Elasticity, Facilitation, and Dynamic Heterogeneity in Glass-Forming Liquids

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We study the role of elasticity-induced facilitation on the dynamics of glass-forming liquids by a coarse-grained two-dimensional model in which local relaxation events, taking place by thermal activation, can trigger new relaxations by long-range elastically mediated interactions. By simulations and an analytical theory, we show that the model reproduces the main salient facts associated with dynamic heterogeneity and offers a mechanism to explain the emergence of dynamical correlations at the glass transition. We also discuss how it can be generalized and combined with current theories.

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Glass-forming liquids display a huge slowing down of the dynamics, characterized by a relaxation time that grows by more than 14 orders of magnitude when the temperature is reduced by just 1/3 from its value at melting [1]. Whereas it is very difficult to find signatures of this dramatic change of behavior in static correlation functions, a clear signal emerges in dynamical spatial correlations and length scales [2]. The associated concomitant growth of timescales and length scales is a hint of the collective nature of the relaxation processes underpinning glassy dynamics. This phenomenon, called dynamic heterogeneity (DH), has been a central one in the research on the glass transition both theoretically and experimentally [2]. However, a full understanding of DH, especially close to the glass transition, is still lacking.

In this respect, an important aspect is certainly dynamic facilitation [2]. This is the property by which a local region that undergoes relaxational motion in a supercooled liquid, or in a similar slow relaxing material, gives rise to or facilitates a neighboring local region to move and relax subsequently. Some theories advocate that facilitation provides a complete explanation of DH [3], whereas others suggest that it is part of a more complex dynamical process associated with the growth of a static length scale [4] (see also the recent discussions in Refs. [5,6]). Despite its important role demonstrated in numerical simulations and experiments [2,6–10], the cause of dynamic facilitation in real systems has not been fully elucidated yet. Furthermore, its interplay with thermal activation events is also not well understood. A theory of glassy dynamics [3,8,11] posits the emergence of kinetic constraints and describes dynamical slowing down using kinetically constrained models (KCM) [12]. In this case, facilitation is the main mechanism at play. However, its effect on dynamics is very dependent on the kind of kinetic constraint chosen, and a first-principle study of the mechanisms that would lead to specific kinetic constraints is currently lacking.

In this Letter, we address these issues by envisioning supercooled liquids as solids that flow, following the pioneering works [13,14], and the recent numerical findings [15–21] which show that anisotropic stress fluctuations with Eshelby-like patterns emerge in the supercooled state. We consider that close to the glass transition dynamics proceeds by local events that take place in a surrounding matrix which is solid on the timescale over which the local event takes place [22,23]. In consequence, the local relaxation event causes an elastic deformation in the surrounding because the system restores mechanical equilibrium that is broken transiently due to the plastic rearrangement [24]. Such deformation changes the arrangements of the particles and can then make some nearby regions more prone to relaxation. Recent works [9,24] have shown that this elastic mechanism indeed leads to dynamic facilitation and/or avalanche-like rearrangements. Reference [25] has used elasticity theory to estimate the energy scale associated with the dynamic facilitation theory of Ref. [3]. Another series of works [26,27] have concentrated on "softness" and elasticity as the fields that mediate facilitation. Here, we focus on the simplest model that encodes this "elasticity-induced facilitation." We show by numerical simulations and theoretical analysis that this mechanism allows us to capture the main salient facts associated with DH. Our model offers a starting point for a quantitative theory of dynamical correlations in glass-forming liquids, and it sheds new light on the theories of the glass transition.

Since glassy phenomenologies, in particular DH, have been universally observed in a wide variety of glassy materials regardless of the details of the microscopic interactions [2,28] (and also for non-time-reversible dynamics such as granular materials), we aim for a coarse-grained simplified description able to capture the essential physical mechanisms. The elastoplastic models (EPMs), which have already been very successful in describing rheology and

yielding transitions of amorphous materials [29,30], provide then a natural setting to study the role of elasticity and plasticity on the dynamics of supercooled liquids. In consequence, we focus on a scalar EPM in which plastic relaxation is not induced by external loading but by thermal activation. We study the observables used to probe equilibrium dynamics and DH of supercooled liquids [2].

