

Supplementary Material for “Why phonon scattering in glasses is universally small at low temperatures”

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DISTRIBUTION OF γ

The coupling between phonons and TLS is of the form

$$H_{int} = \gamma D \sigma_z \epsilon \quad (1)$$

where $\gamma D \sigma_z$ is the elastic dipole operator. This operator is a projection onto the right and left basis states $|\psi_{R,L}\rangle$ of the full dipole operator D .

$$\begin{aligned} D &= \begin{bmatrix} D^{LL} & D^{LR} \\ D^{RL} & D^{RR} \end{bmatrix} \\ &= \frac{1}{2}(D^{LL} + D^{RR})\mathbf{I} + \frac{1}{2}(D^{LL} - D^{RR})\sigma^z + D^{LR}\sigma^x \end{aligned} \quad (2)$$

where $D^{AB} \equiv \langle \psi_A | D | \psi_B \rangle$. I is the identity matrix and thus can be dropped. $D^{LR} = D^{RL}$ depends on the overlap between the right and left wells and hence, is taken to be negligible. As a result, only the σ^z term remains, which is what goes into Eq. (1). *The key is that the σ^z term is half the difference of the elastic dipole moment D in the right and left wells.*

Assuming the magnitude γ_{max} of the elastic dipole to be the same in the right and left well, what can be said about the distribution of the (half) difference $\Delta D_{\alpha\beta}$, assuming some distribution for the relative “orientation” of the dipoles?

The case of vector dipoles, which is directly relevant for dielectric experiments, is instructive. Choose the dipole in the right well, say, to be along the z axis and the dipole on the left well to be randomly oriented in the \hat{n} direction on the unit sphere. The difference of two unit vectors is a vector that need not have unit length. The length of the new vector will multiply γ_{max} and dictate the magnitude of the dipole difference γ between the two wells. The probability distribution for $P(\gamma^2 = \gamma_{max}^2 |\hat{z} - \hat{n}|^2/4)$ is:

$$\begin{aligned} P(\gamma^2) &= \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin(\theta) \delta(\gamma^2 - \gamma_{max}^2 (1 - \cos(\theta))/2) \\ &= \frac{1}{\gamma_{max}^2} \quad 0 < \gamma < \gamma_{max} \end{aligned} \quad (3)$$

or equivalently $P(\gamma) = 2\gamma/\gamma_{max}^2$.

Now consider the elastic dipole. While the most general case is $D_{ij} = a n_i n_j + b m_i m_j$, with \vec{n} and \vec{m} being two unit vectors and a and b being constants, it

is easier consider the simpler case of $D_{ij} = \gamma_{max} n_i n_j$ where \vec{n} is a unit vector. Again we pick \vec{n} to be along the z axis in well 1 and randomly distributed on the unit sphere in well 2. Then $D_{ij}^{(1)} = \gamma_{max} \delta_{zi} \delta_{zj}$ and $D^{(2)} = R^{-1} D^{(1)} R$ where R represents an arbitrary rotation. Thus $\Delta D = D^{(1)} - R^{-1} D^{(1)} R$. The first operation of R is a rotation by an angle θ around a vector in the xy plane. The second operation is a rotation around the z axis which leaves the shape of ΔD unchanged so that the shape of ΔD only depends on θ . For the particular triad $\hat{x}, \hat{y}, \hat{z}$ that diagonalizes ΔD , we get:

$$\Delta D = \gamma_{max} \sqrt{1 - \cos^2(\theta)} \begin{bmatrix} 0 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (4)$$

This implies that ΔD is of the form $\gamma_{max} \sin(\theta) (\hat{z}_i \hat{z}_j - \hat{y}_i \hat{y}_j)$, which is more general than the original tensor but of the same norm. On that basis, what matters is the distribution of the prefactor. The probability distribution of half the magnitude of ΔD is then:

$$\begin{aligned} P(\gamma) &= \frac{1}{2} \int_0^\pi d\theta \sin(\theta) \delta(\gamma - \gamma_{max} \sin(\theta)/2) \\ &= \frac{4\gamma}{\gamma_{max}^2 \sqrt{1 - 4\gamma^2/\gamma_{max}^2}}, \quad 0 < \gamma < \gamma_{max}/2 \end{aligned} \quad (5)$$

Given that the deviation from linearity only occurs for large values of γ , the linear approximation $P(\gamma) \approx 4\gamma/\gamma_{max}^2$ is reasonable. Note that this expression and the normalizing value of γ_{max} differs from that found in Eq. (3) because we are dealing with elastic stress dipoles here rather than vector dipoles.

