Dynamical Phase Transitions and their Relation to Thermodynamic Glass Physics

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We review recent developments in structural-dynamical phase transitions in trajectory space. An open question is how the dynamic facilitation theory of the glass transition may be reconciled with thermodynamic theories that posit a vanishing configurational entropy. Dynamic facilitation theory invokes a dynamical phase transition, between an active phase (close to the normal liquid) and an inactive phase which is glassy, whose order parameter is either dynamic or a time-averaged structural quantity. In particular, the dynamical phase transition in systems with non-trivial thermodynamics manifests signatures of a lower critical point, which lies close to the putative Kauzmann temperature, where any thermodynamic phase transition to an ideal glass state might occur. We discuss these findings, and suggest that the lower critical point of the structural-dynamical phase transition may be related to the large drop in configurational entropy that occurs in the inactive phase of the dynamical phase transition. Increasing supercooling thus brings configurational entropy of the normal liquid much lower, along with the temperature.

I. INTRODUCTION

Understanding the physical origin of the glass transition is a longstanding challenge in condensed matter physics. Cool any liquid sufficiently fast and it will not order into a crystal but remain a liquid before, eventually, it falls out of equilibrium and becomes a glass. The single most important quantity when talking about supercooled liquids is the structural relaxation time $\tau_\alpha$, which measures the average time over which atoms and molecules rearrange such as to lose memory of their initial positions. Over time, a variety of theoretical approaches have been developed to account for the massive slowdown ($\tau_\alpha$ increases by 14 orders of magnitude) in the dynamics of such supercooled liquids as they approach the experimental glass transition temperature $T_g$[1,3]. Compared to crystallisation, little change in structure assumed by the constituent particles occurs during this process, suggesting that, while the origin of the dynamic slowdown could be due to a phase transition related to some kind of change in amorphous order or structure[2], the case that the glass transition is a predominantly dynamical phenomenon is compelling.

The early stages of glassy dynamics (i.e. the first few decades of increase in relaxation time with supercooling) are accurately described by mode-coupling theory (MCT)[4,6]. As input, MCT calculations utilize structural two-point correlations and provide evolution equations for dynamic two-point correlations. In high dimension, a true dynamical transition reminiscent of MCT is found[2]. However, in lower dimensions, the MCT approach fails when many-body correlations become important for relaxation, which occurs typically after 4-5 decades of increase in structural relaxation time$[6,10]$. Recently, a generalised MCT approach has provided a route to address this limitation, although the Herculean task of solving the higher-order coupled equations should not be underestimated[11].

At deeper supercooling, past the (avoided) mode-coupling transition, random first order transition theory (RFOT)[7] and Adam-Gibbs theory[8] both feature a vanishing configurational entropy implying a corresponding divergent static correlation length. The qualitative picture is that of cooperative motion of more and more particles in order for the liquid to relax. Approaches based on replica theory imagine two coupled copies or “replicas” of the same system. At a sufficient degree of supercooling, a phase transition occurs to states which feature a high degree of structural similarity – or overlap upon the application of sufficient coupling[9,10]. Other theories, such as geometric frustration, emphasize locally favoured – or locally preferred – structures (LFS)[9]. These locally favoured structures are geometric motifs that are minima of the local (free) energy. Their concentration appears to increase as a glass-former is cooled down, and they have been identified with emergence of slow dynamics[4]. The geometric frustration theory imagines an avoided phase transition to a state of LFS which would occur e.g. in curved space[9].

II. DYNAMICAL PHASE TRANSITIONS

Here we focus in some detail on an alternative scenario, dynamical facilitation theory[10] in which dynamic arrest is attributed to emerging kinetic constraints. This theoretical approach focuses on the role of real-space fluctuations and dynamic heterogeneities[17], i.e., the coexistence of mobile and jammed regions in a supercooled liquid, at the expense of thermodynamic and structural
FIG. 1. Dynamic heterogeneity and trajectories. Main panel: Dynamic heterogeneity in a simulation of binary hard discs with a size ratio of 1:1.4. Blue particles have moved the least (mean squared displacement $\langle r^2 \rangle < 0.01\sigma^2_{\text{large}}$) and red have moved the most ($\langle r^2 \rangle > \sigma^2_{\text{large}}$). Here the timescale is taken over the structural relaxation time $\tau_\alpha$. Indicated are active and inactive trajectories (the active trajectory looks static).

mechanisms. Like several competing theories, dynamic facilitation is built around the idea of a phase transition, but here its nature is profoundly different: In contrast to conventional thermodynamic phase transitions, where the coexisting phases are characterised by distinct static properties (e.g. the density difference in liquid-gas coexistence), dynamical facilitation starts from the observation that in the supercooled liquid one encounters regions that relax (warm-coloured regions in Fig. 1) on the timescale of $\tau_\alpha$, and some that do not (blue regions in Fig. 1), both of which coexist. The kinetic constraints govern the effective dynamics of localized excitations that sustain motion within the mobile regions.

