Finite-Size Scaling Analysis of the Glass Transition

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We show that finite-size scaling techniques can be employed to study the glass transition. Our results follow from the postulate of a diverging dynamical correlation length at the glass transition whose physical manifestation is the presence of dynamical heterogeneities. We introduce a parameter $B(T, L)$ whose temperature, T, and system size, L, dependences permit a precise location of the glass transition. We discuss the finite-size scaling behavior of a diverging susceptibility $\chi(L, T)$. These new techniques are successfully used to study two lattice models. The analysis straightforwardly applies to any glass-forming system.

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Progress in the study of the "glass transition" has been slow for two main reasons [1]. First, almost by definition, the dynamics of a liquid supercooled through its melting transition towards its glass phase becomes so slow that it is impossible to actually cross a possible phase transition while staying at equilibrium. One relies therefore on thermodynamic or kinetic extrapolations which stop at the glass temperature T_g where the system no longer equilibrates. As a consequence, the mere existence of a genuine transition is still questioned, even though experiments now probe more than a decade of decades of relaxation times.

Second, the nature of the putative transition remains largely unknown as compared to more standard phase transitions. Basically, the situation is such that the formulation of a Ginzburg-Landau type of model is still impossible, because one does not know what would be the correct order parameter, its symmetries, dynamics, correlations, etc. Similarly, at the beginning of the 1990's, two papers apparently settled the related question of the existence of a diverging correlation length at the glass transition, reporting the absence of such a length scale [2,3]. These observations explain why modern developments of statistical mechanics (renormalization and related concepts) have seldom been used in the present context [1].

In this Letter, we show that finite-size scaling techniques first derived from renormalization group concepts in the context of continuous phase transitions are useful to the study of the glass transition, at variance with the common belief. This should allow much more powerful extrapolations to locate a possible transition and a better characterization of its nature, thus making progress on the two important points mentioned above.

The Letter is organized as follows. We first describe the observations underlying our work, before describing the principles of the analysis. As a first application of the method, we then consider two well-known models proposed in the context of the glass transition, namely, the one-dimensional Fredrickson-Andersen model [4] and the three-dimensional Kob-Andersen model [5].

Our work is based upon the recent burst of activity related to the definition, observation, and characterization of dynamical heterogeneities in the dynamics of supercooled liquids [6]. The underlying physics is that particles move in a cooperative manner, the more so the nearer the glass transition. This picture immediately suggests the existence of a dynamic correlation length associated to these dynamical domains, reflecting the length scale on which the particles dynamics have been correlated in a time interval t at temperature T. We shall define below a "coherence length" $\ell(T)$ by choosing the time interval to be equal to the mean relaxation time of the liquid $t_{\rm rel}(T)$. Whether or not $\ell(T)$ is connected to some yet undiscovered structural length scale is an important open problem. Crucially, however, progress can be made if one postulates that $\ell(T)$ actually diverges at some — possibly zero — temperature T_c which will thus nonambiguously define the glass transition temperature one seeks to determine. It should coincide with the temperature at which $t_{\rm rel}(T)$ also diverges.

A measure of $\ell(T)$ in a liquid can be obtained via the spatial decay of the following correlation function:

$$C(r, t) = \langle F(r + r_0, t)F(r_0, t)\rangle - \langle F(r + r_0, t)\rangle\langle F(r_0, t)\rangle, \quad (1)$$

where $F(r, t) = \rho(r, t + t_0)\rho(r, t_0)$; $\rho(r, t)$ is the density at time t and position r, r_0 and t_0 are arbitrary position and time, respectively; $\langle \cdots \rangle$ is a standard ensemble average. Alternatively, $F(r, t)$ can be replaced by any two-time function, $F(r, t) = A(r, t + t_0)B(r, t_0)$, where A and B are physical observables. It is useful to consider $C(r, t_{\rm rel})$, defining $t_{\rm rel}$ in a standard way from the time decay of $\langle F(r, t)\rangle$. In essence, Eq. (1) is a two-time, two-point correlation function measuring correlations in trajectory space [7,8]. Its physical content is similar to most of the measurements of dynamical heterogeneities which amount to first distinguishing between "fast" and "slow" particles and then to checking how these are spatially correlated [6]. Indeed, the correlators $F(r, t)$ precisely tell how fast a particle is, while (1) measures

