

Field-induced superdiffusion and dynamical heterogeneity

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(Received 21 April 2016; published 30 June 2016)

By analyzing two kinetically constrained models of supercooled liquids we show that the anomalous transport of a driven tracer observed in supercooled liquids is another facet of the phenomenon of dynamical heterogeneity. We focus on the Fredrickson-Andersen and the Bertin-Bouchaud-Lequeux models. By numerical simulations and analytical arguments we demonstrate that the violation of the Stokes-Einstein relation and the field-induced superdiffusion observed during a long preasymptotic regime have the same physical origin: while a fraction of probes do not move, others jump repeatedly because they are close to local mobile regions. The anomalous fluctuations observed out of equilibrium in the presence of a pulling force ϵ , $\sigma_x^2(t) = \langle x_\epsilon^2(t) \rangle - \langle x_\epsilon(t) \rangle^2 \sim t^{3/2}$, which are accompanied by the asymptotic decay $\alpha_\epsilon(t) \sim t^{-1/2}$ of the non-Gaussian parameter from nontrivial values to zero, are due to the splitting of the probes population in the two (mobile and immobile) groups and to dynamical correlations, a mechanism expected to happen generically in supercooled liquids.

DOI: 10.1103/PhysRevE.93.060105

Superdiffusion in the presence of an external driving is one of the most intriguing results of microrheological numerical experiments in supercooled liquid [1–3] and of experimental studies on glassy granular media [4]. In supercooled liquids the mean square displacement (MSD) of a tagged particle displays a characteristic intermediate plateau of increasing length when the temperature is lowered, due to caging, while asymptotically the diffusion is always Fickian, $\langle x^2(t) \rangle \sim t$. The surprising finding of [1,2] is that, notwithstanding the slowing down of the dynamics, at low enough temperatures the action of an external force on a probe particle produces a superdiffusive spreading of the probability distribution of displacements: $\sigma_x^2(t) = \langle x_\epsilon^2(t) \rangle - \langle x_\epsilon(t) \rangle^2 \sim t^\gamma$, where $\gamma > 1$ and ϵ is the force acting on the probe. This *anomalous* behavior of the MSD around the drift is really a landmark of non-Fickian diffusion: in the case of Fickian diffusion even the MSD around the drift grows linearly in time, $\sigma_x^2(t) \sim t$, as does the unbiased one. What is the mechanism which triggers a faster-than-Fickian diffusion within an environment with large and broadly distributed relaxation times? At first sight this looks indeed as a quite counterintuitive behavior. The goal of this paper is to provide a clear-cut explanation of the mechanism triggering anomalous diffusion, which in the models studied will appear as a preasymptotic effect, and to show that this transient anomaly in the dynamics is intimately related to dynamical heterogeneities and to the breaking of the Stokes-Einstein relation.

The Stokes-Einstein (S-E) relation connects the diffusion coefficient $D(\beta)$ of a probe to the relaxation time $\tau_{\text{eq}}(\beta)$ of the sample as $D(\beta)\tau_{\text{eq}}(\beta) = \text{const}$ [5]. The physical reason for such a relation is that simple liquids are characterized by a single relevant time scale τ_D : this time scale characterizes the behavior of the system at all scales, from the single molecule diffusion, $D(\beta) \sim \tau_D^{-1}(\beta)$, to the relaxation of the sample $\tau_{\text{eq}}(\beta) \sim \tau_D(\beta)$, which explains the Stokes-Einstein relation. Close to the glass-transition temperature T_g several

time scales appear in the dynamics of the system. In this regime the characteristic diffusion time $\tau_D(\beta) \sim D^{-1}(\beta)$ decouples from the relaxational time, $\tau_{\text{eq}}(\beta) \approx \tau_D(\beta)$, so that one also finds $D(\beta)\tau_{\text{eq}}(\beta) \neq \text{const}$ [6]: this is the Stokes-Einstein violation. A particularly instructive and useful rationalization of this phenomenon was obtained studying kinetically constrained models (KCM) [7–15]. These are lattice models where a mobility field with local update rules subjected to kinetic constraints reproduces the sluggish and heterogeneous dynamics of glasses. The heterogeneous nature of space-time correlations is explained in KCMs in terms of *defects* dynamics. The activity field of KCMs is characterized by rare *mobility* defects which, wandering around in the system, trigger the relaxation of the whole sample. We show here that this very same mechanism also leads to the anomalous transport properties observed in [1,2]. Looking at the motion of a driven intruder, the heterogeneous nature of the medium becomes manifest only in the *out-of-equilibrium* fluctuations $\sigma_x^2(t)$; this is the case also for the diffusion on hierarchical lattices discussed in [16], for some continuous time random walk (CTRW) models [17], and for the field-induced superdiffusion of a tracer in a crowded medium discussed in [18].