To be more specific, let us decompose a large system into distinct subsystems containing a few hundreds of particles. Following the dynamics for an observation time $t_{\text{obs}}$ on the order of a few structural relaxation times, we will find that the resulting trajectories performed by some subsystems will display large cumulative displacements, while others will show very little change in their particle positions. Such a behaviour can be interpreted as the manifestation of a dynamical phase transition between a relatively fast-moving (active) state of trajectories and a slow-moving (inactive) state (cf. the trajectories in Fig. 1). In a suitable dynamical ensemble, it can be shown that a genuine dynamical coexistence can be established, and that a bimodal distribution of active and inactive trajectories emerges.

In a series of papers, idealised lattice models of supercooled liquids, so-called kinetically constrained models (KCMs) have been shown, through both analytical and numerical calculations, to undergo such a dynamical phase transition between active and inactive trajectories. Such models essentially neglect the detail of particle-particle interactions, focusing on the hindrance to relaxation that is a distinctive feature of supercooled liquids. This is represented through simple and idealised on-lattice rules for the motion of particles or the relaxation of spin excitations. The Hamiltonian of such models is often designed to be trivial, yet they exhibit highly nontrivial, glassy dynamics, in particular faster than exponential (i.e. super-Arrhenius) increase of the relaxation time with decreasing temperature and dynamic heterogeneities. The fact that these idealized models exhibit much of the phenomenology of kinetic arrest in liquids (such as super-Arrhenius relaxation and dynamical heterogeneity) provides strong evidence that a thermodynamic transition – absent here by construction – is not necessarily required.

Regardless of the detail of their respective kinetic rules, kinetically constrained models show dynamical phase transitions of the kind illustrated above and in Fig. 1. Crucially, dynamical phase coexistence can be observed only if an external field (often denoted by $s$) is applied, $s_{\text{coex}}(T) > 0$ for any value of the temperature $T$. A formal understanding of the resulting $s$-ensemble relies
on the application of theory of large deviations to non-equilibrium steady states, where the transition is determined from the non-analyticities of cumulant generating functions that can be interpreted as the non-equilibrium analog of free energies\cite{20}. Interestingly, such dynamical transitions seem to be robust to the application of external driving forces that break detailed balance\cite{27,29}.

At this point it is helpful to recall that both thermodynamic approaches and dynamic facilitation are “qualitative theories” that aim to capture the dominant mechanism through which relaxation in supercooled liquids is hampered. They are not first-principle microscopic theories that yield explicit expressions, which makes it hard to discriminate both approaches based on experimentally accessible data and computer simulations. Their simultaneous success thus leaves us with a conundrum: how can the observed — albeit subtle — structural changes occur— and why? Evidence in support of each approach has been presented. For example, several recent studies\cite{27} have shown that the deeply supercooled state presents a low configurational entropy that monotonically decreases with temperature as expected in the Adam-Gibbs/RFOT scenario, both in advanced Monte-Carlo simulations\cite{31} and colloidal experiments\cite{32,33}, as well as a growing static length scale in experiments on molecules\cite{34}.

On the other hand, support for dynamic facilitation comes from computer simulations of atomistic models (including three dimensional Lennard-Jones binary mixtures and hard spheres\cite{35,36}) which are less idealized than KCMs and in which kinetic constraint are not present by construction but emerge from interparticle forces. For instance, the famous Kob-Andersen binary Lennard-Jones model\cite{37} was found to undergo a dynamical transition between a relatively fast-moving active state and a very slow-moving inactive state, i.e. the behaviour indicated in Figs. 1 and 2. Some success was found even in experiments, where KCMs were shown to explain calorimetric effects in the glass transition\cite{38}, while colloids exhibit the dynamical phase transition of Fig. 3\cite{39}.

