac{r_2}{r_1},$$
$$\chi_* = 4\pi PU\sqrt{\frac{a}{\omega}}, \quad (13)$$

increases by the factor $\sqrt{a/\omega}$ as compared to the zero field case (3).

According to the Levitov's analysis of the delocalization problem in the case of the $1/R^3$ energy hopping amplitude [32], the parameter χ_* is a decisive parameter for the delocalization. As follows from Eqs. (12) and (13), when $\chi_* \ll 1$ the first resonance occurs at the distance $R_a \sim r_* e^{1/\chi_*} \gg r_*$. Thus, the first hop of the excitation occurs to the distance R_a, taking some time t_1 determined by the inverse hopping amplitude $t_1 \sim R_a/U \propto e^{3/\chi_*}$. Then, the time required for the second hop, when the next resonance appears, is exponentially large compared with t_1 [32]. Therefore, the delocalization is exponentially slow, if any.

In the opposite case $\chi_* > 1$, the number of resonances exceeds unity in each spherical layer $r_1 < R < 2r_1$ $(r_1 > r_*)$ and the resonant sites form *an infinite cluster* meaning an existence of the delocalized state [8].

The inverse time of a single hop between two nearest resonant neighbors in the delocalization regime can be treated as a relaxation rate for the DE located at a certain site. The distance $R_a < r_*$ between these neighbors can be estimated from the relation $W(R_a) \approx 1$ (see Eq. (12)). This radius is determined as

$$R_a \approx (Pa)^{-1/3} \quad (14)$$

and the typical hopping amplitude corresponding to the distance R_a estimates the inverse lifetime (the relaxation rate) for the DE

$$\tau_*^{-1} \approx T(R_a) = \frac{U}{R_a^3}\sqrt{\omega/a} = (a\omega)^{1/2}\chi. \quad (15)$$

4. Many body delocalization, dephasing and relaxation. Here we will discuss the case of the zero field and a finite temperature $T > 0$. The Anderson localization of excitations proved in Sect. 2 takes place only if every resonant pair RP can be treated independently. This assumption is valid provided that one can neglect the interaction between different excited TLS. We will see that this is not the case at any finite temperature $T > 0$. Below we will show that at any finite temperature the long-range $1/R^3$ interaction of excited TLS leads to the irreversible dynamics and relaxation. This relaxation is essentially of a many-body origin. In other words, one should take into account simultaneous transitions in two or more RP (see Fig.3) and thus at least four TLS will participate in an elementary process [8, 27, 33, 34].

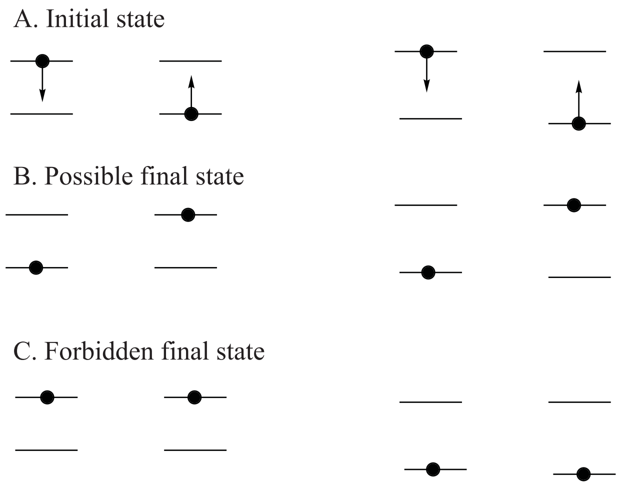

Fig.3. The interaction of resonant pairs leads to the many-body delocalization (A) The excitation is transferred due to the simulteneous flip-flop transitions of four TLS to their new states (B). The transitions involving two TLS belonging to different resonant pairs (C) are not allowed because of the energy mismatch

Any RP has four energy levels. Two of them correspond to the flip-flop configuration mentioned above (see Fig.2B). The two other states of a pair correspond to the configuration where both TLS are either in their excited or their ground states. In fact, the typical energy of excited TLS in a resonant pair is given by the thermal energy T, while their flip-flop transition amplitude V_{ij} is generally much smaller due to the weakness of the interaction Eq. (3). Therefore the flip-flop interaction Δ_{0p} can connect only pairs of TLS, where one of them is in its excited state and the other one is in its ground state (Fig. 2). The other two states are separated from the flip-flop pair by the large energy gap of order of the temperature.

