The nature of slow dynamics in a minimal model of frustration-limited domains

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We present simulation results for the dynamics of a schematic model based on the frustration-limited domain picture of glass-forming liquids. These results are compared with approximate theoretical predictions analogous to those commonly used for supercooled liquid dynamics. Although model relaxation times increase by several orders of magnitude in a non-Arrhenius manner as a microphase separation transition is approached, the slow relaxation is in many ways dissimilar to that of a liquid. In particular, structural relaxation is nearly exponential in time at each wave vector, indicating that the mode coupling effects dominating liquid relaxation are comparatively weak within this model. Relaxation properties of the model are instead well reproduced by the simplest dynamical extension of a static Hartree approximation. This approach is qualitatively accurate even for temperatures at which the mode coupling approximation predicts loss of ergodicity. These results suggest that the thermodynamically disordered phase of such a minimal model poorly caricatures the slow dynamics of a liquid near its glass transition.

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I. INTRODUCTION

Several microscopic scenarios have been proposed as underlying mechanisms for dynamical arrest in supercooled liquids \[1, 2, 3, 4, 5\]. Because this dramatic slowing down is accompanied by the onset of dynamical heterogeneity \[6, 7, 8\], a promising candidate model should account for the spontaneous segregation of rapidly relaxing and slowly relaxing domains. The theory of frustration-limited domains has been developed with this condition in mind \[9, 10\]. The basic units of this theory are microscopic regions of low internal energy whose spatial extent is limited by long-ranged interactions or constraints. Nelson and coworkers suggested that such domains form in simple “atomic” liquids (such as metallic glass-formers) \[11\]. For small clusters of particles, icosahedral arrangements are energetically preferred over closest-packed configurations representative of the crystalline state \[12\]. It is thus argued that a supercooled atomic liquid is rich in low-energy icosahedral clusters which, for geometric reasons, cannot extend indefinitely. In this case, frustration is a consequence of the vanishing curvature of Euclidean space. One can imagine that the nature of local order and source of frustration are somewhat different for other, more complicated materials.

The frustration-limited domain theory has several attractive features. By associating relaxation kinetics with the interfacial area of domain walls, it predicts a crossover in the temperature dependence of structural rearrangement times from Arrhenius to a super-Arrhenius form. Such a crossover is prominent in experiments with fragile glass-formers \[13\]. Further, the theoretical scaling exponent for the asymptotic super-Arrhenius temperature dependence appears to be consistent with experimental data. Finally, the notion of frustration-limited domains resonates with the picture of heterogeneous dynamics that has emerged from experiments \[6, 7, 8\] and simulations \[14, 15, 16\]. While local structure within low-energy clusters may be effectively frozen on a molecular time scale, relaxation can be facile at the strained interfaces between domains. This argument is the essence of Stillinger’s “tear and repair” picture of shear flow in fragile liquids \[17\].

Although these dynamical predictions of frustration-limited domain theory are suggestive, they are fundamentally thermodynamic in nature and thus indirect. In order to make such arguments precise, Grousson et al. have recently focused on the explicit dynamics of minimal models exhibiting frustration-limited domains \[18\]. Specifically, they have simulated stochastic dynamics of several classical spin models that pit short-ranged, ferromagnetic interactions against long-ranged, antiferromagnetic interactions. In addition to confirming super-Arrhenius relaxation for these models at low temperatures, they have demonstrated that the “fragility” of the dynamics (i.e., the degree of deviation from Arrhenius form) varies continuously with the relative strength of the long-ranged frustration. Quite recently, Grousson et al. have performed dynamical mode coupling calculations for the same models, but no direct comparisons to their earlier simulations were made \[19\]. In this paper, we perform similar calculations that are compared directly to numerical simulations.

Schmalian and coworkers have also argued that such a Coulomb-frustrated ferromagnet should display essential features of glassy dynamics \[20, 21, 22\]. In their analysis, it is a proliferation of metastable states that drives vitrification. With the aid of replica mean field theory, this perspective predicts a scaling of fragility with frustration strength that agrees well with the simulation results of Ref. \[18\]. But like the theory of frustration-limited domains, this analysis is thermodynamic in nature, re-
lying on an assumed correspondence between particular subensembles of high free energy and genuine dynamical bottlenecks.