In the following, we review recent work on the role of local structure and its implications for a possible route to reconcile both theoretical approaches. It is based on the observation that in atomistic models of glass formers kinetic constraints are emerging (coarse-grained) interactions that are necessarily accompanied by a structural signature. This implies (weak) spatial interactions between excitations absent in idealized KCMs. Such a lower critical point provides a mechanism for the emergence of a correlation length that is compatible with dynamic facilitation. Numerical evidence so far has been obtained for three different model glass formers: Kob-Andersen\cite{19,20}, Wahström\cite{21} (both are binary mixtures with Lennard-Jones pair potentials), and polydisperse hard spheres\cite{32,33}.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3}
\caption{Phase transitions in trajectory space for the Kob-Andersen binary Lennard-Jones mixture. Left column: $s$-ensemble. (a) Probability distributions $p(c)$ for the density of mobile particles $c$ for two trajectory lengths. The non-concave shape indicates a phase transition in trajectory space as becomes obvious from the bimodal distribution (b) at the field $s^*$ that maximises the fluctuations $(c^2_s) - \langle c_s \rangle^2$. (c) Average fractions of mobile particles (solid lines) and bicapped square antiprism cluster population (dashed lines) vs. the biasing field $s$. Right column: (d-f) as left column but for the $\mu$-ensemble. Here the bicapped square antiprism (depicted in (f)) is the locally favoured structure. Throughout, red and blue lines refer to trajectory lengths $t_{\text{obs}} \approx 5\tau_\alpha$ and $t_{\text{obs}} \approx 10\tau_\alpha$, respectively. Reproduced with permission from Ref. 19.}
\end{figure}

### III. COUPLING OF LOCAL STRUCTURE AND GLOBAL DYNAMICS: THE $\mu$-ENSEMBLE

The first link that we want to establish is between the dynamical phase transitions of dynamical facilitation and the structural changes observed in the liquid. Is there any relation between the glassy trajectories of the inactive dynamical phase and the emergence of local structural order?

To answer this question and in the spirit of statistical mechanics, we seek to define order parameters that quantify the corresponding behavior. Specifically, these are:

\begin{equation}
C[X] = \sum_{t=1}^{t_{\text{obs}}} N\dot{c}(t-1,t), \quad N[X] = \sum_{t=0}^{t_{\text{obs}}} N\dot{u}(t),
\end{equation}

measuring the time-averaged population $C$ of mobile par-
trajectories, introduce external fields. To elucidate the conceptual behavior, we introduce Gaussian [cf. Fig. 3(a,d) for the binary Kob-Andersen mixture]. We also define the fractions $s = C/(Nt_{\text{obs}})$ and $n = N/[N(t_{\text{obs}}+1)]$ which take on values between zero and unity. The crucial feature is that the order parameters are extensive both in space and time.

At first glance, as shown in Fig. 3, the probability distributions of these fractions are somewhat unremarkable; they appear as Gaussians as dictated by the central limit theorem, reflecting fluctuations in the supercooled liquid. However, pushing into the tails of the distributions (low values of $C$ and high values of $N$) we soon realize that they decay much slower than what would correspond to a Gaussian [cf. Fig. 3(a,d) for the binary Kob-Andersen mixture]. To elucidate the conceptual behavior, we introduce external “fields” $s$ and $\mu$ which promote rare trajectories,

$$\langle C \rangle_s = \frac{1}{Z(s)} \langle C e^{-sC} \rangle, \quad \langle N \rangle_\mu = \frac{1}{Z(\mu)} \langle N e^{\mu N} \rangle$$

with corresponding “dynamic” partition functions $Z$ ensuring normalization. For obvious reasons these two ensembles are dubbed $s$-ensemble and $\mu$-ensemble. Eq. (2) is a form of importance sampling. The theory behind it connects to the mathematics of large deviations and will not be reviewed here. Sampling sufficiently many trajectories in computer simulations at non-vanishing fields is a challenge that requires advanced sampling techniques. Two candidates are transition path sampling and cloning algorithms. Results shown here have been obtained from a combination of transition path sampling with replica exchange, for details see.

Numerical curves corresponding to Eq. (2) are plotted in Fig. 3(c,f) and show that there is a qualitative change as we increase both $s$ and $\mu$ which becomes more abrupt as the trajectory length is increased. The corresponding susceptibilities peak at $s^*$ and $\mu^*$. These are hallmarks for a phase transition, which here occurs in the space of trajectories. Fig. 4 shows the joint distribution of $n$ and $c$ for different values of the fields, for which we can discern two basins separated by a barrier. This demonstrates that the transition occurs between a phase that has many mobile particles and a low occupation of LFS (which we identify with the normal supercooled liquid), and a phase with very few mobile particles and a high population of LFS. The later phase shows properties normally associated with a glass. This interpretation is further supported by looking at the reweighted marginal distributions at $s^*$ and $\mu^*$ [Fig. 3(b,e)], which exhibit two peaks. Both phases are also termed active and inactive, respectively. In particular, the $\mu$-ensemble probes the active-inactive transitions explored in the $s$-ensemble: inactive trajectories correlate strongly with trajectories rich in locally favoured structures.