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 RP with $\Delta_p, \Delta_{0p} \ll T$. 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 in the resonant pair can be strongly coupled (see Eq. (2)). In the remainder of this paper we will consider only flip-flop configurations of RP. We will treat RP as a new kind of the two-level system with the energy asymmetry $\Delta_p =| E - E' |$ and the tunneling amplitude $\Delta_{0p}(R)$.

Thus below we investigate the relaxation of this novel RP type of the two-level system for which the distribution function of parameters Δ_p and Δ_{0p} is defined as [8, 30]

$$P^{(2)}(\Delta_p, \Delta_{0p}) = \\ = \langle \delta(\Delta_p - \mid E - E' \mid) \delta(\Delta_{0p} - \frac{U}{R^3}\frac{\Delta_0 \Delta_0'}{EE'}) \rangle. \quad (16)$$

The brackets denote two averaging, namely, thermal averaging and averaging over the distribution of parameters of original TLS. In addition, the integration over the distance R is implied. Evaluation of the pair distribution function Eq. (16) within the logarithmic accuracy [8, 33] reads

$$P^{(2)}(\Delta_p, \Delta_{0p}) \approx (PT)(PU)/\Delta_{0p}^2. \quad (17)$$

Remember that resonant pairs are only those pairs for which $\Delta_p \leq \Delta_{0p}$. Therefore each resonant pair has the single characteristic energy given by its flip-flop amplitude Δ_{0p}. Since resonant pairs are mainly formed by TLS with $\Delta \sim \Delta_0 \sim T$, the characteristic pair transition amplitude can be estimated as U/R^3, where $R \sim (U/\Delta_{0p})^{1/3}$ is the size of the pair.

Resonant pairs interact with each other. We will show following Refs. [8, 33] that this interaction inevitably leads to the energy delocalization induced by collective flip-flop transitions similar to that shown in Fig.3. It is convenient to separate the whole sequence of resonant pairs into the infinite set of strips $k = 1, 2, 3, ...$ These strips are defined in the space of characteristic energies of resonant pair. Each strip k is formed by resonant pairs having transition amplitudes within the range $(\Delta_{0p}(k)/2, \Delta_{0p}(k))$, with $\Delta_{0p}(k) = T/2^k$. One can estimate the concentration of RP N_k within the strip k making use of the distribution function Eq. (17)

$$N_k \approx (PT)(PU) = N_*. \quad (18)$$

It is important that the RP concentration in each strip k is completely independent of the value $\Delta_{0p}(k)$. Separating strips corresponding to all permissible values Δ_{0p}, one can cover completely the whole ensemble of resonant pairs. Within any strip the RP concentration is constant N_* Eq. (18). Therefore the average distance between RP within any strip does not depend on the kind of it. This distance is equal to $R_* \approx N_*^{-\frac{1}{3}}$. Since the interaction between RP is of the same origin as the interaction between TLS, it is given by the same expression U/R_*^3. Therefore, the typical energy of the interaction between RP within any strip is given by

$$U_* \approx U/R_*^3 \approx T\chi^2. \quad (19)$$

Since RP is a kind of TLS, one can introduce a concept of a flip-flop configuration for two RP shown in Fig. 3 and derive an expression for the transition amplitude $\Delta_0^{(4)}$ between the levels of this configuration. Because interacting pairs are both resonant this transition amplitude is given by their characteristic interaction Eq. (19). Consider the effect of this flip-flop interaction on RP belonging to different strips.

