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 τ_α is controlled by a growing static length scale ξ 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 τ_α 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.

When a liquid is cooled sufficiently rapidly to avoid crystallization, its viscosity η increases. In general, $\eta \simeq G\tau_\alpha$, where G is the (plateau) shear modulus and τ_α characterizes the relaxation time of density fluctuations. As the temperature T is lowered, G evolves mildly but τ_α increases by about 15 order of magnitude [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 slows down. The time τ_D over which a particle diffuses its own radius increases, albeit not as much as τ_α. The decoupling of these two quantities (the Stokes-Einstein breakdown) is significant, but comparatively mild: $\mathcal{S} = \tau_\alpha/\tau_D$ is increased by only a few orders of magnitude at T_G [2, 3].

The dependence of τ_α on T is used to classify glassy liquids [4]. Writing $\tau_\alpha = \tau_0 \exp(E/k_B T)$ where τ_0 is a vibrational time (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 [4], which is linear for strong liquids but highly curved for fragile ones. Quantitatively, fragility is defined as $m = d\log(\tau_\alpha)/d\log(T_G/T)$ evaluated at T_G itself. Strong liquids have $m \approx 25$; very fragile ones have $m \approx 120$.

There are competing explanations of the increase in the activation energy E in fragile liquids, which occurs with no obvious change of static structure [5]. Several theories, including the Adam-Gibbs scenario [6], Random First Order Theory (RFOT) [7–9], and those involving locally favored structures [10, 11], posit that this increase stems from the growth of a purely static length scale ξ, characterizing 'hidden order' in the many-body free-energy landscape, not captured by traditional probes of static structure such as pair correlations.

Specifically, in modern interpretations of RFOT [7, 8], ξ 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 density pattern which keeps returning to its ini-

tial state. In this view, regardless of how rapidly these local moves permute the particles within the patch, τ_α is controlled by the fact that the system can relax fully its density fluctuations only via collective rearrangements on the scale ξ, 'breaking' the hidden order. The resulting collective activation energy is $E_{\text{coll}} \simeq c_0(T)\xi(T)^\psi$, where $c_0(T) \simeq c_0(T_G)$ is non-singular, and ψ is some exponent. It can be expressed in terms of thermodynamic quantities alone and is thus independent of the details of the dynamics [48] (see below).

Although there is clear empirical evidence for a growing static length scale $\xi(T)$ (e.g., [12] and references therein), its role in the dynamics is debated. Alternative theories propose that the increase of τ_α stems from growing barriers to the elementary rearrangements required to explore the landscape [13–17]. Such barriers effectively add a term to the activation energy $E_{\text{el}} = c_1(T)$. Unlike E_{coll}, these kinetic barriers can depend greatly on the detailed dynamical rules governing the system. While the definition of 'elementary rearrangements' can be multi-particle (see below), E_{el} describes local physics and cannot diverge. However it might grow strongly enough near T_G to control the glass transition via $\tau_\alpha \propto \tau_{\text{el}} \simeq \tau_0 \exp[E_{\text{el}}/k_B T]$. For example in elastic models [13], rearrangements require a certain strain, giving $E(T) \sim G_\infty(T)$ where G_∞ is the high-frequency (vibrational) shear modulus. Empirically these quantities are indeed strongly correlated [13, 14], as seen by plotting $\log(\tau_\alpha)$ *vs.* $(T_G G_\infty(T))/(T G_\infty(T_G))$, which appears almost perfectly linear even for fragile liquids [14, 18].