the corresponding spatial correlations. The use of the dynamic function $F(r, t)$ makes an arbitrary criterion to distinguish between particles unnecessary. Variations of the correlator (1) have been studied. In Refs. [7,9], the density fluctuations were replaced by an arbitrarily defined overlap between configurations, and by particle displacements in Refs. [10–12]. Is is found that $\ell(T)$ decreases with T, although simulations are yet too limited to confirm a possible divergence at low temperature. Note last that the commonly discussed "non-Gaussian parameter" is related to Eq. (1) when A and B refer to the particle positions, but for $r = 0$. As such, it contains no information about $\ell(T)$.

The observation of a growing coherence length scale has deep consequences which we start to explore in this Letter. This indeed suggests that finite-time local dynamical functions $F(r, t)$ become long-range correlated when the glass phase is approached. As such they play a role similar to that of the order parameter in a standard continuous phase transition, the novelty being that this order parameter is a dynamic quantity, as dictated by the physics of the problem we consider. This statement, which is central to our approach, has to be distinguished from the use of the nonergodicity parameter, $\langle F(r, t \to \infty)\rangle$, to define the glass state [1]. Having identified an "order parameter," it becomes a simple task to extend the tools developed for conventional transitions to study the problem of the glass transition, starting here with finite-size scaling techniques.

Define first the parameter $B(T, L)$ as

$$B(T, L) = 1 - \frac{\langle\varphi^4\rangle_L - 4\langle\varphi^3\rangle_L\langle\varphi\rangle_L + 6\langle\varphi^2\rangle_L\langle\varphi\rangle_L^2 - 3\langle\varphi\rangle_L^4}{3[\langle\varphi^2\rangle_L^2 - 2\langle\varphi^2\rangle_L\langle\varphi\rangle_L^2 + \langle\varphi\rangle_L^4]}, \tag{2}$$

with $\varphi = \varphi(t_{\rm rel}(T)) \equiv L^{-d}\int d^d r F(r, t_{\rm rel}(T))$, where d is the space dimensionality. The crucial information is that the average $\langle\cdots\rangle_L$ is performed in a system of finite linear size L. By construction, $B(T, L) = 0$ when the fluctuations of φ are Gaussian, since it is built from the fourth cumulant of the probability distribution function of φ, $P_L(\varphi)$. This distribution is Gaussian when $L \gg \ell(T)$ as a consequence of the central limit theorem. However, $P_L(\varphi)$ becomes non-Gaussian when $L < \ell(T)$, and $B(T, L)$ is therefore nonzero in that case. Moreover, it is natural to expect that $B(T, L)$ becomes a scaling function of the variable $\ell(T)/L$ in the regime $\ell(T), L \gg a$, where a is the microscopic length scale of the problem, e.g., the particle size in a simple liquid. Also, $P_L(\varphi)$ should be independent of L and $\ell(T)$ in the regime $a \ll L \ll \ell(T)$. The latter assumption implies, in particular, that $B(T_c, L)$ is a constant "universal" number since $L \ll \ell(T_c) = \infty$ is automatically satisfied at the transition. Therefore, the curves $B(T, L)$ versus T for various L cross at T_c. The definition (2) is naturally inspired by the classical paper [13], where a numerical finite-size scaling analysis of the Ising model was performed. In that case, φ was the order parameter of the paramagnetic-ferromagnetic transition, i.e., the magnetization density.

We also define the susceptibility

$$\chi(T, L) = \frac{L^d}{T}[\langle\varphi^2\rangle_L - \langle\varphi\rangle_L^2], \tag{3}$$

again explicitly retaining its L dependence. Similarly, one expects this function to exhibit a scaling behavior for large L and $\ell(T)$. When the correlator $C(r, t)$ is itself a simple function of the ratio $r/\ell(T)$, one gets

$$\chi(L, T) \approx L^d \tilde{\chi}\left[\frac{\ell(T)}{L}\right], \tag{4}$$

where the scaling function $\tilde{\chi}(x)$ behaves as $\tilde{\chi}(x \ll 1) \approx x^d$ and $\tilde{\chi}(x \gg 1) \approx$ const. The assumption made on the correlator is supported by all known simulation results so far [7,9,11]. It is satisfied also in the two models studied below. This point certainly needs further investigation in realistic supercooled liquids.