The strips with the energy smaller than U_* satisfy the conditions of the Anderson delocalization because the characteristic flip-flop transition amplitude U_* between RP is greater or equal to their energy disordering, determined by their characteristic energy Δ_{0k}. The elementary process of an energy hop between pairs is demonstrated in Fig.3. It is important that in order to transfer the energy, the simultaneous transition of all four TLS is required, while the transition of two TLS belonging to different resonant pairs (Fig.3C) is forbidden because of the energy mismatch of order of the thermal energy T. Thus RP excitations in the cluster formed by the strip with the energy U_* are delocalized. The relaxation rate of excitations within this strip is given by the inverse characteristic interaction

$$\tau_*^{-1} \sim U_* \approx T(PU)^2. \quad (20)$$

$$\tau_2^{-1} \approx U(R_*) \approx T\chi^2. \quad (21)$$

This dephasing rate decreases linearly with the temperature. Compared with the phonon-induced channel providing a T^2 -dependence (see Ref. [11]), the dephasing governed by the mechanism concerned predominates at sufficiently low temperatures.

Regardless of the relaxation mechanism, the spectral diffusion rate is defined as [9, 11]

$$\left\langle \left| \dot{E} \right| \right\rangle \sim \tau_2^{-2}. \quad (22)$$

On the other hand, in a system with the $1/R^3$ interaction this rate can be expressed as the ratio of the spectral diffusion amplitude χT and thermal TLS relaxation time τ_1 [9, 11]

$$\tau_2^{-2} = \chi T/\tau_1. \quad (23)$$

Using (21) and (23) one can estimate the relaxation rate for thermal TLS as [8]

$$\tau_1^{-1} = \chi^3 T. \quad (24)$$

5. Relaxation at finite temperature and strong field. Previously, we have shown that the $1/R^3$ interaction of excitations and the external alternating field can independently result in the delocalization and in the irreversible TLS dynamics in the regime of arbitrary strong disordering. Under conditions of the real experiment both the TLS interaction and the external alternating (measuring) field are significant. Below we consider the combined contribution of two those effects on the TLS relaxation.

Our analysis starts with the very low frequency of the external field, where the field can be treated as static. Then the frequency will substantially increase to its maximum value $\omega \sim T$. The case of larger frequencies $\omega > T$ is beyond the scope of this paper because of almost unavoidable heating. The results of our consideration are placed into the Table.

Frequency-dependent relaxation rate

Frequency range	τ_1^{-1}	τ_2^{-1}
$0 < \omega < (T\chi^2)^2/a$	$T\chi^3$	$T\chi^2$
$(T\chi^2)^2/a < \omega < a\chi^2$	$(a\omega)^{1/2}\chi$	$(a\omega T^2)^{1/4}\chi$
$a\chi^2 < \omega < T\chi$	$a\chi^2$	$(aT)^{1/2}\chi^{3/2}$
$T\chi < \omega < a$	$(a/\omega)T\chi^3$	$(a/\omega)^{1/2}T\chi^2$
$a < \omega < T$ or $a < T\chi$	$T\chi^3$	$T\chi^2$

Below we consider the case of a relatively large field amplitude $a > T\chi$ where the field effect is significant. Such a regime has been realised in metallic glasses [35] where the nonlinear behavior has been revealed. The condition $a > T\chi$ means that the fluctuation of a TLS energy induced by the external field exceeds the interaction-induced fluctuation. We also assume that the field amplitude is less than the thermal energy to avoid heating. Thus in our consideration below the field amplitude a satisfies the inequality

$$T\chi < a < T. \quad (25)$$

5.1. *Quasistatic field.* When the frequency of the external field ω is very small, the field is almost static and we can ignore it. Then the relaxation and dephasing rates are defined by the zero field limit Eqs. (21), (24), respectively. In this regime the fluctuations of a TLS energy are due to the self-consistent spectral diffusion alone. The spectral diffusion rate is given by Eq. (22). The rate of a TLS energy fluctuation induced by the external field is $a\omega$. The external field can be treated as static until the spectral diffusion rate exceeds the fluctuation rate due to the field $\tau_2^{-2} > a\omega$, which takes place at $\omega < (T\chi^2)^2/a$ (see the first row in the Table).

