

Thermodynamics of an extended Fredrickson-Andersen model

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(Received 27 October 1998)

We describe an extension of the Fredrickson-Andersen model (or n -spin facilitated kinetic Ising model) which exhibits a transition similar to a glass transition. Our extension incorporates long-range effects. Exact or numerically accurate results are obtained only for one dimension, but most conclusions apply more generally. The model exhibits a discontinuous drop in the specific heat as it is cooled through the transition temperature. This leads to an excess internal energy and a residual entropy at zero temperature. The disorder associated with the residual entropy can be seen in diffuse scattering which is characteristic of disordered systems.
[S1063-651X(99)07303-1]

PACS number(s): 05.50.+q, 82.20.Mj, 64.70.Pf

I. INTRODUCTION

Despite advances in the description of glasses, supercooled liquids, and the glass transition by means of a variety of approaches, there is a lack of complete understanding of the phenomenon [1–7]. Strictly speaking, glasses are not in thermal equilibrium. The glass transition is not a standard phase transition and is not connected with the development of long-range order. The dynamics of supercooled liquids shows a slowing down due to cooperative motions. Within a narrow temperature range around the transition temperature T_g , the dynamics become so slow that thermal equilibrium cannot be observed by an experiment. Despite this basic problem, measurements of a glass's thermal properties (like the specific heat) on the slowest possible time scales yields quasithermodynamic properties of that glass.

Insight into some aspects of the glass transition is obtained from the spin-facilitated kinetic Ising model, introduced by Fredrickson and Andersen [8–11]. In order to reduce the degrees of freedom they carried out a coarse graining on both time and length scales. Therefore the system is separated into a virtual lattice of mesoscopic cells, each containing a sufficiently large number of microscopic particles. This model has been the subject of several numerical [12–17] and analytical studies [18–20]. The model shows a dramatic slowing of the dynamics at low temperatures. This slowing occurs because nearest-neighbor interactions between lattice sites can inhibit the change of state at a lattice site and therefore interactions can restrict the phase space of all possible configurations. Here, we suggest an extension of the usual Fredrickson-Andersen model whose thermodynamic properties may approximate the thermodynamic properties of glasses. Our extension of the Fredrickson-Andersen model postulates that both nearest-neighbor and long-ranged effects are needed to inhibit the change of state at a lattice site. To simplify the calculation, the long-ranged effects are taken to be of infinite range, so they can be described using mean-field theory. The physics

of our model is illustrated for a one-dimensional lattice, where many exact results can be obtained. Many qualitative aspects of our one-dimensional results remain valid for three dimensions.

Section II summarizes the Fredrickson-Andersen model. Section III describes a mean-field solution of the Fredrickson-Andersen model. Section IV presents our modified model and its solution in one dimension. Section V generalizes the conclusions.

II. FREDRICKSON-ANDERSEN MODEL

The Fredrickson-Andersen model associates each mesoscopic cell of a glass with an Ising spin on a lattice. If the local density is higher (lower) than a reference density, the associated spin is down (up). The spin-up sites are fluidlike and the spin-down sites are solidlike. The thermal equilibrium probabilities for a site being spin up, p , or spin down, q , are

$$p = 1 - q = \frac{\exp(-\beta h)}{2 \cosh(\beta h)}, \quad (1)$$

where β is the inverse temperature and h and $-h$ are the effective energies of the liquidlike and the solidlike cells.

Thermal equilibrium of this kinetic model is approached through individual spin flips. A spin-up site flips to a spin-down site at a rate r_- . The spin-flip rate for the reverse process is r_+ . The temperature dependent spin-flip rates satisfy the condition of detailed balance, so

$$qr_+ = pr_- . \quad (2)$$

Thermal equilibrium will be achieved as long as the spin-flip rates (r_{\pm}) do not vanish. However, the glasslike behavior and thermodynamic ambiguity of the Fredrickson-Andersen model is achieved by allowing some spin-flip rates to vanish. This slows (and perhaps stops) the approach to thermal equilibrium.

The core of the Fredrickson-Andersen model is the criterion which determines the nonzero spin-flip rates. Let ν_i be the number of spin-up sites which are nearest neighbors to

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site i . The Fredrickson-Andersen model postulates that the spin at site i can change orientation ($r_{\pm} \neq 0$) only if

$$\nu_i \geq n_{\text{FA}}, \quad (3)$$

where n_{FA} with $0 \leq n_{\text{FA}} \leq z$ (z is the coordination number) is an integer parameter of the model. Sites which are allowed to change their spin direction are ‘‘mobile’’ while spins which cannot flip are ‘‘immobile.’’ Since a spin flip at site j changes ν_i at all sites i neighboring site j , mobile spins can become immobile and immobile spins can become mobile.