The suggested analysis requires extrapolations towards the glass phase, as all equilibrium measurements in supercooled liquids do. However, the new quantities $B(T, L)$ and $\chi(T, L)$ defined above are much richer than, say, an average relaxation time since they contain information on the full distribution $P_L(\varphi)$, so that new informations on the location and the nature — divergence of length and time scales — of the glass transition can be gained.

We now use the analysis suggested above to study two lattice models. We start with the one-dimensional version of the spin facilitated Ising model introduced in Ref. [4]. Spin facilitated models have been introduced as caricatures of real supercooled liquids, in the sense that they are nondisordered spin models displaying glassy dynamics with only very simple static correlations. We have chosen this version of the model since it is reasonably well characterized [4,8,14]. Moreover, as a one-dimensional lattice model, it is relatively easy to simulate on very large time and length scales, nicely confirming the expected scaling behavior. These results thus constitute a very interesting first application of the methods described above. The model is defined by the Hamiltonian

$$H = \sum_{i=1}^{N} s_i, \tag{5}$$

where $s_i = 0, 1$ are two-state observables located at the sites of a chain of size N with periodic boundary conditions. Glassiness originates from the chosen dynamics, since the static properties of the noninteracting Hamiltonian (5) are completely trivial. In a standard Metropolis algorithm, the transition rates are $w(s_i \to 1 - s_i) = \min(1, e^{-\beta\Delta E})$, where ΔE is the energy change in the transition $s_i \to 1 - s_i$. Here, the rates are given instead by $w'(\Delta E) = (s_{i-1} + s_{i+1})w(\Delta E)$ meaning that a spin can flip only if it has at least one nearest neighbor whose value is 1. This kinetic constraint implies glassiness. From (5), one has $\langle s_i\rangle = (1 + e^{1/T})^{-1}$, so that the density of

spin 1 becomes small at low T therefore allowing fewer and fewer transitions. This leads to an Arrhenius relaxation time, $t_{\rm rel} = e^{3/T}$. Also, the dynamics becomes more and more cooperative when approaching the glass transition at $T_c = 0$, with a diverging coherence length $\ell(T) = e^{1/T}$ [8,14].

We simulate this model using a continuous time algorithm in the temperature interval $T \in [0.125, 1.0]$, approximately corresponding to 13 decades in relaxation times. For each temperature, a large system of $N = 2 \times 10^4$ spins is studied, and averages are performed in subsystems of finite-size $L \in [2, 1024]$ for a number of independent configurations ranging from 200 to 20 000. Fortunately, more statistics is required at higher temperatures where deviations from Gaussian behavior are smaller. A natural choice for the dynamic function $F(i, t)$ in the definition (2) is the persistence of the spin s_i at site i. The mean persistence is given by $\langle F(i, t)\rangle_{L=\infty} = \exp[-\sqrt{t/t_{\rm rel}(T)}]$, which defines the relaxation time.

Our results for this model are presented in Figs. 1 and 2. The inset of Fig. 1 shows the variation of $B(T, L)$ as a function of $1/T$ for various sizes L. As expected, $B(T, L)$ crosses over from $B(T, L) = 0$ at high T to a nonzero value at low T, the locus of the crossover being L dependent. The various curves clearly cross at $T_c = 0$ only. One observes in the main figure an excellent collapse of the data, showing that $B(T, L)$ satisfies the expected scaling form $B(T, L) \approx \tilde{B}(\ell(T)/L)$. Since $P_L(\varphi)$ is bimodal when $L \ll \ell(T)$, it is a simple task to compute $B(T_c = 0, L) = \tilde{B}(x \to \infty) = 0.566\ldots$. The inset of Fig. 2 presents the susceptibility $\chi(T, L)$ as a function of $1/T$ for various sizes L. Again, one clearly sees the expected behaviors: $\chi(T, \infty)$ diverges when $T \to 0$ as $e^{1/T} = \ell(T)$, while for finite L the divergence is smeared when $\ell(T) > L$. The main figure shows the perfect collapse of the data suggested by Eq. (4) when $\chi(L, T)L^{-1}$ is plotted as a function of $\ell(T)/L$ (remember that $d = 1$).

