Density of states for interacting tunneling units in the absence of long-range order

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We consider the effect of interactions between two-level (four-level) tunneling states. We allow the tunneling units to interact via a potential $J(r) = \pm a/r^n$, where a is a constant and r is the distance between the tunneling units, obtain the free energy $\bar{F}$ up to and including the second virial coefficient, and neglect higher virial coefficients. We derive the density $P(E)$ of the elementary excitation energies E arising from $\bar{F}$ exactly for a random distribution of tunneling units. We obtain that $P(E) \propto E^{(6-2n)/n}$ for low E. The specific heat $C_p \propto T^{(6-n)/n}$ and the thermal conductivity $\kappa \propto T^{(4n-6)/n}$ for low temperatures T such that the product $\kappa C_p \propto T^3$ independent of n. In particular, for $n = 3$ we find that $P(E)$ is approximately constant for low E and that $C_p \propto T$ and $\kappa \propto T^2$ for low T. Our calculations also give a large T^3 term in C_p and a somewhat flatter portion in κ for higher temperatures, both of these arising from the interacting tunneling units. The low-T dielectric susceptibility χ is predicted to have a $-\ln T$ term in it, provided that $T > T_0$, where T_0 depends on the tunneling matrix element and the interaction between a pair of near-neighbor tunneling units. For $T \ll T_0$, both C_p and κ are predicted to be proportional to $\exp(-\text{const}/T)$ and χ approaches a constant value.

I. INTRODUCTION

The low-temperature thermodynamic properties of tunneling states have received a great deal of interest recently. The primary reason for this is that many different materials exhibit a broad spectrum of low-energy excitations which have been attributed to originate from tunneling states. Such materials include glasses,[1-3] amorphous solids,[4] and tunneling impurities in alkali halides.[5,6]

The low-temperature properties of glasses[1-3] have been described in terms of two-level tunneling states, which are characterized by an asymmetry energy ϵ and a tunneling energy Δ. A constant density of states for the low-energy excitations had to be postulated in order to obtain the experimentally observed results on the thermal conductivity and the specific heat. It has been suggested that this constant density of states arises from essentially isolated tunneling states. Its existence has not been justified from a fundamental point of view and is one of the outstanding problems in the theory of glasses.

Tunneling impurities in alkali halides have been explored in the limit of infinite dilution.[7,8] It was found that the impurities tunnel between a number of equivalent directions of orientation determined by the host crystal lattice and by the impurity potential.[9] One such tunneling impurity is, for example, the CN^- ion dissolved in various alkali halides.[10] Whereas in the case of infinite dilution it is expected that the tunneling impurities give rise to the well-known Schottky anomaly in the low-temperature specific heat, as the impurity concentration is increased the density of states develops into a broad distribution of low-energy excitations, somewhat similar to that observed in glasses, as was demonstrated in recent experiments on $KBr_{1-x}(CN^-)_x$.[10,11] This broad distribution arises from the interactions between the tunneling units, presumably in distinction to the broad distribution in glasses, which has been attributed to essentially isolated noninteracting tunneling states. The purpose of this paper is to study the effect of interactions between the tunneling units and to obtain the density of states of the elementary excitations under certain idealized conditions outlined below.

We consider a set of two-level and four-level tunneling units which are characterized by a tunneling matrix element Δ and interact via a potential J_{ij} between tunneling units at sites i and j. The interaction potential may, for example, arise from either a strain-strain interaction[12,13] having a form $J_{ij} = a(\theta, \phi)/r_{ij}^{-n}$, where r_{ij} is the distance between the tunneling units at sites i and j, and $a(\theta, \phi)$ is a function depending upon the polar angle θ and the azimuthal angle ϕ between the strain or electric dipole moments.

For simplicity, our calculations are done assuming that $J_{ij} = \pm a/r_{ij}^{-n}$, where a is a constant and the plus and minus signs have equal probabilities. In Appendix A we show that the qualitative results for the density of states are not changed by this assumption. Furthermore, we assume that the interacting tunneling units are randomly and uniformly distributed throughout the volume of the solid and that there is no long-range order between the interacting tunneling units. The absence of long-range order can be qualitatively justified on the basis that (i) we are dealing with a competing interaction potential with equal probability for each sign of the potential, (ii) the concentration of the tunneling units is sufficiently low, and (iii) the transverse fields arising from the tunneling matrix elements Δ inhibit the phase transition. The assumption of no long-range order would probably not be valid for a noncompeting potential, unless Δ is sufficiently large.

Using the above assumptions we expand the free energy into a virial expansion, keeping terms up to and including the second virial coefficient and neglecting all higher-

order contributions. Using this truncated free energy, we obtain the density of states $P(E)$ of the elementary excitation energies E exactly for a fixed value of Δ and for $J_{ij} = \pm a / r_{ij}^{-n}$. We find that for very low energies, $P(E) \propto E^{(6-2n)/n}$. In particular, when $n = 3$, which is the case for a strain-strain[12,13] or electric dipole interaction, we find that $P(E) \propto [1 + 3(E/\Delta)^2 + \cdots]$; thus $P(E)$ is approximately constant for low E. From the density of states we calculate the low-temperature specific heat C_p, the thermal conductivity κ, and the dielectric susceptibility χ. We find that for low temperatures T, $C_p \propto T^{(6-n)/n}$ $(1 + A_2 T^2)$ and $\kappa \propto T^{(4n-6)/n}(1 - A_3 T^2)$, where A_2 and A_3 are positive constants. It is interesting to note that for $n = 3$ the leading term in C_p is proportional to T and the leading term in κ is proportional to T^2, not unlike the experimental observations in glasses.[4] We also find that there is a large excess T^3 contribution to the specific heat in addition to the usual Debye contribution, and also that there is a shoulder, or flat portion, in the thermal conductivity because of the increased density of states with increasing energy. The low-temperature dielectric constant is predicted to have a $-\ln T$ dependence in it. All of these results obtained are for low temperatures, but with T still greater than some T_0, where T_0 depends on both the interaction between two near-neighbor tunneling units and Δ. For $T \ll T_0$, C_p and κ both become proportional to $\exp(-\text{const}/T)$, and $\chi \to \text{const}$.

The contents of this paper is briefly as follows. In Sec. II we derive the expression for the free energy and the density of states for both the two-level and four-level interacting tunneling units. We find that the density of states has identical qualitative behavior for the two cases. In Sec. III we derive the expression for the low-temperature specific heat, the thermal conductivity, and the dielectric constant.

II. DERIVATION OF FREE-ENERGY AND THE DENSITY-OF-STATES

A. Free energy of the interacting tunneling units

In this subsection we calculate the free energy of the interacting tunneling units up to the second virial coefficient. We consider a two-level tunneling unit with the bottom of the two wells differing by an energy 2ϵ, where ϵ is the asymmetry energy. Let the states of the system be specified by $| R \rangle$ and $| L \rangle$ representing the states when the tunneling unit is in the right and left wells, respectively. With the use of the complete analogy between this problem and the two-level spin-$\frac{1}{2}$ system, the Hamiltonian H_1 for tunneling unit 1 is found to be[14]

$$H_1 = -\epsilon_1 \sigma_1^z - \Delta_1 \sigma_1^x , \qquad (2.1)$$

where the σ's are the Pauli spin matrices. The $| \uparrow \rangle$ and $| \downarrow \rangle$ represent the spin-up (right) and spin-down (left) states. The last term in Eq. (2.1) allows tunneling between the $| \uparrow \rangle$ and $| \downarrow \rangle$ states, and Δ_1 is the overlap energy due to tunneling.

