Abstract

Although several theories relate the steep slowdown of glass formers to increasing spatial correlations of some sort, standard static correlation functions show no evidence for this. We present results that reveal for the first time a qualitative thermodynamic difference between the high temperature and deeply supercooled equilibrium glass-forming liquid: the influence of boundary conditions propagates into the bulk over larger and larger lengthscales upon cooling, and, as this static correlation length grows, the influence decays nonexponentially. Increasingly long-range susceptibility to boundary conditions is expected within the random first-order theory (RFOT) of the glass transition, but a quantitative account of our numerical results requires a generalization of RFOT where the surface tension between states fluctuates.
Supercooled liquids show a dramatic slowdown of their dynamics upon cooling (14 decades increase of viscosity [1] within a narrow temperature range) without any obvious structural or thermodynamic change [2]. The dynamic correlation function of a supercooled liquid furthermore becomes progressively more nonexponential as the temperature is reduced. Several theories relate these phenomena to increasing spatial correlations of some sort [3, 4, 5, 6]. However, static correlations have so far been unable to distinguish qualitatively the high temperature and the deeply supercooled liquids (though indirect evidences have been inferred from the specific heat [7] and the linear dielectric susceptibility [8]). Inspired by critical phenomena, it is natural to expect that the slowing down of the dynamics is related to the vicinity of a thermodynamic phase transition, where some kind of long-range order would set in [9]. This is the spirit of different recent theories [4, 6, 10, 11, 12, 13], but appears at odds with others [5, 14], at least at first sight. In particular, the crucial physical mechanism at the root of random first-order theory (RFOT) [4] is the emergence of long range amorphous order, whose precise definition and quantitative characterisation is however far from obvious. Dynamic heterogeneities [15] do show a growing dynamic correlation length accompanying the glass transition, both experimentally [16] and numerically [17] and refs. therein]. This is certainly a first important step, but not sufficient to prune down — even at a qualitative level — different theories of the glass transition. In particular, it is not clear whether this phenomenon is due to an underlying static or purely dynamic phase transition.

The approach followed here is based on the very definition of a thermodynamic phase transition, where the effect of boundary conditions becomes long-ranged. The problem is that for glasses there are no natural boundary conditions, since these should be as ‘random’ as the bulk amorphous states that they favor. A possible solution has been proposed in [10], and further discussed in [9], in the context of RFOT, but the scope and some conclusions of the gedanken experiment proposed in [10] are more general [18, 19]. Starting from a given equilibrium configuration, one freezes the motion of all particles outside a cavity of radius $R$ and studies the thermodynamics of the mobile particles with boundary conditions imposed by the frozen ones. Defining a suitable overlap (inside the cavity) between the original equilibrated configuration and the configurations equilibrated with the boundary pinning field, the existence of “order” on a scale $\xi$ implies a large overlap (or ‘point-to-set’ correlation) for $R \ll \xi$ and a small one for $R \gg \xi$. 


Such correlation length arises in RFOT [10], if one assumes (or finds by approximate calculations [20, 21]) that there are many amorphous metastable states, $N \approx \exp[R^{d}\Sigma] \text{ inside the cavity (} \Sigma(T) \text{ is the configurational entropy, or complexity)}. \text{ The particles in the cavity can be either in the original state } \alpha \text{ in which the outside particles have been frozen or in any of the other states. The probability to flip to a different state } \gamma \text{ is determined by the balance between the free-energy loss due to mismatch between } \alpha \text{ and } \gamma, \Upsilon(T) R^\theta, \text{ and the gain from the entropic term } T R^d \Sigma(T) (\Upsilon \text{ is a generalized surface tension and } \theta \leq d - 1). \text{ A crossover between a boundary dominated regime (large overlap) and an entropy dominated regime (small overlap) is expected for } R = \xi \propto (\Upsilon/T \Sigma)^{1/d - \theta}. \text{ Note that this length diverges at the Kauzmann temperature } T_K \text{ where } \Sigma(T_K^+) \to 0.\text{ This gedanken experiment was realized numerically by three of us in ref. [22]. This first study indeed suggested a growing static length, but this length was very small, and no sharp transition between high and low overlap was observed, in contrast with expectations based on RFOT. Is RFOT possibly in contradiction with numerical experiments? What is the physical meaning of the increasingly long range susceptibility to boundary conditions if it is characterized by such a small length? In order to answer these crucial questions we have performed new simulations that measure the local overlap at the center of the cavity. In fact, a major difficulty in interpreting the results of ref. [22] is that the overlap was computed as an average over the whole cavity. As a result, the overlap drop due to the (putative) change of state of the cavity is mingled with the decay of the overlap expected from a trivial weakening of the surface pinning field for larger spheres. This effect is indeed present even in the single state case [19] at all temperatures. As we shall show, analyzing the behavior of the local overlap at the center of the cavity yields sharper results which allow us to answer, at least partially, the above questions.}

