Interacting Defect Model of Glasses: Why Do Phonons Go So Far?

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We present a model of insulating glasses in which defects interact strongly via the elastic strain field. The interactions are oscillatory at short range and cross over to $1/r^3$ interactions at longer length scales. This crossover is marked by a strong renormalization of the density of states and is consistent with the long phonon mean free path found at low temperatures.

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Despite their microscopic disorder, glasses below 1 K exhibit amazingly long phonon mean free paths as deduced from thermal-conductivity experiments.[1] In SiO_2, for example, the mean free path $l \sim 60$ μm at 0.5 K. In general at low frequencies, and hence low temperatures by the dominant-phonon approximation, $l \sim 150\lambda$, where λ is the phonon wavelength and the proportionality constant can vary by a factor of 3 either way from material to material. Around 200 GHz the mean free path drops sharply and becomes considerably shorter ($l \sim \lambda$) at higher frequencies.[1-4] Other than to say that the concentration of scatterers is dilute, the standard model of two-level systems[5] provides no real explanation for why l is so long at low temperatures. In this paper we address this puzzle using a model of strongly interacting defects.[6] Since we confine ourselves to insulating glasses, the interactions are elastic. Using a renormalization-group scheme, we show that these interactions have a crossover as a function of length scale. This crossover is marked by a decrease in the density of states and hence an increase in the phonon mean free path at longer length scales which can be associated with lower-energy scales.

Before presenting the details we briefly review the thermal-conductivity and specific-heat experiment,[1] and their implications. Below 1 K the specific heat in insulating glasses is slightly superlinear in temperature ($C \sim T^{1+\delta}$, $\delta \sim 0.1$-0.3) and the thermal conductivity is slightly subquadratic ($\kappa \sim T^{2-\epsilon}$, $\epsilon \sim 0.05$-0.2). Between 3 and 10 K the thermal conductivity exhibits a plateau and then continues to rise at higher temperatures. The specific heat also displays an anomaly in this temperature range: there is a bump in C/T^3 vs T.

What do the plateau and bump imply about the physics of glasses? No hints come from the two-level system (TLS) model. If we assume that phonons carry the heat[2] in this temperature range, then the plateau represents the previously mentioned crossover from a long mean free path at low frequencies to a short mean free path at higher frequencies. Such behavior is consistent with a rise in the density of states at some energy E_0 since this would increase both the number of scatterers as well as the number of excitations contributing to the specific heat.[7,8] Fitting the bump in C/T^3 for various materials sets E_0 in the range from 10 to 40 K, consistent with the rise in the density of states measured by neutron scattering[9] and Raman scattering.[10,11]

With the above considerations in mind, we sketch an alternative approach to the problem of glasses. We assume that glasses contain some sort of defects that have some low-lying energy excitations with no restriction on the energy range. We want our model to be sufficiently general so that we do not need to specify a microscopic model of the source of excitation. We assume that these defects couple linearly to the strain field:

$$H = \sigma_{\alpha\beta}(\mathbf{r})\epsilon_{\alpha\beta}(\mathbf{r}) , \tag{1}$$

where $\epsilon_{\alpha\beta}(\mathbf{r})$ is the symmetric strain field and $\sigma_{\alpha\beta}(\mathbf{r})$ is the stress field associated with the defects. The indices α and β range over the real-space directions x, y, and z and the sum over repeated indices is understood. In the TLS model[5] $\sigma_{\alpha\beta}$ is replaced by ΓS, where S is a spinlike TLS operator represented by Pauli matrices. Γ is a vector in spin space and a matrix in real space. The spin representation is that of the energy eigenstates of the two-level system. The off-diagonal components in spin space correspond to transitions between energy levels. The diagonal components do not involve transitions and are Ising type. We use this basis in the calculation described later.

The defects interact with each other via the strain field. Here we break with tradition and propose that it is interactions, not the intrinsic splitting of the levels of a single defect, which dominate the energy scale. Using second-order perturbation theory to calculate the off-diagonal effective interaction between two defects which have the same energy splitting ΔE, one finds[12]

$$H_{\text{eff}}(\mathbf{r}-\mathbf{r}',\Delta E) = \sum_{\lambda,\mathbf{p}} \frac{1}{\rho c_\lambda^2} \left(\frac{c_\lambda^2 p^2}{(\Delta E)^2 - c_\lambda^2 p^2} \right) e^{-i\mathbf{p}\cdot(\mathbf{r}-\mathbf{r}')} \eta_{\alpha\beta}^{(\lambda)} \eta_{\gamma\delta}^{(\lambda)} \sigma_{\alpha\beta}(\mathbf{r}) \sigma_{\gamma\delta}(\mathbf{r}') , \tag{2}$$

