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Low Temperature Properties of Amorphous Solids Induced by the Nuclear Quadrupole Interaction

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It is investigated how the nuclear degrees of freedom of the tunneling system (TS) inherent in an amorphous solid influence its dielectric properties. The nuclear quadrupole electric moment of the TS interacting with the crystal field determines the tunneling TS features. At sufficiently high temperatures, the TS is described by the standard tunneling model (STM). At sufficiently low temperatures, this interaction breaks down the coherence of the tunneling. This effect is responsible for the anomalous low-temperature behavior of the sound velocity and low-frequency dielectric permittivity. In addition, the effect explains the anomalous high sensitivity of the dielectric permittivity to the external magnetic field. If the magnetic field is strong enough, the tunneling becomes coherent and the STM can be applied to describe the TS. The approach developed explains the temperature and magnetic field dependence of the real part in the dielectric permittivity, revealed recently in an experiment. We predict an anomalous temperature and magnetic field dependence of the ultrasonic adsorption and thermal conductivity, and in particular, ultrasound absorption and thermal conductivity of glasses below 10 mK.

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I. INTRODUCTION

The standard phenomenological tunneling model (STM) successfully explains many anomalous thermal, acoustic, and dielectric properties of glasses below 1 K [1]. In particular, this is the linear and quadratic temperature dependence on the heat capacity and thermal conductivity, respectively [2–4]. These anomalies are attributed to special low-energy excitations inherent almost in all amorphous solids and disordered crystals [2, 3]. In the STM, these excitations are described by the non-interacting two-level tunneling systems (TLSs) [1–3]. The TLS can approximately be treated as an effective particle (which may contain many atoms) moving in a double-well potential (DWP). Any deviations from the STM predictions have been usually reduced to the TLS interaction [4–7].

The STM predicts a logarithmic temperature dependence of the resonant contribution to sound velocity$\delta v'/v \sim \ln T$. This dependence is known to provide us one of the main evidence for the TLS existence in glasses [2, 8]. The similar dependence should take place for the resonant contribution to dielectric permittivity $\delta\varepsilon'/\varepsilon \sim \ln T$[3, 9].

However, it is experimentally established that the logarithmic dependence for the permittivity crosses over to a temperature-independent one at a few mK and takes a constant value at lowest temperature. Sometimes, the constant is called a dielectric saturation [10, 11]. Similarly, below 8mK, the logarithmic dependence for the sound velocity breaks as well [12].

A decade ago, Strehlow et al. established for multi-component glasses that the application of the external magnetic field restores the logarithmic dependence predicted by the STM and thus, the dielectric saturation disappears [13]. Further investigations revealed an anomalous high sensitivity of dielectric properties in multi-component glasses to magnetic field below 50 mK [14, 15].

To explain such remarkable behavior of glasses in the magnetic field, S. Ketteman et al. have generalized the STM, assuming that a charged tunneling particle moves in the Mexican hat-like potential rather than in the one-dimensional DWP [16]. The magnetic field effect is understood qualitatively, but an agreement with experiment requires the coherent behavior of 10^5TLS. This looks unlikely. The similar model is also employed in [17] to treat qualitatively the magnetic field dependence of two-pulse echo in glasses at low temperatures [18].

An alternative, more plausible, model describing the behavior of glasses in the magnetic field was proposed by Würger, Fleischmann and Enss [19]. They considered a direct coupling between the nuclear spins of tunneling entities and the applied magnetic field. It is important to underline that the model [19] also requires that the tunneling atoms possess a nuclear quadrupole electric moment interacting with the crystal field. A key argument in favor of this model is an isotope effect in the polarization echo experiment [20]. In this experiment, the magnetic field affects amorphous glycerol when hydrogen, which has no quadrupole moment, is substituted with deuterium, which has non-zero quadrupole moment. Thereupon, one may conclude that the quadrupole electrical moment of the tunneling entity is a key attribute responsible for the effect.

