Equilibrium ultrastable glasses produced by random pinning

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Ultrastable glasses have risen to prominence due to their potentially useful material properties and the tantalizing possibility of a general method of preparation via vapor deposition. Despite the importance of this novel class of amorphous materials, numerical studies have been scarce because achieving ultrastability in atomistic simulations is an enormous challenge. Here we bypass this difficulty and establish that randomly pinning the position of a small fraction of particles inside an equilibrated supercooled liquid generates ultrastable configurations at essentially no numerical cost, while avoiding undesired structural changes due to the preparation protocol. We study the melting of these configurations following a sudden temperature jump into the liquid phase. In homogeneous geometries, enhanced kinetic stability is accompanied by large scale dynamic heterogeneity, while a competition between homogeneous and heterogeneous melting is observed when a liquid boundary invades the glass at constant velocity. Our work demonstrates the feasibility of large-scale, atomistically resolved, and experimentally relevant simulations of the kinetics of ultrastable glasses.

Recently, glasses with remarkable thermodynamic and kinetic stability have been prepared by vapor deposition on substrates maintained below the conventional glass transition temperature, $T_g$, of the bulk liquid.1–4 It is estimated that these “ultrastable” glasses occupy states that are so low in the energy landscape that it would take several decades of conventional annealing of amorphous samples to prepare materials with equivalent properties.1 In addition to potential technological applications,1,5 these novel materials raise new challenges for theory and thus opportunities for gaining a deeper theoretical understanding of amorphous materials.6

To rationalize the formation of ultrastable glasses, Ediger and coworkers have hypothesized that deposition on a cold substrate combined with enhanced mobility at the free surface allows the system to effectively burrow into deeper free energy minima.1 Computational studies using facilitated lattice models,7 simulations of Lennard-Jones systems,8,9 as well as theoretical analysis based on random first order transition theory10,11 have demonstrated phenomenology in harmony with these ideas. By inhabiting deeper basins on the energy landscape, stable glasses may be closer to the putative Kauzmann or “ideal glass transition” temperature, $T_K$. This idea has been quantified by experimental estimation of fictive temperatures lying well below $T_g$.1 Ultrastable glasses may thus provide an experimental means of producing an amorphous material with very low configurational entropy.

As difficult as it is to experimentally produce annealed glasses at temperatures that approach $T_K$, it is all the more so in silico. Even the most powerful computers and computational methodologies cannot simulate model supercooled liquids that approach $T_g$, let alone $T_K$, although work mimicking vapor deposition protocols8,9 as well as the use of biased sampling of trajectories,12,13 have moved us closer to this goal. Recently, theoretical investigations have been put forward that potentially make accessing and testing the behavior of “ideal glass” states possible using concepts borrowed from studies of fluids in porous media. Specifically, the physics of a fluid in the presence of a small fraction of randomly pinned particles has been shown numerically14 and theoretically15 to share essentially the same glassy physics as bulk supercooled liquids, which justifies more generally the use of this strategy in studies of glass formation.14–19

Random pinning presents two distinctive features with respect to bulk liquids that are central to our study. First, glassy dynamics and the transition to glassy states occur at temperatures that are higher than in bulk,16 because pinning a fraction $f$ of the particles restricts the available configurational space.14,15 Second, particle configurations produced by randomly pinning particles within a thermalized supercooled liquids are, by construction, at thermal equilibrium.20 Together, these two features suggest that equilibrium configurations created by random pinning correspond to a degree of supercooling at finite $f$ that cannot be achieved by conventional means. We hypothesize that these pinned systems must share many properties with ultrastable glasses formed in the laboratory via nonequilibrium vapor deposition techniques. Crucially, because we obtain pinned configurations directly from bulk fluids, preparation of stable configurations is numerically very easy. This allows us to explore large systems sizes, long relaxation timescales, and various geometries, while automatically avoiding any undesired structural changes due to preparation protocol such as anisotropy or concentration fluctuations.9,21,22 Mimicking experimental work characterizing ultrastable glasses, we first demonstrate ultrastability using conventional calorimetry. We then explore with atomistic resolution the kinetics of melting of stable glasses into the liquid phase following a sudden temperature jump, both in homogeneous and inhomogeneous geometries.

