Interaction-Driven Relaxation of Two-Level Systems in Glasses

Doru Bodea[1] and Alois Würger[2]

[1]*Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer Straße 38, 01187 Dresden, Germany*
[2]*CPMOH, CNRS-Université Bordeaux 1, 351 cours de la Libération, 33405 Talence, France*

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We study how the elastic interaction of two-level systems contributes to their relaxational motion. Evaluating the Mori-Zwanzig memory function in terms of a perturbation series in powers of the couplings J_{ij}, we find a null result at second order, which means that interacting pairs of two-level systems do not give rise to relaxation, yet a finite relaxation rate does occur in fourth order; i.e., a diffusive band is formed by resonant triples. Our results provide a simple explanation for several puzzling experimental observations. Regarding the temperature dependence of the sound velocity $\delta v \sim \ln T$ in the kHz range, we find that its slope below and above the maximum takes opposite signs but the same absolute value, in agreement with the measured ratio $1:-1$. Below the relaxation plateau, the internal friction is shown to vary linearly with T, in agreement with experiment.

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Sound and microwave propagation in amorphous solids at temperatures below a few Kelvin show a rich and unusual behavior [1–4]. At very low T, the sound velocity increases logarithmically with temperature, reaches a maximum at about hundred millikelvin, and then decreases as $\ln T$, whereas the sound attenuation, or internal friction Q^{-1}, increases with a power law T^{α} and then takes a constant value over more than one decade in temperature; the edge of the absorption plateau depends on the external frequency ω. A similar behavior has been reported for real and imaginary parts of the dielectric function $\varepsilon(\omega)$.

These temperature and frequency dependencies are to a large extent determined by the Debye relaxation of quantum two-level systems (TLS) that are present in almost all amorphous solids [1–5]. The interaction of the traveling sound or microwave with the TLS induces a deviation from the thermal equilibrium occupation of the tunneling states. The subsequent relaxation of this perturbation has been studied in terms of phonon absorption and emission, with a rate γ_{ph} that is proportional to T^3 [5]. One thus finds that the maximum of the sound velocity and the lower edge of the absorption plateau occur at the crossover temperature T_ω where $\omega \sim \gamma_{\mathrm{ph}}$. Although at first sight rather seducing, this picture fails to account for the experimental observations on a more quantitative level [6–11]. First, from the one-phonon process one expects for the slopes of the permittivity and the sound velocity below and above T_ω, the ratio $2:-1$, whereas experiment rather shows a ratio of $1:-1$ [7,8]. Second, the internal friction is predicted to vary at low temperature as $Q^{-1} \sim T^3$, whereas a much weaker, close to linear, dependence has been measured [8,9].

As another striking feature, the permittivity of several dielectric and insulating glasses has been shown to depend on an applied magnetic field [12–14]. After several theoretical attempts based on the Ahanorov-Bohm phase of the atoms involved in tunnel systems [15–17], coherent echoes gave evidence that the magnetic-field effects are related to nuclear spins [18]; their coupling to TLS is mediated by nuclear quadrupole moments [19]. Yet in another twist of the story, it was shown that neither the resonant susceptibility nor phonon-driven relaxation account for the magnetic-field dependence of the dielectric function [20], the origin of which seems to be more complex than that of the echoes.

In this Letter we derive a novel relaxation mechanism that is based on triples of interacting TLS, and that resolves the above discrepancies concerning sound propagation. It has been known for a while that the spectral diffusion observed in saturation and echo experiments is due to the longitudinal coupling of TLS [21–23]. More recently, out-of-equilibrium states with very slow kinetics have been observed after a dc electric field sweep [24–26]; there is conclusive evidence that the resulting ''dipole gap'' and the dependencies on sweep and waiting times arise from slowly tunneling TLS with interaction [24,27]. Yet note that these works do not establish an interaction-driven relaxation mechanism.

