Spatial Structures and Dynamics of Kinetically Constrained Models of Glasses

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Kob and Andersen's simple lattice models for the dynamics of structural glasses are analyzed. Although the particles have only hard core interactions, the imposed constraint that they cannot move if surrounded by too many others causes slow dynamics. On Bethe lattices, a dynamical transition to a partially frozen phase occurs. In finite dimensions there exist rare mobile elements that destroy the transition. At low vacancy density v, the spacing Ξ between mobile elements diverges exponentially or faster in $1/v$. Within the mobile elements, the dynamics is intrinsically cooperative, and the characteristic time scale diverges faster than any power of $1/v$ (although slower than Ξ). The tagged-particle diffusion coefficient vanishes roughly as Ξ^{-d}.

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In many liquids, dramatic slowing down occurs on cooling, equilibrium cannot be achieved, and structural relaxation becomes complicated and spatially heterogeneous [1–3]. Despite a great deal of effort, these remarkable phenomena associated with the "glass transition" are still far from understood. Indeed, the most basic issues are unresolved: Is the rapid slowing down due to the proximity to an equilibrium phase transition, albeit in a restricted part of phase space? Or is the underlying cause entirely dynamical? In either case, are there characteristic length scales that grow substantially near the glass transition?

Theoretical developments have been hampered by a shortage of models that capture essential features yet are simple enough to be analyzed. Exceptions are *kinetically constrained* models [4,5] based on the ansatz that the glass transition is caused by geometrical constraints on dynamical rearrangements with static correlations playing no role (see, e.g., [6]). These are also models for granular media (see, e.g., [7]) for which slow dynamics occur already at densities well below close packing [8].

We focus on one of the simplest kinetically constrained models, the Kob-Andersen model (KA) [9]. The KA is a single component lattice gas with no static interactions other than hard core exclusion and dynamics given by a continuous time stochastic process. Each particle attempts, at a fixed rate, to move to a randomly chosen empty neighboring site, *but* the jump is allowed only if both before and after the move the particle has no more than some number, m, of neighboring particles. This corresponds to *vacancies* moving only if the initial and final sites have at least $s = z - m - 1$ neighboring vacancies, with z the coordination number of the lattice. Since this dynamic satisfies detailed balance, the trivial distribution that is uniform over all configurations with a fixed number of particles is stationary; there can thus be no equilibrium transition. Nevertheless, the dynamics is sluggish and heterogeneous at high density [9,10]. For a three-dimensional cubic-lattice with $s = 2$, fits of the self-diffusion coefficient of a tagged-particle, D_S, strongly suggest a dynamical glass transition at $\rho_c \simeq 0.881$, above which D_S appears to vanish and the structural relaxation time to diverge [9].

Our analysis shows that there are four classes of behavior for general KA models. These can be characterized by the dependence of D_S on the *vacancy density* v: (N) *normal* with $D_S \sim v^q$ as $v \to 0$; (C) *collective freezing* with $D_S \to 0$ faster than any power of v as $v \to 0$; (T) *dynamical transition* with $D_S \to 0$ as $v \searrow v_c$ with a nonzero critical $v_c = 1 - \rho_c$; and (F) *always frozen* with a finite fraction of the particles never moving for any v. Normal behavior occurs if a finite cluster of q vacancies can move through an otherwise totally filled lattice, e.g., a triangular lattice with $s = 1$ in which vacancy pairs ($q = 2$) are mobile. This yields D_S proportional to the cluster concentration, v^q. At the opposite extreme, a square lattice with $s = 2$ is always frozen, as a four particle square can never move. We focus on the interesting intermediate cases, C and T. We show that a dynamical transition takes place on (treelike) Bethe lattices—even with finite size loops. But such a transition *cannot* occur on finite-dimensional lattices; if neither normal nor always frozen, these exhibit collective freezing with the dominant dynamical processes involving a number of vacancies that diverges as $v \to 0$. This class (C) includes the original $s = 2$ cubic-lattice.

We first analyze Bethe lattices: infinite treelike graphs with fixed connectivity z, which crudely approximate

high-dimensional or high-coordination-number lattices. Monte Carlo simulations of 10^4 sites with $z = 4$ and $s = 1$ (crudely mimicking the $s = 1$ square lattice) suggest a dynamical correlation length (as defined in [11]) that diverges at a critical density $\rho_c \simeq 0.89$ [12]. Approaching this apparent dynamical transition, the local-density correlation function displays two-step relaxation, as for supercooled liquid [13].

