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Dynamic first-order phase transition in kinetically constrained models of glasses

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We show that the dynamics of kinetically constrained models of glass formers takes place at a first-order coexistence line between active and inactive dynamical phases. We prove this by computing the large-deviation functions of suitable space-time observables, such as the number of configuration changes in a trajectory. We present analytic results for dynamic facilitated models in a mean-field approximation, and numerical results for the Fredrickson-Andersen model, the East model, and constrained lattice gases, in various dimensions. This dynamical first-order transition is generic in kinetically constrained models, and we expect it to be present in systems with fully jammed states.

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An increasingly accepted view is that the phenomenology associated with the glass transition [1] requires a purely dynamic analysis, and does not arise from an underlying static transition (see however [2]). Indeed, it has been suggested that the glass transition manifests a first-order phase transition in space and time between active and inactive phases [3]. Here we apply Ruelle's thermodynamic formalism [4, 5] to show that this suggestion is indeed correct, for a specific class of stochastic models. The existence of active and inactive regions of space-time, separated by sharp interfaces, is dynamic heterogeneity, a central feature of glass forming systems [6]. This phenomenon, in which the dynamics becomes increasingly spatially correlated at low temperatures, arises naturally [7] in models based on the idea of dynamic facilitation, such as spin-facilitated models [8, 9], constrained lattice gases [10, 11] and other kinetically constrained models (KCMs) [12]. Fig. 1 illustrates the discontinuities in space-time order parameters at the dynamical transition in one such model, together with the singularity in a space-time free energy, as a function of a control parameter to be discussed shortly.

The thermodynamic formalism of Ruelle and coworkers was developed in the context of deterministic dynamical systems [4]. While traditional thermodynamics is used to study fluctuations associated with configurations of a system, Ruelle's formalism yields information about its trajectories (or histories). The formalism relies on the construction of a dynamical partition function, analogous to the canonical partition function of thermodynamics. The energy of the system is replaced by the dynamical action (the negative of the logarithm of the probability of a given history); the entropy of the system by the Kolmogorov-Sinai entropy [13], and the temperature by an intrinsic field conjugate to the action. This formalism has been exploited recently to describe the chaotic properties of continuous-time Markov processes [5].

In this work, we define the dynamical partition sum [4, 5] for our stochastic systems by

$$Z_K(s,t) = \sum_{\text{histories}} \text{Prob(history)} e^{-s\hat{K}(\text{history})}, \quad (1)$$

where the sum is over histories from time 0 to time t; the probability of a history is Prob(history); and $\hat{K}$(history) is the number of configuration changes in that history. $\hat{K}$(history) is a direct measure of the activity in a history: an active trajectory has many changes of configuration,

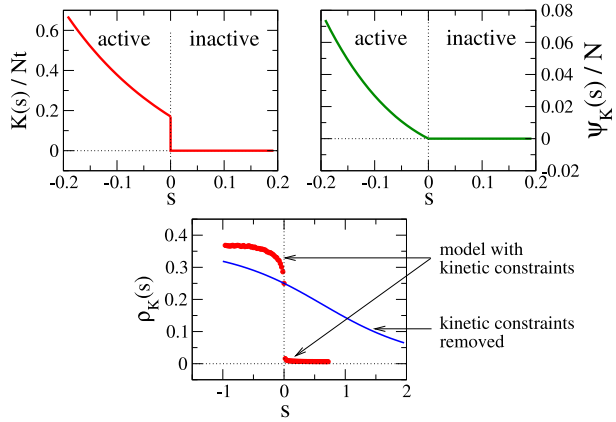

FIG. 1: First order transition in terms of the field s. (Top) The dynamical order parameter $K(s)$ (the average number of configuration changes in a trajectory) and its large-deviation function $\psi_K(s)$ for the FA model, calculated in a mean-field approximation, for $d = 3$ and $T = 0.5$: see Eqs. (6-9). The large deviation function is singular at $s = 0$ and the order parameter K has a first-order jump. The dynamics has two phases, an active one for $s < 0$ and an inactive one for $s > 0$. Physical dynamics take place at $s = 0$, where the two dynamic phases coexist. (Bottom) An alternative order parameter $\rho_K(s)$ (the average number of excited sites in a trajectory: see Fig. 2) in the $d = 1$ FA model at $T = 0.91$, calculated numerically in a finite system ($N = 100$ sites). The transition is absent when the kinetic constraints are removed.

