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Herve M. Carruzzo and Clare C. Yu

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Why phonon scattering in glasses is universally small at low temperatures

Herve M. Carruzzo and Clare C. Yu
University of California, Irvine, Irvine, CA 92697
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We present a novel view of the standard model of tunneling two level systems (TLS) to explain the puzzling universal value of a quantity, $C \sim 3 \times 10^{-4}$, that characterizes phonon scattering in glasses below 1 K as reflected in thermal conductivity, ultrasonic attenuation, internal friction, and the change in sound velocity. Physical considerations lead to a broad distribution of phonon-TLS couplings that (1) exponentially renormalize tunneling matrix elements, and (2) reduce the TLS density of states through TLS-TLS interactions. We find good agreement between theory and experiment for a variety of individual glasses.

Amorphous solids are ubiquitous and technologically important, yet they still hold mysteries such as the universal values of phonon scattering. Below 1 K, phonon scattering reflected in the thermal conductivity (scaled with natural units) [1], the internal friction (in the relaxation regime) [2], the change in the sound velocity, and the resonant ultrasonic attenuation [3] are quantitatively very similar, regardless of the insulating glassy material. This universality is quite surprising, and, though it has been known for quite some time, remains a puzzle. Why does phonon scattering in these materials show such a lack of sensitivity to their composition and structure?

The standard model of tunneling two level systems (TLS) [4, 5] qualitatively describes the behavior of glasses below 1K. It postulates the existence of *independent* entities that tunnel between the two minima of a double well potential with a wide distribution of tunneling matrix elements and energy asymmetries. However, this model does not quantitatively explain the measurements cited above that depend on the coupling of phonons to tunneling two level systems (TLS). In particular, these measurements all find a rather universal value for a dimensionless coupling constant, C , given by

$$C = \frac{\bar{P}\gamma^2}{\rho v^2} \quad (1)$$

where \bar{P} is the density of states of tunneling entities, γ is the strength of their coupling with phonons, ρ is the mass density of the material and v is the sound velocity given by $v^{-3} = \frac{1}{3} \sum_s v_s^{-3}$, where v_s is the sound velocity for polarization s . Within the TLS model, the internal friction Q^{-1} is given by $Q^{-1} = \frac{\pi}{2} C$, while the change in sound velocity in the relaxation regime is $\Delta v/v = -\frac{1}{2} C \ln(T/T_o)$ where T_o is an arbitrary reference temperature. Finally, the scaled thermal conductivity [1] is universal because it depends on ratio of the mean free path ℓ of a phonon to its wavelength λ in the following way: $\ell/\lambda = 1/(2\pi^2 C)$. Measurements of these quantities find values of C between 2×10^{-4} and 5×10^{-4} . The universal value of C is quite surprising given that the parameters entering C are nominally independent and vary significantly from glass to glass. A universal value for this quantity thus implies a degree of coincidence that strains credulity, as noted by

Leggett [6]. Another surprise is the energy scale of the coupling between the sound waves and the TLS which is about 1 eV in insulating glasses, an energy scale that does not match any other in the problem.

Yu and Leggett [7] (YL) made the first attempt to understand this coincidence. They assumed that phonon mediated interactions between TLS dominate the physics. While their mean field scenario did indeed explain the universality, the predicted universal value for C is of order 1, whereas the observed value is 10^{-4} . This failure of a simple mean field theory approach is rather surprising. What determines the value for C ? A variety of rather complicated approaches have been proposed to resolve this question [8–11]. These include renormalization group approaches [8, 11], a random first order phase transition associated with the glass transition [9], two different types of TLS that couple differently to phonons [12], and vibrational instability of harmonic oscillators associated with the boson peak [10]. These models have been able to arrive at the right order of magnitude for C , but the use of a variety of assumptions and estimates have precluded the ability to predict the value of C for different specific glasses.

