Universal properties of glasses in a pseudospin model

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The properties of a pseudospin model of an orientationally disordered crystal are studied. The low-temperature behavior of this model is determined by excitations which are equivalent to two-level systems in glasses. It thus becomes possible to explain the glasslike behavior observed in such materials.

The properties of glasses at low temperatures differ significantly from the properties of ordinary crystals.[1] The differences are seen primarily in the temperature dependence of the heat capacity ($C \sim T$), that of the thermal conductivity ($k \sim T^2$), and the logarithmic time dependence of the heat capacity at temperatures $T \lesssim 1$ K. At higher temperatures ($1 \lesssim T \lesssim 10$ K), there is a plateau ($k = \text{const}$) on the curve of the thermal conductivity, while there is a hump on the temperature dependence (C/T^3). In an effort to explain these anomalous features, it has been hypothesized that these materials contain two-level systems in which transitions between the levels occur by tunneling.[2,3] The two-level systems are characterized by a random energy ϵ and a random tunneling amplitude Δ_0, with a given distribution

$$P(\epsilon,\ \Delta_0) = P_0 \epsilon (\Delta_0)^{-1} (\epsilon^2 - \Delta_0^2)^{-1/2}. \tag{1}$$

At present, the microscopic origin of the two-level systems corresponding to (1) is not totally clear.

An important role in resolving this question may be played by a study of the properties of orientationally disordered crystals, which exhibit a glasslike behavior at low temperatures.[1] In these materials, the tunneling states are associated with impurities having rotational degrees of freedom. At low temperatures, transitions between the rotational levels of an individual impurity center (a rotator) are of a tunneling nature. The interaction of rotators gives rise to a spread of excitation energies. There is, on the other hand, no reason to expect the tunneling amplitudes of the various impurities to acquire a distribution of the type in (1) as a result of an interaction. Analysis of the experimental data on the $(KBr)_{1-x}(KCN)_x$ system, which is the system which has been studied best, shows[4] that the two-level systems responsible for the low-temperature anomalies in the properties cannot be identified with one-impurity excitations.

In this letter we use a simple pseudospin model of an orientationally disordered crystal to analyze some excitations which are more complex than one-impurity excitations. We show that these excitations are characterized by a distribution as in (1) if the interaction of the rotators falls off with the distance as R^{-3}.

We consider the rotational levels of an individual impurity center in the two-well approximation, associating with each impurity center a spin operator $S = 1/2$. The spins σ_i (the density of spins is n) are in a random external field $\omega(-\omega_i\sigma_i^z)$, which is distributed symmetrically and uniformly over the energy interval $(-W, W)$. Each spin interacts with the neighboring spins by a long-range interaction U_{ik} $(-U_{ik}\sigma_i^z\sigma_j^z)$, which falls off with the distance (r_{ik}) between spins i and k in accordance with $|U_{ik}| \approx U_0/r_{ik}^3$. The sign of the interaction can be either positive or negative; the important point is that the angular average of the interaction vanish. We assume that the minimum distance between spins is $(U_0/W)^{1/3}$. We take tunneling into account by introducing a weak transverse field Δ_0, the same for each spin $\Delta_0(\Delta_0\sigma^x;\ \Delta_0 < U_0 n$; and $U_0 n$ is the interaction of the spins at intermediate range).

Important for our purposes is the satisfaction of the following condition, which states that the interaction between spins is weak in comparison with the spread in random energies:

$$\xi < 1;\ \ \xi = U_0 n/W \ln(W/\Delta_0). \tag{2}$$

In particular, limitation (2) allows us to ignore the dipole gap in the density of excitations,[5] since it may come into play at energies $\epsilon \lesssim U_0 n \exp[-W/(U_0 n)]$, where a repulsion of levels plays an important role (more on this below).

