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Low Temperature Properties of Amorphous Materials: Through a Glass Darkly

A wide variety of amorphous materials exhibit similar behavior in their thermal properties. Examples of such universal features include a linear specific heat and a T^2 thermal conductivity below about 1 K. While there is at present no generally accepted model of their behavior at higher temperatures, for the past fifteen years the low-temperature behavior has been attributed to the existence in these materials of tunneling two-level systems. The two-level system model is generally believed to receive strong support from a series of remarkable effects (phonon echoes, etc.) observed in the acoustic behavior of these materials.

In this Comment we point out (a) that while the observed effects are (mostly) consistent with the two-level system model, they do not uniquely establish it, (b) that the model cannot explain the dramatic *quantitative* universality in the low-temperature behavior of amorphous materials, and (c) that the neglect to lowest order within the model of the long-range interactions induced by the strain field, while self-consistent, is not imposed by the data. We sketch an alternative scenario for the properties of amorphous materials in which the role played by these interactions is essential. This scenario, if it can be quantitatively implemented, holds out promise of being able to explain in a unified way not only the quantitative universality below 1 K but also the behavior at higher temperatures.

1. INTRODUCTION

The problem posed by the low temperature properties of glass can be stated as follows: If you take a bunch of atoms, form a crystal and measure the properties of that crystal, you will probably find textbook characteristics such as a Debye specific heat. If, on the other hand, you take those same atoms, jumble them up and form an amorphous solid, you will find properties quite different from

Comments Cond. Mat. Phys.
1988, Vol. 14, No. 4, pp. 231-251

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those of a crystal. In fact, it will not matter very much which particular atoms you chose. The properties of amorphous solids tend to be universal, independent of the particular material and its chemistry. In other words, the basic problem is: what is it about the amorphous nature of these materials that gives rise to such universal features?

The purpose of this Comment is to discuss and define this problem with regard to the low temperature properties of insulating glasses.^{1,2} We begin with a brief review of experiments on specific heat and thermal conductivity. We then describe the standard theory of two-level systems which was proposed in 1972 by Anderson, Halperin and Varma,³ and independently by W. A. Phillips.⁴ In spite of its success, this model has severe inadequacies. As we shall describe, these shortcomings are threefold: (1) There is no experimental proof that local tunneling centers exist. (2) Two-level systems are unable to explain certain universal features seen in glasses. (3) The standard theory includes interactions between two-level systems only where a noninteracting model would clearly give unphysical results. Finally, we will discuss some of our preliminary results on a model which treats the interactions between defects as the fundamental feature. We suggest that the type of interactions depends on length scales. In the low temperature regime collective modes result. These have a logarithmic density of states that agrees well with experiment.

Low Temperature Properties of Insulating Glasses

At low temperatures crystalline materials have a T^3 thermal conductivity due to boundary scattering. Not surprisingly, amorphous materials exhibit very different behavior. What is surprising is that a wide variety of glassy substances, e.g., SiO_2 , Se, polymers and varnishes, have very similar properties. In 1971 Zeller and Pohl⁵ pointed out that below 1 K the specific heat C is roughly linear in temperature (Fig. 1) and the thermal conductivity κ goes as T^2 (Fig. 2). Actually the specific heat is slightly superlinear in insulating glasses ($C \sim T^{1+\delta}$, $\delta \sim 0.1-0.3$) and the thermal conductivity is slightly subquadratic ($\kappa \sim T^{2-\epsilon}$, $\epsilon \sim 0.05-0.2$). Between 3 and 10 K the thermal conductivity exhibits a plateau and then continues to rise at higher temperatures. The specific heat also displays an