We propose a novel explanation based on three aspects implicit in the standard TLS model that were ignored in the original model [4, 5] and were only partially considered subsequently. First, the coupling between phonons and TLS implies that the TLS can interact with each other [7]. Second, this coupling produces an exponential renormalization of the tunneling matrix element due to phonon overlap between the two wells (a kind of polaron effect) [13]. Third, phonons actually couple to the *difference* between the elastic dipole moments in the two wells. If the elastic dipole moment in each well has a random orientation, the difference will also be random and will vary from TLS to TLS, leading to a broad distribution of couplings γ . Our model explains the universal value of C as well as the observed (~ 1 eV) value of the TLS-phonon coupling at low frequencies.

We begin by introducing the model for a set of entities that can tunnel between two states, e.g., “right” and “left” well, randomly distributed in an elastic medium and interacting with phonons:

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

where the free phonon Hamiltonian $H_{\text{ph}} = \sum_{ks} \hbar \omega(ks) \psi_{ks}^\dagger \psi_{ks}$. ψ_{ks}^\dagger and ψ_{ks} are the phonon creation and annihilation operators for wavevector k and polarization s , and $\omega(ks)$ is the phonon dispersion relation [14]. ε_i is the energy asymmetry between the two wells and Δ_i^o is the tunneling matrix element of the i^{th} TLS. In the TLS-phonon interaction (last term in Eq.(2)), $\epsilon_k^i = \sum_s \xi_{ks}(i) \psi_{ks} + \xi_{ks}^*(i) \psi_{ks}^\dagger$ represents the scalar strain field, where $\xi_{ks}(i) = i \sqrt{\frac{\hbar \omega(ks)}{2 \rho v^2}} (\sum_{ab} D_{ab}^i e_{ab}^s(k)) e^{ikr_i}$ 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. $e_{ab}^s(k) = \frac{1}{2} (\hat{k}_a \hat{e}_b^s + \hat{k}_b \hat{e}_a^s)$, \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. For simplicity we ignore the distinction between transverse and longitudinal TLS-phonon couplings.

Our model differs from the standard one in the distributions of the parameters ε_i , Δ_i^o and γ_i . In the standard model, the energy asymmetry between the right and left wells, ε_i , and the tunneling matrix element Δ_i^o are assumed to vary from site to site such that the probability per unit volume to find a TLS with a given value of ε_i and Δ_i^o is:

$$P(\varepsilon, \Delta^o) = \bar{P} / \Delta^o \quad (3)$$

with $0 < \varepsilon < \varepsilon_{\text{max}}$, and $\Delta_{\text{min}}^o < \Delta^o < \Delta_{\text{max}}^o$. $\bar{P} = n_o / (\varepsilon_{\text{max}} \ln(\Delta_{\text{max}}^o / \Delta_{\text{min}}^o))$, and n_o is the density of TLS per unit volume. Typically, \bar{P} is an adjustable parameter fitted to experiments. The distribution of tunneling parameters is assumed to arise from a flat distribution of the tunneling barrier heights [4]. The coupling to phonons is chosen to be identical for all TLS, i.e., $\gamma_i = \gamma$, and is used as a fitting parameter. The final assumption is that the interaction term between TLS and phonons is small, permitting the use of perturbation theory to compute the quantities above [15].

In contrast to the standard model, we note that the phonon-TLS interaction implies that TLS can interact with one another via the strain field [7, 13]. To effect this, we integrate out the phonons with energies higher than the tunneling matrix elements, i.e., $\hbar v k_m > \Delta_o^{\text{max}} \sim 10$ K. (The precise value of Δ_o^{max} is not critical since it ultimately only enters logarithmically.) The result of the integration is [16]:

