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PAPER

How active forces influence nonequilibrium glass transitions

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Abstract

Dense assemblies of self-propelled particles undergo a nonequilibrium form of glassy dynamics. Physical intuition suggests that increasing departure from equilibrium due to active forces fluidifies a glassy system. We falsify this belief by devising a model of self-propelled particles where increasing departure from equilibrium can both enhance or depress glassy dynamics, depending on the chosen state point. We analyze a number of static and dynamic observables and suggest that the location of the nonequilibrium glass transition is primarily controlled by the evolution of two-point static density correlations due to active forces. The dependence of the density correlations on the active forces varies non-trivially with the details of the system, and is difficult to predict theoretically. Our results emphasize the need to develop an accurate liquid state theory for nonequilibrium systems.

1. Introduction

Nonequilibrium glass transitions have recently emerged as a new type of dynamic arrest occurring in particle systems driven out of equilibrium by active forces [1, 2]. The initial theoretical interpretation, based on the analysis of simple glass models driven by active forces [3], has been confirmed in several computer simulations of more realistic active matter models [1, 4–14, 49]. A number of alternative theoretical approaches have now been proposed to describe this phenomenon [15–17]. Just as in equilibrium [18, 19], nonequilibrium glass transitions bear no connection to the jamming transition [20], which corresponds instead to a geometric transition taking place in the absence of any driving mechanism.

Although the system is driven far from thermal equilibrium, the corresponding slow dynamics exhibits all the characteristic signatures of supercooled liquids approaching an equilibrium glass transition [21], such as caging, dynamical slowing down, non-exponential time correlation functions and dynamic heterogeneity [22]. A unique feature that distinguishes active from equilibrium glasses is the emergence of collective effective temperatures [3, 23, 24]. Nonequilibrium glass transitions represent an experimentally relevant concept, because they may explain various dynamic phenomena observed experimentally in both dense active colloidal suspensions [25, 26], active granular materials [27], and in biological systems [2, 28–31].

The above summary demonstrates that the existence of nonequilibrium glassy dynamics is well-established. For a number of model systems, phase diagrams, microstructure, dynamic timescales and length scales have been thoroughly analyzed. However, not much is known quantitatively about how active forces influence the glass transition. In a very trivial sense, adding active forces to an equilibrium material must suppress the glass transition, as the amount of driving energy then increases [1, 10]. This is equivalent to increasing the temperature at equilibrium. However, the outcome of departing from thermal equilibrium *at constant driving energy* is much less trivial, and is in fact not understood. There exist conflicting results in the literature, suggesting that glassy dynamics is either suppressed (as in hard sphere systems [4, 5]), or enhanced (as in Lennard-Jones particles [9, 11]) when going out of equilibrium. It is unclear whether these seemingly distinct behaviors are due to a change in the pair interaction, to the details of the active forces or microscopic dynamics, or to a genuine physical effect. If real, then, these results beg the question as to what physical quantity is the main indicator to reveal how active forces modify the location of the glass transition.

In this work, we devise a simple molecular dynamics model for self-propelled particles where acceleration or slowing down of the dynamics can both be observed by changing active forces at constant density of the system. This directly shows that it is in fact very difficult to predict whether active forces will fluidify or glassify a given material, as they can do both. Having a model displaying both types of response to active forces allows us to directly investigate how the departure from equilibrium influences the glassy dynamics, and which microscopic quantity is responsible for their evolution. Our numerical analysis reveals that active forces have a strong impact on the microstructure of the fluid, which can be readily quantified by two-point static density correlations functions. The nontrivial evolution of the static structure then primarily accounts for the evolution of the glass transition, underlying the need to develop a more accurate liquid state theory for active fluids.

The paper is organized as follows. In section 2 we define our model system of purely repulsive, non-aligning, self-propelled particles and provide the details of the numerical simulations. The main features of the glassy dynamics and the evolution of the apparent glass transition line upon increasing departure from equilibrium are reported in section 3. Next, in sections 4 and 5 we analyze two sets of equal-time steady-state correlations, velocity correlations and two-point density correlations, respectively. We conclude in section 6 with a discussion of the correlations between the evolution of the glassy dynamics and of the steady-state structure upon increasing departure from equilibrium.

2. Interpolating between hard and soft active particles

Earlier studies of glassy dynamics in model active systems used either a hard-sphere interaction [4, 5] or a Lennard-Jones interaction [9–11]. Both families of studies reported opposite results regarding the influence of active forces on the glass transition.

To continuously interpolate between these limiting cases, we use the Weeks–Chandler–Andersen truncation of the Lennard-Jones potential [32], a strategy used before in equilibrium studies [33]. This choice creates a purely repulsive system that allows us to continuously move from simulating a hard sphere-like system at very low temperatures and moderate densities (when the typical nearest-neighbor distance is slightly larger than the range of the potential), to simulating a Lennard-Jones-like system at moderate temperatures and large densities (when the typical nearest-neighbor distance is smaller than the range of the potential). The comparison with earlier works suggest that our model should display both an acceleration or a slowing down of the glassy dynamics, depending on the density regime, allowing us to revisit and unify previous studies.

To model an active liquid, we use the so-called active Ornstein–Uhlenbeck particles model [34] introduced independently in [35, 36]. In this model, the dynamics is overdamped and the particles move under the combined influence of the interparticle interactions and the self-propulsion. The self-propulsion is modeled as an internal driving force evolving according to the Ornstein–Uhlenbeck process. Thus, the equations of motion are given by