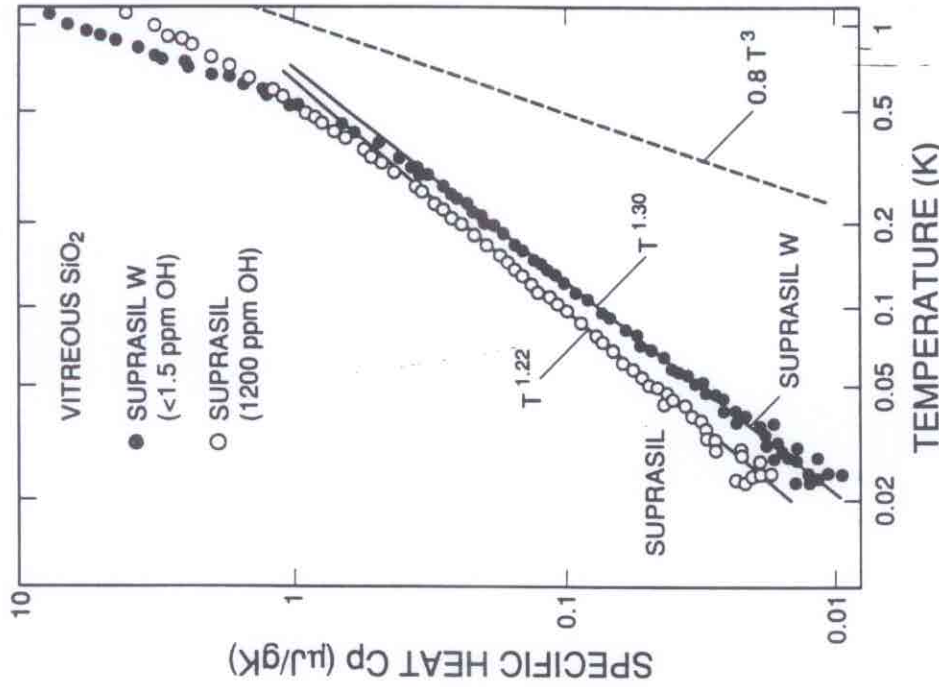


FIGURE 1 Low temperature specific heat of vitreous SiO_2 from Ref. 27. Notice that the specific heat is slightly superlinear.

anomaly in this temperature range. Namely, there is a bump in C/T^3 versus T . This is shown in Fig. 3. The dashed lines on this plot are the Debye specific heats calculated from sound velocity measurements. Notice that the measured specific heat is greater than the Debye value. This excess is often referred to as the anomalous specific heat. The universality of the thermal conductivity

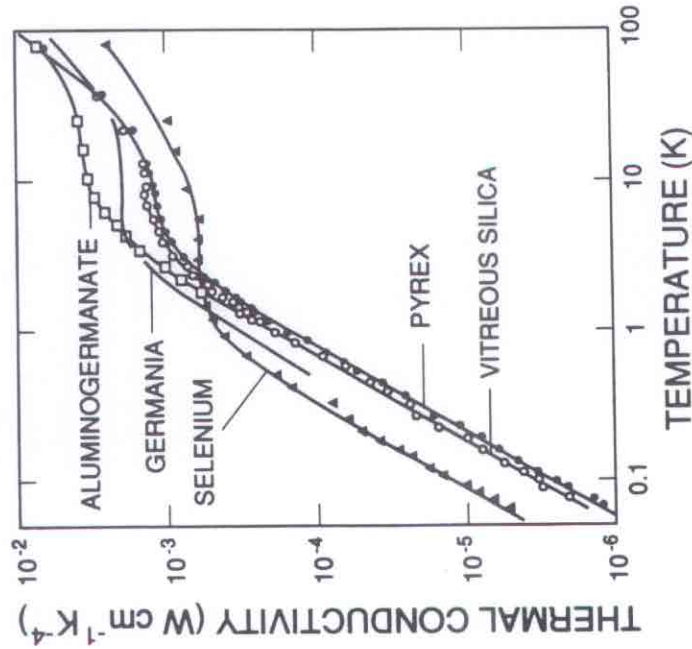


FIGURE 2 Low temperature thermal conductivity of a variety of amorphous materials from Ref. 5. Below 1 K the thermal conductivity is slightly less than quadratic.

goes beyond its T^2 dependence. Freeman and Anderson⁶ have demonstrated that the thermal conductivity of different materials can be scaled onto one curve both above and below the plateau. They did this by rescaling κ by a constant which depends on the Debye temperature and the average sound velocity, and by rescaling the temperature T by the Debye temperature.

Standard Theory of Two-Level Systems

Glassy properties below 1 K are usually viewed within the confines of the standard model of two-level systems.¹⁻⁴ The motivation behind this model is to think of a glass as a metastable configuration of atoms. In this jumble there are probably a few atoms or groups

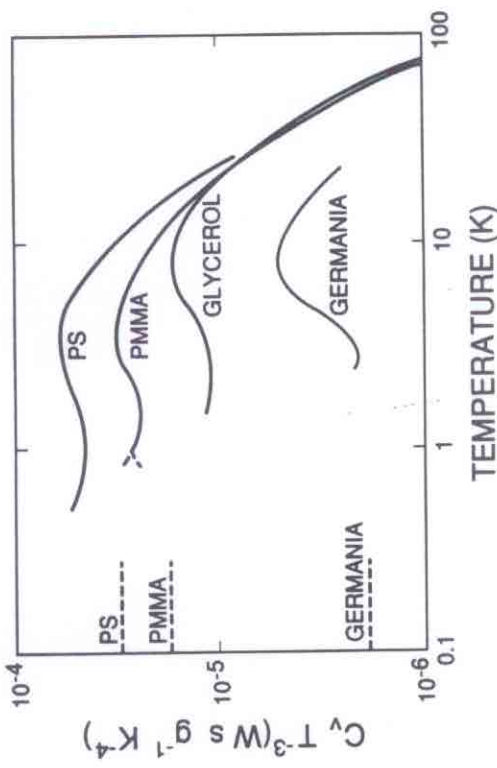


FIGURE 3 $C_v T^3$ versus T from Ref. 5. Notice the bump is between 3 and 10 K.