$$H = H_{\text{ph}, k < k_m} + \frac{1}{2} \sum_i \varepsilon_i \sigma_i^z + \frac{1}{2} \sum_i \Delta_{\text{max}}^o e^{-\gamma_i^2 / \gamma_o^2} \sigma_i^x + \frac{1}{\sqrt{V}} \sum_{k < k_m, i} \gamma_i \epsilon_k^i \sigma_i^z + \frac{1}{2} \sum_{i \neq j} \sigma_i^z J_{ij} \sigma_j^z \quad (4)$$

where

$$\gamma_o = \frac{\sqrt{2}}{3} \sqrt{\rho v^2 v_o \hbar \omega_D} \quad (5)$$

and v_o is a unit volume of the chemical formula unit of the glass as would be used to define a Debye frequency, ω_D [1]. The third term shows that the tunneling matrix element has been renormalized downward by a polaron effect [13] in which the overlap of the phonon wavefunctions between potential wells exponentially reduces the effective tunneling. Unlike the standard model where the tunneling depends on the WKB exponent that incorporates the barrier height, in our model the exponent of the tunneling matrix element depends on the TLS-phonon coupling γ_i . The fourth term contains the remaining TLS-phonon interaction which is weak and can be treated with perturbation theory as in the standard model. The last term shows that a TLS-TLS interaction term has been generated which is quite complex due to the tensorial nature of elastic dipole moments. Following YL, J_{ij} is simplified to $J_{ij} = \frac{1}{\rho v^2} s_i \gamma_i s_j \gamma_j / r_{ij}^3$ where r_{ij} is the distance between TLS i and j , and $s_i = \pm 1$ is a spin representation of the orientation of the elastic dipoles (see [17] for the full expressions).

To motivate our second assumption, let us review why the YL scenario failed to give the correct value of C . YL assumed $\gamma_i = \gamma \forall i$ so that $J_{ij} = \frac{\gamma^2}{\rho v^2} s_i s_j / r_{ij}^3$ in Eq. (4). The $1/r^3$ interactions together with a simple random mean field theory produces a density of states independent of the original density of TLS given by:

$$\bar{P} \approx \frac{\rho v^2}{\gamma^2} \quad (6)$$

If we plug this into the expression for C , Eq. (1), we get $C \sim 1$; universal but 4 orders of magnitude too large. In addition, \bar{P} is two orders of magnitude too large compared to the density of states from specific heat measurements. \bar{P} could be reduced by increasing the strength of the interactions γ but this will not solve the $C \sim 1$ problem if the same value of γ controls the attenuation of low frequency phonons.

To fix this problem, we note that contrary to the standard model, the coupling between TLS and phonons should actually have a broad distribution. To see why, note that in Eq. (2), the TLS elastic dipole moment couples to the phonons via a σ^z term, so that it is the *difference* between the dipole moments in the right and left wells of the TLS that couples to the strain field. Assume now that the dipole moment in each well has the same magnitude (γ_{max}), but a different orientation. The difference between the dipole moments in the two wells is itself a dipole moment with magnitude γ . For two randomly oriented vector dipoles, the magnitude of the difference vector will have a uniform distribution $P(\gamma^2) = 1/\gamma_{\text{max}}^2$ [16]. The case of elastic tensor dipoles is more difficult

but leads to a similar distribution, albeit with an increase in probability for large values of γ (see [16]).

The maximum possible value of the coupling, γ_{max} , is taken to be larger than the γ inferred from acoustic experiments, leading to stronger interactions between TLS and hence, a lower density of states \bar{P} . In addition, these random TLS-phonons couplings produce a wide distribution of tunneling amplitudes due to the factor, $\exp(-\gamma_i^2/\gamma_o^2)$, multiplying Δ_o^{max} . TLS with large values of γ_i have very small tunneling rates so they will not be seen, e.g., in internal friction measurements. (The choice of a single value Δ_o^{max} for the tunneling matrix element prefactor is based on the view that while glasses have no obvious order, any region is very similar to any other [18].)

