Absence of Marginal Stability in a Structural Glass

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Marginally stable solids have peculiar physical properties that were first analyzed in the context of the jamming transition. We theoretically investigate the existence of marginal stability in a prototypical model for structural glass formers, combining analytical calculations in infinite dimensions to computer simulations in three dimensions. While mean-field theory predicts the existence of a Gardner phase transition towards a marginally stable glass phase at low temperatures, simulations show no hint of diverging time scales or length scales, but reveal instead the presence of sparse localized defects. Our results suggest that the Gardner transition is deeply affected by finite dimensional fluctuations, and raise issues about the relevance of marginal stability in structural glasses far away from jamming.

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Many types of fluids (molecular, colloidal, metallic) transform into amorphous glasses [1,2]. In the glass phase, they present thermodynamic [3], transport [4], vibrational [5], and mechanical [6] properties that are not observed in crystals. These "low-temperature anomalies" are observed in a wide range of systems with very different particle types or interactions, and several theoretical approaches were developed to understand them [4,7–10], making specific assumptions about the nature of the excitations responsible for the anomalies.

A different proposal recently emerged from the convergence of two lines of research, based on the idea that collective excitations associated to marginal stability could be the key concept underlying these properties. First, it was realized that systems close to a jamming transition are marginally stable, in the sense that the number of mechanical interactions in the system is precisely tuned [11]. It was later proposed that marginal stability persists away from jamming [12–14]. Second, an extension of the random first order transition theory [15–17] to amorphous hard spheres in large dimensions was obtained [18]. It predicts a Gardner phase transition [19,20] between a normal glass phase and a marginally stable one characterized by an excess of low-frequency modes [21] and unusual rheological properties [22–24]. The marginal stability of the Gardner phase could provide a universal explanation for glass anomalies [18,25]. Marginal stability implies that the system responds in a strong and system-spanning way to a weak, localized perturbation [12], implying the existence of delocalized soft modes [21], and diverging susceptibilities [22,26].

These recent results provide new opportunities to explain the properties of amorphous materials, motivating ongoing efforts to understand whether marginal stability generally holds in these materials. Hard [27–29] and soft [14] spheres very close to the jamming transition have been analyzed, showing that marginal stability and the Gardner transition may be relevant in colloidal and granular glasses. However, molecular and metallic glasses are usually modeled by longer-ranged, continuous pair interactions for which no jamming transition takes place [1,2]. In this context, much less is known about the role of marginal stability [30], and the existence of a Gardner phase has not been established. Therefore, it is not known whether marginal stability can be used to understand the low-temperature anomalies in generic structural glasses.

To address this important question, we combined theoretical and numerical analysis of the low-temperature vibrational properties of a standard model for atomic glasses. At the mean-field level, a marginally stable Gardner phase is predicted, which is then conceptually unrelated to jamming. However, our numerical simulations of the same model in three dimensions contrast with these predictions. We find no sign of a phase transition within the entire glass phase. Instead, we detect sparse localized defects at low temperature, but they do not give rise to growing time scales and length scales that would accompany the emergence of marginal stability at a Gardner phase transition.

Mean-field theory.—We consider a monodisperse system of d-dimensional particles interacting through a continuous pair potential $v(r) = \epsilon(\sigma/r)^{4d}$. This is the repulsive part of the Lennard-Jones potential, generalized to an arbitrary dimension d. The exponent for the inverse power law is larger than d to ensure that the virial coefficients remain finite in any dimension. We use σ and ϵ as our unit length and energy, respectively. The state of the system is uniquely controlled by $\Gamma = \hat{\varphi}/T^{1/4}$, where T is the temperature and $\hat{\varphi} = \rho V_d 2^d/d$ is the rescaled packing fraction (ρ is the number density, and V_d the volume of a d-dimensional sphere of diameter unity). We fix the

packing fraction $\hat{\varphi} = 1$ and vary the temperature, thus exploring the entire phase diagram.

In the limit $d \to \infty$, the thermodynamic properties of the liquid and glass can be computed exactly [15,18]. In this limit, the system exhibits a sharp dynamical transition of the mode-coupling type [18,31] at a temperature T_d at which the relaxation time of the liquid diverges. Below T_d, the system is trapped in one of the exponentially-many minima of the free-energy landscape. We compute the properties of a typical equilibrium liquid at a temperature $T_g \leq T_d$. As temperature decreases, the glass is confined near the state selected at T_g in a "restricted" equilibrium, and thus follows an equation of state different from the liquid. We compute exactly the free energy of this glass $f_g(T, T_g; \Delta, \Delta_r)$ at the replica symmetric level [32] thanks to a state-following construction [18,32,33]. It depends on two parameters: Δ is the long-time limit of the mean-squared displacement within the followed glass state, and Δ_r is the relative mean-squared displacement between the original equilibrium configuration at T_g and the one followed to T. The free energy f_g is stationary with respect to Δ and Δ_r. The average pressure and energy of glasses are obtained by taking derivatives of f_g with respect to density and temperature, respectively.