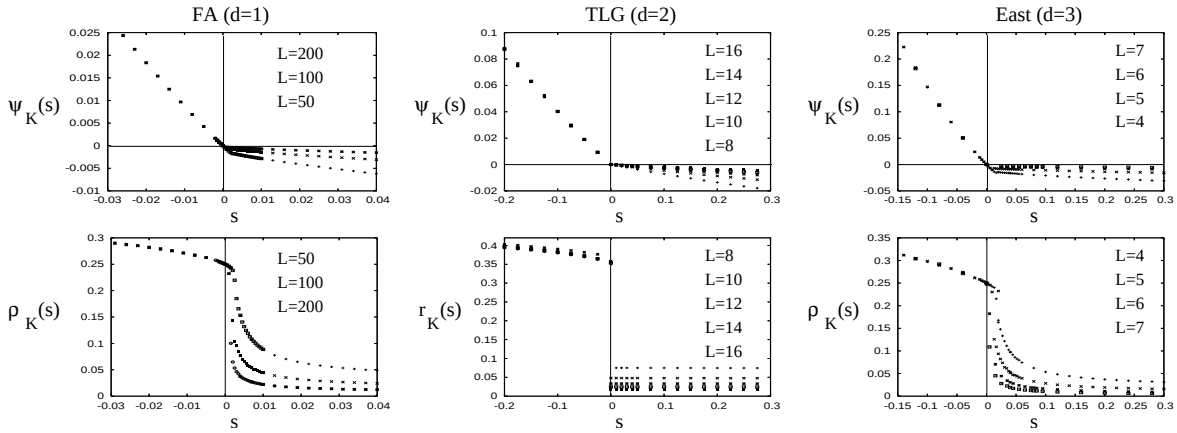

FIG. 2: Large-deviation function ψ_K and order parameters as a function of system size, for the FA model ($d = 1$), the TLG ($d = 2$) and the East model ($d = 3$). For the FA and East models, we show the average density of excited sites $\rho_K(s) \equiv Z(s,t)^{-1} \sum_{\text{histories}} \text{Prob(history)} e^{-s\hat{K}(\text{history})} \rho(\text{history})$, where $\rho(\text{history}) = (Nt)^{-1} \int_0^t \mathrm{d}t' \sum_i n_i(t')$. For the TLG, we replace $\rho(\text{history})$ by $r(\text{history}) = (Nt)^{-1} \int_0^t \mathrm{d}t'\, r(\mathcal{C}(t'))$, to obtain the averaged escape rate $r_K(s)$. We use representative conditions: $T = 0.91$ for the FA and East models, and density 0.5 in the TLG. Results were obtained by the dynamical method of [20].

an inactive one has few or none. At long times

$$Z_K(s,t) \propto e^{t\psi_K(s)}, \tag{2}$$

where $\psi_K(s)$ is the large deviation function for $\hat{K}$. The quantity $-\psi_K(s)$ is a free energy per unit time, for trajectories [14]. When t is large, the derivatives of $\psi_K(s)$ give the cumulants of $\hat{K}$. For example, the mean number of configuration changes in a trajectory of length t is $K(0)$, where

$$K(s) = -t\frac{\mathrm{d}}{\mathrm{d}s}\psi_K(s). \tag{3}$$

For some models, it is reasonable to use the dynamical action, or dynamical complexity, $\hat{Q}_+ \equiv \ln[\text{Prob(history)}]$ [5], as an order parameter for dynamical activity, in place of $\hat{K}$. In that case the large deviation function is the topological pressure and the average of $(-\hat{Q}_+/t)$ at $s = 0$ is the Kolmogorov-Sinai entropy [4, 5]. In [3], the action was denoted by $\mathcal{E} \equiv -Q_+$ and the quantity $1+s$ was denoted by b. In the following, we focus on $\hat{K}$, which simplifies the analysis.

