Geometry-Induced Superdiffusion in Driven Crowded Systems

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Recent molecular dynamics simulations of glass-forming liquids revealed superdiffusive fluctuations associated with the position of a tracer particle (TP) driven by an external force. Such an anomalous response, whose mechanism remains elusive, has been observed up to now only in systems close to their glass transition, suggesting that this could be one of its hallmarks. Here, we show that the presence of superdiffusion is in actual fact much more general, provided that the system is crowded and geometrically confined. We present and solve analytically a minimal model consisting of a driven TP in a dense, crowded medium in which the motion of particles is mediated by the diffusion of packing defects, called vacancies. For such nonglass-forming systems, our analysis predicts a long-lived superdiffusion which ultimately crosses over to giant diffusive behavior. We find that this trait is present in confined geometries, for example long capillaries and stripes, and emerges as a universal response of crowded environments to an external force. These findings are confirmed by numerical simulations of systems as varied as lattice gases, dense liquids, and granular fluids.

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Active microrheology monitors the response of a medium while in the presence of a tracer particle (TP) manipulated by an external force. It has become a powerful experimental tool for the analysis of different systems such as colloidal suspensions [1–5], glass forming liquids [6–10], fluid interfaces [11], or live cells [12,13]. It constitutes a striking realization of a key problem in statistical physics which in a much broader context aims at determining the response of a medium to a perturbation created by a driven TP [14–16]. A considerable amount of knowledge has been gathered on the forms of the so-called force-velocity relation, that being the dependence of the TP velocity v on the value of the applied force F, both in the linear and the nonlinear response regimes.

Behavior beyond the force-velocity relation was recently addressed numerically in the pioneering work of Ref. [7], which studied via molecular dynamics simulations the dynamics of an externally driven, or biased TP in a glass-forming liquid (a dense binary Yukawa liquid). It was recognized that while the TP moves ballistically, i.e., $\langle X_t \rangle \sim vt$, the variance $\sigma_x^2 = \langle (X_t - \langle X_t \rangle)^2 \rangle$ of the TP position X_t along the bias grows surprisingly in a superdiffusive manner with respect to time t, so that $\sigma_x^2 \sim t^\lambda$, where λ is within the range 1.3–1.5. For such systems, this effect was found *only* in the close vicinity of the glass transition while regular diffusion was recovered away from the transition [8], suggesting that such anomalous behavior could be a distinct feature of being close to the glass transition.

A number of attempts have been made to explain these findings, based either on a random trap model [7], mode coupling theory [8], or continuous-time random walks (CTRWs) [10]. All of these approaches rely on the notion of a complex energy landscape and thereby assume that the system is close to the glass transition. However, they do not provide a quantitative nor qualitative understanding of the superdiffusive behavior. In particular, the question of whether superdiffusion is the ultimate regime or only a transient is still open [9,10].

Here, we show that in fact superdiffusion in active microrheology settings can appear away from the glass transition, and even *independently* of glassy properties. Based on a simple model that does not involve any complex energy landscape or kinetic constraints, we demonstrate that superdiffusion emerges universally in confined crowded systems. We fully quantify this superdiffusion, show that it is long-lived, highlight the key role of the system's geometry, and provide a clear physical mechanism underlying such behavior.

Our starting point is a minimal model of a crowded system in which the motion of particles and of the TP is mediated by so-called "diffusive" packing defects, or vacancies that are sufficiently large in size to allow their diffusive motion to proceed through direct swapping of their positions with the host medium particles. Note that the existence of such defects is tacitly assumed in various models of crowded systems [17–19]. Within this picture, the TP moves only when a defect arrives to its location.

More precisely, we consider a lattice gas model where the particles in the medium perform symmetric random walks on a d-dimensional lattice constrained by hard-core interactions between the particles, so that there is at most one particle per lattice site. The TP performs a random walk biased by an external applied force $\mathbf{F} = F\mathbf{e}_1$ (see Fig. 1 and the Supplemental Material [20] for a detailed description of the dynamics), hindered by hard-core interactions with the host particles. Note that this description represents a combination of two paradigmatic models of nonequilibrium statistical mechanics: asymmetric (for the TP) and symmetric (for the host particles) simple exclusion processes [21]. Until now, only the force-velocity relation has been analyzed for such a model [22,23], with the exception of single file systems for which the variance has been calculated [24,25] and infinite 2D systems, where, however, only a particular limit of the variance was considered [26,27] (see also Ref. [28] for qualitative arguments in various geometries). Below, we describe the main steps of a method of calculation that allows us to determine the full dynamics of the variance in dense systems. We present the results and highlight their physical meaning in the main text, while we provide the technical details of calculations in the Supplemental Material [20].