Model and preparation of “equilibrium pinned
We study the properties of a two-dimensional binary Lennard-Jones mixture, employing a 65:35 mixture of particles with interactions parameters as in the model of Kob-Andersen,23 which has been previously shown to be stable against crystallization and serves as a model supercooled liquid.24,25 Studying the two-dimensional system allows us to more directly visualize the spatial variation in observables. Exploratory studies of the three-dimensional system suggest that our results do not sensitively depend on dimensionality. We prepare many independent equilibrated supercooled configurations with \( N = 10^4 \) and density \( \rho = 1.2 \) at initial temperature \( T_i = 0.425 \). For the bulk, the mode-coupling temperature (which roughly coincides with the computer glass transition) is \( T \approx 0.4 \) (see Methods for a definition of reduced units). In these configurations, we fix the position of a percentage \( f \) of particles (see Methods). This produces equilibrium configurations at state point \( (f, T_i) \), which are our numerical analog of the samples that are vapor-deposited below the glass temperature, in the sense that both protocols produce amorphous configurations with low energy. To characterize these configurations, we either use finite rates to heat/cool them and study their calorimetric properties, or sudden temperature jumps into the liquid phase, \( T > T_i \), to follow the kinetics of “melting” of the glass into the liquid. We refer to our pinned samples as “pinned glasses,” even though they actually correspond to equilibrium samples. These deeply supercooled states come at no numerical cost. Indeed, the ordinary challenge of finding a good configuration satisfying a difficult constraint (such as low temperature) is reversed here because we define the hard constraint after the standard bulk configuration has been prepared (similar to the idea of planting, see e.g. Ref. 26).

**Calorimetric measurements establish ultrastability of randomly pinned samples**

To quantify the thermodynamic stability of our pinned samples, we mimic the experimental technique of scanning calorimetry. To this end we heat, then cool, then reheat the sample again at constant rate \( \dot{\gamma} = dT/dt \) from initial temperature \( T_i \) to a maximum temperature of \( T = 2.0 \). In Fig. 1(a) we show the results of this temperature scans for samples with \( f = 5 \) and \( f = 10 \). Upon initial heating, the system stays on the “stable glass branch” until the glass transition temperature is crossed. The system then melts and returns to the equilibrium liquid branch. On cooling, the samples do not return to the stable glass branch but to a different one which is much higher in energy. Little hysteresis is observed upon reheating, showing that the glass configurations produced by conventional cooling are much less glassy. We also observe that the simulations for larger \( f \) need to be heated to a much higher temperature before they “melt”, confirming that the glass temperature increases with \( f \). The difference in energy between the original sample and re-annealed samples also increases with \( f \). Differential scanning calorimetry experiments on ultrastable glasses look quite similar, in the sense that the experimental conditions under which these glasses were prepared determine how different the two glass branches are.

By taking the numerical derivative of the energy, we extract the heat capacity \( c_V \) for pinned samples, shown in Fig. 1(b). We see that the peak temperature signaling the glass transition moves to higher temperature with increasing \( f \), and the peak height also grows with \( f \). By contrast, heat capacities for the re-annealed samples (dotted) display a much smaller peak, providing verification that ordinary annealed samples occupy much higher energy states. The peaks in \( c_V \) of the re-annealed glasses...
lie approximately under those from the initial heating. This is in contrast to what is seen for ultrastable glasses, where the re-annealed samples melt at lower temperature than the vapor deposited ones.\(^1\) This is probably because our numerical heating rates are much faster than in experiments, resulting in \(c_V\) peaks that are very broad, and thus much harder to resolve.

The energy gap between stable and re-annealed pinned glasses depends on the annealing rate, and it shrinks for slower cooling (by definition the gap should vanish when \(\gamma \to 0\)). In Fig. 1(c) we show the energy gap \(\Delta E\) measured at \(T = T_i\), normalized by the average initial energy \(E_0\). In all cases, we observe \(\Delta E > 0\), with gaps that increase with \(f\). Extrapolation of these curves, even for modest \(f\), would suggest that only cooling rates smaller by several orders of magnitude could produce similarly stable configurations. This directly demonstrates the advantage of using pinned glasses, which can truly be described as extremely “old” glasses that we obtain at essentially no numerical cost, in excellent analogy with vapor-deposited glasses.\(^1\)