In terms of Pauli matrices, the two-level Hamiltonian reads $\frac{1}{2}\Delta_0\sigma_x + \frac{1}{2}\Delta\sigma_z$, where Δ is the asymmetry energy between the two positions $\sigma_z = \pm 1$ and quantum tunneling with amplitude Δ_0 is accounted for by the off-diagonal operator σ_x, resulting in the energy splitting $E = \sqrt{\Delta_0^2 + \Delta^2}$. Transverse coupling to a heat bath gives rise to the relaxational part of the dynamical susceptibility [5]

$$\chi_{\mathrm{rel}}(\omega) = \frac{n_0}{k_B T}\left\langle \frac{1-u^2}{\cosh(E/2k_BT)^2}\,\frac{iu^2\gamma}{\omega + iu^2\gamma}\right\rangle, \qquad (1)$$

where angular brackets $\langle ... \rangle$ indicate the ensemble average with respect to the parameter distribution $P(\Delta_0, \Delta) = P_0/\Delta_0$, with n_0 the number of TLS per unit volume. We have introduced the shorthand notation $u = \Delta_0/E$ and $\bar{u} = \Delta/E$, and split a factor u^2 from the rate. After replacing, in

the rate γ, the splitting E by the thermal energy k_BT, we integrate over E, u and find

$$\chi_{\rm rel}(\omega) = P_0\left[\frac{1}{2}\ln\left(1+\frac{\gamma^2}{\omega^2}\right) + i\arctan\frac{\gamma}{\omega}\right]. \quad (2)$$

In the limiting cases $\gamma \ll \omega$ and $\gamma \gg \omega$, one readily obtains the known results for sound propagation, after inserting the phonon-driven rate of thermal TLS, $\gamma_{\rm ph} = (3/2\pi)(g^2/\varrho v^5\hbar^4)(k_BT)^3$, with the mass density ϱ and the sound velocity v of the host, and the TLS deformation potential g [5,6].

Here we study relaxation due to the elastic interaction of two-level systems. The tunneling motion between the two wells at $\sigma_z = \pm 1$ gives rise to an elastic deformation field and an effective interaction $\frac{1}{2}J_{ij}\sigma_z^i\sigma_z^j$, where

$$J_{ij} = \frac{g^2}{\varrho v^2 r_{ij}^3} = \frac{U_0}{r_{ij}^3} \quad (3)$$

varies with the inverse cube of the distance r_{ij} of TLS i and j, the parameter U_0 being of the order eV(nm)^3.

Relaxation phenomena and the corresponding rates are best dealt with in the framework of the Mori-Zwanzig projection method and a perturbative evaluation of the memory function [28]. Here we use the formulation given in Eqs. (2.12–2.36) of Ref. [29]; we merely have to replace the phonon coupling potential with the TLS pair interaction $f = \frac{1}{2}\sum_{i,j}J_{ij}\sigma_z^i\sigma_z^j$. The resulting rate $u^2\gamma_{\rm Tr} = \cosh^2(E/2k_BT)M''(0)$ is given by the zero-frequency limit of the spectrum of the longitudinal memory function $M(t)$.

Truncating at second order in the perturbation f, one finds for a given TLS i

$$M_2(t) = \sum_{\{\pm\}}\sum_j J_{ij}^2 G_i^{\pm}(t)G_j^{\pm}(t), \quad (4)$$

with the uncoupled one-particle correlation functions

$$G_i^{\pm}(t) = u_i^2\frac{\cosh[E_i/(2k_BT) \pm itE_i/\hbar]}{\cosh[E_i/2k_BT]}.$$

Yet a simple estimate shows that only a few percent of TLS have a resonant neighbor with $|E_i - E_j| \le J_{ij}$. Inserting this constraint and integrating the distribution $\mathcal{P}(J_{ij}) \propto J_{ij}^{-2}$ over the range where the interaction is effective, $\hbar\gamma_{\rm ph} \le J_{ij} \le k_BT$, one readily finds the probability of resonant pairs [30],

$$W_2 = \frac{8\pi}{3}P_0U_0\ln\frac{k_BT}{\hbar\gamma_{\rm ph}}. \quad (5)$$

For typical parameters $\gamma_{\rm ph} \sim 10^7$ sec^{-1} (at about 1 K) and $P_0U_0 \sim 10^{-3}$, one obtains the numerical value $W_2 \approx 3\%$ [30–32].