The true thermodynamic properties of the Fredrickson-Andersen model do not describe a glass transition. We illustrate this thermodynamic limitation for a square lattice where z is 4. For this case, if n_{FA} is 3 or 4, rectangular spin-down regions of the lattice are permanent and cannot change at any temperature. This is not reasonable since initial conditions determine the possible configurations for all future times. (‘‘Configuration’’ denotes the orientation of a set of spins.) However, if n_{FA} is 0, 1, or 2, the thermodynamic properties are those of a set of free spins. To prove this result, one can apply (with notational modifications) the theorem by van Enter [21] which describes accessible states in a related class of models called ‘‘bootstrap percolation’’ [22] (or ‘‘diffusion percolation’’ [23]). The van Enter theorem applied to the Fredrickson-Andersen square lattice for $n_{\text{FA}}=2$ (the only interesting case) shows that sequences of spin flips at mobile sites connect essentially all possible spin configurations. Furthermore, we have generalized this theorem to show that the energy fluctuation needed to flip essentially any spin is finite. [Formally, given any $\epsilon > 0$ and a random distribution of spins with spin-up probability $p > 0$, an arbitrary spin can be flipped with a probability greater than $1 - \epsilon$ by a finite energy fluctuation $E(\epsilon, p)$.] Thus the partition function must include essentially all spin configurations, and the thermodynamics are unaffected by the spin-flip restriction when $n_{\text{FA}}=2$. Of course, when p is small, the energy fluctuation needed to flip spins can be so large that relaxation times would be too long to be observed. In this sense the Fredrickson-Anderson model may afford a realistic description of glasses. We are only noting that the formally derived exact thermodynamic properties of this model will not describe the quasithermodynamics of glasses. Generalization of these comments applies to the Fredrickson-Andersen model on other lattices and for three dimensions.

III. MEAN-FIELD THEORY

We apply the simplest possible version of a mean-field theory to the Fredrickson-Andersen model by replacing the number of spin-up neighbors of site i [ν_i of Eq. (3)] by its average over all the sites, called $\bar{\nu}$. In thermal equilibrium

$$\bar{\nu} = zp, \quad (4)$$

with p given by Eq. (1). Since every site is characterized by the same $\bar{\nu}$, a phase transition takes place at a critical spin-up probability

$$p_c^{\text{MF}} = n_{\text{FA}}/z. \quad (5)$$

Solving Eq. (1) for T gives

$$\frac{1}{T} = \beta = \frac{1}{2h} \ln \left(\frac{1-p}{p} \right) \quad (6)$$

so if $p_c^{\text{MF}} < 1/2$ and $h > 0$, the corresponding mean field transition temperature T_c^{MF} is positive and finite. Above the transition temperature, the system has the properties of a set of free spins, since every spin is mobile. However, cooling below T_c^{MF} cannot lower the energy or the entropy because all spin-flip rates vanish for $p < p_c^{\text{MF}}$.

This mean-field model restricts the spin configurations to those in which the fraction of spin-up sites is at least p_c^{MF} . The thermodynamic properties of this model with a restricted configuration space are unusual. The ground state is highly degenerate. This means the ‘‘third law’’ of thermodynamics is violated, and there is a residual nonzero entropy even at zero temperature. Also, since the system’s internal energy approaches the lowest allowed energy at T_c^{MF} , the heat capacity vanishes at temperatures below T_c^{MF} .

Mean-field theory becomes accurate in the limit of long-ranged interactions. Thus one might expect a fairly sharp glass transition for a long-ranged generalization of the Fredrickson-Andersen model. Long-ranged influences can play an important role in real glasses because topological requirements associated with partial ordering are effectively long ranged.

Mean-field theory also becomes accurate in the limit of large dimensionality. In the infinite-dimensional limit, where $z \rightarrow \infty$, the mean-field results become exact.