To our knowledge, there are a few theoretical studies on the nuclear quadrupole effect in glassy dielectric properties [21–26]. In [21]; the authors attempted to apply their theory to the experiment performed at temperatures $T > 5mK$. In [21] the quadrupole energy b (energy acquired by a nuclear quadrupole in the crystal field) and the Zeeman energy m (energy of a nuclear spin in the magnetic field) are supposed to be smaller than 0.5 mK. The condition $b, m << T$ allowed them to use the perturbation approach. The correction to the permittivity induced by the magnetic field is found to be $\delta\varepsilon'/\varepsilon' \sim 1/T^4$ [21]. This is too small as compared with the dependence $\delta\varepsilon'/\varepsilon' \sim 1/T$ obtained in experiment [21, 27].

A discrepancy between two theoretical results is due to the following reasons. In [21, 26], the authors assume that the tunneling systems with parameters $b, m \ll T$ contribute to the permittivity if $E \approx T$ for these systems. Here E is the TLS energy without the quadrupole and Zeeman terms. However, it is shown in [25] that the tunneling systems have $E \approx m \approx b \ll T$.

One should also pay a special attention to the following circumstance. In [13, 15], the permittivity was measured as a response to the external electromagnetic field with the frequencies $\nu > 100\,Hz$. It was clearly explained in Refs. [13, 15, 23–25] that the resonant part of the permittivity alone is measured in the experiments made at the frequencies >100 Hz. It is important that the relaxation part of the response does not contribute to the permittivity. This is because the relaxation rate is too small and the population of the tunneling system is conserved during the oscillation period of the external field. At the same time, the static permittivity, which comprises both the resonant and relaxation contributions, is calculated in [21, 26]. Thus, in our opinion, these results seem have no direct relation to the experiment.

The case $T \leq m, b$was investigated in Refs. [23, 24]. It was established that the nuclear quadrupole interaction breaks down the coherent tunneling at the temperatures $T \leq 5\, mK$. This effect is shown to be responsible for the dielectric saturation. Also, it is found in these papers that the application of the sufficiently strong external magnetic field restores the coherent tunneling. Thus, if $m \gg b$, the recovery of the $\ln T$ dependence inherent in the STM should take place.

The acoustic and dielectric properties in glasses are known to have a number of common features [1–4]. In particular, the STM predicts that the correction to the sound velocity induced by the resonant TLS-phonon interaction behaves as $\delta v/v \sim \ln T$[28, 29]. It was experimentally found in [29] that the logarithmic temperature dependence is violated. Instead, the saturation effect is observed.

It follows from the above that the nuclear quadrupole interaction below $50\, mK$ (and in particular, in the mK region) seems to govern the properties of amorphous solids. In particular, the real part of the permittivity ε' is strongly affected by this interaction. On the other hand, due to the Kramers-Kronig relation, this influence should be reflected in the imaginary part of the permittivityε". For this reason, it is important to investigate how ε" is subjected to the quadrupole interaction.

Due to similarity between the acoustic and dielectric glass properties mentioned above, we investigate here how the nuclear quadrupoles influence the ultrasonic absorption. In acoustics, the parameter ε" determines the sound absorption. At low temperatures, this is most effective relaxation channel responsible for the thermal conductivity. For this reason, in this manuscript, we will investigate the influence of both the quadrupole interaction and external magnetic field upon the thermal conductivity of amorphous solids below 5 mK. Note that the dielectric absorption in the microwave region should manifest the similar features.

II. TUNNELING MODEL AND QUADRUPOLE INTERCATION

A TLS may be described by the standard pseudospin 1/2 Hamiltonian [1, 2]

$$h_{TLS} = -\Delta_0 \cdot s^x - \Delta \cdot s^z. \tag{1}$$

Here Δ_0 is a tunneling splitting coupling the two energy minima in a double-well potential, and Δ is the level asymmetry. Within the STM, the tunneling parameters obey the universal distribution

$$dP\left(\Delta, \Delta_0\right) = \frac{P}{\Delta_0} d\Delta d\Delta_0, \tag{2}$$

where P is a constant.

Let tunneling particle have its own degree of freedom associated with its nuclear spin$I$. If the spin$I \geq 1$, the particle can carry an electric quadrupole moment, which interacts with the crystal field. In the simplest approximation, the electric field gradient (EFG) is assumed to have an axial symmetry. This assumption singles out the directions $\mathbf{u}_l$and $\mathbf{u}_r$in the left and the right wells of the DWP [19, 22]. In this case, the quadrupole interaction in each well can be described as

$$\hat{H}_{r(l)} = b\left[\left(\hat{I}^{\mathbf{u}_{r(l)}}\right)^2 - I\left(I+1\right)\right]. \tag{3}$$

Here b is a quadrupole interaction constant, while $\hat{I}^{\mathbf{u}_l}$and $\hat{I}^{\mathbf{u}_r}$ are the corresponding nuclear spin projection operators.