We consider facilitated models [12] defined by a binary field $n_i = 0, 1$ and a non-interacting Hamiltonian $H = \sum_i n_i$, where $i = 1, \ldots, N$ are the sites of a lattice. The dynamics at site i is subject to a kinetic constraint $C_i(n_j)$ which is a function of the neighbours n_j of i. That is, site i changes its state with a rate proportional to C_i. For example, in the Fredrickson-Andersen (FA) model [8, 12], $C_i = 1$ if any of the nearest neighbours j of i are in the "excited" state, $n_j = 1$; otherwise $C_i = 0$. Sites with $C_i = 1$ make the transitions $0 \to 1$ with rate c and $1 \to 0$ with rate $(1-c)$, where c is the equilibrium concentration of excitations at temperature T, $c \equiv \langle n_i \rangle = (1 + e^{1/T})^{-1}$. One can also consider exchange dynamics $1_i 0_j \leftrightarrow 0_i 1_j$ between nearest neighbour sites i and j, so that the total density of excited sites is conserved. The result is a constrained lattice gas [10, 11, 12]. For example, the two vacancy facilitated lattice gas, or (2)-TLG [11], is defined on a triangular lattice, with exchange dynamics and a constraint C_{ij} which vanishes unless the two common nearest neighbours of sites i and j are vacant.

Dynamic heterogeneity in KCMs is a consequence of space-time correlations in the dynamics. Within the glassy regime, trajectories contain space-time regions with relatively fast dynamics, and others which rearrange slowly [7]. Slow regions can be non-finite in extent, that is, KCMs contain states for which the number of sites with $C_i \neq 0$ is subextensive in the system size N. These configurations are important for our purposes. Transitions out of these states occur with a rate that is subextensive in N. For systems obeying detailed balance, we show below that if any such states exist the partition sum (1) is dominated by trajectories localised in them, at large N and t and for all positive s. The order parameter $\hat{K}$ is subextensive for these trajectories, so that $K(s)/N$ vanishes for $s > 0$ in the large N limit. Conversely, for $s < 0$, the dominant trajectories visit states in which a finite density of sites have $C_i \neq 0$, and $K(s)/N$ is finite. These arguments establish the existence of the transition shown in Fig. 1.

For a more quantitative analysis, we identify $\psi_K(s)$ as the largest eigenvalue of a time evolution operator [5, 15]. The dynamics of the stochastic model are specified by a master equation

$$\partial_t P(\mathcal{C},t) = -r(\mathcal{C})P(\mathcal{C},t) + \sum_{\mathcal{C}'} W(\mathcal{C}' \to \mathcal{C})P(\mathcal{C}',t), \tag{4}$$

where $\mathcal{C}$ denotes a configuration of the system, the

$W(\mathcal{C}' \to \mathcal{C})$ are the transition probabilities between configurations, and $r(\mathcal{C}) = \sum_{\mathcal{C}'} W(\mathcal{C} \to \mathcal{C}')$ is the rate of escape from $\mathcal{C}$. We write this equation in an operator form $\partial_t P = \mathbb{W} P$, where the matrix elements of $\mathbb{W}$ are $\mathbb{W}(\mathcal{C}, \mathcal{C}') = W(\mathcal{C}' \to \mathcal{C}) - r(\mathcal{C})\delta_{\mathcal{C},\mathcal{C}'}$.