As a second application of the techniques described above, we study the kinetically constrained lattice gas introduced in Ref. [5]. The model consists of hard spheres on a cubic lattice of linear size N. In this case, temperature plays no role, and the relevant control parameter is the density of particles, ρ. This lattice gas is also complemented by kinetic rules which make the dynamics glassy. In a nonconstrained lattice gas, particles move to an empty nearest neighbor site with unit rate. In the present model, particles can move to an empty nearest neighbor, provided the particle has fewer than four neighbors before and after the move. This kinetic rule aims at reproducing dynamics of a supercooled liquid, where particles can hardly escape the cage formed by their neighbors. As a simple lattice model for the glass transition, it has been much studied since its introduction. Simulations have reported a dynamical arrest at a density $\rho_c \approx 0.881$ where the relaxation time apparently diverges as a power law [5]. Very recently, however, a lower bound for the diffusion constant was analytically derived for this model, showing that the divergence of the relaxation time at a finite density found in simulations is only apparent [15]. The growth of $t_{\rm rel}(\rho)$ with ρ is in fact extremely abrupt, $\ln\ln t_{\rm rel}(\rho) \propto (1 - \rho)^{-1}$, with an associated coherence length $\ln\ln\ell(\rho) = c/(1 - \rho)$, where c is a numerical constant. The extremely fast increase of $t_{\rm rel}$ can easily be numerically confused with a true divergence at a finite density. This more difficult problem represents thus a highly selective test for the methods described in this Letter. We now show that finite-size scaling allows one to distinguish between a true and an apparent divergence of the relaxation time in this model.

We use again a continuous time Monte Carlo algorithm to study a model of linear size $N = 24$ for densities

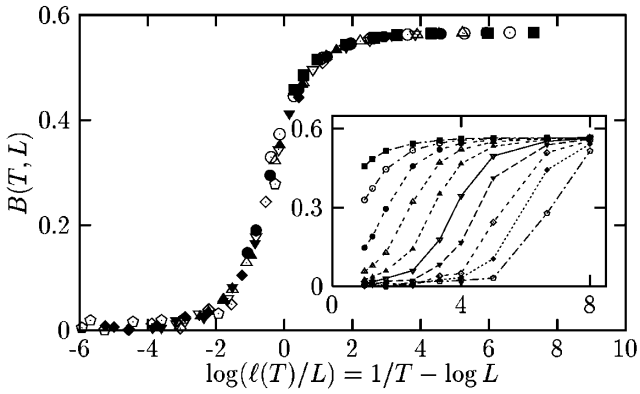

FIG. 1. Inset: The parameter $B(T, L)$ as a function of the inverse temperature $1/T$ for various system sizes $L = 2, 4, \ldots, 1024$ (top to bottom) cross at the glass transition $T_c = 0$. Main: The parameter $B(T, L)$ as a function of the scaling variable $\ln(\ell(T)/L)$ with $\ell(T) = \exp(1/T)$. Both figures are for the one-dimensional spin facilitated Ising model.

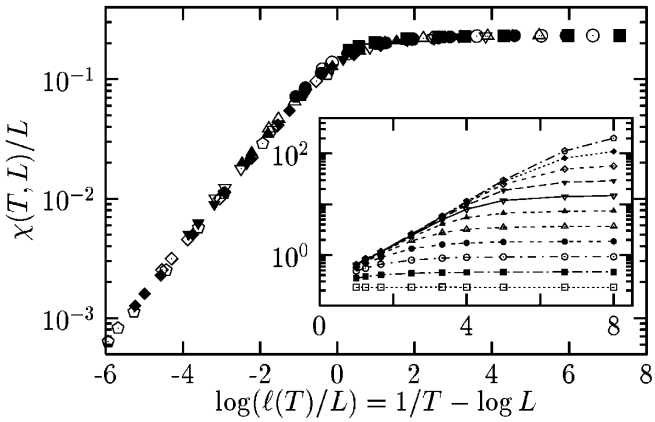

FIG. 2. Inset: The susceptibility $\chi(T, L)$ as a function of the inverse temperature $1/T$ for the same sizes as Fig. 1 increasing from bottom to top. Main: The rescaled susceptibility $\chi(T, L)L^{-1}$ as a function of the scaling variable $\ln(\ell(T)/L)$ with $\ell(T) = \exp(1/T)$. Both figures are for the one-dimensional spin facilitated Ising model.