$$\dot{r}_i = \xi_0^{-1} [F_i + f_i], \quad (1)$$

$$\tau_p \dot{f}_i = -f_i + \boldsymbol{\eta}_i. \quad (2)$$

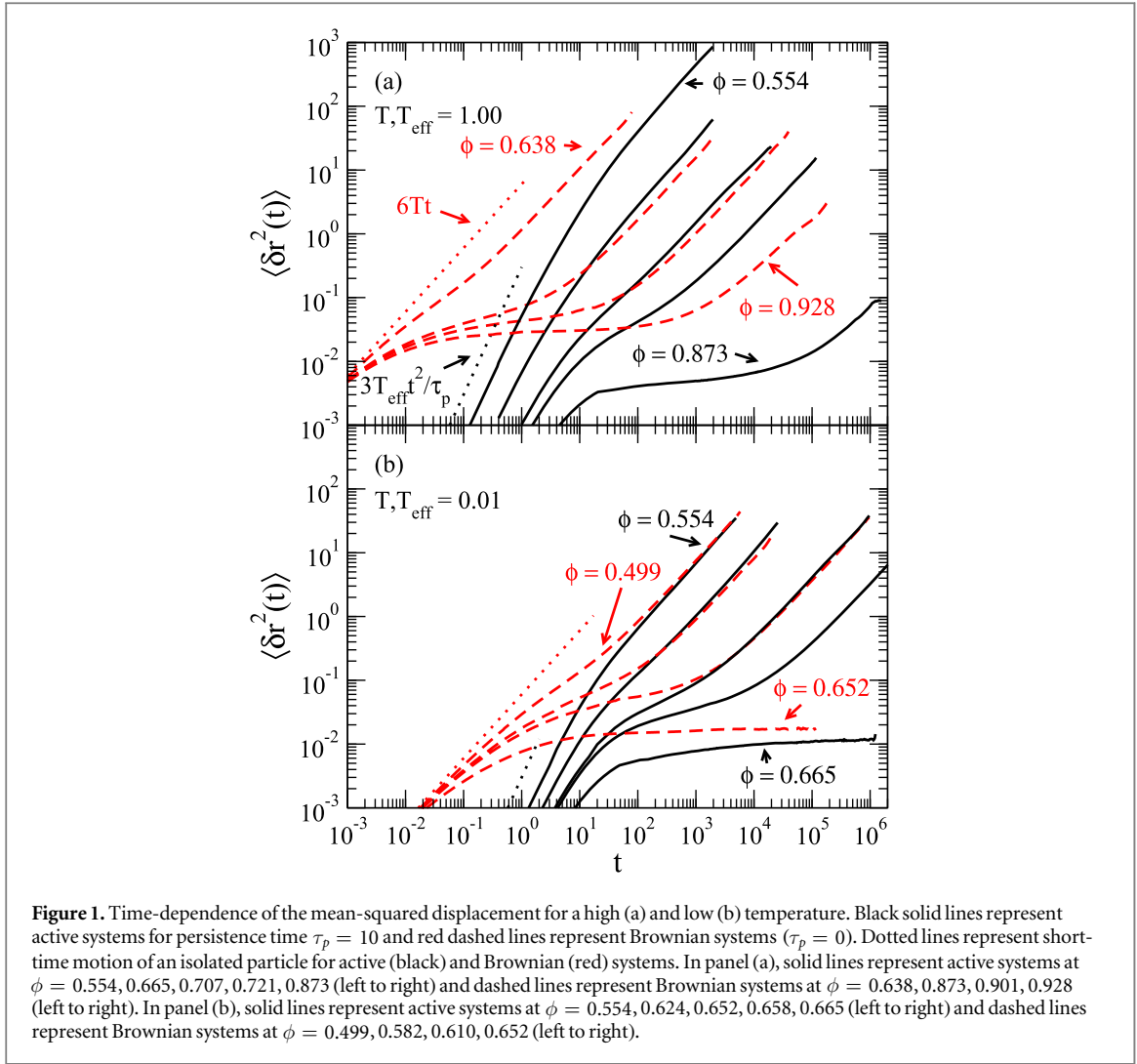
In equation (1), r_i is the position of particle i , ξ_0 is the friction coefficient of an isolated particle, F_i is the force acting on particle i originating from the interactions, and f_i is the self-propulsion force acting on particle i . In equation (2), τ_p is the persistence time of the self-propulsion and $\boldsymbol{\eta}_i$ is an internal Gaussian noise with zero mean and variance $\langle \boldsymbol{\eta}_i(t) \boldsymbol{\eta}_j(t') \rangle_{\text{noise}} = 2\xi_0 T_{\text{eff}} I \delta_{ij} \delta(t - t')$, where $\langle \dots \rangle_{\text{noise}}$ denotes averaging over the noise distribution, T_{eff} is the single-particle effective temperature, and I is the unit tensor. In the following, we set the friction coefficient to unity, $\xi_0 = 1$. Notice that f_i is the unique driving force in equation (1), which does not contain an additional Brownian noise term.

The name ‘single-particle effective temperature’ for T_{eff} originates from the fact that an isolated particle moving under the influence of the self-propulsion evolving according to equation (2) performs a persistent random walk with the long-time diffusion coefficient equal to $D_0 = T_{\text{eff}}$ (we use a system of units such that the Boltzmann constant k_B is unity). We note that for a system of interacting self-propelled particles other effective temperatures can be defined based on different fluctuation-dissipation ratios [3, 23, 37]. These effective temperatures are, in general, different from the single-particle effective temperature. Since we will not be concerned with these collective effective temperatures in this work, in the following, for brevity, we will refer to T_{eff} as the effective temperature. Importantly, T_{eff} controls the amount of energy injected into the system, and it represents the analog of the thermal bath for an equilibrium system.

The interparticle forces originate from a potential, $F_i = -\sum_{j \neq i} \nabla_i V_{\alpha\beta}(r_{ij})$ where

$$V_{\alpha\beta}(r) = 4\epsilon \left[\left(\frac{\sigma_{\alpha\beta}}{r} \right)^{12} - \left(\frac{\sigma_{\alpha\beta}}{r} \right)^6 \right], \quad (3)$$

for $r \leq \zeta_{\alpha\beta} = 2^{1/6} \sigma_{\alpha\beta}$ and zero otherwise. In equation (3), α, β denote the particle species A or B , $\epsilon = 1$ (which sets the unit of energy), $\sigma_{AA} = 1.4$, $\sigma_{AB} = 1.2$, and $\sigma_{BB} = 1.0$ (which sets the unit of length). Our unit of time is



$\sigma_{AA} \xi_0 / \epsilon_{AA}$. We simulated $N = 1000$ particles composing a 50:50 mixture in a volume V using periodic boundary conditions in three spatial dimensions. The truncation of the potential is at the potential minimum, and thus the interparticle force is purely repulsive. The repulsive character of the force combined with the finite range of the potential implies that in the low temperature limit the system becomes equivalent to a hard sphere system consisting of a binary mixture of spheres of diameters ζ_{AA} and ζ_{BB} .

As our control parameters, we use the volume fraction $\phi = \pi N [\zeta_{AA}^3 + \zeta_{BB}^3] / (12V)$, the effective temperature T_{eff} , and the persistence time of the self-propulsion τ_p . Since there is no thermal noise, when $\tau_p \rightarrow 0$ this model system becomes equivalent to a Brownian system at a temperature $T = T_{\text{eff}}$. Therefore, τ_p quantifies the increasing departure from equilibrium as τ_p increases from zero. In this work, we investigate the dependence of the glassy dynamics on the persistence time and we also compare the results obtained for active systems with those obtained from overdamped Brownian dynamics simulations at a temperature T . We vary the persistence time between $\tau_p = 0$ and $\tau_p = 10$ and the effective temperature between $T_{\text{eff}} = 0.01$ and $T_{\text{eff}} = 1.0$.

3. Glassy dynamics and phase diagram

In figure 1 we show the development of glassy dynamics upon increasing the volume fraction, at two values of the effective temperature, the highest and the lowest temperature investigated, $T_{\text{eff}} = 1.0$ and $T_{\text{eff}} = 0.01$, respectively. We illustrate the changes in the dynamics by showing the mean-squared displacement

$$\langle \delta r^2(t) \rangle = \frac{1}{N_A} \left\langle \sum_{i=1}^{N_A} [r_i(t) - r_i(0)]^2 \right\rangle, \quad (4)$$

where the summation is over the particles of type A and N_A is the number of these particles. Here and in the following, we restrict our discussion to the larger particles, particles A, and we note that analyzing the dynamics

of the B particles leave the conclusions unchanged. Furthermore, to simplify notation we do not use conventional subscripts when referring to quantities pertaining to particles A only. Thus, we use $\langle \delta r^2(t) \rangle$ rather than $\langle \delta r_A^2(t) \rangle$ and later in section 5 we will use, for instance, $g(r)$ rather than $g_{AA}(r)$.