INTEGRATING OUT THE PHONONS

In a system of TLS interacting with phonons, the timescale describing the TLS is much slower than that of phonons with energies $\hbar\omega(k) \gg \Delta^o$ where $\omega(k)$ is the phonon spectrum and Δ^o is the tunneling matrix element of the TLS. Since the tunneling matrix element is at most a few Kelvin, the vast majority of the phonons are fast compared to the TLS. This situation allows us to integrate out the fast phonons from the problem, resulting in an effective low energy Hamiltonian with the TLS parameters renormalized. We detail here the steps involved. The starting point is the Hamiltonian for an ensemble of TLS interacting with phonons:

$$H = H_{\text{ph}} + \frac{1}{2} \sum_i (\varepsilon_i \sigma_i^z + \Delta_i^o \sigma_i^x) + \frac{1}{\sqrt{V}} \sum_{ik} \gamma_i \epsilon_k^i \sigma_i^z \quad (6)$$

where the free phonon Hamiltonian $H_{\text{ph}} = \sum_{ks} \hbar\omega(k_s) \psi_{ks}^\dagger \psi_{ks}$. ψ_{ks}^\dagger and ψ_{ks} are the phonon creation and annihilation operators for mode k and polarization s . In the TLS-phonon interaction (last term in Eq. (6)), $\epsilon_k^i = \sum_s \xi_{ks}(i) \psi_{ks} + \xi_{ks}^*(i) \psi_{ks}^\dagger$ represents the scalar strain field, where $\xi_{ks} = i \sqrt{\frac{\hbar\omega(k_s)}{2\rho v^2}} D_{ab}^i e_{ab}^s(k) e^{ikr_i}$ and $\gamma_i D_{ab}^i$ is the TLS elastic dipole moment with strength γ_i shown as an explicit factor. ρ is the density of the material and v is the speed of sound. $e_{ab}^s = \frac{1}{2}(\hat{k}_a \hat{e}_b^s + \hat{k}_b \hat{e}_a^s)$, \hat{k} is the unit wavevector and \hat{e} is the polarization unit vector. r_i denotes the position of the i^{th} TLS, and $\sigma_i^{x,z}$ are Pauli matrices. ε_i and Δ_i^o are the asymmetry energy and tunneling matrix element of the i^{th} TLS respectively.

Since the Hamiltonian Eq. (6) is at most quadratic in the phonon fields, integrating out the high frequencies amounts to a shift of the phonon coordinate (by completing the square). The trick is that this shift needs to be different for each state (i.e., the right and left well states), since the linear coupling has the opposite sign (because of the σ_i^z operator). This shifting of the coordinates is achieved by a unitary transformation (also known as a polaron transformation, see, e.g., [1]):

$$U = e^{-\frac{1}{\sqrt{V}} \sum_{ks,i} \sigma_i^z \gamma_i (\xi_{ks}(i) \psi_{ks} - \xi_{ks}^*(i) \psi_{ks}^\dagger) / (\hbar\omega(k_s))} \quad (7)$$