We now introduce a second two-level tunneling unit and allow a pair interaction J_{ij} between them. We assume that if both tunneling units are in the same state (both right or left) the energy is lower (or possibly higher) by J_{ij} compared with the case when they are in the opposite states (one right, one left). The right and left states are chosen arbitrarily. For a strain-strain or electric dipole interaction we are dealing with a J_{ij} which is on the average equally likely to be positive or negative. The Hamiltonian H_{12} for the pair of two-level tunneling units 1 and 2 becomes

$$H_{12} = -\epsilon_1 \sigma_1^z - \epsilon_2 \sigma_2^z - \Delta_1 \sigma_1^x - \Delta_2 \sigma_2^x - J_{12} \sigma_1^z \sigma_2^z . \qquad (2.2)$$

By excluding the terms of the form $\sigma_1^x \sigma_2^x$ we exclude processes in which both units tunnel simultaneously. Even though such terms may be important for the dynamics of the system, here we are interested primarily in the static properties.

For N tunneling units, the Hamiltonian becomes

$$H_0 = -\frac{1}{2} \sum_{i,j} J_{ij} \sigma_i^z \sigma_j^z - \sum_i \epsilon_i \sigma_i^z - \sum_i \Delta_i \sigma_i^x . \qquad (2.3)$$

In Eq. (2.3) it is explicitly assumed that Δ_i and ϵ_i will not be affected by the interaction J_{ij} between the tunneling units. In a realistic physical situation this assumption may not be valid[15] because a strong strain interaction between the units may distort the local environment at site i.

We next expand the free energy F into a virial expansion, keeping terms only up to the second virial coefficient. We write

$$F = F^{(1)} + F^{(2)} + F^{(3)} + \cdots \qquad (2.4a)$$

$$= \left\langle \sum_i f_i + \sum_{i,j} f_{ij} + \sum_{i,j,k} f_{ijk} + \cdots \right\rangle_c , \qquad (2.4b)$$

where $F^{(l)}$ is the contribution to the free energy from the lth virial coefficient, and the angular brackets $\langle \ \rangle_c$, indicate a configurational average over all the random values of J_{ij}. f_i represents the one-vertex terms, f_{ij} represents the two-vertex terms, and f_{ijk} represents the three-vertex terms in the free energy. We will only keep the first two terms in Eq. (2.4b), denoting these as the truncated free energy $\bar{F}$. We have

$$-\beta F^{(1)} = -\beta \sum_i \langle f_i \rangle_c = \sum_i \langle \ln \text{tr} \exp(-\beta H_i) \rangle_c , \qquad (2.5)$$

where tr represents the trace, and H_i is given in Eq. (2.1). Equation (2.5) is readily evaluated to give

$$-\beta F^{(1)} = \left\langle \sum_i \ln\{ 2 \cosh[\beta(\epsilon_i^2 + \Delta_i^2)^{1/2}] \} \right\rangle_c . \qquad (2.6)$$

The expression for $F^{(2)}$ is somewhat more complicated since we have to include all two-vertex contributions to the free energy. We do this by expanding the free energy into a cumulant expansion and resumming all two-vertex terms. The result is

$$-\beta F^{(2)} = \sum_{i,j} \langle \ln \text{tr} \exp(-\beta H_{ij})$$
$$- \ln \text{tr} \exp[-\beta(H_i + H_j)] \rangle_c , \qquad (2.7)$$

where H_i is given by Eq. (2.1) and H_{ij} is given by a generalization of Eq. (2.2).

The four eigenvalues λ_ν ($\nu = 1$ to 4) of the H_{ij} are obtained by solving the 4×4 matrix

$$\underline{H}_{ij} = \begin{bmatrix} \underline{A}_{ij} & -\underline{B}_{ij} \\ -\underline{B}_{ij} & \underline{C}_{ij} \end{bmatrix} , \tag{2.8}$$

with

$$\underline{A}_{ij} = \begin{bmatrix} -J_{ij} - \epsilon_i - \epsilon_j & -(\Delta_j + \Delta_i)/2 \\ -(\Delta_j + \Delta_i)/2 & J_{ij} - \epsilon_i + \epsilon_j \end{bmatrix} , \tag{2.9a}$$

$$\underline{B}_{ij} = \frac{1}{2} \begin{bmatrix} \Delta_i + \Delta_j & 0 \\ 0 & \Delta_i + \Delta_j \end{bmatrix} , \tag{2.9b}$$

$$\underline{C}_{ij} = \begin{bmatrix} J_{ij} + \epsilon_i - \epsilon_j & -(\Delta_i + \Delta_j)/2 \\ -(\Delta_i + \Delta_j)/2 & -J_{ij} + \epsilon_i + \epsilon_j \end{bmatrix} . \tag{2.9c}$$

In principle, the eigenvalues of H_{ij} can be obtained for all ϵ_i and ϵ_j. We will, however, be interested in the limit as $\epsilon_i = \epsilon_j \rightarrow 0$, and $\Delta_i = \Delta_j = \Delta$. This assumption limits this paper to a consideration of two-level tunneling with zero asymmetry energy and equal values of Δ, with a distribution of the interaction energy J_{ij}. An extension of our results to an assumed identically distributed Δ is quite straightforward. Similarly, the qualitative behavior of our low-T results are unchanged by allowing the values of Δ_i to be different at each site i, as will be seen below from the four eigenvalues of H_{ij} when $\Delta_i \neq \Delta_j$. However, the asymmetry energy ϵ must remain zero for our results to be valid, since otherwise the low-T specific heat becomes proportional to $\exp(-2\epsilon/T)$.

The four eigenvalues of H_{ij} are, for positive J_{ij}, in order of increasing energies,

$$\lambda_1^{(0)} = -(J_{ij}^2 + 4\Delta^2)^{1/2} , \tag{2.10a}$$

$$\lambda_2^{(0)} = -J_{ij} , \tag{2.10b}$$

$$\lambda_3^{(0)} = +J_{ij} , \tag{2.10c}$$

$$\lambda_4^{(0)} = (J_{ij}^2 + 4\Delta^2)^{1/2} . \tag{2.10d}$$

For $\Delta_i \neq \Delta_j$ the four eigenvalues λ_ν' ($\nu = 1$ to 4) of Eqs. (2.10) become

$$\lambda_1' = -[J_{ij}^2 + (\Delta_i + \Delta_j)^2]^{1/2} ,$$

$$\lambda_2' = -[J_{ij}^2 + (\Delta_i - \Delta_j)^2]^{1/2} ,$$

$$\lambda_3' = [J_{ij}^2 + (\Delta_i - \Delta_j)^2]^{1/2} ,$$

and

$$\lambda_4' = [J_{ij}^2 + (\Delta_i + \Delta_j)^2]^{1/2} .$$

We find that use of the eigenvalues λ_ν, i.e., $\Delta_i \neq \Delta_j$, does not change the qualitative result obtained for the low-temperature thermal properties of the system.