Now, consider the probability $P(\mathcal{C}, t, K)$ that the system is in configuration $\mathcal{C}$ at time t, having made K transitions since time 0. We define $\tilde{P}(\mathcal{C}, t, s) \equiv \sum_K e^{-sK} P(\mathcal{C}, t, K)$, so that $Z(s,t) = \sum_{\mathcal{C}} \tilde{P}(\mathcal{C}, t, s)$. The time evolution of $\tilde{P}$ obeys a Master equation $\partial_t \tilde{P} = \mathbb{W}_K \tilde{P}$, where [5, 15]

$$\mathbb{W}_K(\mathcal{C}, \mathcal{C}') = e^{-s} W(\mathcal{C}' \to \mathcal{C}) - r(\mathcal{C})\delta_{\mathcal{C},\mathcal{C}'}. \tag{5}$$

The calculation of $\psi_K(s)$ reduces to finding the largest eigenvalue of $\mathbb{W}_K$.

The FA model dynamics can be described using the Doi-Peliti [16] occupation number formalism. The operator (5) then reads [17, 18]:

$$\mathbb{W}_K^{(\mathrm{FA})} = \sum_{\langle i,j \rangle} \hat{n}_i \left[c \left(e^{-s} a_j^\dagger - 1 \right) + \left(e^{-s} - a_j^\dagger \right) a_j \right]. \tag{6}$$

where $a_i^\dagger$ and a_i are bosonic creation and annihilation operators, $\hat{n}_i = a_i^\dagger a_i$ and $\langle i, j \rangle$ indicates that the sum is over pairs of nearest neighbours. For simplicity we have allowed for multiple occupancy per site: changing the dynamics in this way has little effect at low temperatures [17]. The single occupancy case can be formulated in a similar manner [18], and the results are analogous.

We begin with a simple mean-field treatment of $\mathbb{W}_K^{(\mathrm{FA})}$. We discard fluctuations and define $W_K^{(\mathrm{FA})}(\phi, \overline{\phi})$ by making the replacements $a_i \to \langle a_i \rangle \equiv \phi$ and $a_i^\dagger \to \langle a_i^\dagger \rangle \equiv \overline{\phi}$ in the operator $\mathbb{W}_K^{(\mathrm{FA})}$. Then we can estimate the largest eigenvalue of $\mathbb{W}_K^{(\mathrm{FA})}$ by finding the stationary points of $W_K^{(\mathrm{FA})}(\phi, \overline{\phi})$. The Euler-Lagrange equations are

$$0 = 2\overline{\phi}\phi \left(\phi - e^{-s} c \right) + \phi \left(c - e^{-s} \phi \right), \tag{7}$$

$$0 = 2 \left(\overline{\phi} - e^{-s} \right) \overline{\phi}\phi + c\overline{\phi} \left(1 - e^{-s} \overline{\phi} \right). \tag{8}$$

There are two solutions that correspond to steady states of the dynamics: (i) $\phi = c\overline{\phi} = c\overline{\phi}_*$ with $\overline{\phi}_* = \frac{3}{4} e^{-s} + \frac{1}{4}\sqrt{9e^{-2s} - 8}$; and (ii) $\overline{\phi} = \phi = 0$. For solution (i), $W_K^{(\mathrm{FA})}(\phi, \overline{\phi})$ is positive for $s < 0$ and negative otherwise; for solution (ii), $W_K^{(\mathrm{FA})}(\phi, \overline{\phi}) = 0$. Our estimate for the large deviation function is given by the larger value of $W_K^{(\mathrm{FA})}(\phi, \overline{\phi})$, so that

$$\psi_K^{(\mathrm{m.f.})}(s) = \begin{cases} Nd \left(c\overline{\phi}_* \right)^2 \left(e^{-s} \overline{\phi}_* - 1 \right) & (s < 0) \\ 0 & (s > 0) \end{cases} \tag{9}$$

where d is the spatial dimension, and $\overline{\phi}_*$ is defined above. Figure 1(top) shows $\psi_K^{(\mathrm{m.f.})}(s)$ and $K(s)$. The active phase is stable for $s < 0$; the inactive one is stable for $s > 0$. The large deviation function $\psi_K^{(\mathrm{m.f.})}(s)$ is continuous at $s = 0$, but its derivative $K(s)$ displays a first-order jump.