The first step of the calculation consists in solving an auxiliary problem that involves a single vacancy. It relies on the remark done in Ref. [29] that for 2D systems, in the absence of driving force and for small values of the density of vacancies ρ_0, the dynamics of the TP can be deduced from the analysis of the joint dynamics of the TP and a single isolated vacancy. As a matter of fact, this is still true for a biased TP in any dimension. In the Supplemental Material [20], we show how to account for both the nontrivial waiting time distribution and the anticorrelation

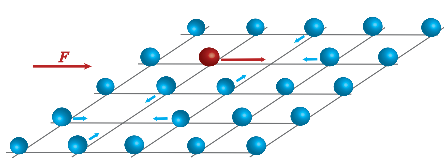

FIG. 1 (color online). The model; a discrete lattice in which sites are occupied by identical hard-core medium particles (blue spheres). The red sphere denotes the tracer particle (TP) which, in addition to hard-core interactions, is subject to an external force $\mathbf{F} = F\mathbf{e}_1$, and thus has asymmetric hopping probabilities. The arrows of different size depict schematically the hopping probabilities; a larger arrow near the TP indicates that it has a preference for moving in the direction of the applied field. Jumps are possible only when a vacancy (in concentration ρ_0) is adjacent to a particle.

effects between consecutive steps of the driven tracer, which are in fact induced by the dynamics of a single diffusing vacancy. The determination of the propagator of the TP in the presence of a single vacancy is shown to reduce to the calculation of first-passage time distributions of this vacancy at the site occupied by the TP. It is important to realize that the vacancy itself performs a simple random walk, symmetric everywhere except in the vicinity of the TP, because of the presence of the bias. These first-passage time distributions can be determined by using standard methods of random walks with defective sites [30]. In the second step of the calculation, the variance of the TP in the presence of the density of vacancies ρ_0 is deduced from the knowledge of both the single vacancy propagator and the first-passage time distributions mentioned above. Such exact asymptotic expressions of the variance σ_x^2 are finally obtained for various geometries and for arbitrary values of the dimensionless force $f = \beta\sigma F$ (where σ is the lattice step and β is the reciprocal temperature). These are valid at large times and low vacancy densities, and are summarized below.

Superdiffusive regime.—First, our approach predicts the following large-t behavior of the variance σ_x^2 in the leading order of ρ_0:

$$\lim_{\rho_0\to 0}\frac{\sigma_x^2}{\rho_0}\underset{t\to\infty}{\sim} 2a_0^2 t \times \begin{cases} (4/3\sqrt{\pi}L)t^{1/2} & \text{2D stripe,} \\ (2\sqrt{2/3\pi}/L^2)t^{1/2} & \text{3D capillary,} \\ \pi^{-1}\ln(t) & \text{2D lattice,} \\ A + \coth(f/2)/(2a_0) & \text{3D lattice,} \end{cases} \tag{1}$$

where a_0 is an f-dependent constant

$$a_0 = \frac{\sinh(f/2)}{\cosh(f/2)[1 + \frac{2d\alpha}{2d-\alpha}] + d - 1}, \tag{2}$$

$A = \hat{P}(\mathbf{0}|\mathbf{0};1) + 2(13\alpha - 6)/[(2+\alpha)(\alpha - 6)]$, d is the system dimension, $\alpha = \lim_{\xi\to 1^-}[\hat{P}(\mathbf{0}|\mathbf{0};\xi) - \hat{P}(2\mathbf{e}_1|\mathbf{0};\xi)]$, and $\hat{P}(\mathbf{r}|\mathbf{r}_0;\xi)$ is the generating function (discrete Laplace transform) of the propagator of a symmetric simple random walk (see the Supplemental Material [20] for the explicit expressions). These surprisingly simple exact expressions unveil the dependence of the variance on time, width L of the stripe or of the capillary, and on the reduced driving force f. Figure 2 shows an excellent quantitative agreement between the analytical predictions and the numerical simulations: The time, width, and driving force dependences are unambiguously captured by our theoretical expressions.