For an isolated active particle or, alternatively, in a non-interacting system, the mean-squared displacement can be calculated analytically,

$$\langle \delta r^2(t) \rangle = 6T_{\text{eff}} [\tau_p (e^{-t/\tau_p} - 1) + t]. \quad (5)$$

At short times, the particle motion is ballistic and $\langle \delta r^2(t) \rangle \simeq 3(T_{\text{eff}}/\tau_p)t^2$. The long-time motion is diffusive and $\langle \delta r^2(t) \rangle \simeq 6T_{\text{eff}}t$. Comparing the long-time result with that for an isolated Brownian particle, $\langle \delta r^2(t) \rangle = 6Dt$, we see that the long time diffusive motion of an isolated active particle at an effective temperature T_{eff} matches that of the isolated Brownian particle at a temperature $T = T_{\text{eff}}$.

As shown in figure 1, the ballistic and diffusive regimes are still observed in mean-squared displacements in systems of interacting active particles (solid lines, $\tau_p = 10$). However, for active particles both the short-time dynamics and the long-time dynamics change with the volume fraction. The change in the short-time dynamics is induced by correlations between active particles velocities and positions, discussed further in section 4. These correlations are an important feature of active systems [9]. At constant effective temperature, their magnitude decreases with decreasing persistence time and the correlations vanish in the Brownian limit. Generally, for active systems both the short-time ballistic motion and the long-time diffusive motion slow down with increasing volume fraction. The slowing down of the short-time dynamics is more pronounced at higher effective temperatures (note, however, that at higher effective temperatures the volume fractions are also somewhat larger). Generally, with increasing volume fraction, at intermediate times a plateau begins to develop and glassy dynamics emerge.

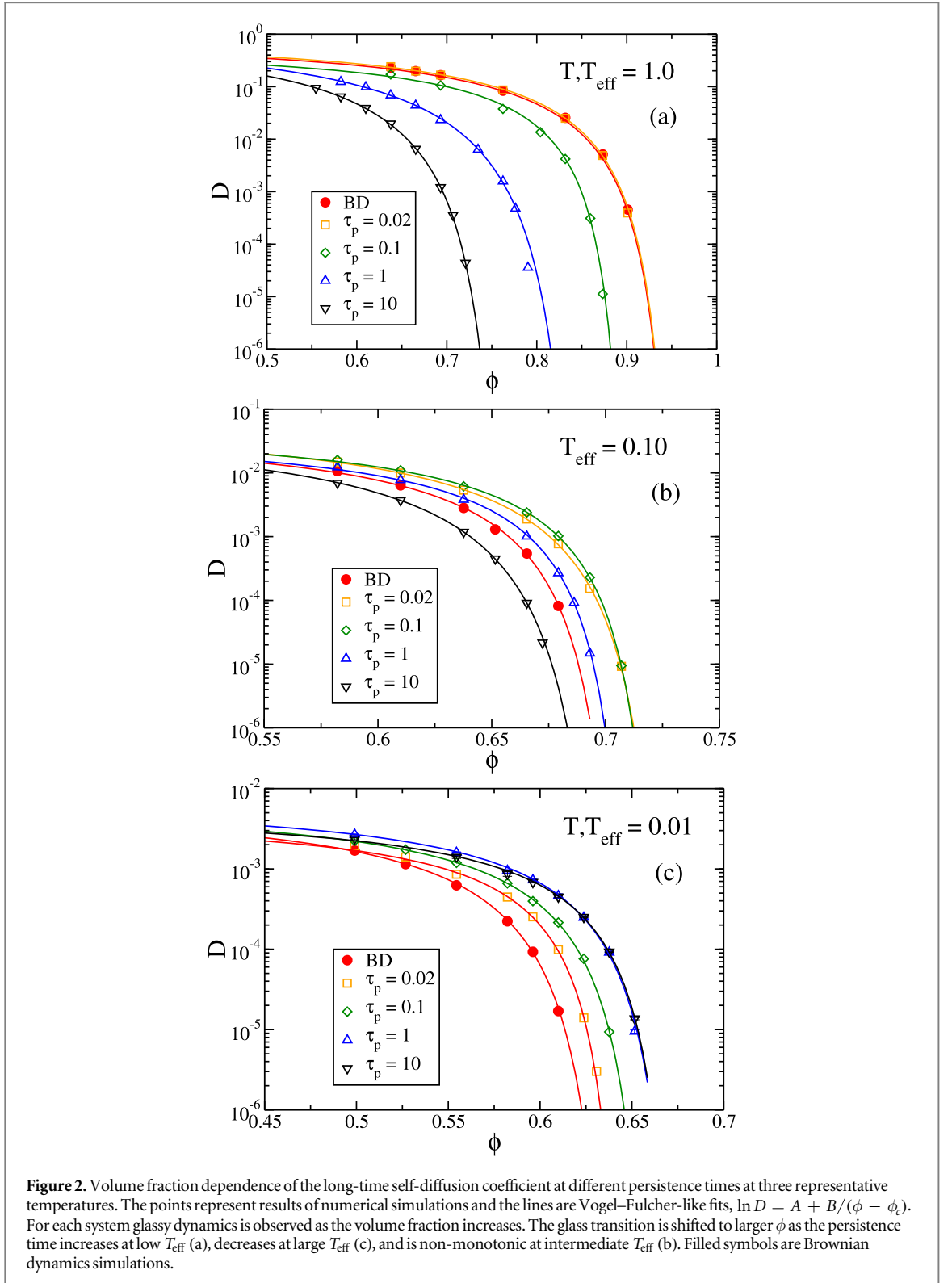
Figure 1 also shows that for Brownian systems we have two diffusive regimes, for short times $\langle \delta r^2(t) \rangle = 6Tt$ (dotted red line) and for long times $\langle \delta r^2(t) \rangle = 6Dt$, where D is the long-time self-diffusion coefficient. At intermediate times a plateau develops between the short time diffusive motion and the long time diffusive motion. The presence of the plateau indicates caging of individual particles.

While emerging glassy dynamics in active systems is generally similar to that in Brownian systems, we note some important quantitative differences. First, in active systems there is a significant slowing down of the short-time ballistic motion whereas in Brownian systems the short-time diffusive motion is independent of the volume fraction (and it depends only trivially on the temperature). Second, plateau heights in active systems are different from those in Brownian systems. This is especially prominent at the higher temperature where the plateau height for the densest $\tau_p = 10$ active system is around an order of magnitude smaller than for the densest Brownian system (note that the density of the active system is quite a bit lower than that of the Brownian system). For $T_{\text{eff}} = 0.01$, a well-defined plateau is not observed and instead the mean-squared displacement exhibit a very slow subdiffusive behavior. These latter facts suggest that upon increasing departure from equilibrium the effective interparticle interaction changes significantly. We comment on this point further in section 5.