Figure 1 shows the four eigenvalues of H_{ij} as a function of the interaction potential J_{ij}. We note some interesting results exhibited in this figure. For $J_{ij} = 0$ the energy difference between the ground state and first excited state, i.e., the excitation energy, has a gap of 2Δ. As J_{ij} increases, the excitation energy decreases and goes to zero proportionally to Δ^2/J_{ij} for $J_{ij} \rightarrow \infty$. This shows that, whereas the noninteracting tunneling units have a gap in their excitation energies, the interacting ones have very-low-energy excitations, provided J_{ij} is sufficiently
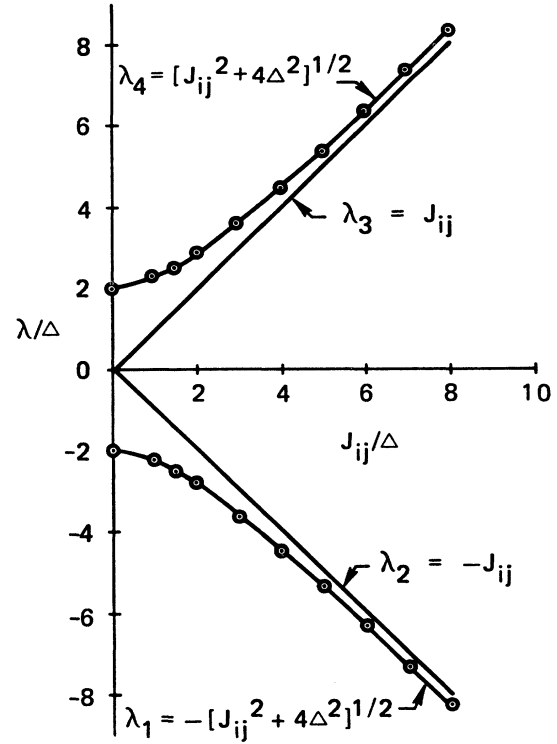

FIG. 1. Four eigenvalues of the Hamiltonian H_{12} as a function of the interaction potential J_{ij} between the pair of tunneling states. We note that the difference between the two lowest eigenvalues becomes smaller as J_{ij} increases.

large. The physical meaning of this result is that for fixed Δ, the strongly interacting pairs will mostly contribute to the low-temperature thermodynamic properties of the tunneling states. This shows that the interacting tunneling states will have a completely different excitation spectrum from a *classical* magnetic system (when spin waves are neglected). In the latter, the elementary excitations will have large energies for large J_{ij}, whereas in the former they will not. The understanding of the physics presented in this paper is simpler if this important difference between the energies of the interacting pair of tunneling states and the interacting pair without tunneling is realized at the outset.

The density of the low-energy excitations depends on, and is uniquely determined by, the probability distribution of the J_{ij}'s. Using the eigenvalues, Eqs. (2.10), with Eq. (2.7), we obtain, for $\epsilon = 0$,

$$-\beta F^{(2)} = \sum_{i,j} \langle \ln 2\{ \cosh(\beta J_{ij}) + \cosh[\beta(J_{ij}^2 + 4\Delta^2)^{1/2}] \}$$

$$-2\ln[2\cosh(\beta\Delta)] \rangle_c . \tag{2.11}$$

We let the effective number of neighbors be z_0, and assume that all the J_{ij}'s have identical probability distributions; Eq. (2.11) then becomes

$$-\beta F^{(2)} = \frac{Nz_0}{2} \langle \ln[\cosh(\beta E_+)\cosh(\beta E_-)]$$

$$-2\ln\cosh(\beta\Delta) \rangle_c , \qquad (2.12)$$

where

$$E_\pm = [(J_{ij}^2 + 4\Delta^2)^{1/2} \pm J_{ij}]/2 . \qquad (2.13)$$

We note that E_- is one-half of the difference between the ground-state and first-excited-state energies for positive J_{ij}, and E_+ is the same quantity for negative J_{ij}. We also remark that $F^{(2)}$ goes to zero in the limit as $J_{ij} \to 0$, as it should.

B. Density of states

To evaluate the configurational-average free energy of the pairs given by Eq. (2.12), we have to obtain the density of the excitation energies E_+ and E_-. For this purpose we assume that the tunneling units are randomly and uniformly distributed over the volume of the pair, $V_0 = \frac{4}{3}\pi r_0^3$, where r_0 is some effective mean distance between the pairs. A reasonable assumption for V_0 would be that if we put one impurity at the origin, the second impurity should, on the average, just be within V_0. If we let r_1 be the near-neighbor distance, then we make the reasonable assumption that

$$r_0^3 = r_1^3/c , \qquad (2.14)$$

where c is the fractional concentration of the tunneling states. c is assumed to be small compared to unity. We also assume that the excluded volume due to the limitation in the distance of closest approach of a near neighbor is $V_1 = \frac{4}{3}\pi r_1^3$. (In a realistic case the excluded volume will depend upon the lattice structure and will not be $4\pi r_1^3/3$.)

We next consider a random potential which is of the form $J_{ij} = a(\theta,\phi)/r_{ij}^n$, where $a(\theta,\phi)$ is a function which depends upon the angles θ and ϕ between the interacting strain dipoles or electric dipoles.[13] For example, for a dipole-dipole interaction, $a(\theta,\phi) \propto (1 - 3\cos^2\theta)$. $a(\theta,\phi)$ is assumed to have a form such that

$$\int a(\theta,\phi)\sin\theta\, d\theta\, d\phi = 0 ,$$

i.e., that its average over all angles vanishes. Under these conditions, we show in Appendix A that the density of the excitation energies can be obtained by using the expression for J_{ij} of the form

$$J_{ij} = \pm a/r_{ij}^{-n} , \qquad (2.15)$$

with each sign having a probability of $\frac{1}{2}$. We let the near-neighbor and largest value of J_{ij} be $J_1 = a/r_1^3$, and also let $J_0 = a/r_0^3$. Using the relations $P_1(V) = (V_0 - V_1)^{-1}$ and $P_2(J) = P_1(V)|dJ/dV|^{-1}$, we have

$$P_2(J) = \frac{4\pi/n}{2(V_0 - V_1)} |a|^{3/n} |J|^{-(3+n)/n} , \qquad (2.16)$$

with $J_0 < |J| < J_1$. The factor of 2 in the denominator of Eq. (2.16) arises from the consideration of the positive

as well as negative values of J. The use of the equation $E_\pm = [(J^2 + 4\Delta^2)^{1/2} \pm J]/2$ gives $J = \pm(E_\pm^2 - \Delta^2)/E_\pm$. We let

$$A_\pm = [(J_1^2 + 4\Delta^2)^{1/2} \pm J_1]/2 , \qquad (2.17a)$$

$$B_\pm = [(J_0^2 + 4\Delta^2)^{1/2} \pm J_0]/2 , \qquad (2.17b)$$

and then $P(E_\pm) = P_2(J)|dE_\pm/dJ|^{-1}$. We obtain

$$P_\pm(E_\pm) = \frac{(4\pi/n)|a|^{3/n}}{V_0 - V_1} \frac{E_\pm^{2(3-n)/n}(E_\pm^2 + \Delta^2)}{|E_\pm^2 - \Delta^2|^{(3+n)/n}} , \qquad (2.18)$$

where in Eq. (2.18), $A_- \le E_- \le B_-$ and $B_+ \le E_+ \le A_+$.

Equations (2.17) and (2.18) can be combined into a single expression as follows:

$$P(E) = \frac{(2\pi/n)|a|^{3/n}}{V_0 - V_1} \left[\frac{1}{(E-\Delta)^2} + \frac{1}{(E+\Delta)^2} \right]$$

$$\times \left[\frac{E^2}{|E^2 - \Delta^2|} \right]^{(3-n)/n} , \qquad (2.19)$$

with the energy E restricted to two regions, $A_- \le E \le B_-$ and $B_+ \ge E \ge A_+$. Equation (2.19) gives the density of states $P(E)$ for a general r^{-n} potential. In particular, for $n = 3$, we have

$$P(E) = \frac{2\pi|a|}{3(V_0 - V_1)} \left[\frac{1}{(E-\Delta)^2} + \frac{1}{(E+\Delta)^2} \right] . \qquad (2.20)$$

Equation (2.20) shows a large increase in the density of states near $E \to \Delta$, and a divergence for $E = \Delta$. However, examining Eq. (2.13) gives $E = \Delta$ only when $J = a/r^3 = 0$, which occurs only when the distance between the tunneling states approaches infinity. We can understand the increase in $P(E)$ near $E \to \Delta$ by considering the limit as $V_0 - V_1 \to \infty$. As the distance between the tunneling states increases, more and more pairs will have small values of J, and since the number having small J is proportional to r^3 the density of states becomes strongly peaked about $E = \Delta$. Thus as $c \to 0$, $P(E)$ becomes more and more concentrated near $E = \Delta$. Thus

$$\lim_{c \to 0} P(E) \approx \delta(E - \Delta) . \qquad (2.21)$$

Equation (2.21) can be shown as arising from Eq. (2.20) using the appropriate limits of integration.