where $\eta_{\alpha\beta}^{(\lambda)} = (\hat{\mathbf{p}}_\alpha \hat{\mathbf{e}}_\beta^{(\lambda)} + \hat{\mathbf{p}}_\beta \hat{\mathbf{e}}_\alpha^{(\lambda)})/2$. The sum over λ is over the longitudinal and transverse phonon polarizations. ρ is the density and c is the speed of sound. $\hat{\mathbf{p}}_\alpha$ is the αth component of the unit phonon wave vector and $\hat{\mathbf{e}}_\beta$ is the βth component of the unit phonon polarization vector. For $\Delta E = 0$, the effective interaction becomes dipolar, i.e., it goes as g/r^3, where $g \sim \gamma^2/\rho c^2$. Taking $\gamma \sim 1$ eV, we estimate $g \sim 5\times 10^4$ K Å^3.

The crossover is associated with a change in the off-diagonal interaction between defects from oscillatory to dipolar with increasing length scale.[13] (The diagonal interaction is dipolar at all length scales.) Imagine that we divide a sample into blocks of size r_1 such that the distance between the centers of the blocks is also r_1. Suppose that we know the density of states in each block and hence the energy-level spacing. We then work out the interaction between blocks via phonons. Having done this, we can group our blocks into bigger blocks of size r_2 and repeat the process. By iterating, we go to longer and longer length scales. There are two natural length scales in the problem. The first, which we will denote by r_d, is the average nearest-neighbor distance between bare defects. The second, which we will call r_0, is given by $(g/\hbar c)^{1/2}$.

We start by considering individual defects, i.e., blocks of size $r \sim r_d$. Suppose the intrinsic energy splitting ΔE_0 of a bare defect is much less than the phonon energy $\hbar c/r_d$. (r_d is the relevant phonon wavelength at this length scale.) Then the strain interaction will dominate the energy splitting and the effective splitting will be $\Delta E \sim g/r^3$. According to mean-field theory,[6,14,15] the density of states $n(E)$ will be $\sim 1/g$ and $l \sim \lambda$. At longer length scales, $r_d \ll r \ll r_0$, the level separation in the blocks of size r is $\Delta E \sim [n(E) r^3]^{-1} \sim g/r^3$. The phonon energy, $\Delta\omega_{\mathrm{ph}} \sim \hbar c/r$, is much less than ΔE. Hence the phonons are too weak to cause transitions and are ineffective at causing off-diagonal interactions between blocks. At longer length scales, $r \gtrsim r_0$, $\Delta\omega_{\mathrm{ph}} \gtrsim \Delta E$, phonons are effective mediators of the interaction between blocks, and the interaction goes as $1/r^3$. Notice that setting the phonon energy equal to the typical energy splitting ($\hbar c/r = g/r^3$) and solving for r gives r_0. Thus r_0 provides a natural crossover from a region of ineffective interaction to a region of dipolar interaction. We believe this crossover is associated with the plateau in the thermal conductivity and the bump in C/T^3. Using our previous value of $g \sim 5\times 10^4$ K Å^3, we estimate $r_0 \sim 13$ Å and $\hbar\omega_0 = \hbar c/r_0 \sim 24$ K. This is consistent with the value for the rise in the density of states ($E_0 \sim$ 10–40 K).

Interactions also strongly renormalize the density of states as one goes through the crossover. We now present a calculation which is consistent with this assertion. We start by dividing our sample up into concentric spherical shells like those of an onion. Imagine randomly pasting postage stamps of various sizes on the layers of the onion. These represent the defects. We assume that without interactions the energy levels of the bare defects are degenerate. Thus $\Delta E_0 = 0$ initially. The density of states N_1 resulting from off-diagonal interactions between the initial core and the first layer is calculated. From N_1 we find the energy splitting ΔE_1 of this composite which forms the new core. This is the ΔE we use in $H_{\mathrm{eff}}(\mathbf{r},\Delta E)$ to calculate the density of states in the next iteration. By iterating this procedure, we go to longer and longer length scales and watch how the density of states changes as we do this. In general, the density of states N_m resulting from iterating through m shells is given by a definition akin to that found in mean-field theory:[6,14,15]

$$N_m = \left[\pi \sum_{i=1}^{m} \int_{i\text{th shell}} d^3r\, H_{\mathrm{eff}}(\mathbf{r},\Delta E_{i-1})\right]^{-1}, \tag{3}$$

where $H_{\mathrm{eff}}(\mathbf{r},\Delta E)$ is given by Eq. (2) and $\mathbf{r}$ is measured from the center of the core. The sum i is over the layers of the onion and ΔE_{i-1} is given by

$$\Delta E_m = [N_m \times (\text{volume of core with } m \text{ shells})]^{-1}. \tag{4}$$

