"Tunneling Two-Level Systems" Model of the Low-Temperature Properties of Glasses: Are "Smoking-Gun" Tests Possible?

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ABSTRACT: Following a brief review of the "two-level (tunneling) systems" model of the low-temperature properties of amorphous solids ("glasses"), we ask whether it is in fact the unique explanation of these properties as is usually assumed, concluding that this is not necessarily the case. We point out that (a) one specific form of the model is already experimentally refuted and (b) that a definitive test of the model in its most general form, while not yet carried out, would appear to be now experimentally feasible.
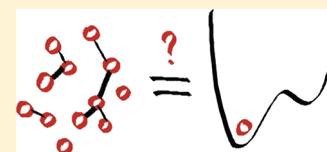
1. INTRODUCTION

Structurally amorphous materials ("glasses") constitute a substantial fraction of all terrestrial matter, yet our overall understanding of their behavior is not at all comparable to that which we have for crystalline matter. While both the transition to the glassy state and the thermodynamic and response behavior at ambient temperatures have some universal features that have been the subject of a huge amount of literature (see, e.g., refs 1 and 2 respectively), a particularly intriguing problem is posed by the behavior of glasses below about 1 K. Ever since the pioneering experiments of Pohl and co-workers in the early 1970s,[3] it has been recognized that in this regime, the properties of glasses not only are qualitatively different from those of crystalline solids but show a remarkable degree of universality. For example, almost without exception, the specific heat of an arbitrary (insulating) amorphous solid well below 1 K is approximately linear in T, the thermal conductivity is approximately quadratic in T, and the ultrasonic behavior is consistent with a Q factor that at zero temperature is independent of frequency and surprisingly large (more on this below).

Very soon after the original experiments,[3] a plausible model to explain them was published independently by Phillips[4] and by Anderson, Halperin, and Varma.[5] This model, which has become known as the "tunneling two-level system" (TTLS) model, postulates that because of the structurally amorphous nature of the system, there exist some entities (single atoms, groups of atoms, or in some cases even single electrons) that have available two nearly degenerate configurations and can tunnel between them (a more quantitative description is given in the next section). With a plausible choice of the distribution of the relevant parameters, the TTLS model can account for the qualitatively universal features noted above; furthermore, by analogy with other well-known examples of "two-state" systems in, for example, atomic physics and NMR, it naturally predicts various nonlinear phenomena such as acoustic saturation and echoes, all of which have been at least qualitatively verified by experiment.[6] The original model has undergone considerable elaboration over the last 40 years; in particular, an interesting series of papers[7] by Peter Wolynes and his collaborators has attempted to explain the generic existence of the postulated two-level systems (TLSs) as a natural consequence of processes occurring in the glass transition. These successes have persuaded the overwhelming majority of the relevant community that the TTLS model is the unique explanation of the low-temperature properties of glasses.

Viewed from the above perspective, the aim of the present article may seem rather quixotic: to ask whether the TTLS explanation is indeed as unique as it is usually taken to be and to try to suggest definitive ways of testing the model against a particular (loosely defined) class of alternative hypotheses. Obviously, to do this, it is necessary to define exactly what we mean by the "TTLS model" (and, in particular, what it excludes), and this will be done in section 2. In section 3, we sketch some reasons for skepticism about the model and introduce a (very generic) class of alternative hypotheses, and in section 4, we propose an experiment that we believe should discriminate unambiguously between this class and the TTLS model. Throughout, we confine ourselves to insulating glasses and concentrate on the linear ultrasonic properties, which we believe are among the most unambiguously predicted consequences of the model.

2. DEFINITION OF THE TTLS MODEL; SOME SIMPLE CONSEQUENCES

There exist in the literature a number of good accounts of the TTLS model and its most important experimental predictions.[6,8] For present purposes, a sufficient definition runs roughly as follows: For the purpose of calculating the low-energy states of glasses (those relevant to the equilibrium and near-equilibrium properties well below 1 K), an adequate effective Hamiltonian is of the form

$$\hat{H} = \hat{H}_{ph} + \hat{H}_{TLS} + \hat{H}_{int} \tag{1}$$

Here, $\hat{H}_{ph}$ is the phonon Hamiltonian, given in terms of phonon annihilation and creation operators a_i^+ and a_i by

$$\hat{H}_{ph} = \sum_{k\alpha} \hbar\omega_{k\alpha}a_{k\alpha}^+a_{k\alpha} \qquad \omega_{k\alpha} = c_\alpha|k| \tag{2}$$

