Does a growing static length scale control the glass transition?

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Several theories of the glass transition propose that the structural relaxation time $\tau_\alpha$ is controlled by a growing static length scale $\xi$ that is determined by the free energy landscape but not by the local dynamical rules governing its exploration. We argue, based on recent simulations using particle-radius-swap dynamics, that only a modest factor in the increase in $\tau_\alpha$ on approach to the glass transition may stem from the growth of a static length, with a vastly larger contribution attributable instead to a slowdown of local dynamics. This reinforces arguments that we base on the observed strong coupling of particle diffusion and density fluctuations in real glasses.

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When a liquid is cooled sufficiently rapidly to avoid crystallization, its viscosity $\eta$ increases. In general, $\eta \simeq G\tau_\alpha$, where $G$ is the (plateau) shear modulus and $\tau_\alpha$ characterizes the relaxation time of density fluctuations. As the temperature $T$ is lowered, $G$ evolves mildly but $\tau_\alpha$ increases by about 15 order of magnitude \cite{1} until, at the transition temperature $T_G$, it becomes too large to measure experimentally. The liquid then becomes a glass, and falls out of equilibrium. Near the glass transition, the diffusion of a tagged particle also becomes very slow. The characteristic time $\tau_D$ over which a particle diffuses its own radius increases, albeit not as much as $\tau_\alpha$. The decoupling of these two quantities (referred to as the Stokes-Einstein breakdown) is significant, but comparatively mild: the ratio $S = \tau_\alpha / \tau_D$ is increased by only a few orders of magnitude at $T_G$ \cite{2,3}.

The dependence of $\tau_\alpha$ on $T$ is used to classify glassy liquids \cite{4}. Writing $\tau_\alpha = \tau_0 \exp(E/k_B T)$ where $\tau_0$ is a vibrational time scale (in the picosecond range) and $E$ some activation energy, one finds that $E$ is constant in some liquids, called strong, but increases up to a factor 5 under cooling in other liquids, called fragile. Fragility is best shown in the ‘Angell plot’ of $\log(\tau_\alpha)$ vs. $T_G/T$ \cite{4}, which is linear for strong liquids but highly curved for fragile ones. Quantitatively, fragility is defined as $m = \log(\tau_\alpha)/\log(T_G/T)$ evaluated at $T_G$ itself. Strong liquids have $m \simeq 25$; very fragile ones have $m \simeq 120$.

There are competing explanations of the increase in the activation energy $E$ in fragile liquids, which occurs with no obvious change of static structure \cite{5}. Several theories, including the Adam-Gibbs scenario \cite{6}, Random First Order Theory (RFOT) \cite{7,8}, and those involving locally favored structures \cite{9,10}, posit that the increase in $E$ stems from the growth of a purely static length scale $\xi$, characterizing some ‘hidden order’ in the many-body free-energy landscape that is not captured by traditional probes of static structure such as pair correlations.

In particular, in modern interpretations of RFOT \cite{5,8}, $\xi$ is a ‘point-to-set’ correlation length set by the minimum scale on which alternative packings are available to a patch of fluid whose environment is held frozen. Shorter scale motions do cross local barriers, but cannot discover a new pattern for the density which keeps returning to its initial state. In this view, regardless of how rapidly these local moves can permute the particles within the patch, $\tau_\alpha$ is controlled by the fact that the system can relax fully its density fluctuations only via collective rearrangements on the scale $\xi$ that ‘break’ the hidden order. The resulting collective activation energy is $E_{\text{coll}} \simeq c_0(T)\xi^a(T)$, where $c_0(T) \simeq c_0(T_G)$ is non-singular, and $\psi$ is some exponent. It can be expressed in terms of thermodynamic quantities alone and is thus independent of the details of the dynamics \cite{15} (see more on that below).

Although there is clear empirical evidence that a growing static length scale $\xi(T)$ exists (see e.g. \cite{12} and references therein), its role in the dynamics is debated. Alternative theories instead propose that the increase of $\tau_\alpha$ stems from growing barriers to the elementary rearrangements required to explore the landscape \cite{13–17}. Such barriers effectively add a term to the activation energy $E_\text{el} = c_1(T)$. Unlike $E_{\text{coll}}$, these kinetic barriers can depend greatly on the details of the dynamical rules governing the system. While the definition of ‘elementary rearrangements’ can be multi-particle (see more on that below), $E_\text{el}$ describes local physics and cannot diverge. However it might grow strongly enough near $T_G$ to control the glass transition via $\tau_\alpha \simeq \tau_0 \exp[E_{\text{el}}/k_B T]$. For example in elastic models \cite{13}, rearrangements require a certain strain, giving $E(T) \sim G_{\infty}(T)$ where $G_{\infty}$ is the high-frequency (vibrational) shear modulus. Empirically these quantities are indeed strongly correlated \cite{13,14}, as seen by plotting $\log(\tau_\alpha)$ vs. $(T_G G_{\infty}(T))/(T G_{\infty}(T_G))$, which now appears almost perfectly linear even for fragile liquids \cite{14,15}.