**I. BEHAVIOUR OF THE OVERLAP AT HIGH AND LOW TEMPERATURE**

We study a soft-sphere model [23] that we can equilibrate below the Mode Coupling transition temperature $T_{MC} = 0.226$ [24] and for large systems (see methods). After equilibration, several independent reference configurations are chosen as starting points for runs with all but $M$ particles frozen. These mobile particles are confined inside a sphere of radius $R$ such that the inside density equals the bulk density. After the confined runs reach
equilibrium, the local overlap at the center $q_c(R)$ is measured. To define $q_c(R)$, we partition
the simulation box in many small cubic boxes of side $\ell$, such that the probability of finding
more than one particle in a single box is negligible. Let $n_i$ equal the number of particles in
box $i$, then
\begin{equation}
q_c(R) = \frac{1}{\ell^3 N_i} \sum_{i \in v} \langle n_i(t_0) n_i(t_0 + \infty) \rangle
\end{equation}
where the sum runs over all boxes within a small volume $v$ at the center of the sphere, $N_i$ is
the number of boxes, and $\langle \ldots \rangle$ means thermal average. To minimize statistical uncertainty
without losing the local nature of $q_c(R)$ we choose $N_i = v/\ell^3 = 125$. Normalization is such
that the overlap of two identical configurations is 1 on average, while for totally uncorrelated
configurations $q_c = q_0 \equiv \ell^3 = 0.062876$.

We show $q_c(R) - q_0$ for several temperatures in Fig. 1. The decay becomes slower at
lower temperature: the effect of boundary conditions propagates on larger length-scales.
This clearly shows the growth of static order that expands into the bulk in the deeply
supercooled phase. The most striking result, however, is that at low temperatures the
decay is no longer the simple exponential that prevails in a standard liquid state [19]. The
relaxation can be fit by a “compressed exponential”,
\begin{equation}
q_c(R) - q_0 = \Omega \exp \left[ -(R/\xi)^\zeta \right], \quad \zeta \geq 1,
\end{equation}
where $\zeta$ increases at low temperatures (see Fig. 2a) above its high temperature liquid value
$\zeta = 1$ (see Fig. 2b). Larger values of $\zeta$ means a sharper crossover between large and small
overlaps. The best fit parameters are given in Table I. Interestingly, the value of the length
scale $\xi$ found here is significantly larger than those in [22]. This is partly due to the fact
that $\zeta > 1$ at low temperatures (see appendix for further discussion). The overlap $q_c(R)$
therefore appears as a thermodynamic quantity able to single out the deeply supercooled
liquid in terms of (a) a large correlation length $\xi$ and (b) an anomalous nonexponential
relaxation, characterized by the exponent $\zeta > 1$.

The growth of $\xi$ strongly suggests that the liquid state should be thought of as a mosaic
of regions (transiently) ordered on a lengthscale $\xi$. Furthermore, following suggestions based
on RFOT [4, 10, 11], it is natural to conjecture that we are probing the growth of positional
amorphous order. Interestingly, in this scenario, all finite point static correlations remain
featureless, whereas the point-to-set correlations [9, 25], captured by $q_c(R)$, grow. The
simplest version of RFOT predicts at low temperature a sharp cross-over of $q_c(R)$ for $R = \xi$
FIG. 1: Overlap at the centre of the mobile cavity vs. radius $R$ of the cavity, for temperatures $T = 0.482$ (diamonds), 0.350 (triangles), 0.246 (squares) and 0.203 (circles). Lines are fits to eq. (2).