We neglect frustration and mimic the spin degrees of freedom represented by the $\sigma_{\alpha\beta}(\mathbf{r})$ associated with the defects in the layers by real numbers which can be positive or negative, depending on which choice minimizes the interaction energy with the core whose stress components have been set positive. We fix the magnitude of the coupling constants and watch how the density of states changes relative to them. γ_s is the magnitude of the traceless symmetric part of $\sigma_{\alpha\beta}$ and γ_c is that of $\mathrm{Tr}(\sigma_{\alpha\beta})/3$ for both the core and the shell defects. (γ_s and γ_c are related to the longitudinal and transverse couplings γ_l and γ_t, e.g., $\gamma_s = \sqrt{2}\gamma_t$). We carry out the iteration scheme with the aid of a computer. A typical onion has 200 layers, each of which is 5 Å since this is the typical length of a structural unit. Our input consists of ρ, c_l, c_t, and γ_s. The first four quantities are measured experimentally, though the value of γ_s is subject to some uncertainty. To determine γ_c, we use $(\gamma_c,\gamma_s)^2 = \frac{3}{2}(\gamma_l/\gamma_t)^2 - 2$ and the empirical fact that $\gamma_l/\gamma_t \sim 1.5$ at long length scales.

The numerical results indicate that the density of states is strongly renormalized by the strong interactions between neighboring defects. To gain some understanding of these results, we note that at short range $\Omega = \Delta E r/\hbar c$ is large and the dominant interactions go as $(\Omega^2/r^3)\cos\Omega$. (Ω can be thought of as $\Delta E/\hbar\Delta\omega_{\mathrm{ph}}$ in the block rescaling picture.) The dependence on Ω^2 implies that the interaction between two regions depends on the magnitude of the splitting arising from interactions within each region. Using (3) and (4) and ignoring angular factors, we obtain the following approximate recursion relation for $\Omega_m(r) = \Delta E_m r/\hbar c$:

$$\Omega_m(r) \sim \left(\frac{r_0}{r}\right)^2 \left[\ln\left(\frac{r_2^1}{r_1^1}\right) + \sum_{i=2}^{m} [\Omega_{i-1}(r_2^i)\sin\Omega_{i-1}(r_2^i) - \Omega_{i-1}(r_1^i)\sin\Omega_{i-1}(r_1^i)]\right], \tag{5}$$

where the sum is over shells making up the core. r_1^i and r_2^i are the inner and outer radii of the ith shell, and we used $r \sim r_2^m$. $r_0^2 \sim g/\hbar c_t$ and the coupling g is roughly given by $\gamma_s^2/\rho c_t^2$. For $r_0/r \gtrsim 1$, the lack of cancellation between the inner and outer radii of each shell as well as the absence of frustration allows each stage to reinforce the next and leads to a power series in $r_0^2 \sim g$ as well as $\Omega \gg 1$. This also results in a large denominator in (3) and hence a small density of states. Thus it is the strong interactions between near-neighboring defects which increase the energy splittings and "blow a hole" in the low-energy density of states. For long length scales where $(r_0/r) \ll 1$, Ω goes to zero as r^{-2}. Thus we expect Ω to go through a maximum at an r somewhat greater than r_0 (see Fig. 1), and the interactions will change from oscillatory to smooth.

As Ω goes to zero at long length scales, H_{eff} approaches gr^{-3} behavior. If we allow energy scales to be related to length scales via $E \sim gr^{-3}$, then $N(E) \sim [A + g\ln(E_0/E)]^{-1}$, where E_0 is on the order of the crossover energy and A contains the cumulative effects of the inner shells.[6,15,16] A is the dominant term and sets the order of magnitude of the density of states. This is just a restatement of the fact that it is the strong short-range interactions that renormalize $N(E)$. The behavior of $N(E)$ at low energies agrees with the flat density of states seen in experiment, and the logarithmic deviations are in the right direction, c.f. the superlinear specific heat and the subquadratic thermal conductivity. The inset of Fig. 1 shows how the renormalization of the density of states and of l/λ proceeds as one goes to longer length scales in a single onion. Using Fermi's golden rule, we have defined $l/\lambda = (2\pi^2 gN)^{-1}$, where we have taken $g = \gamma_s^2/\rho c_t^2$ and N is the final density of states (after 200 layers).

