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Jamming in a model glass: interplay of dynamics and thermodynamics

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Abstract

The relationship between extended structures, glassy dynamics and the presence of an underlying thermodynamic transition is explored in the context of lattice models of loops. We review the physical picture which emerges from numerical studies of loop models. It shows dynamical barriers and critical fluctuations combining to produce exponentially slow relaxations close to the critical point. The picture is further sharpened by comparing two loop models with equal dynamical barriers but different equilibrium fluctuations.

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Most liquids, when supercooled, freeze into a glass. The approach to the glass transition is marked by a precipitous slowing down of the dynamics with no obvious structural changes. Experimental evidence is mounting that extended spatial structures play a significant role in the slow dynamical response [1]. The presence of dynamical heterogeneities, clusters of fast moving particles, near the glass transition has been dramatically demonstrated in experiments on colloids [2] and simulations of Lennard–Jones liquids [3]. This raises the intriguing possibility that extended structures are the key to understanding the nature of the glass transition.

A long-standing issue in the context of supercooled liquids is whether the slow dynamical response associated with the freezing into a glass is related to an underlying thermodynamic transition? Then the discovery of dynamical heterogeneities begs the question: Is the slow response of glasses associated with ordering of these spatially

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extended structures? To address this question, in a recent set of publications [4], we have analyzed a non-disordered, frustrated lattice model of loops where we explicitly establish the existence of a critical point with diverging barriers. The physical picture that emerges is one of jamming of extended structures coupled to their critical fluctuations leading to activated dynamics. Below we review the salient features of this scenario and further substantiate it by performing simulations on a related model. This new model is chosen so as to accentuate the interplay of dynamics and thermodynamics.

1. Loop glass models

We define two loop glass models based on the three-coloring model on a two-dimensional hexagonal lattice introduced by Baxter [5]. The bonds are colored either A , B or C with a constraint which forbids bonds of the same color meeting at a vertex. The number of configurations which satisfy the constraint is exponential in the number of lattice sites; in other words the model has an extensive entropy. The coloring constraint gives rise to naturally occurring extended structures in the system: non-intersecting loops formed by pairs of colors.

Within the ensemble of three-colorings, it is possible to induce a phase transition by introducing either (a) an *interaction* which favors ordered arrangements of one of the colors (LI model), or (b) a *field* which orients one of the colors in a particular direction (LF model). The model that we have studied previously is the LI model [4].

The phase transitions in these loop models are best described in terms of the height representation. A two-component height function can be defined for the three-coloring model on the vertices of the dual triangular lattice whereby loops of alternating colors play the role of contour lines. Details of the height map can be found in Ref. [6]. The interaction and field terms couple only to one component of the height ($h_1(x, y)$) through the global tilt, ρ , of the surface [4].

In the LI model, we introduce a long-range interaction energy between the C colors, with a parameter μ measuring the strength of this interaction. In the height language, this translates to an energy function which is *quadratic* in ρ and is minimized by the maximal tilt value $\rho = L$; L is the linear system size. Tuning the energy coupling in LI induces a tilting transition of the h_1 surface [4]. The transition occurs at a finite value of $\mu = \mu_*$, where the curvature of the free energy vanishes signaling a critical point. The average tilt remains zero until this critical point and then jumps to its maximal value at μ_* . By constructing a master equation for the tilt [4], the dynamics of the transition was shown to be dominated by barriers leading to a Vogel–Fulcher type divergence of relaxation times.

In the LF model, we introduce a field term which couples *linearly* to the tilt and also favors a maximally tilted surface. The LF model is the 3—*color* version of the dimer model introduced by Kasteleyn [7] which undergoes a continuous tilting transition of the Pokrovsky–Talapov (PT) kind [8]. The tilting transition in the LF model is expected to be in the same class as the PT transition. This can be argued on the basis of the energies being identical and the entropies of the two models being qualitatively similar.

Both entropies show a maximum at zero tilt [5] and go to zero with a finite slope at the maximum tilt.

2. Results for the LI model

The color constraint imposes severe restrictions on the excitations of the model and, therefore, on the choice of dynamics, since no local color updates are possible. To investigate the transition from the rough untilted surface to the flat tilted state, we choose, as before [4], a dynamics involving loops of all sizes.¹ In one Monte Carlo step we randomly choose a site and either an A - B , B - C or a C - A loop passing through it. We then switch the colors along the chosen loop, using the usual Metropolis rules. For example, a $\dots CACA \dots$ loop becomes an $\dots ACAC \dots$ loop, with probability 1 if the energy change is non-positive, and with rate $\exp(-\Delta E)$ when ΔE is positive.

