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Interdefect elastic interaction in glass fiber and the theory of tunneling systems

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In a thin fiber of radius R, the long-range elastic dipolar interaction between two dipoles at a distance r from each other is modified from $V \propto 1/r^3$ to $V \propto \exp(-r/R)$. This offers opportunities to test the recent theory of tunneling systems in glasses based on the long-range interdefect interactions. Several possible experiments are proposed.

At low temperatures the thermal, acoustic, and dielectric properties of glassy materials can generally be understood on the basis of the tunneling-system or two-level-system (TLS) model.[1-4] In this model it is assumed that there exist atoms (or clusters of atoms) each with two potential minima. The tunneling motion between the two minima constitutes the low-temperature dynamics. Based on a few simple and plausible assumptions about the broad distributions in energy splitting between the two levels and in the tunneling rates, the TLS model provides a satisfactory phenomenological interpretation of the low-temperature behavior of glasses.[4]

In the form of the TLS model as it was originally proposed and generally accepted, the tunneling entities are regarded as primarily noninteracting objects and essentially independent of one another. The interaction between the TLS is considered important only in special contexts, such as in causing level broadening and spectrum diffusion.[5-7] While the existence of the inter-TLS interactions is not questioned, their importance is often underestimated if not entirely overlooked.

Recently, Yu and Leggett[8] proposed that such interactions could well be the key to a deeper understanding of the low-temperature properties of glasses. It is suggested, for example, that the near-flat distribution in energy splitting (flat density of states) may be a consequence of the long-range ($\propto 1/r^3$) interdefect elastic dipolar interaction.[9-11] According to the theory, the interaction renders the density of states essentially flat and slowly increasing with energy, as it has been generally observed. Such a scenario is consistent with the results of both numerical and approximate analytical calculations.[9,11] It is further speculated that the interaction theory may explain a number of long-standing puzzles about the TLS. Consider, for example, the acoustic attenuation in glasses at low temperatures. It is found that the dimensionless ratio λ/l, where λ is the wavelength of sound and l its mean free path, is essentially independent of material and roughly equal[12] to $\frac{1}{150}$. Within the TLS model[4] this quantity is given by, up to numerical factors, $\gamma^2 n/\rho v_s^2$, where γ, the deformation potential, measures the coupling strength between a TLS and the local strain, n is the TLS density of states, ρ the mass density of the glass, and v_s the speed of sound. All four quantities vary from one glass to another, but the combination $\gamma^2 n/\rho v_s^2$ show much smaller variations. Within the noninteracting TLS model this can only be attributed to a curious coincidence. It becomes more natural in the interaction theory, where the coupling strength γ and the density of states n are determined by the interdefect elastic interaction and are therefore not independent of the elastic modulus ρv_s^2. This was suggested by Freeman and Anderson[12,13] and has received tentative support both from preliminary calculations based on the interaction model[14] and from numerical simulations.[15]

Although the interaction theory is still in its infancy, it has already provided refreshing insight into many familiar results of the TLS model. It shares some attractive features with the recently proposed microscopic theory of the tunneling systems in KBr:KCN.[16] On the other hand, so far, the theory has been built to interpret the existing experimental data; it has yet to make new predictions that can be used to discriminate itself from the conventional noninteracting TLS model. In view of the success and promise of the interaction theory, it is of urgent interest to design and perform experiments that can test the basic premise of the theory, namely, that the long-range interdefect interaction is vital to the formation and the properties of the TLS.

In this paper we discuss this question from the theoretical point of view. We show that experiments performed on glass fibers are potentially useful in testing the interaction theory. Here we assume a fiber to be a homogeneous rod of glass with a radius R on the order of microns. Further, we assume the fiber to be "glassy" in the sense that, apart from its shape and size, it is indistinguishable from bulk glass; in particular, it should have its share of TLS. We find that within such a fiber, the interdefect interaction is no longer $\propto 1/r^3$ but rather $\propto \exp(-r/R)$ for two defects separated by a distance $r > R$. Thus, the long-range interaction is effectively turned off among those defects that are more than R apart. If the interaction theory is correct and the *long-range* nature of the interaction is indeed responsible for the properties of low-energy TLS, then the behavior of TLS in a fiber should be very different from that in the bulk, at least in the low-energy limit. If, on the other hand, the long-range interaction is unimportant and the conventional picture is correct, then no such difference is expected. This is because the conventional theory implicitly assumes the size of a TLS to be of the atomic scale. Since the radius of a fiber is much larger than the intrinsic size of a tunneling system, the behavior of a TLS should not be affected by the size of the fiber.