Combining the two activation terms gives $\ln(\tau_\alpha/\tau_0) \simeq a_0T^\psi + a_1$, with $a_0 = c_0/k_B T$ and $a_1 = c_1/k_B T$. This estimate coincides in form with an upper bound on the relaxation time \cite{10}; this bound shows that a ‘diverging $\xi$’ is rigorously required for an ideal glass transition ($\tau_\alpha/\tau_0 \to \infty$). It is then tempting to conclude that growth of $\xi$ controls the real glass transition ($\tau_\alpha/\tau_0 \to 10^{15}$).

In this work we present arguments for the contrary view, proposing instead that most of the increase in $\tau_\alpha$
stems from an increase upon cooling of $E_{el}$. Our main argument is based on recent numerical observations [20–23] in which a judicious choice of dynamics is shown to equilibrate systems far deeper inside the glass ($T \ll T_G$) than previously achievable. Indeed this dynamics almost abolishes the glass transition. We shall infer from this outcome that $E_{el}$ rather than $E_{coll}$ is dominant in controlling $\tau_\alpha$. This finding applies, strictly speaking, only to the polydisperse systems studied numerically in [20–23]. We then give a more general and complementary argument, to the effect that the Stokes-Einstein factor should obey $S \geq \exp[E_{coll}/k_BT] = \tau_\alpha/\tau_{el}$. It follows that the growth of $\xi$ contributes only a few decades at most to the 15 decades increase in $\tau_\alpha$ on the approach to $T_G$. Our findings are depicted schematically in Fig. 1.

![FIG. 1: Left panel: schematic of the free energy landscape $F(|\mathbf{r}|)$ for models where the dominant kinetic barriers $E_{coll}$ are collective, and appear on a length scale $\xi$. Local barriers $E_{el}$ are much smaller than $E_{coll}$. In this scenario introducing swap moves in the dynamical rules should have little effect on structural relaxation, since it leaves $E_{coll}$ unchanged; also there should be a very large decoupling between particle diffusion (that can occur by permuting particles, which requires to overcome local barriers only) and relaxation of density fluctuations (for which collective barriers must be overcome). Both predictions appear to be contradicted by observations, suggesting that local barriers dominate the slowdown (right panel). (In this case the definition of $E_{coll}$ and $\xi$ may become somewhat arbitrary.)](image-url)

Swap algorithms. Recent Monte-Carlo schemes have managed to equilibrate liquids at temperatures where traditional algorithms would need at least a $10^{10}$ speed-up to allow equilibration [20, 22]. These swap algorithms consider polydisperse particles. In addition to the usual translational moves, they allow for moves in which two particles of different radius exchange places. This can also be viewed as a radius-swap at fixed particle positions (in which case there is no direct contribution to tagged-particle diffusivity). One then has particles whose sizes are an extra set of dynamical variables, with update rules that preserve detailed balance. Accordingly the free energy landscape $F(|\mathbf{r}|)$ as a function of the particle positions $|\mathbf{r}|$ is, after integration over sizes, well defined and independent of the chosen dynamical rules. The swap rules can involve either well-separated particles as reported in [22, 23], or be local, for instance restricted to nearest neighbour swaps [20]. For the dynamical rules of [22, 23], the time scale $\tau_\alpha^\pm$ on which density fluctuations relax with swaps [49] can be measured and compared to the value $\tau_\alpha$ without swaps allowed. The latter case behaves like molecular dynamics simulations, and display the standard glass transition. Remarkably though, with the swap dynamics switched on the glass transition is essentially gone: $\tau_\alpha^+(T_G)/\tau_\alpha$ is only around $10^2 - 10^3$. This contrasts with $\tau_\alpha/\tau_0 \approx 10^{15}$ for the standard dynamics at $T_G$.

Implications of these observations. Local and long-range swap dynamics do appear to give very similar results for $\tau_\alpha$ [24], as we justify in S.I. This suggest that either type of swap dynamics can mollify local barri-
the glass transition. To support this view, we now present a polydispersity-free argument that again points toward control predominantly by local barriers.

For concreteness we suppose RFOT to correctly identify the static length scale \(\xi\). Recall that in this approach, each configuration is a mosaic of states, whose characteristic size \(\xi\) results from a competition between configurational entropy and “surface tension” between states. The activation barrier to nucleate a new density configuration on a length scale \(r\) then varies as \(cr^\psi\): the dynamics is fast on short length scales, but slow on long length scales. However for \(r < \xi\), a single state is thermodynamically favored. Local rearrangements are possible, but these will not fully relax density fluctuations. Processes that can relax density fluctuations occur on a scale \(\xi\), the smallest on which alternative density patterns can appear \[8\].