Our model, called EPM-Q, is defined on a $L \times L$ square lattice where each site represents a mesoscopic coarse-grained region of the equilibrium supercooled liquid. To each site i we associate a local shear stress σ_i, a local energy barrier $\Delta E(\sigma_i)$ activating the plastic relaxation event [31–33], and an orientation $\psi_i \in (0, \pi/2]$ for the Eshelby elastic interaction. The following dynamical rules of the model encode the effects of local thermal activation, local plastic rearrangement, and long-range and anisotropic elastic interaction. At each time step, we pick a site i at random uniformly among the L^2 sites. If $|\sigma_i|$ is greater than or equal to a threshold value $\sigma_c = 1$ then the site i rearranges with probability one, whereas if $|\sigma_i| < \sigma_c$ then the site i rearranges with probability $e^{-\Delta E(\sigma_i)/T}$, where T is the temperature. Because of this local plastic event, σ_i is updated by a local stress drop: $\sigma_i \to \sigma_i - \delta\sigma_i$, where $\delta\sigma_i = (z + |\sigma_i| - \sigma_c)\mathrm{sgn}(\sigma_i)$; $z > 0$ is a random number drawn by a distribution $\rho(z)$. The sign function $\mathrm{sgn}(x)$ takes into account that if $\sigma_i > 0$ (or $\sigma_i < 0$) local yielding is activated by a barrier at σ_c (or $-\sigma_c$). The stress drop $\delta\sigma_i$ at site i is then redistributed on the surrounding sites using the Eshelby kernel [34] with the (random) orientation ψ_i. A new orientation is drawn uniformly at random after each plastic event. We stress that the random orientation introduced in this Letter is a crucial aspect to describe the physics of quiescent (isotropic) supercooled liquids [35,36], and differs from the aligned Eshelby kernel used in previous elastoplastic modelings of sheared materials [30]. We repeat the above attempt L^2 times, which corresponds to the unit of time. Our choice of $\rho(z)$ and $\Delta E(\sigma)$ is based on previous literature [22,32,33,37]:

$$\rho(z) = \frac{1}{z_0} e^{-z/z_0}, \qquad \Delta E(\sigma) = K(\sigma_c - |\sigma|)^a, \qquad (1)$$

where $a = 1.5$ [38,39], and the mean value z_0 and the generalized stiffness K are set to one for simplicity [40]. Note that the mean-field facilitated trap models introduced in Refs. [41,42] share important conceptual similarities with the EPM-Q. The results presented below are obtained in the stationary state for systems with $L = 64$ unless otherwise stated. See Supplemental Material (SM) [43] for more details [43]. As in usual investigations of glassy dynamics, we have studied the intermediate scattering function [58]. To make the connection with studies on KCMs [11], and since it is particularly well suited for lattice systems, we have focused on the average of the persistence function $P(t) = (1/L^2)\sum_i p_i(t)$, where $p_i(t)$ is equal to one if the site i did not relax from time zero to time t and zero otherwise [59]. Both correlation functions behave in a qualitatively and quantitatively analogous way. We show the latter in Fig. 1(a) and the former in SM [43].

