Frustrated Interactions and Tunneling: Two-Level Systems in Glasses

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It is argued that the standard model of weakly interacting two-level systems (TLS) is compatible with the proposal that low-temperature properties of glasses are determined by strongly interacting defects. *Typical* strongly interacting defects have their tunneling suppressed, and observable tunneling occurs only for TLS which *appear* to be weakly coupled because their interactions are frustrated. Localized TLS are generated rather than collective modes because tunneling lowers the energy of the system. This explains both the success and the universality of the TLS model.

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Glasses have anomalous low-temperature properties that have been interpreted as evidence for the existence of two-level tunneling systems (TLS) [1,2]. Despite the enormous success of TLS theory, the model is *ad hoc* and one is led to ask what fundamental mechanism gives rise to the universal nature of the TLS seen in many different materials. In this paper the question is addressed by combining ideas of Yu and Leggett [3,4] and Klein [5] stressing the role of dipolar interactions between defects with an understanding of the role of frustration in quantum systems. A simple, intuitively appealing scenario is constructed that leads to universal properties at low temperatures and also explains why a theory of weakly interacting TLS is valid.

The TLS model postulates that amorphous materials contain weakly interacting tunneling centers with two energy levels of splitting ε coupled by a matrix element Δ_0, where the ε are uniformly distributed (constant density of states at low energies) [1,2]. The evidence that this model applies to glasses is very strong [6]. The constant density of excitations as a function of energy leads to a specific heat C that is linear in the temperature T and to a thermal conductivity κ that varies as T^2 for $T \lesssim 10$ K, as observed experimentally. In addition, phonon echoes [7] and saturation of ultrasonic absorption [8] have been observed. These latter experiments probe the two-level nature of the tunneling systems. Finally, tunneling of a TLS in a polycrystalline bismuth film has been observed directly [9]. Therefore, there can be little doubt that TLS exist.

Despite these successes of the TLS model, recently Yu and Leggett (YL) have argued that the model is seriously incomplete (or wrong) because the interactions between the TLS are large enough that they should dominate the physics [3,4]. They point out that several aspects of the behavior of glasses are not easily understood within the framework of the TLS model. These include the nearly universal value of the ratio of the phonon mean free path l to the thermal wavelength λ at temperatures $T<1$ K for many different materials ($l/\lambda \sim 150$) [10], the slight deviation of the temperature dependence of the specific heat C and of the thermal conductivity κ from the TLS predictions, and the presence of a "plateau" in log-log plots of κ as a function of T as well as a "bump" in plots of C/T^3 vs T near ~ 10 K in nearly all glasses.

YL attempt to understand these universal features using a model of defects with strong dipolar interactions. Universal features are expected to arise from such a model because dipolar interactions in three dimensions give rise to a density of states per unit energy that is independent of the number of interacting objects [1,3]. Evidence for such a view includes the fact that glassy crystals such as $KBr_x(CN)_{1-x}$ [11] and $Ba_{1-x}La_xF_{2+x}$ [12] display roughly concentration-independent glassy properties over broad concentration ranges [13]. However, in the scenario of YL it is unclear how the band of excitations induced by the interactions can explain absorption saturation, echoes, and other evidence of localized, saturable excitations.

In this paper these two views are reconciled and a scenario is constructed that leads both to the usual TLS model to describe low-temperature properties and to a natural understanding of universality of low-temperature properties of amorphous materials as a consequence of strong interactions. The new idea in this paper is that the frustrated dipolar interactions between TLS lead not to collective modes but rather to apparently free TLS. Tunneling is frozen out for those TLS which feel a substantial "effective field" from interactions; the only TLS that tunnel are those whose interactions are completely frustrated [14]. The number of these frustrated TLS is determined by the dipolar interactions and displays universal properties. Configurations with localized TLS are selected out because quantum-mechanical tunneling lowers the energy. Frustration insures that many states are energetically degenerate in the classical limit $\hbar \to 0$. Since tunneling is suppressed exponentially as mass increases, the lowest-energy configuration has localized excitations.

These ideas are illustrated with a model similar to that of YL but with tunneling explicitly included, with Hamiltonian [3,4,15–17]

$$H=\sum_{i,j} J_{ij} S_{iz} S_{jz} + \sum_i \varepsilon_i S_{iz} - \sum_i t_i S_{ix}\ , \tag{1}$$

where

$$S_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad S_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$

the sums are over the defects, the J_{ij} describe the interactions between two TLS at sites i and j, and ε_i and t_i respectively describe the asymmetry and the tunneling matrix element of the ith TLS. The TLS are represented as quantum-mechanical $S=\frac{1}{2}$ spins. For dipolar interactions, one expects the magnitudes of the J_{ij} to fall off with separation r_{ij} between sites i and j as J_0/r_{ij}^3. It is useful to define h_i, the effective field at site i:

$$h_i = \varepsilon_i + \sum_j J_{ij} S_{jz} . \tag{2}$$

For example, if all spins except that at site m are held fixed, then the tunneling of the mth spin is described by [18]

$$H_m = h_m S_{mz} - t_m S_{mx} . \tag{3}$$

The interest here is in very low temperatures T, so it is reasonable to consider the energy of this quantum-mechanical system at $T=0$. First, note that interactions suppress tunneling. For instance, if one diagonalizes the Hamiltonian (1) for the case of two spins which each have $\varepsilon=0$ and tunneling matrix element t, then ΔE, the splitting between the ground state and first-excited state, obeys

$$\Delta E(J,t) = (4t^2 + J^2)^{1/2} - J . \tag{4}$$

Thus, as J/t becomes large the tunnel splitting decreases as $\sim 2t^2/J$. Thus, strong interactions lock the spins together and suppress the tunneling. If many TLS are strongly coupled, then either one spin can tunnel but in a very asymmetric well, or else the tunneling of many degrees of freedom must occur simultaneously, which leads to a tunneling rate that decreases exponentially with the number of simultaneous tunnelers. Thus, if infinitely many TLS were, e.g., ferromagnetically coupled, then no tunneling between low-lying states would occur.

However, one expects the J_{ij} to be random, leading to frustrated interactions. The key point is that this frustration leads to the presence of TLS that are basically decoupled from the rest of the system so that they are free to tunnel. Many authors have stressed the importance of frustration in determining the properties of random systems, but the discussion is usually couched in terms of frustrated *loops* [19]. Here it is important to think about the degree of frustration at a *site* [20]. By doing this one sees that completely frustrating the interactions at one site allows the spin at that site to lower its energy via tunneling. If the frustrated bonds are arranged differently so that individual spins are not free to tunnel, then the energy is higher. Thus, the low-energy excitations arising because of frustration are *localized* so that the energy gain due to tunneling can be maximized.

To illustrate how frustration can lead to localized tunneling centers, consider the cluster depicted in Fig. 1. In this cluster, dashed lines correspond to antiferromagnetic ($J_{ij}=+J$) and full lines to ferromagnetic bonds ($J_{ij}=-J$), where $J>0$ is the bond strength. The model is described by the Hamiltonian (1) with $\varepsilon_i=0$ and the $J_{ij}=\pm J$ for nearest neighbors, and it is assumed $t_i \ll J$ for all sites i. This system exhibits frustration—along each plaquette with a question mark in its interior it is impossible to satisfy all the bonds simultaneously. In the limit $J/t \to \infty$ the ground state is degenerate. The states with lowest exchange energy have three frustrated bonds arranged in one of the four ways shown in Fig. 2. In configurations 2(a) and 2(b) no site has more than one unsatisfied bond, whereas in 2(c) and 2(d) the site marked with an asterisk star has two satisfied and two unsatisfied bonds and is hence completely frustrated. In fact, 2(d) is obtained by flipping the spin (*) in 2(c). A tunneling energy of order t is gained if the system has arranged its bonds so that the site (*) is completely frustrated, whereas the other configurations lead to tunneling energies of order t^2/J. Therefore, up to corrections of order t^2/J the ground state of this cluster is a linear combination of configurations 2(c) and 2(d), with energy $-7J-t$. Thus, the ground state has a fully frustrated spin that is free to tunnel.

Of course, Eq. (1) is much more complex than this cluster because of the random, long-range couplings. However, so long as the degeneracy of the $\hbar \to 0$ limit of the Hamiltonian is large enough so that configurations with fully frustrated spins have the same classical energy as those without such spins [21], then for nonzero $\hbar$ the configuration with largest (most negative) tunneling contribution to the energy (and hence localized TLS) is

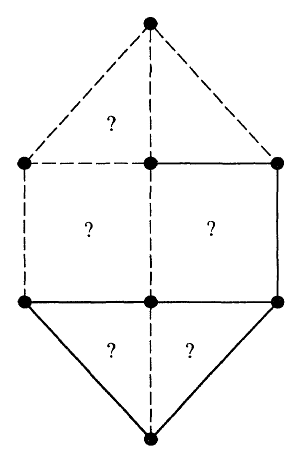

FIG. 1. Cluster of eight spin-$\frac{1}{2}$ particles, denoted by solid dots. Dashed (solid) lines denote antiferromagnetic (ferromagnetic) Ising interactions of strength J. Frustrated plaquettes are labeled with a question mark.

