Solvable Models of Supercooled Liquids in Three Dimensions

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We introduce a supercooled liquid model and obtain parameter-free quantitative predictions that are in excellent agreement with numerical simulations, notably in the hard low-temperature region characterized by strong deviations from mode-coupling-theory behavior. The model is the Fredrickson-Andersen kinetically constrained model on the three-dimensional M-layer lattice. The agreement has implications beyond the specific model considered because the theory is potentially valid for many more systems, including realistic models and actual supercooled liquids.

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The ubiquity of glass in nature and technology has driven research in this area for decades but there is still no agreement on the basic mechanism by which a supercooled liquid forms a glass [1–3]. An essential difficulty for deciding over competing theories is the fact that often they make only *qualitative* statements. For instance, the classic thermodynamics vs dynamics controversy resolves around the putative divergences of susceptibilities and correlation lengths but, while the debate has produced many conceptual developments [1,2], there are no precise *quantitative* predictions to be matched with experiments and numerical simulations. In other words, models of supercooled liquids in three dimensions, either realistic or on lattice, have been studied previously only by means of numerical simulations. In the following, we present a model in three dimensions in which the gap between theory and experiments can instead be filled. Furthermore, the agreement has implications beyond the specific model considered because the theory is potentially valid for many more physical systems, including the most realistic models and actual supercooled liquids.

Mode-coupling theory (MCT) [3] is widely popular in the experimental literature because it captures many qualitative features of the physics of liquids upon supercooling, notably two-step relaxation and stretched exponential decay. Furthermore, it agrees quantitatively with numerical simulations, although one has to replace the values of some MCT parameters with values extracted from data fits [4–7]. The essential problem is that it predicts a dynamical arrest transition at a temperature where actual systems are still in the liquid phase. In spite of this serious drawback, many believe that the MCT transition is still relevant and determines a crossover from power law to exponential increase of the relaxation time that is widely observed. Further support to this scenario comes from the fact that simple liquid models in the limit of infinite physical dimension d display a sharp transition qualitatively similar to the one of MCT, although MCT itself is quantitatively wrong in this limit [8]. In that case, the sharp transition is clearly a mean-field artifact due to the $d \to \infty$ limit and it should become a crossover as soon as the dimension is finite. Similarly, the success of kinetically constrained models (KCMs) in reproducing the physics of supercooled liquids is often attributed to the presence of an avoided MCT-like transition that becomes sharp when one switches from lattices in finite dimensions to the Bethe lattice where mean-field theory is correct [9–16].

Building on the analogy between MCT and spin-glass models with one step of Parisi's replica-symmetry breaking (1RSB) discovered more that 30 years ago [17], it has been recently proposed that to fix MCT one has to replace it with a set of stochastic dynamical equations called stochastic-beta relaxation (SBR) [18,19]. SBR describes the β regime, i.e., the timescale when dynamic correlations stay close to a plateau value, and it has a simple and intuitive interpretation: it is basically MCT supplemented with random spatial fluctuations of the temperature that are effectively quenched on the β timescale. SBR is promising, as it seems to cure the drawbacks of MCT, displaying, in particular, the crossover and also dynamical heterogeneities, without spoiling its successes, in particular, two-step relaxation and stretched exponential decay [20,21]. SBR is a universal theory potentially valid for many different microscopic models, the specific model determining the values of its few (five) quantitative parameters. In the following, we show that SBR holds for the models we study. To do so, we have

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first computed the five SBR parameters, then solved numerically the dynamical stochastic equations, and finally performed Monte Carlo simulations to be compared with the theoretical predictions. It turned out that SBR provides an accurate quantitative parameter-free description of the dynamics, i.e., the models are solvable beyond mean-field theory.

The model considered is a particular realization of the classic Fredrickson-Andersen (FA) KCM [9,10]. The FA model is made by Ising spins on the sites of a lattice that are independent, the Hamiltonian being $H = \sum_i s_i$, but obey a kinetically constrained dynamics: a spin can flip only if it has at least m of its c nearest neighbors in the excited (up) state. An equilibrium configuration is thus easily generated numerically at time zero and one typically measures the persistence. More precisely, we define the local persistence $\phi_i(t)$ as equal to one if $s_i(t') = -1$ for all $0 \le t' \le t$ and zero otherwise, thus the averaged persistence is the number of *negative* sites that have never flipped at time t divided by the total number of spins [22]. The FA model on the Bethe lattice is known to exhibit a dynamical arrest transition of the MCT type [11–16]: at the critical temperature T_c the persistence remains blocked to a plateau value $\phi_{\rm plat}$ that is approached in a power-law fashion. The FA dynamical transition is intimately related to bootstrap percolation and both T_c and $\phi_{\rm plat}$ can be computed from its solution on the Bethe lattice, as discussed in the Supplemental Material [23]. In particular, for connectivity $c = 4$ and $m = 2$ the average persistence $\phi(t)$ obeys at $T_c = 0.480\,898$