In Fig. 5(b), we plot the reweighted distribution of $n$ for another model system, polydisperse hard sphere. We now have a closer look at the trajectories populating the first peak (red shaded area, LFS-poor) and the second peak (blue shaded area, LFS-rich). As shown in Fig. 5(c), the static structure as measured by the pair distribution function is very similar (note, however, the splitting of the second peak). In contrast, the dynamics as measured by the intermediate scattering function [ISF, Fig. 5(d)] and the mean-square displacement [Fig. 5(e)] are different, demonstrating that the average dynamics in the LFS-poor phase is “fast” while in the LFS-rich phase it is much slower. There is thus again a strong correlation between the overall population of LFS and the global dynamics.

Experimental evidence, using colloidal suspensions, has now been found for this structural and dynamical phase transition. More recent studies suggest that the inactive/locally favoured structure-rich phase obtained in the space of trajectories also correlates strongly with particularly low (potential) energy states, and that when decreasing the temperature, the inactive, LFS-rich phases tend to dominate the statistics, with the coexistence values of $s^*$ and $\mu^*$ approaching zero as the temperature is decreased. In this sense, guiding trajectory sampling with the usage of time-integrated observable can be an efficient way to identify low energy states, more present in the arrested glassy phases.
and LFS-rich (blue) region of P(μ∗) obtained from trajectories sampled within the LFS-poor (red) and glass peak (blue). (b) Radial distribution functions obtained from trajectories sampled within the LFS-poor (red) and LFS-rich (blue) region of P(n). Arrows indicate the positions of peaks. (c) Intermediate scattering function and (d) mean-squared displacement for the two phases. Reproduced from Campo, M. and Speck, T. J. Chem. Phys., 152 014501 (2020), with the permission of AIP Publishing.

FIG. 5. Trajectory properties in hard spheres. (a) Distribution function Pp,n(μ∗) of the population of LFS at coexistence. Shaded areas indicate the region around the liquid peak (red) and glass peak (blue). (b) Radial distribution functions obtained from trajectories sampled within the LFS-poor (red) and LFS-rich (blue) region of P(n). Arrows indicate the positions of peaks. (c) Intermediate scattering function and (d) mean-squared displacement for the two phases. Reproduced from Campo, M. and Speck, T. J. Chem. Phys., 152 014501 (2020), with the permission of AIP Publishing.

IV. REWEIGHTING FOR DEEPLY SUPERCOOLED CONFIGURATIONS

These ideas have been recently tested in the case of a canonical atomic glass former (the Kob-Andersen binary mixture), and it has been shown that other systems exhibit similar behaviour, such as hard spheres and the Wahnström binary Lennard-Jones model. In particular, it has been shown that the large deviations of time-integrated structural observables give access to low energy configurations that sample the tails of the probability distribution of inherent state energies. Through reweighting, this sampling recovers the thermodynamical properties of the system (such as the configurational entropy) down to very low temperatures, without the need of sampling the dynamics at low temperatures directly.

Additionally, this approach shows that the dynamical phase transition between trajectories poor/rich in structure sampled in the trajectory ensemble corresponds to a transition between two distinct metastable amorphous states at high/low inherent state energies respectively: one corresponds to the supercooled liquid sampled in conventional dynamics; the other to a secondary amorphous state, with low energy, low configurational entropy, rich in structure and very slow dynamics, see Fig. 6. This second amorphous state is more metastable than the conventional supercooled liquid: however, the difference in stability (as measured by the value μ∗ of the conjugated field μ at coexistence between the two phases) is a function of the temperature and decreases as the temperature is reduced. Extrapolations are consistent with the scenario that μ∗(T) → 0 at a finite crossover temperature T∞ ≈ TK.

However, we emphasise that in the case of the other models which have been investigated, in particular the Wahnström binary Lennard-Jones model does not lead to T∞ ≈ TK, μ∗(T) → 0, but that indeed μ∗(T) does not seem to follow a straight line (Fig. 7). The observation is that while the topology of the phase diagram is preserved, the actual degree of metastability of the supercooled liquids, as quantified by the coexistence value of the dynamical chemical potential as a function of temperature, actually depends on the details of the system. Yet, we find that the emerging, long-lived structural of the structure-rich phase has a direct physical meaning: as discussed in Ref. 7, configurations extracted from the structure-rich phase display a more rigid response, related to the enhanced stability of the locally favoured structures.