The next step is to approximate the TLS-TLS interaction term in Eq. (4) using a poor man's random mean field theory [16]. The effective field felt by a given TLS is the sum of the fields from all the surrounding TLS, most of which are 'frozen' at low temperature: $\varepsilon_i \equiv \varepsilon_i(J_{ij} = 0) + \sum_{j \neq i} J_{ij} \sigma_j^z$. Since we assume that the local asymmetry variations are small compared to the interactions between TLS, we can neglect the $\varepsilon_i(J_{ij} = 0)$ term so that the asymmetry energy ε_i arises entirely from interactions. If we assume the σ_j^z to be uncorrelated, the sum will have approximately a Gaussian distribution with zero mean (the J_{ij} are equally positive and negative). The variance is given by: $Var(\varepsilon_i) \approx (\frac{\gamma_{max}^2}{2\rho v^2})^2 (\frac{4\pi}{3V_o})^2$ where V_o is the average volume per rearranging region. For energies small compared to the variance, the Gaussian distribution is essentially flat and thus the probability of finding a TLS with a given (small) ε_i is $P(\varepsilon_i) \approx 3\rho v^2 V_o / (2\pi)^{2/3} \gamma_{max}^2$. The density of states per unit energy and unit volume is then simply [16]:

$$\tilde{n}_o \approx \rho v^2 / 3\gamma_{max}^2 \quad (7)$$

With this random mean field approximation, the Hamiltonian in Eq. (4) reduces to that of an independent TLS model. The effect of interactions between TLS has been subsumed into n_o , the distribution for the energy asymmetry per unit volume which is now expressed in terms of material parameters. Together with the distribution of γ , $P(\gamma^2) = 1/\gamma_{max}^2$, and the expression for Δ^o , $\Delta^o \equiv \Delta_o^{max} e^{-\gamma^2/\gamma_{max}^2}$, we have an independent TLS model quite similar to the standard model. The key difference is that γ controls the value of the tunneling matrix element Δ^o in addition to the coupling between TLS and phonons. It is more convenient to change variables from $(\varepsilon$ and $\gamma)$ to $(\varepsilon$ and $\Delta^o)$. This gives:

$$P(\varepsilon, \Delta^o) = \frac{\bar{P}}{\Delta^o} \quad (8)$$

where

$$\bar{P} = \frac{1}{3} \frac{\rho v^2}{\gamma_{max}^2} \left(\frac{\gamma_o}{\gamma_{max}} \right)^2 \quad (9)$$

γ is now given in terms of Δ^o by:

$$\gamma = \gamma_o \ln^{1/2} \left(\frac{\Delta_o^{max}}{\Delta^o} \right) = \frac{\sqrt{2}}{3} \sqrt{\rho v^2 v_o \hbar \omega_D} \ln^{1/2} \left(\frac{\Delta_o^{max}}{\Delta^o} \right) \quad (10)$$

Let us bring all the pieces together and write our effective non-interacting Hamiltonian:

$$H = H_{\text{phonon}} + \frac{1}{2} \varepsilon \sigma^z + \frac{1}{2} \Delta^o \sigma^x + \sum_{k < k_m} \gamma(\Delta^o) \epsilon_k \sigma^z \quad (11)$$

where γ is an explicit function of Δ^o given by Eq. (10) and the distribution of parameters is given by Eq. (8). Experimental quantities of interest should be computed with these expressions, though it is easier to do so by simplifying Eq. (10) for γ as follows. With $\Delta_o^{max} \sim 10$ K, $\ln^{1/2}(\frac{\Delta_o^{max}}{\Delta^o})$ in γ varies from about 1.5 for $\Delta^o = 1$ K to about 5 for $\Delta^o = 5 \times 10^{-11}$ K (which corresponds to an oscillation time of 1 second). Since Δ^o dictates which TLS can respond on an experimental timescales, γ can be replaced by $\gamma_{eff} = \alpha \gamma_o$ with α equal to some constant in the range from 1 to 5. For concreteness, we will use $\alpha = 2.5$.