From Eq. (2.20) we note the interesting results that, for an r^{-3} potential, the density of elementary excitations is approximately constant for energies $(E/\Delta)^2 \ll 1$. We have

$$P(E) = \frac{2\pi|a|}{3(V_0 - V_1)\Delta^2} \left[1 + \frac{3E^2}{\Delta^2} + O(E^4) \right] . \qquad (2.22)$$

A graph for the logarithm of the density of states as a function of the energy E for $n = 3$ is shown in Fig. 2. The approximately constant density of states for the very-low-energy elementary excitations arises naturally in our calculation when it is assumed that the tunneling states interact via a r^{-3} potential. This model is, to our knowledge, the first microscopic model that predicts a constant density of states of the elementary excitations
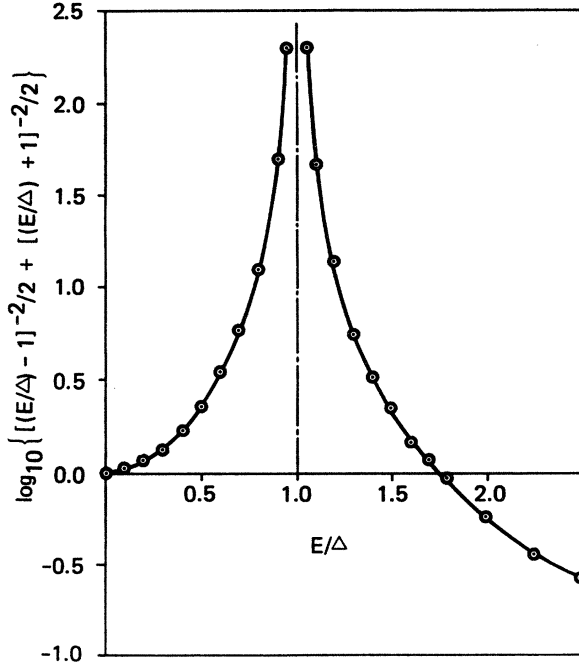
FIG. 2. Logarithm of the density of states $P(E)$ as a function of the energy E for a fixed value of the tunneling energy. $P(E)$ is multiplied by a constant K such that $KP(E)=1$ for $E=0$.
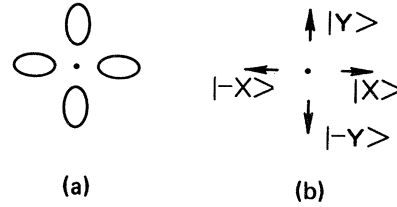

FIG. 3. (a) Shows the four potential minima assumed for the tunneling states. The depths of the four potentials are assumed to be identical. (b) Graphical representation of the four states. The states are, for convenience, designated as $|x\rangle$, $|-x\rangle$, $|y\rangle$, and $|-y\rangle$.

from fundamental considerations. We also remark that for $n=3$, one-half of the states have energies less than Δ and one-half have energies greater than Δ. By examining Eq. (2.19) we find that the density of states $P(E)$ is proportional to $E^{(6-2n)/n}$ for general n and $E \ll \Delta$.

C. Interacting four-level tunneling units

In Eqs. (2.19) and (2.20) we derive the energy levels and the density of the low-energy excitations for the interacting two-level tunneling units. In this subsection we derive the energy levels and the density of states for the interact-

ing four-level tunneling units for the following simplified model. Consider a completely symmetric tunneling unit in potential wells of equal depth such that it may take four positions: up, down, to the right and to the left; see Fig. 3(a). The tunneling unit is allowed to perform 90° tunneling only (neglecting 180° tunneling). The simplest way to represent the statics of this problem is by considering a four-orientational classical "dipole" whose orientations are shown in Fig. 3(b). Let the four states of the single tunneling unit be designated by $|x\rangle$, $|-x\rangle$, $|y\rangle$, and $|-y\rangle$, as shown in Fig. 3(b). Since the dipoles are allowed to tunnel through 90° degrees, a tunneling matrix element Δ connects the $|\pm x\rangle$ states with the $|\pm y\rangle$ states. Such a connection has to be built into the Hamiltonian. It is assumed that no matrix elements connect the $|x\rangle$ state with the $|-x\rangle$ state, or the $|y\rangle$ with the $|-y\rangle$ state, thus eliminating 180° tunneling.

The Hamiltonian $H_1(4)$ for the single four-level tunneling unit is for wells of identical depth,

$$H_1(4)=-\tfrac{1}{2}\Delta(|x_1\rangle+|-x_1\rangle)(\langle y_1|+\langle -y_1|)+\text{H.c.} ,$$

(2.23)

where H.c. is the Hermitian conjugate of the first part of $H_1(4)$.

The Hamiltonian $H_{12}(4)$ between the pair of four-level interacting tunneling units is

$$H_{12}(4)=-\frac{\Delta}{2}\{[(|x_1\rangle+|-x_1\rangle)(\langle y_1|+\langle -y_1|)+(|x_2\rangle+|-x_2\rangle)(\langle y_2|+\langle -y_2|)]+\text{H.c.}\}$$

$$-J_{12}|x_1\rangle|x_2\rangle\langle x_2|\langle x_1|-J_{12}|y_1\rangle|y_2\rangle\langle y_2|\langle y_1|-J_{12}|-x_1\rangle|-x_2\rangle\langle -x_2|\langle -x_1|$$

$$-J_{12}|-y_1\rangle|-y_2\rangle\langle -y_2|\langle -y_1|+J_{12}|-x_1\rangle|x_2\rangle\langle x_2|\langle -x_1|$$

$$+J_{12}|x_1\rangle|-x_2\rangle\langle -x_2|\langle x_1|+J_{12}|-y_1\rangle|y_2\rangle\langle y_2|\langle -y_1|+J_{12}|y_1\rangle|-y_2\rangle\langle -y_2|\langle y_1| .$$

(2.24)

Again, as in the case of the two-level tunneling states, we neglect processes in which both units tunnel simultaneously.

We form the pair states using the orthogonality of $|x\rangle$, $|-x\rangle$, $|y\rangle$, and $|-y\rangle$, and obtain the energies of the four-level pair by diagonalizing the 16×16 matrix

representing the pair states. The matrix is given in Appendix B. The final result for the partition function Z_4 is from Eq. (A5) of Appendix B,

$$Z_4=4(\cosh(\beta J/2)+\cosh\{\beta[(J/2)^2+4\Delta^2]^{1/2}\})^2$$

$$=[4\cosh(\beta U_+)\cosh(\beta U_-)]^2 ,$$

(2.25)

where

$$U_{\pm} = \{[(J/2)^2 + 4\Delta^2]^{1/2} \pm J/2\}/2 . \tag{2.26}$$

Comparing Eq. (2.26) with Eq. (2.13) we obtain the expression for the partition function of the four orientational states in terms of the partition function Z_2 of the two orientational states,

$$Z_4(J) = [Z_2(J/2)]^2 . \tag{2.27}$$

This result shows that the qualitative behavior of the two-orientational and four-orientational tunneling states is the same. The excitation energy U_- approaches zero for large positive values of J, and U_+ approaches zero for large negative values of J. For an interaction potential $J_{ij} = \pm a/r^3$, the density of states $P(U_{\pm})$ becomes

$$P(U_{\pm}) = \frac{2\pi |a|}{6(V_0 - V_1)} \left[\frac{1}{(U_{\pm} - \Delta)^2} + \frac{1}{(U_{\pm} + \Delta)^2} \right] , \tag{2.28}$$

with the values of A and B in Eqs. (2.17) redefined such that $J_1 \rightarrow J_1/2$ and $J_0 \rightarrow J_0/2$. We thus obtain that the density of the low-energy excitations from a pair of four-level tunneling states is also approximately constant for $U_- \ll \Delta$, provided the interaction $J_{ij} = \pm a/r_{ij}^3$.