The operators in the old and new basis are related by

$$\begin{aligned} \phi_{ks} &\equiv U^{-1} \psi_{ks} U \\ S^n &\equiv U^{-1} \sigma^n U \end{aligned} \quad (8)$$

where $n = x, z$. Explicitly:

$$\psi_{ks} = \phi_{ks} - \sum_i S_i^z \frac{\gamma_i \xi_{ks}^*(i)}{\hbar\omega(k_s)} \quad (9)$$

where we have used the relation $[f(A), B] = [A, B] \frac{\partial f}{\partial A}$, the fact that σ_z commutes with U , and:

$$\sigma_i^x = \cosh(2\alpha) S_i^x + \sinh(2\alpha) S_i^z S_i^x \quad (10)$$

where $\alpha = \frac{1}{\sqrt{V}} \sum_{ks} \gamma_i (\xi_{ks}(i) \psi_{ks} - \xi_{ks}^*(i) \psi_{ks}^\dagger) / (\hbar\omega(k_s))$. Using:

$$e^{A+B} = e^A e^B e^{-\frac{1}{2}[A,B]} \quad (11)$$

which is valid if $[A, [A, B]] = [B, [A, B]] = 0$, we can normal order the Bose operators in Eq.(10):

$$\begin{aligned} : \cosh(2\alpha) : &= e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \tilde{\xi}_{ks}} (e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \phi_{ks}^\dagger} e^{\sum_{ks} 2\tilde{\xi}_{ks} \phi_{ks}} \\ &+ e^{\sum_{ks} 2\tilde{\xi}_{ks}^* \phi_{ks}^\dagger} e^{-\sum_{ks} 2\tilde{\xi}_{ks} \phi_{ks}}) \end{aligned} \quad (12)$$

Likewise:

$$\begin{aligned} : \sinh(2\alpha) : &= e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \tilde{\xi}_{ks}} (e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \phi_{ks}^\dagger} e^{2\tilde{\xi}_{ks} \phi_{ks}} \\ &- e^{\sum_{ks} 2\tilde{\xi}_{ks}^* \phi_{ks}^\dagger} e^{-\sum_{ks} 2\tilde{\xi}_{ks} \phi_{ks}}) \end{aligned} \quad (13)$$

where $\tilde{\xi}_{ks} = \gamma_i \xi_{ks} / \sqrt{V} \hbar\omega(k_s)$. Thus $: \cosh(2\alpha) : = e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \tilde{\xi}_{ks}} (1 - 4 \sum_{ks} \tilde{\xi}_{ks}^* \tilde{\xi}_{ks} \phi_{ks}^\dagger \phi_{ks} + 4 \sum_{ks} \tilde{\xi}_{ks}^2 \phi_{ks}^2 + 4 \sum_{ks} (\tilde{\xi}_{ks}^*)^2 (\phi_{ks}^\dagger)^2 + O(\phi^3))$ and $: \sinh(2\alpha) : = e^{-\sum_{ks} 2\tilde{\xi}_{ks}^* \tilde{\xi}_{ks}} (4 \sum_{ks} \tilde{\xi}_{ks}^* \tilde{\xi}_{ks} \phi_{ks} - 4 \sum_{ks} \tilde{\xi}_{ks}^* \phi_{ks}^\dagger + O(\phi^3))$.

Since the phonons are much faster than TLS tunneling rate, we need only keep zeroth order in the phonon fields. This gives:

$$\sigma_i^x = e^{-\frac{1}{V} \sum_{ks} 2\gamma_i^2 \xi_{ks}^* \xi_{ks} / (\hbar\omega(k_s))^2} S_i^x \quad (14)$$

Evaluating the sum in the exponent yields $\frac{\gamma_i^2}{\gamma_o^2} (1 - \frac{vk_m}{\omega_D})^2$ where $\gamma_o = \frac{\sqrt{2}}{3} \sqrt{\rho v^2 v_o \hbar\omega_D}$. Since vk_m is less than one tenth of ω_D in the present model, k_m can be set to zero in this expression. Finally, substituting Eqs. (8) and Eq. (14) into Eq. (6) as well as relabelling S_i^n by σ_i^n and ϕ_k by ψ_k gives the result:

$$\begin{aligned} H &= H_{\text{ph}, k < k_m} + \frac{1}{2} \sum_i \varepsilon_i \sigma_i^z + \frac{1}{2} \sum_i \Delta_{\text{max}}^o e^{-\gamma_i^2 / \gamma_o^2} \sigma_i^x \\ &+ \frac{1}{\sqrt{V}} \sum_{k < k_m, i} \gamma_i \epsilon_k^i \sigma_i^z + \frac{1}{2} \sum_{i \neq j} \sigma_i^z J_{ij} \sigma_j^z \end{aligned} \quad (15)$$

with J_{ij} given by:

$$J_{ij} = \frac{1}{V} \sum_k \frac{\gamma_i \xi_k^*(i) \gamma_j \xi_k(j)}{\hbar\omega(k)} \quad (16)$$