We solve the resulting set of coupled integro-differential equations given in Ref. [32] to obtain the phase diagram in Fig. 1(a). First, we compute the potential energy E of the equilibrium liquid and the dynamical transition temperature, $T_d = 0.002\,914$. We then compute the energy of glasses prepared at different $T_g \leq T_d$ as a function of temperature. A Gardner transition is detected when the replica symmetric solution becomes unstable [32], signaling the transformation of the simple glass into a marginally stable one. The low-temperature Gardner phase is described by breaking the replica symmetry [20,34], and the transition belongs to the same universality class of the spin-glass transition in a magnetic field [35,36]. The presence of a Gardner transition is, in general, not a universal result [35,37]. In our model, over a large temperature window $0.5 \lesssim T_g/T_d < 1$, a marginally stable Gardner phase exists, while for $T_g \lesssim 0.5 T_d$ no Gardner transition is found. Because our model does not possess a jamming transition, our results show that mean-field theory predicts that marginal stability is not restricted to the vicinity of jamming, but should be broadly relevant for generic structural glasses with continuous interactions.

Numerical simulations.—There is no clear consensus on the influence of finite dimensional fluctuations on the Gardner transition [36,38–42]. Contradictory results were reported in numerical works. The existence of a transition was suggested in $d = 4$ [39], but opposite claims were also made [38,41]. A renormalization group approach [42] found a fixed point in all dimensions $d \geq 3$, while other works found different results [36,43]. Thus, we must confront our theoretical predictions to a direct numerical

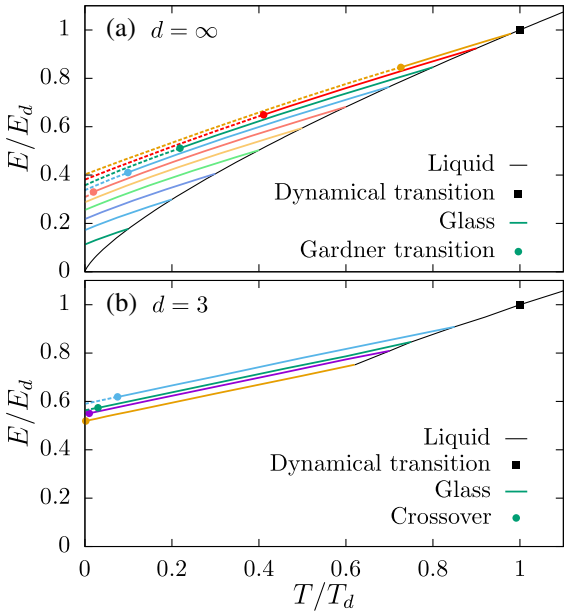

FIG. 1. (a) Mean-field phase diagram. Energy of the equilibrium liquid (black line), and dynamical transition temperature T_d (black square). Various glasses prepared at $T_g < T_d$ are followed out of equilibrium (full lines). When $T_g \gtrsim 0.5 T_d$, these glasses undergo a Gardner transition (bullets) into marginally stable glasses (dashed lines). (b) Numerical phase diagram with equivalent representations of the liquid, dynamic transition and glass lines. Bullets locate the temperature crossover below which localized excitations appear, but do not correspond to a Gardner transition. In both panels, axes are rescaled by the value at T_d: $E_d = E(T_d)$.

investigation of the $3d$ version of the above model. Because the putative transition occurs deep inside the glass phase, it is crucial to prepare well-thermalized glasses, such that the structural relaxation time is larger than the duration of the simulation. This is now possible thanks to the development of an efficient SWAP Monte Carlo technique [44–46].