To obtain the free energy for the pairs we must use Eq. (2.7). The result $-\beta F_4^{(2)}$ for the four-level states is obtained from Eqs. (B7) and (B12) of Appendix B to give

$$-\beta F_4^{(2)} = \sum_{i,j} \langle \ln[4\cosh(\beta U_+)\cosh(\beta U_-)]^2$$

$$-\ln[2\cosh(\beta\Delta)]^2 \rangle_c , \tag{2.29}$$

with the probability density of $U_{\pm}$ given by Eq. (2.28). A comparison of Eq. (2.29) with Eqs. (2.12) and (2.28) with

Eq. (2.20) again shows the similar behavior of the pair of two-orientational and four-orientational tunneling units.

III. DERIVATION OF THE LOW-TEMPERATURE SPECIFIC HEAT, THERMAL CONDUCTIVITY, AND DIELECTRIC CONSTANT

A. Low-temperature specific heat

We let the specific heat C_p be separated into two parts,

$$C_p = C_1 + \bar{C}_2 , \tag{3.1}$$

where C_1 is the contribution from $F^{(1)}$, and C_2 is the contribution from $F^{(2)}$. In the limit as $\epsilon_i \rightarrow 0$ and $\Delta_i = \Delta$, we obtain

$$C_1 = Nk_B(\beta\Delta)^2\text{sech}^2(\beta\Delta) . \tag{3.2}$$

This result gives the well-known Schottky anomaly in the specific heat.

The contribution from the second virial coefficient $\bar{C}_2$ is

$$\bar{C}_2 = \tfrac{1}{2}Nz_0 k_B \beta^2 \langle E_+^2 \text{sech}^2(\beta E_+) + E_-^2 \text{sech}^2(\beta E_-)$$

$$-2\Delta^2\text{sech}^2(\beta\Delta) \rangle_c ,$$

$$\bar{C}_2 \equiv C_2 - Nz_0 k_B(\beta\Delta)^2\text{sech}^2(\beta\Delta) , \tag{3.3}$$

where C_2 is the contribution to the specific heat arising from the interacting term only.

Combining the integration over E_+ and E_- into a single integral, the density of states for a general power-law potential is given by Eq. (2.19), and for a r^{-3} potential it is given by Eq. (2.20). We obtain

$$\bar{C}_2 = Nz_0 k_B \beta^2 \left\{ \left[\left[\int_{A_-}^{B_-} + \int_{B_+}^{A_+} \right] [P(E)E^2\text{sech}^2(\beta E)]dE \right] - \Delta^2\text{sech}^2(\beta\Delta) \right\} . \tag{3.4}$$

For very low temperatures, the major contribution to the specific heat arises from small values of E. We consider the following specific case, which—in our opinion—represents, in some cases, a realistic physical situation. We assume that the near-neighbor interaction is large compared to Δ and compared to $k_B T$. Then for $J_1 \gg \Delta$, the lower limit of the integral in Eq. (3.4) becomes $A_- \simeq \Delta^2/J_1$. In what follows, it is convenient to introduce the temperature

$$T_0 = \Delta^2/k_B J_1 . \tag{3.5}$$

We will arbitrarily classify temperatures less than T_0 as very low temperatures, and temperatures greater than T_0, but small compared to Δ, as low temperatures. C_2 in Eq. (3.4) becomes

$$C_2 \approx \frac{2\pi Nz_0 |a|^{3/n} k_B}{V_0 - V_1} \left[\frac{2}{\Delta} \left[\frac{k_B T}{\Delta} \right]^{(6-n)/n} \int_{T_0/T}^{\alpha\beta\Delta} y^{6/n} \left[1 + \frac{3y^2}{(\beta\Delta)^2} + O(\beta^{-4}) \right] \text{sech}^2(y) \, dy \right] , \tag{3.6}$$

where α is of the order or unity. We note that the leading terms of the low-temperature specific heat are proportional to $T^{(6-n)/n}(1 + A_2 T^2)$, where A_2 is a positive constant. In particular, for $n = 3$ we have

$$C_p = \frac{Nz_0 |a| k_B^2 \pi^3 T}{18\Delta^2(V_0 - V_1)} \left[1 + \frac{21\pi^2}{20} \left[\frac{k_B T}{\Delta} \right]^2 + O(T^4) \right] . \tag{3.7}$$

We remark that, for the case when[16] $n < 3$, the low-T specific heat will be proportional to $T^{1+(6-2n)/n}$, and will increase with a power of T greater than 1. For $n = 3$, our results give that $C_p \propto T$ for low T, and also show that there is a large T^3 dependence in C_p arising from the interacting tunneling states above and beyond the usual Debye contribution.

For $T \ll T_0$ the leading term in C_p is no longer linear in T for $n = 3$, but rather becomes proportional to

$\exp(-\text{const}/T)$. The total specific is obtained by adding Eqs. (3.2) and (3.4).

B. Dielectric susceptibility

To obtain the dielectric susceptibility $\mathcal{X}$ we let

$$\mathcal{X} = \mathcal{X}_1 + \overline{\mathcal{X}}_2 , \tag{3.8}$$

where $\mathcal{X}_1$ and $\overline{\mathcal{X}}_2$ are the electric susceptibilities arising from $F^{(1)}$ and $F^{(2)}$, respectively. The expression for $\mathcal{X}$ is

$$\mathcal{X} = \beta \lim_{E_0 \to 0} \frac{\partial^2 \ln(-\beta F)}{\partial(\beta E_0)^2} ,$$

where E_0 is the externally applied electric field. The relationship between the dielectric constant ϵ (ϵ here is not to be confused with the asymmetry energy of Sec. II) and $\mathcal{X}$ is $\epsilon = 1 + 4\pi\mathcal{X}$.