where the sum over k has an upper cutoff k_{max} such that $\hbar c_i k_{max} \gg k_B \times 1$ K and where the sum over α runs over one longitudinal (l) and two transverse (t) branches; the sound velocities c_l and c_t are constant, reflecting the average isotropy of the glass at sufficiently large length scales. $\hat{H}_{TLS}$ is the part of the Hamiltonian attributable to the postulated TLSs and has the form

$$\hat{H}_{TLS} = \sum_i E_i b_i^+ b_i \tag{3}$$

where the operators b_i^+, and b_i are Pauli operators, that is, they satisfy the (anti)commutation relations

$$\{b_i, b_i^+\} = 1 \qquad [b_i, b_j^+] = 0 \text{ for } i \neq j \tag{4}$$

Thus the eigenvalues of the "occupation numbers" $n_i = b_i^+ b_i$ are 1 and 0 and can be specified independently. Finally, the part of the Hamiltonian expressing the interaction between the phonons and the TLS is of the form

$$\hat{H}_{int} = \sum_{\alpha\beta} \int \hat{e}_{\alpha\beta}(\mathbf{r})\hat{T}_{\alpha\beta}(\mathbf{r}) \, d\mathbf{r} \tag{5}$$

where $\hat{e}_{\alpha\beta} = (1/2)[(\partial\mu_\alpha/\partial x_\beta) + (\partial\mu_\beta/\partial x_\alpha)]$ is the phonon strain operator (u_α = mean "background material" displacement) and the stress operator $T_{\alpha\beta}$ is linear in the "spin" operators $\hat{\sigma}^{(i)}$ of the TLS

$$T_{\alpha\beta}(\mathbf{r}) = \sum_i g_{\alpha\beta\gamma}^{(i)}\hat{\sigma}_\gamma^i \delta(\mathbf{r} - \mathbf{r}_i) \tag{6}$$

where r_i is the position of the TLS i

Several points should be noted about the model described by eqs 1−6. First, it excludes what one might think are some obvious possibilities, most obviously, that the nonphonon part of the Hamiltonian cannot be written in the simple additive form (eq 3), but also (e.g.) that the stress tensor involves combinations of the $\hat{\sigma}^{(i)}$'s higher than linear, such as, $K_{ij}\hat{\sigma}^{(i)}\hat{\sigma}^{(j)}$. Second, at the present stage, it is really only a generic two-level model; the "tunneling" aspect enters only through the choice of the parameters E_i and $g_{\alpha\beta\gamma}^{(i)}$ (see below). Third, -and this is crucial to the subsequent discussion-it is implicit that the coupling term (eq 5) is small enough that in the calculation of any given physical property, it is adequate to confine oneself to the lowest order in $\hat{H}_{int}$ that gives "physically sensible" results (e.g., for the specific heat, zeroth order, and for the ultrasonic attenuation, first order). Finally, it should be noted that it is not necessarily assumed that the Hamiltonian (eq 1) is the one that would be appropriate at a fully microscopic level; bearing in mind that we are interested only in "low-energy" phenomena and hence by implication only in the behavior of phonons with wavelength $\lambda > hc/k_B T$ (which for $T \sim 1$ K is ~1000 Å), we may include the possibility that eq 1 is itself the output of some nontrivial renormalization procedure.

Equations 1−6 may be regarded as the most general definition of the TLS model of glasses. However, to extract quantitative physical predictions from it, one of course needs to specify the distribution of the parameters E_i and $g_{\alpha\beta\gamma}^{(i)}$ (including

any correlations between these two quantities). This is where the tunneling aspect comes in; in the original and simplest version of the model, the two states in question are conceived as corresponding to two spatially separated positions of the center of mass of a group of particles, so that for a given TLS i, in a basis in which these two positions are taken as the eigenstates of $\hat{\sigma}_z^{(i)}$, the Hamiltonian is

$$\hat{H}_i = \frac{1}{2}\begin{pmatrix} \epsilon_i & \Delta_i \\ \Delta_i & -\epsilon_i \end{pmatrix} \tag{7}$$

with ϵ_i as the offset between the potential energy in the two configurations and Δ_i as the matrix element for tunneling between them. Then, we evidently have