**Kinetics of homogeneous melting of ultrastable pinned glasses into the liquid**

We now study the response of pinned glasses after a sudden change in temperature to \(T > T_i\), the subsequent dynamics occurring fully out of equilibrium. We compute the self-overlap function for the \(N_u\) unpinned particles, \(q_s(t, t_w) = \frac{1}{N_u} \langle \sum_m g_m(t, t_w) \rangle\) with

\[
g_m(t, t_w) = \theta \left( |r_m(t + t_w) - r_m(t_w)| - a \right),
\]

where \(t_w\) is the waiting time since the temperature jump, \(\theta(x)\) is the step function which is unity for \(x \leq 0\) and zero for \(x > 0\), and \(r_m(t)\) denotes the position of particle \(m\) at time \(t\). We choose \(a = 0.22\) such as the self-overlap relaxation time \(\tau_s\), defined as \(q_s(\tau_s, 0) = 1/e\), matches the relaxation time \(\tau_{s}\) in equilibrium conditions and as defined in a previous work.\(^{25}\)

The relaxation of samples heated to \(T\) becomes drastically slower as \(f\) is increased, as shown for \(T = 1\) in Fig. 2(a). In addition, a plateau develops and grows in height suggesting a more stable glass.\(^{8,22}\) When the waiting time increases, the particle dynamics accelerates as the glass progressively melts into the liquid, see Fig. 2(b). The system eventually relaxes towards the thermalized liquid state where dynamics becomes stationary. A comparison between the \(t_w = 0\) (glass melting) and \(t_w \to \infty\) liquid relaxation data shows that (i) the relaxation time of the liquid is faster, (ii) its time correlation function shows more pronounced deviation from exponential decay, (iii) and the intermediate plateau height is much lower. While (i) and (iii) reveal kinetic stability of the pinned glasses, (ii) is more surprising at it could naively suggest that the melting relaxation process is less heterogeneous than the equilibrium dynamics. We shall see below that the opposite is actually true.

The overlap relaxation time \(\tau_s(T, f, t_w)\) can serve as an indicator of the progress of glass melting. In Fig. 2(c), we show the overlap relaxation time for each \(f\) after waiting time \(t_w\). The relaxation time is initially large, decays to the equilibrium value, and eventually becomes independent of \(t_w\). We define the transformation time \(t_{trans}\) as the time where \(\tau_s(T, f, t_w)\) has fully decayed to this plateau value, in analogy to its experimental determination.\(^{4,21}\) We observed that \(q_s(t_{trans}, 0, T) \equiv 0.01\) serves as a consistent proxy for \(t_{trans}\) across all \(f\) and \(T\) and is a simpler and more accurate tool for the extraction of \(t_{trans}\) than is inspection of the \(\tau_s\) curves, and so we adopt this practical definition of \(t_{trans}\).

With increasing \(f\), we observe a substantial increase
in $t_{\text{trans}}$ relative to that for unpinned samples. For example, the data in Fig. 2(c) show an increase of a factor of $\approx 10^3$. However, the equilibrium relaxation time $\tau_\alpha$ also increases with $f$. It is sensible to define a “stability ratio”, $S = t_{\text{trans}}/\tau_\alpha$, to characterize the melting of ultrastable glasses, as done in a recent set of experiments.$^4$

In Fig. 3(a) we present the evolution of the stability ratio for all $(f,T)$ points studied. The corresponding wide range of $\tau_\alpha$ is highlighted in Fig. 3(b). We find that $S$ is always larger than about 100, with a maximum value reaching $S \approx 500$ for the most stable systems. For given preparation and melting temperatures $(T_i,T_f)$, we find that $S$ increases with $f$. This result is expected since the initial state then corresponds to a pinned glass that is increasingly stable, as demonstrated in Fig. 1. Our data also indicate that increasing the melting temperature $T$ decreases both $\tau_\alpha$ and $t_{\text{trans}}$, but their ratio remains approximately constant. This suggests that $S$ is a robust way to compare the stability of samples with different preparations.

While $S$ is substantially larger for the equilibrium pinned glasses than for conventionally prepared glass configurations (our tests indicate a gain of about 10), we do not find stability ratios $S$ in the range $10^3 - 10^6$ reported experimentally.$^3$ The main reason is obvious from Fig. 3. Our results reveal that the stability ratio increases only weakly with increasing the glass stability, since the equivalent of several decades of annealing increases $S$ by less than a decade. However, although our pinned glasses correspond to equilibrium, low energy states, we melt them in a relatively high temperature regime such that both $\tau_\alpha$ and $t_{\text{trans}}$ can be numerically measured. Instead, glass melting in experiments is performed at much lower temperatures close to the experimental $T_g$. Extrapolating the data in Fig. 3(a), which increase with decreasing $T$, to $\tau_\alpha$ values 8 orders of magnitude larger could plausibly yield stability ratios comparable to the ones determined experimentally. Note that our equilibrium pinned samples do not display any structural change with respect to the bulk, such as anisotropy, layering, or compositional fluctuations which might occur during vapor deposition.$^3, 21,22$ Such structural changes could also contribute to increasing the stability ratio measured experimentally.