From the long-time limit of the mean-squared displacements we extract the long-time self-diffusion coefficients, $D = \lim_{t \rightarrow \infty} \langle \delta r^2(t) \rangle / (6t)$. In figure 2, we show the dependence of the diffusion coefficients on the volume fraction for a number of active systems characterized by a given value of the persistence time and the effective temperature, (τ_p, T_{eff}) , and for Brownian systems characterized by the temperature T . For the highest temperature, $T_{\text{eff}} = 1.0$, in figure 2(a) we see that the diffusion coefficient decreases with increasing persistence time at a fixed volume fraction. In contrast, at an intermediate temperature, $T_{\text{eff}} = 0.1$, we find a non-monotonic dependence of the diffusion coefficient on the persistence time at a fixed volume fraction. Finally, at the lowest temperature, $T_{\text{eff}} = 0.01$, we find that the diffusion coefficient increases with increasing persistence time at a fixed volume fraction for the range of persistence times investigated. We observe that increasing departure from equilibrium can either promote or suppress the glassy dynamics without changing the pair interaction between the self-propelled particles, as announced in the introduction.

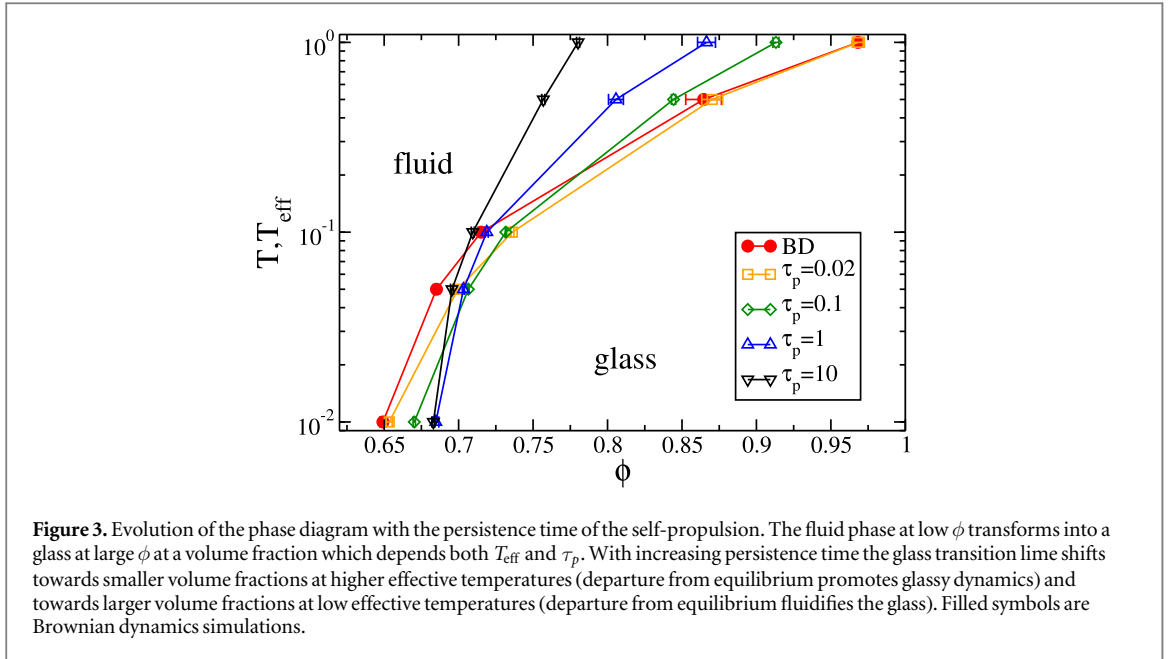
These results suggest that there is a change in the persistence time dependence of the apparent glass transition line in the temperature–volume fraction plane. To determine quantitatively the glass transition line, we fit the diffusion coefficient data to a Vogel–Fulcher-like dependence on the volume fraction, $\ln D = A + B/(\phi - \phi_c)$, where A, B and ϕ_c are fitting parameters. These empirical fits are shown as continuous lines in figure 2. The Vogel–Fulcher-like formula results in reasonable fits to the data and, therefore, reasonable estimates for the glass transition volume fraction $\phi_c = \phi_c(\tau_p, T_{\text{eff}})$. Other fitting functions would provide qualitatively similar results for the evolution of the glass transition lines.

In figure 3 we present the resulting glass–fluid phase diagram in the temperature–volume fraction plane, for different persistence times. As can be inferred from the dependence of the diffusion coefficient on the volume fraction, the glass transition volume fraction ϕ_c monotonically decreases with increasing τ_p for $T_{\text{eff}} = 1.0$, and it



monotonically increases with increasing τ_p for $T_{\text{eff}} = 0.01$. At intermediate T_{eff} there is a non-monotonic change of ϕ_c upon increasing the persistence time, which signals the crossover between the high and low temperature regimes. These findings are consistent with both the previously observed increase of the glass transition temperature for the Lennard-Jones system [11] and the increase of the glass transition packing fraction for the hard-sphere system [4, 5].

Figure 3 shows for the first time that for the same model active system increasing departure from equilibrium can both glassify an active fluid and fluidifies an active glass, depending on the studied thermodynamic state point. This directly demonstrates that the physical intuition that activity fluidifies the glass is incorrect, as activity can also solidify the supercooled fluid. In the following two sections we search for correlations between the



observed non-trivial dependence of the dynamics on the persistence time and the dependence of static (equal-time) correlations on the persistence time. In section 4 we examine the evolution of the correlations between active particles' velocities on the persistence time. In section 5 we investigate the dependence of density fluctuations on the persistence time.

4. Nonequilibrium velocity correlations

While developing a theory for the dynamics of systems of active Ornstein–Uhlenbeck particles [9, 15] we discovered that correlations between velocities of self-propelled particles play an important role in the dynamics of active systems. Subsequent simulational studies [9, 11] showed that in the active system with Lennard-Jones interactions the velocity correlations grow upon increasing departure from equilibrium. The correlations between velocities of different active particles vanish in the limit of zero persistence time and are zero in Brownian systems. Therefore, these nonequilibrium velocity correlations represent the most natural candidate to explain how the glass transition departs from its equilibrium counterpart as the persistence time increases.