IV. EXTENDED FREDRICKSON-ANDERSEN MODEL

Even when long-ranged effects are significant, the simple mean-field theory described above would be an inadequate approximation. It does not properly describe effects associated with the dominant short-ranged interactions. The extended Fredrickson-Andersen model described here assumes both long-ranged and short-ranged effects control the spin-flip rates. The long-ranged interaction is treated in the spirit of mean-field theory, but the short-ranged part is included on the mesoscopic scale.

The extended Fredrickson-Andersen model is characterized as follows.

(1) Each spin is either ‘‘immobile’’ or ‘‘mobile,’’ with no spin flips for the immobile spins and detailed balance determining the ratios of the spin-flip rates for the mobile spins.

(2) As with the original Fredrickson-Andersen model, a spin is mobile if it is embedded in a sufficiently large concentration of spin-up sites. However, these spin-up sites can be either nearest neighbors or distant neighbors. Thus a spin is mobile if *either* (A) The spin is adjacent to at least n_{FA} spin-up sites *or* (B) the number density of spin-up sites is greater than a fixed critical density p_c .

This extended Fredrickson-Andersen model has two parameters; the integer n_{FA} for nearest neighbors and long-ranged criterion p_c . Unlike the mean-field treatment of the Fredrickson-Andersen model, these parameters are independent and they are not related by Eq. (5).

The long-range part of this model [property 2(B)] means a glass transition is possible. At temperatures such that

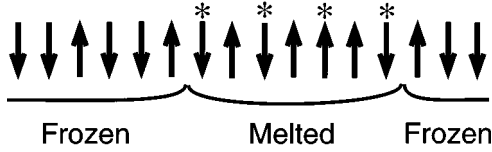


FIG. 1. A spin chain with frozen and melted parts. Mobile spins are highlighted with *.

$p > p_c$, there is no restriction of the dynamics. When $p < p_c$ some of the spins will be static because of the short-range condition [property 2(A)]. If n_{FA} is chosen to be sufficiently large, the restricted dynamics at low temperatures will restrict the number of possible configurations and will alter the thermodynamics.

One dimension

Simple results for the extended Fredrickson-Andersen model can be obtained on a one-dimensional lattice. If $n_{FA} = 1$ (or 0) the thermodynamics for this model are the same as for free spins, so we consider only the case $n_{FA} = 2$. For $T < T_c$ and $n_{FA} = 2$, the one-dimensional chain of spins is separated into “frozen” and “melted” segments, as is illustrated in Fig. 1. These segments are determined by the configuration of the system as it cools through the transition temperature. Spins in the frozen segments are permanently immobile (as long as $T < T_c$). The spins in the melted segments may be either mobile or immobile. However, allowed spin flips can “free up” an immobile spin, so no spin in a melted segment is permanently immobile. The mobile spins in the melted segment are labeled with an asterisk in Fig. 1. Spin configurations in each melted segment are restricted to a subset of all possible configurations. Within this subset, thermal equilibrium is obtained. Since frozen segments are permanent below T_c , temperature variations of the thermal properties are determined by the melted segments.

A melted segment is characterized by its length n . Let $n\pi_n$ be the probability that a random site will lie in a melted segment of length n . For a one-spin melted segment

$$\pi_1 = p_c^2 q_c^4. \quad (7)$$

In Eq. (7), p_c^2 represents the probability that the single melted spin is surrounded by two spin-up sites [as required by property 2(A) when $n_{FA} = 2$]. The q_c^4 represents the probability that both these neighboring spins are adjacent to a pair of down spins, which is required for them to be in frozen segments. Similarly, the probability of finding a melted segment of length 2 is

$$\pi_2 = (p_c^2 + 2p_c q_c) \pi_1 \quad (8)$$

because the two mobile spins can only be in the configurations “up-up,” “up-down,” or “down-up.”

To facilitate our calculation of the internal energy, we write

$$\pi_n = p_c^2 q_c^4 \psi_n(p_c, q_c) \quad (9)$$

and insist that $\psi_n(p, q)$ be treated formally as a function of two independent variables, p and q , even though physically $p + q = 1$. For $n = 1, 2$ we pick

$$\psi_1(p, q) = p + q, \quad (10)$$

$$\psi_2(p, q) = p^2 + 2pq. \quad (11)$$

For longer segments, the $\psi_n(p, q)$ are obtained from a recursion relation which is a generalization of the recursion relation for the Fibonacci numbers;

$$\psi_{n+1}(p, q) = p\psi_n(p, q) + pq\psi_{n-1}(p, q). \quad (12)$$

This generates the π_n because for a portion of a melted segment of length n , the $n+1$ lattice site will also be in that melted segment if it is spin up (probability p_c). However, if the $n+1$ site is spin down (probability q_c), it will be part of the melted segment if a spin-up site and then a spin-down site is added to the segment of length $n-1$.