**Enhanced kinetic stability is accompanied by large-scale dynamic heterogeneity**

Atomistic simulations offer the possibility to observe how the melting process of the glass occurs in homogeneous geometries. In Fig. 4 we show the spatial distribution of $q_m(t = \tau_{1/2}, t_w = 0, T)$, where $\tau_{1/2}$ is the time
at which $q_4(t = \tau_{1/2}, t_w = 0) = 1/2$, and the color of the particles represents $q_m$ coarse-grained over a local region of radius 1.5, for clarity purposes. Fig. 4(a-d) show snapshots for $f = 0, 2.5, 7.5, \text{ and } 12.5$, which allow us to observe how the stable glass initial configuration (in blue) progressively melts into the liquid (in red). We observe that liquid pockets emerge at some random location in the glass, and grow in size until the whole system has melted. Clearly as $f$ (and thus the transformation time) increases, we see that the “nucleation” regions where the liquid first appears become sparser, so that the size of the liquid/glass domains observed at time $t = \tau_{1/2}$ grow in size. Using the language of equilibrium glass transition studies, the melting dynamics becomes spatially correlated over larger distances when kinetic stability increases.

To quantitatively validate this visual impression, we compute the four-point dynamical structure factor which measures the scattering off of these dynamic domains,

$$S_4(k, t, t_w) = \frac{1}{N_k} \sum_{m,n} q_m q_n e^{i k (r_m(0) - r_n(0))},$$

where $q_m$ is shorthand for $q_m(t, t_w)$ defined in Eq. (1). The resulting profiles shown for one temperature in Fig. 4(e) have larger spatial variation measured by $S(k \rightarrow 0, t)$ with increasing $f$. This reveals a growing non-equilibrium length scale for dynamically heterogeneous melting. If we increase the waiting time, we observe from Fig. 4(f) that the dynamic heterogeneity decreases as the sample transforms back into the liquid. Finally, for a given value of $f$, we find that $S_4$ also grows as $T$ is decreased (data not shown).

Fig. 4 shows that glass melting is spatially heterogeneous, with a correlation length scale increasing with the glass stability. Additionally, dynamics is correlated over larger distances (and is thus more “heterogeneous”) for $t_w = 0$ (where time correlations are nearly exponential) than for equilibrium (where time correlations are stretched). Another interesting point is the growth of spatial correlations with $f$ at constant $T$ observed in Fig. 4, which contrasts with recent numerical studies suggesting a much weaker effect on the dynamic correlation length in equilibrium conditions. Our nonequilibrium data therefore confirm the qualitative idea that increasing $f$ moves the system closer to a glass transition.

Physically, our results appear consistent with both a dynamic picture where relaxation is first triggered by a sparse population of “defects” and then propagates in space via dynamic facilitation, and with a thermodynamic picture where the melting occurs via “nucleation and growth” of the liquid into the glass, as envisioned by random first order transition theory.

Coexistence of homogeneous melting and front propagation in inhomogeneous geometries

Experiments on ultrastable glasses in inhomogeneous geometries have demonstrated that when the stable glass is capped with a liquid interface, melting can be initiated from the liquid boundary which propagates at constant velocity into the glass. These observations suggest that “heterogeneous glass melting” can compete with the homogeneous process studied above, in complete analogy with nucleation across a first-order phase transitions.

To investigate this situation, we produce a liquid/glass interface by leaving a strip along the $y$ direction of the system with no pinning ($f = 0$), while the rest of the system is pinned at finite $f$, as before. The unpinned region is the numerical analog of the liquid interface in experiments. In this inhomogeneous geometry, we monitor the self-overlap profiles along the $x$ direction perpendicular