The expression for the free energy as a function of applied field E_0 is obtained for the two-level tunneling units by replacing ϵ in Eqs. (2.2) and (2.3) by pE_0, where p is the dipole moment of the tunneling unit. From Eq. (2.7) we obtain

$$\mathcal{X}_1 = (Np^2/\Delta)\tanh(\beta\Delta) . \tag{3.9}$$

In order to obtain $\overline{\mathcal{X}}_2$ we have to find the corrections, to order E_0^2, to the eigenvalues given by Eq. (2.10). Letting $J_{ij} = J$, we then have, to order E_0^2,

$$\lambda_1 = -(J^2 + 4\Delta^2)^{1/2} - \frac{(pE_0)^2}{2\Delta^2} \frac{[(J^2 + 4\Delta^2)^{1/2} - J]^2}{(J^2 + 4\Delta^2)^{1/2}} , \tag{3.10a}$$

$$\lambda_2 = -J , \tag{3.10b}$$

$$\lambda_3 = J\left[1 - \frac{2(pE_0)^2}{\Delta^2}\right] , \tag{3.10c}$$

$$\lambda_4 = (J^2 + 4\Delta^2)^{1/2} + \frac{(pE_0)^2}{2\Delta^2} \frac{[(J^2 + 4\Delta^2)^{1/2} + J]^2}{(J^2 + 4\Delta^2)^{1/2}} . \tag{3.10d}$$

The dielectric susceptibility $\overline{\mathcal{X}}_2(J)$ for a *single* pair of two-level tunneling units is, for a fixed $J_{ij} = J$,

$$\overline{\mathcal{X}}_2(J) = \frac{2p^2}{D}\left[-\frac{2J}{\Delta^2}e^{\beta J} - \frac{[(J^2 + 4\Delta^2)^{1/2} - J]^2}{2\Delta^2(J^2 + 4\Delta^2)^{1/2}}\exp[-\beta(J^2 + 4\Delta^2)^{1/2}] \right.$$
$$\left. + \frac{[(J^2 + 4\Delta^2)^{1/2} + J]^2}{2\Delta^2(J^2 + 4\Delta^2)^{1/2}}\exp[\beta(J^2 + 4\Delta^2)^{1/2}] \right] - \frac{2p^2}{\Delta}\tanh(\beta\Delta) , \tag{3.11}$$

where

$$D = 2\cosh(\beta E_+)\cosh(\beta E_-) , \tag{3.12}$$

and $E_\pm$ is given by Eq. (2.13). We let

$$\overline{\mathcal{X}}_2 = \langle \overline{\mathcal{X}}_2(J) + \overline{\mathcal{X}}_2(-J) \rangle_c . \tag{3.13}$$

Summing over all pairs gives, after some algebra,

$$\overline{\mathcal{X}}_2 = \frac{Nz_0p^2}{2}\left[\left\langle \frac{E_-^2\tanh(\beta E_+) + E_+^2\tanh(\beta E_-)}{\Delta^2(E_+ + E_-)}\right\rangle_c - \frac{2}{\Delta}\tanh(\beta\Delta)\right]$$

$$\equiv \mathcal{X}_2 - Nz_0p^2\tanh(\beta\Delta)/\Delta , \tag{3.14}$$

where the angular brackets, $\langle\ \rangle_c$, denote an average over E_+ and E_- with the respective probability distributions given by Eq. (2.18), and $\mathcal{X}_2$ is the part of the susceptibility arising from the interacting term only.

Using the relation $E_\pm = \Delta^2/E_\mp$ and the fact that $E_- < \Delta$ and $E_+ > \Delta$, we obtain $\mathcal{X}_2$ for a r^{-3} potential in the form of a single integral,

$$\mathcal{X}_2 = Nz_0p^2\left[\left\langle \frac{2\Delta^2\tanh(\beta E)}{E(E^2 + E^2)}\right\rangle_c - \frac{\tanh(\beta\Delta)}{\Delta}\right]$$

$$= Nz_0p^2\left\{\frac{4\pi a\Delta^2}{3(V_0 - V_1)}\left[\left(\int_{A_-}^{B_-} + \int_{B_+}^{A_+}\right)\left[\frac{\tanh(\beta E)}{E(E^2 - \Delta^2)^2}\right]dE\right] - \frac{\tanh(\beta\Delta)}{\Delta}\right\} .$$

We examine Eq. (3.15) in the following limits. (i) $J \to 0$; for this case $P(E) \approx \delta(E - \Delta)$ and $\overline{\mathcal{X}}_2$ vanishes identically as it should. (ii) We let the fractional concentration c of the tunneling states approach zero. Since $V_0 = \frac{4}{3}\pi r_0^3$ is the effective volume of the pair, we assume that $r_0^3 = r_1^3/c$, then $r_1^3 \ll r_0^3$, and it is sufficient to integrate Eq. (3.14) over a small region of energy near $E \approx \Delta$. Again we obtain that $\mathcal{X}_2 \to 0$.

We let $Q = 4\pi a\Delta^2/[3(V_0 - V_1)]$. Integrating Eq. (3.15) in parts, and letting

$$g(E) = [Q\tanh(\beta E)/(2\Delta^2)][\ln|E^2/(E^2 - \Delta^2)| - \Delta^2/(E^2 - \Delta^2)] ,$$

we obtain

$$\chi_2 = Nz_0 p^2 \left\{ [g(B_-) + g(A_+) - g(A_-) - g(B_+)] \right.$$

$$\left. - \left[Q \left(\int_{\beta A_-}^{\beta B_-} + \int_{\beta B_+}^{\beta A_+} \right) \frac{\text{sech}^2 x}{2\Delta^2} \left[\ln \left| \frac{x^2}{x^2 - (\beta\Delta)^2} \right| + \frac{(\beta\Delta)^2}{(\beta\Delta)^2 - x^2} \right] dx \right] - \frac{\tanh(\beta\Delta)}{\Delta} \right\} . \tag{3.16}$$

We evaluate Eq. (3.16) for low temperatures under the following conditions. We let

$$\beta A_- \approx \beta\Delta^2 / J_1 = (T_0 / T) < 1 ,$$

and $\beta\Delta \gg 1$ and $\beta B_+ \gg 1$. Under these conditions, the T-dependent part of the low-T susceptibility is

$$\chi_2 \approx -Nz_0 p^2 \frac{4\pi |a|}{3\Delta} \frac{1}{V_0 - V_1} \ln \left[\frac{T}{\Delta} \right] . \tag{3.17a}$$

Thus our calculation predicts a $-\ln T$ dependence in the low-temperature susceptibility, provided $T > T_0$. For $T \ll T_0$, i.e., very low temperatures, the temperature-dependent part of the dielectric susceptibility becomes proportional to

$$1 / \cosh^2(\beta E_-) \approx 1 / \cosh^2(\Delta^2 / k_B J_1) .$$

The major contribution to the dielectric constant χ_2 arises in this case from the first part of Eq. (3.16) added to Eq. (3.9) to give for, $\beta\Delta \gg 1$ and $(T/T_0) \gg 1$,

$$\chi \simeq \frac{Np^2}{\Delta} \left[1 + z_0 + \frac{4\pi |a| z_0}{3(V_0 - V_1)\Delta} \ln \left[\frac{J_1}{\Delta} \right] \right] . \tag{3.17b}$$

Thus our calculations predict that the dielectric susceptibility χ_2 increases as $-\ln T$ with decreasing, and low, temperatures, and then flattens out as the temperature is further lowered. A reasonable value for the near-neighbor interaction J_1 arising from strain interactions is $250 \text{ K} < J_1 < 2000 \text{ K}.$[12] Thus, for a value of Δ of the order of 1 K, χ_2 will be given by Eq. (3.17) for T greater than a few millikelvins, provided that $k_B T \ll \Delta$.