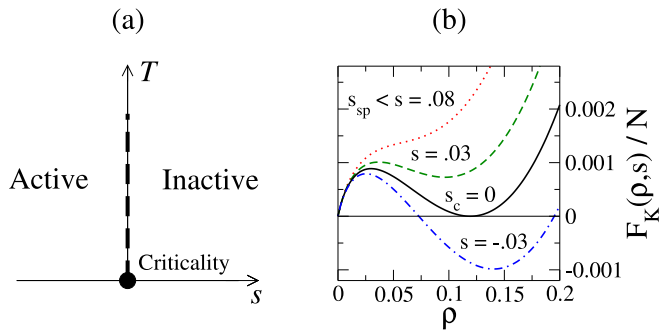

FIG. 3: (a) Space-time phase diagram for facilitated models. $s = 0$ is a first-order transition line between the active phase ($s < 0$) and the inactive phase ($s > 0$). It terminates in a critical point at $T = 0$. (b) Mean-field variational free-energy $\mathcal{F}_K(\rho, s)$, Eq. (11), for the FA model at $T = 0.5$, for several values of s. For $s < 0$ the active phase is dominant and the absolute minimum of $\mathcal{F}_K$ is at $\rho \neq 0$. At $s = 0$ there is dynamic phase-coexistence. For $s > 0$ the inactive phase dominates and $\rho = 0$ minimizes the free-energy. The active minimum no longer exists beyond the spinodal s_{sp}.

The mean-field approximation is clearly very crude. It can be improved systematically with loop corrections, but the coexistence scenario does not change qualitatively: it is a consequence of the kinetic constraints and the existence of states with subextensive escape rates. We now confirm this by means of numerical simulations.

The time evolution associated with the operator $\mathbb{W}_K$ can be obtained dynamically. This operator does not conserve probability [see Eq. (5)] but there are methods to simulate the evolution that it represents [19]. In our case we apply this scheme, analogous to the quantum diffusion Monte Carlo procedure, to continuous time Monte Carlo dynamics [20]. In Fig. 1 we show a plot of $\rho_K(s)$ for the FA model in one dimension. In Fig. 2 we present a finite size scaling analysis of both ψ_K and a dynamical order parameter for the FA model in one dimension, the (2)-TLG model in two dimensions, and the East model in three dimensions (see e.g. [21]). As we approach $N \to \infty$ the crossover between active and inactive phases becomes sharp. Figure 2 shows results for three different models, in different dimensions and with different dynamical constraints; the TLG has a conserved density while the FA and East models do not. Our similar results for these different models demonstrate that space-time phase coexistence occurs quite generally in KCMs.

We have demonstrated that, for any temperature T, the dynamics of KCMs such as the FA or East models take place at dynamical phase coexistence between active and inactive phases. We can construct a phase diagram in terms of T and s, see Fig. 3(a). The $s = 0$ axis is a first-order transition line. It ends in a critical point at $T = 0$. For constrained lattice gases such as the Kob-Andersen model and the (2)-TLG, the phase diagram is similar, with T replaced by density of particles c. In that case the critical point is at the maximum density $c = 1$.

We emphasize that while a zero temperature dynamical critical point is common to many KCMs, this is not a sufficient condition for space-time phase coexistence. For example, consider the pair appearence and annihilation (AA) model of [18], which has the same critical properties as the FA model. Its dynamical rules are $\emptyset A \rightleftharpoons A\emptyset$, $AA \to \emptyset$, $\emptyset\emptyset \to AA$, with rates D, λ and γ, respectively. All states in this model have extensive escape rates, so we do not expect any transition at $s = 0$. Deriving the Euler-Lagrange equations analogous to (7,8), leads to a large deviation function that is analytic at $s = 0$. We have also calculated $\psi_K(s)$ exactly [22] for the AA model in $d = 1$, at the free fermion point $\lambda + \gamma = 2D$ [23]: the large deviation function $\psi_K(s)$ is indeed analytic at $s = 0$ [22].