Neither the conventional TLS theory nor the interaction theory discusses the microscopic nature of the TLS; instead, both assume the existence of some "defects"[8,17] or "tunneling centers." Our discussion bypasses this issue, but assumes that the defects are fairly strongly localized objects, involving atoms within a sphere no larger than a few tens of angstroms at most, and possibly much smaller. There are at least three possible types of long-range interactions between two such defects. First, there is the elastic dipolar interaction to be discussed in detail below. Second, if the defects have nonvanishing electric dipolar moments,[18] there is also an electric dipolar interaction. Finally, in metallic glasses there is an interdefect interaction mediated by the electrons. All three interactions are essentially $\propto 1/r^3$, with possibly complicated angular dependence and oscillating signs. In the interaction theory, it is the $1/r^3$ nature of the interaction that is primarily responsible for the properties of the TLS. Accordingly, we will focus on the long-range aspect of the interaction.

The elementary entity in elastic interaction is an elastic dipole.[19,20] (There is no such thing as an elastic monopole.) Unlike an electric dipole, an elastic dipole is a second-rank symmetric tensor[20] λ_{ij} $(i,j=1,2,3)$, generally with a nonvanishing trace corresponding to the fractional change in volume caused by the dipole. It couples to the local strain u_{ij} so that the energy of an isolated elastic dipole in a uniform strain is $\lambda_{ij}u_{ij}$. (Repeated indices are always summed over.) In an infinite medium, it is straightforward to show that the interaction between two dipoles $\lambda^{(1)}$ and $\lambda^{(2)}$ goes like $1/r^3$. Consider, for example, an isotropic medium with a Young's modulus Y and a Poisson ratio σ. The dipolar interaction is given by

$$V=\lambda_{ij}^{(1)}\lambda_{kl}^{(2)}\frac{1+\sigma}{8\pi Y(1-\sigma)}\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_l}\times\left[\left[(3-4\sigma)\delta_{ij}+\frac{x_i x_k}{r^2}\right]\frac{1}{r}\right]. \quad (1)$$

Results of this type are well known[10] and can be easily obtained using the classical theory of elasticity.[21] More conveniently, we can use the following technique. We note that the elastic interaction is fundamentally the result of the exchange of virtual phonons, in exact analogy with the electrostatic interaction, which is the result of virtual photon exchange. We can write down the Hamiltonian of the combined system of phonons plus two dipoles located at 0 and **r**, eliminate the phonons, and identify the r-dependent part of the energy with the static interaction between the defects.[22] To illustrate, we consider a model system of scalar phonons coupled to two vector dipoles and write the total Hamiltonian as

$$H=\lambda^{(1)}\cdot\nabla u(0)+\lambda^{(2)}\cdot\nabla u(\mathbf{r})+\int d^3x\left[\frac{\rho}{2}\left(\frac{\partial}{\partial t}u\right)^2+\frac{K}{2}(\nabla u)^2\right]. \quad (2)$$

Since the Hamiltonian is quadratic in the phonon degrees of freedom, we can immediately integrate out the phonons to find the static interaction between the dipoles to be

$$V=\lambda_j^{(1)}\lambda_l^{(2)}\int\frac{d^3k}{(2\pi)^3}\frac{k_j k_l}{Kk^2}e^{ikr}. \quad (3)$$

Carrying out the Fourier transform, we have

$$V=\lambda_j^{(1)}\lambda_l^{(2)}\frac{1}{4\pi K}\left[\frac{\delta_{jl}}{r^3}-\frac{3x_j x_l}{r^5}\right]. \quad (4)$$

A similar calculation using the vector phonons reproduces Eq. (1).