It is argued in [23, 24] that a tunneling entity can involve several nuclei carrying quadrupole electrical moment. This assumption looks very probable since up to 200 atoms can be involved into the tunneling in glasses [30]. Let the number of identical nuclear quadrupoles affiliated with tunneling be$n$. Then, Hamiltonian acquires an extra term

$$\hat{h}_b = n\left(\frac{\hat{H}_r + \hat{H}_l}{2} + \frac{\hat{H}_r - \hat{H}_l}{2}s^z\right). \tag{4}$$

At sufficiently low temperatures, only the ground states in the left and the right well should be taken into account. Because of the strong disorder in glasses the EFG has a low symmetry, and therefore, any eigenstate is nondegenerate. In particular, this concerns the ground states. A nuclear component of the total eigenfunction of the tunneling system in each well of the DWP is the eigenfunction of either operator $\hat{I}^{\mathbf{u}_l}$or operator$\hat{I}^{\mathbf{u}_r}$. Since these operators do not commute, the mismatch between the ground states holds for. Let $\eta < 1$ be the overlap integral between the ground states of one quadrupole in the left and the right wells. This parameter depends on the angle between the directions $\mathbf{u}_l$ and $\mathbf{u}_r$ and on the value of nuclear spin$I$. In the case of n quadrupoles, the effective overlap integral is of the order of $\eta_* = \eta^n$.

It is shown in [23, 24] that the effect of nuclear quadrupole breaks down the coherent tunneling at the temperatures $T \leq nb$. This effect results in a strong renormalization of the tunneling splitting

$$\Delta_{0*} = \begin{cases} \Delta_0, & \Delta_0 \gg nb, \\ \eta^n \Delta_0, & \Delta_0 \ll nb. \end{cases} \tag{5}$$

Qualitatively, this means a gap for the effective tunneling splitting within the interval $(\eta^n nb, nb)$. Beyond this interval, the distribution function$P\left(\Delta, \Delta_{0*}\right)$is obtained from Eq. (2) as a result of substituting Δ_0 with Δ_{0*}. Thus, in the first approximation, the low-energy excitations in glasses having quadrupole energy can be described by Hamiltonian (1) with Δ_0 substituted with Δ_{0*}. Then

$$P\left(\Delta_{0*}\right) = \frac{P}{\Delta_{0*}}\left(\theta(\Delta_{0*} - nb) + \theta(\eta^n \cdot nb - \Delta_{0*})\right) = \frac{P}{\Delta_{0*}} g\left(\Delta_{0*}\right). \tag{6}$$

III. RESONANT PHONON ABSORPTION

The standard expression for the resonant single-phonon absorption rate by a tunneling system with the parameters EandΔ_{0*} takes the form [2]

$$\tau_{ph}^{-1}\left(E,\Delta_{0*}\right)=\frac{\pi\gamma^2\omega}{\rho v^2}\frac{\Delta_{0*}^2}{E^2}\tanh\left(E/2T\right), \tag{7}$$

$$E=\left(\Delta^2+\Delta_{0*}^2\right)^{1/2}. \tag{8}$$

Here$\omega=E$is a resonant phonon frequency, γ is a coupling constant between the tunneling system and phonons, ρ is a material density, and v is the sound velocity. Here and below, we put the Planck constant $\hbar=1$.