Thus the second-order expression $M_2(t)$ does not decay. Its spectral function $M_2''(\omega)$ is not a smooth function of frequency but shows sharp resonances at $\omega = \pm(E_i - E_j)/\hbar$ and, in particular, $M_2''(\omega = 0)$ does not define a rate. In physical terms this means that, in the framework of the pair model, the elastic interaction J_{ij} does not give rise to a band of quasidiffusive excitations, and that there is no interaction-driven relaxation rate. We stress that this negative result is not modified when taking spectral diffusion into account [23].

Now we show that relaxation does arise in the next order of the perturbation series for the memory function, involving three-spin processes. Pushing the perturbation expansion for $M(t)$ to fourth order we have

$$M_4(t) = \sum_{\{\pm\}}\sum_{j,k} J_{ij}^2J_{ik}^2G_i^{\pm}(t)\int_0^t d\tau\int_0^\tau d\tau' \times [G_j^{\pm}(t)G_k^{\pm}(\tau-\tau') + G_j^{\pm}(t-\tau')G_k^{\pm}(\tau)], \quad (6)$$

with the shorthand notation $J_k^2 = \bar{u}_i^2J_{ik}^2 + \bar{u}_j^2J_{jk}^2$. The two terms of the integrand correspond to the rainbow and crossing diagrams of Fig. 1, the latter of which contributes little and thus will be discarded.

We estimate the number of resonant triple configurations W_3. For a given TLS i, the number N_2 of thermal neighbors j within the maximum interaction radius $r_{\rm max} = (U_0/\hbar\gamma_{\rm ph})^{1/3}$ is given by $N_2 = \frac{4}{3}\pi r_{\rm max}^3P_0k_BT$; i.e., i, j satisfy the condition $|J_{ij}| \ge \hbar\gamma_{\rm ph}$. A resonant triple occurs if the third TLS k satisfies $|E_i \mp E_j \mp E_k| \le J_k$; this second condition is met with the probability W_2. Thus we have $W_3 = N_2W_2$ and, after inserting $r_{\rm max}$,

$$W_3 = \frac{4\pi}{3}\frac{k_BT}{\hbar\gamma_{\rm ph}}P_0U_0W_2. \quad (7)$$

With the numbers given below Eq. (5) one has $W_3 > (K/T)^2$. This means that at temperatures below 1 K, a given TLS i is involved in several resonant configurations ijk, $ij'k'$,... where the neighbors j, k, j', k' participate, in turn, in more distant triples and thus form a diffusive band.

In other words, all thermal TLS continually exchange energy by three-body resonant processes. At temperatures below 100 mK, one has $W_3 \sim 10^3$; then the memory spectrum $M_4''(\omega)$ is a smooth function and its zero-frequency value defines the relaxation rate $u_i^2\gamma_{\rm tr} = c_i^2M_4''(0)$.

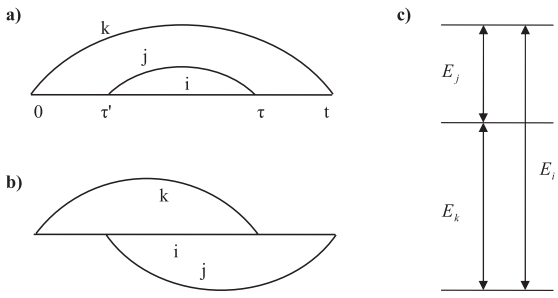

FIG. 1. Fourth-order diagrams contributing to the relaxation pole of the memory function $M_4(t)$. Only the "rainbow" diagram (a) has been retained, the contribution of the crossing diagram (b) is significantly smaller. Solid lines indicate TLS propagators $G^{\pm}$. One of the resonant configurations is shown in (c).