We believe that this picture contradicts the observation in real glasses the Stokes Einstein (SE) factor \(S = \tau_s/\tau_D\) at \(T_G\) is of order 10^3 [3, 27], rather than a much larger value. In practice, much of the violation is thought to arise from dynamic heterogeneity (DH), in which diffusive and structural relaxation times are dominated by the most liquid and most solid regions respectively [28]. However, DH can only increase \(S\): any other mechanism found to contribute to the SE violation therefore gives a lower bound on it. We may therefore write \(S = S_1 S_2 \geq S_1\), where \(S_2\) accounts for DH, and \(S_1\) accounts for the possibility that particles can exchange positions while leaving density fluctuations unchanged [29]. We now argue that \(S_1 \sim \exp[\alpha_1 \xi^\psi]\), by noting first that, even for \(r < \xi\), there is always an exponentially large number of states available for conventional dynamics, corresponding to permutations of the particles within a fixed density pattern. Any such state can be reached by a series of local permutations, which can each be generated by local rearrangements only. Without polydispersity there cannot be any thermodynamic reason for a particular permutation to be preferred, so a tagged particle can move diffusively with \(\tau_D \simeq \tau_0 \exp[\epsilon_{el}/k_B T] = \tau_\alpha/S_1\).

We then have \(S_1 = \exp[\alpha_0 \xi^\psi] < S \simeq 10^3\), implying that the growth of \(\xi\) cannot account for more than about a 3 decade increase in \(\tau_\alpha\) at \(T_G\). Were the glass transition controlled purely by growth of \(\xi\), a much stronger decoupling of \(\tau_D\) and \(\tau_\alpha\) could be expected. This is incompatible with the fact that the fragility defined from \(\tau_D\) is almost as large as that defined from \(\tau_\alpha\) (only 13-25\% [23] smaller for the liquids studied in [3, 27]). In practice therefore, diffusive and density relaxations are much more strongly coupled than RFOT seems to imply, and this points to a major role for \(E_{el}\) at the glass transition. The same arguments apply to any theory for which fast local moves are insufficient to break a hidden order on a growing scale \(\xi\).

In the specific case of RFOT our argument can be restated as follows: for the nucleation picture to hold (whereby a new density pattern appears by an activated barrier crossing at scale \(\xi\)) local equilibrium in the landscape \(F(\{\mathbf{r}\})\) should be reached, on all length scales shorter than \(\xi\), at time scales much smaller than \(\tau_\alpha\). But this local exploration of phase space permutes particles, leading to diffusion on the same fast timescale. Thus in any temperature regime where RFOT is dominant in driving the slowdown in \(\tau_\alpha\), a severe Stokes Einstein breakdown should occur. This is not seen in practice.

Asymptotic equivalence of landscapes. We now return to the case of polydisperse particles and explain why, as we assumed above, the free energy landscape \(F(\{\mathbf{r}\})\) is the same for dynamics with and without swaps, for large enough \(\tau_\alpha\). This implies in turn that \(E_{coll}\) is the same (including its prefactor \(c_1\)) for the two types of dynamics, precisely in the regime in which a growing static length could come to dominate the dynamics. The argument is almost the same as the one just given for monodisperse particles: for large \(\xi\), local rearrangements are rapid in comparison to \(\tau_\alpha\), sampling with Boltzmann weight configurations where groups of particles are permuted locally. Thus traditional (non-swap) dynamics already performs local swaps on time scales much shorter than \(\tau_\alpha\) in this limit. The collective free energy barriers, when large, must thus be described by the same \(F(\{\mathbf{r}\})\) with and without swaps, which is what we assumed above.

Discussion. We are not suggesting that a growing length scale \(\xi\) cannot contribute at all to the slowdown of dynamics in real glasses. But to play more than a supporting role, the resulting order must affect elementary rearrangements themselves, via \(E_{coll}\). For example, in the spirit of elastic models, locally favored structures that grow under cooling [11, 11] could increase \(G_\infty\), which slows down local moves requiring finite strain. Similarly, the observation that radius-swap moves dramatically speed up the dynamics of glasses does not contradict dynamically-facilitated models [30, 31], since in these models the choice of local dynamics is important. However they do need to be consistent with the fact that particle diffusion and density relaxation dynamics are strongly coupled.

In our view, the proof that a static length scale must diverge if \(\tau_\alpha\) does [19] is of limited practical relevance for real liquids near their glass transition, since the proper signal for this divergence is \(\tau_\alpha/\tau_D\) which increases relatively mildly within the experimental range. (Moreover, \(\xi\) need only diverge as \((\ln(\tau_\alpha/\tau_D))/\psi\)). The same conclusion is reached if, instead of considering the dynamics in terms of the rescaled temperature \(T/T_G\), one normalizes it by a natural energy scale \(G_\infty(T)/G_\infty(T_G)\) (which cannot be singular for smooth interactions). As stated earlier, the dynamics shows no sign of divergence if plotted in terms of \((T_G G_\infty(T))/(T_G G_\infty(T_G))\) [13, 18].