Inset: Comparison of $q_c(R) - q_0$ at $T = 0.203$ (filled circles) with the overlap $Q(R) - q_0$ integrated over the whole sphere (open circles, data ref. 22). The local observable $q_c(R)$ shows a much sharper behaviour.

II. RFOT WITH FLUCTUATING SURFACE TENSION

To account for our numerical results, we now propose a natural generalization of RFOT, based on the idea that the effective interface tension, $\Upsilon$, is in fact state-dependent. Repeating
FIG. 2: a) Local overlap at the centre of the mobile cavity vs. $R$ for $T = 0.203$ (circles) with fits to a compressed exponential (full line) and simple exponential (dotted line). The best exponential fit is rather poor. Inset: same data in semilog axes, where a simple exponential looks like a straight line. b) As in panel a) but for $T = 0.350$. The exponential fit (line) is good at this temperature, as can be seen in the inset, which displays the same data in semilog axes.
TABLE I: Best fit parameters as function of temperature. Errors are jacknife estimates. (*) For the highest temperatures, the results quoted are for a simple exponential fit, which gives a very good description of the data with only two parameters. If nonetheless those data are fit with the compressed exponential form, an exponent $\zeta$ compatible with 1 is obtained, albeit with very large (60–80%) error.

The argument of ref. 10 with this extra assumption, the probability that the cavity is found in a state $\gamma$ different from the outside pinning (frozen) state $\alpha$, is:

$$p_{\text{out}}(\alpha) = \frac{\int d\Upsilon e^{Rd\Sigma^* - \beta\Upsilon R^\theta} p_\alpha(\Upsilon|f^*)}{1 + \int d\Upsilon e^{Rd\Sigma^* - \beta\Upsilon R^\theta} p_\alpha(\Upsilon|f^*)}, \quad p_{\text{in}}(\alpha) = 1 - p_{\text{out}}(\alpha), \quad \text{(3)}$$

where $\beta = 1/T$, $f^*$ and $\Sigma^* = \Sigma(f^*)$ are respectively the free energy and configurational entropy of the equilibrium states, and $p_\alpha(\Upsilon|f)$ is the distribution of surface tension for state $\alpha$ with other states of free energy $f$ (see Methods). Since our simulations average over many realizations of the external state $\alpha$, we in fact compute $p_{\text{out}}^{(\alpha)}(R)$ averaged over all possible pinning states:

$$p_{\text{out}}(R) = \sum_\alpha w_\alpha p_{\text{out}}^{(\alpha)}(R) = \sum_\alpha w_\alpha \frac{\int d\Upsilon e^{Rd\Sigma^* - \beta\Upsilon R^\theta} p_\alpha(\Upsilon|f^*)}{1 + \int d\Upsilon e^{Rd\Sigma^* - \beta\Upsilon R^\theta} p_\alpha(\Upsilon|f^*)}, \quad \text{(4)}$$

where $w_\alpha$ is the thermodynamic weight of each state. The leading contribution of the integral over $\Upsilon$ is shown in Methods to be $e^{-\beta y_\alpha R^\theta}$, where $y_\alpha$ is the lower edge of $p_\alpha(\Upsilon|f)$ and where the exponent $\theta$ is possibly renormalized (this might explain why the effective value of $\theta$ can be less than the naively expected value $d-1$). Introducing $P(Y) = \sum_\alpha w_\alpha \delta(Y - y_\alpha)$ we find

$$p_{\text{out}}(R) = \int dY P(Y) \frac{e^{Rd\Sigma^* - \beta Y R^\theta}}{1 + e^{Rd\Sigma^* - \beta Y R^\theta}}, \quad \text{(5)}$$

The simple mosaic result of ref. 10 is recovered setting $p_\alpha(\Upsilon|f) = \delta(\Upsilon - \Upsilon_0)$ and $P(Y) = \delta(Y - Y_0)$. Expression (5) can be simplified further by noticing that the approximation $(1 + e^{Rd\Sigma^* - \beta Y R^\theta})^{-1} \approx \Theta(Y - T\Sigma^* R^{1-\theta})$ holds even for rather small values of $R$ ($\Theta$ is the
step function). Hence:

\[ p_{\text{out}}(R) = \int_0^{T\Sigma^* R^{d-\theta}} P(Y) \, dY. \]  