5.2. *Adiabatic relaxation induced by slow field.* At larger frequency

$$\omega > (T\chi^2)^2/a \quad (26)$$

the field cannot be ignored. It can stimulate transitions of TLS and irreversible relaxation due to energy level crossings of different TLS. At a large field amplitude Eq. (25) crossing between two TLS energy levels E_0 and E_j ($E_0 - E_j - a\cos\omega t = 0$) [36] is possible provided that

$$|E_0 - E_j| < a. \quad (27)$$

The average distance between two TLS satisfying Eq. (27) is given by $R_a \simeq (Pa)^{-1/3}$.

Consider a flip-flop pair of TLS with the close energies satisfying Eq. (27) and separated by the distance R_j. This pair undergoes level crossing during the field oscillation period ω^{-1}. If the pair transition amplitude is large $V^2 = (U/R_j^3)^2 > \dot{E} \simeq a\omega$, the *adiabatic relaxation* takes place. Then according to the Landau–Zener theory the excitation will transfer within a TLS pair with the almost *unity probability* if the size R_j of a pair is sufficiently small (see Eq. (11))

$$R_j < r_* \simeq \left(U/\sqrt{a\omega}\right)^{1/3}. \quad (28)$$

The *adiabatic* regime takes place when each thermal TLS has the large number of adiabatic neighbors, satisfying Eqs. (27), (28). This requires $r_* > R_a$, which leads to the frequency constraint from top (see Table, the second row)

$$\omega < a\chi^2 \quad (29)$$

For given thermal TLS the relaxation rate τ_1^{-1} can be estimated as a frequency of adiabatic level crossings given by the product of the field oscillation frequency ω and the number of adiabatic level crossing events during the single field oscillation. The latter number is given by the number of adiabatic neighbors χ_*, satisfying Eq. (27) and located within the sphere of the radius r_* around given TLS, $\chi_* \simeq Par_*^3 = (a/\omega)^{1/2}\chi \gg 1$

$$\tau_1^{-1} \simeq \chi_*\omega = (a\omega)^{1/2}\chi. \quad (30)$$

It is interesting that the relaxation rate (30) coincides with that of Eq. (15).

In the adiabatic regime we can estimate the dephasing rate using Eqs. (23), (30)

$$\tau_2^{-1} = (T\chi/\tau_1)^{1/2} = \left(a\omega T^2\right)^{1/4}\chi. \quad (31)$$

It is important that dephasing (31) is so fast that the phase coherency between the periodic events of level crossings occurring during the period ω^{-1} can be ignored. This is true in the whole adiabatic frequency domain (the second row of the Table) because the dephasing time is in fact less than the oscillation period (see Eqs. (25), (29), (31))

$$\omega\tau_2 = (\omega/(a\chi^2))^{3/4}(a/T)^{1/4}\chi^{1/2} < 1. \quad (32)$$

5.3. *Non-adiabatic regime at intermediate frequencies.* Consider the *nonadiabatic regime*, which takes place at higher frequencies

$$\omega > a\chi^2. \quad (33)$$

This condition is opposite to Eq. (29). In this regime the vast majority of thermal TLS undergo nonadiabatic level crossings (see Eq. (34) below).

In the adiabatic case we have dealt with the regime $\omega\tau_2 < 1$ (see Eq. (32)). In the nonadiabatic regime characterized by Eq. (33) one should distinguish between two cases $\omega\tau_2 < 1$ and $\omega\tau_2 > 1$ where the phase memory between two subsequent level crossings either exists or does not exist, respectively.

Consider the first case $\omega\tau_2 < 1$ valid at the border of adiabatic and non-adiabatic domains $\omega = a\chi^2$ (see the Table). Here dephasing is fast and phase correlations between periodic level crossings can be ignored. Most efficient flip-flop transitions occur between thermal TLS separated by the average distance R_a Eq. (14). In fact this distance is the characteristic separation of neighboring two-level systems with energy difference less than the field amplitude a, required for level crossing. On the other hand at larger distances a nonadiabatic transition probability decreases with the distance very fast as $V^2 \sim R^{-6}$. Therefore the most efficient energy transfer should occur between nearest neighbors. The relevant transition amplitude reads $V_a = U/R_a^3 \sim a\chi$. In the large frequency case Eq. (33) one has

