SOME COMMENTS ON THE NATURE OF UNIVERSAL PROPERTIES IN LOW–TEMPERATURE GLASSES[1]

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<u>Abstract</u> We discuss the recent theory of Burin and Kagan that attempts to explain the existence of universal low temperature properties in amorphous solids. We suggest a realistic experimental scenario that could be used to test the theory. We comment on the results of an experiment that has already been performed in the proposed geometry.

INTRODUCTION

Many different amorphous solids display a remarkable universal behavior at low temperatures.[1] Examples of such behavior include a specific heat and thermal conductivity that have roughly linear and quadratic temperature dependencies, respectively, below about 1K. [2] In addition to these qualitative similarities, low temperature amorphous solids show dramatic *quantitative* universalities. An example of this type of universality is demonstrated in the relation $\frac{l}{\lambda} \sim 150$, where l is the phonon mean free path and λ is the phonon wavelength. For many amorphous solids, this relation holds to within a factor of 2, below 1K.[3]

The first type of universal behavior, manifested in the qualitative similarities in the specific heat and thermal conductivity of a variety of amorphous solids at low temperatures, can be described by the standard tunneling model.[4] In this picture, the glass is viewed as a metastable configuration of atoms. In such a configuration, it is possible that an atom or group of atoms may reside in either of two equilibrium positions. The potential energy curve for this situation can be represented as a double well potential. At low temperatures, the atom or group tunnels from one equilibrium position to the other. In a basis consisting of states localized in the left and right wells, respectively, the Hamiltonian for the tunneling process may be

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expressed by

$$H = \frac{1}{2} \begin{pmatrix} \epsilon & -\Delta_0 \\ -\Delta_0 & -\epsilon \end{pmatrix}. \tag{1}$$

Here ϵ is the asymmetry energy (difference in energy between the left and right wells), and Δ_0 is the tunneling matrix element that connects the lowest energy states in each well. The standard tunneling model then dictates that the distributions of the asymmetry energy ϵ and the tunneling matrix element Δ_0 are given by

$$P(\epsilon, \Delta_0) = \frac{\overline{P}}{\Delta_0} \tag{2}$$

with a constant $\overline{P}$. These assumptions lead directly to a specific heat that varies linearly with temperature, and a thermal conductivity that varies as T^2.[4]

Various aspects of the standard tunneling model may be questioned. First, the microscopic foundation for the model is not firmly justified. While there has been some recent success in "locating" the tunneling systems in computer models of disordered solids,[5] questions still remain. Though the uniform distribution in the asymmetry energy is quite reasonable, there is no firm justification for the flat distribution in $\log(\Delta_0)$. Furthermore, the standard tunneling model cannot explain the remarkable *quantitative* universality in the ratios of certain parameters, for instance the relation $\frac{l}{\lambda} \sim 150$ as explained above.[6] As a result of these inadequacies, several alternative models have been proposed.[6,7] A common theme in these models is the belief that the interactions between the tunneling systems dominate the energy scale at low temperatures. Recently, Burin and Kagan[8] have devised a model that attempts to explain not only the form of the distribution of tunneling center parameters, but also the quantitative resemblance of certain properties observed in various glasses. In this note, we will briefly discuss the salient features of the model of Burin and Kagan. We will then propose a realistic experimental scenario that we believe can be used to test their model. Lastly, we briefly comment on one experiment that has already been performed that might shed some light on this issue.

THE MODEL OF BURIN AND KAGAN

In the model of Burin and Kagan[8], the strain mediated dipole-dipole interaction between tunneling centers is responsible for the universal properties observed in low temperature glasses. The amorphous medium consists of double well centers distributed randomly in space with an arbitrary distribution of parameters. Unlike

the situation described in standard tunneling model, the parameters describing the randomly distributed tunneling centers, called "primary defect parameters", do not display universal behavior. That is, the distribution of the defect energy is not necessarily flat, and will vary depending on the chemical composition of the glass. The universal properties appear as a consequence of the many body interaction of the primary tunneling systems, leading to the creation of delocalized excitations called "many center excitations". Due to the delocalization, the spectral properties of the many center excitations are independent of the primary defect parameters (thus leading to universal ratios such as $\frac{l}{\lambda} \sim 150$) and demonstrate practically uniform distributions in the energy asymmetry and the logarithm of the tunneling matrix element. The role of the many center excitations increases with decreasing energy (temperature).