POOR MAN'S MEAN FIELD APPROACH TO ESTIMATING THE DENSITY OF STATES n_o

We review here the naive random mean field used for calculating the TLS density of states \tilde{n}_o . We note that this approach gives incorrect results for all quantities involving correlation functions. However, for the purpose of computing the local effective field for low energies of a TLS, it should be fine due to the large distribution of fields obtained. (It should also be noted that at very low energies, a gap in the density of states must develop for stability reasons [2]. This is analogous to the formation of the Coulomb gap that arises in electron glasses [3–5]. This gap occurs well below the energies and hence, temperatures considered in this work.)

The starting point is a Hamiltonian of the form:

$$H = \frac{1}{2} \sum_{i \neq j} J_{ij} \sigma_i^z \sigma_j^z \quad (17)$$

where

$$J_{ij} = \frac{\gamma_i x_i \gamma_j x_j}{\rho v^2 r_{ij}^3} \quad (18)$$

γ_i are the magnitude of the couplings between phonons and TLS. These couplings are distributed between 0 and some maximum value γ_{max} such that $P(\gamma^2)$ equals a constant. x_i is meant as a simplified version of full TLS elastic dipole tensor with random orientation. The distribution of x_i is such that $\langle x_i \rangle = 0$ and $\langle x_i^2 \rangle = 1$. The requirement $\langle x_i^2 \rangle = 1$ comes from the fact that experiments only measure averages over the square of the elastic dipole moment's orientation. r_{ij} is the distance between two TLS randomly distributed at positions r_i and r_j . σ_i^z is the z th Pauli matrix.

In the spirit of the Weiss mean field approach, the local field is written as:

$$h_i = \sum_j J_{ij} m_j \quad (19)$$

where m_i is the ‘‘magnetization’’ induced by the field h_i . $m_i = \tanh(\beta h_i)$ where β is the inverse temperature $1/k_B T$. If the distribution of values for h_i is large, then at low temperatures, most TLS will be saturated (with $m_j = \pm 1$), and those TLS that are not saturated will tend to be isolated and thus will not contribute to the sum. We assume that h_i has a Gaussian distribution $p(h_i) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-h_i^2/2\sigma^2}$. All we need is the variance of this distribution:

$$\sigma^2 = \langle (h_i)^2 \rangle = \sum_{jk} \langle J_{ij} J_{ik} m_j m_k \rangle \quad (20)$$

where $\langle \dots \rangle$ denotes averages over the disorder. We next assume that the J 's and m 's are uncorrelated and use the fact that $m_i^2 = 1$ to get:

$$\sigma^2 = \sum_j \langle J_{ij}^2 \rangle \quad (21)$$

Using the definition for J_{ij} in Eq. (18) yields:

$$\sigma^2 = \left(\frac{1}{\rho v^2} \right)^2 \left\langle \sum_j \frac{\langle x_i^2 \rangle \langle x_j^2 \rangle \langle \gamma_i^2 \rangle \langle \gamma_j^2 \rangle}{r_{ij}^6} \right\rangle_{r_j} \quad (22)$$

where $\langle \dots \rangle_{r_j}$ is an average over all possible positions of the TLS. Now $\langle x_i^2 \rangle = 1$ and $P(\gamma_i^2) = 1/\gamma_{max}^2$ with $\gamma_i \in [0, \gamma_{max}]$, so $\langle \gamma_i^2 \rangle = \gamma_{max}^2/2$. Therefore:

$$\sigma^2 = \left(\frac{\gamma_{max}^2}{2\rho v^2} \right)^2 \left\langle \sum_j \frac{1}{r_{ij}^6} \right\rangle_{r_j} \quad (23)$$