With this simplification, we can now use Eq. (1) to calculate C with $\gamma = \gamma_{eff}$ and Eq. (9) for \bar{P} to obtain:

$$C = \bar{P} \frac{\gamma_{eff}^2}{\rho v^2} = \frac{\alpha^2}{3} \left(\frac{\gamma_o}{\gamma_{max}} \right)^4 \quad (12)$$

The last step is to estimate γ_{max} which requires going beyond elasticity. On general grounds, we expect $\gamma_{max} \sim a \rho v^2 v_o$ with $a < 1$ being a material independent constant [19]. Using elastic stability criteria in disordered systems yields a better estimate [20]:

$$\gamma_{max} = \frac{4}{3} \left(\frac{2}{9\pi} \right)^{2/3} \rho v^2 v_o \sim 0.23 \rho v^2 v_o \quad (13)$$

This and other ways to estimate γ_{max} are further discussed in the Supplemental Material [16].

Table I shows the values of C obtained from Eq. (12) using Eq. (13) for γ_{max} for the insulating glasses for which we have all the required data. We have used $k_b T_D = \hbar \omega_D = \hbar v (\frac{6\pi^2}{v_o})^{1/3}$. v_o is obtained from the material's chemical formula (see ref. 16 in [3]) using $v_o = M/N_A \rho$ where M is the molecular mass and N_A is Avogadro's number. The only independent parameters are ρ , v_l , v_t and v_o . The theory has no adjustable parameters.

While the overall comparison between theory and experiment are good, the discrepancies call for a discussion. First, we did not distinguish between longitudinal and transverse modes. Given that experimentally [3] the ratio $\gamma_l^2/v_l^2 \approx \gamma_t^2/v_t^2$ and that it is the ratio that matters for the TLS-phonon interaction, the errors from this approximation should not be large. In particular, this approximation cannot explain the large discrepancy for

Glass	ρ [kg/m ³]	v [m/s]	v_o [Å ³]	T_D [K]	$C_{exp} \cdot 10^4$	$C_{th} \cdot 10^4$
SiO ₂	2200	4163	45.3	348	2.9	2.9
BK7	2510	4195	41.8	360	3.3	2.5
SF4	4780	2481	40.7	215	2.75	0.9
SF57	5510	2327	55.2	182	2.98	0.9
SF59	6260	2131	40.2	185	2.78	1.0
V52	4800	2511	61.1	190	4.9	0.8
BALNA	4280	2569	39.9	224	4.8	1.2
LAT	5250	3105	68.2	226	3.7	0.3
Zn-glass	4240	2580	45.9	215	3.6	2.0
PMMA	1180	1762	138.4	101	3.7	2.9

TABLE I. C for dielectric glasses computed from Eq. (12). Data from [3].

LAT between C_{th} and C_{exp} because C_{th} for LAT is 10 times lower than $C_{th}=C_{exp}$ for SiO₂, even though the experimental difference between $\gamma_{l,t}$ and $v_{l,t}$ for the two materials is not large. A more likely source of the discrepancies is our estimate of the volume v_o of the molecular formula unit which enters into the Debye temperature and is not well defined. One possibility is to consider v_o as the one adjustable parameter of the theory.

In short, the broad spectrum of TLS-phonon couplings γ produces a distribution of tunneling parameters Δ^o , many with values too small to contribute to ultrasonic measurements due to the exponential dependence of the tunneling on γ^2 . The TLS that have tunneling amplitudes large enough to participate in ultrasonic experiments result in estimates of γ of order 1 eV. This observed energy scale for γ is consistent with $\gamma \sim \sqrt{\rho v^2 v_o \hbar \omega_D}$ from Eq. (10). For example, using values appropriate for SiO₂ ($\rho = 2200$ kg/m³, $v=4200$ m/s, $v_o = 45 \times 10^{-30}$ m³ and $\hbar \omega_D = 350K$), we find $\gamma \sim 0.57$ eV, in close agreement with the experimental values of γ between 0.65 and 1 eV [3]. On the other hand, \bar{P} is determined by the interaction between TLS, regardless of the amount of tunneling suppression. Eq. (9) shows that the scale of \bar{P} is dictated by $\rho v^2 v_o$ and γ_{max} , and hence is lower than what is found using Eq. (6) with the ultrasonic value γ_{eff} . This is why $Q^{-1} \sim 10^{-4}$ is so much smaller than in the original YL approach. It should be possible to experimentally probe the distribution of γ for TLS that couple to superconducting qubits and are altered by strain [22].