We now consider an ideal fiber suspended in vacuum with two dipoles located at cylindrical coordinates (r_1,ϕ_1,z_1) and (r_2,ϕ_2,z_2), respectively. The boundary condition is that the force on the surface of the fiber must be zero or $\partial\sigma_{ik}/\partial x_k=0$ at $r=R$, where σ_{ik} is the stress tensor. We expand the displacement field in terms of the eigenmodes of the system under such a boundary condition. Making, once again, the scalar wave approximation first, we have

$$u(r,\phi,z)=\int\frac{dk}{2\pi}\sum_{m,s}J_m(\beta_{ms}r/R)\times[a_{kms}e^{im\phi}e^{ikz}e^{-i\omega_{kms}t}+\text{c.c.}]. \quad (5)$$

Here J_m, $m=0,1,2,\ldots$, is the mth Bessel function, the constants β_{ms} are determined by the boundary condition at $r=R$ which, in our scalar approximation, is $J_m'(\beta_{ms})=0$, $s=1,2,\ldots$. We have also introduced $\omega_{kms}^2=(K/\rho)(k^2+\beta_{ms}^2/R^2)$, and the a's are complex coefficients. Using this basis and repeating the calculations of Eq. (2) to Eq. (4), we can find the static interaction in a fiber. It is convenient to project the dipoles unto their respective local cylindrical unit-vector basis. The coupling between the two vector dipoles is then given by a dyad. Consider, for example, its zz component. We have

$$V_{zz}=\lambda_z^{(1)}\lambda_z^{(2)}\sum_{m,s}\int\frac{dk}{2\pi K}\frac{kk}{k^2+\beta_{ms}^2/R^2}\times e^{ik(z_1-z_2)}F(r_1,r_2,\phi_1,\phi_2). \quad (6)$$

Here F is independent of k. For large $|z_1-z_2|$, we have

$$V_{zz}\sim-\frac{\lambda_z^{(1)}\lambda_z^{(2)}}{2K}F(r_1,r_2,\phi_1,\phi_2)\times\sum_{m,s}\frac{\beta_{ms}}{R}\exp(-\beta_{ms}|z_1-z_2|/R). \quad (7)$$

In Eq. (7) the root $\beta_{0,1}=0$ is excluded from the sum. This is because the k integral in Eq. (6) is one dimensional, and the contribution from the term with $\beta_{0,1}$ is simply proportional to $\delta(z_1-z_2)$ and has no long-range contribution. Equation (7) is clearly dominated by the smallest nonzero root of the Bessel function, given by $\beta_{1,1}\approx 1.8412$. Similarly, we can examine all the other components of the coupling dyad and show that the interaction is always of short range. Qualitatively the same result is found when the tensor version of the calculation is carried out. The complete result is somewhat complicated and will be published elsewhere. In our calculation we have assumed the linear elasticity theory to be valid and the elastic defects to be pointlike pure dipoles with no high-order components. Both assumptions are expected to be valid in the long distance, weakly interacting regime. The results are explicitly presented here in the simple model of isotropic elastic medium but are of general validity and are applicable to anisotropic, crystalline fibers.

The physics of this result can best be understood using an analogy with the electrostatic interaction. Since our fiber is suspended in vacuum, the phonons in the fiber cannot leak into the environment. There is therefore a total internal reflection of acoustic waves at the surface of the fiber, and the fiber behaves as an ideal acoustic waveguide. Consider a waveguide for the electromagnetic wave made of a good conductor. If we place two point charges in the waveguide, the effective interaction between them is no longer the familiar $1/r$ Coulomb interaction. The image charges induced by the two point charges largely cancel the direct interaction between the test charges, leaving only a weaker, short-range interaction. In the elastic case, the image elastic dipoles also act to cancel the direct interaction, resulting in a similar short-range interaction. To avoid confusion, we note that since the dielectric constant of an insulating glass fiber is not very different from that of vacuum, the electric dipolar interaction is essentially unmodified in a fiber. Fortunately for our application, in glasses the elastic dipolar interaction usually dominates; the electric dipolar interaction is typically weaker by 2 orders of magnitude (see below) and can be neglected as a first approximation. We have performed calculations to show that in a metallic glass the electron-mediated, Ruderman-Kittel-Kasuya- Yosida-like interaction is also modified in a fiber, just like the elastic interaction. Its magnitude, though difficult to estimate reliably, is comparable to or less than that of the elastic interaction. We conclude, then, that the dominant long-range interdefect interactions are effectively turned off at a distance greater than the fiber radius.