Combining the two activation terms gives $\ln(\tau_\alpha/\tau_0) \simeq a_0 \xi^\psi + a_1$, 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 τ_α [19]; this bound shows that a *diverging* ξ is required for an ideal glass transition ($\tau_\alpha/\tau_0 \to \infty$). It is then tempting to conclude that *growth* of ξ 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 τ_α 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 that E_{el} rather than E_{coll} is dominant in controlling τ_α. 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 $\mathcal{S} \geq \exp[E_{coll}/k_B T] = \tau_\alpha/\tau_{el}$. It follows that the growth of ξ contributes only a few decades at most to the 15 decades increase in τ_α on the approach to T_G. Our findings are depicted schematically in Fig.1.
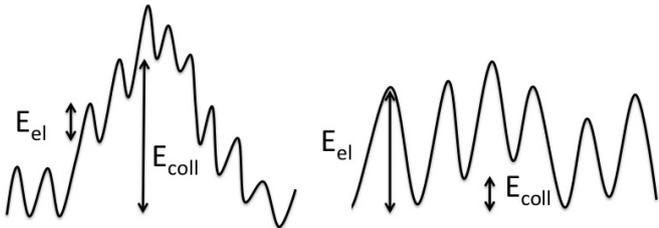
FIG. 1: Left panel: schematic of the free energy landscape $F(\{\mathbf{r}\})$ for models where the dominant kinetic barriers E_{coll} are collective, and appear on a length scale ξ. Local barriers E_{el} are much smaller than E_{coll}. In this scenario introducing swap moves in the dynamical rules should 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 ξ may become somewhat arbitrary.)

Swap algorithms. Recent Monte-Carlo schemes have successfully equilibrated liquids at temperatures where traditional algorithms would need at least a 10^{10} speed-up to allow equilibration [20–25]. These swap algorithms consider polydisperse particles. In addition to the usual translational moves, they allow 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). Particles sizes are then 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 particle positions $\{\mathbf{r}\}$ is, after integration over sizes, well defined and independent of the chosen dynamical rules [49]. The swap rules can involve either well-separated particles as in [22, 23], or be local, for instance restricted to nearest neighbour swaps [26]. For the dynamical rules of [22, 23], the time scale τ_α^+ on which density fluctuations relax with swaps [50] can be compared to the value τ_α^- 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: $\tau_\alpha^+(T_G)/\tau_0$ is only around $10^2 - 10^3$. This contrasts with $\tau_\alpha^-/\tau_0 \simeq 10^{15}$ 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 τ_α [23], as we justify in SI. This suggest that either type of swap dynamics can mollify local barriers, leading to much faster relaxation than the conventional dynamics of real glasses. It follows that the latter is dominated by E_{el}. Indeed, switching from standard to swap dynamics simply enhances the local move set: swapping is a specific local move involving two particles only [51]. The underlying free-energy landscape $F(\{\mathbf{r}\})$ is unaffected. As such, so long as one first equilibrates the system, any static thermodynamic length ξ must be identical with both dynamics. Indeed, a major selling-point of the swap algorithms is that they *allow full equilibration* over an unprecedentedly wide temperature range. We further consider (shown below) that for large enough τ_α the collective barriers E_{coll} must be identical for swap and non-swap dynamics. We can now compare the dynamics with $(+)$ and without $(-)$ swaps. Since $\tau_\alpha^\mp = \tau_{el}^\mp \exp[E_{coll}(\xi)/k_B T]$, if glass physics is dominated by the growing length scale ξ, then $\tau_\alpha^-/\tau_\alpha^+$ should depend only weakly on T: the only such dependence is through the factor τ_{el}^-/τ_{el}^+. But in the simulations, $\tau_\alpha^-/\tau_\alpha^+$ increases without apparent limit as T is lowered: this increase can be tracked for about 4 decades before it becomes too large to measure [22, 23]. In contrast, the growth of τ_α^+, which bounds above the slowing down caused by the growth of ξ, only shows a 2 or 3 decade increase at T_G. However, it continues to grow by several more decades as T is further decreased.

We offer the following interpretation for these results. The relaxation time τ_α^+ describing the with-swap dynamics, for which local barriers are mollified, may ultimately be controlled by the growth of ξ, although other effects are plausible [52]. However, with the swap-free dynamics relevant to real glasses, the resulting contribution to τ_α^-/τ_0 is smaller than 3 decades at the glass transition. Thus the lion's share of the growth in τ_α stems not from a growing static length but from growing local barriers. Only these barriers are affected by introducing local swaps, so that the resulting collapse of the glass transition points directly to their dynamical importance.