of atoms which can sit more or less equally well in either of two equilibrium positions. This can be represented schematically by a double well potential as shown in Fig. 4. At low temperatures the atom or group tunnels back and forth between the two minima. The difference between the right well and the left well is given by the asymmetry energy ϵ . The barrier height is denoted by V and d is the distance between minima. It is easy to write down a Hamiltonian for this system. If we use a right-well-left-well basis, the Hamiltonian is a 2×2 matrix:

$$H = \frac{1}{2} \begin{pmatrix} \epsilon & -\Delta_o \\ -\Delta_o & -\epsilon \end{pmatrix}. \quad (1)$$

Here Δ_o , the tunneling matrix element, can be expressed using the standard WKB formula

$$\Delta_o = \omega_o e^{-W} \quad (2)$$

where

$$W \sim \frac{\sqrt{2MVd}}{\hbar}. \quad (3)$$

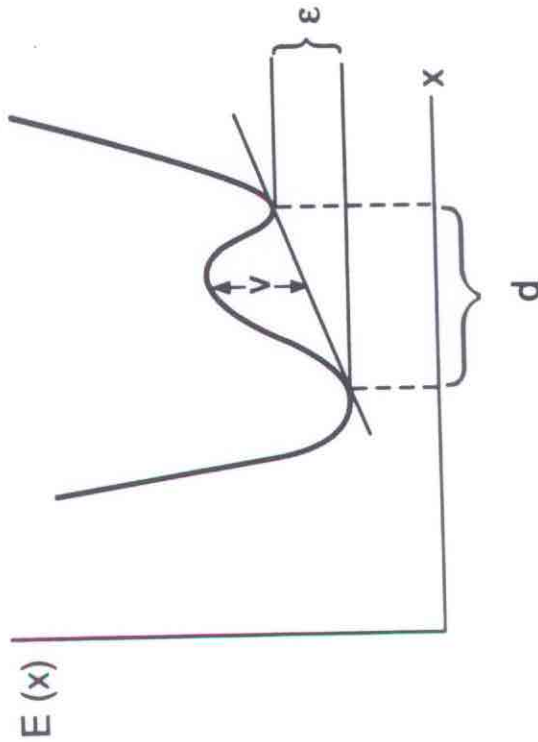


FIGURE 4 Double well potential for a two-level system.

M is the mass of the atom or the tunneling entity. ω_0 is on the order of the Debye frequency since atomic motion is involved in the tunneling. In order for tunneling centers to be seen experimentally, they must have low barrier heights, i.e., $V \ll 0.1$ eV. When a transition occurs in a two-level system, a phonon is emitted or absorbed to conserve energy. This implies that $\epsilon \approx kT$. Typical estimates of the number of two-level systems are in the range of 100 ppm. (This comes from integrating up to 10 K the density of states deduced from specific heat measurements (see below).)

Two important assumptions enter into this model. The first is that the distribution of the exponential argument W of the tunneling matrix element is flat, i.e., $P(W)$ is constant. The second is that the distribution of asymmetry energies $N(\epsilon)$ is constant at least at low energies. With these forms for $N(\epsilon)$ and $P(W)$, the density of states $n(E)$ of the energy eigenvalues of the two state Hamiltonian in Eq. (1) is constant.

How do two-level systems account for a linear specific heat and a T^2 thermal conductivity? It is relatively easy to derive a linear specific heat using a constant density of states n_0 . The calculation

is similar to that used for electronic heat capacities. One finds that $C(T) = \pi^2 k_B^2 n_0 T/6$. To calculate the thermal conductivity, we must first ask what is carrying the heat at low temperatures. Zaitlin and Anderson⁷ showed experimentally that phonons are responsible for thermal transport below about 20 K. Thus, the expression for $\kappa(T)$ is

$$\kappa(T) = \frac{1}{3} \int_0^{\omega_D} C_{\text{ph}}(T, \omega) v l(T, \omega) d\omega \quad (4)$$