FIG. 5. From bulk to heterogeneous melting via front propagation of the liquid phase. (a-d) Relaxation profiles for systems where $f = 5$ (a,b) or $f = 10$ (c,d) everywhere except in the middle of the sample (dashed lines) where $f = 0$. The samples are heated to $T$ and the overlap profiles $q(t, x)$ are shown as a function of the time $t$ (color code as in Fig. 4). Relaxation occurs rapidly in the center, and the liquid slowly invades the glass until homogeneous melting occurs and the interface disappears. (e) Time slices for the data in panel (d) for times increasing from top to bottom. (f) The temperature evolution of the melting front velocities $v_{\text{melt}}$ essentially tracks that of the structural relaxation time, $v_{\text{melt}} \sim \tau^{-1}$. 


to the liquid strip, \( q(t, x) \equiv \langle q_m(t, t_w = 0) \rangle_y \) for particles \( m \) with horizontal positions in a strip of width 0.8 centered at \( x \). The results in Fig. 5(a-d) demonstrate that relaxation occurs very rapidly in the unpinned region, creating a liquid/glass (red/blue) interface, as desired. As time increases, the position of the interface (white) moves and the melted section propagate into the pinned glass. When \( t \sim t_{\text{trans}} \), the pinned glass may relax homogeneously and the interface disappears. The system is entirely fluid when \( t \gg t_{\text{trans}} \). Representative overlap profiles\(^7,34\) are shown in Fig. 5(e). We locate the “propagating front” from the overlap value \( q(t, x_{\text{from}}) = 0.2 \), and fit its position for a given \( (T, f) \) to a linear function of time, which defines the “melting velocity”, \( v_{\text{melt}} = v_{\text{melt}}(T, f) \).

The temperature dependence of \( v_{\text{melt}} \) is shown for four values of \( f \) in Fig. 5(f). At higher temperatures, it was not possible to observe a melting front over sufficient time range. This is exemplified in Fig. 5(a), where at \( \log_{10}(t) \approx 3.2 \), the sample has fully melted at a position far from the interface, indicating that the time scale for the bulk relaxation is shorter than the time taken for the front to propagate over a significant distance. Our numerical results indicate that \( v_{\text{melt}} \) is approximately proportional to \( \tau^{-1}_\alpha \), see Fig. 5(f), suggesting that the same relaxation mechanisms that govern melting front propagation allow for density relaxation in the pinned bulk. It is similarly found in experiments that \( \ell \equiv v_{\text{melt}}\tau_\alpha \) is a microscopic length scale with weak temperature dependence.\(^4\) In our simulations, \( \ell \) has molecular dimensions.

The observed competition between front propagation (speed \( v_{\text{melt}} \)) and homogeneous melting (timescale \( t_{\text{trans}} \)) suggests a maximum length scale over which heterogeneous melting can be observed, \( \xi \approx v_{\text{melt}}t_{\text{trans}} \). Interestingly this can be rewritten as \( \xi = \ell \times S \), showing that \( \xi \) is mainly controlled by the absolute value of the stability ratio \( S \). Since the values observed in experiments can be up to \( S \approx 10^6 \), a reasonable assumption of molecular dimensions for \( \ell \) results in a maximal length scale for front propagation of the order of microns, as found.\(^4\) Our prediction that \( \xi \propto S \) is experimentally verifiable. Since this massive length scale directly results from kinetic stability, it should not be viewed as a large correlation length scale characterizing ultrastable glasses.

**Discussion and perspectives**

We have shown that the random pinning of particles in a two-dimensional model supercooled liquid produces highly stable in silico glasses termed “pinned glasses”. Our approach does not address specificities due to the experimental vapor deposition process, but allows instead a detailed exploration of the physical properties of ultrastable glasses. We have demonstrated that pinned glasses behave in many ways as experimentally realized ultrastable glasses. Both systems lie deep in the energy landscape, are characterized by peculiar calorimetric properties, and enhanced kinetic stability. Both exhibit a competition between homogeneous and heterogeneous glass melting, which results, for inhomogeneous geometries, in a liquid front invading the glass over a large length scale controlled by the ratio \( S = t_{\text{trans}}/\tau_\alpha \) between the bulk transformation time and the equilibrium relaxation time.

An important difference between the two systems is that pinned samples are formed in equilibrium, with no structural change induced by the preparation protocol, i.e. we can fully decouple stability and structure, in a way that might be experimentally realized in colloidal materials.\(^35,36\) We have attributed the larger stability ratio values measured experimentally both to this distinction and to the fact that melting simulations are performed in a different temperature regime than in experiments. Our work nevertheless shows that preparing energetically favorable configurations via random pinning is a promising route to understand the properties of vapor-deposited ultrastable glasses. Both types of systems thus appear as a type of amorphous materials that cannot be prepared by conventional annealing protocols, which confers them with extraordinary physical properties.