On the other hand, with local barriers mollified by swap dynamics as in [22, 23], the growth of \(\xi\) could become paramount, and it is possible that RFOT is the correct theory of such dynamics. For realistic dynamics however theories of local barriers appear to be needed to describe the glass transition. One such theory is provided by elastic models [13, 14], for which \(E_{coll}\) is proportional to \(G_\infty\). However the spatial description of local moves
remains very crude in these models. Importantly, they can involve finite collections of particles, for purely dynamical reasons. For example, it may require less strain energy to permute three particles forming a triangle than two neighboring particles.

Understanding the geometry of such elementary rearrangements is a challenge. Recently there has been a convergence of different approaches to compute the properties of soft collective vibrational modes in hard sphere liquids: a real space description [32, 33], mean-field approximation [31, 32] and exact calculations in infinite dimensions [33, 34]. (The discussion below may apply to other liquids by replacing the packing fraction \( \phi \) by \( T \).) It is found that some elastic modes become stable only above an onset packing fraction \( \phi_0 \) [33, 35, 34]. For \( \phi < \phi_0 \), one expects flow to be fast as the system can relax without jumping over free energy barriers along unstable directions of phase space. By contrast, flow must be activated for \( \phi > \phi_0 \). This phenomenon is captured in part by the mode-coupling theory of liquids and by mean-field theory [4, 41, 42]. One expects a growing dynamical length scale at such intermediate \( \phi \) because the last modes to remain unstable as \( \phi \) increases are increasingly collective. For larger \( \phi \), the simplest guess is that activated elementary rearrangements occurs along similar modes to those whose stability is exchanged at \( \phi_0 \). This is consistent with the observation that the dynamical length scale almost saturates (at the scale of collective motions of a few tens of particles) once \( \tau_\alpha \) has increased by a few orders of magnitude [44].

This line of thought is consistent with the observations of swap algorithms. Indeed if elementary rearrangements become more collective because local moves are too costly, then dynamical correlations can disappear if effective local moves such as radius-swaps are allowed for. Alongside much faster relaxation, dynamical correlations near the mode coupling temperature should then be eliminated. This is indeed observed: dynamical correlations become very small when swaps are used [22]. Furthermore, dynamical correlations near \( T_G \) also become very small. This observation undermines the RFT result that dynamical heterogeneities near \( T_G \) in liquids reflect \( \zeta \) [4], since \( \zeta \) is unaffected by the choice of dynamics. Instead it supports the view that for non-swap dynamics, elementary rearrangements are spatially more extended than \( \zeta \), further indicating that \( T_G \) lies outside the temperature range where RFOT might apply.

Another point to clarify concerning elastic models is why \( G_\infty \) increases under cooling. From that perspective, the exchange of stability of some elastic modes at \( \phi_0 \) can be shown to lead to an increase in shear modulus under cooling [4, 33, 15], and is thus consistent with mean-field approaches. Other factors susceptible to stiffen the material may be very system-specific, including the previously mentioned growth of locally-favored structures [10, 11].

A promising avenue that appears consistent with the swap-algorithm observations would thus be to combine elastic models with a more detailed description of collective dynamical modes beyond their linear regime. This could also account for other key facts of the dynamics in liquids, in particular the presence of growing dynamical length scales, the correlations between activation energy and high frequency shear modulus, and the correlations between entropy and dynamics [46].

**Conclusion.** The recent observation that swap algorithms can essentially eliminate the glass transition without changing the free energy landscape casts doubt on theories in which a growing static length scale, determined solely by this landscape and setting a minimum scale for finding new density patterns, controls the slowdown of relaxations in glasses. Instead, these observations cap the contribution from this source at a few decades of growth in the structural relaxation time, with a much larger factor arising instead from the growing barriers to local rearrangement. This concurs with a similar cap derived from the Stokes Einstein violation for real glasses which is likewise only a few orders of magnitude, requiring tagged particle diffusion (asymptotically unaffected by the growth of \( \xi \)) and structural relaxation to remain strongly coupled near the glass point. Among theories of the local barrier physics, a combination of elastic models with an improved description of collective dynamic modes may offer a promising route forward.

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Note that in [23], one may expect a transition for swap dynamics at some

T_{MCT} \rightarrow T_{MCT}^* > T_{MCT}^*.

Indeed for the swap dynamics stability is more demanding since radii can smoothly evolve (if polydispersity is continuous). This change of fragility can be deduced from the power law fit reported between \( \tau_D \) and \( \tau_\alpha \) in [23, 27].