This paper addresses the extent to which the slow relaxation in such simplified models truly resembles that of molecular glass-forming liquids. For this purpose we investigate in detail the dynamics of a model closely related to those studied in Refs. [18] and [20]. The model and dynamical propagation rules we consider, which are free of artificially quenched disorder and kinetic constraints, are described in Sec. 2. For several values of the frustration strength, we compare the time dependence of spin correlations computed in simulations with those predicted by approximate theoretical approaches.

Two self-consistent dynamical equations obtained from theory are discussed in Sec. 3. They correspond to resummations of an exact, infinite diagrammatic series for time correlations. The first resummation, yielding exponential relaxation at each wave vector, is a direct dynamical generalization of Brazovskii’s static result for this class of models [23]. Calculations based on this straightforward approach agree remarkably well with simulation results, described in Sec. 4, even for temperatures approaching a thermodynamic transition to a fully ordered state. The second resummation is formally analogous to the idealized mode coupling theory of liquids. As such, it predicts loss of ergodicity at finite temperature. By contrast, we find no evidence of nonergodic behavior or two-step relaxation for simulated disordered states of this model.

Surprisingly, then, the Hartree approach is the more accurate approximation for slowly relaxing disordered states of the model system. Detailed comparison of simulation and theory confirms that sluggishness indeed arises from de Gennes narrowing (i.e., from dramatic changes in static correlations), in contrast to complex dynamical mechanisms such as mode coupling. In this respect, the minimal model we have studied does not capture important aspects of supercooled liquid dynamics, specifically the intermediate time plateau and long time stretching of dynamical correlators. Implications of this result are discussed in Sec. 5, along with issues related to fragility and local conservation of magnetization. In section 6 we conclude.

II. MODEL

We consider fluctuations of a field $\phi(r)$ at position $r$ in three dimensions, with energy [23, 24, 25]

$$\beta \mathcal{H}[\phi(r)] =$$

$$\int dr \left[ \phi(r) \left( \tau + k_0^{-2} (\nabla^2 + k_0^2)^2 \phi(r) + \frac{\lambda}{4!} \phi^4(r) \right) \right]. \quad (1)$$

Here, the energy scale $\beta^{-1}$ characterizes typical fluctuations of a surrounding heat bath. The physical meaning of the field $\phi$ may be somewhat abstract in the context of supercooled liquids, for example representing the degree of a particular local packing symmetry. The application of Eq. (1) to diblock copolymer melts is more intuitive [27, 28, 20]. In this case, $\phi$ represents the local excess number density of one monomer type, and $\tau$ describes the preferential affinity of monomers for others of the same type. Because our interest in this model is motivated by the work in Ref. [18], we will imagine that $\phi$ simply represents a coarse-grained, scalar spin density. Here, $\tau$ is a dimensionless temperature measuring the distance from an underlying critical temperature when $k_0 = 0$.

For any physical interpretation, the wave vector $k_0 \neq 0$ characterizes long-ranged order of a low-temperature, microphase separated state. The coefficient $\lambda$ multiplying the $\phi^4$ term in Eq. (1) will later be used to order terms in perturbation series, although in calculations its numerical value will be of order unity.

For wave vectors near $k_0$, the action in Eq. (1) corresponds to that studied by Schmalian and Wolynes [20, 21, 22] and (in a hard-spin lattice version) by Grousson et al. [18]. In their work, frustration is explicit in competing interactions of square-gradient $(J |\nabla \phi(r)|^2)$ and Coulomb $(Q \phi(r) \phi(r')/|r - r'|)$ forms. The relative strengths of these interactions determine the periodicity of the ground state, $k_0 \propto (Q/J)^{1/4}$, in which spin-up and spin-down domains alternate in stripes or lamellae [26]. In the model defined by Eq. (1) this frustration is instead implicit in nonzero $k_0$, but has the same physical effect. Namely, homogeneous domains are energetically favored at small length scales, while net magnetization is effectively constrained to vanish at larger length scales. In the context of diblock copolymers, this effective constraint reflects the stoichiometry imposed by polymer connectivity [25, 28, 23].