A first glimpse of the unusual nature of the dynamics can be caught in the time history of the tilt for $\mu < \mu_*$, shown in Fig. 1. The initial coloring state corresponds to a perfectly flat surface with no tilt. As the state evolves via loop updates the average tilt is seen to remain at zero. This is observed for all $\mu < \mu_*$. As μ is increased, the fluctuations about the zero tilted state increase. Concurrently with this, the time histories show the system being trapped in higher tilted states for increasingly longer times.

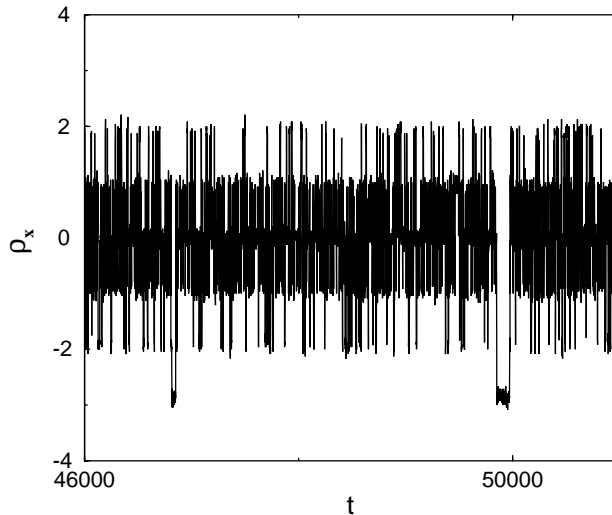


Fig. 1. Monte Carlo time history of the x -component of the tilt at $\mu = 0.275$.

¹ Unlike our previous work here we show results for simulations in which we only update loops with windings equal to $(0,0)$, $(1,0)$ or $(0,1)$.

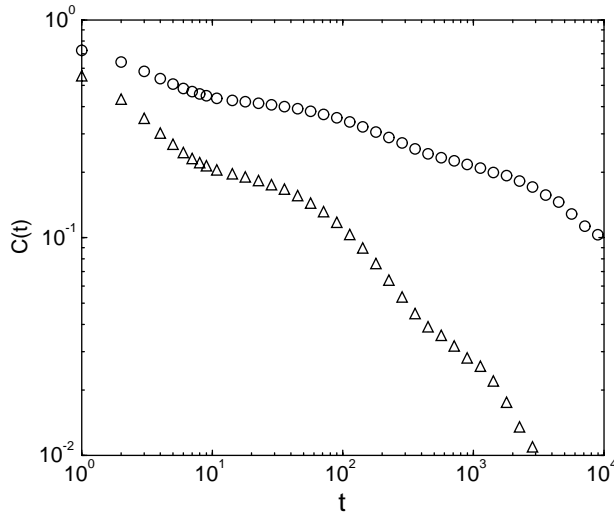


Fig. 2. Log-log plot of the tilt-tilt autocorrelation function at $\mu = 0.265$ (Δ) and 0.275 (\circ) for $L = 24$.

These features can be quantified by studying the equilibrium tilt-tilt autocorrelation function, $C(t)$, which measures the correlation between tilt fluctuations separated by a time interval t [4].

It is clear from the curves shown in Fig. 2, that the relaxation time increases sharply as μ is increased. To characterize this increase of timescales we adopt an operational definition of the relaxation time τ via $C(\tau) = C_0$, where C_0 is an arbitrary small constant. This definition of τ was used earlier in Refs. [9,10]. The timescale τ grows exponentially as the critical point μ_* is approached, and can be fitted well to the Vogel-Fulcher form, $\exp(A/(\mu_* - \mu))$. Furthermore, the decay of $C(t)$ shows a multiple-plateau structure modulating a slow decay; see Fig. 2. This we associate with a broad distribution of relaxation timescales τ_ρ associated with different tilts ρ . The connection between the two can be established by a saddle point analysis [4].

The origin of the well-separated timescales of relaxation τ_ρ can be traced to the dynamical barriers associated with packing of system spanning loops into an ordered structure. A tilted state ρ encounters an energy barrier to go to $(\rho - 1)$ while spatial constraints block transitions to $(\rho + 1)$; both these barriers grow exponentially with ρ leading to an exponential growth of τ_ρ . Finally, this exponential growth combines with the divergence of the tilt fluctuations at $\mu = \mu_*$ giving rise to a Vogel-Fulcher divergence of the relaxation time τ .

Our study of the LI model reinforces the idea that glassy dynamics can arise near a critical point due to dynamical barriers combined with critical fluctuations. To explore this idea further, we next discuss our results for the LF model, which has the same dynamical barriers, while the thermodynamic transition is in a different universality class.