A number of important conclusions can be drawn from this result. (i) Superdiffusion with an exponent $\lambda = 3/2$ takes place in confined, quasi-1D geometries, those being, infinitely long 3D capillaries and 2D stripes. This result is quite counterintuitive: indeed, in the absence of driving force it is common to encounter diffusive, or even

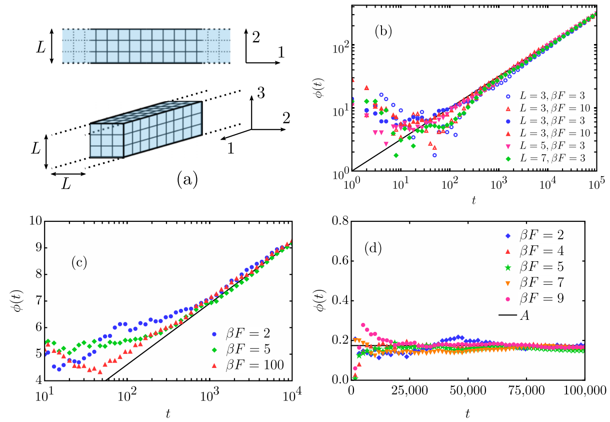

FIG. 2 (color online). Studied geometries and reduced variance as a function of time in the superdiffusion regime. (a) Sketch of stripe- and capillarylike geometries. (b) Simulations in capillaries [empty symbols, $\phi(t) = \sqrt{3\pi/2}L^2/(4a_0^2\rho_0 t)\sigma_x^2(t)$] and stripes [filled symbols, $\phi(t) = 3\sqrt{\pi}L/(8a_0^2\rho_0 t)\sigma_x^2(t)$] with density $\rho_0 = 10^{-5}$, and theoretical prediction (solid line, $\sqrt{t}$). (c) Simulations on a 2D lattice with density $\rho_0 = 10^{-5}$ and $\phi(t) = \frac{\pi}{2a_0}[\sigma_x^2(t)/(\rho_0 a_0 t) - (2a_0/\pi)(\ln 8 + \gamma - 1) - 2a_0\pi(5-2\pi)/(2\pi - 4) - \coth(f/2)]$ and theoretical prediction (solid line, $\ln t$). (d) Simulations on a 3D lattice with density $\rho_0 = 10^{-6}$ and $\phi(t) = [\sigma_x^2(t)/(\rho_0 t) - a_0\coth(f/2)]/(2a_0^2)$ and theoretical prediction [solid line: A, defined after Eq. (1)].

subdiffusive growth of the fluctuations of the TP position in such crowded molecular environments [19], however, not superdiffusion. (ii) The superdiffusion in such systems emerges beyond (and therefore cannot be reproduced within) the linear response-based approaches: The prefactor in the superdiffusive law is proportional to f^2 when $f \to 0$. Despite the presence of the superdiffusion, the Einstein relation is nonetheless valid for systems of arbitrary geometry due to subdominant (in time) terms whose prefactor is proportional to f. (iii) In unbounded 3D systems σ_x^2 grows diffusively and not superdiffusively. (iv) For $d = 1$ (single files), one finds $\alpha = 2$, so that $a_0 = 0$, and no superdiffusion can take place, in agreement with [25]. As a matter of fact, in this case the variance grows subdiffusively. Finally, this shows that superdiffusion is geometry-induced and the recurrence of the random walk performed by a vacancy is a necessary but not sufficient condition in order for superdiffusion to occur.

Giant diffusion regime.—The exact analytical result in Eq. (1) provides explicit criteria for superdiffusion to occur. Technically, this yields the behavior of the variance when the limit $\rho_0 \to 0$ is taken before the large-t limit. It, however, does not allow us, due to the nature of the limits involved, to answer the question whether the superdiffusion is the ultimate regime (or just a transient), which requires the determination of $\lim_{t\to\infty}\sigma_x^2$ at fixed ρ_0. Importantly, we find that the order in which these limits are taken is crucial in confined geometries ($\lim_{t\to\infty}\lim_{\rho_0\to 0}\sigma_x^2 \neq \lim_{\rho_0\to 0}\lim_{t\to\infty}\sigma_x^2$). The methodology described above is still applicable but it is crucial to realize that now, between two consecutive visits of the TP, a given vacancy experiences an effective bias due to the motion of the TP resulting from its interaction with the rest of the vacancies. In particular, the first-passage time densities involved in the single vacancy problem mentioned previously are modified. Quite surprisingly, this effective bias, even if arbitrarily small in the $\rho_0 \to 0$ limit, dramatically affects the ultimate long-time behavior of the variance in confined geometries.