$$V_a^2 < \dot{E} = a\omega \quad (34)$$

and transitions are *nonadiabatic*. According to the Landau–Zener theory, their probability per one energy level crossing, occurring a few times for the period ω^{-1}, is $W_a \simeq V_a^2/(a\omega) = (a/\omega)\chi^2$. This probability defines the TLS relaxation rate as the inverse average time between two successful transitions

$$\tau_1^{-1} = W_a\omega \simeq a\chi^2. \quad (35)$$

Then, using Eq. (23), one finds

$$\tau_2^{-1} = a^{1/2}T^{1/2}\chi^{3/2} \quad (36)$$

The above derivation remains valid until breaking the condition Eq. (32) at

$$\omega > a^{1/2}T^{1/2}\chi^{3/2}. \quad (37)$$

In the opposite case of $\omega\tau_2 > 1$, the field periodicity is significant so that the formalism of DE introduced in Sect. 3 (see also Ref. [31]) becomes applicable.

The further analysis depends on the relationship between DE energy splitting ω and the spectral diffusion amplitude $T\chi$ (see Eq. (25)). We start with the case of lower frequencies (see the Table)

$$\omega \le T\chi. \quad (38)$$

where all pairs of TLS with an energy difference less than the field amplitude a inevitably undergo DE level crossing during the spectral diffusion quasi-period τ_1. These crossing levels are coupled by the transition amplitude of dressed excitations Eq. (10).

In the regime of Eqs. (37), (38) the relaxation is induced by non-adiabatic level crossing caused by the spectral diffusion. On the other hand, the spectral diffusion is caused by the relaxation dynamics of TLS, so the process is self-consistent [8, 27, 33, 34].

Assume that there is an existing characteristic relaxation rate of thermal TLS τ_1^{-1}. Transitions of thermal TLS change other TLS energies giving rise to the spectral diffusion. The spectral diffusion leads to level crossing of DE, stimulating irreversible transitions with the output rate r_{out}. The self-consistent relaxation mechanism requires the input rate τ_1^{-1} to be equal to this output rate.

The rate of transitions induced by the spectral diffusion can be estimated as the number of transitions induced by level crossings during the quasi-period of the spectral diffusion τ_1 multiplied by the frequency of spectral diffusion cycles τ_1^{-1}. Energy level crossing for two DE happens when (cf. Eq. (8), Fig.2)

$$E_1 - E_2 = n\omega \quad (39)$$

(where n is an integer number). The total number of such crossings due to the spectral diffusion induced energy fluctuation $T\chi$ for the time τ_1 is given by

$$N_{\tau_1} \simeq (T\chi/\omega). \quad (40)$$

Multiple crossings for the same TLS pair give a logarithmic correction to Eq. (40) and can be neglected in the qualitative scaling approximation [8, 33, 34]. This is due to the specific behavior of the spectral diffusion induced by the $1/R^3$ interaction. For this spectral diffusion the characteristic energy fluctuation is directly proportional to the time [9, 11]. Therefore this is the anomalous diffusion process compared to the normal diffusive $t^{1/2}$ behavior of the displacement. For this super- diffusion case the probability of energy to return back to its initial value is not so large as in the case of the normal diffusion.

We consider nonadiabatic flip-flop transitions between only neighboring thermal DE separated by the distance R_a (see Eq. (14)), because the nonadiabatic transition probability to larger distances drops with the distance very fast (R^{-6}). The probability of the nonadiabatic transition during one level crossing for such a pair of thermal DE induced by the spectral diffusion with the rate $<|\dot{\varepsilon}|>\approx \tau_2^{-2}$ can be estimated using the Landau-Zener theory in the nonadiabatic limit. The transition probability can be found using the transition amplitude Eq. (10) for $R = R_a$

$$W_* = (V_a\tau_2)^2 < 1. \qquad (41)$$

The relaxation rate is expressed as

$$\tau_1^{-1} = r_{out} = N_{\tau_1}W_*/\tau_1 \simeq a\chi^2. \qquad (42)$$

One can show that if we take the input relaxation rate slower than Eq. (42), the output rate will be faster than the input one, and, if we take the input rate larger than Eq. (42) the output rate will be smaller than the input rate. Therefore the solution (42) is stable and the only possible. Thus, the *nonadiabatic single particle* relaxation described by Eqs. (35), (36) takes place within the whole frequency domain $a\chi^2 < \omega < T\chi$ (Table, the third row).