The treelike structure of Bethe lattices enables analytic study by iteration. Arranging the tree with $k = z - 1$ branches going up from each node and one going down, we focus on the following events for a chosen *node* and rearrangements *restricted* to the *subtree above it*: ($\mathcal{A}$) node is occupied by a particle which cannot move up even if the site below it is empty; ($\mathcal{B}$) node is empty, but a particle can never move onto it from below; (C) node is occupied by a particle which can only move up if the node below it is empty; ($\mathcal{D}$) node is none of the above. Self-consistent equations for the probabilities of these events can be found by combining k branches together with the site below them to which they are all linked [12]. For $s = 1$ with $z = k + 1 = 4$, this yields a critical density, $\rho_c \cong 0.888825$ in agreement with simulations. Below ρ_c, $\mathrm{Prob}[\mathcal{A}] = \mathrm{Prob}[\mathcal{B}] = 0$; particles are mobile and diffusive at long times. At ρ_c, $\mathrm{Prob}[\mathcal{A}] = A_c \neq 0$ and the fraction of immobile particles jumps discontinuously, as does the *extensive configurational entropy*, $S_C = \ln[\text{ different ergodic componets}]$ [12]. Above ρ_c, $\mathrm{Prob}[\mathcal{A}] - A_c \sim \sqrt{\rho - \rho_c}$, the singularity indicative of diverging length and time scale. The KA dynamical transition on this Bethe lattice (such as $1/r^2$ percolation in one-dimension [14]) thus has both critical and "first order" characteristics. Interestingly, infinite-range *quenched random p*-spin models, conjectured to be related to the glass transition, also exhibit a dynamical transition with similar features [13].

The basic features found above hold for generic KA models on Bethe lattices with $s \neq 0, k$, including those with *finite size loops* [12], i.e., for the so-called pure Husimi trees [15] and for Bethe lattices where single sites are replaced by $L \times L$ squares (these interpolate among the Bethe lattice, $L = 1$, and the square lattice, $L = \infty$). The only exceptions are cases in which particles with two occupied neighbors cannot move ($m = 1$): the transition is then continuous and $\rho_c = 1/k$, the critical density for conventional site percolation. This equivalence is due to the fact that the dynamical transition is associated with the emergence of an infinite cluster of permanently blocked particles. In general, a frozen phase will exist only if an infinite cluster remains after all the particles mobile under KA rules have been iteratively removed. This is a type of *bootstrap percolation*, already introduced in this [9] and broader [16–18] contexts. For Bethe lattices, an exact solution [12] (see [18] for a simpler example) yields the same critical density found above.

We now turn to finite dimensions and show that the dynamical transition is destroyed by exponentially rare processes, which will only occur in sufficiently large systems. Nevertheless, at high densities, the dynamic is intrinsically collective and extremely slow. Furthermore, the ghost of the Bethe lattice transition may cause a sharp crossover at an apparent critical density.

We focus initially on the simple $s = 1$ square lattice case. First, let us define a configuration as *framed* if all its boundary sites are empty (see, for example, the square on the right of Fig. 1) and as frameable if, by an allowed sequence of moves, a framed configuration can be reached (see, for example, the square on the left of Fig. 1). A key observation is that the lines of vacancies on the boundaries of a framed square can be shifted (moving vacancies starting from the corner) so that any nearest-neighbor pair of particles inside the square can be sandwiched between two lines of vacancies. The position of this pair of particles can then be exchanged and the vacancy lines afterwards shifted back to their original positions. The net result is a pair exchange of particles with all other particles returned to their original positions. By combining such pair exchanges, all frameable configurations with the same density can be connected. Thus, these form an irreducible component $\mathcal{Z}_\rho$ of the configuration space. We must now show that almost all random configurations with a given $\rho < 1$ belong to $\mathcal{Z}_\rho$ in the thermodynamic limit. This can be done iteratively by observing that an l by l frameable configuration that has at least two vacancies externally adjacent to each of its sides is also an $l + 2$ by $l + 2$ frameable configuration; see Fig. 1. Starting from a two by two frameable "nucleus" of vacancies, one can thus grow an L by L frameable configuration, if the requisite vacancies are present in each concentric shell. The probability that this occurs is $P_L(\rho) = (1 - \rho)^4 \prod_{l=2}^{L/2} [1 - \rho^{2l} - 2l\rho^{2l-1}(1 - \rho)]^4$, which converges to a nonzero probability, $P_\infty(\rho)$. Thus for all ρ, infinite frameable squares exist with all frameable configurations within them being reachable. For small vacancy densities, $P_\infty(\rho) \simeq e^{-2\hat{K}_F/v}$ with $\hat{K}_F \simeq 4.48$. Moreover, $P_L(\rho)$ depends weakly on L for $L > \xi(\rho) \sim \ln(1/v)/v$; ξ is thus the *core* size of frameable regions. Note that P_L is the probability that a frameable square can be constructed around a nucleus at a *fixed*