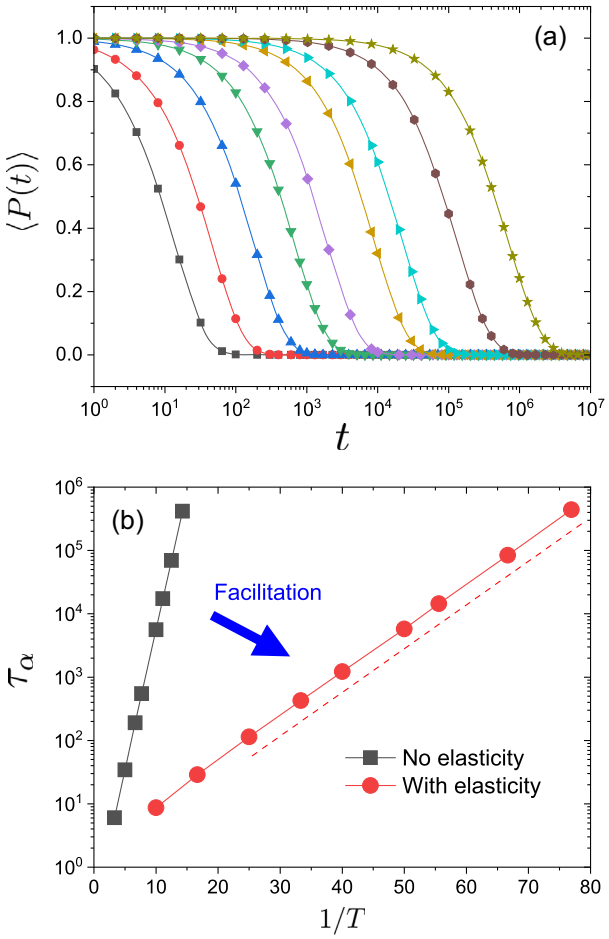

FIG. 1. (a) The average persistence function $\langle P(t)\rangle$ for $T = 0.100$, 0.060, 0.040, 0.030, 0.025, 0.020, 0.018, 0.015, and 0.013 (from left to right). (b) The relaxation time τ_α for the models with (circle) and without (square) elastic interaction. τ_α is defined by $\langle P(\tau_\alpha)\rangle = 1/2$. The dashed straight line defines an (average) activation energy barrier $\Delta E(\bar{\sigma}^{\mathrm{act}})$.

Similarly to what is found for dynamical correlation functions in supercooled liquids, $\langle P(t)\rangle$, where $\langle\cdots\rangle$ is the time average at the stationary state, decays in an increasingly sluggish way with decreasing T, thus capturing the slowing down of the dynamics. The shape of the relaxation function is simpler than the one of the realistic liquid models. This is due to the simplicity of the model and can be cured by generalizing it, as we shall discuss later. By plotting the relaxation time τ_α (defined as $\langle P(\tau_\alpha)\rangle = 1/2$) as a function of $1/T$ in Fig. 1(b) we find that τ_α diverges in an Arrhenius way when lowering the temperature. For comparison, we also plot τ_α obtained from the model *without* elastic interactions, i.e., in the absence of stress redistribution. Remarkably, this relaxation time is larger,

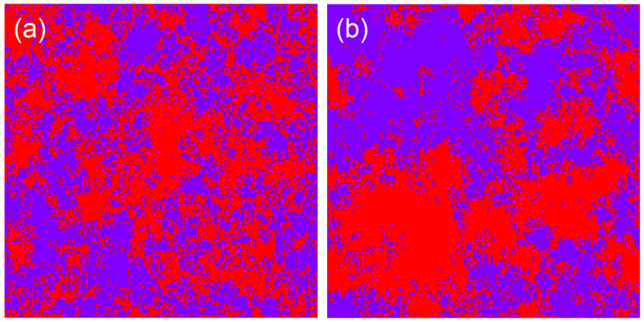

FIG. 2. Snapshots for local persistence $p_i(\tau_\alpha)$ when $P(\tau_\alpha) \approx 1/2$ for $T = 0.040$ (a) and $T = 0.013$ (b), respectively. The system size is $L = 128$. Red and blue sites correspond to mobile $[p_i(\tau_\alpha) = 0]$ and immobile $[p_i(\tau_\alpha) = 1]$ sites, respectively.

showing that elastic interactions substantially diminish the energy barrier. This is direct evidence that elastic interactions facilitate and accelerate dynamics in the model. This conclusion is achieved thanks to the coarse-grained model approach, where we can turn elasticity on and off. This is virtually impossible for molecular simulations since elasticity is an emergent property of the material. The second direct evidence is provided by studying the morphology of dynamical correlations. Figure 2 shows the patterns formed by the local persistence $p_i(t)$ at two different temperatures: clearly, the dynamics is spatially heterogeneous over lengths that increase when lowering the temperature (see also movies in SM [43]). The patterns in Fig. 2 strongly resemble the ones found in realistic (atomistic, colloidal, granular) systems [2]. The counterparts of Fig. 2 in the absence of elastic interactions (not shown) display no spacial dynamical correlations at all.