To obtain $\psi_n(p, q)$ for any n , one can write the recursion relations for the $\psi_n(p, q)$ in matrix form

$$\begin{pmatrix} \psi_{n+1} \\ \psi_n \end{pmatrix} = \begin{pmatrix} p & pq \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \psi_n \\ \psi_{n-1} \end{pmatrix}. \quad (13)$$

Eigenvalues of the matrix are

$$\gamma_{\pm} = \frac{p}{2} \pm \left[\left(\frac{p}{2} \right)^2 + pq \right]^{1/2}. \quad (14)$$

The $\psi_n(p, q)$ must be a sum of powers of these eigenvalues,

$$\psi_n(p, q) = a\gamma_+^{n-1} + b\gamma_-^{n-1}. \quad (15)$$

The coefficients a and b are obtained using Eqs. (10) and (11) for $\psi_1(p, q)$ and $\psi_2(p, q)$, yielding

$$\psi_n(p, q) = (p+q) \left(\frac{\gamma_+^{n-1} + \gamma_-^{n-1}}{2} \right) + \left(\frac{p^2 + 3pq}{2} \right) \left(\frac{\gamma_+^{n-1} - \gamma_-^{n-1}}{\gamma_+ - \gamma_-} \right). \quad (16)$$

The functions $\psi_n(p, q)$ yield more than the π_n . All the statistics of the melted segments can be expressed in terms of these functions. Each term in the $\psi_n(p, q)$ polynomial corresponds to a different spin configuration. For example, $\psi_2(p, q) = p^2 + 2pq$ means the melted segment of length 2 has one configuration with two spins up (the p^2 term) and two configurations with one spin up and one spin down (the $2pq$ term).

Knowledge of the $\psi_n(p, q)$ allows us to calculate the thermodynamic properties of this one-dimensional system. The probability that a site is in a melted segment is obtained using Eq. (9)

$$P_{\text{melt}} = \sum_{n=1}^{\infty} n \pi_n = p_c^2 q_c^4 \sum_{n=1}^{\infty} n \psi_n(p_c, q_c). \quad (17)$$

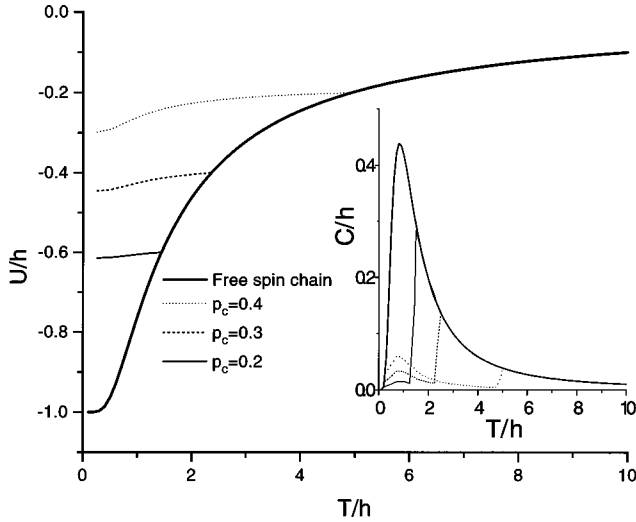


FIG. 2. The internal energy U and the heat capacity C for a free-spin chain and for $p_c = 0.2, 0.3, 0.4$.

The form for the $\psi_n(p, q)$ given in Eq. (16) means P_{melt} can be obtained simply by summing geometric series and simplifying using $p_c + q_c = 1$. The result,

$$P_{\text{melt}} = p_c^2(1 + 3p_c - 4p_c^2 + p_c^3), \quad (18)$$

is shown as a function of p_c as one of the curves in Fig. 3.