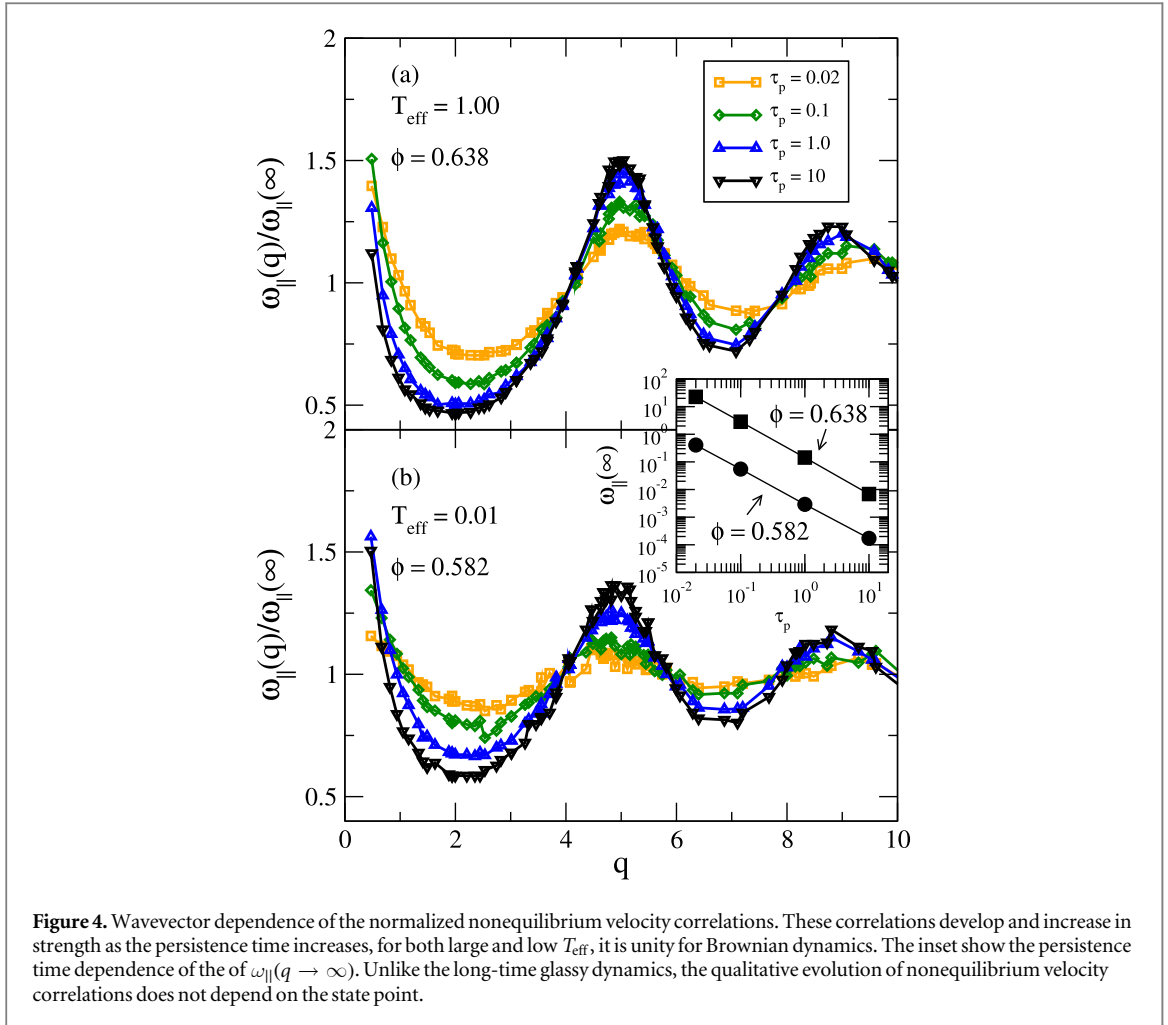
In [15] a mode-coupling-like theory is derived for active particle systems. As with the mode-coupling theory of the glass transition for passive particle systems, the structure of system is the input to equations that describe the decay of density fluctuations. For passive particle systems the only needed input is the static structure factors, but for active particle systems the input is not only the static factors but also a wavevector-dependent function that characterizes static velocity correlations, which only exists for nonequilibrium self-propelled particles.

In [9, 15], we showed that this function is important for the dynamics of an active system. For a binary mixture, in analogy with the partial static structure factors that characterize the number density fluctuations, in principle we need to introduce three different functions corresponding to the correlations of velocities of the A particles, velocities of the B particles, and the AB cross-correlations. As stated earlier, we restrict the discussion to the larger particles and only examine the correlation between velocities of the A particles, which is defined as follows:

$$\omega_{\parallel}(q) = \hat{q} \cdot \left\langle \sum_{i,j=1}^{N_A} (f_i + F_i)(f_j + F_j) e^{-iq(r_i - r_j)} \right\rangle \cdot \hat{q}, \quad (6)$$

where both summations are over the particles of type A , $\hat{q} = \mathbf{q}/|\mathbf{q}|$ and $\xi_0^{-1}(f_i + F_i)$ is the instantaneous velocity of particle i .

The large wavevector limit of this function, $\omega_{\parallel}(\infty) = \lim_{q \rightarrow \infty} \omega_{\parallel}(q)$, exactly determines the short-time behavior of the mean-squared displacement in the system of interacting active particles, $\langle \delta r^2(t) \rangle \simeq 3\omega_{\parallel}(\infty)t^2$. According to the approximate mode-coupling-like theory sketched in [9] and then detailed in [15], the complete function $\omega_{\parallel}(q)$ enters into the expressions for the so-called vertices of the irreducible memory function, and thus, together with the static structure factor, it determines the long-time dynamics. If the function $\omega_{\parallel}(q)$