$$E_i = (\epsilon_i^2 + \Delta_i^2)^{1/2} \tag{8}$$

What is the distribution of the offsets ϵ_i and the tunneling matrix elements Δ_i? As regards the former, it seems very reasonable to take it to be simply constant (uniform distribution; from a microscopic point of view, there is nothing special about zero bias). As for the Δ_i, the default assumption would seem to be that the principal dependence comes from the WKB exponent in the expression for the tunneling amplitude; then, rather general arguments indicate that provided we are interested mainly in low-energy states, this exponent can again be considered to be uniformly distributed. With these rather weak assumptions, the distribution of parameters in the two-dimensional (ϵ, Δ) space is

$$\rho(\epsilon, \Delta) = \frac{const}{\Delta} \tag{9}$$

with the constant of course material-dependent. This gives a constant density of states as a function of the total energy splitting E

$$\rho(E) = \bar{P}_0 \tag{10}$$

We also need to consider the distribution of the TLS−phonon coupling constants $g_{\alpha\beta\gamma}^{(i)}$. The usual assumption in the literature is that (a) any difference between the l and t components of g lies only in the overall magnitude not the form, and (b) strain of either kind mainly affects the offset ϵ, so that any effect on the tunneling matrix element Δ may be neglected. If this is so, then in the energy representation for the Pauli matrices $\sigma^{(i)}$ (which we will use from now on unless explicitly otherwise stated), the matrix form of $g_{\alpha\beta\gamma}^{(i)}$ is identical to that of $\hat{H}_{TLS}$ in the original (position) representation, that is

$$g_{\alpha\beta\gamma}^{(i)} = \frac{g_{\alpha\beta}^{(i)}}{E_i}(\epsilon_i\delta_{z\gamma} + \Delta_i\delta_{x\gamma}) \tag{11}$$

with the parameters $g_{\alpha\beta}^{(i)}$ having some random distribution uncorrelated with Δ_i, ϵ_i, for example, a Gaussian with zero mean. When combined with 9, eq 11 says that the distribution of the off-diagonal (in the energy representation) terms in the $g^{(i)}$ is strongly peaked toward small values (an important qualitative feature of the model, which was already recognized in the earliest papers). We will refer to the distribution of parameters given by eqs 9 and 11 as the "canonical" distribution, and distinguish between the generic TLS model defined by eqs 1−6 and the TTLS model defined by eqs 1−6 plus this distribution.

3. WHAT COULD BE WRONG WITH IT? AN ALTERNATIVE SCENARIO

Let us start with a semiphilosophical point, which will probably be dismissed out of hand by most right-thinking physicists: the fact that the TTLS model appears to explain so adequately most of the properties of amorphous solids does not prove that it is correct! Technically, to argue that "Theory T predicts experimental result(s) E: we see E; therefore, theory T is correct" would of course be to commit the logical fallacy known as "affirming the consequent". Of course, this fallacy is formally committed every day in the pages of physics journals, and generally, people do not worry too much about it. Why not? We suspect because typically in these cases, there is an unspoken extra premise: "it is extremely unlikely that any theory other than T would predict experimental results E", which when combined with "we see E" indeed permits us to draw with high confidence the conclusion that T is correct. The question then arises; in the case of the use of the TTLS model to explain the behavior of glasses, is the unspoken extra premise correct? We believe that this is less obvious than it may seem.

First, let us consider the various kinds of nonlinear behavior in the ultrasonics (saturation, echoes, hole-burning, ...), which we suspect are in most people's minds the most convincing evidence in favor of the model. Setting aside the rather complicated question of the degree of quantitative agreement of the data with the TTLS model, one may ask whether the prediction of the qualitative features, that is, the mere existence of these phenomena, is unique to the model? We believe that the answer is no and indeed that it is probable that almost *any* model of the system energy levels and stress matrix elements other than the familiar harmonic oscillator one will suffice to reproduce them; calculations on simple one-particle systems tend to confirm this prejudice. A simple "hand-waving" argument goes as follows: in quantum mechanics, to obtain a response to a near-monochromatic field that *fails* to saturate, we need not just an infinite sequence of equidistantly spaced energy levels but also that the relevant matrix elements (in the case of ultrasound absorption, those of the stress tensor operator) between these levels increase sufficiently fast with energy. While we are accustomed to the fact that these conditions are both adequately satisfied for the simple harmonic oscillator, they are really more like the exception than the rule. In other words, it may be that what is "special" about the nonphonon modes in glasses is not that they are well-described as TTLS but that they are *not* well-described as simple harmonic oscillators!