(6)

The overlap at the centre of the cavity is given by

\[ q_c(R) = p_{\text{in}}(R) q_1 + p_{\text{out}}(R) q_0, \]

where \( q_1 \) is the self-overlap of the cavity (which is not necessarily 1 at finite temperatures). Using equation (6), we finally obtain

\[ q_c(R) - q_0 = (q_1 - q_0) \int_0^{\infty} dY P(Y). \]  

(7)

At this point one needs to make some assumption about \( P(Y) \). An interesting parameterization is:

\[ P(Y) = \frac{\nu}{Y_c} \left( \frac{Y}{Y_c} \right)^{\nu-1} \exp\left[-(Y/Y_c)^\nu\right] = -\frac{d}{dY} \exp\left[-(Y/Y_c)^\nu\right] \]  

(\( \nu > 0 \)), which describes either a stiff distribution (small values of \( Y \) suppressed) at large \( \nu \) or a soft one (small values of \( Y \) enhanced) for small \( \nu \). \( Y_c \) is the typical value of \( Y \). The standard mosaic picture corresponds to \( \nu \to \infty \). Equation (7) now reads, explicitly:

\[ q_c(R) - q_0 = (q_1 - q_0) \exp\left[-(R/\xi)^{\nu(d-\theta)}\right] \]  

(9)

where the usual RFOT relation between \( \xi \) and \( \Sigma \) is recovered: \( \xi = (Y_c/T\Sigma^*)^{1/(d-\theta)} \). As in the Adam-Gibbs treatment [28], the growth of the length is caused by the decreasing of the configurational entropy at low temperature. The expression (9) is the compressed exponential form (2) used above to fit our numerical data. Within this framework, the thermodynamic anomaly \( \zeta = \nu(d-\theta) \) is directly related (at low temperatures) to the exponent \( \nu \) describing the surface tension distribution.

A non trivial \( P(Y) \) could arise for two main reasons. One is strong pre-asymptotic corrections to standard RFOT: though the surface tension between two typical states could be self-averaging in the \( R \to \infty \) limit, fluctuations can be state dependent and important at finite \( R \). This is often what happens for interfaces in random media (see SI). The second possibility is that the effective interface tension fluctuates even in the large \( R \) limit. If the exponent \( \theta \) turns out to be less than \( d-1 \), as suggested by phenomenological arguments in ref. [4] then the interface should be a highly fluctuating object as e.g. in the droplet theory of spin-glasses [29]. We find that \( \zeta \), and therefore \( \nu \), increase when \( T \) decreases indicates that the crossover of \( q_c(R) \) becomes sharper at larger sizes (see Fig. 1). This is compatible with a finite-size effect scenario. This behaviour is also expected within the RFOT scenario that
predicts a vanishing surface tension at the mode-coupling transition \( T_{MC} \), which behaves as a spinodal point. Coherent amorphous order droplets should therefore be fractal around \( T_{MC} \) and compact below [30], which suggests an increase of the effective value of \( \nu \) as \( T \) decreases. A first principle RFOT computation of \( q_c(R) \) for the model we simulated would be very instrumental to clarify this issue.

III. CONCLUSIONS AND OUTLOOK

We have unveiled a qualitative difference between the high temperature and deeply supercooled equilibrium regimes: the influence of boundary conditions propagates into the bulk on an increasingly large lengthscale upon cooling. Furthermore, the growth of this length is accompanied by a sharpening of the decay. We have developed a theoretical framework, based on a generalization of RFOT, that explains these results as a one-state to multi-state transition governed by the surface tension distribution. The sharpening of the decay at low temperature corresponds to more and more regions developing a large surface tension. From a more general perspective, our numerical results strongly support a mosaic picture where the super-cooled liquid is characterized by a “hidden” static order [32] on an increasingly larger scale upon cooling. Although this is a natural consequence of RFOT, other theoretical approaches may also account for these phenomena at least on a qualitative level: in particular the frustration limited domain theory [13] and, perhaps surprisingly, some kinetically constrained models (see [18] for a discussion of this point). The main difference is the physical origin of the growing static lengthscale: within RFOT and at variance with other approaches, it is tightly linked to the decrease of the configurational entropy. Our work opens the way to a quantitative study of this issue and, hence, to a clear-cut test of RFOT as a valid theory of the glass transition. From a more theoretical point of view, several crucial questions remain elusive: can a RFOT-like transition exist outside mean-field? How precisely can amorphous metastable states be defined? Does the notion of effective surface tension between these states make sense? A definitive test of the mosaic scenario requires to find a way to measure directly this surface tension and its distribution in the deeply supercooled phase. Work in this direction is in progress.
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IV. METHODS