Let us define R_o such that $\frac{4}{3}\pi R_o^3$ is the volume V_o per TLS and replace the average of the sum by an integral: $\langle \sum_i \dots \rangle_{r_i} \rightarrow \frac{1}{V_o} \int_{R_o}^{\infty} 4\pi r^2 \dots dr$. Thus:

$$\left\langle \sum_j \frac{1}{r_{ij}^6} \right\rangle_{r_j} \approx \frac{1}{V_o} \int_{R_o}^{\infty} \frac{4\pi r^2}{r^6} dr = \left(\frac{4\pi}{3V_o} \right)^2 \quad (24)$$

The variance is then $\sigma^2 = \left(\frac{\gamma_{max}^2}{2\rho v^2} \right)^2 \left(\frac{4\pi}{3V_o} \right)^2$. For the small values of h_i that we are interested in, we can set $h_i = 0$ in the Gaussian distribution: $P(h_i) \approx 1/\sqrt{2\pi\sigma^2} \approx \frac{\rho v^2}{\gamma_{max}^2} \frac{3V_o}{(2\pi)^{3/2}}$. Finally, to obtain the density of states n_o , we have to divide by the volume per TLS, V_o . We also need to remember that in the TLS model, the energy asymmetry is taken to be positive, so we must ‘fold’ the negative values onto the positive ones, which gives a factor 2. The result is then:

$$\tilde{n}_o = \frac{\rho v^2}{\gamma_{max}^2} \frac{6}{(2\pi)^{3/2}} \approx \frac{\rho v^2}{3\gamma_{max}^2}. \quad (25)$$

where we have approximated $\frac{6}{(2\pi)^{3/2}} \sim 0.38\dots$ by $1/3$.

ESTIMATING γ_{max} USING STABILITY CRITERION

In this section we present various ways to estimate γ_{max} . The maximal value of a dipole moment is governed by what are essentially stability arguments. The elastic medium has to be able to handle the stress from the local dipole. It then boils down to figuring out what is the maximal atomic strain allowed. The first step is to estimate the strain produced at the atoms neighboring a given dipole of strength γ at the origin. That is easy to do. The result is:

$$\delta^2 = \frac{3\gamma^2}{2(\rho v^2 v_o)^2} \quad (26)$$

δ represents a typical strain created in the surrounding medium by the dipole. The question has thus shifted to figuring out what the maximum δ should be. There are a few options: (In what follows, any numerical estimate is done for SiO_2 .)

1. The elastic medium becomes unstable well before the average displacement of its constituents reaches the lattice spacing. So assume that the strain δ has some fixed value $\alpha_1 \sim 0.2 - 0.25$. Then $\gamma_{max} = \alpha_1 \sqrt{\frac{2}{3}} \rho v^2 v_o$. For C to be $\sim 10^{-4}$, then α_1 needs to be $1/4$. This is quite a reasonable number.
2. A better estimate might be obtained from a ‘‘reverse Lindemann criterion’’. The maximum strain fluctuations that the glass can sustain occur at the glass transition temperature T_g . So δ should be set as a multiple of these maximal strain fluctuations which are given by:

$$\langle e^2 \rangle_{T > T_D} = \frac{3\hbar\omega_D}{\rho v^2 v_o} \frac{T_g}{T_D} \quad (27)$$

where e is the strain field. Taking $\langle e^2 \rangle_{T > T_D}$ as the maximum value for δ^2 gives the following estimate