Of course, there are other features of the experimental data whose prediction might at first sight seem unique to the TTLS model, such as time-dependent specific heats[9] and the equality, up to a numerical factor, of ultrasonic absorption in the low-temperature resonant and high-temperature (low-frequency) relaxation regimes (see section 4); while our prejudice is that a more general scenario should be able to reproduce at least the qualitative aspects of these phenomena, it must be said at once that a quantitative calculation is at present lacking.

However, it is not always appreciated that to a large extent the same situation exists for the established model. Indeed, while it is probably true that just about all of the existing experimental data are consistent with the generic TLS model, it can often be made so only by a choice of parameter distributions that violate the more restrictive assumptions in eqs 9 and 11, which were taken above to define the TTLS

version. A typical example is the specific heat; the experimental dependence on temperature is actually not linear but rather resembles a power law with exponent 1.2−1.3, and this behavior is clearly inconsistent with eq 10 and hence with the TTLS ansatz in eq 9. More generally, once one abandons the defining ansatz in eqs 9 and 11 of the TTLS version, the TLS model becomes so "squishy" that a cynic might be forgiven for anticipating that it can be made consistent with just about any experimental data.

A rather different kind of motivation for challenging the uniqueness of the TTLS model as an explanation of the low-temperature properties of glasses lies in the striking quantitative universality of some of these properties, in particular, the dimensionless (and surprisingly large, $\sim 10^4$) Q factor that describes the transverse ultrasonic absorption and frequency shift in the MHz−GHz range.[10,11] In the TTLS model, where the relevant expression is a product of four independent factors, this universality can be attributed only to a mind-boggling degree of coincidence; more generally, what it seems to suggest is that the low-temperature, long-wavelength properties of glasses emerge as a result of some rather nontrivial renormalization process that iterates to a fixed point. An initial attempt to implement such a renormalization scheme has been made in ref 12, where it has been shown that with two unproved but plausible generic ansatzes, it is possible to reproduce something like the observed value of Q (cf. also ref 13). Of course, this consideration does not in itself imply that the final Hamiltonian that emerges from the renormalization process does not itself possess the TLS structure described by eqs 1−6, but from an intuitive point of view, this seems rather unlikely.

With this motivation, let us consider, as a counterpoint to the TTLS model a scenario[12] that is about as far from it as possible without describing simply a collection of harmonic oscillators, namely, one in which while the phonon contribution to the total Hamiltonian is still given by eq 2, the "nonphonon" term $\hat{H}_{TLS}$ (eq 3) and the coupling term (eq 4) are replaced by random matrices in the many-body Hilbert space with some appropriately specified statistical properties. Needless to say, we do not necessarily expect that this model will be the correct one; the truth may well lie somewhere between it and the TLS version, but it serves as a convenient point of comparison.

We would like to emphasize strongly that the conjecture made in this paper is *not* that TLSs in amorphous solids never exist. Indeed, there are a few systems in which their existence may be established rather directly by experiment. A particularly striking example comes from the beautiful experiments of the Bordeaux group[14] on the spectroscopy of single terrylene molecules in polyethylene (PET), in particular, from the "spectral trail" experiments, which give rather direct evidence that in a substantial fraction of the molecules studied ($\sim 40\%$), the part of the environment that gives rise to the shifts in the resonance frequency jumps between two (and only two) discrete configurations. However, what is interesting is that to the extent that one takes these experiments as definitive evidence for the presence of TLS in bulk amorphous PET, by the same token, one will have to take the similar experiment of ref 15 as equally definitive evidence for their *absence* in amorphous solid toluene, and that consideration would suggest that it would be extremely interesting to investigate whether the subdegree thermal and acoustic behavior of toluene is that of a typical amorphous solid; while a negative answer would

strengthen the case for the TLS model, a positive one would definitively refute it.