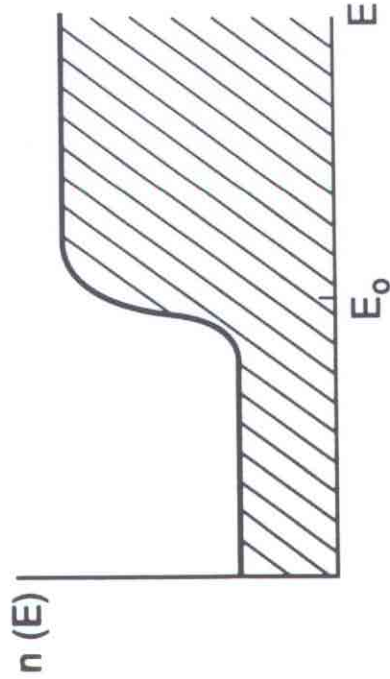
where $C_{\text{ph}}(T, \omega)$ is the phonon specific heat, v is the sound velocity, and ω_D is the Debye frequency. $l(T, \omega)$ is the phonon mean free path as a function of temperature and frequency. The dominant process which limits the phonon mean free path at low temperatures is resonant scattering. In this process a phonon carrying energy $\hbar\omega$ is absorbed by a two-level system of energy $E \sim \hbar\omega$. As a result the two-level system makes a transition from its ground state to its excited state. It turns out that as a result of resonant scattering, $l \sim \omega^{-1} \sim T^{-1}$. When this is combined with $C_{\text{ph}} \sim T^3$, we see that $\kappa(T) \sim T^2$. However, we note an important feature which, while not incompatible with two-level systems, is not explained by the standard model. Namely, all experimental data is consistent with a phonon mean free path below 1 K that is 150 times the phonon wavelength λ to within a factor of 2. This remarkable quantitative universality seems not to have made its full impact on the glassy community.

Crossover

We now turn to the plateau in the thermal conductivity and bump in C/T^3 (Figs. 2 and 3). These features are empirically correlated, i.e., they appear in the same temperature range (3–10 K). What do they imply about the physics of glasses? No hints come from the two-level-system model. Instead let us assume that the phonons continue to carry the heat in this temperature range. Using Eq. (4) and assuming the phonons have a Debye specific heat, we can ask how $l(T, \omega)$ must behave as a function of frequency in order to give rise to a plateau. The answer is shown in Fig. 5(a). As we have already remarked, at low frequencies $l \sim 150\lambda$ where λ is the



(a)



(b)

FIGURE 5 (a) This is the phonon mean free path versus frequency that is needed to fit the thermal conductivity. (2) Density of states of glassy excitations needed to fit the bump in C/T^3 and the thermal conductivity plateau.

phonon wavelength. Around 200 GHz the mean free path drops sharply. Some^{8,9} have attributed this to Rayleigh scattering ($l \sim \lambda^{-4}$) although the exponent could be greater than 4. At higher frequencies (above the plateau) l is of order λ .^{6,8,10} Thus one can think of the plateau as representing a crossover region from a long mean free path at low frequencies to a short mean free path at higher frequencies.

What is needed to account for such behavior? One ingredient is a rise in the density of states at some energy E_0 (Fig. 5(b)).^{11,12} Such an increase in the number of scatterers would shorten the phonon mean free path. An increase in the number of excitations would also account for a bump in C/T^3 which can be thought of as a crossover from a low temperature value of the linear specific heat to a high temperature value. Fitting the bump for various materials sets E_0 in the range 10–40 K. This is consistent with the rise in the density of states measured by neutron scattering¹³ and Raman scattering.^{14,15} That E_0 is considerably higher than the temperature at which the bump occurs can be understood by noting that excitations give a sizable contribution to the specific heat well below their characteristic frequency. To get a quantitative fit to the bump, one also needs to include strong scattering such as Rayleigh scattering. Including only a rise in the density of states which fits the specific heat gives too high a plateau.¹¹ These two ingredients, the rise in $n(E)$ and strong scattering, do not “solve” the problem of the crossover. They merely help to define what a successful theory should explain: the bump in C/T^3 and the plateau in the thermal conductivity between 3 and 10 K. In other words, what picks out this energy scale?

2. PROBLEMS WITH THE MODEL OF TWO-LEVEL SYSTEMS AT LOW TEMPERATURES

No Direct Experimental Evidence

Even if we confine ourselves to the regime below 1 K, there are some serious problems with the two-level system model. In the first place, not only is there no direct experimental evidence that anything is tunneling; if one looks at it carefully, there is no *un-*

ambiguous proof that two-level systems, in the sense usually assumed, exist at all. This point needs some discussion. The principal evidence usually cited in favor of the two-level system model, apart from the fact that it can explain the linear specific heat and T^2 thermal conductivity as described above, is a remarkable set of experimental observations on the behavior of ultrasound propagation in glasses in this regime. Take phonon echoes as an example.¹⁶ These are the ultrasonic analog of spin echoes seen in NMR. In this case two ultrasound pulses a time τ apart are followed by an echo at time 2τ . This implies that there are long-lived states (lifetime $> 2\tau$) which have time-reversal invariance. It does not imply that there are necessarily two-level systems since multi-level systems which behave as spin $S \geq 1/2$ can also give phonon echoes.