In two dimensions and higher, the presence of nonzero $k_0$ in Eq. (1) has a subtle but profound effect on the thermodynamics of the paramagnetic state. Specifically, the large entropy of fluctuations near $|k| = k_0$ significantly reduces the free energy of the disordered phase. Within a Hartree approximation, this contribution is sufficient to make the paramagnetic state stable or metastable for all finite $\tau$. As a consequence, the transition to a phase with long-ranged order is first (rather than second) order and occurs at a temperature $\tau_{tr} < 0$. This effect was first recognized by Brazovskii [23] and has been summarized lucidly by Binder and Fredrickson [24]. Its qualitative features have been subsequently confirmed in experiments with diblock copolymers [25]. Because, in this picture, statistics of the disordered state are dominated by fluctuations near $k_0$, we expect the model of Eq. (1) to belong to the same universality class as those of Refs. [18] and [20, 21, 22]. Later, we will demonstrate that slow dynamics of this state are dominated by the very same fluctuations.

The action in Eq. (1) is not a true Hamiltonian, and thus has no intrinsic dynamics. We consider two commonly used stochastic propagation rules which generate a canonical ensemble of fluctuations consistent with Eq. (1)

\[\beta \mathcal{H}[\phi(r)] = \int dr \left[ \phi(r) \left( \tau + k_0^{-2} (\nabla^2 + k_0^2)^2 \phi(r) + \frac{\lambda}{4!} \phi^4(r) \right) \right]. \quad (1)\]
Attention will be primarily focused on a simple Langevin equation,
\[
\frac{\partial \phi(\mathbf{r})}{\partial t} = -\frac{\delta \mathcal{H}[\phi]}{\delta \phi(\mathbf{r})} + \eta(\mathbf{r}, t),
\]
where \(\eta(\mathbf{r}, t)\) is a random force whose statistics are Gaussian, and
\[
\langle \eta(\mathbf{r}, t)\eta(\mathbf{r}', t') \rangle = 2\beta^{-1}\delta(\mathbf{r} - \mathbf{r}')\delta(t - t').
\]
In Eq. 3 angled brackets denote an average over all possible realizations of the random force. The above equation of motion, along with the energetics of Eq. 1 and the statistics of Eq. 3, has been studied previously, most notably in the context of nucleation and nonequilibrium pattern formation following a rapid quench to \(\tau < \tau_{\text{tr}}\) [24, 25, 31, 33]. To our knowledge, the detailed equilibrium dynamics of the paramagnetic phase very close to the transition (i.e., \(\tau \gtrsim \tau_{\text{tr}}\)) have not until now been fully explored.

The dynamics generated by Eq. 2 do not conserve the field \(\phi(\mathbf{r}, t)\). Because the slow relaxation of supercooled liquids results in part from the conservation of hydrodynamic densities, this feature of Eq. 2 may be somewhat troubling (particularly in the context of diblock copolymers, in which the number density is clearly conserved). For this reason, we consider a second form of dynamics that conserves \(\phi(\mathbf{r}, t)\) by construction. Trajectories of these dynamics are chains of microstates generated by a Metropolis Monte Carlo algorithm. In detail, a random displacement of the field \(\phi(\mathbf{r})\) is attempted at discrete time steps, and accepted with probability
\[
P_{\text{acc}} = \min\left[1, \exp\left(-\beta \Delta \mathcal{H}\right)\right],
\]
where \(\Delta \mathcal{H}\) is the change in energy produced by the displacement. Local conservation of the field is achieved by restricting the choice of random displacements to those which do not alter the local net magnetization. Further details of this procedure will be described in Sec. 4. Simulation results presented in that section demonstrate that the decay of spin correlations produced by Eqs. 2 and 11 are nearly identical, within an arbitrary rescaling of time in the Monte Carlo chain of states. The physical meaning of this fact will be discussed in Sec. 5.

III. THEORY

In this section we discuss two approximations for relaxation of the field \(\phi(\mathbf{r}, t)\). Specifically, we derive closed equations of motion for the correlation function
\[
C_k(t - t') = \langle \phi_k(t)\phi_{-k}(t') \rangle,
\]
where Fourier components of the field are defined in the standard way:
\[
\phi_k(t) = \int d\mathbf{r} \phi(\mathbf{r}, t)e^{ik\cdot\mathbf{r}}.
\]

The first equation of motion is linear in \(\phi_k(t)\) and resembles phenomenological theories of high temperature liquid state dynamics, such as the wave vector dependent viscoelastic theory [34]. The second is nonlinear and has the form of the idealized mode-coupling approximation to the dynamics of density fluctuations in supercooled liquids [1]. This equation contains the feedback mechanism responsible for the interruption of particle diffusion at intermediate time scales due to constraints imposed by slowly reorganizing local environments (the “cage” effect).