Let us mention a few more systems in which either theory or experiment or both provides (or has been thought to provide) strong arguments for the presence of TLS. (1) It has long been believed that the behavior of KBr−KCN mixtures can be satisfactorily explained[16] in terms of TLS associated with the two possible orientations of the KCN complex. (2) In the disordered oxide barriers that are nowadays often used in Josephson junctions, there is rather direct evidence that there exist TLS carrying electric dipole moments, which moreover can be tuned by the application of external strain.[17] What is interesting is that rather frequently, these systems turn out to have a value of Q^{-1} considerably smaller than the "universal" figure of $\sim 3 \times 10^{-4}$. Our conjecture, therefore, would be that for these comparatively rare cases, the renormalization process discussed in ref 12 is ineffective, while for the majority of cases, it takes place and leads not only to the universal (maximum allowed) value of Q^{-1} but also to a structure of the output many-body levels, which is in general of non-TLS form. (3) Finally, it may be claimed that the isotope effects observed[18] in (natural or deuterated) glycerol are definitive proof of the TLS model. This brings us back to the point raised at the beginning of this section: While we of course agree that the TLS model gives a natural and elegant explanation[19] of the data, the interesting question is whether it is unique in doing so, and this may be regarded as a special case of the issue raised above concerning nonlinear effects more generally.

To repeat, in this paper, we are not disputing that there are some amorphous solids in which TLSs exist; we are not even necessarily disputing that TLSs may exist, at some level, in all amorphous solids. What we *are* disputing is the claim that the TLS model, in the precise sense in which we have defined it in the last section, is the unique and universal explanation of the behavior, in particular, the thermal and ultrasonic behavior, of amorphous solids below 1 K.

4. ULTRASONIC ABSORPTION: A SMOKING GUN?

The classic work on ultrasound propagation within the TTLS model, with the distribution of parameters given by the "canonical" form (eqs 9 and 11), is the paper of Jäckle.[20] Here, we briefly review the main conclusions, confining ourselves to the small-amplitude (linear) regime. To facilitate the discussion, it is convenient to define the quantity $\tau_c(T)$, the characteristic relaxation time of a symmetric ($E_j = \Delta_j$) TLS with splitting E equal to $k_B T$; this is given by the appropriate special case of eq 7 of ref 20 and may be verified to be proportional to T^3. A closely similar discussion may be given of dielectric-loss experiments (see ref 8).

Within the model, there are two mechanisms for ultrasound absorption that operate in parallel. The first is resonant absorption; the relevant expression follows directly from standard "golden-rule" perturbation theory in $\hat{H}_{int}$ and is given by[8]

$$Q_{res}^{-1}(\omega) = \pi C \tanh\left(\frac{\hbar\omega}{2k_B T}\right) \qquad C \equiv \frac{\overline{P}_0 \gamma^2}{\rho c^2} \qquad (12)$$

where $\overline{P}_0$ is the (constant) TTLS density of states (eq 10) and γ is the rms value of the coupling constant $g_{\alpha\beta}$. (Actually, the quantities Q, C, c and γ should each have a suffix $\alpha = l$ or t denoting the polarization of the sound mode in question; we omit this to avoid cluttering up the formulas).

The second process, "relaxation" absorption, is a little more subtle. The phonon modulates the energy splitting of the TLSs and thereby throws their occupations out of thermal equilibrium; the ensuing relaxation to equilibrium extracts energy from the sound wave and thus leads to damping. The relevant formula (equivalent to eqs 21 and 22 of ref 20) is

$$Q_{rel}^{-1}(\omega) = const \sum_j \frac{(-\partial n_0/\partial E_j)\omega\tau_j}{1 + \omega^2\tau_j^2} \gamma_j^2 \left(\frac{e_j}{E_j}\right)^2 \qquad (13)$$

In the present context, we will be interested primarily in the limit $\omega \ll \tau_c^{-1}(T)$ (but need to note that in the opposite limit, the absorption is smaller by a factor of $(1/\omega\tau_c)$). In that limit, using the perturbation theory formula for τ_j^{-1} ($\propto (\Delta_j/E_j)^2$) and the canonical distribution (eq 9), we find, remarkably, that the distribution of τ^{-1} is, apart from a numerical constant, simply proportional to τ, and this then implies the simple result

$$Q_{rel}^{-1} = \frac{\pi}{2} C \qquad (14)$$

Comparing eq 14 with eq 11, we see that the relaxation contribution to the absorption (inverse Q factor) in the low-frequency, high-temperature regime is predicted to be exactly half of its value in the high-frequency, low-temperature ($\hbar\omega \gg k_B T$) regime!