Stokes-Einstein violation. The arguments above are restricted to polydisperse systems, as simulated by radius-swap algorithms. However for conventional swap-free dynamics the relaxation in these systems is no different from classical numerical models of structural glasses [22, 23]. This suggests that polydispersity is immaterial to whether a growing static length controls 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 ξ. Recall that in this approach, each configuration is a mosaic of states, whose character-

istic size ξ 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 $c_0 r^\psi$: the dynamics is *fast* on short length scales, but slow on long ones. However for $r < \xi$, a single state is thermodynamically favored. Local rearrangements are possible, but will not fully relax density fluctuations. Processes that can relax density fluctuations occur on a scale ξ, the smallest on which alternative density patterns can appear [8].

We believe that this picture contradicts the observation that in real glasses the Stokes Einstein (SE) factor $\mathcal{S} = \tau_\alpha/\tau_D$ at T_G is of order 10^3 [3, 27], rather than much larger. 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 $\mathcal{S}$: any other mechanism found to contribute to the SE violation therefore bounds it below. We may therefore write $\mathcal{S} = \mathcal{S}_1\mathcal{S}_2 \geq \mathcal{S}_1$, where $\mathcal{S}_2$ accounts for DH, and $\mathcal{S}_1$ allows the possibility that particles can exchange positions while leaving density fluctuations unchanged [29]. We now argue that $\mathcal{S}_1 \sim \exp[a_1\xi^\psi]$, 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 permutations, 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 $\tau_D \simeq \tau_0 \exp[E_{\rm el}/k_B T] = \tau_\alpha/\mathcal{S}_1$.

We then have $\mathcal{S}_1 = \exp[a_1\xi^\psi] < \mathcal{S} \simeq 10^3$, implying that the growth of ξ cannot account for more than about a 3 decade increase in τ_α at T_G. Were the glass transition controlled purely by growth of ξ, a much stronger decoupling of τ_D and τ_α could be expected. This is incompatible with the fact that the fragility defined from τ_D is almost as large as that defined from τ_α (only 13-25% [53] 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, pointing to a major role for $E_{\rm el}$ 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 ξ.

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 ξ) local equilibrium in the landscape $F(\{\mathbf{r}\})$ should be reached, on all length scales shorter than ξ, at time scales much smaller than τ_α. 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 τ_α, 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 τ_α. This implies in turn that $E_{\rm coll}$ 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 ξ, local rearrangements are rapid in comparison to τ_α, 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 τ_α 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 ξ 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 $E_{\rm el}$. For example, in the spirit of elastic models, locally favored structures that grow under cooling [10, 11] could increase G_∞, 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 τ_α does [19] is of limited practical relevance for real liquids near their glass transition, since the proper signal for this divergence is τ_α/τ_D which increases relatively mildly within the experimental range. (Moreover, ξ need only diverge as $(\ln(\tau_\alpha/\tau_D))^{1/\psi}$.) 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 $(T_G G_\infty(T))/(T G_\infty(T_G))$ [14, 18].

On the other hand, with local barriers mollified by swap dynamics [22, 23], the growth of ξ could become paramount, and RFOT is possibly the correct theory of such dynamics. For realistic dynamics however, theories of local barriers appear necessary to describe the glass transition. One such theory is provided by elastic models [13, 14], for which $E_{\rm el}$ is proportional to G_∞. 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 rear-

rangements 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 [34, 35] and exact calculations in infinite dimensions [36–38]. (The discussion below may apply to other liquids by replacing the packing fraction ϕ by T.) It is found that some elastic modes become stable only above an onset packing fraction ϕ_0 [33, 38, 39]. For $\phi < \phi_0$, one expects flow to be fast: the system can relax without crossing 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 [7, 40–43]. One expects a growing *dynamical* length scale at such intermediate ϕ because the last modes to remain unstable as ϕ increases are increasingly collective. For larger ϕ, the simplest guess is that activated elementary rearrangements occurs along similar modes to those whose stability is exchanged at ϕ_0. This is consistent with observations that the dynamical length scale almost saturates (at the scale of collective motions of a few tens of particles) once τ_α 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. 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 RFOT result that dynamical heterogeneities near T_G in liquids reflect ξ [5], since ξ is unaffected by the choice of dynamics. Instead it supports the view that for non-swap dynamics, elementary rearrangements are spatially more extended than ξ, further indicating that T_G lies outside the temperature range where RFOT might apply.