To quantify DH we measure the dynamical susceptibility [60], $\chi_4(t) = L^2(\langle P^2(t)\rangle - \langle P(t)\rangle^2)$, in Fig. 3(a). We observe essentially the same time and temperature evolution found in molecular dynamics simulations of supercooled liquids [2]. To study the relationship between time and length scales, we plot the peak of χ_4 as a function of the logarithm of τ_α in Fig. 3(b). This curve displays a striking similarity with the ones obtained from experimental data (see, e.g., Fig. 4 of Ref. [61]): after a fast increase during the first decades of slowing down, the increase of the dynamical correlation length becomes slower, possibly logarithmic, with respect to τ_α. This is a highly nontrivial result that can be found only in some tailored KCMs [11], and it has been argued to hold for the random first order transition (RFOT) theory [62,63]. In both cases, the bending shown in Fig. 3(b) is associated with cooperative dynamics. As we shall explain later, in our model, the reason is different (although it shares some similarities with KCMs).

We now offer a theoretical explanation for the phenomenological behavior presented above. Our starting point is the kinetic elastoplastic theory developed in Ref. [64].

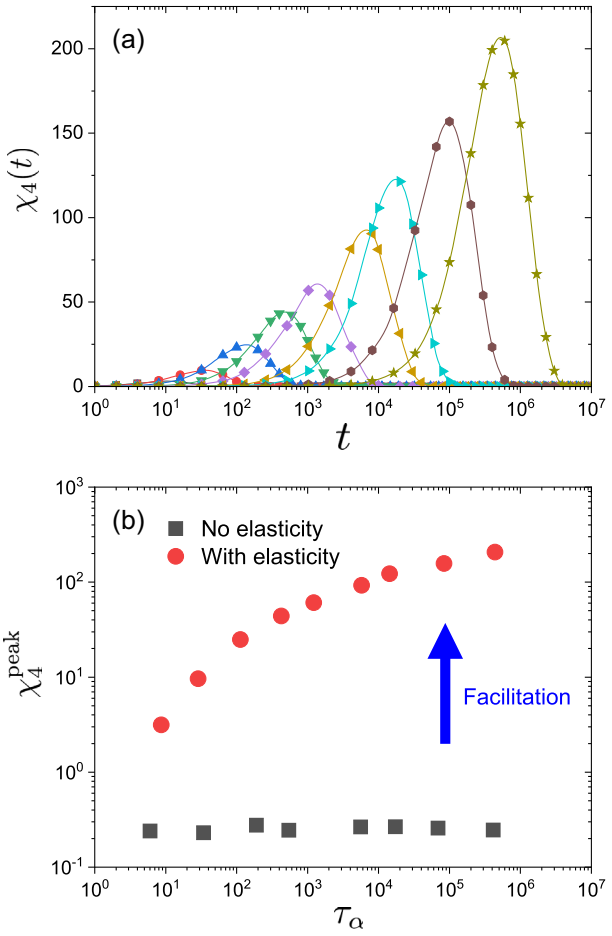

FIG. 3. (a) Time and temperature evolution of $\chi_4(t)$ for $T = 0.100$, 0.060, 0.040, 0.030, 0.025, 0.020, 0.018, 0.015, and 0.013 (from left to right). (b) The peak value of χ_4 as a function of τ_α for the models with (circle) and without (square) elastic interactions.

Using translation invariance, one finds that the kinetic equations of Ref. [64] boil down to the following Hébraud-Lequeux-like model [32,65,66] for the probability distribution $P(\sigma, t)$ of the local stress σ at a given site (see SM [43]):

$$\frac{\partial P(\sigma,t)}{\partial t} = D(t)\frac{\partial^2 P(\sigma,t)}{\partial \sigma^2} - \nu(\sigma,\sigma_c)P(\sigma,t) + \Gamma(t)y(\sigma), \quad (2)$$

where the three terms on the right-hand side in Eq. (2), respectively, correspond (from left to right) to (i) the redistribution of the stress due to elastic interactions, (ii) a loss term due to rearrangements that change the local value of σ, and (iii) a gain term due to rearrangements in which the local value of the stress becomes equal to σ after stress drops that take place with rate $\Gamma(t) = \int_{-\infty}^{\infty} d\sigma\, \nu(\sigma,\sigma_c)P(\sigma,t)$. The strength of the first term $D(t)$ is related to the total relaxation rate by $D(t) = \alpha\Gamma(t)$ [64]. In our case, in which the Eshelby orientations

are randomly oriented, $\alpha \simeq 0.110$ (see SM [43]), whereas $\nu(\sigma, \sigma_c)$ reads