$$\phi(t) - \phi_{\rm plat} \approx \frac{1}{(t/t_0)^a}, \qquad t \gg 1, \tag{1}$$

where $\phi_{\rm plat} = 21/32$. At present, analytic expressions of t_0 and a are not available, but they can be estimated from numerical simulations as $a \approx 0.352$ and $t_0 \approx 2.30$. It is well known that the sharp transition is instead avoided when the FA model is studied on regular lattices in two and three dimensions, but no first-principle theoretical description of the dynamics can be obtained in those cases. As we will show in the following, such a description can instead be obtained on the finite-dimensional lattice we consider here.

We studied the FA model (with $m = 2$) on the (random) lattice in three dimensions yielded by the application of M-layer construction of [28] to the diamond cubic lattice (that has connectivity $c = 4$, see Fig. 1). The M-layer construction can be applied to any lattice: to obtain a random instance, one considers M copies of the original lattice, say the square lattice in $d = 2$ as in the figure, rewires through a random permutation the M links corresponding to a given link on the original ($M = 1$) lattice, and finally repeats the procedure for each link of the original lattice, as shown in Fig. (1). It can be easily seen that short loops in the lattice are rare for large values of M and the lattice is locally treelike. Loops are nevertheless present at

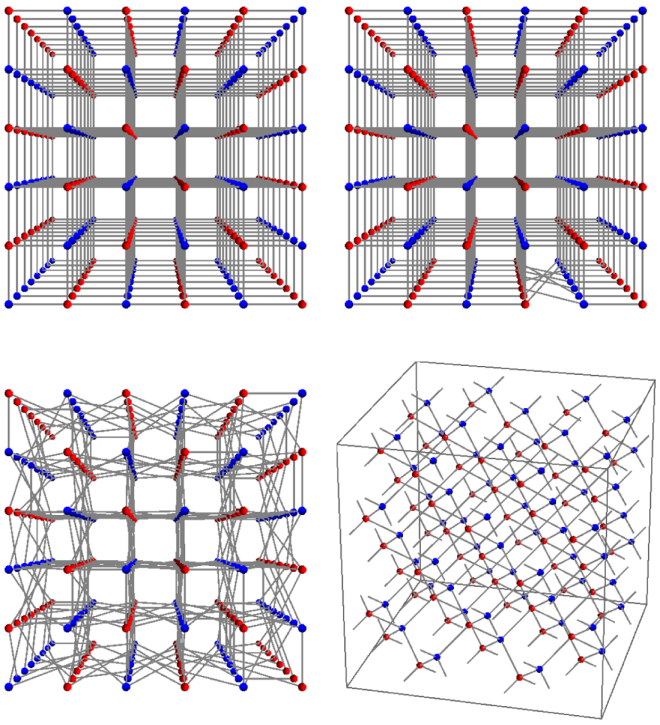

FIG. 1. (Left to right, top to bottom) The M-layer procedure: (1) M-independent copies of a lattice (a regular 2D lattice in the figure) are stacked on top of each other (view from the top); (2) the edges corresponding to a given link in the original lattice are rewired between the M copies; (3) the procedure is repeated for every link of the original lattice. For large M, the graph is locally a Bethe lattice, but at large distances it has the properties of finite-dimensional lattice. (4) The three-dimensional diamond cubic lattice.