Treating \(\lambda\) as a perturbation parameter, the solution to Eq. 2 may be written as an infinite series of terms, each representing a collection of field interactions and periods of free propagation. As a result, \(C_k(t)\) and its associated response function, \(G_k(t) = -\beta dC_k(t)/dt\) can be expanded in powers of \(\lambda\). (We focus exclusively on the portion of the phase diagram in which dynamics are ergodic, so that \(C_k(t)\) and \(G_k(t)\) are related by the fluctuation-dissipation theorem.) Fig. 1 shows diagrammatic representations of the first two terms in the series for \(C_k(t)\). The development of this expansion, as well as the partial series summations underlying our approximations, have been discussed thoroughly in the context of other models. We will describe physically significant highlights of the procedure and refer the reader to Refs. [12, 35] for details.

A linear equation of motion for \(C_k(t)\) results from summing only terms in the series whose diagrams have the basic topology shown in Fig. 1(a). In the irreducible segments of these “tadpole” diagrams, all interactions coincide in time. Consequently, such a summation renormalizes only the static portion of the basic tadpole diagram of Fig. 1(a). The dynamics predicted by this Hartree resummation scheme are identically those of a variationally optimized harmonic reference system [37]. They are thus obtained more directly by assuming Gaussian statistics for \(\phi_k(t)\). Specifically, we multiply the Fourier transform of Eq. 2 by \(\phi_{-k}(0)\) and average over the noise history,
Eq. 7 is the first member of a complicated hierarchy of equations relating multi-point fluctuations to correlations of higher order. But if we assume that \( \phi_k(t) \) is a Gaussian random variable, the hierarchy closes immediately:

\[
\frac{\partial C^\text{H}_k(t)}{\partial t} = -\mu^\text{H}_k C^\text{H}_k(t).
\]  

(8)

Here, the renormalized mass \( \tau^\text{H} \) that appears in the expression for the structure factor \( \rho^\text{H}_k = 1/C^\text{H}_k(0) = \tau^\text{H} + k_0^{-2}(k^2 - k_0^2)^2 \) is determined self-consistently by

\[
\tau^\text{H} = \tau + \frac{\lambda}{2} \sum_{k'} C^\text{H}_k(0).
\]  

(9)

This result is precisely Brazovskii’s static approximation\[22\]. The relaxation described by Eq. 8 while simply exponential, occurs with rates that are significantly renormalized by the entropy of fluctuations near \( |k| = k_0 \).

More elaborate, nonlinear approximations for \( C_k(t) \) result from incorporating diagrams with more complicated topologies\[23\]. The mode coupling approximation (MCA) is an example, including diagrams with the “sunset” shape of Fig. 14. Summing all terms that renormalize the propagators (but not the vertices) of the basic sunset diagram yields the MCA. Because these contributions involve more than one unique time variable, they are capable in principle of capturing nontrivial memory effects. As shown in Ref.\[35\], the self-consistent result of this resummation is

\[
\frac{\partial C^\text{MCA}_k(t)}{\partial t} = -\mu^\text{MCA}_k C^\text{MCA}_k(t) + \\
\frac{\lambda^2 \beta}{6} \int_0^t dt' \sum_{k',k''} \left[ C^\text{MCA}_k(t-t') C^\text{MCA}_{k'}(t-t') \right] \\
\times C^\text{MCA}_{k-k'-k''}(t-t') \frac{\partial C^\text{MCA}_{k''}}{\partial t'}
\]  

(10)

The final, nonlinear term of Eq. 10 explicitly couples the dynamics of fluctuations at different wave vectors, so that the decay of \( C^\text{MCA}_k(t) \) is not simply exponential. The mode-coupling estimate of the static structure factor, \( C^\text{MCA}_k(0) = 1/\mu^\text{MCA}_k \), is determined by a self-consistent equation involving both the “tadpole” and “sunset” diagrams. We avoid this static calculation by instead replacing \( \mu^\text{MCA}_k \) in Eq. 10 with the exact form of \( C^\text{MCA}_k(0) \) from numerical simulations. This procedure is commonly employed in mode-coupling studies of supercooled liquids.