We simulate a continuously-polydisperse system composed of $N = 1500$ particles at number density $\rho = 1$. We perform selected simulations with $N = 12\,000$. to analyze finite size effects. Particles interact via the repulsive pair potential $v(r_{ij}) = \epsilon(\sigma_{ij}/r_{ij})^{12} + F(r_{ij})$, where $F(r_{ij})$ guarantees the continuity of the potential up to the second derivative at the numerical cutoff distance $r_{\text{cut}} = 1.25\sigma_{ij}$, beyond which $v = 0$ [46]. The particle diameters are drawn from the normalized continuous distribution $P(\sigma_m \leq \sigma \leq \sigma_M) \sim 1/\sigma^3$. The size ratio of $\sigma_m/\sigma_M = 0.45$ was optimized to provide an excellent glass-forming ability. Similarly, we use a nonadditive interaction rule for the cross diameters $\sigma_{ij} = (\sigma_i + \sigma_j/2)(1 - \eta|\sigma_i - \sigma_j|)$, with $\eta = 0.2$. Length, time, and energy are respectively expressed in units of $\bar{\sigma} = \int \sigma P(\sigma) d\sigma$, $\sqrt{\epsilon/m\bar{\sigma}^2}$ and ϵ. The mode-coupling crossover temperature $T_d \approx 0.1$ is determined by fitting the relaxation time τ measured with standard dynamics to $\tau \sim (T - T_d)^{-\gamma}$ [46]. Using the SWAP

Monte Carlo technique, equilibrium can be ensured (using standard criteria [46]) down to $T \approx 0.6 T_d$.

To numerically mimic the state following scheme, the SWAP Monte Carlo technique is used to produce $N_s = 50$ independent equilibrium configurations at each T_g(0.062, 0.07, 0.075, 0.082, 0.092). We then generate $N_{\rm th} = 10$ copies of each configuration, that differ only by the initial velocities of particles (two such copies are referred to as A and B). Each of the $N_s \times N_{\rm th}$ samples is simulated at T_g in the NVE ensemble during a time t_q, depending on T_g ($t_q = 1000$ for $T_g = 0.062, 0.07$; $t_q = 100$ for $T_g = 0.075$; $t_q = 0$ for $T_g = 0.085$). The time t_q is chosen such that particles in different copies have time to explore their cages without diffusing. After t_q, the glass is instantaneously cooled to a temperature $T < T_g$ with a Berendsen thermostat (coupling parameter $\tau_T = 10$) [47]. Waiting times t_w are measured since the quench. We find that after $t_w \approx 100$, the temperature stabilizes to the desired value. For the highest $T_g = 0.092$ studied, diffusion is not totally suppressed at equilibrium. Glasses were first cooled down to $T = 0.07$ with a cooling rate $\gamma = 10^{-7}$ before making copies. We then used the same protocol as for $T_g = 0.07$ to obtain the data.

The Gardner transition is a second-order phase transition accompanied by diverging time scales and length scales characterizing vibrational dynamics. The transition signals profound changes in the structure of the landscape and the emergence of marginal stability. Mean-squared displacements (MSD) therefore represent the central observables for such an investigation [27,48]:

$$\Delta(t, t_w) = \frac{1}{N'} \sum_{i=1}^{N'} \langle |\boldsymbol{r}_i(t + t_w) - \boldsymbol{r}_i(t_w)|^2 \rangle,$$
$$\Delta_{AB}(t) = \frac{1}{N'} \sum_{i=1}^{N'} \langle |\boldsymbol{r}_i^A(t) - \boldsymbol{r}_i^B(t)|^2 \rangle, \quad (1)$$

where the brackets indicate averages over thermal fluctuations and disorder. They respectively represent the standard MSD and the relative MSD between two copies of the same glass. Since smaller particles may escape their cage more easily, we concentrate on the $N' = N/2$ larger particles.

The typical behavior of the MSDs after a quench is shown in Fig. 2(a). Both quantities converge to their long-time limits, Δ_{AB}^{∞}, Δ^{∞} after a time of order 100 (set by the thermostat). No sign of slower relaxation or aging behavior is detected at any state point, which indicates that the time dependence of the observables is not pertinent. The absence of slow relaxation contrasts dramatically with hard sphere simulations [27], and it directly reveals the absence of marginal stability throughout the glass phase.