V. PUTTING IT ALL TOGETHER

We have shown that the dynamical phase transition, albeit of a (time-integrated) structural quantity, allows access to states very deep in the energy landscape and seems to have a lower critical point somehow close to the putative Kauzmann transition of Adam-Gibbs and RFOT theory. How can it be that a method of dynamical phase transitions, starting from such a wildly different standpoint, can actually start to relate to the Kauzmann-type scenario?

Some insight may be gleaned from examining certain physical quantities in the two phases – the structure-rich inactive phase and the structure-poor, active phase, which lies close to the normal, unbiased, liquid. We begin with the inherent state energy, in Fig. 8. It is clear that, at relatively high temperature (e.g. T = 0.6 in the Kob-Andersen model), the typical inherent state energy of the structure-rich inactive phase is very low (relative to the structure-poor equilibrium liquid). Upon dropping the temperature, in the structure-rich inactive phase, the inherent state energy decreases more gradually with reducing the temperature, while that of the structure-poor active phase falls markedly.

In the particular case of the Kob-Andersen mixture, the simulations in trajectory space allow us to be substantially more quantitative. The average inherent state energy of the structure-poor liquid is found to be well modelled by

\[ \phi_{\text{poor}}(T) = \phi_{\infty} - \frac{J^2}{2T} \]  

in the regime where inherent states are well defined (i.e.
FIG. 6. Results of trajectory sampling (μ-ensemble) computer simulations of the Kob-Andersen binary Lennard-Jones mixture. (a) Temperature versus μ phase diagram. Two distinct structural-dynamical phases are found at coexistence at a finite value μ∗ of the field μ when sampling trajectories of different durations tobs (filled and empty circles): these are poor and rich in structure (LFS poor/rich), represented in the insets (with red and dark blue particles indicating the LFS regions). The scaled value μ∗tobs, however, lies on a single line. A linear extrapolation (dashed line) indicates that at a temperature Tc ≈ TK one would observe the transition from one phase to the other directly in the thermal average of structural quantities (μ = 0) without recurring to large deviations, under the form of intermittency. (b) Dynamical coexistence in the temperature versus concentration of LFS per trajectory plane. The coexistence region (determined by several numerical methods, in blue) has a non-trivial temperature dependence and narrows as the temperature is reduced. The equilibrium supercooled liquid approaches the coexistence region gradually and is always located close to the LFS-poor boundary. The extrapolation of the line of susceptibility maxima (green stars) and the equilibrium line meet at a temperature close to TK, suggesting a cross-over between the LFS-poor and the LFS-liquid. More information in Ref. 40. Reproduced from Ref. 40.

for temperatures well below the onset of slow dynamics), with J being a fitted characteristic energy scale. In the same regime, the average inherent energy of the structure-rich states follows approximately

\[ \phi_{\text{rich}}(T) \approx \gamma T + \phi_0, \]

(4)

with fitting parameters γ and φ₀. These specific forms suggest the existence of a crossover temperature Tₓ at which the structure-poor and the structure-rich state become indistinguishable, which for the particular values of the fitting parameters results to be Tₓ ≈ 0.33. This is a suggestive insight of the trajectory sampling approach in the context of atomistic glassformers: the structural-dynamical transition appears to terminate at a very low temperature in a critical point close to, but above, the estimates of the temperature at which the relaxation times diverge (the Vogel-Fulcher-Tamman fit suggests a divergence at TVFT ≈ 0.325).