A second example is the attenuation and velocity shift of ultrasound.^{1,2,17,18} The two-level system model predicts that the absorption due to resonant scattering by the two-level systems has the form

$$I^{-1}(\omega, T) = A_I \omega \tanh\left(\frac{\hbar\omega}{2kT}\right) \quad (5)$$

where A_I is a constant. Existing experimental data are consistent with this formula (though it should be noted that with one exception¹⁸ experiments to date have been restricted to the regime $\hbar\omega \ll kT$). It is customary to relate the sound attenuation to a shift in the sound velocity via a Kramers-Kronig transformation which is essentially an integral over frequency. Assuming that a formula of the form (5) is valid for the frequency range of integration, then it implies, by a Kramers-Kronig relation, a shift in the velocity of low frequency sound of the form

$$\frac{\Delta v}{v} = \frac{2vA_I}{\pi} \ln\left(\frac{T}{T_0}\right) \quad (6)$$

where v is the sound velocity, A_I is a constant, and T_0 is a reference temperature. Equation (6) is also consistent with experiment. However, the question arises whether the observation of (5), and hence (6), necessarily implies a two-level system model. In fact it

does not: it is easy to construct models of many-body systems which reproduce (5) and (6) but are not (or at least not obviously) reducible to the form of two-level systems. In passing we note that the assumption leading from (5) to (6) actually implies that $A_v = A_I$. In contrast, while the experimental results are consistent with (5) and (6) individually, to fit them A_v must be 40% larger than A_I .¹⁸ The most natural explanation is that the density of states is not exactly constant. To adjust $n(E)$ in order to resolve this discrepancy, along with the slight deviation of the thermal conductivity exponent from 2, only emphasizes the *ad hoc* nature of the model.

A third example of an experiment that is cited as evidence of two-level systems is saturation of the sound attenuation.^{1,2,18,19} By saturation we mean that the attenuation at a given frequency falls to zero as the acoustic intensity increases. This is consistent with the standard model in which only a finite number of slowly relaxing two-level systems can resonantly absorb sound. As the number of phonons exceeds the number of these two-level systems, the excess ultrasound is not attenuated, i.e., the attenuation saturates. However, this experiment does not prove that two-level systems exist. One might well consider a more general model involving many-body energy levels. The saturation of the attenuation requires only that if particular phonon α can be absorbed with conservation of energy by a transition $A + \alpha \rightarrow B$ between two many-body levels A and B , then there exists no further level C such that the transition $B + \alpha \rightarrow C$ also conserves energy (and, of course, the level B is sufficiently long-lived). Almost any structure of the many-body levels other than the one derived from simple harmonic oscillators fulfills this condition.

We could continue to discuss experiments but the first point should be clear: there is no definitive experimental proof that two-level systems exist or that anything is tunneling. It is true that the two-level system model is *sufficient* to explain most of the anomalous features of ultrasound propagation cited above; it is not at all obvious that it is *necessary*. Of course, it is improbable in the extreme that it is simply an accident that the two-level system model gets these features right. The really interesting question is whether it does so because it is unique or rather because it is a

subclass of a more general class of models for the many-body energy levels and matrix elements which, in general, do not have the property of being factorizable into a product of two-state descriptions. Surely such more general models can indeed explain at least some of the cited effects.

Standard Model Does Not Explain Universal Behavior

A second problem of the two-level system model is that, while by construction it is *consistent* with the observed universality of glassy properties, it does nothing to *explain* them. It is generally agreed that the two-level system model fails to explain the behavior seen above 1 K. However, it is important to emphasize that it also fails to address some of the most remarkable universalities seen below 1 K where the two-level system model is generally believed to be valid. For example, why should there even be excitations below 1 K? The other energy scales in the problem are much higher, e.g., the phonon energy scale is on the order of the Debye energy ($\Theta_D \sim 300$ K) and the electronic energy scale involves bond energies which are typically a few thousand degrees. Possibly one could believe that there are a few two-level systems with 1 K energy splittings. But why are there so many in such a wide variety of glasses? Why is their density of states so flat? Why is the ratio l/λ so close to 150 for so many glasses?

Another common trend is the coefficient of the $\ln T$ term in the ultrasound velocity shift of Eq. (6). If we confine ourselves to genuinely amorphous insulators (thereby excluding both crystalline Si and KBr-KCN mixtures, where a similar term occurs) then the measured variation in this quantity, among the twenty or so different amorphous materials for which values appear in the literature, is a factor of about 4. (This is essentially the same feature as the universality of the scaled thermal conductivity.) In the two-level system model the parameters which determine this quantity, namely the density of two-level systems and the coupling constants of the phonons to them, are independent parameters of the model (and individually vary by factors on the order of 10–20) so that this universality can be attributed only to a coincidence.