We study the driven dynamics of a tracer particle in two kinetically constrained models: the one-dimensional Fredrickson-Andersen (FA) [7–9] and Bertin-Bouchaud-Lequeux (BBL) models [10,13]. Both have been studied and used as models of supercooled liquids. The local structure in FA is described by a binary variable $n_i \in \{0,1\}$: sites with $n_i = 1$ are active while those with $n_i = 0$ are inactive. The update of n_i is possible only when at least one of its neighbors is already active, namely, one needs $n_{i+1} = 1$ or $n_{i-1} = 1$. When possible, the update $1 \rightarrow 0$ is always accepted, while $0 \rightarrow 1$ takes place with probability $e^{-\beta}$, where β is a dimensionless inverse temperature. The dynamics obeys detailed balance, with an energy function $E = \sum_i n_i$, so that the equilibrium state has no correlations between sites. The FA model exhibits

nontrivial correlated dynamics for $\beta \gtrsim 1$. The concentration of active sites is $c = \langle n_i \rangle = [1 + e^\beta]^{-1}$. Lengths are in units of the lattice spacing which we set to unity. The BBL model is described by a continuous variable: the density of mass $\rho_i \in [0, \infty)$. The elementary step of the BBL dynamics is the simultaneous update of the density in a couple of neighboring sites ρ_i and ρ_j : $\rho'_i = q(\rho_i + \rho_j)$ and $\rho'_j = (1 - q)(\rho_i + \rho_j)$, with $q \in [0, 1]$. The update is possible only when the densities of the two sites fulfill the constraint $(\rho_i + \rho_j)/2 < \rho_{th} = 1$: the BBL is a kinetically constrained mass transport model. The random variable q , which introduces stochasticity in the dynamics of the density field, is extracted from a distribution $\psi_\mu(q)$ characterized by the parameter μ . In the present study we consider the value $\mu = 0.3$, which produces a diffusive dynamics of the mobility defects, which are represented by the *active* links where the kinetic constraint is fulfilled. Details on the dynamics of the mobility defects for different values of μ can be found in [10,13]. In the BBL model the definition of *active* links is naturally encapsulated into the definition of the model. In the FA model a link between two sites i and $i + 1$ is *active* when both are active, $n_i = n_{i+1} = 1$.

Following [11,12] we model microrheological experiments by assuming that the driven tracer can only move between two adjacent sites when these form an active link. Since the updates of n_i in FA and of the mass field ρ_i in BBL do not depend on the position of the probe, the latter behaves as a *ghost* particle: it is influenced by the background but has no influence on it. In order to induce a directed motion of the probe we unbalance in both models the probability of its forward and backward displacements: $p_\epsilon^{\rightarrow} = 1/2 + \epsilon$, $p_\epsilon^{\leftarrow} = 1/2 - \epsilon$, with $\epsilon \in [-1/2, 1/2]$. The dynamics of a ghost probe in both the FA and the BBL can be then fully understood in terms of the mobility defects dynamics [8,11]. The dynamics of the probe is ruled by two relevant time scales: the mean *persistence* time, which is the time the probe has to wait on average before being hit for the first time by a defect, and the average *exchange* time, which is the time between two successive meetings with a defect. The difference between these two time scales is both the signature of heterogeneous dynamics and the key ingredient of the anomalous transport of a probe. There is only one difference for the defect dynamics of the two models: while in the BBL the diffusion coefficient of defects does not depend on their concentration c , in the FA model the diffusion of mobile defects depends on the temperature and hence on their concentration. Henceforth, in order to present a unified discussion of the two models, time is measured in the units of τ_0 , which is the time scale on which a mobile defect moves one step ($1/c$ and 1 for the FA and the BBL models, respectively).