We obtain next the internal energy (per site) and the specific heat. These quantities give physical insight into the properties of this model and its possible relevance to real materials. At temperatures equal to and above the transition temperature, the internal energy and heat capacity describe free spins. Below the transition temperature, only the melted segments contribute to changes in the internal energy and the specific heat. Thus we need only consider the temperature dependence of the melted segment contribution to the internal energy, which is

$$U(T)_{\text{melt}} = \sum_{n=1}^{\infty} \pi_n u_n(T), \quad (19)$$

where the internal energy of a melted segment of length n is

$$u_n(T) = h \frac{1}{\psi_n(p, q)} \left(p \frac{\partial}{\partial p} - q \frac{\partial}{\partial q} \right) \psi_n(p, q) \quad (20)$$

because the differentiations “count” the number of spin-up and spin-down sites. For example, using Eq. (20) and $p + q = 1$ gives $u_1(T) = h(p - q)$ and $u_2(T) = h(2p^2)/(p^2 + 2pq)$ as one expects. The sums needed to obtain $U(T)_{\text{melt}}$ must be done numerically. They converge quickly, and Fig. 2 shows the temperature dependence of the internal energy for $p_c = 0$ (free spins) and for $p_c = 0.2, 0.3, 0.4$. The corresponding specific heats $C(T) = dU(T)/dT$ are shown in the inset. As one can see in Fig. 2, the suppression of spin flips dramatically reduces the specific heat below the glass transition.

Associated with the lower specific heat is an excess internal energy at $T=0$, represented by the difference between the internal energy of the model and the internal energy of

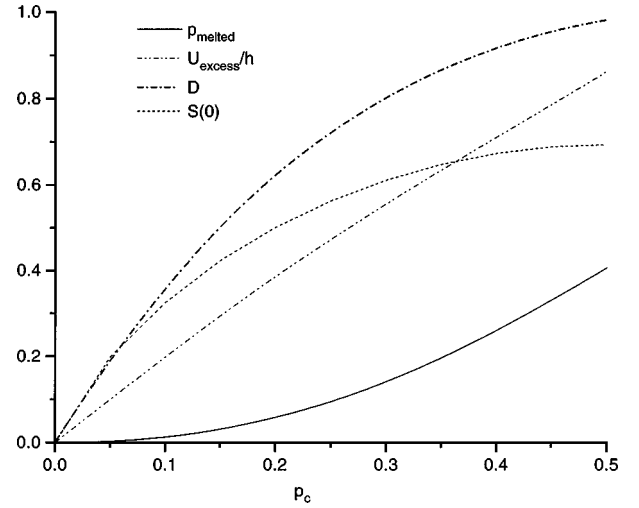


FIG. 3. The probability for a spin to be within a melted section P_{melt} , the excess internal energy U_{excess}/h , the scattering ratio D , and the residual entropy $S(0)$ as functions of p_c .

the free spins. Because the change in the free-spin internal energy from the transition temperature to zero temperature is $-2hp_c$,

$$U_{\text{excess}} = 2hp_c - [U(T_c)_{\text{melt}} - U(0)_{\text{melt}}].$$

The excess internal energy can be evaluated exactly. Using Eq. (19),

$$U(T_c)_{\text{melt}} = -hp_c^2(1 - 5p_c + 4p_c^2 + p_c^3). \quad (21)$$

For $T=0$, the configuration restrictions for melted segments means that $u_n(0) = -h$ for odd n and $u_n(0) = 0$ for even n . Thus

$$U(0)_{\text{melt}} = -h \sum_{n=\text{odd}>0}^{\infty} \pi_n. \quad (22)$$

Simplifying gives

$$U(0)_{\text{melt}} = -h \frac{p_c^2 q_c^2}{1 + p_c^2} (1 - p_c q_c^2). \quad (23)$$

The p_c dependence of U_{excess} obtained using the above four equations is shown in Fig. 3.

There is also a residual zero-temperature entropy, $S(0)$, which is associated with the disorder which is frozen into the system. It can be evaluated starting from

$$S(T) = S(T_c) - [S(T_c)_{\text{melt}} - S(T)_{\text{melt}}]. \quad (24)$$

To find $S(0)$, two of the terms in Eq. (24) can be found exactly

$$S(T_c) = -(p_c \ln p_c + q_c \ln q_c) \quad (25)$$

and because segments with an even number of sites have a doubly degenerate ground state,

$$\begin{aligned}
S(0)_{\text{melt}} &= \ln(2) \sum_{n=\text{even}>0}^{\infty} \pi(n) \\
&= \ln(2) \frac{p^3 q^2 (2 - 2p + 2p^2 - p^3)}{(1 + p^2)}. \quad (26)
\end{aligned}$$