Importance of Interactions

The third problem with this model is its failure to take into account interactions between two-level systems. Individual two-level systems couple to phonons. Thus, they should interact with each other by exchanging phonons. In a few instances interactions have been included perturbatively²⁰ or within the context of mean field theory,²¹ in order to treat certain aspects of glasses. For the most part, however, it is assumed that the main characteristic features of glassy behavior can be understood from a noninteracting model.

Let us estimate the strength of this interaction. The Hamiltonian which describes the coupling of a two-level system to the strain field is^{1–4}

$$H = \gamma S e \quad (7)$$

where e is the strain field and the two-level system operator S acts like a spin-1/2 operator. γ is the coupling constant and has a typical value of 1 eV. Using second order perturbation theory, one can show that the effective interaction between two two-level systems exchanging phonons is dipolar, i.e., it goes as g/r^3 where $g \sim \gamma^2/\rho v^2$. ρ is the mass density and v is the sound velocity. Taking $\gamma \sim 1$ eV, we estimate $g \sim 5 \times 10^4 \text{ K}\text{\AA}^3$.

The justification which is usually invoked for treating the interaction as a perturbation goes roughly as follows²⁰: Consider a volume of radius r^3 surrounding a particular two-level system. The chance of finding a second two-level system in this volume whose energy splitting is within ΔE of the first is of order 1 only if $\Delta E n(E) r^3 \geq 1$, i.e., if $\Delta E \geq [n(E)r^3]^{-1}$. On the other hand, the interaction will have a substantial effect on the level structure only if it is of order ΔE or greater. This requires $g/r^3 \geq [n(E)r^3]^{-1}$, i.e., $gn(E) \geq 1$. Now it is easy to show that it is precisely this combination of parameters which determines the ultrasonic attenuation and velocity shift; in fact, up to a factor of order unity we have $\lambda/l \sim gn(E)$. Thus the fact that the experimentally observed value of λ/l is of order 10^{-2} (cf. above) supposedly shows that the effects of the interaction between two-level systems are indeed a small,

perturbation. Although this argument is self-consistent, it is by no means obvious that it is the only way to view the problem. Indeed, the fact that $gn(E) \ll 1$ may itself be a result of interactions.

3. AN ALTERNATIVE MODEL

With the above considerations in mind, we will sketch in the rest of this paper a possible alternative approach to the problem of glasses. We emphasize that what we are going to present is not a theory but at best a scenario, that is, a series of guesses which we cannot at present justify but which we believe, if true, would explain a great many of the puzzling features of these materials.

We start by asking what basic features do glasses have in common that is more general than two-level systems. Experimentally we know that at low temperatures there are contributions to the entropy which are not due to phonons. There is no reason *a priori* why this should be true but it certainly is so for the vast majority of amorphous materials. We therefore assume that glasses contain some sort of defects that have some low lying energy excitations with no restriction on the energy range. We want our model to be sufficiently general that we do not need to specify a microscopic model of the source of excitation. These defects could, for example, be associated with single square wells. After all a glass is typically 10% less dense than its crystalline counterpart. The extra space could mean that the atoms sit in rather flat-bottomed potentials. In any case, whatever the defects are, we assume that they couple linearly to phonons in the way described by Eq. (7) with S schematically representing the defect operator. In order to resist the temptation of thinking of S as a spin operator, we will let B represent the defect operator. The strain-mediated defect-defect interaction is dipolar:

$$H_{\text{eff}} = \sum_{ij} \frac{g}{r_{ij}^3} B_i B_j \quad (8)$$

where $g = \gamma^2/\rho v^2$.

Here we break with tradition and take the position that it is interactions, not the intrinsic splitting of the levels of a single

defect, which dominate the energy scale. The basic idea is that these interactions between defects will give rise to collective modes. A mean field theory approach²¹ to the problem of a system of defects interacting via g/r^3 yields a low energy density of states that is independent of the number of defects, i.e., $n(E) \sim 1/g$. This is obviously not universal since g is material dependent. In particular, when we take account of the difference between transverse and longitudinal phonons, the proper expression for $n(E)$ depends on v_t, v_l, γ_t and γ_l , where the subscripts l and t denote longitudinal and transverse, respectively. We can, however, look at the dimensionless ultrasonic attenuation $\sim \lambda/l$. As we mentioned before, this will be proportional to $gn(E)$ and hence will be a function only of the ratios (v_t/v_l) and (γ_t/γ_l) . These ratios fluctuate much less from material to material than do the absolute magnitudes of the individual sound velocities and couplings. Hence the dimensionless attenuation is reasonably constant in accordance with the behavior noted earlier for the coefficient of the logarithm in Eq. (6). There is, however, a snag in this scenario. Namely, $\lambda/l \sim gn(E) \sim 1$, i.e., $l \sim \lambda$. This agrees with the behavior of the thermal conductivity at high temperatures but not with the behavior at low temperatures or in the crossover region.