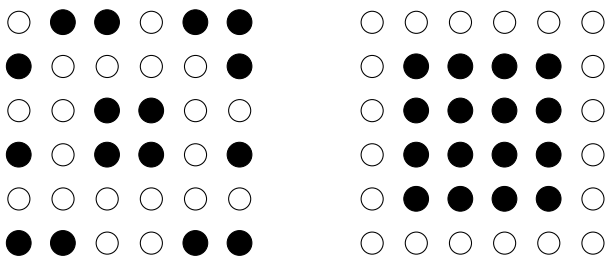

FIG. 1. On the left, there is a 4×4 framed configuration with two vacancies (white circles) adjacent to each side. After moving the external vacancies to the corners, the framed square can be expanded to the 6×6 framed configuration on the right.

position. On the other hand, the probability, $F_L(\rho)$, that a large square is frameable is roughly given by considering the $O([L/\xi]^2)$ possible positions of a nucleus within the square. Although the probabilities of a configuration being frameable around each of these possible nuclei are not independent, if the nuclei are separated by more than a core diameter, they are roughly independent. Indeed, it can be proven that F_L asymptotes to one for $L \gg \Xi$, with the long crossover length scale $\Xi(\rho)$ given, as expected from the above argument, by $\Xi^2 P_\xi(\rho) \sim \Xi^2 P_\infty(\rho) \sim \xi^2$.

We have shown that at any density almost all configurations of sufficiently large systems can be reached from one another. This *excludes* the possibility of a dynamical transition [19]. The phase space of systems with linear size, L, larger than the crossover length Ξ is covered almost entirely by a single ergodic component, while the phase space for $L < \Xi$ is typically decomposed into many disjoint parts. The framing analysis yields an *upper bound* for Ξ.

A lower bound for Ξ can be obtained from bootstrap percolation arguments. From a random configuration with density ρ, iteratively remove all particles with less than three occupied neighbors. If any particles remain, they must form a system-spanning cluster of frozen particles, and the configuration space will thus be broken up into many disjoint pieces. This happens with high probability for $\ln L < K_F/\upsilon$ with $K_F = \pi^2/18$ [16,20], yielding a lower bound for Ξ with the same density dependence as the above upper bound, but a different constant K_F less than $\hat{K}_F$ of the upper bound. However, a better definition of framed configurations yields a $\hat{K}_F$, which is identical to K_F [12]. The corresponding frame is *minimal* and the asymptotic form of the crossover length $\Xi \sim e^{\pi^2/18\upsilon}$ *exact* (up to subdominant factors).

All other finite-dimensional KA models can be analyzed by generalizing the notion of frameable configurations; we thereby rule out *any* dynamical transitions. For d-dimensional hyper-cubic lattices, $s = 0$ and $s > d - 1$ correspond, respectively, to N and F, while all the nontrivial cases, $1 \le s \le d - 1$, are class C, collective freezing [12]. Analogous expansion arguments yield upper bounds on their crossover lengths: $\Xi < \exp^{os}[K_U(s,d)/\upsilon^{1/d-s}]$, with $\exp^{os}$ as the exponential function iterated s times and $K_U(s,d)$ as a positive constant given in [12]. Bootstrap percolation yields a lower bound of the same form but with a different constant $K_L(s,d)$ [21]. The scale ξ, above which the probability that a nucleus is expandable saturates, is generally of order $\ln\Xi$, up to log-log factors.

We now turn to the dynamics, focusing again on the $s = 1$ square lattice model at high density. Vacancies will typically be far apart or in small clusters that cannot move. However, vacancies can move within the cores of large frameable regions. And these cores of size $\xi \sim \ln(1/\upsilon)/\upsilon$ are themselves mobile because they are likely to find vacancies on *all* the ξ successive line segments needed for them to move in a given direction. A generic particle cannot move substantial distances except when a mobile core passes by; assuming independent core motion, a tagged-particle will thus diffuse with $D_S \sim D_M n_M$, where $n_M \sim 1/\Xi^2$ is the density of the mobile cores which diffuse with $D_M \simeq \xi^2/\tau_\xi$, $1/\tau_\xi$ being the typical relaxation rate of a core. Using our framing analysis and a generalization of the technique of [22], we have proved that, indeed, $D_S > 0$ for all ρ [12].