Assuming that defects are independent random walkers the dynamics of the probe can be described as a CTRW [11]. In this approximation the histogram of probe displacements, which corresponds to the self part of the van Hove function $G_s(r, t) = \langle \delta(r - [x(t) - x(0)]) \rangle$ (the angular brackets indicate the average over different trajectories), can be exactly written with the following formula [11]:

$$G_s(x, t) = \mathcal{P}(t)\delta(x) + \int_0^t p(t-s)P_{1st}(x, s)ds. \quad (1)$$

In Eq. (1) the probability of persistence times is denoted by $p(t-s)$, $P_{1st}(x, s)$ is the propagator for trajectories which start with a jump event (the subscript 1st indicates that at least one displacement took place) and $\mathcal{P}(t)$ is the persistence function, i.e., the probability that a probe is not hit by a mobility defect for a duration of t when the observation starts at an arbitrary time. From Eq. (1) is clear that the population (or the probability) is split into two groups: probes which at time t have already started to move and probes which at t are still at rest (we refer to a population of probes since we can think of having many probes evolving in parallel and starting from random positions). Since in both FA and BBL models defects behave as independent random walkers, persistence equals the survival probability $\mathcal{P}(t) = e^{-c\sqrt{t}}$, with c the concentration of walkers [19]. The distribution of persistence times $p(t)$ which enters Eq. (1), is obtained as $p(t) = -d\mathcal{P}(t)/dt \propto ce^{-c\sqrt{t}}/\sqrt{t}$. The distribution of exchange times, which is in turn proportional to $-dp(t)/dt$ [12], reads to leading order in c as $\psi(\tau) \propto ce^{-c\sqrt{\tau}}/\tau^{3/2}$. The representation of the probe dynamics as a CTRW is very insightful and will be our main tool to understand the relationship between dynamic heterogeneity and anomalous diffusion of a driven tracer. For both the FA and the BBL models the relaxation time scale τ_{eq} is defined by the stretched-exponential cutoff of the persistence and exchange time distributions: $\tau_{eq}/\tau_0 = c^{-2}$.

We now present our numerical results about anomalous diffusion in the FA and BBL models, which are shown in Figs. 1 and 2. By looking at Fig. 1 we notice that a driven probe ($\epsilon = 1/2$) has a linear drift in both FA and BBL, but at the same time the non-Gaussian parameter $\alpha_\epsilon(t)$ for noncentered distributions signals important deviations from Gaussianity up

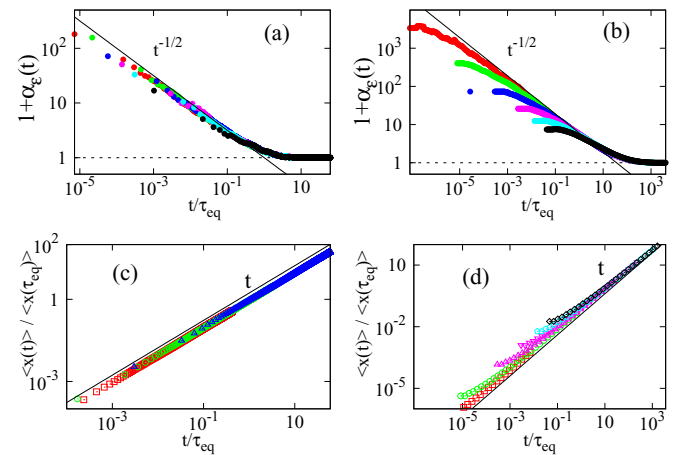


FIG. 1. Data collapse for the non-Gaussian parameter $\alpha_\epsilon(t)$ obtained plotting $1 + \alpha_\epsilon(t)$ vs t/τ_{eq} for the different concentrations of mobility defects (different symbols) in (a) FA and (b) BBL. The continuous straight line emphasizes the behavior $\alpha_\epsilon(t) \sim t^{-1/2}$ in the preasymptotic regime. The different concentrations of mobility defects are $c = 10^{-2}$, 6.7×10^{-3} , 5.2×10^{-3} , 3.8×10^{-3} , 2.8×10^{-3} , and 1.9×10^{-3} for the FA model; $c = 2 \times 10^{-1}$, 1.1×10^{-1} , 5.1×10^{-2} , 1.6×10^{-2} , 2.7×10^{-3} , and 2.8×10^{-4} for the BBL model. Also shown is the drift of the probe in (c) FA and (d) BBL; data are at the same concentrations of mobility defects of (a) and (b). Collapse is obtained plotting $\langle x_\epsilon(t) \rangle / \langle x_\epsilon(\tau_{eq}) \rangle$ vs t/τ_{eq} .