5.4. *High frequencies.* When the external field frequency ω exceeds the scale $T\chi$, the spectral diffusion does not necessarily lead to DE energy level crossing for closely located pairs, considered in Sect. 5.3. The single-particle delocalization in this case does not occur similarly to the case studied in Sect. 2 because the field frequency is large compared to that needed for the delocalization (see the Table, line 2). On the other hand, there exists a finite concentration of excited DE (excited TLS dressed by some certain number of the external field quanta, see Fig. 2) and many-body relaxation should be similar to that described in Sect. 4.

Following the approach of Sect. 4 one can introduce the concept of the resonant pair of DE, instead of RP (resonant pairs of TLS). A resonant pair of DE (RPDE) is defined as a pair of DE separated by the distance R_{12} with sublevel energies E_1 and E_2, which obeys the resonant condition for some integer $n < a/\omega$ (see Fig.2)

$$\Delta_{pn} = |E_1 - E_2 + n\omega| < T\,(R). \qquad (43)$$

Here the transition amplitude $T\,(R) \approx (U/R^3)\sqrt{\omega/a}$ is taken from Eq. (10). Like a resonant pair, one can treat RPDE as a new kind of two-level system. The parameter Δ_{pn} is the asymmetry energy for RPDE. Then, strictly following the derivation between Eq. (16) and Eq. (18) one can estimate the concentration of RPDE for any given value of transition amplitude $T\,(R)$. This concentration is given by the expression

$$N^* \approx T\chi^2\sqrt{\frac{a}{\omega}}. \qquad (44)$$

The concentration N^* is independent of the given parameter $T(R)$ just like the concentration of RP N_* defined by Eq. (18) does not depend on the parameter Δ_{0p}. The appearance of the factor $\sqrt{a/\omega}$ in Eq. (44) in comparison with Eq. (18) can be understood as follows. The concentration N^* compared with N_* acquires the factor $n_* = a/\omega$ since resonance condition (43) should be valid for at least one integer $|n| < n_*$ so that the number of possible distinguishable resonances for the single pair multiplies the resonance probability by the factor n_*. On the other hand, the resonance probability decreases by the factor $1/\sqrt{n_*}$ proportionally to the reduction in the transition amplitude Eq. (10). The overall effect is just the increase in the density of resonant pairs by the factor $\sqrt{a/\omega}$ (cf. Eq. (13)). Accordingly, the interaction between these resonant DE pairs enhances by the same factor and the new dephasing rate is given by this universal interaction

$$1/\tau_2 = T\chi^2(a/\omega)^{1/2}. \qquad (45)$$

Making use of Eq. (23) one can estimate the relaxation rate as

$$\tau_1^{-1} = \left(T\chi\tau_2^2\right)^{-1} = (a/\omega)T\chi^3. \qquad (46)$$

When the frequency exceeds the amplitude $a < \omega <$ $< T$, excitation dressing becomes negligibly small and we return to the linear regime (the last row of the Table). The regime when the frequency exceeds the temperature is realized in several echo measurements [2]. This regime can lead to heating and requires a special study.

6. Summary. We have described the self-relaxation rate of TLS at different temperatures, external field amplitudes, and frequencies. We found that the relaxation rate is either temperature independent (see the Table), or decreases with the temperature as T. The phonon stimulated relaxation $\tau_1^{-1} \propto T^3$ (see Ref. [9]) is much slower in the low temperature limit and the self-relaxation described above should dominate when $T \to 0$.

The suggested theory predicts the parametric dependences of relaxation rates in various regimes. In contrast to the theory for the phonon-stimulated relaxation (see the review of Hunklinger and Raychaudchary [9] and references therein) we are not able to determine the numerical coefficients for each rate process shown in the Table. This missed factor can be of order of unity, but it can also be much greater or much smaller than unity, like 0.01 or 10^2. The example of the latter situation is given by the tunneling rate involving two-phonon processes, examined by Kagan and Maksimov [37] for the quantum diffusion problem, where they found the large numerical prefactor of 1000 due to the large factorial factor involved.