A. Simulation

We have studied the soft-sphere binary mixture \([23]\), a fragile model glass-former. In addition to capturing the essential features of fragile glasses, this model can be thermalized below the Mode Coupling temperature with the swap Monte Carlo algorithm of \([31]\). Particles are of unit mass and belong to one of two species \(\mu = 1, 2\), present in equal amounts and interacting via a potential

\[
V = \sum_{i>j} v_{ij}(|\mathbf{r}_i - \mathbf{r}_j|) = \sum_{i>j} \left[ \frac{\sigma_{\mu(i)} + \sigma_{\mu(j)}}{|\mathbf{r}_i - \mathbf{r}_j|} \right]^{12},
\]

where the radii \(\sigma_\mu\) are fixed by the conditions \(\sigma_2/\sigma_1 = 1.2, (2\sigma_1)^3 + 2(\sigma_1 + \sigma_2)^3 + (2\sigma_2)^3 = 4\ell_0^3\), and \(\ell_0\) is the unit of length. The particle density is \(\rho = N/V = \ell_0^{-3}\). A smooth long-range cut-off is imposed setting \(v_{ij}(r) = B_{ij}(a - r)^3 + C_{ij}\) for \(r > r_c = \sqrt{3}\) and \(v_{ij}(r) = C_{ij}\) for \(r > a\), where \(a, B_{ij},\) and \(C_{ij}\) are fixed by requiring continuity up to the second derivative of \(v_{ij}(r)\). Temperature is measured in units of energy. To obtain the reference configurations, 4 to 8 replicas of systems with \(N = 2048\) or \(N = 16384\) where equilibrated in a cubic box with periodic boundary conditions. Then the overlap was computed in systems with frozen boundaries and \(M\) mobile particles, with \(M = 20, 30, 40, 50, 100, 150, 200, 300, 400, 800, 1600, 3200\) mobile particles \((1.684 \leq R \leq 9.142)\). The results are averaged over 8 to 32 outer states (reference configurations). Data were collected for at least 10 relaxation times (up to \(10^6\) Monte Carlo steps), after discarding an initial portion of at least one relaxations time. Error bars were obtained from a jacknife estimate from sample-to-sample fluctuations.
B. Overlap

To obtain equation (3), write the partition function for the mobile cavity surrounded by pinning state α

$$Z_c = e^{-\beta R^d f_\alpha} + \sum_{\gamma \neq \alpha} e^{-\beta R^d f_\gamma - \beta R^\theta \Upsilon_{\alpha\gamma}}, \quad (10)$$

so that

$$p^{(\alpha)}_{\text{out}}(R) = \frac{\sum_{\gamma \neq \alpha} e^{-\beta R^d f_\gamma - \beta R^\theta \Upsilon_{\alpha\gamma}}}{Z_c}. \quad (11)$$

Introducing $N_\alpha(f, \Upsilon) = \sum \delta(f - f_\gamma) \delta(\Upsilon - \Upsilon_{\alpha\gamma})$, the sum can be written

$$\sum_{\gamma \neq \alpha} e^{-\beta R^d f_\beta - \beta R^\theta \Upsilon_{\alpha\gamma}} = \int df \int d\Upsilon \, e^{-\beta R^d f - \beta R^\theta \Upsilon} N(f, \Upsilon), \quad (12)$$

$$= \int df \int d\Upsilon \, e^{-\beta R^d f - \beta R^\theta \Upsilon + R^d \Sigma(f)} p_\alpha(\Upsilon | f), \quad (13)$$

where in the last equality we have defined $p_\alpha(\Upsilon | f) = N_\alpha(f, \Upsilon) / N(f)$, and $N(f) = \exp[\Sigma(f)]$ is the number of states with free energy $f$. Equation (3) follows approximating the integral over $f$ with the saddle-point method, which picks $f = f^*$ as the dominant contribution. $p_\alpha(\Upsilon | f)$ is the fraction of states $\gamma$ (inside the cavity) with free energy $f$ and effective interface tension $\Upsilon$, which we assume to be $R$-independent.