**Methods**

The Koh-Andersen Lennard-Jones system\(^23\) is a binary system of particles with pairwise Lennard-Jones interactions, such that particles \( i \) and \( j \) separated by a distance \( r_{ij} \) interact with the potential \( V(r_{ij}) = \epsilon_{\alpha\beta} (\sigma_{\alpha\beta}/r_{ij})^{12} - (\sigma_{\alpha\beta}/r_{ij})^6 \), where \( \alpha \) and \( \beta \) represent the particle type, \( A \) or \( B \) for particle \( i \) and \( j \) respectively. Interaction parameters are given by \( \epsilon_{AA} = 1, \epsilon_{AB} = \epsilon_{BA} = 1.5, \epsilon_{BB} = 0.5 \), and \( \sigma_{AA} = 1, \sigma_{AB} = \sigma_{BA} = 0.8, \sigma_{BB} = 0.88 \). The interaction between particle types \( \alpha \) and \( \beta \) is truncated and shifted up at distance \( 2.5\sigma_{\alpha\beta} \). All particles have equal mass \( m \). Energies, distances, and times are reported in reduced units proportional to \( \epsilon_{AA}, \sigma_{AA} \) and \( \tau = \sqrt{m\sigma_{AA}^2/\epsilon_{AA}} \). We study the two-dimensional variant of the model, for which a 65:35 ratio of \( A \) to \( B \) particles was previously shown to be a robust model supercooled liquid, resistant to crystallization.\(^24,25\) We generated configurations using Molecular Dynamics at a series of decreasing temperatures following the procedure of Ref. 25 to generate configurations at density \( \rho = 1.2 \) with \( N = 10000 \) down to \( T = 0.45 \), and then generated configurations at \( T = 0.425 \) by running simulations on these configurations for an additional \( 1.25 \times 10^7 \tau \) using an integration time-step of \( dt = 0.0005\tau \). In all cases, simulations were performed using LAMMPS,\(^37\) and the temperature was maintained using a Nosé-Hoover thermostat with a time constant of \( 100dt \).\(^38\)

We wished to perform simulations with a fraction \( f = f/100 \) of particles fixed in place in a “uniformly random” manner similar in spirit to Ref. 14 to avoid sampling problems arising from localized extra-slow dynamics due to clusters of pinned particles. In order to do this, we chose a minimum distance between pinned particles \( d_{\text{min}}(f) \) and then randomly picked a set of particles commensurate with this minimum pair difference. In practice, we were able to do this for the \( f \) of interest by using \( d_{\text{min}}(f) = 0.85d_{\text{avg}}(\phi = 0.55) \), where \( d_{\text{avg}}(\phi) \) is the diameter \( fN \) particles would have if they were placed in the same box with packing fraction \( \phi = f\pi(4d_{\text{avg}}/2)^2 \). In this way the radial distribution function for the pinned particles is similar to that of a simple liquid with packing fraction \( \phi = 0.55 \), which was chosen heuristically. The results presented in this work appear robust to the specific choice here,
or indeed if the particles to pin are chosen totally at random, and this particular sampling should be viewed as a numerical convenience with no incidence on our results.

In order to perform the heating and cooling experiments shown in Fig. 1, twelve simulations were run for every value of $f$ and $\gamma$, by choosing four uncorrelated sets of $\tilde{f}N$ random particles from three independent starting configurations. These configurations were heated from $T_i = 0.425$ to $T = 2.0$ with the thermostat temperature raised in a linear fashion at a rate $\gamma$. The samples were subsequently cooled back to $T = 0.425$ and reheated to $T = 2.0$, also at a rate $\gamma = dt/dt$. For other data in this work, the thermostat temperature was instantly raised to a “melting temperature” $T$ into the liquid phase. For any data point or curve shown here, at least 8 simulations were performed, with 4-6 independent configurations using 2-5 sets of randomly pinned particles. The simulation temperature stabilized at $T$ by 1000$\Delta t$, corresponding to about 10 times the thermostat relaxation time. For all of these simulations, the simulation integration time-step was kept at $\Delta t = 0.005$ without any adverse consequences.

Author Contributions L.B., G.M.H., and D.R.R. conceived the study. G.M.H. designed, executed, and analyzed computational experiments. G.M.H., D.R.R., and L.B. analyzed the results and wrote the paper.

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