Evaluating the convolution integrals in $M_4(t)$ we find

$$\gamma_{\rm tr} = \pi \sum_{j,k} u_j^2 u_k^2 \frac{J_{ij}^2 J_k^2}{2E_k^2} \frac{c_i}{c_j c_k} \delta(E_{ijk}), \tag{8}$$

where the factor $\delta(E_{ijk})$ assures the resonance condition, i.e., that a combination of the three energies E_i, E_j, E_k gives zero, and where the temperature factors $c_i = \cosh(E_i/2k_BT)$ provide an effective cutoff at $E_i \sim k_BT$ and thus restrict the relaxation process to thermal TLS. An estimation of the average rate is obtained by decoupling the double sum, $\gamma_{\rm tr} = \pi\lambda J_0/\hbar$, with the quantities $J_0 = \sum_j \langle u_j^2 J_{ij}^2 \delta(E_{ijk})\rangle$ and $\lambda = \sum_k \langle u_k^2 J_k^2/E_k^2\rangle$. The minimum value of relevant couplings $J_0 = U_0/r_0^3$ is determined by the condition that there is at least one resonant triple within a sphere of radius r_0, i.e., $1 = \frac{4}{3}\pi r_0^3 W_2 P_0 k_B T$. Thus one has $J_0 = \frac{4}{3}\pi W_2 P_0 U_0 k_B T$. The parameter λ is obtained by inserting $\sum_k \langle ... \rangle = \frac{4}{3}\pi P_0 U_0 \int dE \int dJ J^{-2}(...)$, with integration boundaries given by the inequalities $\hbar\gamma_{\rm ph} \le J_{ik} \le E_k \le k_BT$, resulting in $\lambda = W_2$. Whence the rate

$$\gamma_{\rm tr} = \frac{2\pi^2}{3} W_2^2 P_0 U_0 \frac{k_BT}{\hbar}. \tag{9}$$

With typical parameters $W_2 \sim 3\%$ and $P_0U_0 \sim 10^{-3}$ we obtain the numerical value $\gamma_{\rm tr} \sim 10^6\ (T/{\rm K})\,{\rm sec}^{-1}$, which is not unreasonable in view of the minimum interaction $J_0/\hbar \sim 10^7\ (T/{\rm K})\,{\rm sec}^{-1}$. Beyond the mean-field treatment of the elastic couplings, one would expect a broad relaxation spectrum about the mean value $\gamma_{\rm tr}$.

In Figs. 2 and 3 we plot real and imaginary parts of the complex susceptibility $\chi = \chi' + i\chi''$, with the rate $\gamma = \gamma_{\rm ph} + \gamma_{\rm tr}$ and the values

$$\gamma_{\rm tr} = 10^6\ (T/{\rm K})\,{\rm sec}^{-1},$$
$$\gamma_{\rm ph} = 3 \times 10^7\ (T/{\rm K})^3\,{\rm sec}^{-1}.$$

In principle $\gamma_{\rm tr}$ should be put to zero at temperatures above 1 K; yet in this range one has $\gamma_{\rm tr} \ll \gamma_{\rm ph}$ and the interaction-driven rate is irrelevant in any case. Figure 2 shows for two different frequencies $\omega = 10^3$ and 10^4 sec^{-1} the real part $\chi'(\omega)$, to which we have added the "resonant" contribution $\chi'_{\rm res}(\omega) = -2P_0 \ln T$; one easily distinguishes three domains with a logarithmic temperature dependence $\chi' = \xi P_0 \ln T$ but different prefactors ξ.