Now the integral in equations (3) and (4) can be simplified using the saddle point method, which is a very good approximation even for rather small values of $R$. Because of the exponential term in $R^\theta$, the integral is dominated by the lowest values of $\Upsilon$ supported by the distribution $p_\alpha(\Upsilon | f)$. There are two possibilities cases: (a) If this function has a left edge, i.e. it vanishes for $\Upsilon < y_\alpha$, one finds up to subleading terms: $\int d\Upsilon \, p_\alpha(\Upsilon | f) \, e^{-\beta \Upsilon R^\theta} \simeq e^{-\beta y_\alpha R^\theta}$. Otherwise: (b) there are arbitrarily small effective tensions $\Upsilon$. Remarkably, in this case, depending on the form of $p_\alpha(\Upsilon | f)$ at small $\Upsilon$, one can obtain a renormalization of $\theta$. For instance in the case $p_\alpha(\Upsilon | f) \simeq \exp(-c_\alpha / \Upsilon^a)$, where $a$ is a positive exponent, one finds $\int d\Upsilon \, p_\alpha(\Upsilon | f) \, e^{-\beta \Upsilon R^\theta} \simeq e^{-\beta y'_\alpha R^{\theta'}}$, where $y'_\alpha$ is a constant dependent of the outside state $\alpha$ and temperature and $\theta' = a \theta / (a + 1) < \theta$.

APPENDIX A: COMPARISON WITH THE RESULTS OF REF. 22

Following the same numeric protocol of this work, in [22] the influence of the boundary conditions on the total overlap within the sphere was studied. It was observed that the decay
of $q_{\text{tot}}(R)$ is described sufficiently well in the single state framework and the standard RFOT scenario was ruled out. In this work we show however that at low enough temperatures neither the standard RFOT nor the single state scenario account for the local overlap data and a generalized RFOT theory is presented which is far more successful.

One might retrospectively wonder if the data in [22] could have been used to discriminate between the one-state and the generalized RFOT scenarios. The answer is no. Allowing for a $R$ dependence of $q_0$ and $q_1$ of the type suggested in [22]:

$$q_{0;1}(R) = 3(1 - q_{0;1}^*) \left[ \frac{1}{x} - \frac{2}{x^2} + \frac{2(1 - e^{-x})}{x^3} \right] + q_{0;1}^*$$  \hspace{1cm} (A1)

with $x \equiv R/\lambda_{0;1}$, the generalized RFOT prediction for $q_{\text{tot}}(R)$ reads now:

$$q_{\text{tot}}(R) = q_0(R) + (q_1(R) - q_0(R)) \exp \left[ -\left(\frac{R}{\xi}\right)^{\zeta} \right]$$  \hspace{1cm} (A2)

In the figure below we show that the generalized RFOT encoded in (A2) and the one-state prediction given by the formula (6) of ref. [22] fit the total overlap data at $T = 0.203$ at a comparable level of accuracy. Following Occam’s razor principle (“entia non sunt multiplicanda praeter necessitatem”), one should choose one-state theory, which is the one with the smallest number of parameters.

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FIG. 3: Total overlap of the mobile cavity at $T = 0.203$ (data taken from ref. [22]). The dashed line shows the fit with one-state theory while the solid line shows the fit with the generalized RFOT prediction [A2].

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[32] We call it “hidden” since, as discussed in the introduction, all simple static correlation functions investigated until now have never shown direct evidence of a growing length.

[33] Actually, some $R$-dependence would not affect the result. The important requirement is that this dependence does not lead to values exponentially large or small in $R$.