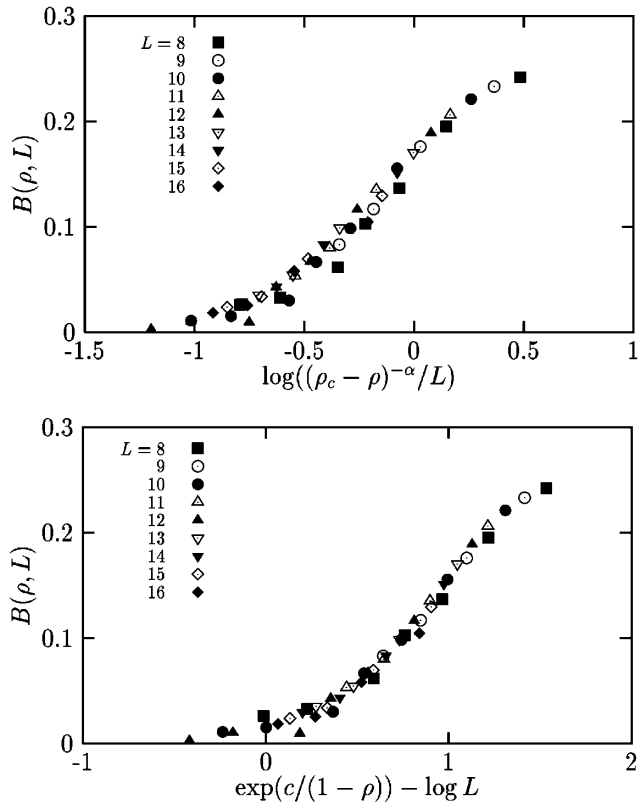

FIG. 3. The parameter $B(T, L)$ as a function of the reduced variable $\ln(\ell(\rho)/L)$ with $\ell = (\rho_c - \rho)^{-\alpha}$ (top) or $\ell = \text{expexp}[c/(1-\rho)]$ (bottom). A much better collapse is obtained in the bottom part, with one less free parameter. Systematic deviations are visible in the top part. Both are for the kinetically constraint lattice glass of Ref. [5].

$\rho \in [0.75, 0.87]$, covering almost 8 decades of relaxation times. We use $F(i, t) = \rho(i, t)\rho(i, 0)$ as a dynamical quantity, where $\rho(i, t)$ is the density at site i and time t. The averages are performed in subsystems of linear sizes L, using 10^4 independent initial conditions at each density. The correlator (1) is well described by a simple exponential form, $C(r, t_{\text{rel}}) = e^{-r/\ell(\rho)}$. The latter measurements are also a check against spurious finite-size effects, since they show that $N \gg \ell(\rho)$ for the densities studied here.

We show our results in Fig. 3 where the parameters $B(\rho, L)$ are represented as a function of two possible scaling variables ℓ/L. The top figure assumes a power law divergence of the coherence length $\ell(\rho) \approx (\rho_c - \rho)^{-\alpha}$ at a finite density $\rho_c < 1$, while the bottom figure makes use of the results of Ref. [15], with a divergence at $\rho_c = 1$ only. The best collapses obtained in the two cases are shown, using the values $\rho_c = 0.882$, $\alpha = 0.59$, and $c = 0.167$. The second scaling is clearly superior, the first one having systematic deviations either at large or small sizes. The figure shown here is a compromise between those. Moreover, the second scaling has one less free parameter. The same conclusions are drawn from the susceptibility (3). These three arguments thus discriminate both possibilities, and we numerically confirm the absence of a dynamical arrest at finite density in this model.

To summarize, we have shown that finite-size scaling techniques can be used to study the glass transition. This follows from the identification of local two-time observables $F(r, t)$ as relevant "order parameters" with interesting fluctuations and correlations. We have defined two quantities, $B(T, L)$ and $\chi(T, L)$, whose scaling behavior allows one to locate the glass transition in a much more accurate manner than an extrapolation of simpler observables like the relaxation time. The new method straightforwardly applies to any glass-forming systems, including off-lattice models. More generally, our results raise the hope that more concepts and methods borrowed from continuous phase transitions studies can also be used.

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