As the inherent state energies of structure-rich and poor states converge, we can follow the decrease of configurational entropies of the two disordered states with decreasing temperature. In particular, considering that configurational entropy s, inherent state energy φ, and (configurational) temperature are related by \( ds/d\phi = \)
The emerging picture is that at a low enough temperature the equilibrium, structure-poor supercooled liquid becomes indistinguishable from a very low entropy, very low energy structure-rich metastable state. Remarkably, this picture contains elements of both the Ada-Gibbs/RFOT scenario (i.e. a steadily decreasing configurational entropy to a disordered low entropy state at very low temperatures) and the dynamical facilitation scenario (i.e. the structure-rich state is at the same time an inactive state with vanishing configurational entropy). The crucial question revolves around the location and nature of the crossing point of the inherent state energy and configurational entropy. This is ultimately related to the fate of the structural-dynamical phase coexistence in trajectory space, and in particular the location of the lower critical point of the dynamical phase transition. Several possibilities arise (see more detailed discussions in Refs. 40 and 3). Simulation results of three particulate systems (the Kob-Andersen binary mixture, the Wahnström mixture, and polydisperse hard-spheres) indicate that the coexistence terminates at temperatures close (albeit slightly higher) than the estimated temperature at which the relaxation times are predicted to diverge, e.g. $T_{VFT}$. Whether the coexistence terminates at nonzero dynamical chemical potential $\mu_c$ is still an open question, whose resolution will depend on more accurate, low temperature measures and finite size studies. In any case, a critical point is accompanied by a diverging correlation length influencing the unbiased liquid even for $\mu_c > 0$ and providing a mechanisms for increasing static correlations.

As to the exact relationship between the dynamical phase transition and any thermodynamic glass transition it is clear that the structure-rich inactive phase is a state with exceedingly low configurational entropy, and so in some sense lies at least close to any “ideal glass” state with vanishing configurational entropy.

How does this square with the kinetically constrained models? Interestingly, by adding “softness”, i.e. by softening the constraints of the East model (a KCM), Elmatad and Jack were able to show a profound difference in its dynamical phase diagram (Fig. [25]). The unmodified East model has its dynamical phase transition at $s = 0$, but the softness led to a lower critical point reminiscent of that in the Kob-Andersen model in Fig. [3]. The dynamical phase transition moreover shifted to positive value of the dynamical field $s$. Related results were obtained by Turner et al. [25] with plaquette models, which...
might also be thought of as “KCMs with thermodynamics”. Here the same model showed the $s$—ensemble type transition, and the $\varepsilon$-coupling of the Replica theory.

Finally, it is interesting to note that the buildup of structural correlations in the structure-rich state has repercussions on the orientational correlations as well. Since only relatively small systems of some hundreds of particles can – with the present numerical methods—efficiently sample the structural-dynamical transition at low temperatures, the emerging orientational correlations involve the entirety of the sampled regions of space once the structure-rich state is accessed. It is interesting to note, however, that finite-size and compositional constraints prevent structure-rich configurations from forming complex equilibrium crystalline structures such as the Laves phases of polydisperse hard spheres or crystallising like binary glass formers.

VI. CONCLUSIONS

The notion of metastability implies local equilibrium on finite timescales. Supercooled liquids are metastable disordered states, with complex energy landscapes whose topology is believed to influence the emergent relaxation patterns. Here we have revisited recent results that connect purely dynamical and thermodynamical descriptions of glassy behaviour within a particular framework designed to deal with metastable states, i.e. the theory of large deviations of structural and dynamical observables. The key outcome is that over suitably long observation timescales the dynamics of supercooled liquids explores trajectories that can be characterised either by high mobility and modest structural order or low mobility and enhanced local structural features. Interestingly, a first-order transition in trajectory space can be associated with this behaviour, and it is common to different model of structural glasses, i.e. additive and non-additive Lennard-Jones mixtures and purely repulsive size-dispersed hard spheres.

Most importantly, the transition is strongly affected by decreasing the temperature: at lower and lower temperatures, the inactive, structure-rich trajectories are less and less distinguishable from the active, structure-poor ones. A characterisation of the energy landscape explored by the two classes shows that while the active trajectories sample relatively high energy and entropy regions, the inactive ones explore a narrow region of low energy and low entropy. It is this insight that allows us to bring together two apparently disparate viewpoints of the glass transition: the mosaic of low entropy regions of the Adam-Gibbs/RFOT scenario and the dynamical facilitation framework. The population of locally favoured structures per trajectory is then used as a reaction coordinate to explore metastability, as it couples at the same time with inherent state energies and particle mobility.

Clarifying the relationship between local structure, local configurational entropy and mobility excitations appears as a key task for a more complete theory of dynamic arrest. This will include developing a systematic framework for coarse-graining the model-specific aspects and predicting physically relevant quantities, such as the activation energies advocated by the dynamical facilitation or the size and shape of the cooperatively rearranging regions of the RFOT/Adam-Gibbs scenario. Ensembles analogous to the $\mu$-ensemble can be devised for other quantities which have been defined to quantify glassy systems, including soft spots, aggregated softness fields, two-body excess and patch entropy, local bonding and packing, and community inference and may help to elucidate the relationships between these different descriptors of glassy heterogeneities and their relationship with the dynamics.

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