Another point to clarify concerning elastic models is why G_∞ increases under cooling. From that perspective, the exchange of stability of some elastic modes at ϕ_0 can be shown to lead to an increase in shear modulus under cooling [7, 35, 45], and is thus consistent with mean-field approaches. Other factors that 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 ξ) 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.

[1] M. D. Ediger, C. A. Angell, and S. R. Nagel, J. Phys. Chem. **100**, 13200 (1996).

[2] M. D. Ediger, Annual Review of Physical Chemistry **51**, 99 (2000).

[3] S. F. Swallen, K. Traynor, R. J. McMahon, M. Ediger, and T. E. Mates, The Journal of Physical Chemistry B **113**, 4600 (2009).

[4] C. Angell, Journal of Non-Crystalline Solids **131**, 13 (1991).

[5] L. Berthier and G. Biroli, Reviews of Modern Physics **83**, 587 (2011).

[6] G. Adam and J. H. Gibbs, The Journal of Chemical Physics **43**, 139 (1965).

[7] V. Lubchenko and P. G. Wolynes, Annual Review of Physical Chemistry **58**, 235 (2007).

[8] J.-P. Bouchaud and G. Biroli, The Journal of Chemical Physics **121**, 7347 (2004).

[9] T. Kirkpatrick and D. Thirumalai, Reviews of Modern Physics **87**, 183 (2015).

[10] D. Kivelson, S. A. Kivelson, X. Zhao, Z. Nussinov, and

G. Tarjus, Physica A: Statistical Mechanics and its Applications **219**, 27 (1995).

[11] H. Tanaka, The European Physical Journal E **35**, 113 (2012).

[12] S. Albert, T. Bauer, M. Michl, G. Biroli, J.-P. Bouchaud, A. Loidl, P. Lunkenheimer, R. Tourbot, C. Wiertel-Gasquet, and F. Ladieu, Science **352**, 1308 (2016).

[13] J. C. Dyre, Reviews of Modern Physics **78**, 953 (2006).

[14] D. H. Torchinsky, J. A. Johnson, and K. A. Nelson, J. Chem. Phys. **130**, 064502 (pages 11) (2009).

[15] S. Mirigian and K. S. Schweizer, The Journal of chemical physics **140**, 194506 (2014).

[16] J. P. Garrahan, P. Sollich, and C. Toninelli, Dynamical heterogeneities in glasses, colloids, and granular media **150**, 111 (2011).

[17] A. Lemaître, Physical review letters **113**, 245702 (2014).

[18] T. Hecksher and J. C. Dyre, Journal of Non-Crystalline Solids **407**, 14 (2015).

[19] A. Montanari and G. Semerjian, Journal of statistical physics **125**, 23 (2006).

[20] L. Berthier, D. Coslovich, A. Ninarello, and M. Ozawa, Physical Review Letters **116**, 238002 (2016).

[21] R. Gutiérrez, S. Karmakar, Y. G. Pollack, and I. Procaccia, EPL (Europhysics Letters) **111**, 56009 (2015).

[22] A. Ninarello, L. Berthier, and D. Coslovich, arXiv preprint arXiv:1704.08864 (2017).

[23] L. Berthier, P. Charbonneau, D. Coslovich, A. Ninarello, M. Ozawa, and S. Yaida, arXiv preprint arXiv:1704.08257 (2017).

[24] T. S. Grigera and G. Parisi, Physical Review E **63**, 045102 (2001).

[25] L. Fernández, V. Martin-Mayor, and P. Verrocchio, Physical Review E **73**, 020501 (2006).

[26] L. Berthier, Private communication (2017).

[27] S. F. Swallen and M. Ediger, Soft Matter **7**, 10339 (2011).

[28] G. Tarjus and D. Kivelson, The Journal of chemical physics **103**, 3071 (1995).