Glass	ρ [kg/m ³]	v [m/s]	v_o [Å ³]	T_g [K]	T_D [K]	C_{exp} 10 ⁴	\bar{P}_{exp} [10 ⁴⁵ /J·m ³]	C_g 10 ⁴	\bar{P}_g [10 ⁴⁵ /J·m ³]	C_e 10 ⁴	\bar{P}_e [10 ⁴⁵ /J·m ³]
SiO ₂	2200	4163	45.3	1473	348	2.9	0.8	2.8	0.9	2.9	1.0
BK7	2510	4195	41.8	836	360	3.3	1.1	9.2	3.1	2.5	0.9
SF4	4780	2481	40.7	693	215	2.75	1.1	3.8	1.8	0.9	0.4
SF57	5510	2327	55.2	687	182	2.98	1.0	3.6	1.8	0.9	0.5
SF59	6260	2131	40.2	635	185	2.78	1.0	3.7	2.2	1.0	0.6
V52	4800	2511	61.1	593	190	4.9	1.7	5.2	2.3	0.8	0.3
BALNA	4280	2569	39.9	520	224	4.8	2.1	7.6	3.6	1.2	0.6
LAT	5250	3105	68.2	723	226	3.7	1.4	5.0	1.7	0.3	0.1
Zn-glass	4240	2580	45.9	570	215	3.6	2.2	7.2	3.8	2.0	1.1
PMMA	1180	1762	138.4	374	101	3.7	0.6	3.7	1.4	2.9	1.1

TABLE I. Predictions for C and \bar{P} for dielectric glasses using two ways of estimating γ_{max} as described in the text. Data from [6] and references therein.

for γ_{max} :

$$\gamma_{max} = \alpha_2 \sqrt{2\rho v^2 v_o k_b T_g} \quad (28)$$

where α_2 is a proportionality factor of order 1. This estimate for γ_{max} gives $C \sim 10^{-4}$ with $\alpha_2 = 1$, but to fit C to the value for SiO₂ requires $\alpha_2 = 1.5$. It is noteworthy that Lubchenko et al. [7] have obtained the same expression (without the factor 2) for γ_{max} based on a slightly different elastic stability argument.

- The expression for $\langle e^2 \rangle$ was written on purpose in an odd way proportional to $\hbar\omega_D/T_D$. The reason is that the quantity that multiplies T_g/T_D is just 8/3 times the expression for $\langle e^2 \rangle_{T=0}$. It thus seems natural to think that an equally good approximation would be to approximate δ as a multiple of $\sqrt{\langle e^2 \rangle_{T=0}}$. In this case, the estimate for γ_{max} becomes:

$$\gamma_{max} = \alpha_3 \sqrt{\rho v^2 v_o \hbar\omega_D} \quad (29)$$

In this case, γ_{max} is directly proportional to γ_o and C becomes entirely material independent. α_3 needs to be about 5 in order to get $C \sim 10^{-4}$.

- Another way to estimate γ_{max} is to use Alexander's elasticity theory [8] of disordered systems. In his model, internal stresses permeate the glass and, in particular, negative bond tension, i.e., compression, can occur that will lead to buckling. In this case, the energy of a bond of length R is $\frac{T}{2R}u^2 + \frac{k}{4R^2}u^4$ where u is the deviation from the normal length R , T is the bond tension and k is the elastic spring constant which is $\omega_D\rho v_o$. When the tension T is negative (corresponding to compression), the bond buckles, resulting in $u = \sqrt{\frac{2TR}{k}}$. Using $\omega_D = vk_D$ and $k_D = (\frac{9\pi}{2})^{1/3}/R$, the strain e can be expressed as $e = (\frac{2}{9\pi})^{1/3} \sqrt{\frac{2TR}{\rho v^2 v_o}}$. Equating TR to γ_{max} , and

e to δ , i.e., stating that the elastic medium should support the strain e , we obtain

$$\gamma_{max} = \frac{4}{3} \left(\frac{2}{9\pi}\right)^{2/3} \rho v^2 v_o \quad (30)$$

Again, we should assume that there is a factor of proportionality of order one. This is the expression used in the main text.

Table I lists the predictions for \bar{P} and C using the glass transition estimate, \bar{P}_g and C_g , with $\alpha_3 = 1.5$, as well as the prediction using the estimate from stability, \bar{P}_e and C_e . In each case, both \bar{P} and C show fluctuations consistent with the experimental data. The values derived from the estimate based on the glass transition temperature are closer to the experimental values but also show more significant deviations in some cases, e.g., BK7 and Zn-glass. All these various estimates yield the correct order of magnitude for C and \bar{P} which underscores the robustness of our approach.

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