The relaxation time, τ_ξ, of a core is dominated by the time to reach the most severe *bottleneck* in configuration space. Since all configurations are equiprobable, this is proportional to $\exp(\Delta S)$, the ratio of the number of accessible (frameable) configurations of the core to the number in the bottleneck [12]. The worst case scenario in which there is a *single* bottleneck configuration corresponds to $\Delta S = S_{\text{total}} \simeq \ln\xi!$, yielding an upper bound $\tau_\xi < \xi!$. A lower bound is obtained by noting [12] that, to equilibrate a frameable square of size ξ, one has to pass through configurations with the nucleus in a *corner* of the square. Using a transfer matrix technique, we obtain for $\ell \times \ell$ minimally frameable squares an entropy difference $\Delta S \approx \Upsilon\sqrt{\ell} + \alpha \ln\ell + C$, with $\Upsilon = 2\sqrt{6+\sqrt{22}} - 2\sqrt{3} \cong 3.075$ and α computable in principle [12]. Simulations of frameable squares with $\ell = 4$ to $\ell = 16$ yield equilibration times with $\log\tau_\ell$ increasing slower than ℓ and in good agreement with the square root law with the *predicted* coefficient Υ plus logarithmic corrections. We thus conjecture that the entropic bottlenecks for moving the core of a mobile region are equivalent, up to subdominant factors, to the entropy loss associated with forcing the nucleus of such core to a corner.

As long as bottlenecks for motion of the mobile cores are not too much larger than the above estimates, $\tau_\xi \ll \Xi^2$ and the dominant contribution to $D_S \sim D_M n_M$ will arise from the low density of mobile regions rather than the long time needed for motion within them. Thus, to leading order in υ^{-1}, $\ln D_S \approx 2K_F/\upsilon$ for the square lattice model with $s = 1$. Numerical simulations at high densities fit this form well; see Fig. 2. In particular, $\lim_{\upsilon\to 0} \upsilon \ln D = 2K_F \cong 0.9 - 0.95$ cf. the predicted $2K_F \cong 1.1$. For the other KA models, we obtain similar relationship between D_S and n_M. For the $s = 2$ cubic case, this yields $\ln\ln D_S^{-1} \propto 1/\upsilon$, a result consistent with a recent finite size scaling analysis [23].

Thus far, we have focused on the high density limit. Yet simulations on KA and other kinetically constrained models suggest a transition at a nontrivial critical density [9]. This is a natural consequence of the existence of a dynamical transition on Bethe lattices: in finite dimensions, the transition will be replaced by a crossover, which, in some cases, could be quite sharp. In the $s = 1$ square lattice case, a three-vacancy element can move along a network of other vacancies, provided these are linked no more weakly than via second neighbors (along axes or diagonals). At low density, there will always be a percolating vacancy network of such type. As the density

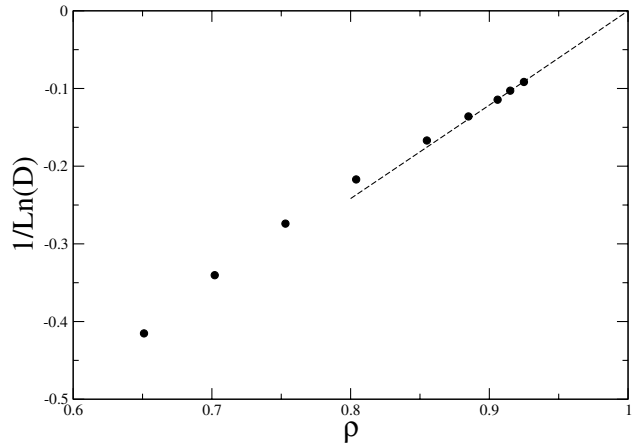

FIG. 2. $(\ln D)^{-1}$ as a function of ρ for 400×400 square lattice with $s = 1$. Straight line shows $\ln D_S \propto 1/v$ behavior.

increases, diffusion slows down as the vacancy percolation cluster, on which mobile elements rely, shrinks. Beyond the second-neighbor vacancy percolation transition at ρ_{2P}, the *simple* mobile elements can no longer move. But by *growing*, mobile elements can still form; they have to become large enough to find a percolating cluster on which objects of their size can move. At high densities, such mobile elements will be the cores of (minimal) frameable regions that we have discussed above. If the crossover from small mobile objects and behavior similar to that on Bethe lattices is sharp enough, a substantial range of "critical" behavior of D_S—and of relaxation times—might be observed near to an *apparent* transition at $\rho_G \approx \rho_{2P}$. This reasoning can be generalized to other KA models—indeed, a sharper crossover should occur in higher dimensions—and could be an explanation of the apparent dynamical transition found for the original cubic-lattice KA [9].

We have shown how simple kinetically constrained models exhibit concretely some of the qualitative features conjectured to be the cause of the dramatic slowing down in structural glasses. In particular, finite-dimensional models can "almost" have a dynamical phase transition; such exist on Bethe lattices and percolation arguments suggest it is replaced by a crossover—possibly very sharp—in finite dimensions. The dominant dynamical processes involve cooperative rearrangements of regions whose size diverges in the high density limit; nevertheless, these yield a nonvanishing self-diffusion coefficient at all densities. But the length and time scales of these processes grow extremely rapidly at high densities. This is reminiscent of the super-Arrhenius increase of the relaxation time as fragile liquids are cooled.

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