Crude Scaling Argument

The change in behavior at the crossover can be understood as a change in length scales. We suspect that the type of interaction between defects depends on length scales. Before we explain what we mean by this, let us set up the following picture. Imagine that we divide a sample into blocks of size r_1 such that the distance between the centers of the blocks is also r_1 . We then work out the interaction between blocks via phonons. Having done this, we can group our blocks into bigger blocks of size r_2 . These might be somewhat different from those of the smaller blocks because of the phonon interactions. We can repeat the process of working out the interactions between blocks and forming bigger blocks. By iterating, we go to longer and longer length scales. Second order perturbation theory yields the usual dipolar interactions between blocks, namely, g/r^3 . We suspect that the crossover is associated with a change in the interaction at a certain length scale. This is

roughly given by $r_o \sim \sqrt{g/\hbar v}$ since this is the only length scale associated with g . Although we can think of several possible scenarios which suggest r_o as a crossover, we do not feel confident enough to present any of them here, though in all of them, long-range interactions (g/r^2) dominate for long length scales ($r > r_o$). We do, however, believe this crossover is associated with the plateau in the thermal conductivity and the bump in C/T^3 . Using our previous value of $g \sim 5 \times 10^4 \text{ K}\text{\AA}^3$, we estimate $r_o \sim 13 \text{ \AA}$ and $\hbar\omega_o = g/r_o^3 \sim 24 \text{ K}$. This is consistent with the value of the energy at which the rise occurs in the density of states ($E_o \sim 10\text{--}40 \text{ K}$).

Density of States

Below E_o the long-range part of the interactions lead to collective modes involving many defects. The density of states for these modes differs from that for individual defects. A stability argument suggests that the renormalized density of states at low energies is given by²³

$$n(E) \sim \frac{1}{g \ln(T_d/E)} \quad (9)$$

where T_d is a characteristic temperature of the defects. It is not the glass transition temperature but rather is on the order of $\hbar\omega_o$.

To get a rough idea of where the logarithm comes from, consider the Hamiltonian

$$H = \sum_i \phi_i n_i + \frac{1}{2} \sum_{i \neq j} V_{ij} n_i n_j \quad (10)$$

where the subscripts i and j refer to sites or blocks. ϕ_i is the on-site energy. V_{ij} represents the interaction between sites. Suppose, for the moment, that each site has two energy levels. Then n_i refers to which level is occupied on site i . The energy shift at site i due to interactions is given by

$$\Delta E_i = \frac{1}{2} \sum_j V_{ij} n_j \quad (11)$$

We will take $V_{ij} = g/r_{ij}^3$. Let $n_i \sim \langle n_i \rangle \sim 1$ and replace the sum by an integral over d^3r normalized by the volume Ω . This three-dimensional integral gives

$$\Delta E \sim \frac{g}{\Omega} \ln(R_2/R_1) \quad (12)$$

where R_1 and R_2 are the lower and upper limits of the radial integral. They are related to energy scales via $T_d \sim g/R_1^3$ and $E \sim g/R_2^3$. Making these substitutions and noting that $n(E) \sim 1/(\Delta E \cdot \Omega)$ yield the logarithmic density of states found in (9). This mean field argument is crude but it gives the same answer as more technically correct approaches.^{23,24} It is straightforward to remove the restriction to two energy levels per site and to include the coupling between transitions on two different sites. The important point is that the logarithm is a result of being in three dimensions.

This logarithmic density of states leads to corrections in the specific heat and the thermal conductivity. Instead of being linear in temperature, the specific heat behaves according to

$$C \sim \frac{T}{\ln(T_d/T)} \quad (13)$$

This is superlinear because the density of states rises slowly with increasing energy. As T or E increases, more states contribute to the specific heat than if $n(E)$ were flat. Thus the specific heat is superlinear in agreement with experiment. Similarly, the thermal conductivity goes as

$$\kappa \sim T^2 \ln(T_d/T) \quad (14)$$

This is subquadratic in agreement with experiment. As the density of states rises with increasing energy, so does the scattering rate or inverse mean free path. Hence, the thermal conductivity does not rise as fast as T^2 which it would do if $n(E)$ were constant. Another consequence of the logarithmic density of states is that, the 40% discrepancy between measurements of the sound velocity²⁵ and the ultrasonic attenuation is reduced if not resolved. Although

our expression for the density of states helps to explain why the attenuation is reduced at low temperatures, the value it gives for λ/l is still too large.