At very low temperatures $T < T_\omega$, both rates are smaller than the external-field frequency $\gamma < \omega$; then the relaxation contribution is negligible, and the slope is given by the resonant contribution only, $\xi = -2$. At high temperatures $T > T_*$, where $\omega < \gamma_{\rm tr} < \gamma_{\rm ph}$, relaxation is dominated by the phonon-driven mechanism; with $\chi'_{\rm rel}(\omega) = 3P_0 \ln T$ one readily obtains the slope $\xi = 1$. These two cases are well known [5,6]; a novel feature occurs in the intermediate range $T_\omega < T < T_*$, where the interaction-driven relaxation mechanism is dominant, satisfying $\omega < \gamma_{\rm tr}$ and $\gamma_{\rm ph} < \gamma_{\rm tr}$. Because of the linear temperature dependence

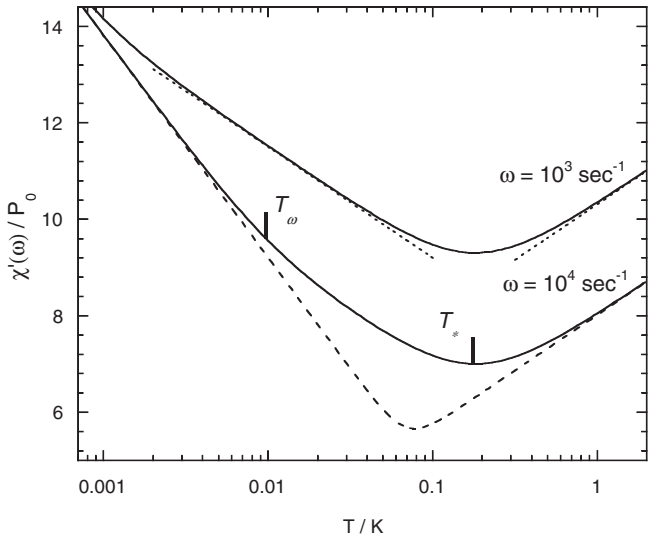

FIG. 2. Real part of the complex susceptibility as a function of temperature, for $\omega = 10^3$ and 10^4 sec^{-1}. The slopes $\xi = \pm 1$ are indicated by the dotted lines. The dashed curve is calculated with the phonon rate $\gamma_{\rm ph}$ at $\omega = 10^4$ sec^{-1} and shows the slope ratio 2: − 1.

of the rate $\gamma_{\rm tr} \propto T$, the relaxation susceptibility varies as $\chi'_{\rm rel} = P_0 \ln T$ resulting in the slope parameter $\xi = -1$. These different laws are summarized as

$$\chi' = \begin{cases} -2P_0 \ln T & (T < T_\omega) \\ -P_0 \ln T & (T_\omega < T < T_*) \\ P_0 \ln T & (T_* < T) \end{cases}. \tag{10}$$

The minimum of χ' occurs at $\gamma_{\rm tr} = \gamma_{\rm ph}$, i.e., at the temperature T_* that does not depend on frequency, whereas

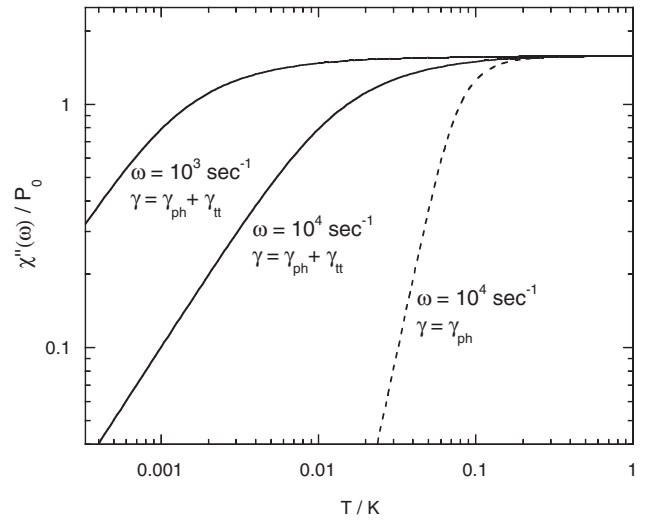

FIG. 3. Imaginary part of the complex susceptibility as a function of temperature, for $\omega = 10^3$ and 10^4 sec^{-1}. The plateau value is $\pi/2$. At low temperatures, relaxation of TLS triples with rate $\gamma_{\rm tt}$ results in the linear law $\chi''(\omega) \propto T$. The dashed curve indicates pure phonon relaxation with the rate $\gamma_{\rm ph} \propto T^3$, leading to $\chi''(\omega) \propto T^3$.