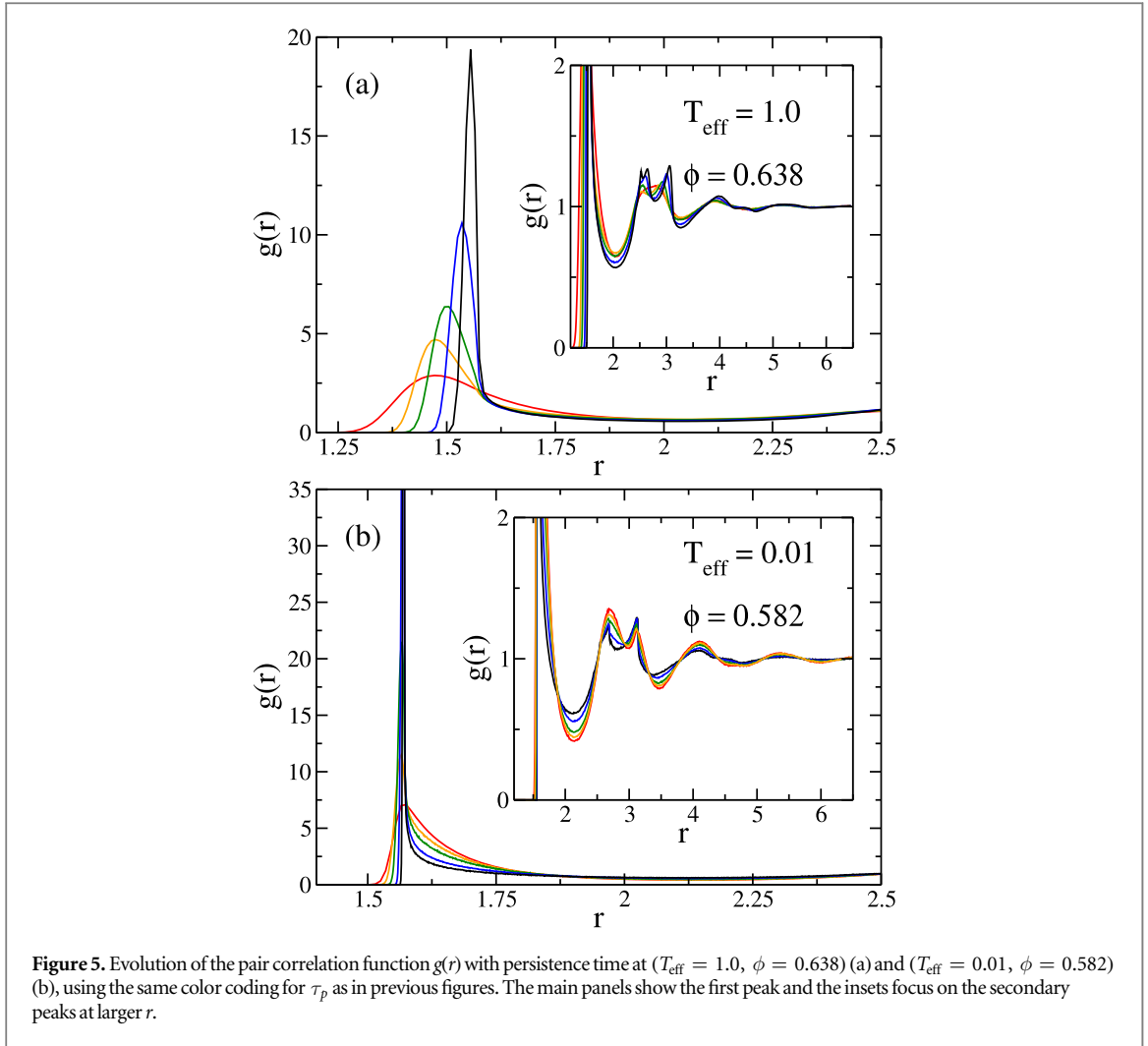


evolved differently upon increasing the persistence time at high and low effective temperatures (i.e. for Lennard-Jones-like and hard-sphere-like systems), it would suggest that the velocity correlations are responsible for the non-trivial changes in the phase diagram shown in figure 3.

Previous work [11] showed that upon increasing both the persistence time and the importance of the interparticle interactions, the overall scale of the velocity correlations decreases significantly while their wavevector dependence becomes more pronounced. The overall scale of the velocity correlations can be characterized by their large wavevector limit, $\omega_{\parallel}(\infty)$. We recall that figure 1 indicates that the short-time dynamics of the self-propelled particles slows down upon increasing the volume fraction at fixed persistence time, for both high and low effective temperatures. This slowing down of the short-time dynamics is the direct consequence of the decreasing of $\omega_{\parallel}(\infty)$ upon increasing the volume fraction at constant persistence time and effective temperature, and thus increasing the importance of the interparticle interactions.

The focus of this work is the dependence of the (long-time) glassy dynamics on the departure from equilibrium. To this end, in the inset to figure 4 we show the evolution of $\omega_{\parallel}(\infty)$ with the persistence time for $T_{\text{eff}} = 1.0$ and 0.01. In both cases we observe approximate power-law dependence of $\omega_{\parallel}(\infty)$ on τ_p , $\omega_{\parallel}(\infty) \propto \tau_p^{-1.25}$. Thus, while $\omega_{\parallel}(\infty)$ at $T_{\text{eff}} = 1.0$ is about 50 times larger than that at $T_{\text{eff}} = 0.01$, its dependence on the persistence time is the same at both temperatures, and, therefore, $\omega_{\parallel}(\infty)$ seems unconnected to the more complicated evolution of the glassy dynamics reported in figures 2(a)–(c).

In the main panels of figure 4 we show the evolution of the wavevector dependence of $\omega_{\parallel}(q)$ upon increasing the persistence time for $T_{\text{eff}} = 1.0$ and 0.01. To this end we plot $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$ for different values of τ_p . Upon increasing the persistence time we observe that the oscillations of $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$ are becoming more pronounced, which indicates growing local velocity correlations. However, this increase occurs at every effective temperature investigated, independently of the evolution of the long-time glassy dynamics. Thus, growing local velocity correlations and glassy dynamics appear to be largely uncorrelated. In other words, velocity correlations accompany the nonequilibrium glass transition, they presumably quantitatively affect its location, but they do not seem to be the main factor responsible for the nontrivial evolution of the phase diagram shown in figure 3.



5. Two-point density correlations

Since the dependence of the velocity correlations on the persistence time is uncorrelated to the evolution of the glassy dynamics, we turn our attention to other, more conventional static correlations, namely density correlations. In this section we investigate equal-time correlations of the density fluctuations by examining the pair correlation function in real and Fourier space. We show that the dependence of pair correlations upon increasing the departure from equilibrium strongly correlates with the evolution of the glassy dynamics reported in figures 2(a)–(c), and we provide a physical interpretation of the results.

5.1. Pair correlation function $g(r)$

In figure 5 we show the evolution of pair density correlations in the direct space upon increasing the persistence time. Specifically, we show the pair correlation function characterizing correlations between the A particles,

$$g(r) = \frac{1}{N_A} \sum_{i=1}^{N_A} \sum_{j \neq i}^{N_A} \langle \delta(r - |r_i - r_j|) \rangle, \quad (7)$$

where both summations are over the A particles.

For both high and low effective temperatures we observe a very strong sharpening and growth of the first peak of the pair correlation function. This may be interpreted as an increasing ‘adhesion’ between self-propelled particles upon increasing the persistence time. The height of the first peak of $g(r)$ is 4.5 to 7 times larger for $\tau_p = 10$ than for the Brownian limit at the same packing fraction. While the quantitative details of the evolution of the first peak change with the effective temperature, qualitatively the evolution is very similar. Thus, the sharpening and growth of the first peak of the pair correlation function is a general feature of self-propelled particles which does not seem to be correlated with the non-trivial evolution of the glass transition line.

On the other hand, the position of the first peak has a very different behavior in the two panels shown in figure 5. It shifts to larger distances for $T_{\text{eff}} = 1.0$ but remains essentially at the same position for $T_{\text{eff}} = 0.01$. As a