[29] P. Charbonneau, Y. Jin, G. Parisi, and F. Zamponi, Proceedings of the National Academy of Sciences **111**, 15025 (2014).

[30] J. P. Garrahan and D. Chandler, Phys. Rev. Lett. **89**, 035704 (2002).

[31] C. Toninelli, G. Biroli, and D. S. Fisher, Phys. Rev. Lett. **98**, 129602 (2007).

[32] C. Brito and M. Wyart, EPL (Europhysics Letters) **76**, 149 (2006).

[33] C. Brito and M. Wyart, The Journal of Chemical Physics **131**, 024504 (2009).

[34] E. DeGiuli, E. Lerner, C. Brito, and M. Wyart, Proceedings of the National Academy of Sciences **111**, 17054 (2014).

[35] E. DeGiuli, A. Laversanne-Finot, G. A. Düring, E. Lerner, and M. Wyart, Soft Matter **10**, 5628 (2014).

[36] S. Franz, G. Parisi, P. Urbani, and F. Zamponi, Proceedings of the National Academy of Sciences **112**, 14539 (2015).

[37] A. Altieri, S. Franz, and G. Parisi, Journal of Statistical Mechanics: Theory and Experiment **2016**, 093301 (2016).

[38] T. Maimbourg, J. Kurchan, and F. Zamponi, Physical Review Letters **116**, 015902 (2016).

[39] P. Charbonneau, J. Kurchan, G. Parisi, P. Urbani, and F. Zamponi, Nature Communications **5** (2014).

[40] P. Rabochiy, P. G. Wolynes, and V. Lubchenko, The Journal of Physical Chemistry B **117**, 15204 (2013).

[41] T. S. Grigera, A. Cavagna, I. Giardina, and G. Parisi, Phys. Rev. Lett. **88**, 055502 (2002).

[42] S. Franz and A. Montanari, J. Phys. A **40**, F251 (2007).

[43] A. Andreanov, G. Biroli, and J.-P. Bouchaud, EPL (Europhysics Letters) **88**, 16001 (2009).

[44] C. Dalle-Ferrier, C. Thibierge, C. Alba-Simionesco, L. Berthier, G. Biroli, J.-P. Bouchaud, F. Ladieu, D. L'Hôte, and G. Tarjus, Phys. Rev. E **76**, 041510 (2007).

[45] D. Nakayama, H. Yoshino, and F. Zamponi, Journal of Statistical Mechanics: Theory and Experiment **2016**, 104001 (2016).

[46] M. Wyart, Phys. Rev. Lett. **104**, 095901 (2010).

[47] H. Jacquin, B. Kim, K. Kawasaki, and F. Van Wijland, Physical Review E **91**, 022130 (2015).

[48] This statement holds true if the order parameter Q on a correlation volume ξ^d evolves smoothly at each numerical step, as it ensures that configurations with $Q = Q^*$ for which the free energy barrier is maximum are visited. This must be true for any Monte-Carlo where moves involves n_0 particles with $n_0 << \xi^d$.

[49] Here $F(\{\mathbf{r}\}) = -k_B T \ln(Z(\{\mathbf{r}\}))$ where $Z(\{\mathbf{r}\}) = \sum_\sigma \exp(-\beta U(\{r\}, \{\sigma\}))$. The sum is over all choices of particle radii consistent with the imposed polydispersity, and $U(\{r\})$ is the total potential energy.

[50] Note that in [23], τ_D and τ_α are both treated as one-particle properties. Here we use τ_α to describe collective density relaxations.

[51] A different instance of local dynamical rules profoundly affecting the glass transition is discussed in [47].

[52] One may expect a transition for swap dynamics at some temperature T_{MCT}^+ where local minima appear in $F(\{\mathbf{r}\})$ with $T_{MCT}^- > T_{MCT}^+$. Indeed for the swap dynamics stability is more demanding since radii can smoothly evolve (if poly-dispersity is continuous).

[53] This change of fragility can be deduced from the power law fit reported between τ_D and τ_α in [3, 27].