We gather the results for Δ_{AB}^{∞} and Δ^{∞} in Fig. 2(b). The standard MSD changes linearly with T, as expected. The behavior of the relative distance is qualitatively the same

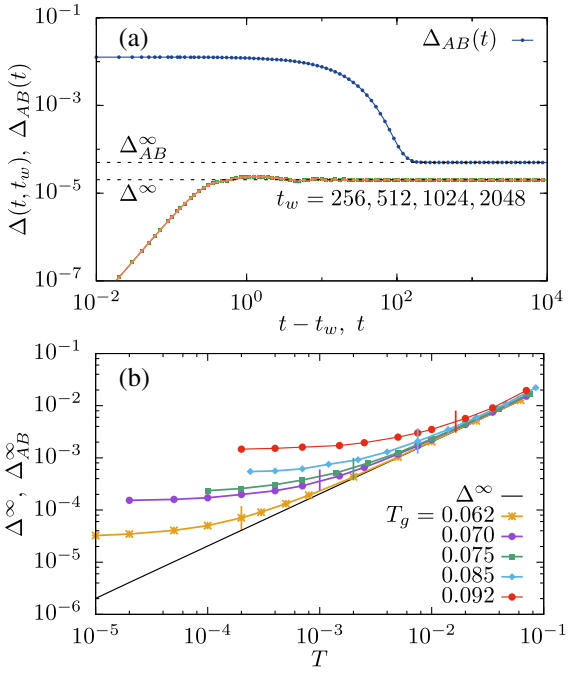

FIG. 2. (a) Mean-squared displacement $\Delta(t, t_w)$ and mean-squared distance $\Delta_{AB}(t)$ as a function of $t - t_w$ and t for $T_g = 0.062$ and $T = 10^{-4}$ rapidly converge to their long-time limits (dashed lines). (b) Long-time limits of Δ^{∞} and Δ_{AB}^{∞} as a function of temperature, for different T_g. Both quantities differ below $T^*(T_g)$, indicated by vertical segments.

for all T_g. The equality $\Delta_{AB}^{\infty} \approx \Delta^{\infty}$ holds at high enough T, meaning that the structure of the basin is relatively simple. There is a crossover temperature $T^*(T_g)$ (vertical segments), below which $\Delta_{AB}^{\infty} > \Delta^{\infty}$. The distance between two copies is then much larger than the vibrations they can perform individually, suggesting that the copies get quenched in distinct minima. This splitting of MSDs was observed in hard spheres [27,28] and identified as a Gardner transition. We report in Fig. 1(b) the crossover temperatures and the glass energy. The similarity between the two phase diagrams in Fig. 1 is obvious.

The absence of slow relaxation in Fig. 2(b) reveals the lack of a growing time scale. To address length scales, we study the global fluctuations of the relative MSD. The variance of these fluctuations defines the susceptibility $\chi_{AB} = N[\langle \tilde{\Delta}_{AB}^2 \rangle - \langle \tilde{\Delta}_{AB} \rangle^2]/[\langle \Delta_{AB}^{i2} \rangle - \langle \Delta_{AB}^{i} \rangle^2]$, where $\tilde{\Delta}_{AB}$ is the plateau value of the relative MSD for a given pair AB, and Δ_{AB}^{i} its single particle version. The normalization in χ_{AB} ensures that $\chi_{AB} = 1$ for spatially uncorrelated motion and that χ_{AB} is a direct measure of the correlation volume. If the crossover at T^* corresponded to a Gardner transition, the susceptibility would diverge near T^*. The results in Fig. 3(a) are very similar for all T_g values; the susceptibility increases very weakly as temperature decreases. Within our error bars, there are actually very little global fluctuations above the floor level. This directly

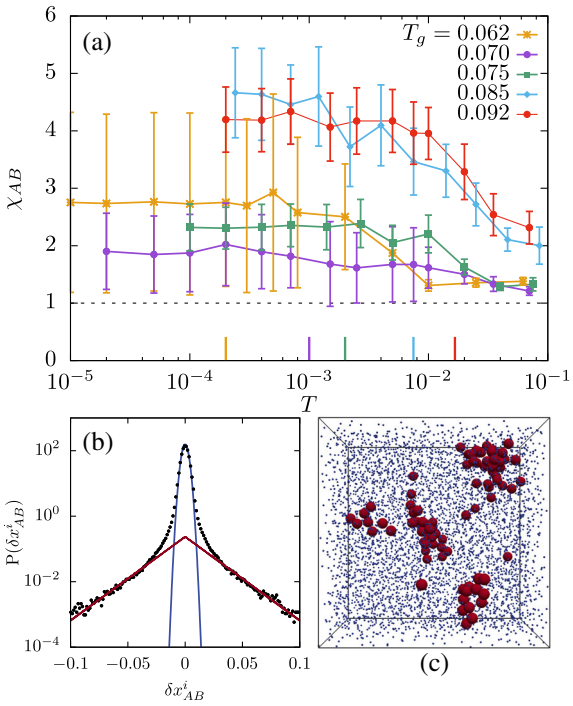

FIG. 3. (a) The susceptibility χ_{AB} for different T_g raises mildly above the floor level (dashed line) across T^* (vertical segments). The error bars are computed using the jackknife method. (b) The van Hove function of relative displacements has a narrow Gaussian core of width given by Δ^∞ (solid blue), and exponential tails (solid red) at $T_g = 0.062$ and $T = 10^{-4}$. (c) A corresponding snapshot with $N = 12000$ showing the few particles having displacements outside the Gaussian range (red spheres) among a majority of particles undergoing small amplitude Gaussian vibrations (blue dots).