Finally, since our effective Hamiltonian in Eq. (11) and the form of $P(\varepsilon, \Delta^o)$ in Eq. (8) reduce to those of the standard TLS model, all the results of the standard model carry over with, at most, logarithmic temperature corrections of those quantities that depend on the TLS-phonon coupling since γ has a logarithmic dependence on Δ^o as shown in Eq. (10). In particular, the specific heat has the same temperature dependence as in the standard TLS model. The thermal conductivity κ at low tem-

peratures is limited by the scattering of phonons from TLS resulting in a logarithmic temperature correction: $\kappa \sim T^2/(1 + 2 \ln(\Delta_{max}^o/2k_B T))$.

Everything discussed so far applies for temperatures below 1K. Let us briefly discuss what happens above $k_B T = \Delta_{max}^o \sim 10$ K. The following estimate shows that the tunneling barrier height V is comparable to Δ_{max}^o . If we ignore the effect of phonons on tunneling, the bare tunneling matrix element is given in the WKB approximation by:

$$\Delta_{max}^o = \hbar \omega_D e^{-\sqrt{2M}Vd/\hbar} \quad (14)$$

Solving for V with $\hbar \omega_D \sim 350K$ yields $\sqrt{2\rho v_o}Vd/\hbar = \ln(35)$. Using the numbers for SiO₂ with the barrier height V in Kelvin and d in Å, we get $1.6\sqrt{V}d \sim \ln(35)$, which means that $V \sim 5K$ for $d = 1\text{Å}$ and $V \sim 20K$ for $d = 0.5\text{Å}$. Thus, it is plausible that the barrier height is in the 1-30K range which corresponds roughly to the temperature where there is the plateau in the thermal conductivity and the boson peak in the specific heat.

At temperatures much greater than the barrier height, thermal fluctuations make tunneling and the tunnel barrier irrelevant. So tunneling no longer reduces the density of states and thus, for $k_B T \gg V$, we have $\bar{P} = \tilde{n}_o = \frac{\rho v^2}{3\gamma_{max}^2}$. Δ^o decouples from γ and the relevant coupling to phonons is the average of γ^2 which is $\gamma_{max}^2/2$. Therefore, in this regime, $C = \bar{P}\langle\gamma^2\rangle/\rho v^2 = 1/6$ and is universal. The ratio of the mean free path to the wavelength becomes:

$$\frac{\ell}{\lambda} = \frac{1}{2\pi^2 C} \sim 0.3 \quad (15)$$

This is observed in the thermal conductivity in the temperature range above the plateau [1]. The intermediate temperature regime ($\sim 3 - 10$ K) corresponding to the plateau is very much material dependent and other processes come into play here [23].

In conclusion, we have elucidated aspects implicit in the standard TLS model that include strongly interacting TLS [6, 7, 13], exponentially renormalized tunneling matrix elements [13], and a heretofore unrecognized broad distribution of TLS-phonon couplings. This produces the correct order of magnitude for Q^{-1} and the coupling γ seen in acoustic experiments. Variations in the predicted values of Q^{-1} from material to material are only slightly larger than in experiments. At high temperatures, where tunneling is irrelevant, we predict $\ell/\lambda \sim 1$, consistent with thermal conductivity experiments.

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