While this result is significant in its own right, we are primarily interested in its potential applications to glasses. It is easy to estimate the interaction strength between two defects in glass. Consider first the elastic dipolar interaction, which can be estimated to be[10] $V \sim g/r^3$ with $g = \gamma^2/\rho v_s^2$. Taking the values for SiO_2 we find $g = 1.2 \times 10^{-35}$ erg cm^3. It is known that at least some TLS have an intrinsic electric dipole moment on the order of $p = 0.5$ D.[18] The electric dipolar interaction can thus be estimated to be $\sim p^2/r^3$ with $p^2 = 2.5 \times 10^{-37}$ erg cm^3 and is smaller than the elastic interaction by nearly 2 orders of magnitude. In a fiber of radius $R = 1$ μm, the long-range elastic interaction is essentially $1/r^3$ up to a distance of 1 μm, where it has a magnitude of $\sim 10^{-23}$ erg and corresponds to a frequency of $\nu \sim 1500$ Hz. Beyond 1 μm the interaction decays rapidly and we have effectively no *long-range* interaction weaker than $h\nu$ with ν on the order of 10^3 Hz.

The number of experiments that can be carried out in a fiber is fairly limited. First of all, to maintain the one dimensionality of the fiber, the experiment must be done on essentially isolated fibers (or a loose bunch of fibers), not on a tight bundle. Second, the experiment must be done at low temperatures, and the requirement of good thermal contact with a refrigerator is in direct conflict with the requirement that the fiber be isolated mechanically from the environment. Notwithstanding these obvious difficulties, we suggest several possible experiments. It may be possible to measure the mechanical Q of a vibrating fiber, thereby determining indirectly the effective phonon attenuation at the frequency of vibration without having to attach a transducer to the fiber. The lowest bending mode has a frequency[23] $\omega \approx 1.76(Y/\rho)^{1/2}R/L^2$, where L is the length of the fiber. For a micron radius fiber with a length $L \sim 1$ cm, this is on the order of 100–1000 sec^{-1} and is tunable to some extent by changing L. As we have estimated earlier, such a frequency is of the same order of magnitude as the minimum energy of $1/r^3$ interaction in the same fiber, $E_{\min}$ (defined to be the energy scale at which the $1/r^3$ interaction crosses over to the short-range regime), and is ideally suited for determining whether the near-universal value of $\lambda/l \sim \frac{1}{150}$ is indeed a consequence of the long-range elastic interaction. For if the interaction theory is correct, there should be a change in slope on the Q vs ω plot near $\omega \sim E_{\min}/\hbar$, signaling a crossover from the bulk behavior to the one-dimensional behavior. Such a result, if observed, would be a strong support for the interaction theory. On the other hand, because the frequency is low, the experiment is effectively in the very-high-temperature limit. The estimated Q due to the resonant absorption of the TLS is of the order of $150kT/\hbar\omega$ or about 10^8. Thus the damping is likely to be dominated by other sources.[24] A further complication is the relaxational contribution[25] of TLS, which is expected to be large.

It may also be possible to perform an electric echo experiment[18] on a loose bunch of fibers. Since there is no need of making mechanical contact with the sample, this has some advantage over the acoustic measurements. On the other hand, so far the interaction theory has not made any predictions about the outcome of such an experiment, and it is not clear what the signature of the interaction model might be. Optical dephasing measurement[26] is another possibility. Here the complication is the coat surrounding the fiber core, which makes the fiber much thicker, with a radius generally on the order of 100 μ. Finally, noise measurements in very small metallic-glass samples[27] can be used to monitor the motion of essentially individual TLS. It will be interesting to study the correlation of different TLS in the light of the interaction theory to see if a signature can be found.

In conclusion, we have demonstrated that the interdefect interaction is of short range in a fiber for distances greater than the fiber radius. This offers intriguing opportunities for designing experiments to test the interaction theory for the low-temperature properties of glasses.

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