large distances, thus at any finite M the lattice is finite-dimensional, although it looks like a Bethe lattice at short distances. Given that for each site $i = 1, \ldots, N$ of the original lattice ($M = 1$) there are M spins s_i^α, $\alpha = 1, \ldots, M$, the total number of sites is $N_{\rm tot} = M \times N$ and the natural local order parameter is the average over the layers of the local persistence minus the plateau value

$$g(x,t) \equiv \left(\frac{1}{M}\sum_{\alpha=1}^{M} \phi_i^\alpha(t)\right) - \phi_{\rm plat}, \tag{2}$$

where x is the spatial coordinate of site i. For M large but finite it is natural to expect that any observable takes the same value it has on the Bethe lattice with small $O(1/M)$ corrections: the model should be solvable by the Bethe approximation. This is indeed true, except at the Bethe lattice critical temperature: while the Bethe approximation predicts that the averaged order parameter $g(t)$ never reaches zero, the three-dimensional nature of the M-layer lattice implies that this cannot be true and leads to a dramatic deviation from mean-field behavior. This is indeed confirmed by numerical data. In Fig. 2 we plot the time decay of the shifted persistence $\langle g(x,t)\rangle$ where

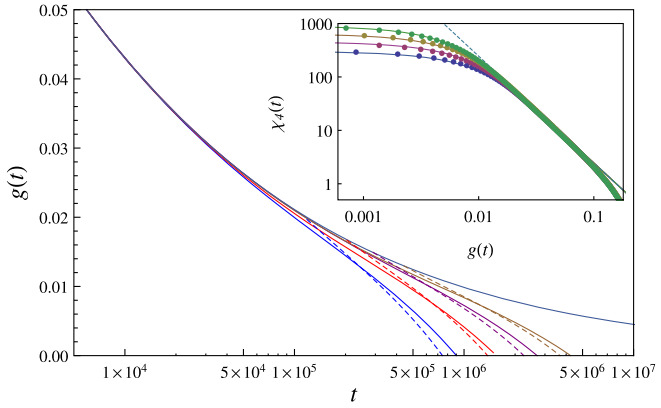

FIG. 2. Persistence vs time in $d = 3$. Solid lines from bottom to top: data for $M = 3000, M = 50\,000, M = 100\,000, M = 200\,000$ and for the Bethe lattice curve ($M = \infty$). The data follow the Bethe curve at initial times and deviate from it at later times increasing with M. The dotted lines represent the corresponding SBR predictions describing the data when they start to deviate from the mean-field curve (see text). (Inset) Parametric plot of $\chi_4(t)$ vs $g(t)$ in $d = 3$ for the above values of M. The SBR predictions (solid lines) are in excellent agreement with the numerical data (points) in the large-time regime where $g(t)$ deviates from MF. At small intermediate times [large $g(t)$] both SBR and the numerical data approach the MF asymptote (dashed straight line), numerical data deviate on smaller (microscopic) times.

here and in the following the angle brackets mean average with respect to (i) different instances of the random lattice generated by the M-layer construction, (ii) different initial equilibrium configurations, and (iii) different thermal histories. Monte Carlo simulations were carried out at the critical temperature of the Bethe lattice $T_c = 0.480\,898$ for systems with $M = 3000$ (with size of the original diamond cubic lattice $L = 8$), $M = 50\,000$, $M = 100\,000$, and $M = 200\,000$ ($L = 4$). At initial times, the data follow the mean-field (MF) curve corresponding to $M = \infty$ (obtained from numerical simulations on the Bethe lattice (see the Supplemental Material [23]) and deviate from it at larger times (increasing with M) reaching the plateau value ($g = 0$) in finite time.

The difficult problem is to obtain theoretical predictions in the region where the data deviate from the mean-field curve and to proceed it is useful to examine the role of fluctuations. Within the M-layer, a mean-field approximation to fluctuations can be obtained as a sum over non-backtracking walks of Bethe lattice fluctuations [28]. At the critical temperature, this yields [23]

$$\langle g(x,t)g(y,t)\rangle - \langle g(x,t)\rangle\langle g(y,t)\rangle \overset{\text{MF}}{\approx} \frac{t^{a(2-d/2)}}{M} f\left(\frac{x-y}{\xi(t)}\right), \tag{3}$$

where $f(x)$ is a scaling function, the correlation length diverges with time as $\xi(t) \propto t^{a/2}$, and d is the space dimension. We can now invoke the Ginzburg criterion and argue that the MF approximation $\langle g(x,t)\rangle \overset{\text{MF}}{\approx} 1/(t/t_0)^a$ is accurate as long as fluctuations around the mean are small. Generically, they are small due to the $1/M$ prefactor, but we see that there is a timescale t_G when, due to the $t^{a(2-d/2)}$ prefactor, they become comparable with the (squared) order parameter $\langle g(x,t)\rangle \overset{\text{MF}}{\approx} 1/(t/t_0)^a$,

$$\frac{t_G^{a(2-d/2)}}{M} \approx \frac{1}{t_G^{2a}} \to t_G \approx M^{\frac{1}{a(4-d/2)}}. \tag{4}$$