As the microphase transition point is approached from high temperature, we expect that only modes near the ordering wave vector \( k_0 \) will remain important. In this regime, a reduced model without reference to the coupling of specific length scales should capture the qualitative behavior of Eq. 10. Such a schematic model is similar to that studied by Leutheusser for structural glass-forming liquids\[37\]. (Indeed, Eq. 10 is only slightly different from that encountered in the idealized mode-coupling theory of supercooled liquids\[1\]. In particular, the memory kernel involves a two-point correlation function raised to the third, rather than second, power.) Restricting attention to \( |k| = k_0 \) and neglecting coupling to other wave vectors, Eq. 10 reduces to the dynamical equation exactly describing the p-spin model of a mean field spin glass (with \( p = 4 \)). Since for \( p > 2 \) the critical properties of such models are essentially \( p \)-independent, we expect that near a critical point, Eq. 10 will exhibit a plateau and an eventual transition to nonergodic behavior\[38\].

The character of the slow dynamics resulting from the theories underlying Eq. 8 and Eq. 10 are fundamentally different. The dynamical Hartree theory (Eq. 8) may exhibit a rapid slowing of dynamics as a function of inverse temperature only if the statics, as expressed through the renormalized mass \( \tau^\text{H} \), are strongly temperature dependent. On the other hand, due to the nonlinearity of Eq. 10, a slight change in the structure factor may result in a dramatic change in relaxation times. It is well known that glass-forming liquids show little change in static structure as the glass transition is approached\[39\].

Thus, theories of the type given in Eq. 8 are not relevant near the glass transition. In the following sections, the predictions of Eq. 8 and Eq. 10 will be compared with simulations for the Coulomb-frustrated system.

The renormalized perturbation theories developed in this section are strictly valid only in the limit of weak coupling, i.e., for small \( \lambda \) or large, positive \( \tau \). In our simulation work, we fix \( \lambda = 1 \). It is thus instructive to ask what value of \( \tau \) these theories are expected to break down. To answer this question, we follow the arguments of Hohenberg and Swift\[24\]. Specifically, we compare the Hartree approximation to the renormalized mass, \( \tau^\text{H} \), with corrections introduced by mode coupling (i.e., the renormalized sunset diagram). These corrections are comparatively small when

\[
|\tau| \lesssim 0.2 k_0^{-7/5}.
\]  

(11)

In this regime the Hartree and mode coupling approximations are controlled, and differ only quantitatively from one another. For larger \( |\tau| \), however, the two approximations can differ substantially, as we will see in numerical results presented in the following section. It has been noted previously that the static Hartree approximation can be accurate beyond its strict range of validity. There is thus no guarantee that a range of \( \tau \) exists in which the mode coupling approximation significantly improves upon an appropriately chosen harmonic reference system.
IV. SIMULATIONS

In order to follow the dynamics of Eq. 2 or Eq. 4 numerically, it is necessary first to coarse-grain the field \( \phi(\mathbf{r}, t) \) in space. This procedure yields a (periodically replicated) finite set of dynamical variables, whose time evolution may be integrated approximately over short intervals. We select a coarse-graining length \( a = 2\pi/nk_0 \), and define new fields at lattice points \( \mathbf{r}_i \):

\[
\Phi_i(t) = a^{-3} \int_{v_i} d\mathbf{r} \phi(\mathbf{r}, t),
\]

where \( v_i \) is bounded by a cube of side length \( a \) centered at \( \mathbf{r}_i \). In the calculations described below, \( n = 8 \), so that a domain of wavelength \( \pi/k_0 \) comprises several “soft spins” \( \Phi_i \). To lowest order in \( a \) and a small time increment \( \Delta t \), these renormalized fields evolve according to

\[
\Phi_i(t + \Delta t) = \Phi_i(t) - \Delta t[(\tau + k_0^{-2}(L + k_0)^2)\Phi_i(t) + \frac{\lambda}{3!}\Phi_i^3(t)] + \eta_i(t).
\]

Here, the lattice approximation to the Laplacian operator acting on a function of space, \( \mathcal{L}f(x_i) = a^{-2} \sum_{j \in \text{nn}} f(x_j) - f(x_i) \), is taken to include a sum over nearest neighbors only (denoted nn). After coarse-graining, statistics of the random force remain Gaussian, with \( \langle \tilde{\eta}(\mathbf{r}_i, t_m)\tilde{\eta}(\mathbf{r}_j, t_n) \rangle = 2\beta^{-1}(\Delta t/a^3)\delta_{ij}\delta_{mn} \).