More precisely, we show that the superdiffusive regime is always transient for an experimentally relevant system with ρ_0 fixed, while the long-time behavior obeys

$$\lim_{t\to\infty}\frac{\sigma_x^2}{t} \underset{\rho_0\to 0}{\sim} \begin{cases} B & \text{quasi-1D,} \\ 4a_0^2\pi^{-1}\rho_0\ln(\rho_0^{-1}) & \text{2D lattice,} \\ 2a_0^2[A + \coth(f/2)/(2a_0)]\rho_0 & \text{3D lattice,} \end{cases} \tag{3}$$

i.e., is always diffusive. The constant B depends on the driving force f and on the geometry (quasi-1D in this case) of the system. This long-time diffusive behavior has several remarkable features: In dense quasi-1D systems the variance is independent of ρ_0, meaning that the corresponding longitudinal diffusion coefficient $D_\parallel$ is enhanced in comparison to the transverse one $D_\perp$ by a factor $1/\rho_0$, which may attain giant values in systems with $\rho_0 \ll 1$. In 2D this effect is negligible and $D_\parallel$ is only a factor $\ln(\rho_0^{-1})$ larger than $D_\perp$. In unbounded 3D systems no such strong anisotropy between $D_\parallel$ and $D_\perp$ will emerge.

Full dynamics: scaling regime and crossover.—Finally, our approach provides the complete time evolution of the variance in the regime corresponding to $\rho_0 \ll 1$ and at a sufficiently large time t, that interpolates between the two limiting regimes of superdiffusion and giant diffusion listed above. In this regime, it is found that

$$\sigma_x^2 \sim \begin{cases} tg(\rho_0^2 t) & \text{quasi-1D,} \\ -\frac{2a_0^2}{\pi}\rho_0 t\ln((\rho_0 a_0)^2 + 1/t) & \text{2D lattice,} \\ 2a_0^2[A + \coth(f/2)/(2a_0)]\rho_0 t & \text{3D lattice,} \end{cases} \tag{4}$$

where the scaling function g is explicitly calculated and satisfies $g(x) \underset{x\to 0}{\sim} x^{1/2}$ and $g(x) \underset{x\to\infty}{\sim}$ const. Figure 3 reveals excellent quantitative agreement between the analytical predictions and the numerical simulations. Several comments are in order. (i) Equation (4) encompasses both limiting behaviors (1) and (3), and shows explicitly that the limits $t \to \infty$ and $\rho_0 \to 0$ in quasi-1D and 2D systems do not commute; (ii) In such systems the superdiffusion persists up until the onset of the giant diffusive behavior at times $t_\times \sim 1/\rho_0^2$, which can be very large when $\rho_0 \ll 1$. Superdiffusion is therefore very long-lived in such systems. Despite its transient feature, we thus expect superdiffusion to be a robust characteristic of confined crowded