The crucial aspect in the formation of the many center excitations is the fact that the strain mediated interaction between the primary tunneling centers decays as $1/R^3$. In three dimensions, the average number of primary tunneling centers forming a multicenter excitation increases logarithmically with glass volume

$$N(V) \sim \alpha \log(V) . \tag{3}$$

This logarithmic behavior allows Burin and Kagan to study the formation of many center excitations with a renormalization group approach. A similar procedure was first used by Levitov[9] in the study of the delocalization of vibrational modes caused by the electric dipole interaction. The logarithmic divergence of $N(V)$ indicates *criticality* $(N(V) \sim V^\alpha)$, and, hence, *delocalization.*[9] According to this argument, a "modified dipole-dipole interaction", in three dimensions, $1/R^{3+\eta}$, prohibits the formation of delocalized multicenter excitations for $\eta > 0$. As a result, the distribution of primary tunneling centers gains importance and, following the argument of Burin and Kagan, no universal behavior of glasses is expected. In contrast, for $\eta < 0$ the number of primary centers forming a delocalized excitation diverges.

While we will not recapitulate the detailed arguments of Burin and Kagan, we would like to highlight some important features of their argument. Starting from the Hamiltonian

$$H = -\sum_i \omega_i S_i^z - \frac{1}{2} \sum_{ij} U_{ij} S_i^z S_j^z - \sum_i \Delta_{0i} S_i^x, \tag{4}$$

where ω_i is the asymmetry energy (previously referred to as ϵ), Δ_{0i} is the tunneling matrix element of the ith primary tunneling center, and $U_{ij} = \frac{u_{ij}}{R_{ij}^3}$ gives the interaction strength between primary centers. The asymmetry energy is distributed in the

interval $(-W/2, W/2)$, the average scale for the tunneling amplitudes of the primary centers is Δ_{0*}, and $U_0 = \langle |u_{ij}| \rangle$ gives the characteristic scale for interactions of the primary centers. In the standard tunneling model, the parameters ω_i and Δ_{0i} have preassigned distributions, whereas here Burin and Kagan assume no specific form for the distribution of these primary defect parameters. It is, however, assumed that

$$W \gg U_0 n \gg \Delta_{0*} \tag{5}$$

where n is the density of tunneling centers. This allows Burin and Kagan to first neglect the tunneling, and to include its effects in a perturbative fashion in the parameter Δ_{0*}/W.

Now the density of states for the asymmetry energy in the presence of TLS-TLS interactions, $P(\Delta)$, is considered (here Δ is the asymmetry energy modified by the interactions). At zero temperature, the system should be in the ground state. This means that the energies of the multicenter excitations should be positive. For single particle excitations, neglecting for now the last term in the Hamiltonian (4), this fact is embodied in the stability criteria

$$\Delta_i = \omega_i + \frac{1}{2}\sum_j U_{ij} S_j^z > 0 \ . \tag{6}$$

This type of stability condition may be extended to include n centers. For example, pair excitations have the stability requirement

$$\Delta_{ij} = \Delta_i + \Delta_j - U_{ij} > 0 \ , \tag{7}$$

three center interactions $\Delta_{ijk} = \Delta_i + \Delta_j + \Delta_k - U_{ij} - U_{ik} - U_{jk} > 0$ and so on. Burin and Kagan first consider a restricted range of interaction, R_0, limited enough to consider the intercenter interactions as a weak perturbation. Next, the decrease in the density of single particle excitations caused by the stability criteria for pair excitations is calculated,

$$P_1(\Delta) = \frac{1}{V}\sum_i \langle \delta(\Delta - \Delta_i) \prod_i \theta(\Delta_{ij}) \rangle. \tag{8}$$

Here, V is the system volume, and $\theta(\Delta_{ij})$ is a step function that enforces the stability requirement for pair excitations. The density of single particle excitations is then approximated as

$$P_1(\Delta) \sim P_0 \left(1 - P_0 \int d\mathbf{R_{12}} \int d\Delta_1 \langle \theta(\frac{u_{ij}}{R_{12}^3} - \Delta_1 - \Delta) \rangle_u \theta(R_0 - R_{12}) \right), \tag{9}$$

where $\langle...\rangle_u$ denotes an average over the u_{ij}. Burin and Kagan assume that $P(\Delta)$ has no singularity at $\Delta = 0$, allowing for the replacement of $P(\Delta_1)$ with $P_0 = P(0) \approx n/W$ because the main contribution to the above integral comes from small values of Δ_1. The above integral may be performed, yielding

$$P_1(\Delta) \approx P_0(1 - 2\chi\xi), \tag{10}$$

where

$$\chi = \pi P_0 U_0 \ll 1 \tag{11}$$
$$\xi = \ln(R_0/R_{min}) \tag{12}$$

with R_{min} defined through $U_0/R_{min}^3 \approx W$. A similar calculation for the density of states for pair excitations yields