The simulation algorithm described above (which is very similar to those of Refs. 22, 31) has several advantages over the numerical approach of Ref. 18, which employs “hard spins” \( (\Phi_i = \pm 1) \) and explicit frustration. First, spins interact only with nearest and next-nearest neighbors, providing linear scaling of computational effort with system size. Because cumbersome techniques associated with long-ranged forces are not required, a larger set of dynamical variables may be considered. In our calculations, the periodically replicated unit cell includes 643 spins arranged on a cubic lattice. More importantly, the coarse-graining procedure allows the dimensions of the unit cell to scale with the physically relevant length \( k_0^{-1} \). As a result, the unit cell spans several correlation lengths, even for very small values of \( k_0 \). By contrast, in the work of Ref. 18, the unit cell is comparable to a single natural lamellar spacing for several of the simulated states (particularly those corresponding to “fragile” systems). In those cases, significant finite size effects are possible. In effect, Grousson et al. cut off slowly relaxing fluctuations at small \( k \) rather than the rapidly relaxing fluctuations at large \( k \) that are integrated out in our approach. In Sec. 5 we discuss the dynamical implications of such a cutoff.

Using Eq. 13 we have computed the dynamics of several states of the model system at temperatures above the microphase separation transition \( (\tau \gtrsim \tau_c) \). We focus on three values of \( k_0 \) (0.1, 0.5 and 1.0) corresponding to a somewhat broader range of model parameters than was considered in Ref. 18. In each case, \( \lambda = 1 \), so that the microscopic dynamics is in principle strongly nonlinear. Representative configurations of the system are depicted in Fig. 2 typifying the high-temperature paramagnetic phase (a), the disordered phase near the microphase separation transition (b), the ordered lamellar phase (c), and a nonequilibrium state produced by rapid quenching of a disordered system to low temperature (d).

For each value of \( k_0 \) we consider, the relaxation of spin correlations slows dramatically near the transition to microphase separation. The time required for single-spin correlation,

\[
C(t) = \langle \Phi_i(0)\Phi_i(t) \rangle = N^{-1} \int \frac{d\mathbf{k}}{8\pi^3} C_k(t),
\]

with \( C(t) \equiv \langle \Phi_i(0)\Phi_i(t) \rangle = N^{-1} \int \frac{d\mathbf{k}}{8\pi^3} C_k(t) \), to decay to 10% of its initial value, \( \tau \), is plotted as a function of \( \tau \) in Fig. 3. The growth of relaxation times as \( \tau \) approaches \( \tau_c \) is sharpest for the smallest value of \( k_0 \). Indeed, critical fluctuations are suppressed least strongly in this case, as evidenced by the onset of sluggishness near \( \tau = 0 \).

Grousson et al. have likened systems corresponding to large and small values of \( k_0 \) to “strong” and “fragile” glass formers, respectively. For fragile cases, they have even shown that the temperature dependence of \( C(t) \) is well fit by the Vogel-Fulcher form found for supercooled liquids. Although this functional form suggests an emerging importance of activated processes, the dramatic growth of relaxation times is in fact well captured by the harmonic reference system described in Sec. 3. Numerical solutions of Eq. 2 (plotted as solid lines in Fig. 3), corresponding to this Hartree approximation, are especially accurate in the most fragile case \( (k_0 = 0.1) \). Even for the least fragile case \( (k_0 = 1) \), computed rates differ from predicted values by at most a factor of two.
FIG. 3: Time $\bar{t}$ required for spin correlations to decay to 10% of their initial values, as a function of scaled temperature $\tau$. Circles, squares, and diamonds show numerical results for $k_0 = 0.1$, $k_0 = 0.5$, and $k_0 = 1.0$, respectively. Solid lines are predictions of the Hartree approximation described in the text. Arrows indicate temperatures at which this approximation is expected to break down for each value of $k_0$, as estimated using Eq. 11.