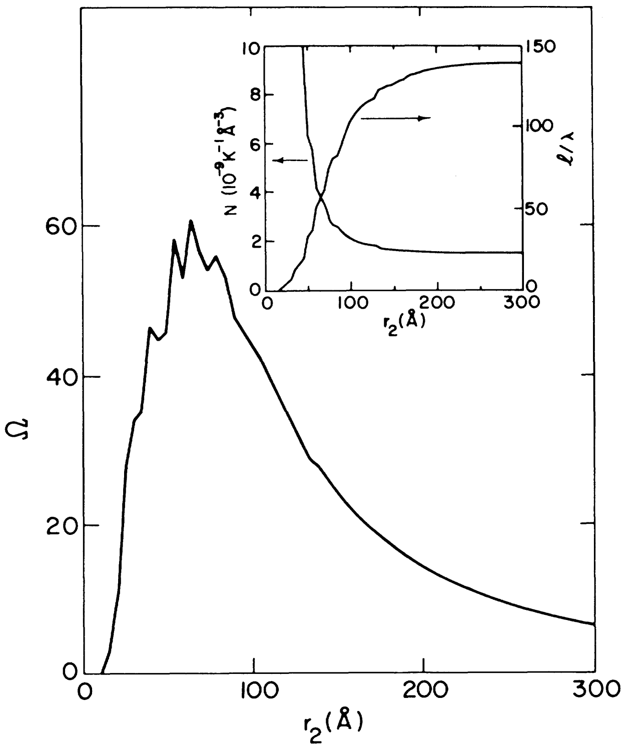

FIG. 1. The scaling parameter Ω vs the radius r evaluated for a single onion using parameters appropriate for SiO_2. Inset: Density of states vs r, the l/λ vs r for a single onion.

Since it is the short-range interactions involving the inner layers that dominate the magnitude of the density of states, fluctuations in the size, number, and placement of the defects in these layers lead to fluctuations in the final results of each onion. To provide some uniformity, we have set a lower bound on the fraction of solid angle occupied by defects, though l/λ is not very sensitive to this fraction. We also average the dimensionless attenuation λ/l over several hundred onions. Assuming that a suitable average is taken over the disorder, we note by dimensional arguments that $\langle l/\lambda \rangle$ can only be a function of the value of Ω after the first iteration, namely Ω_1. Our calculation indicates that different materials with different γ's have comparable values of l/λ due to the similarity of their initial values for Ω. If we use experimentally deduced values[16] of γ_s, then $\Omega_1 \sim 1$ and one does not start far enough above the crossover to get much renormalization of l/λ. However, the couplings may change as one goes to longer length scales. Using values of γ_s within roughly a factor of 2 of those quoted in the literature,[16] we have studied several different materials and find values of l/λ in agreement with experiment.

Our calculation of $\langle l/\lambda \rangle$ is a sensitive function of γ as can be seen from the upper curve in Fig. 2. As we men-

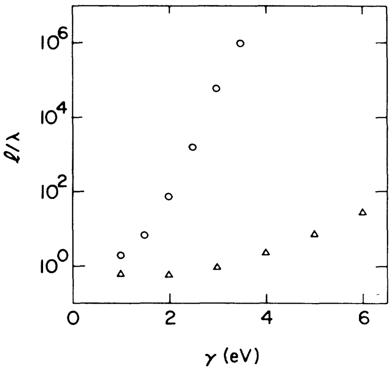

FIG. 2. l/λ vs γ with parameters appropriate for SiO_2. The dimensionless attenuation has been averaged over 400 onions with a minimum defect fraction of 0.4. The circles correspond to adjusting the sign of the stress components of each defect to optimize the interaction energy with the core. The triangles correspond to randomly choosing the signs but constraining each shell to give a positive contribution to the energy. A real system that is frustrated should lie between these two curves.

tioned before, iterating (5) where $(r_0/r) \gtrsim 1$ has a reinforcing effect which leads to a power series in $r_0^2 \sim g$ and hence roughly exponential dependence on γ^2. Including frustration would reduce the amount of positive feedback. However, since there is a net minimization of the energy, there will still be a net decrease in the density of states. We can get some idea of the importance of optimizing the stress configuration by looking at a case where it is substantially reduced. Figure 2 is a plot of l/λ vs γ with parameters appropriate for SiO_2 in which the attenuation has been averaged over 400 onions. If we assume that the logarithm of the attenuation has a Gaussian distribution, then there is roughly an error of $N^{-1/2} \sim 5\%$ error in $\ln(\langle l/\lambda \rangle)$. The lower curve shows the results of randomly choosing the sign of the stress components of the defects but with the constraint that the sum in (3) contain the absolute value of the contribution of each shell. This is equivalent to allowing each shell to make a global rotation of its stress components. We expect a frustrated system which minimizes the energy to have an l/λ which lies between these two curves. For SiO_2 this agrees with experimental values for l/λ for $\gamma_s \gtrsim 2$ eV.

To summarize we have presented a model of insulating glasses in which defects strongly interact via the elastic strain field. The dominant interactions are oscillatory ones at short distances which strongly renormalize the density of states. For a variety of materials this results in $l/\lambda \gg 1$ in agreement with experiment. Furthermore, there is a natural crossover at longer length scales to $1/r^3$ interactions. We have argued that this crossover can be associated with the plateau in the thermal conductivity and the bump in C/T^3.

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