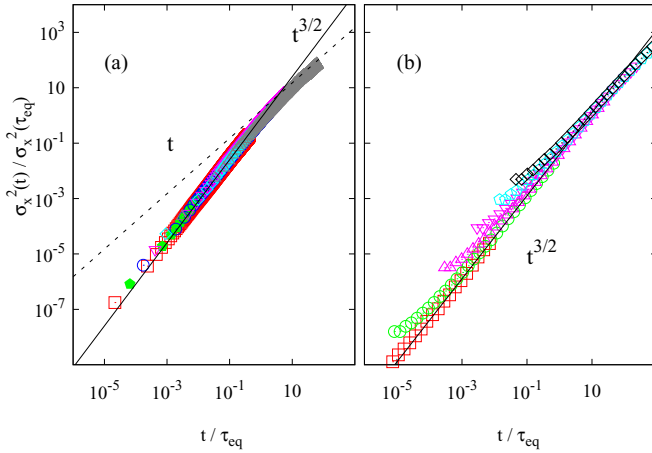


FIG. 2. Mean square displacement around the drift $\sigma_x^2(t)$ for a driven probe ($\epsilon = 1/2$) in (a) FA and (b) BBL. Different symbols represent different concentrations c of the mobility defects, which are, respectively for the two models, the same as in Fig. 1. Collapse of the curves is obtained by plotting $\sigma_x^2(t)/\sigma_x^2(\tau_{\text{eq}})$ vs t/τ_{eq} . Full line is the $t^{3/2}$ scaling; the dashed line in (a) is the Fickian behavior $\sigma_x^2(t) \sim t$.

to $t \sim \tau_{\text{eq}}$. For a Gaussian distribution with mean $\langle x_\epsilon(t) \rangle$ and variance $\sigma_x(t)$ the fourth noncentered moment reads $\langle x_\epsilon^4(t) \rangle = \langle x_\epsilon(t) \rangle^4 + 6\langle x_\epsilon(t) \rangle^2 \sigma_x^2(t) + 3\sigma_x^4(t)$, which allows us to define $\alpha_\epsilon(t)$ as

$$\alpha_\epsilon(t) = \frac{\langle x_\epsilon(t) \rangle^4}{\langle x_\epsilon(t) \rangle^4 + 6\langle x_\epsilon(t) \rangle^2 \sigma_x^2(t) + 3\sigma_x^4(t)} - 1, \quad (2)$$

where $\sigma_x^2(t) = \langle x_\epsilon^2(t) \rangle - \langle x_\epsilon(t) \rangle^2$. It can be easily seen that with zero drift the standard definition of the non-Gaussian parameter is recovered. For aesthetic reasons in Fig. 1 we plotted $1 + \alpha_\epsilon(t)$. Equation (1) tells us that the overall drift comes from the convolution of the drift of the moving probes with the distribution of persistence times:

$$\langle x_\epsilon(t) \rangle = \int_0^t ds p(t-s) \langle x_\epsilon(s) \rangle_{1\text{st}}. \quad (3)$$

From the inspection of Eq. (3) it is possible to single out the different physical mechanisms [20] which determine the linear behavior of the drift, $\langle x_\epsilon(t) \rangle \sim t$, in the two regimes $1 \ll t \ll \tau_{\text{eq}}$ and $\tau_{\text{eq}} \ll t$. In the latter, due to the exponential cutoff of $\psi(\tau)$, for $t \gg \tau_{\text{eq}} = c^{-2}$ the drift $\langle x_\epsilon(s) \rangle_{1\text{st}}$ is linear. In this regime one can approximate $\int_0^t ds p(t-s) \langle x_\epsilon(s) \rangle_{1\text{st}} \sim (1 - \mathcal{P}(t)) \langle x_\epsilon(t) \rangle_{1\text{st}} \sim \langle x_\epsilon(t) \rangle_{1\text{st}}$, which shows that the total drift is also linear. On the contrary, in the former regime (which we will call “preasymptotic” henceforth), persistence and exchange distributions can be approximated by power-law distributions: $\psi(\tau) \sim \tau^{-3/2}$ and $p(t) \sim t^{-1/2}$. This leads to a subdiffusion of the moving probe as $\langle x_\epsilon(t) \rangle_{1\text{st}} \sim \sqrt{t}$. The physical reason is that moving probes are repeatedly hit by a mobile defect $t^{1/2}$ times. On the other hand the fraction of moving probes increases too, as $c\sqrt{t}$, due to the heavy tail of $p(t)$. How these two effects combine can be read in the explicit expression of Eq. (3) in the regime $t \ll \tau_{\text{eq}}$, i.e., $\langle x_\epsilon(s) \rangle \sim c \int_0^t ds (t-s)^{-1/2} \sqrt{s}$; the change of variable $s \rightarrow s/t$ in the last integral yields immediately $\langle x_\epsilon(s) \rangle \sim ct$.