Activated barrier crossing is manifestly absent on a harmonic landscape, strongly implying that slow dynamics are driven by static renormalization, rather than by fundamental changes in the structure of trajectory space.[5] The static structure factor, $C_k(0)$, in fact becomes more sharply peaked in a way that mirrors the sudden growth in $\bar{t}$. In other words, the slowing of relaxation appears to be an example of de Gennes narrowing.[34]

The time dependence of spin correlations provides further evidence for this interpretation. Specifically, even when relaxation is very slow, correlations decay nearly exponentially at each wave vector. In Fig. 4, $C_k(t)$ is plotted for many $k$ values for a system very near microphase separation ($k_0 = 0.5$, $\tau = -0.1$). Included wave vectors span a range from the lowest accessible spatial frequency ($k = 2\pi/L$) to several multiples of $k_0$. In no case is relaxation detectably caged or stretched at long times. The single-spin correlation function, $C(t)$, in Eq. 14 is a superposition of all $C_k(t)$, and thus does not decay as a single exponential. At long times, however, relaxation is dominated by the slowest modes, those with wave vectors lying in a spherical shell with $|k| \approx k_0$, and is very nearly exponential. Since static correlations are strongest for these modes, especially near the microphase separation transition, $C(t)$ is nonexponential only over a small range of the total decay.

Remarkably, spin relaxation is essentially identical for a very different choice of microscopic propagation rules which conserve the field $\Phi$. In this Monte Carlo dynamics, described in Sec. 2, each trial move simultaneously displaces $\Phi$ at a randomly chosen site $i$ and at a a site $j$ randomly chosen from the nearest neighbors of $i$. The displacement at $i$, $\Delta \Phi_i$, is exactly compensated by that at $j$, i.e., $\Delta \Phi_j = -\Delta \Phi_i$. In this way, $\Phi$ is conserved at all length scales greater than or equal to the lattice spacing $a$. In general, such a constraint can influence dynamical behavior dramatically. For instance, scaling exponents for unstable domain growth in similar models depend intimately on the conservation of order parameters.[33] The relaxation described above, however, is modified by the constraint only at very short times.

The insensitivity of slow dynamics to field conservation in this model was anticipated by Sachdev, who noted that the corresponding constraint couples strongly only to fluctuations with very small wave vector ($|k| \approx 0$).[10] The long-lived correlations in Fig. 4, however, are governed not by these modes, but instead by fluctuations of finite wave vector ($|k| \approx k_0$), which couple relatively...
Hartree picture. For example, the fragility parameter 

tivated barrier crossing may be well rationalized in the 

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than novel relaxation mechanisms. Indeed, several dy-

divides further evidence that “vitrification” in this model 

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weakly to the constraint. Conservation is thus of only 

modest importance at long times in the disordered phase. 

These facts can lead in principle to dramatic finite size 

effects in numerical simulations. Specifically, the smallest 

periodically replicated unit must accommodate fluctuations 

with wavelengths several times k₀. Otherwise, the 

absence of truly long wavelength fluctuations will generate 

spuriously strong coupling of conservation constraints 

to the slowest accessible modes. As a result, artificial dy-

namical features may appear at long times. For model 

energetics that are similar to but different from Eq. 1, 

stretched exponential relaxation has been computed from 

simulations in which system dimensions are comparable 

to the smallest possible results of numerical simulations: 

τ = −0.14, τ = −0.12, τ = −0.1, τ = −0.08, τ = −0.06, 

τ = −0.04, τ = −0.02, τ = 0, (in order from top to bottom in the plot). In each case, k₀ = 0.5.

defined empirically by

\[
\bar{\tau} \propto \exp \left( \frac{T_K D}{T - T_K} \right)
\]  

appears to scale as \( D \sim Q^{1/3} \) in the simulations of Ref. [18]. Here, \( T_K \) is a fitted Kauzmann temperature at which relaxation times appear to diverge. Schmalian and Wolynes have suggested that this scaling arises from the entropy of activation for structural rearrangement of mesoscopic domains [20, 21, 22]. The Hartree approximation offers a simpler explanation. Fitted relaxation times follow \( \bar{\tau} \sim \tau_H^{-1} \approx \exp(D/\tau) \) rather well. It is easy to show by dimensional scaling that the renormalized mass is a function only of \( \tau/Q^{1/3} \), yielding immediately \( D \sim Q^{1/3} \). This slightly different scaling form fits the results of Grousson et al. equally well [18]. For the simulations of larger systems we have presented, the Hartree prediction appears to be superior.