We do also expect large numerical prefactors for our expressions. There are several reasons to have them. First, each factor χ possibly involves the spherical integration factor 4π or $4\pi/3$ in addition to PU factor. In this paper we have included the factor 4π into the definition of our factor χ Eq. (4) contrary to the previous work. This is done because this factor naturally appears in the localization criterion (3) and correspondingly it can enter the rate expressions of the Table. Thus the parameter of interaction weakness $PU \sim 10^{-4} - 10^{-3}$ can be increased by the order of magnitude. Accordingly, the "linear" expression for the relaxation rate Eq. (24) agrees better with the results of the systematic internal friction measurements performed by Classen, Burkert, Enss, and Hunklinger [6]. We believe that the factor $\chi = 4\pi PU$ reflects the absolute values of relaxation rates better than the smaller factor $\chi = PU$ itself. The accurate calculations of the numerical factors are beyond the scope of our qualitative study. The alternative explanation for a quantitative disagreement [6] can be based on the strong non-linearity. For instance, the extra-factor a/ω in the relaxation rate (see Eq. (46)) can account for the difference of theory and experiment as well. The comparison of our predictions with experiment can still be performed using the experimental data for different glasses, with different values of the parameter χ. Note that the preliminary numerical analysis of the Floquet state delocalization, described in Sect. 3, in the equivalent one-dimensional model supports our expectations of the large numerical prefactor, exceeding unity by at least one order of magnitude.

Another possible problem of the direct application of our theory to the experiment is that the interaction-induced relaxation leads to the equilibration within the TLS subsystem rather than for the whole system. This is similar to spin-spin and spin-lattice relaxations in the NMR problem. Then in order to describe the heat balance between TLS and phonons, one should introduce the separate TLS and phonon temperatures and perform the thermal balance analysis for the whole system. This study is beyond the scope of this paper.

The crossover temperature between two regimes depends on the external field parameters a and ω. It usually reduces with decreasing the field amplitude and can also decrease with decreasing the field frequency (see the Table). This knowledge helps us to understand the absence of the interaction-stimulated relaxation reported by Pohl and coworkers in Ref. [38]. In this work, the strain field amplitude was made extremely small $\varepsilon \leq 10^{-8}$, while the frequency $\omega \sim 0.5\,\mathrm{MHz}$ is higher than in other group studies [4–6] and belongs to the range described by the fourth column of the Table, when the relaxation rate decreases with increasing ω.

Our results agree with the recent low-temperature measurements of the dielectric constant by Ladieu and coworkers [24, 39], which can be interpreted assuming that the TLS relaxation rate becomes temperature independent. This shows the dependence on the external field amplitude $\sqrt{a}$ (the second row of the Table). Another very interesting discovery of Ladieu and coworkers includes the sensitivity of the TLS relaxation rate to the sample thickness. The suggested theory is essentially three dimensional. If the thickness of the sample will be less than the typical distance between resonant pairs (around few tens of nanometers) then the irreversible relaxation will remarkably slow down. This agrees with the observations of Ref. [24, 39]. More accurate theoretical analysis of the data is necessary for the careful interpretation.

Our predictions can be directly verified using the non-equilibrium measurement technique developed by Osheroff and coworkers [5, 40], which is based on the analysis of the system response to the large sweep of the external electric field E_{DC} taking different times τ_s. Our results for the field-stimulated relaxation are valid in this case if one takes $a \sim E_{\mathrm{DC}}\mu_{\mathrm{TLS}}$ (where $\mu_{\mathrm{TLS}} \sim 1\mathrm{D}$ is the typical TLS dipole moment) and $\omega \simeq 1/\tau_s$. Since the maximum possible amplitude a is very high $a \geq 0.1\,\mathrm{K}$, while the minimum "frequency" is as low as $\omega \sim 1\,\mathrm{s}^{-1}$, all the regimes described in the Table can be attained and analyzed.

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