Since

$$d\Delta=\frac{EdE}{\left(E^2-\Delta_{0*}^2\right)^2}, \tag{9}$$

the distribution function (6) in the variables E,Δ_{0*} reads

$$d\left(E,\Delta_{0*}\right)=P\frac{EdEd\Delta_{0*}}{\Delta_{0*}\left(E^2-\Delta_{0*}^2\right)^{1/2}}g\left(\Delta_{0*}\right). \tag{10}$$

Therefore, the *total* phonon absorption rate is given by the expression

$$\tau_{ph}^{-1}\left(\omega\right)=P\frac{\pi\gamma^2}{\rho v^2}\tanh\left(\frac{\omega}{2T}\right)\int_0^{\omega}\frac{g\left(\Delta_{0*}\right)\Delta_{0*}d\Delta_{0*}}{\left(E^2-\Delta_{0*}^2\right)^{1/2}}. \tag{11}$$

The integral in Eq.(11) can be estimated as follows

$$\omega\left(\sqrt{1-\left(nb/\omega\right)^2}\theta\left(\omega-nb\right)+1-\sqrt{1-\min\left(\left(\eta^n nb/\omega\right)^2,1\right)}\right). \tag{12}$$

Substituting Eq.(12) into Eq.(11), one obtains

$$\tau_{ph}^{-1}\left(\omega\right)\approx P\frac{\pi\gamma^2}{\rho v^2}\tanh\left(\frac{\omega}{2T}\right)\omega\begin{cases}1, & \omega>nb;\\ \left(\eta^n\cdot nb/\omega\right)^2, & \eta^n nb<\omega<nb;\\ 1, & \omega<\eta^n nb.\end{cases} \tag{13}$$

Thus, beyond the frequency interval ($\eta^n nb<\omega<nb$), the nuclear quadrupole interaction does not change the expression for the resonant sound absorption and coincides with that given by the STM. However, within this frequency interval, the absorption experiences a decrease. The decrease is noticeable since the parameter $\left(\eta^n\cdot nb/\omega\right)^2\ll 1$. Also let us note that, for the STM, $\tau_{ph}^{-1}\sim\omega$ at the temperatures $T<nb$. Instead, the involvement of the quadrupole interaction yields $\tau_{ph}^{-1}\sim 1/\omega$ for the frequencies within the quasi gap.

Estimate (13) is based on the qualitative distribution (6), which has a quasi gap for the effective tunneling splitting within the frequency interval ($\eta^n nb<\omega<nb$). A more quantitative analysis of the phonon lifetime is given in the next section based on the toy nuclear spin model spins described in [24].

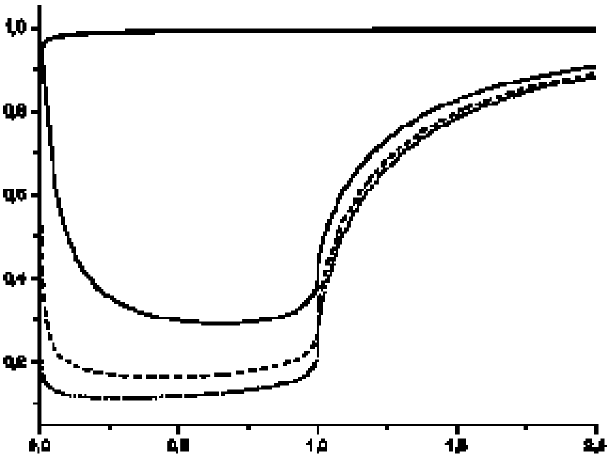

FIG. 1: The ratio $\tau_{toy}^{-1}(\omega)/\tau_{STM}^{-1}(\omega)$as a function of the relative frequencyω/nb. The thick solid curve corresponds to the case $n = 0$. The thin solid curve corresponds to the case $n = 2$. The dotted line curve corresponds to the case $n = 4$ and the dash-dot line curve corresponds to the case $n = 6$.

IV. NUMERICAL SIMULATION BASED ON THE TOY MODEL

In the toy model described in [24], the nuclear spins are substituted with the identical oscillators. This "bosonization" approach to nuclear spins is justified when one deals with the low energy states of many spins [31]. Within the toy model (see all details in Ref. [24]), the distribution function for the effective tunneling splitting Δ_{0*} is given by the expression

$$P\left(\Delta_{0*}\right) = \frac{1}{\Delta_{0*}}\begin{cases} 1, & \Delta_{0*} > bn; \\ \frac{1}{1+3n\ln(1/\eta)\left(\frac{\tilde{\Delta}_0}{bn}\right)^2\left[1-\left(\frac{\tilde{\Delta}_0}{bn}\right)^2\right]^{1/2}}, & \Delta_{0*} < bn, \end{cases} \tag{14}$$

Here, n and $b$have the same sense as in the sections above, and $\tilde{\Delta}_0$ is the implicit solution of the equation

$$\Delta_{0*} = \Delta_0 \exp\left\{-n\ln\left(1/\eta\right)\left[1-\left(\frac{\Delta_0}{bn}\right)^2\right]^{3/2}\right\}. \tag{15}$$

It is easy to see that distribution (15) reveals a dip within the interval $(\eta^n nb < \omega < nb)$. Thus, Eq. (15) correlates with Eq. (6).