Let us now turn from the absorption to the shift of the ultrasound frequency due to interaction with the TLS. While the absolute value of this shift is of course not experimentally accessible, its temperature dependence is, and as shown in ref 8, a Kramers−Kronig analysis applied to the above results unambiguously predicts that for fixed ω, one should find both for $\omega\tau_c \gg 1$ ($T \ll T_0$ where T_0 is the temperature at which $\omega\tau_c \sim 1$), and for $\omega\tau_c \ll 1$ ($T \gg T_0$) the relative velocity shift should be given by

$$\frac{\delta c}{c} = A \ln\left(\frac{T}{T_0}\right) \qquad (15)$$

with A equal to C (eq 12) on the low-temperature side and to $-2C$ on the high-temperature side. This is *not* the behavior seen experimentally; while $\delta c/c$ indeed passes through a maximum for $T \sim T_0$, the (negative) slope for $T > T_0$ has a magnitude approximately equal to its (positive) value for $T < T_0$.[21] Reference 21 notes that this and some other discrepancies with the TLS predictions cannot be fixed by "minor modifications" of the TTLS model.

While the above discrepancy may be a definitive refutation (at least for the relevant experimental systems, vitreous silica and BK7) of the TLS model when supplemented with the canonical parameter distribution (eq 9), (i.e., of the TTLS model), is it a refutation of the TLS model as such? Because one needs to fit the experimentally observed frequency as well as temperature dependence, this is not immediately clear. Therefore, we may need to look elsewhere for our "smoking gun".

Consider then the absorption in the high-frequency, low-temperature regime ($\hbar\omega \gg k_B T, \hbar\tau_c^{-1}$). In this regime, the only non-negligible contribution to absorption should be from the resonance mechanism, and a little thought shows that for quite general forms of the distribution of TLS splittings $\rho(E)$, it should have the general form

$$Q^{-1}(\omega) = \text{const}\left(\rho(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_{\text{B}}T}\right)\right) \tag{16}$$

(where the constant involves the γ's etc.). Thus, if we fix ω and vary T, we should predict quite independently all of the unknown parameters

$$Q^{-1}(T) = \text{const}\tanh\left(\frac{x}{2}\right) \qquad \left(x \equiv \frac{\hbar\omega}{k_{\text{B}}T}\right) \tag{17}$$

This really is a smoking gun; violations of this prediction cannot be fixed by making ad hoc adjustments to the TLS model. Conversely, if the behavior in eq 16 is indeed found, this would (pace the "affirmation of the consequent" objection!) be convincing evidence in favor of the model.

What is the current experimental situation? Many reviews give the impression that the TTLS prediction (eq 17) is well-verified. However, to the best of our knowledge, until very recently, there was only one experiment[22] that evaded the low-frequency limit $x \equiv (\hbar\omega/k_{\text{B}}T) \ll 1$, and even that went up only to $x \sim 1.2$, a point at which the correction to the high-temperature limit formula is only about 20% (cf. Figure 2 of ref 22). Therefore, one may reasonably ask whether the data is equally consistent with an alternative formula, e.g. one that would follow from the alternative scenario sketched at the end of section 3. Actually, without further specification of this scenario, the question is ill-defined because unlike in the TLS scenario, the stress matrix element T_{mn} may depend on E_m as well as the difference $E_m - E_n$. If for simplicity we postulate that T_{mn} (or rather its statistical distribution) depends only on the difference $E_m - E_n$, then for fixed ω, we recover a formula similar to eq 17, namely

$$Q^{-1}(T) = \text{const}(1 - \exp -x) \qquad \left(x \equiv \frac{\hbar\omega}{k_{\text{B}}T}\right) \tag{18}$$

In Figure 2 of ref 12 is plotted a comparison of both eqs 17 and 18 with the data of ref 22; while the former may seem to represent it slightly better, the difference would seem to lie within the presumed error bars. (In passing, we note that appreciably better agreement for both the velocity shift and the thermal conductivity is obtained for the "alternative" scenario than that for the TTLS model.[12])

In the past few years, a number of experiments, primarily motivated by interest in designing high-Q superconducting circuits, have been performed on the *dielectric* loss of various amorphous materials; some of these have operated in the regime $\hbar\omega/k_{\text{B}}T > 1$ (see, e.g., ref 23). Unfortunately, while many of these papers claim evidence for (tunneling) TLSs, none has, to our knowledge, tested explicitly for the characteristic TLS temperature dependence (eq 17) over a range where the discrepancy with eq 18 would become visible. We believe that such a test, preferably on a material with the universal value of $\sim 3 \times 10^{-4}$ of Q^{-1} (cf. end of section 3) is now feasible and would be a definitive test of the TLS model for such a system.

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Notes
The authors declare no competing financial interest.

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