3. Results for the LF model

Simulations were performed for the LF model using the same Metropolis dynamics. The time history of the tilt, shown in Fig. 3, clearly shows a non-zero value of the average tilt for finite μ . This feature is consistent with the PT transition and is in sharp contrast to the behavior of the LI model. The temporal fluctuations about the average also show different characteristics and can again be quantified by measuring $C(t)$.

The correlation functions obtained from the LF model are shown in Fig. 4. The relaxation is exponential with a timescale which increases exponentially with μ , but does not seem to diverge at a finite value of μ .

The observation of an exponential decay of $C(t)$ indicates that the asymptotic decay is dominated by a single timescale. A detailed study [11] of the different tilt relaxation timescales τ_ρ show, that $\tau_{\langle\rho\rangle}$ ($\langle\rho\rangle$ being the equilibrium tilt at a given μ) dominates over the rest. This is very different from the LI model. The origin of the differences in the dynamical behavior can be traced back to the behavior of tilt and its fluctuations near the critical point [11]. In the LF model, the tilt changes continuously as a function of μ in contrast to the LI model where the tilt stays pinned at zero and then changes discontinuously to unity at the critical point. This behavior, in conjunction with the dynamical barriers, leads to a single timescale dominating the dynamics of the LF model and multiple timescales contributing to the LI model near the critical point [11].

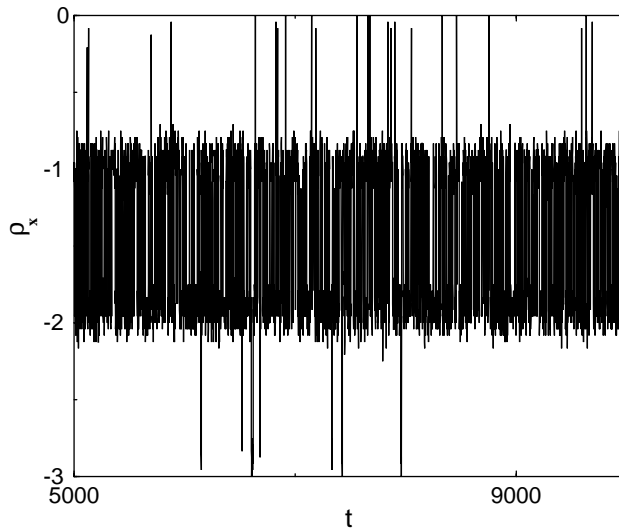


Fig. 3. Monte Carlo time histories of the tilt for the LF model at $\mu = 0.3$. Here $\langle\rho\rangle = -1.49$.

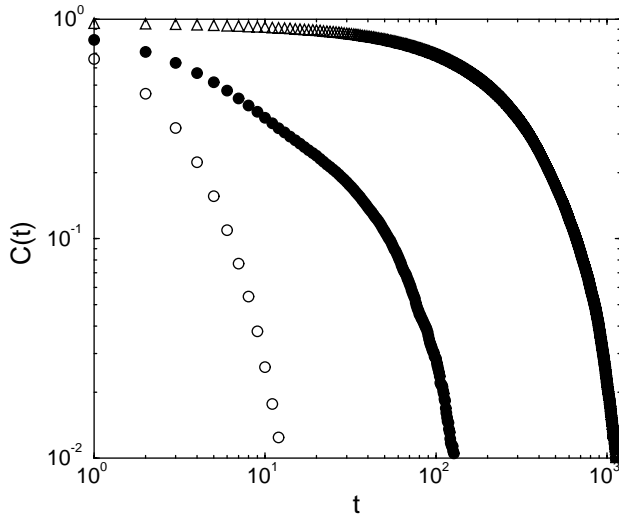


Fig. 4. Log-log plot of the tilt-tilt autocorrelation function for $\mu = 0.3$ (\circ), 0.4 (\bullet), and 0.5 (\triangle) for $L = 24$.

4. Conclusions

In summary, we have demonstrated that an interplay of dynamics and thermodynamics in loop glass models lead to slow dynamics. The jamming of extended structures leads to diverging barriers and a hierarchy of timescales in these models. Thermodynamics plays an important role as well. The LI and LF models have very different equilibrium phase transitions and this is seen to influence the dynamics. While in the LI model the decay of the autocorrelation function is slower than exponential, in the LF model the decay is exponential. Currently we are investigating in detail the critical dynamics of the LF model by constructing a master equation for the tilt analogous to our work on the LI model. Such an effective theory can be used to explore the dynamics close to the critical point where it is difficult to perform simulations.

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