Let us consider excitations of the system at very low temperatures:

$$T < \Delta_0. \tag{3}$$

We single out a subsystem of spins which are involved in excitations with low energies $\epsilon \lesssim \eta\Delta_0 (\eta \gtrsim 1)$, and we project the total Hamiltonian of the system onto the states of this subsystem. By singling out the low-energy excitations, we can restrict the discussion to the longitudinal (Ising) part of the Hamiltonian $(\tilde{H}_0)$, since the tunneling part $(\hat{V})$ redefines the excitation energies at the scale $\Delta_0 \lesssim \eta\Delta_0$. In the case $\Delta_0 = 0$ the state of the system is characterized by certain values of all spins $(\sigma_i^z = \sigma_i^{(0)})$. The simplest (one-particle) excitation arises in the system when a single spin flips $(\sigma_i^z = \sigma_i^{(0)} \Rightarrow \sigma_i^z = -\sigma_i^{(0)})$. This excitation is characterized by an energy

$$\Delta_i = -2\sigma_i^{(0)}(\omega_i + \sum_k U_{ik}\sigma_k^{(0)}). \tag{4}$$

A spin i is part of the subsystem if the condition of a low excitation energy, $\Delta_i \lesssim \eta\Delta_0$, is satisfied. The concentration of such spins (singles) is $n_i \simeq n\eta\Delta_0/W$. More complex excitations (doubles) are associated with the flip of two spins $(\sigma_i^z = \sigma_i^{(0)}$, $\sigma_k^z = \sigma_k^{(0)} \Rightarrow \sigma_k^z = -\sigma_k^{(0)}$, $\sigma_i^z = -\sigma_i^{(0)})$ and are characterized by an energy

$$\Delta_{ik}^{(2)} = \Delta_i + \Delta_k + 4U_{ik}\sigma_i^{(0)}\sigma_k^{(0)}. \tag{5}$$

We are interested in only those pairs for which the interaction lowers the excitation energy $(U_{ik}\sigma_i^{(0)}\sigma_k^{(0)} < 0)$. In this case the energy $\Delta_{ik}^{(2)}$ may be small in comparison with Δ_0, under the conditions $\Delta_i,\ \Delta_k \gg \Delta_0$. Pairs of spins which have this property should also be included in the subsystem. The concentration of spins which are parts of pairs,

$n_2 \simeq n_1 \xi$, is small in comparison with the concentration of singles, by virtue of inequality (2). The contributions of triple excitations and of more complex excitations are small quantities of even higher order in the parameter ξ, and we will ignore them. Double excitations must be taken into account in the temperature range in (3), because of the pronounced tunneling-induced splitting of the energy of one-impurity excitations.

In the Hamiltonian projected onto states of the subsystem for the pair in (5), we should consider only the states $\sigma_i^z = \sigma_i^{(0)}$, $\sigma_k^z = \sigma_k^{(0)}$; $\sigma_k^z = -\sigma_k^{(0)}$, $\sigma_i^z = -\sigma_i^{(0)}$, which we will describe by means of an effective spin σ_{ik}, whose projections $\sigma_{ik}^z = \pm 1/2$ correspond to these states. In the temperature range in (3), the spins which are not part of the subsystem are in a state corresponding to the lowest energy [the probability for the excited state is exponentially small: $W_+ < \exp(-\eta\Delta_0/T)$. Incorporating these spins in the Ising approximation leads to simply a redefinition of the random field acting on the spins in the subsystem. As a result, we are left with a system of singles and doubles which are interacting weakly (because of the low concentrations, n_1, $U_0 n_1 \ll \Delta_0$). The single–double and double–double interactions differ negligibly from the seed interaction of singles; incorporating these differences does not alter the results.