Diffusive Modes

As we mentioned, in this scenario the modes of the defect system are essentially collective and have no clear "two-state" characteristics. Is this compatible with a linear or nearly linear specific heat? The following example of a case in which we can plausibly get a linear specific heat from purely collective modes may be helpful. A more detailed version of the following argument can be found in the paper by one of us.²⁵ We start by considering a locally conserved quantity $A(r,t)$ which obeys the diffusion equation

$$\frac{\delta A(r,t)}{\delta t} = D \nabla^2 A(r,t) \quad (15)$$

where D is the diffusion constant. To satisfy this equation, $A(r,t)$ must be a quantity whose density is conserved but whose current is not. One choice which may be relevant to our problem is the local energy density. Within certain assumptions, it turns out that its minimum contribution to the thermodynamic energy is

$$E(T) \sim \sum_k \int d\omega \frac{1}{e^{\beta\omega} - 1} \left(\frac{\omega D k^2}{\omega^2 + (D k^2)^2} \right) \quad (16)$$

The Bose factor comes from the fluctuation-dissipation theorem. The term in parentheses is the imaginary part of the diffusive correlation function. At high temperatures Eq. (16) gives the classical Dulong-Petit specific heat. At low temperatures the exponential factor allows only small values of ω to contribute and the specific heat is linear in temperature. This suggests that a linear specific heat may be a consequence of considerations more general than those embodied in the two-level model. It is unclear whether it is possible to make a more concrete connection between this observation and the scenario for glasses sketched above. However, if Eq. (16) is to be consistent with the logarithmic density of states

of Eq. (9), there must be an anomalous diffusion constant, i.e., $D(k,\omega) = D_0(k/k_0) \ln^2(T_d/\omega)$.

Absence of a Phonon Bottleneck

Finally, we discuss an experiment which is naturally explained by our model of collective modes but not by two-level systems. This is the absence of a phonon bottleneck. To understand why we would expect a phonon bottleneck within the context of two-level system, consider a system of two-level systems with some of them having energy splitting $\hbar\Omega_0$. If we flood the system with monochromatic phonons of energy $\hbar\Omega_0$, these will be absorbed by resonant two-level systems and later reemitted. Since reabsorption and reemission will happen repeatedly, the phonons of energy $\hbar\Omega_0$ will be quite long-lived. This is a phonon bottleneck. However, such a bottleneck is not seen in glasses. Golding and Graebner found that the phonons equilibrate quite rapidly.²⁶ This is easy to understand using the concept of collective modes. Think of these modes as many-body states representing a band of energies. These states cannot be simply represented as a product of two-level systems. However, it is quite plausible that they couple resonantly to phonons which induce transitions between pairs of collective states. This results in experimental features which resemble those of two-level systems. However, unlike two-level systems, such a system excited by monochromatic phonons can decay to any one of a number of intermediate states accompanied by the emission of energy-conserving phonons. As a result of this decay process, we expect the phonons to equilibrate rapidly, and they do. A major question for the model is whether the structure of levels and matrix elements necessary for this explanation to work is compatible with the considerations given above concerning the effect of the long-range $1/r^3$ interactions.

4. SUMMARY

To summarize, the widely accepted model of two-level systems is inadequate in many respects. There is no experimental proof that such systems exist. The standard model fails to explain several

universal trends. In addition it ordinarily does not take into account interactions between two-level systems. We believe it is important to include interactions between defects from the very beginning and we have sketched preliminary work on a model which does this. We found that as the length scale increases, there is a natural crossover at ω_0 to a regime where long-range interactions dominate. This crossover is associated with the plateau in the thermal conductivity and the bump in C/T^3 . Interactions result in collective modes which have a logarithmic density of states that agrees with the low temperature specific heat, thermal conductivity and ultrasonic measurements. We also gave an example where a linear specific heat was due to diffusive modes rather than two-level systems or free electrons. The thoughts we have suggested do not in any sense constitute a theory. They are merely intended to provoke discussion and to demonstrate that the low temperature properties of glasses continue to pose a challenging problem.

Acknowledgments

We have enjoyed helpful discussions with A. C. Anderson, P. Bhattacharyya, J.-M. Duan, J. J. Freeman, Y. Fu, B. Golding, and J. E. Graebner. We would like to thank the Institute for Theoretical Physics at Santa Barbara for its hospitality from April to June of 1987 when some of this work was done. This work was supported in part by NSF grant DMR83-15550 and NSF grant PHY82-17853 supplemented by funds from NASA.

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