Thus, on this timescale MF theory must fail and, most notably, the spurious transition will be avoided, for instance, the dressed propagator on the lhs of (3) will deviate from the bare expression on the rhs and the actual correlation length will cease to grow. We note that the Ginzburg time grows with M and thus deviations from the MF occur at later time for increasing values of M in agreement with the data of Fig. 2. Most importantly, since $t_G \approx M^{\{1/[a(4-d/2)]\}}$ is large for large M, the order parameter is $O(1/t_G^a)$ small and the correlation length is $O(t_G^{a/2})$ large: this grants that deviations from mean-field theory are described by an effective Landau theory, because one can retain only the lowest orders in the Taylor expansion of the order parameter and its space and time derivatives. Following the arguments and computations of [18,19], we argue that the effective theory is SBR, meaning that the generic K-point average obeys for $1 \ll M < \infty$,

$$\langle g(x_1,t_1),\ldots,g(x_K,t_K)\rangle \approx [\hat{g}(x_1,t_1),\ldots,\hat{g}(x_K,t_K)], \tag{5}$$

where $\hat{g}(x,t)$ on the rhs is the solution of the SBR equations

$$\sigma + s(x) = -\alpha\nabla^2\hat{g}(x,t) - \lambda\hat{g}^2(x,t) + \frac{d}{dt}\int_0^t \hat{g}(x,t-s)\hat{g}(x,s)ds. \tag{6}$$

The separation parameter σ measures the distance from the critical point and vanishes at $T = T_c$. The square brackets mean average with respect to the field $s(x)$ that is a quenched random fluctuation of σ, Gaussian and delta-correlated in space,

$$[s(x)] = 0, \qquad [s(x)s(y)] = \Delta\sigma^2\delta(x-y). \tag{7}$$

The SBR equations have to be solved with the small-time condition $\lim_{t\to 0}\hat{g}(x,t)(t/t_0)^a = 1$, where λ and a are related by the MCT relationship $\lambda = \{[\Gamma^2(1-a)]/[\Gamma(1-2a)]\}$. In practice, for times smaller than t_G, the observables on the lhs of Eq. (5) can be accurately approximated with the values they have on the Bethe lattice, while on times of order t_G they are described by the rhs. This explains the peculiar initial conditions of the SBR equations: the short-time behavior on times $O(t_G)$ matches long-time behavior for times $1 \ll t \ll t_G$, i.e., the mean-field result given by Eq. (1).

Equation (5) embodies the power of the effective theory approach: on the lhs, we have a model with a complex microscopic dynamics for which no analytic treatment of dynamics is available (not even on the Bethe lattice); on the rhs, we have a (numerically) solvable set of equations that were derived in [18,19] starting from symmetry considerations (essentially the detailed balance property of the dynamics) but *without* reference to any specific microscopic model. The microscopic details determine the actual values of the five SBR parameters a, t_0, α, $\Delta\sigma$, and σ that are needed to get quantitative predictions. Using recent developments on bootstrap percolation on the Bethe lattice [29] and some lattice-dependent geometrical constants, we obtain (details in the Supplemental Material [23]):

$$\Delta\sigma^2 = \frac{0.285}{M}, \quad \alpha = 0.411, \quad \sigma = 0.222 \times (T_c - T). \tag{8}$$

Within SBR, mean-field theory is recovered setting $\Delta\sigma^2 = 0$, in this case $\hat{g}(x,t)$ is constant in space, the gradient term plays no role, and one recovers the critical MCT equation [3], in particular, for $\sigma \geq 0$ ($T < T_c$) $\phi(t)$ never goes below the plateau value. The M-layer construction allows one to have a finite but small $\Delta\sigma$ so that the MCT transition is avoided and $\phi(t)$ crosses the plateau at a finite time for all values of σ. The SBR predictions corresponding to the data shown in Fig. 2 were obtained solving numerically (by space-time discretization) Eq. (6) for many instances of the $s(x)$ in a box of size L. From the figure, we note that the quality of the SBR predictions increases with M and is excellent for $M = 200\,000$, especially considering that there is no single fitting parameter.