We deal with the tunneling by perturbation theory. In a first approximation, there is a tunneling of singles, $\hat{V}_1 = \Sigma' \Delta_0 \sigma_i^x$ (the summation here is over only those spins which are part of the subsystem). Since the interaction of singles at intermediate range is weak in comparison with their characteristic energy Δ_0, and since singles which lie close to each other constitute doubles, we can assume that the singles are isolated in calculating the spectrum of excitations. The energy of an isolated spin cannot be lower than Δ_0, because of the tunneling-induced splitting. At the temperatures in (3), the contribution of singles to the thermodynamic characteristics of the system is thus exponentially small and can be ignored in comparison with the contribution of double excitations, for which the tunneling-induced splitting is weaker and arises only in second-order perturbation theory. The tunneling part of the Hamiltonian for the doubles is[5]

$$\hat{V}_2 = \Sigma' \Delta_{0ik}^{(2)} \sigma_{ik}^x, \quad \Delta_0^{(2)} \approx U_0 \Delta_0^2 / (\Delta_i \Delta_k r_{ik}^3). \tag{6}$$

To eliminate the "nonphysical" increase in the double tunneling amplitude as Δ_i, $\Delta_k \Rightarrow 0$, we introduce the limitations Δ_i, $\Delta_k > \Delta_0$. These limitations reflect the repulsion of levels. In second- and higher-order perturbation theories, corrections to the interaction U and to the tunneling amplitude Δ_0 which are small, proportional to the parameter ξ, arise. We will ignore them.

Let us examine the double excitations in more detail. Their interaction is also weak, because of their low concentration n_2, and we can use the approximation that the doubles are isolated. Each double is characterized by a tunneling amplitude $\Delta_0^{(2)}$ and an energy $\epsilon^{(2)} = [(\Delta_0^{(2)})^2 + (\Delta^{(2)})^2]^{1/2}$. The double excitations are described by a distribution with respect to energy and tunneling amplitude. This distribution is found by integration:

$$P(\epsilon^{(2)}, \Delta_0^{(2)}) \approx n^2/W^2 \int_{r_{\min}} dr_{ik} \int_{\Delta_0} d\epsilon_i \int_{\Delta_0} d\epsilon_k$$

$$\times \delta(\epsilon^{(2)} - ((\epsilon_i + \epsilon_k - U_0/r_{ik}^3)^2 + (\Delta_0^{(2)})^2)^{1/2}) \tag{7}$$

$$\times \delta(\Delta_0^{(2)} - U_0\Delta_0^2/(\epsilon_i\epsilon_k r_{ik}^3)), \quad r_{\min} = (U_0/W)^{1/3}.$$

At the energies of interest here, $\epsilon^{(2)} < \Delta_0$, we find

$$P(\epsilon^{(2)}, \Delta_0^{(2)}) \approx \tilde{P}_0 \epsilon^{(2)} (\Delta_0^{(2)})^{-1} ((\epsilon^{(2)})^2 - (\Delta_0^{(2)})^2)^{-1/2},$$

$$\Theta(\Delta_0 - \Delta_0^{(2)})\Theta(\Delta_0^{(2)} - \Delta_0^2/W), \quad \tilde{P}_0 = nU_0W^{-2}. \tag{8}$$

Distribution (8) coincides with distribution (1), for two-level systems in glasses, over a wide energy range. It follows that in the temperature range in (3) the heat capacity of the system should exhibit the anomalies in the temperature and time dependence which are characteristic of glasses. The low-temperature thermal conductivity can be estimated by working from the one-phonon interaction of spins with the medium, which is diagonal in σ^z. As a result, we find the known result for glasses: $k \sim T^2$. Our model can explain the plateau in the thermal conductivity and the hump in the heat capacity at temperatures $T > \Delta_0$. Specifically, excitations of singles, whose concentration is greater than that of doubles, come into play at these temperatures. The scattering of phonons strengthens sharply, and the growth of the thermal conductivity with the temperature should come to a halt. The hump in the heat capacity can also be associated with an increase in the concentration of excitations.

In summary, we have shown that the model of pseudospins with an r^{-3} interaction in a strong random field (a similar model was recently proposed by Yu and Leggett[6] to describe glasses) demonstrates a glasslike behavior at sufficiently low temperatures and can explain the low-temperature properties of the glasslike phase of orientationally disordered crystals.

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