$$\nu(\sigma, \sigma_c) = \frac{1}{\tau_0}\theta(|\sigma| - \sigma_c) + \frac{1}{\tau_0} e^{-[\Delta E(\sigma)/T]}\theta(\sigma_c - |\sigma|), \quad (3)$$

where $1/\tau_0$ is the rate of the plastic event. The first term on the right-hand side in Eq. (3) is due to spontaneous relaxation when the system is locally unstable (beyond σ_c or below $-\sigma_c$), whereas the second one is due to thermally activated relaxation [32]. We show in SM [43] that for $T \to 0$ the stationary $P(\sigma)$ has a symmetric bell shape and is nonzero for $-\bar{\sigma} < \sigma < \bar{\sigma}$ with $\bar{\sigma}$ strictly less than σ_c (leading to a gap in the distribution of excitations [67,68]). Its analytic expression is reported in SM [43]. We plot it in Fig. 4 as a dashed curve and compare it to $P(\sigma)$ obtained from numerical simulations of the two-dimensional model at different temperatures. The agreement is very good. Figure 4 numerically confirms that the support of $\lim_{T\to 0} P(\sigma)$ is within the interval $[-\bar{\sigma}, \bar{\sigma}]$ with $\bar{\sigma} < \sigma_c$, and show a numerical value of $\bar{\sigma}$ very close to the one we computed analytically. The stress $\bar{\sigma}$ defines the smallest typical energy barrier $\Delta E(\bar{\sigma})$. As shown in SM [43], the latter determines the relaxation rate Γ for $T \to 0$:

$$\Gamma \simeq \frac{1}{\tau_0} e^{-[\Delta E(\bar{\sigma})/T]}. \quad (4)$$

The above results lead to a scenario in which there is a spatially heterogeneous distribution of local stresses given by $P(\sigma)$. This leads to a distribution of energy barriers in the system. The sites having the smallest barriers, corresponding to $|\sigma| \simeq \bar{\sigma}$, are the ones triggering rearrangements

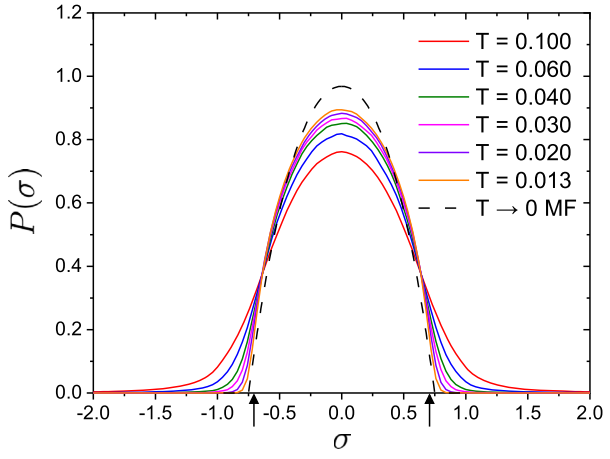

FIG. 4. Probability distribution function of the local stress $P(\sigma)$. The dashed curve indicates the solution of the mean-field (MF) theory at $T \to 0$. The vertical arrows indicate the location of $\bar{\sigma}^{\rm act}$ and $-\bar{\sigma}^{\rm act}$ extracted from $\Delta E(\bar{\sigma}^{\rm act})$ with the activation energy barrier in Fig. 1(a). Recall that $\sigma_c = 1$.

and controlling the relaxation time at small temperatures. Our numerical findings presented before fully agrees with this picture: indeed, the value of the stress $\bar{\sigma}^{\rm act}$ that would lead to the Arrhenius behavior in Fig. 1(b), $\tau_\alpha \sim 1/\Gamma \sim e^{\Delta E(\bar{\sigma}^{\rm act})/T}$, is identical (or very close) to the edge $\bar{\sigma}$ of the support of the stress distribution, see Fig. 4. The effect of elasticity-induced facilitation on the relaxation timescale, shown in Fig. 1(b), is correctly reproduced by our analysis of Eq. (2) which predicts $\tau_\alpha \sim 1/\Gamma \sim e^{\Delta E(0)/T}$ in the absence of stress redistribution, hence a larger barrier $\Delta E(0) > \Delta E(\bar{\sigma}^{\rm act})$ (see SM [43]).