the second crossover at $\gamma_{tr} = \omega$ results in the law $T_\omega \propto \omega$. The above discussion applies to moderate external-field frequencies of the order of kHz. A different behavior is expected in the MHz range, where the minimum of χ' occurs at $\gamma_{ph} = \omega$, and the only crossover temperature varies with frequency as $T_\omega \propto \omega^{1/3}$.

Regarding the imaginary part, one finds two different laws at frequencies in the kHz range ($T_\omega < T_*$),

$$\chi''_{rel} = \begin{cases} P_0\gamma_{tr}/\omega \propto T & (T < T_\omega) \\ (\pi/2)P_0 & (T_\omega < T) \end{cases}, \tag{11}$$

whereas at high frequencies ($T_* < T_\omega$), there is an intermediate phonon-driven relaxation regime,

$$\chi''_{rel} = \begin{cases} P_0\gamma_{tr}/\omega \propto T & (T < T_*) \\ P_0\gamma_{ph}/\omega \propto T^3 & (T_* < T < T_\omega) \\ (\pi/2)P_0 & (T_\omega < T) \end{cases}. \tag{12}$$

The resonant contribution $\chi''_{res} = \pi P_0 \tanh(\hbar\omega/2k_BT)$ is insignificant in this frequency range.

These results account for several experimental observations that remained without explanation so far. There has been a longstanding controversy concerning the slopes of the logarithmic temperature dependence of both sound velocity $v = v_0 - \frac{1}{2}(g^2/\varrho v_0^2)\chi'(\omega)$ and dielectric constant $\varepsilon' = \varepsilon'_0 + \frac{1}{2}p^2\chi'(\omega)$. In various experiments it was found that the ratio of the slopes below and above the minimum of χ' is close to $1:-1$, whereas from phonon-driven relaxation one expects $2:-1$ [5,6]. This discrepancy is resolved by the interaction-driven relaxation that leads to the ratio $1:-1$; cf. Eq. (10). The fact that the low-temperature slope is not always well defined, could be simply due to the crossover at T_ω.

The imaginary part of the susceptibility describes microwave absorption $\varepsilon''(\omega) = p^2\chi''(\omega)$ and sound attenuation (or internal friction) $Q^{-1} = (g^2/\varrho v_0^2)\chi''(\omega)$. At temperatures below T_ω, pure phonon relaxation gives $Q^{-1} \propto T^3$. Yet experimental studies in the kHz range show a much weaker dependence [8–11], which is close to the linear law $Q^{-1} \propto T$ that arises from the interaction-driven relaxation mechanism in Eq. (11). Classen *et al.* measured the exponent α of the power law $Q^{-1} \propto T^\alpha$ below 30 mK as a function of the external-field frequency; below 2 kHz they found the constant $\alpha = 1$, whereas at higher frequency the exponent strongly increases and reaches $\alpha > 2$ at 20 kHz [9]. This experimental finding agrees with Eqs. (11) and (12), that predict $\alpha = 1$ at low frequency ($T_\omega < T_*$) and an intermediate value $1 < \alpha < 3$ at higher frequencies where $T_* < T_\omega$.

In summary, we have shown that resonant triples of interacting TLS give rise to a novel relaxation mechanism. Its rate γ_{tr} varies linearly with temperature and dominates the phonon rate $\gamma_{ph} \propto T^3$ below a few hundred mK, thus providing a simple explanation for the observed slope ratio $1:-1$ of the logarithmic law for the sound velocity and the linear temperature dependence of the internal friction Q^{-1}. The magnetic-field dependence of the dielectric function and the thermal conductivity will be discussed elsewhere.

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