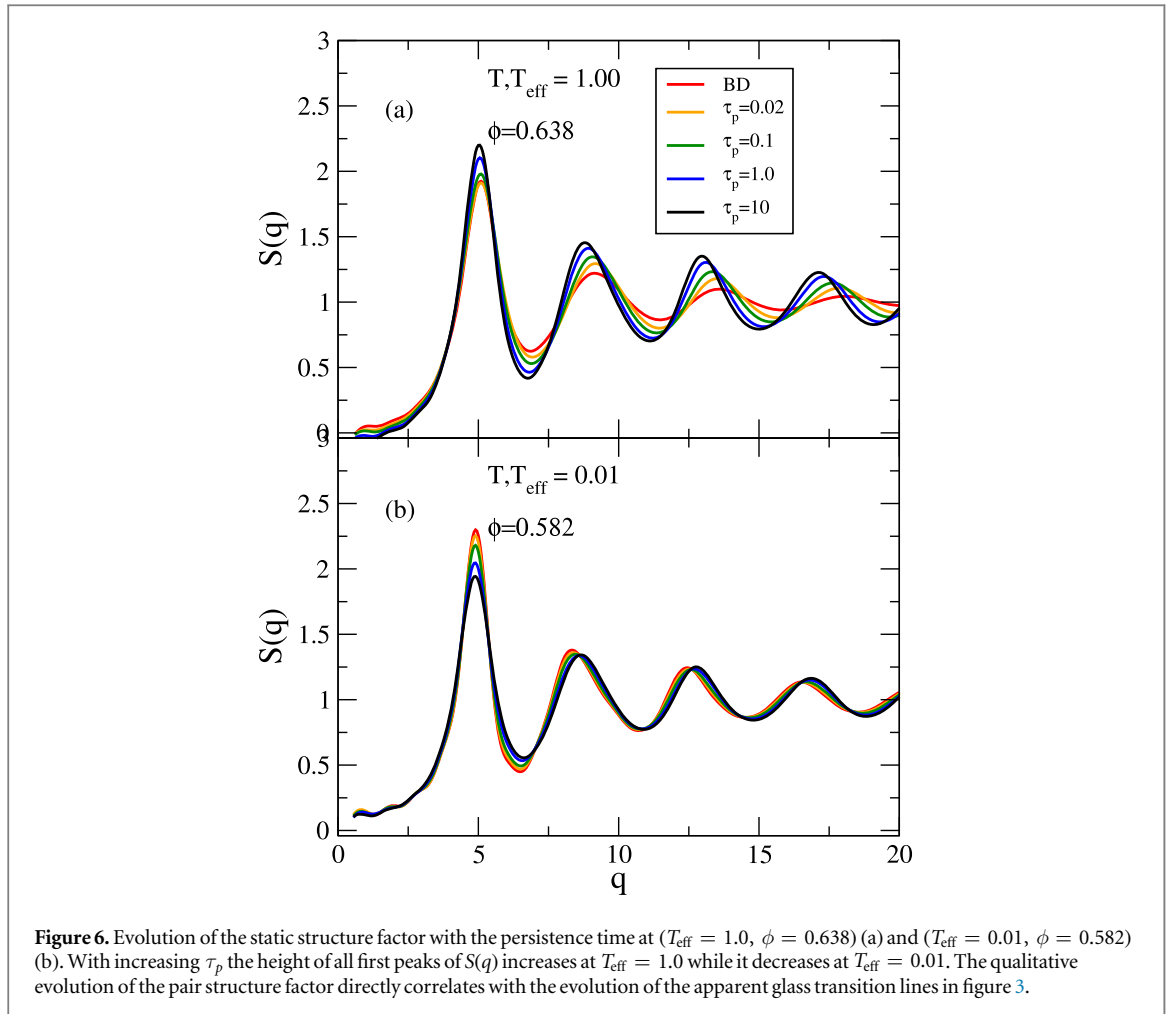


Figure 6. Evolution of the static structure factor with the persistence time at ($T_{\text{eff}} = 1.0$, $\phi = 0.638$) (a) and ($T_{\text{eff}} = 0.01$, $\phi = 0.582$) (b). With increasing τ_p the height of all first peaks of $S(q)$ increases at $T_{\text{eff}} = 1.0$ while it decreases at $T_{\text{eff}} = 0.01$. The qualitative evolution of the pair structure factor directly correlates with the evolution of the apparent glass transition lines in figure 3.

result, the effective diameter of the particle increases with τ_p for $T_{\text{eff}} = 1.0$ but is approximately constant for $T_{\text{eff}} = 0.01$. This effective inflation of the particles for the higher temperature may be held responsible for the slowing down of the dynamics as τ_p increases, as the system becomes effectively more crowded. Strikingly, this effect is absent for the lower temperature.

In the insets in figure 5, we show that the secondary peaks of the pair correlation function change in a qualitatively different way upon increasing departure from equilibrium. Specifically, the amplitude of the secondary peaks increases with increasing τ_p for $T_{\text{eff}} = 1.0$ (indicating enhanced local structure) while it decreases with increasing τ_p for $T_{\text{eff}} = 0.01$ (indicating decreasing local structure). This qualitatively different change does correlate with the evolution of the glass transition lines, where enhanced (suppressed) structure seems to promote (suppress) the glassy dynamics. As shown in the next subsection, the rather subtle changes of the secondary peaks in the direct space translates into more visible changes of the static structure factor in the Fourier domain.

5.2. Static structure factor $S(q)$

The contrast between the evolution of the density correlations with increasing departure from equilibrium at high and low temperatures is easier to observe in the Fourier domain. We show in figure 6 the partial static structure factor for the A particles,

$$S(q) = \frac{1}{N_A} \left\langle \sum_{i=1}^{N_A} \sum_{j=1}^{N_A} e^{-iq \cdot (r_i - r_j)} \right\rangle, \quad (8)$$

where both summations are over the A particles.

We observe in figure 6 that the amplitude of all the peaks of the static structure factor increases with increasing the persistence time for the higher effective temperature, $T_{\text{eff}} = 1.0$. In contrast, the amplitude of the peaks decreases with increasing the persistence time for the lower effective temperature, $T_{\text{eff}} = 0.01$. Therefore, the evolution of the peak height of the structure factor correlates directly with the evolution of the fitted glass transition volume fraction, ϕ_c . We also find that the oscillations of the structure factor decay slower with

increasing τ_p , which directly reflects the sharpening and growth of the first peak of the pair correlation function discussed in section 5.1 above.

The correlation identified between glassy dynamics and structure factor implies that to understand the gross features of nonequilibrium glass transitions in active systems, one must first understand how increasing departure from equilibrium affects the static structure of the active fluid at the level of two-point quantities. This first step is crucial, as some theories for the nonequilibrium glass transitions use the nonequilibrium pair correlation $S(q)$ as input for a dynamical theory of the driven glassy dynamics [9, 15]. This initial step is usually not emphasized in equilibrium studies [21], as there exist very accurate theories predicting the equilibrium $S(q)$ from the sole knowledge of the pair interaction [38].

5.3. Physical interpretation: potential of mean force $u(r)$

There have been a few attempts to develop a theory for the static structure of active fluids. One way to develop such a theory is to start with an approximate mapping of the nonequilibrium active fluid onto an effective equilibrium fluid [36, 39–41]. Having an effective equilibrium model one could then use the well established framework of the equilibrium liquid state theory to calculate the pair correlation function and the static structure factor. Farage *et al* [39] showed that such a procedure results in very accurate predictions for the pair correlation function, at least far from the glassy regime. However, Rein and Speck [42] found that the structure of the simulated nonequilibrium active fluid can be quite different from that of the simulated effective equilibrium fluid. Obviously, more work is needed in this area [43]. In particular, we note that no comparison between functions characterizing density correlations obtained from simulations and the same functions calculated using liquid state theory was performed in the glassy regime that is of interest for the present work.

A different way to evaluate the static structure of an active fluid is to use the very recently proposed ‘integration through transients’ approach [17] which was originally developed to describe sheared colloidal suspensions [44]. So far, this approach was only used to analyze transient glassy dynamics (i.e. the dynamics after active forces have been turned on) but in principle it could be used to calculate equal-time steady-state properties.

In fact, to get some insight into the evolution of the density correlations upon increasing the persistence time it would be nice to have an effective, persistence time-dependent potential. In other words, we would like to investigate a potential that, if used in an equilibrium simulation, would result in the nonequilibrium pair correlation functions shown in figure 5. Finding an effective potential resulting in a given pair correlation function is a separate project. Moreover, it is not even obvious that such a potential could always be found for our nonequilibrium pair correlation functions.

Instead, here we look at the evolution of the generalized potential of mean force,

$$u(r) = -T_{\text{eff}} \ln[g(r)], \quad (9)$$

on the persistence time to obtain some physical intuition about the effective interaction experienced by the particles. Recall that $u(r)$ only represents a gross approximation to the real pair interaction between the particles, even at thermal equilibrium. We present numerical results for $u(r)$ in figure 7. At both high and low temperatures, the evolution of $u(r)$ signals the emergence of short-range adhesive forces, manifested through the development of a narrow negative well near the interparticle distance.