Finally, in models that obey detailed balance with respect to a probability distribution $p_{\mathcal{C}}$, such as the ones considered here, $\psi_K(s)$ can be calculated through a variational method. The master operator Eq. (5) is made symmetric by a similarity transformation, $\mathbb{H}_K(\mathcal{C},\mathcal{C}') \equiv p_{\mathcal{C}}^{-1/2}\mathbb{W}_K(\mathcal{C},\mathcal{C}')p_{\mathcal{C}'}^{1/2} = e^{-s}\sqrt{W(\mathcal{C}'\to\mathcal{C})W(\mathcal{C}\to\mathcal{C}')} - r(\mathcal{C})\delta_{\mathcal{C},\mathcal{C}'}$. Since $\mathbb{H}_K$ is symmetric and has the same eigenvalues as $\mathbb{W}_K$, we can apply a variational principle:

$$\psi_K(s) = \max_{V_{\mathcal{C}}} \frac{\sum_{\mathcal{C}} V_{\mathcal{C}}\mathbb{H}_K(\mathcal{C},\mathcal{C}')V_{\mathcal{C}'}}{\sum_{\mathcal{C}} V_{\mathcal{C}}V_{\mathcal{C}}}. \qquad (10)$$

Using a trial distribution in which only one of the $V_{\mathcal{C}}$ is finite shows that the largest diagonal element of $\mathbb{H}$ is a lower bound on $\psi_K(s)$, so that $\psi_K(s) \geq -\min_{\mathcal{C}} r(\mathcal{C})$. Hence, if there exists a state for which the escape rate $r(\mathcal{C})$ is subextensive, then $\psi_K(s)/N \geq 0$ at large N. Further, $\psi_K(s)$ is non-increasing [since $\hat{K}$ is non-negative] and $\psi_K(0) = 0$, so the existence of a subextensive escape rate establishes immediately that $\psi_K(s)/N = K(s)/N = 0$ for all $s > 0$ in the large N limit, as we asserted above.

As an example of how to obtain $\psi_K(s)$ from Eq. (10) consider the FA model in a mean-field geometry, such as the complete graph. The transition rates in this case are $W(n \to n+1) = cn$, $W(n+1 \to n) = n(n+1)/N$, where $n = \sum_i n_i$ is the total number of excitations. For the variational state we assume $V_n = e^{Nf(n/N)}$, for some function $f(\rho)$ of the excitation density $\rho \equiv n/N$. In the limit of large N, the leading contribution to (10) comes from values of ρ that maximize $f(\rho)$, and Eq. (10) reduces to $\psi_K(s) = -\min_\rho \mathcal{F}_K(\rho, s)$, where

$$\mathcal{F}_K(\rho, s) = N\rho\left(\rho - 2e^{-s}\sqrt{c\rho} + c\right). \qquad (11)$$

$\mathcal{F}_K$ is a Landau free-energy function for the order parameter ρ. The minimum of $\mathcal{F}_K$ occurs at $\rho_* = c(\overline{\phi}_*)^2$, and from Eq. (10) the result of Eq. (9) is recovered. As shown in Fig. 3(b) the function $\mathcal{F}_K(\rho, s)$ behaves in the characteristic way associated with a first-order phase-transition.

We have shown that the dynamics of KCMs is characterised by the coexistence in space-time of active and inactive dynamical phases. In our view, this dynamical phase coexistence underlies the heterogeneous particle dynamics observed in glass formers. Thus, experimentally obervable phenomena such as transport decoupling [6] arise from the fluctuations associated with this dynamic phase equilibrium. The extension [3] of our results to atomistic models will clarify the degree to which the theoretical framework described here captures the generic features of glassy dynamics.

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