It is due to this nontrivial mechanism that, even in the preasymptotic regime, we can observe a linear drift.

Figure 2 shows then that the non-Gaussianity of $G_\epsilon(x, t)$ is manifest in the transport properties of the intruder when one looks at the MSD around the drift $\sigma_x^2(t) = \langle x_\epsilon^2(t) \rangle - \langle x_\epsilon(t) \rangle^2$: it grows superdiffusively as $\sigma_x^2(t) \sim t^\gamma$ with $\gamma \approx 3/2$ for $1 \ll t \ll \tau_{\text{eq}}$. The exponent $3/2$ is the same which characterizes the field-induced superdiffusion of a tracer in a crowded medium [18], while the faster-than-linear spreading of the van Hove function that we point out is clearly related to force-induced giant diffusivity discussed in [14]. The population splitting scenario, already well established in the context of supercooled liquid models [11,14,21] and recently discussed also in the context of CTRW with aging dynamics [22,23], allows one to perfectly understand both qualitatively and quantitatively not only the observed superdiffusion (Fig. 2), but also the preasymptotic behavior of the non-Gaussian factor (Fig. 1).

From the definition of $G_s(x, t)$ in Eq. (1) we have that the MSD around the drift reads

$$\sigma_x^2(t) = -\langle x_\epsilon(t) \rangle^2 + \int_0^t ds p(t-s) \langle x_\epsilon^2(s) \rangle_{1\text{st}}. \quad (4)$$

We already know that in the preasymptotic regime $\langle x_\epsilon(t) \rangle \sim ct$, so that we only need to know $\langle x_\epsilon^2(s) \rangle_{1\text{st}}$. The latter is obtained from the asymptotic behavior of moments for a biased CTRW [24–27]. In a CTRW with waiting time distribution $\psi(\tau) \sim \tau^{-(1+\beta)}$ and $0 < \beta < 1$ it holds $\langle x_\epsilon^2(t) \rangle \sim t^{2\beta}$; since in our case $\beta = 1/2$, we have $\langle x_\epsilon^2(s) \rangle_{1\text{st}} \sim s$. By plugging this last result into Eq. (4) and retaining the leading contribution to $p(t-s)$ when $t \ll \tau_{\text{eq}}$, one finds

$$\sigma_x^2(t) \sim c \int_0^t ds \frac{s}{\sqrt{t-s}} \sim ct^{3/2}, \quad (5)$$

where the asymptotic behavior of the integral has been evaluated by simply changing the variable $s \rightarrow s/t$. The term $\langle x_\epsilon(t) \rangle^2 = O(c^2)$ appearing in Eq. (4) has been dropped in Eq. (5) because it is subleading in the regime $c \ll 1$, $t \ll \tau_{\text{eq}}$. We obtained analytically the superdiffusive behavior tracing it back to the subsequent hits with the mobile defects, which are in turn encoded in the heavy tails of $\psi(\tau)$. To estimate the preasymptotic behavior of the non-Gaussian parameter $\alpha_\epsilon(t)$ we need to know the fourth-order noncentered moment, which from Eq. (1) reads

$$\langle x_\epsilon^4(t) \rangle = \int_0^t ds \frac{c}{\sqrt{t-s}} \langle x_\epsilon^4(s) \rangle_{1\text{st}} \sim ct^{5/2}. \quad (6)$$

The scaling in Eq. (6) comes again from the asymptotic behavior of moments in a CTRW characterized by the distribution of waiting times $\psi(\tau) \sim \tau^{-(1+\beta)}$ and $0 < \beta < 1$: $\langle x_\epsilon^4(s) \rangle \sim s^{4\beta}$ [25,26]. Since in our case $\beta = 1/2$ we have $\langle x_\epsilon^4(s) \rangle \sim s^2$. For small values of c the denominator of the non-Gaussian parameter reads at leading order

$$\langle x_\epsilon(t) \rangle^4 + 6\langle x_\epsilon(t) \rangle^2 \sigma_x^2(t) + 3\sigma_x^4(t) \sim 3c^2 t^3, \quad (7)$$

so that, combining Eq. (6) with Eq. (7) we find

$$\alpha_\epsilon(t) \sim c^{-1} t^{-1/2}, \quad (8)$$

which is perfectly consistent with the numerical behavior of $\alpha_\epsilon(t)$ shown in Fig. 1.