demonstrates that spatial correlations between particle motion remain microscopic across the crossover T^*, which is thus not accompanied by a growing correlation length. This is consistent with the absence of slow dynamics in Fig. 2(a). A similar value of χ_{AB} was observed in larger systems of $N = 12000$ particles at specific state points. We studied the spatial correlation function for the relative MSD [27], of which the volume integral is χ_{AB}, and we did not find hints of a growing length scale at any temperature. We conclude that T^* does not coincide with the emergence of a marginally stable phase.

To understand the origin of the crossover observed in Fig. 2(b), we resolve the vibrational dynamics at the particle scale. We measure the distribution of relative particle displacements, $P(\delta x^i_{AB})$, where $\delta x^i_{AB} = x^i_A - x^i_B$ is the relative motion of particle i between copies A and B along the x direction. We average over the three directions of space. The van-Hove function is nearly Gaussian when $T^* < T < T_g$ with a width controlled by Δ^∞. Close to T^* and below, the distribution remains Gaussian in its core, but it exhibits tails that are well fitted by an exponential, as shown in Fig. 3(b). We evaluate the statistical weight of the particles contributing to the tails by integrating the exponential fit. It varies between 1% and 3% for all state points and typically increases with T_g. This corresponds to a small subset of particles that get frozen in slightly distinct positions below T^* in two copies. The error bars for χ_{AB} are large because the number of particles in the tails is small, and it fluctuates significantly from one pair AB to another. We have checked that these mobile particles encompass all particles, not only small ones that are more mobile. To gather spatial information on these few mobile particles, we select particles with a relative displacement δx^i_{AB} outside of the Gaussian core of the distribution and visualize them in snapshots. A typical snapshot obtained for $N = 12000$ is shown in Fig. 3(c). We represent the vast majority of particles with Gaussian displacements as small points and highlight particles contributing to the tails with larger red spheres. Strikingly, these mobile particles are clustered into sparse, localized defects. When T_g increases, the number of mobile particles as well as their characteristic displacement Δ^i_{AB} increases weakly. This directly accounts for the shift of T^* with T_g. These few localized clusters thus dominate the behavior of the relative displacement Δ^∞_{AB}, which is averaged over particles, and they are responsible for its separation from Δ^∞ in Fig. 2(b). Our key conclusion is that the emergence of these localized clusters at T^* does not correspond to a Gardner phase transition, and glasses below T^* are not marginally stable.

In our system, the marginal stability described within mean-field approaches is strongly suppressed by finite-dimensional fluctuations. Our results differ dramatically from previous work on hard sphere systems [27,28]. This surprising lack of universality contrasts with the universality of glass formation [2]. One possible explanation for this difference is that structural glasses may generically become marginal only when pushed towards specific "critical" transitions, such as jamming [49]. The jamming transition appears robust down to $2d$, with the same critical properties as in $d = \infty$ [50]. This could explain why a Gardner transition is observed in finite-dimensional hard sphere glasses near jamming, whereas glass formers with continuous interactions reach nonmarginal inherent structures at zero temperature. After this work was completed, two other works appeared reporting consistent findings [51,52].

Our results raise two types of questions. First, the presence of a marginally stable glass phase seems highly dependent on the details of the particle interactions and on dimensionality. To better understand the nature of the Gardner transition, one should better investigate the crossover between continuous and discontinuous interactions using, for instance, well-chosen particle interactions [53,54]. One should also investigate the crossover towards nonmean-field behavior by using either dimensionality [18] or the interaction range [48,55] as tuning parameters. Second, it would be interesting to connect the present

results with other observations of localized defects [10], such as soft localized modes controlling the low-frequency part of the vibrational spectrum in amorphous solids [56,57], localized defects controlling relaxation in supercooled liquids [58–61], or the shear-transformation zones [62–65] controlling the mechanical behavior of amorphous solids.

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