$$P_2(\Delta) \approx P_0\chi\xi \tag{13}$$

and, in general, for many center excitations, $P_n \sim (\chi\xi)^{n-1}$. The crucial point is that as R_0 increases, the product $\chi\xi$ becomes larger ($\chi\xi \sim \mathcal{O}(1)$), signaling the decrease in the importance of the single particle (primary tunneling center) properties, and the onset of many center excitations. Using a renormalization group approach, Burin and Kagan calculate the density of states $P(\Delta, \Delta_0)$ including the influence of the many body terms. They find, after tunneling effects are included, a distribution that may be approximately written as $P(\Delta, \Delta_0) = \overline{P}/\Delta_0$ with a value of $C \equiv \overline{P}U_0 \sim 10^{-3} - 10^{-4}$ that is in agreement with experiments.[3] This distribution results from the consideration of many body effects; *the primary (noninteracting) set of tunneling systems do not show this universal behavior*. The universal parameter C appears due to the delocalization caused by the TLS-TLS interaction that produce many center excitations, effectively "washing out" the details of the chemical structure of the particular glass under study.

A PROPOSED TEST

A crucial aspect of the theory of Burin and Kagan is the long range $1/R^3$ dipole-dipole force between the TLS resulting in delocalization. This fact is responsible for universal low temperature properties in glasses. This was originally conjectured by Yu and Leggett.[6] Following the reasons given below Eq. (3), one has to put the system out of criticality in order to test this conjecture. One may do this by either

changing the form of the TLS interaction or by confining the spatial geometry of the primary tunneling centers.

First work in the latter direction has been done by Fu.[10] He proposed a study of the properties of a free-standing, thin glass wire. He showed that in a thin fiber of radius R_*, the long ranged $1/R^3$ force is modified to $U \sim \exp(-R/R_*)$ for two defects separated by a distance $R > R_*$. Thus, the hypothesis that long range forces are responsible for the universal properties observed in glasses may be tested in a thin glass fiber, where the dipole-dipole force is no longer long ranged.

As far as we know, no experimental studies have been made on such a system. The reasons for this are twofold. First, it is very difficult to do low temperature studies on free fibers. Here, the coat surrounding the fiber will greatly increase the fiber radius, and the need for good thermal contact with a refrigerator will make isolation of the fiber difficult (or impossible). Second, the temperature must be extremely low in order to see the effects of the confining wire. One recent estimate of the temperature needed to see the effects proposed by Fu is $T \sim 10^{-7}$ K for a wire with a $1\mu m$ diameter.[11]

Is it possible to test, in a realistic way, theories like the one outlined in the section above? We believe the answer is yes. The crucial point to note in the theory of Burin and Kagan is the need for a TLS-TLS coupling that varies as $1/R^3$ *in three dimensions*. The story is drastically different if all the TLS are confined to quasi-two dimensions, while the coupling between them still varies as $1/R^3$. Consider a layer of thickness a, where a is of atomic dimensions. If such a layer is glassy, and lies on a substrate that is thick and contains no TLS dynamics, then we are approximately in the regime where the TLS dynamics are confined to two dimensions, while their interaction still varies as $1/R^3$. We implicitly assume that sound waves are not affected by the interface between the amorphous layer and the bulk. To see how this situation varies from the usual one, consider the parameter ξ of the last section for $R \gg a$

$$\xi = \frac{1}{4\pi} \int \frac{d\mathbf{R_{ij}}}{R_{ij}^3} \approx \frac{a}{2}\left(\frac{1}{R_{min}} - \frac{1}{R_0}\right). \tag{14}$$

Since R_{min} is of the order of the size of the primary tunneling centers, the parameter ξ is always $\mathcal{O}(1)$, and never shows the logarithmic growth characterized by the usual situation in three dimensions. Accordingly, in the geometry proposed above, the parameter $\chi\xi$ defined in the last section will *always be small*, obviating the importance of the many center excitations. In such a case, the intrinsic "primary"

distributions should dominate, and universal properties will be lost (if one accepts the arguments of Burin and Kagan).

The experiment we suggest has already been performed, albeit not for the purpose that we discuss here. The hole burning experiment of Orrit, Benard, and Möbius on an ionic dye in a Langmuir-Blodgett monolayer is an experiment of the type we propose above.[12] In this experiment, persistent holes in the excitation spectrum of resorufin adsorbed on an ammonium salt monolayer were measured. The monolayer is disordered due to preparation effects and the holes showed signatures of glassy behavior. In fact, Pack and Fayer[13] were able to explain this data qualitatively by assuming a standard tunneling model description of monolayer. *This contradicts the theory of Burin and Kagan*, which (as we have pointed out above) predicts results at variance with the standard model in a two dimensional system with $1/R^3$ TLS-TLS interactions. It would be important, however, for a variety of such experiments to be performed with different "glassy" monolayers, so that a definitive conclusion can be reached.

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