Strong anomalous diffusion takes place when the scaling assumption $G_s(x,t) = \mathcal{F}[x/\ell(t)]/\ell(t)$ for the van Hove function and the scaling of moments $\langle x^n(t) \rangle \sim \ell^n(t)$ cannot be written in terms of a single length $\ell(t)$ [24]. In the present case (FA and BBL) this phenomenon takes place due to population splitting. Looking at the distribution of moving probes we have found the numerical evidence (not shown) that $P_{1st}(x_\epsilon, t)$ is a half-Gaussian: at different times a perfect collapse of data is obtained with $\ell(t) = t^{1/2}$. Taking otherwise into account the whole population of probes, i.e., also the contribution $\mathcal{P}(t)\delta(x)$, in Eq. (1), of probes which never jumped up to time t , we have $\sigma_x^2(t) \sim t^{3/2} \neq \ell^2(t) = t$: diffusion is *strongly* anomalous [24]. Let us stress that this strong anomalous diffusion is not due, as usual, to a multiscaling property of the probability distribution of displacements [24]: it comes from the splitting of probes population into slow persistent ones, not moving roughly until $t \sim \tau_{eq}$, and those which at $t \ll \tau_{eq}$ have already been repeatedly hit by mobility defects. Precisely the same mechanism is at the origin of the S-E violation $D\tau_{eq} = e^{-\beta}$: a probe diffuses much more than one step on the relaxation time scale due to repeated interactions with the same mobile defect. Such a mechanism is also responsible for dynamic heterogeneity: all regions that relax because they are hit by the same mobile defect within the time scale τ_{eq} become dynamically correlated. We have therefore shown that dynamic correlations, violation of S-E, and *strong* anomalous superdiffusion are directly connected in KCMs [11] (see also [16]). While on the one hand we expect field-induced superdiffusion to be general in systems where a population-splitting phenomenon of the kind discussed here is taking place, on the other hand the precise value of the superdiffusive exponents in a generic model may depend on the force ϵ pulling the probe (see [2]). Our choice to study both in the FA and the BBL models the dynamics of a “ghost” probe, i.e., a probe which does not have any influence on the background mobility field, was done precisely to get rid of this—in general to be expected—dependence of the

superdiffusive exponent on ϵ . In the presence of a ghost probe neither the persistence time distribution $p(t)$ nor the exchange time distribution $\psi(\tau)$ depend on ϵ ; because of that, even the exponents of the anomalous dynamics are not carrying any dependence on ϵ .

In conclusion we have related the superdiffusive behavior of driven probes to the splitting of their population in frozen ones and moving ones repeatedly hit by the same mobility defect. Our analysis provides an explanation of the results found in atomistic models [1,2] and link them to the phenomena of dynamic heterogeneity and Stokes-Einstein violation. Furthermore, it offers a theoretical derivation of the superdiffusion exponent $3/2$. This value, compatible with our numerical results for both the FA and BBL models, is also surprisingly close to the value 1.45 found in the supercooled Yukawa mixture of [2]. Also remarkable is the qualitative agreement between the behavior of the non-Gaussian factor $\alpha_\epsilon(t)$ characterizing the driven preasymptotic dynamics of a probe in one-dimensional FA and BBL models and the one found for the unbiased dynamics of a probe in a three-dimensional supercooled Lennard-Jones mixture (see Fig. 3 of [1]). All these similarities point toward the presence of universal features, still to be investigated, which are intrinsically related to dynamical heterogeneity and emerge at low temperatures independently of the dimensionality and specific interactions of the models. We expect indeed our findings to hold generically beyond the simple models we focused on. In fact, the key ingredients are the population-splitting scenario and the anomalous diffusion of mobile probes induced by dynamic correlations, which are phenomena known to be present generically in supercooled liquids.

We wish to thank E. Barkai, J.-L. Barrat, O. Bénichou, and A. Heuer for useful discussions and comments. G.G. acknowledges support from the ERC Grant GLASSDEF No. ADG20110209. G.B. acknowledges support from the ERC Grant NPRGGLASS.

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