The dynamical scenario predicted by the simplified mode coupling theory of Sec. 3, on the other hand, is not borne out in our simulations. Most significantly, we observe neither loss of ergodicity nor two-step relaxation over the relevant range of \( \tau > \tau_T \). These failures of the dynamically nonlinear approximation are evident in Fig. 5 comparing the MCA and simulation results for the same system and thermodynamic states considered in Fig. 3. Interestingly, the MCA predicts trapping at values of \( \tau \) for which the Hartree approximation remains reasonable. The infinite series of terms incorporated in the mode coupling approximation thus adds little realism to the lower order description, and eventually leads to incorrectly anomalous behavior. This series of terms must be compensated to a large degree by omitted terms at each order. There have been suggestions that similar cancellation occurs in mode coupling expansions of supercooled liquid dynamics [22]. In that case, however, signatures of idealized mode coupling (i.e., two-step relaxation) survive despite the existence of omitted relaxation channels. In our model, no such signatures are evident.

We emphasize that the thermodynamic states we have simulated lie exclusively in the disordered phase of our model system. We have confirmed this fact by computing the work to reversibly impose long-range order (i.e., nonzero \( \langle|\Psi_k|\rangle \) at \( |k| = k_0 \)). Although the static Hartree approximation suggests that the states of lowest temperature considered for each \( k_0 \) have global free energy minima in ordered configurations, the computed free energy of the paramagnetic state is in fact lower for each case. This quantitative failure of Brazovskii’s approximation is not surprising, as the relevant states lie outside the strict range of validity of the approximation for \( \lambda = 1 \) [23, 24]. Our analysis thus leaves unexplored a narrow window of the disordered state extremely close to \( \tau = \tau_T \). While it is possible that qualitatively new dynamical behavior arises in this region, it cannot account for the dramatic slowing down we have demonstrated at higher temperature, which is driven by extreme struc-
FIG. 6: Comparison of MCA (solid lines) with numerically simulated dynamics (symbols) (a) and MCA (solid lines) with Hartree approximation (dashed lines) (b). Results are shown for thermodynamic states identical to those plotted in Fig. 5. According to Eq. 11, contributions of mode coupling become significant around $\tau \simeq -0.06$ (fifth line from the top). Note that this temperature is very near the critical temperature at which MCA predicts loss of ergodicity.

V. CONCLUSIONS

We have examined in detail a simple model that is closely related to the frustration-limited domain theory of Kivelson, Tarjus and coworkers, and to the uniformly frustrated “stripe-glass” model studied by Schmalian and Wolynes. The disordered phase of this model system indeed displays some hallmarks of molecular glass-forming liquids. But its dynamics differ qualitatively from generic glassy behavior in several respects.

While relaxation times increase dramatically in these models in a non-Arrhenius fashion as temperature is lowered, we find that an optimized harmonic reference system captures the time dependence of fluctuations semi-quantitatively. In contrast to the vitrification of molecular liquids, the onset of this sluggishness is not accompanied by significant power law or stretched exponential decay of correlations in time. Perhaps most importantly, we find that the slow decay of dynamical correlations is driven by significant changes in static structure. An idealized mode-coupling theory captures these changes less accurately than the simpler harmonic approach, predicting caging and eventual trapping at temperatures where the simulated dynamics remain exponential and ergodic. Together, these results strongly suggest that slow dynamics of this model system arise from the same Gaussian fluctuations that drive the microphase separation transition first order. This conclusion differs markedly from those of Kivelson et al., Grousson et al., and of Schmalian and Wolynes, which invoke activated barrier crossing to explain the dramatic temperature dependence of relaxation times. Uniform frustration alone thus appears insufficient to account for the unusual relaxation properties of supercooled liquids. It remains possible, of course, that such frustration plays a key role, but in concert with other important physical mechanisms and constraints.

The simulations described in this paper constitute a thorough study of the thermodynamically disordered phase of a uniformly frustrated model. They raise some interesting questions about such models and leave others unaddressed. In particular, while glassy behavior is not manifest for $\tau > \tau_{tr}$, we can not rule out the existence of caging or trapping extremely close to or below the microphase separation temperature (since relaxation at these temperatures is prohibitively slow). To explore this possibility, we have simulated several initially disordered states with $\tau < \tau_{tr}$. Their nonequilibrium evolution towards long-ranged order exhibits the self-similar aging characteristic of coarsening phenomena, but for numerically accessible time scales does not resemble the relaxation of a supercooled liquid. Nonexponential equilibrium relaxation could yet emerge at temperatures above but very near the order-disorder transition. Eastwood and Wolynes have in fact recently suggested that, for certain spin models, surface tension effects drive the onset of activated dynamics nearly to the ideal glass transition temperature. Although quite interesting, such behavior would itself be uncharacteristic of glass-forming liquids, as it would appear only within an extremely narrow thermodynamic range.
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