The total phonon relaxation rate is then obtained if one substitutes Eq. (14) into Eq. (11). Doing the numerical estimation of the relaxation rate, one takes the representative value of the parameter $\eta = 1/3$ (see. Ref. [24]). We consider the cases when the number of quadrupoles contained in the tunneling particles is $n = 0, 2, 4, 6$. The frequency ω is measured in the units of nb.

V. DISCUSSION AND CONCLUSION

In Secs. **III** and **IV,** we have established that the nuclear quadrupole interaction strongly affects the ultrasonic absorption in glasses. The analytical estimation (13) describes the phonon absorption rate in the presence of the quadrupole interaction. The strong increase of the phonon lifetime for the frequencies $\eta^n nb < \omega < nb$ is found. Assume that $T < \omega$; then, we have the prefactor $\tanh(\omega/2T) = 1$. Next, within the STM, $\tau_{ph}^{-1} \sim \omega$; however, the involvement of the nuclear quadrupole interaction results in a different dependence $\tau_{ph}^{-1} \sim 1/\omega$ for the frequencies within the quasi gap. This finding is confirmed by the numerical simulation in Sec. **IV**. It follows from the results obtained in this section that the phonon lifetime increases as the quadrupole interaction becomes stronger.

The involvement of the nuclear quadrupole interaction reduces the overlap between the states in the left and the right wells. Therefore, the coupling between the tunneling systems and phonons decreases as a result of the quadrupole interaction. Ex facte, this should result in decreasing the resonant ultrasonic absorption and increasing the phonon lifetime. However, this conclusion is incorrect since the phonon lifetime depends not only on the tunneling splitting but also on total energy splitting E. This is evidently confirmed by the direct estimation of the phonon absorption rate τ_{ph}^{-1} as the frequency becomes smaller than $\eta^n nb$. The proper reason for the effect is an appearance of the quasi gap in the distribution function for the tunneling splitting.

The thermal conductivity in glasses is due to phonons, which are resonantly scattered by the tunneling system At a given temperature T the heat is mainly transferred by the phonons with frequencies$\omega \sim T$. It is known that the thermal conductivity $\kappa(T) \sim \tau_{ph}^{-1}(T)$. It directly follows from Eq. (13) that the thermal conductivity should be increased by a large factor $(T/\eta^n nb)^2$as compared with the STM one within the temperature region $\eta^n nb < T < nb$. The numerical simulation for the thermal conductivity performed within the toy model resembles the behavior of τ^{-1} presented in the figure.

This conclusion contradicts to the result obtained in [32] where the effect for the thermal conductivity was found to be approximately 4%. The main reason for the discrepancy between our results and that of [32] is that in [32] an approximation, valid only if $nb \ll T$, is used. At the same time, the results are applied to the case $nb \approx T$. Also, neither Ref. [21] nor Ref. [32] takes the strong renormalization of the distribution function given by Eq. (13) into account.

Let us now consider how the external magnetic field influences the phonon relaxation in the temperature region where the quadrupole effect is of great importance. For sufficiently strong magnetic field, the Zeeman splitting exceeds the quadrupole interaction. Therefore, the latter can be neglected. Under such conditions, eigen nuclear states in both DWP wells are the same and completely determined by the magnetic field. Therefore, the overlap integral $\eta \approx 1$. Therefore, application of the magnetic field should result in a strong decrease in the phonon mean free path and the thermal conductivity. So, in a strong magnetic field, i.e., $m > nb$, the quadrupole effect is suppressed, and we should expect both the standard phonon resonant absorption rate and standard thermal conductivity.

The corresponding experiments at ultra-low temperatures without and in magnetic field can elucidate the effect of the quadruple interaction on the properties of multi-component glasses.

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