The melted-segment contribution to the entropy at T_c can be obtained numerically in a variety of ways. We used

$$S(T)_{\text{melt}} = - \frac{dF}{dT}, \quad (27)$$

where the free energy function for the melted segments is

$$F = -T \sum_{n=1}^{\infty} \pi_n \ln Z_n \quad (28)$$

and the partition function for each chain segment is

$$Z_n = (e^{\beta h} + e^{-\beta h})^n \psi_n(p, q). \quad (29)$$

The residual entropy obtained from these expressions is shown as a function of p_c in Fig. 3. The entropy expression in Eq. (24) implies a continuous entropy function even at the transition temperature T_c . This means we have assumed there is no ‘‘collapse’’ of the system at the transition temperature. Such a collapse would occur if the positions of the frozen and melted segments were specified. The entropy is continuous because all allowed partitions of the spin chain into frozen and melted segments is included in the counting of configurations. By assuming the system is the sum of all possible configurations, this model is translationally invariant even below T_c .

Despite the formal translational invariance, the extended Fredrickson-Andersen model is disordered in the sense that it leads to diffuse elastic scattering. The diffuse scattering occurs because the model has a complicated spin-spin correlation function, $\langle \sigma_i \sigma_j \rangle$, where σ_i is the spin variable for site i , $\sigma = +1$ corresponds to spin up, $\sigma = -1$ corresponds to spin down, and $\langle \rangle$ denotes a thermodynamic average. The total scattering probability is the sum of a forward (or Bragg) scattering part and a diffuse scattering part. The total scattering probability is proportional to $\langle \overline{\sigma^2} \rangle$, where the ‘‘bar’’ indicates an average over all lattice sites. Assuming the scattering amplitude from a site is proportional to σ , the forward scattering probability is proportional to $\langle \overline{\sigma} \rangle^2$. Letting D denote the ratio of the diffuse scattering to the total scattering, and noting that $\sigma^2 = 1$,

$$D = 1 - \frac{\langle \overline{\sigma} \rangle^2}{\langle \sigma^2 \rangle} = 1 - \left(\frac{U(T)}{h} \right)^2. \quad (30)$$

At high temperatures, the diffuse scattering fraction D is caused by thermal fluctuations. If all the spins could align at low temperatures, $-U/h$ would be unity and the diffuse scattering fraction would vanish. Since the lowest value of the internal energy is $-h + U_{\text{excess}}$ the diffuse scattering does not vanish even when $T \rightarrow 0$. This zero-temperature diffuse scattering is not associated with thermal fluctuations. Instead, it is caused by the disorderlike correlations which are frozen into the lattice. The diffuse scattering fraction is shown as a function p_c in Fig. 3.

V. GENERALIZATION

Many of our results obtained in one dimension apply more generally. In particular, the modified Fredrickson-Anderson model yields a similar phase transition with T_c given by Eq. (6) on a square lattice (with n_{FA} is 3 or 4) or a cubic lattice (with $n_{\text{FA}} = 4, 5, \text{ or } 6$). For $T < T_c$ on the square or cubic lattices, there is a reduced specific heat. This leads to an excess internal energy at $T=0$ (compared to free spins). Associated with the reduced specific heat is a residual entropy at $T=0$ given by Eq. (24). There is also a diffuse scattering fraction given by Eq. (30). Unfortunately, numerical estimates for these quantities are not easy to obtain in two or three dimensions.

The phase transitions described here are all associated with mean-field theory or (equivalently) infinite-ranged effects or (equivalently) infinite dimensions. It is logical to ask if there are reasonable finite-ranged models which would lead to physically reasonable glasslike thermodynamic properties.

Note added in proof. We wish to thank Professor Michael Schultz for a critical reading of this manuscript. Dr. Schultz has urged us to remind readers that our calculations describe a simple solvable model—not a real glass. Although the model was constructed with glass and the glass transition in mind, unavoidable simplifications mean true glasses exhibit more complex and varied properties than are described by our model. Relaxation time scales have not been addressed in our solutions. The sharp transition temperature of our model bears only an approximate relation to experimentally observed glass transitions.

ACKNOWLEDGMENTS

The authors wish to thank R. G. Palmer for useful comments and discussions. This work is a part of a project supported by the Deutsche Forschungsgemeinschaft (Sonderforschungsbereich 418), by the German Academic Exchange Service (DAAD, Doktorandenstipendium HSP III), and by the NSF under Grant No. DMR 9628224.

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