C. Low-temperature thermal conductivity κ

The contribution to the thermal conductivity is from relaxation arising from resonant scattering by the tunneling units as well as from the usual relaxation processes. The contribution from the relaxation processes is of higher order in T than the resonant scattering contribution, and therefore at low temperatures it is a good approximation to consider the latter only. The expression for the thermal conductivity is[3,17]

$$\kappa(T) = \frac{1}{3} \sum_{i=1}^{3} \int C_i(\omega) v_i(\omega) l_i(\omega) d\omega , \tag{3.18}$$

where the summation over i is over the three phonon modes, $C(\omega)$ is the frequency-dependent specific heat, $v(\omega)$ is the sound velocity, and $l(\omega)$ is the mean free path. We approximate $v(\omega)$ by a constant velocity $\bar{v}$, and

$$l(\omega) = v(\omega) / \tau(\omega) \equiv \bar{v} / \tau(\omega) .$$

We write

$$C(\omega) = A\omega^4 e^{\beta\hbar\omega} / (e^{\beta\hbar\omega} - 1)^2 ,$$

with

$$A = 3\hbar^2 k_B V / (2\pi^2 k_B T^2 v^3) .$$

We have[17]

$$\tau_{\text{res}}^{-1} = \frac{2\pi}{h} \int P(E)[1 - 2n(E)] \left[\left[\frac{hk}{2\rho v_i} \right]^{1/2} M_{ll'} \right]^2$$

$$\times \delta(E - hvk) dk , \tag{3.19}$$

where $M_{ll'}$ is the matrix element of transition between states l and l', and $n(E) = (e^{\beta E} + 1)^{-1}$. In Appendix C we consider the matrix element M_{12} between the ground state and the first excited state, the most important term at low temperatures. We obtain

$$M_{12} = \frac{-2\Delta\gamma}{\{2(J^2 + 4\Delta^2)^{1/2}[(J^2 + 4\Delta^2)^{1/2} - J]\}^{1/2}} , \tag{3.20}$$

where $\gamma = \frac{1}{2}(\partial\epsilon / \partial\epsilon_p)$, where ϵ_p is the local strain. In terms of the energy E, M_{12} can be rewritten

$$M_{12} = \frac{-\Delta\gamma}{(E^2 + \Delta^2)^{1/2}} , \tag{3.21}$$

$$\tau_{\text{res}}^{-1} = \sum_i \frac{\pi}{h\rho v_i^2} \left[\frac{\Delta^2 \gamma^2}{E^2 + \Delta^2} \right] P(E)E \tanh(\beta E) . \tag{3.22}$$

For low temperatures only the low energies contribute appreciably. Thus $\Delta^2 / (E^2 + \Delta^2) \rightarrow 1$, and

$$M_{12} \approx -\gamma . \tag{3.23}$$

The low-temperature thermal conductivity is now calculated by using Eq. (3.23) in Eq. (3.18) and considering the resonant scattering contribution only. Thus we have

$$\kappa \approx \int_{A_-}^{B_-} \frac{C(\omega)d\omega}{P(\omega)\omega \tanh(\hbar\omega)} . \tag{3.24}$$

For low temperatures, only small values of ω contribute. Equation (2.19) shows that $P(\omega)$ is proportional $\omega^{(6-2n)/n}$ for small ω. Substituting this result into Eq. (3.24) gives

$$\kappa(T) \propto T^{(4n-6)/n} . \tag{3.25}$$

It is interesting to note, using Eq. (3.6), that the product

$$\kappa C_p \propto T^3 \tag{3.26}$$

independently of the value of n for low enough T. We also note from Eq. (3.25) that for $n = 3$, $\kappa(T) \propto T^2$. This result is not unlike those observed in glassy materials. For somewhat higher ω,

$$P^{-1}(\omega) \propto [1-3(\hbar\omega/\Delta)^2] \ ,$$

and the thermal conductivity will be proportional to

$$\kappa = T^{(4n-6)/n}[1-A_3(n)T^2] \ , \tag{3.27}$$

where $A_3(n)$ is a positive constant for fixed n. Thus the predicted thermal conductivity will increase slower than $T^{(4n-6)/n}$ for higher temperatures. For even higher temperatures the resonant scattering contribution is proportional to T^5; however, our results are not expected to be valid since we are not considering the relaxation contribution to the scattering. Our calculations show that there is a shoulder (flat portion) in the thermal conductivity at $k_B T/\Delta \approx 0.2$. This shoulder arises from the increased density of states with increasing energy.

IV. CONCLUSIONS

We study the effect of interactions on a set of low concentrated two-level and four-level tunneling states under certain strongly restrictive conditions. We assumed that the tunneling states have no long-range order and are sufficiently dilute that the first two virial coefficients give the major contribution to the free energy. The interaction potential $J(r)$ is assumed to have a form $J(r) \propto \pm a/r^n$, with n as a parameter. We find that the interaction between the tunneling units strongly modifies the spectrum of the elementary excitations of the system at low energies, and gives an n-dependent density of states $P(E)$ of the elementary excitation energies E. In particular, for $n=3$, the density of states is approximately a constant for low E, and our results give a specific heat linear in T and a thermal conductivity proportional to T^2. Although these results are not unlike those obtained in glasses, we have specifically refrained from making any detailed comparisons with experiments. The primary reason for this is that the purpose of this paper is to demonstrate the effect of the interactions and how they modifiy the density of states, even though it is not clear what role such interactions play in realistic glassy systems. A further difficulty of comparing our results with experiments is that the effective number of neighbors z_0 and the effective volume V_0 of the pair of enter as parameters into our problem, and it is by no means clear what these parameters should be in a realistic model of impurities distributed in alkali halides.[10,11] The calculations are performed using a single value of the tunneling matrix element Δ and it is assumed that Δ does not change with the interaction potential J. These assumptions may also not be realistic for glassy systems.[15,16] This is another reason for refraining from making detailed comparisons with experiment.

APPENDIX A

In this appendix we prove that if we start with a potential

$$J(r) = a(\theta,\phi)r^{-n} \ , \tag{A1}$$

the density of states will be qualitatively the same as that for a potential $J(r) = \pm a/r^3$. It is assumed that

$$\int a(\theta,\phi)\sin\theta \, d\theta \, d\phi = 0 \ ,$$

for otherwise the system may undergo a phase transition, contrary to our assumption.

With Eq. (A1) we have a set of random potentials for random r, θ, and ϕ, and to obtain the configurational average $\langle \ \rangle_c$ we have to integrate over all three of these variables. First, we perform the integration over r, and at the end of our calculation, we perform the integration over θ and ϕ.

The only change in Eq. (2.16) is replacing $|a|$ by $|a(\theta,\phi)|$. Similarly, in Eqs. (2.19) and (2.20) we replace $|a|$ by $|a(\theta,\phi)|$. Finally, the average of the thermodynamic quantity $X(E)$ is

$$\langle X(E) \rangle = \int X(E)P(E,\theta,\phi)\sin\theta \, d\theta \, d\phi \tag{A2}$$

$$= \langle X(E) \rangle_0 \int |a(\theta,\phi)/a|^{3/n}\sin\theta \, d\theta \, d\phi \ , \tag{A3}$$

where $\langle X(E) \rangle_0$ is the average value of $X(E)$ calculated with a potential $J(r) = \pm a/r^n$. The result of Eq. (A3) is that a is replaced by some constant a'.