The above scenario also offers an explanation for the development of DH. Once a site with $|\sigma| \simeq \bar{\sigma}$ triggers a local rearrangement, stress is redistributed around and can lead to subsequent relaxations. This is how dynamic facilitation takes place. A rough argument suggests that sites at linear distance $\ell < \xi(T)$, where $[1/\xi^d(T)]/T \sim O(1)$, are dynamically correlated as the induced change in their local barrier can affect their relaxation time substantially. Hence, following Ref. [69], one finds a peak of $\chi_4 \sim \xi^d \sim (1/T) \sim \log \tau_\alpha$. This conjectured behavior of χ_4 is in qualitative agreement with our numerical findings, and it indeed leads to bending in the log-log plot of χ_4 versus τ_α (see also SM [43]). The mechanism described above for DH is different from the ones at play in KCMs [11] and argued to hold for RFOT [62,63]. Relaxation in EPM-Q is due to a combination of activation and elasticity instead of subdiffusion of rare conserved defects [11]. In this new mechanism, avalanches of motion have a finite size and appear intermittently [70]. Determining from atomistic simulations of glass-forming liquids which one of these mechanisms holds is a very interesting open challenge, see, e.g., Refs. [6–8].

In summary, we have shown that the simple EPM-Q model, which encodes "elasticity-induced facilitation," is able to reproduce the salient features associated with the growth of dynamical correlation in glass-forming liquids. Our results offer new perspectives on the theories of the glass transition, in particular on the theoretical proposals describing the dynamics of supercooled liquids as "solids that flow" [13,25,71–73]. The main difficulty of those scenarios was that they were thought to be unable to explain the emergence of dynamical heterogeneity and dynamical correlations, particularly the growth of the four-point correlation function that is the central observable measured in experiments [61] and molecular simulations [60]. We have shown concretely in a simple model that it is the elastic interaction between local relaxations on top of the solid state that provides the emergence of dynamic facilitation and leads to dynamical heterogeneity even in models with local barriers, consolidating molecular simulation studies [9,14,22]. Some other important facts are instead missed in our approach, but as we argue below, more realistic versions of the model should be able to capture them.

For instance, DH is not only associated with heterogeneity in space but also with stretched exponential relaxation [74]. In the EPM-Q presented in this study, there is no heterogeneity (or disorder) in the solid state, as all the parameters associated with local relaxation events [σ_c, z_0, and K in Eq. (1)] are the same on all sites. In a more realistic version, which considers the disorder of the amorphous solid [66], they should be random variables to be redrawn after a local relaxation. This would lead to a more heterogeneous distribution of barriers and hence of relaxation times, providing a possible mechanism for stretched relaxation. Furthermore, a more realistic model should also consider that local stress relaxation is not instantaneous [75], and that a complete description should be tensorial instead of scalar [76–78].

Another important generalization concerns the behavior of the relaxation time and the nature of the "local relaxation event." Even if the elementary dynamical process were truly local and noncooperative, the typical value of the local energy barriers could depend on the temperature [13]. Taking into account that elastic constants correlate with slow dynamics [13,71–73,79], introducing a realistic T dependence of K and σ_c would provide a simple way to describe DH and super-Arrhenius behavior at the same time. Another possibility is that the single (mesoscopic) site relaxation process of our model could actually correspond to a cooperative many-particle rearrangement as the one envisioned to take place in RFOT. Within this perspective, the local degrees of freedom in our model correspond to what has been called *activons* (rearrangements on the point-to-set correlation length scale) in Ref. [5] and are represented as local traps in Refs. [6,42]. It would certainly be interesting to estimate the typical coarse-graining length scale associated with a local relaxation event (corresponding to a single site in the model). This could be done by measuring the size of shear transformation zones [39,80] or by a mapping from a molecular simulation to an elastoplastic model [81–83]. Moreover, in order to study the connection with KCMs thoroughly, in particular with models with softened kinetic constraint [84], it would be interesting to study whether EPM-Q-like models present the space-time dynamical transition predicted in the dynamic facilitation scenario [85], and how it relates to the physical ingredients of EPM-Q, such as elasticity and local structural disorder [86].

EPMs have been fruitfully used to develop a scaling theory of avalanches in sheared amorphous solids [29]. The EPM-Q model paves the way for an analogous study of the avalanches of motion in supercooled liquids [87] which will allow a thorough comparison with the in-depth characterization of DH of Ref. [6].

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