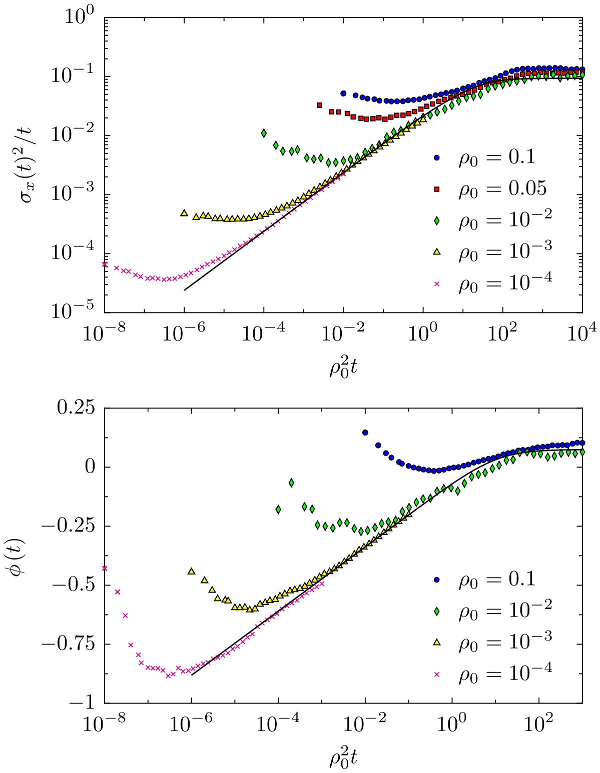

FIG. 3 (color online). Top: rescaled variance as a function of rescaled time $\rho_0^2 t$ on stripelike lattices ($L = 3$) for different densities [solid line, $g(\rho_0^2 t)$]; see the Supplemental Material [20]. Bottom: rescaled variance $\phi(t) = \sigma_x^2(t)/(\rho_0 t) - (2a_0^2/\pi)\ln(1/\rho_0^2 a_0^2)$ as a function of rescaled time $\rho_0^2 t$ on a 2D infinite lattice for different densities [solid line, $h(\rho_0^2 t)$] with $h(x) = (2a_0^2/\pi)\ln[a_0^2 x/(1 + a_0^2 x)] + a_0 \coth(f/2) + 2a_0^2\pi(5 - 2\pi)/(2\pi - 4) + (2a_0^2/\pi)(\ln 8 + \gamma - 1)$.

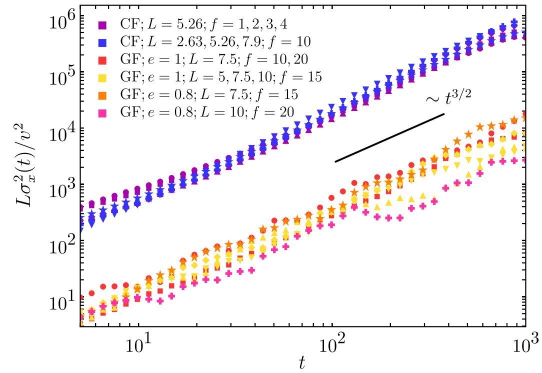

FIG. 4 (color online). Rescaled variance $L\sigma_x^2(t)/v^2$ as a function of time obtained from off-lattice simulations for different widths of stripes L and forces f (it can be shown that $v \sim a_0$ in the superdiffusion regime). CF: molecular dynamics of colloidal fluids in confined striplike geometries. GF: simulations of dense monodisperse granular fluid in confined striplike geometries; e stands for the restitution parameter. More details on off-lattice simulations are given in the Supplemental Material [20].

systems and we anticipate that the ultimate diffusive behavior might be in practice difficult to observe.

The physical mechanism responsible for the emergence of the geometry-induced superdiffusion, revealed by our exact approach, can be ascertained in the large f limit by a mean-field version of the model, which stipulates that after each interaction between the TP and a vacancy, all the other vacancies remain uniformly distributed. The model can then be reformulated as an effective CTRW that takes into account *explicitly* the dynamics of the diffusive vacancies. This is in contrast to the CTRW approach presented in Ref. [10] for glassy systems, which infers the mean and the variance of the waiting-time distribution from the numerical data. In the quasi-1D case, the waiting time of the first jump of the TP is extracted from the distribution $-dS_1/d\tau$, where S_1 is the well-known survival probability of an immobile target in a sea of diffusing predators $S_1(\tau) \propto \exp(-\rho_0\sqrt{\tau})$ [31]. Waiting times of subsequent jumps are then drawn from the distribution $-d[T(\tau)S_1(\tau)]/d\tau$, where $T(\tau)$ is the survival probability of an immobile target chased by a single random walker that starts near the target [31]. Using the waiting time distribution described here, standard calculations show the following. (i) Superdiffusion with an exponent $\lambda = 3/2$ appears as a result of repeated interactions between the TP and a *single* vacancy in quasi-1D systems. This explains, in particular, why no superdiffusion takes place in strictly single-file systems, for which the cumulative displacement of the TP due to interactions with a single vacancy amounts to at most one lattice step. (ii) Diffusive behavior is established ultimately, when other vacancies start to interact with the TP, after a cross-over time which scales as $1/\rho_0^2$. Note that while this mean-field approach reproduces the scaling properties of the variance with respect to the time and the density, it is unable to predict the correct width and driving force dependencies provided by our exact treatment.

Altogether, our results show that the emergence of superdiffusion of a driven TP crucially depends on the system's geometry, an aspect so far disregarded in this context. In order to quantitatively confirm our predictions on further crowded nonglassy systems, we performed off-lattice simulations investigating the dynamics of a biased TP confined to a controlled quasi-1D geometry for models of monodisperse dense liquids (colloidal particles) and monodisperse granular fluids (using an algorithm similar to the one presented in Ref. [32]). In Fig. 4 we plot the properly rescaled variance, where a clear data collapse is visible. This validates the time, width, and driving force dependences that feature in our analytical expression (1) also for off-lattice systems. Finally, our analysis shows that superdiffusion is not the hallmark of glassy systems but is a generic feature of driven dynamics in confined crowded systems.

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