APPENDIX B

In this appendix we obtain the expression for the 16×16 matrix $\underline{M}$ representing the Hamiltonian given by Eq. (2.23). To obtain the matrix we start with a representation involving the pair states formed from the product of $|\pm x_i\rangle$ and $|\pm y_i\rangle$ where $i=1$ and 2. The matrix is

$$\underline{M} = \begin{bmatrix} \underline{A} & \underline{0}_{4\times4} & \underline{E} & \underline{E} \\ \underline{0}_{4\times4} & \underline{B} & \underline{E} & \underline{E} \\ \underline{E} & \underline{E} & \underline{C} & \underline{0}_{4\times4} \\ \underline{E} & \underline{E} & \underline{0}_{4\times4} & \underline{D} \end{bmatrix} \ , \tag{B1}$$

where $\underline{0}_{n\times n}$ is the n-dimensional null matrix, and

$$\underline{A} = \begin{bmatrix} \underline{a} & \underline{b} \\ \underline{b} & \underline{0}_{2\times2} \end{bmatrix}, \quad \underline{B} = \begin{bmatrix} -\underline{a} & \underline{b} \\ \underline{b} & \underline{0}_{2\times2} \end{bmatrix}, \quad \underline{C} = \begin{bmatrix} \underline{0}_{2\times2} & \underline{b} \\ \underline{b} & -\underline{a} \end{bmatrix},$$

$$\underline{D} = \begin{bmatrix} \underline{0}_{2\times2} & \underline{b} \\ \underline{b} & \underline{a} \end{bmatrix}, \quad \underline{E} = \begin{bmatrix} \underline{c} & \underline{0}_{2\times2} \\ \underline{0}_{2\times2} & \underline{c} \end{bmatrix}, \tag{B2}$$

where

$$\underline{a} = \begin{bmatrix} -J & 0 \\ 0 & J \end{bmatrix}, \quad \underline{b} = \begin{bmatrix} -\Delta & -\Delta \\ -\Delta & -\Delta \end{bmatrix}, \quad \underline{c} = \begin{bmatrix} -\Delta & 0 \\ 0 & -\Delta \end{bmatrix} \ . \tag{B3}$$

Obtaining the eigenvalues of $\underline{M}$ is nontrivial. They are

$$\lambda_{1,2} = \pm(J^2 + 16\Delta^2)^{1/2} \ ,$$

$$\lambda_{3,4} = \pm J \ ,$$

$$\lambda_5 = \lambda_6 = \lambda_7 = \lambda_8 = 0 \ , \tag{B4}$$

$$\lambda_{9,10} = \lambda_{11,12} = \tfrac{1}{2}[J \pm (J^2 + 16\Delta^2)^{1/2}] \ ,$$

$$\lambda_{13,14} = \lambda_{15,16} = -\tfrac{1}{2}[J \pm (J^2 + 16\Delta^2)^{1/2}] \ ,$$

where λ_j is the jth eigenvalue $(1 \leq j \leq 16)$.

The partition function Z_4 for the pair of interacting four-level tunneling states is obtained using (B4).

$$Z_4 = 4 + 2\cosh(\beta J) + 2\cosh[\beta(J^2 + 16\Delta^2)^{1/2}]$$

$$+ 8\cosh(\beta J/2)\cosh[(\beta/2)(J^2 + 16\Delta^2)^{1/2}]$$

$$= [2(\cosh(\beta J/2) + \cosh\{\beta[(J/2)^2 + 4\Delta^2]^{1/2}\})]^2 . \quad (B5)$$

Comparing Z_4 with the partition function of the interacting pair of two-level tunneling states, Z_2, we obtain

$$Z_4(J) = [Z_2(J/2)]^2 . \quad (B6)$$

The partition function Z_4 may be written

$$Z_4 = [4\cosh(\beta U_+)\cosh(\beta U_-)]^2 , \quad (B7)$$

where

$$U_\pm = \tfrac{1}{2}\{[(J/2)^2 + 4\Delta^2]^{1/2} \pm J/2\} . \quad (B8)$$

Next we evaluate the eigenvalues of the 4×4 matrix $\underline{H}_i$ for the four-level tunneling unit as follows:

$$\mathrm{Det}(\underline{H}_i - \lambda\underline{1}_{4\times4} = \mathrm{Det}\begin{bmatrix} \underline{\Delta} & \underline{b} \\ \underline{b} & \underline{\Delta} \end{bmatrix} , \quad (B9)$$

where

$$\underline{\Delta} = \begin{bmatrix} \lambda\underline{1}_{2\times2} & \underline{0}_{2\times2} \\ \underline{0}_{2\times2} & \lambda\underline{1}_{2\times2} \end{bmatrix} , \quad (B10)$$

and $\underline{b}$ is given by Eq. (B3)

The eigenvalues of Eq. (B9) are

$$\lambda_{1,2} = 0,0, \quad \lambda_{3,4} = \pm2\Delta . \quad (B11)$$

Thus the partition function Z_i for the single noninteracting four-level tunneling state i is

$$Z_i = 2 + 2\cosh(2\beta\Delta_i) = 4[\cosh(\beta\Delta_i)]^2 , \quad (B12)$$

and for $\Delta_i = \Delta_j = \Delta$, we have

$$\ln \mathrm{tr}\, \exp[\beta(H_i + H_j)] = 2\ln[4\cosh(\beta\Delta)] . \quad (B13)$$

Equation (B13) is used in Eq. (2.28) to obtain the second virial coefficient for the interacting tunneling states.

APPENDIX C

In this appendix we calculate the matrix element of transition between the energy states shown in Fig. 1 and represented by the eigenvalues given by Eq. (2.10). We express the Hamiltonian as

$$H = H_0 + H' \quad (C1)$$

where H_0 is the Hamiltonian of the pair given by Eq. (2.3) for the two-level states. H' is the perturbation arising from the strain fields ϵ_p introduced by the phonons.[4,17] Thus,

$$H' = \frac{1}{2}\frac{\partial H_0}{\partial \epsilon_p} . \quad (C2)$$

We let

$$C' = \frac{1}{2}\frac{\partial J_{ij}}{\partial \epsilon_p}, \quad \gamma = \frac{1}{2}\frac{\partial \epsilon}{\partial \epsilon_p}, \quad \delta = \frac{1}{2}\frac{\partial \Delta}{\partial \epsilon_p} . \quad (C3)$$

For the case when $\epsilon_1 = \epsilon_2$, we have

$$\underline{H}' = \frac{1}{2}\begin{bmatrix} -C'-2\gamma & -\delta & -\delta & 0 \\ -\delta & C' & 0 & -\delta \\ -\delta & 0 & C' & -\delta \\ 0 & -\delta & -\delta & -C'+2\gamma \end{bmatrix} . \quad (C4)$$

Next we let $\underline{T}$ be the matrix which diagonalizes $\underline{H}_0$, such that $\underline{T}\underline{H}_0\underline{T}^{-1} = \underline{\Delta}_1$, where $\underline{\Delta}_1$ is a diagonal matrix whose eigenvalues are given by Eq. (2.10). Let $M_{\alpha\beta}$ be the matrix element of transition between the states α and β, where α and β go from 1 to 4. Let Ψ_α be the αth eigenfunction of H_0 associated with the αth eigenvalue. Then $M_{\alpha\beta} = (\underline{T}\underline{H}'\underline{T}^{-1})_{\alpha\beta}$, or

$$M_{\alpha\beta} = \langle \Psi_\alpha | H' | \Psi_\beta \rangle . \quad (C5)$$

The eigenvectors Ψ_α of H_0, in the order of the eigenvalues listed in Eq. (2.10), are

$$\Psi_1 = [\lambda(\lambda - J)]^{-1}(\Delta, \tfrac{1}{2}(\lambda - J), \tfrac{1}{2}(\lambda - J), \Delta) ,$$

$$\Psi_2 = (1/\sqrt{2})(1, 0, 0, -1) ,$$

$$\Psi_3 = (1/\sqrt{2})(0, 1, -1, 0) , \quad (C6)$$

$$\Psi_4 = [\lambda(\lambda + J)]^{-1/2}(\Delta, -\tfrac{1}{2}(\lambda + J), \tfrac{1}{2}(\lambda + J), \Delta) ,$$

where $\lambda = (J^2 + 4\Delta^2)^{1/2}$. The matrix element M_{12} connecting the ground state with the first excited state is

$$M_{12} = -\frac{2\gamma\Delta}{\{[2(J^2 + 4\Delta^2)^{1/2}][(J^2 + 4\Delta^2)^{1/2} - J]\}^{1/2}} . \quad (C7)$$

This matrix element is used in Eq. (3.19) to calculate the thermal conductivity.

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