SBR is a powerful theory that provides not only the average dynamical order parameter but, according to Eq. (5), also all possible fluctuations. To demonstrate this, in the inset of Fig. 2, we plot parametrically the $\chi_4(t)$ function that yields the fluctuations of the persistence density

$$g(t) \equiv \frac{1}{N_{\rm tot}} \sum_{i,\alpha} g_i^\alpha(t), \quad \chi_4(t) \equiv N_{\rm tot}(\langle g^2(t)\rangle - \langle g(t)\rangle^2). \tag{9}$$

According to the MF expression (3), $\chi_4(t)$ should diverge with time as t^{2a} [leading to a MF asymptote $\chi_4(g) \propto g^{-2}$], instead on the Ginzburg timescale t_G, over which $g(t)$ deviates from MF and reaches zero, $\chi_4(g)$ deviates from the MF law and remains finite. Note that the agreement between numerical data and the numerical solution of the SBR equations is even better in the parametric representation.

SBR can be applied in other dimensions as well, we have considered (see Supplemental Material [23]) the $d = 0$ case that corresponds to finite-size effects in mean-field models on fully connected or sparse random graphs with 1RSB at the so-called dynamical temperature T_d [30]. The interplay between the parameters M and L can be also be clarified in terms of the SBR equations [23].

To discuss the results in a broader context, we note that the M-layer construction can be applied virtually to all supercooled liquid lattice models, including different KCMs [31] and plaquette models [32,33], leading to analogous solvable non-MF models described by SBR. Furthermore, SBR can provide quantitative theoretical predictions for generic tunable models [34–40] that in earlier studies could only be studied by means of numerical simulations. On the other hand, the lattice for $M \gg 1$ is rather different from the original $M = 1$ lattice one is ideally interested in: the latter has many short loops, while the former has very few. Thus, the condition $M \gg 1$ alters artificially the three-dimensional geometry at the microscopic scale and one may ask if this hampers the applicability of SBR to realistic models and actual supercooled liquids. To clarify this point, we stress that $M \gg 1$ is a sufficient but *not* necessary condition. A necessary condition in a generic system is that the dynamical correlation length is large enough to justify the use of a coarse-grained description: numerical simulations do indeed report correlation lengths significantly larger than the microscopic scale in supercooled liquids [41–44], while unfortunately they cannot be measured in current experimental settings. The natural framework to discuss coarse-grained observables is Wilson's renormalization-group (RG) theory, where each system corresponds to a particular point in the space of RG Hamiltonians that display all possible powers of the order parameter and its spatial and time derivatives and thus depends on a infinite number of coupling constants. In practice, these additional terms lead to higher powers of $\hat{g}(x,t)$ and higher order spatial and time derivatives in Eqs. (6). SBR assumes that these additional terms can be neglected and this can be motivated by the following RG argument. The absence of a sharp dynamical transition in finite dimension implies the absence of a stable fixed point (FP), as a consequence, all Hamiltonians flow under RG toward the high-temperature FP, however, if the correlation length is large it will take many RG steps for it to decrease to one. Since standard dimensional analysis implies that the coupling constants of the additional terms decrease close to the Gaussian fixed point (they are irrelevant operators in RG jargon), it is possible that on the scale of the correlation length the coarse-grained theory is driven near SBR by the RG flow. This explains why many different systems, including experimental ones, are potentially described by SBR and thus share the same qualitative features that do not depend on the actual values of the SBR parameters: notably, power law to exponential increase of the β time and dynamical heterogeneities below the avoided transition.

SBR is thus potentially valid for $M = 1$ as well, because long-range correlations may develop also in the presence of short-range interactions. The deviations at smaller M in the plots are indeed expected because the SBR parameters, being model dependent, change with M. The values we

computed in Eqs. (8) have actually $1/M$ corrections that can be also computed systematically through a feasible but tedious power expansion. The only special feature of the large-M regime is that the SBR parameters can be computed exactly from the Bethe lattice, while the computation in the $M = 1$ case is less straightforward (one should take into account the presence of small loops), but it is still feasible in principle.

While it is satisfying to compute the SBR parameter independently as we have done here, one could also extract some or all of them from fits. This means that SBR can be a useful tool to rationalize experimental data in the region where the widely used ideal MCT scalings fail. The outcome would still be highly nontrivial because the SBR [Eqs. (6)] yields predictions for many more quantities than those needed to determine the SBR parameters through fits. In particular, here we considered only one temperature, but one may consider a whole range of temperatures [20] [corresponding to different values of σ in Eqs. (6)] and also study spatial correlations [20] and finite-size effects.

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