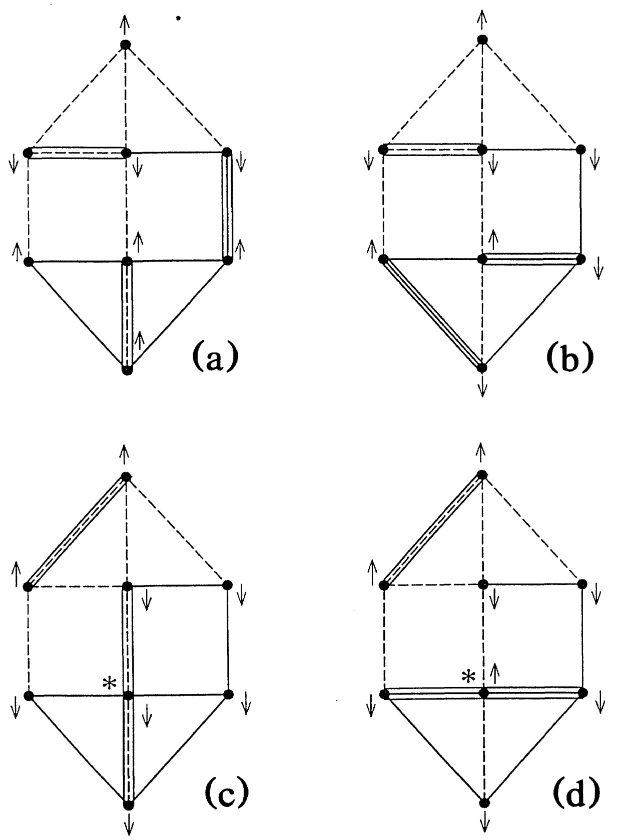

FIG. 2. Four configurations of the cluster shown in Fig. 1 with the lowest exchange energy $\sum_{i,j} J_{ij} S_{iz} S_{jz}$. Each frustrated plaquette has one of the three frustrated bonds, which are denoted with thick lines. The effective field at the site labeled (*) in (c) and (d) is zero and hence this spin is free to flip and lower the energy by t. States (a) and (b) are related by the simultaneous flip of two spins, but this process leads to an energy gain only of order t^2/J. Thus the ground state of this cluster is a linear superposition of (c) and (d) (up to corrections of order t/J). This example illustrates why it is energetically favorable to arrange frustrated bonds so that a *site* is maximally frustrated, producing a localized TLS.

selected out.

Large degeneracy is important so that the effective field at a given site can be made small without sacrificing interaction energy. It is reasonable that dipolar interactions lead to large degeneracy because the entire configuration must be determined self-consistently. Spins more distant than a radius R can contribute to the effective field an amount $\int_R^\infty dr\, r^{d-1}/r^3$, which for $d=3$ diverges logarithmically. It is also vital that at least some of the classically degenerate configurations have spins with small effective fields. Stability considerations yield bounds for the distribution of effective fields that are discussed below.

Since the dipolar interaction falls off with the separation r as r^{-3}, the interaction energy of a system with N defects is proportional to N [1,3], and the total density of states per unit energy and unit volume is $\tilde{n} \sim 1/J_0$ [3]. Estimates of the interaction constant J_0 [3] yield $\tilde{n} \sim 1/J_0 \sim 2\times 10^{-5}/\mathrm{K\,Å}^3$, which is roughly consistent with the observed density of excitations estimated from the specific heat for temperatures above the plateau in the thermal conductivity [15].

Here it is proposed that the plateau in the thermal conductivity and the bump in the specific heat occur when the collective modes (thermally assisted rearrangements of many spins) freeze out, so that the properties below 1 K are determined by the number of tunnelers n_{TLS} rather than the total density of states $\tilde{n}$. The effective field distribution for a system of classical spins coupled by random dipolar interactions has not been calculated, but upper bounds have been obtained from stability arguments [22-24]. The density of local fields $P(h)$ (and hence the number of frustrated spins n_{TLS}) obeys

$$P(h) < \frac{1}{J_0 \ln(J_0/a_0^3 h)}, \tag{5}$$

where a_0 is the defect separation, so that J_0/a_0^3 is the bandwidth. If one assumes that the system's $P(h)$ is close to this upper bound and $a_0 > 1$ Å, then Eq. (5) yields a value of n_{TLS} that is at least an order of magnitude larger than observed. A realistic calculation is thus required to demonstrate this crossover theoretically; such a calculation would provide compelling evidence for the importance of strong interactions.

The scenario presented here is consistent with several experimental observations. First, it is consistent with the large body of experiments indicating that a model involving weakly interacting TLS yields a good description of the low-temperature properties of glasses. Second, it is consistent with the roughly concentration-independent properties observed in glassy crystals [11,12]. Third, it yields a natural framework for understanding the universal properties such as the density of TLS as well as the specific heat bump and the thermal conductivity plateau at ~ 10 K.

In this paper it has been argued that *typical* two-level systems in glasses are coupled by strong dipolar interactions, but that strongly interacting TLS do not tunnel and the only TLS that contribute to the low-temperature properties are the fully frustrated ones that appear decoupled from the others. Weakly interacting TLS are generated rather than collective modes because tunneling lowers the energy of the system. This proposal explains the success of standard weakly interacting TLS theory, while at the same time supporting the hypothesis that the universal properties of glasses arise because of strong interactions between defects.

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