However, at $T_{\text{eff}} = 1.0$ the potential of mean force reveals a striking feature, that is not observed at $T_{\text{eff}} = 0.01$. We observe that the repulsive interaction becomes much steeper with increasing the persistence time, and the spatial range of the repulsion extends to larger distances. The particles, therefore, effectively become stiffer and, perhaps more dramatically, they also appear larger with increasing persistence time. It is reasonable to assume that this apparent ‘expansion’ of the particles directly accounts for the observed shift of the glass transition towards smaller volume fractions with increasing τ_p for $T_{\text{eff}} = 1.0$. In other words, glassy dynamics is promoted in this regime because self-propulsion makes the active suspension effectively more crowded than its equilibrium counterpart. The evolution of the local structure for $T = 1.0$ also explains the drastic decrease in the height of the intermediate-time plateau in the mean-squared displacements shown in figure 1(a).

For $T_{\text{eff}} = 0.01$, an apparent expansion of the particles is barely visible. This is physically reasonable, as the equilibrium system is already very close to the hard sphere interaction, and it is difficult to make this hard-core interaction steeper. In this regime, the main effect of increasing the persistence time is the emergence of a short-range effective adhesion between the self-propelled particles. The emergence of adhesion through self-propulsion was noted before in the fluid regime [25, 45], and we find here that it is quite pronounced for dense assemblies of self-propelled hard spheres as well, as surmised in [5]. In that case, it is not surprising that the glass transition is shifted to larger ϕ , as this behavior is well-known from equilibrium studies of adhesive spheres [46]. Therefore, the physical reason that self-propelled hard spheres undergo a delayed glass transition is not that they are ‘driven’ out of equilibrium by active forces, it is much more subtle. Instead, active forces induce an effective

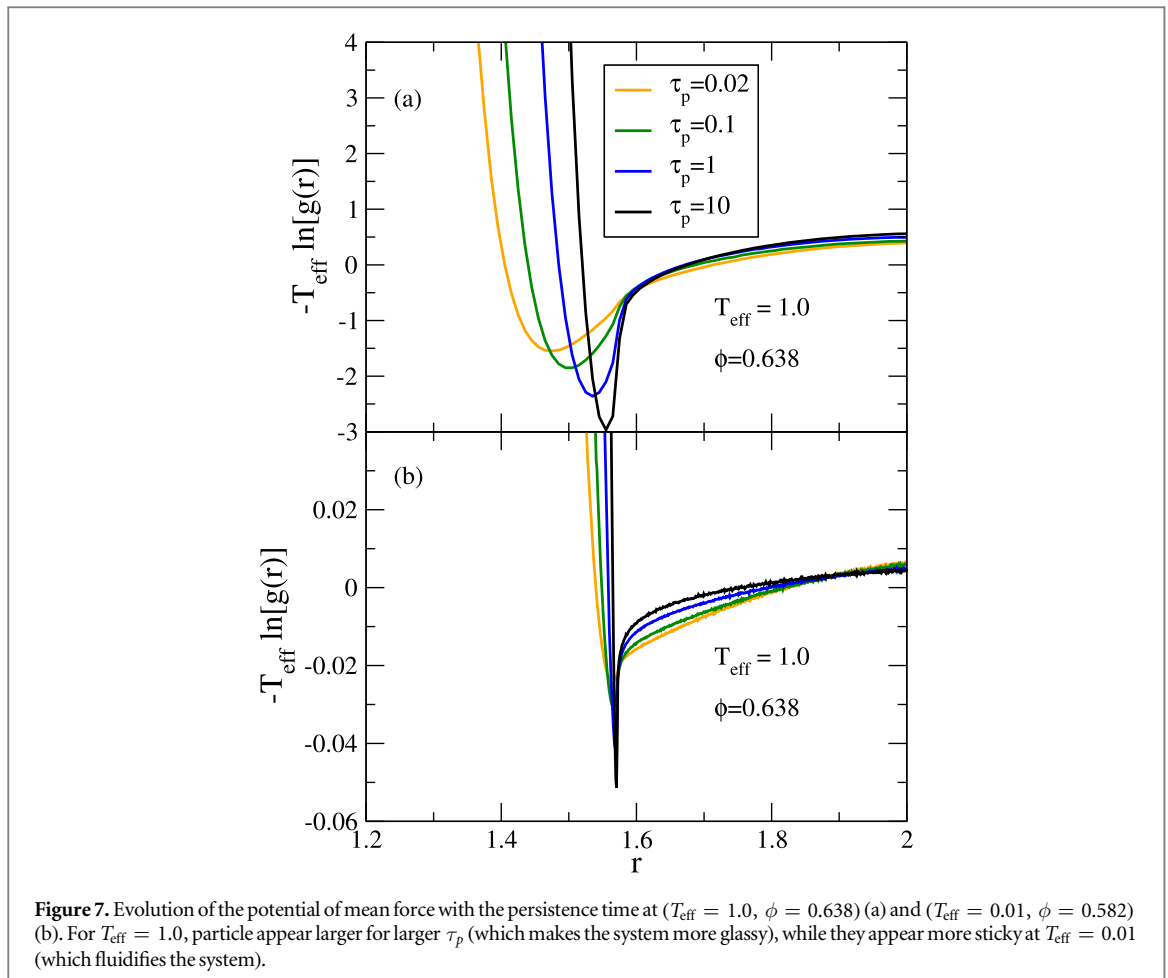


Figure 7. Evolution of the potential of mean force with the persistence time at ($T_{\text{eff}} = 1.0$, $\phi = 0.638$) (a) and ($T_{\text{eff}} = 0.01$, $\phi = 0.582$) (b). For $T_{\text{eff}} = 1.0$, particles appear larger for larger τ_p (which makes the system more glassy), while they appear more sticky at $T_{\text{eff}} = 0.01$ (which fluidifies the system).

adhesion between the particles, which modifies the local structure of the fluid towards the one of equilibrium sticky spheres. It is this effective stickiness which eventually fluidifies the dense fluid. This interpretation also accounts for the subdiffusive plateau dynamics reported in figure 1(b), which stems physically from the existence of two length scales controlling the intermediate time dynamics, the cage size and the adhesion range.

Finally, we note that the above described changes in the potential of mean force and the pair correlation function are not well captured by the approximate mappings of active systems onto effective equilibrium systems described in the literature. In particular, these approximations fail badly for low T_{eff} as they fail to predict effective adhesion between the particles. They can, however, very qualitatively reproduce the effective ‘inflation’ of the particles for soft interactions, and the qualitative change in the steepness. These results show that deriving more accurate effective interactions for nonequilibrium active particles is an important research goal.

6. Discussion

Systems of interacting self-propelled particles undergo a glass transition which has many of the features associated with glass transitions in equilibrium systems. The transition is characterized by a dramatic slowing down upon a small change in control parameters, either temperature, effective temperature, or density. When glassy dynamics set in, the particles become confined in cages of neighboring particles. Consequently, a plateau develops in the mean-squared displacement and the intermediate scattering function. Importantly, profound changes in the dynamics are accompanied by very small changes in the static structure of the particles as observed in two-body correlation functions.

There are, however, differences in the specifics of the transition that become apparent as one increases the departure from equilibrium, which for our system is achieved by increasing the persistence time τ_p . For larger τ_p there is a decrease in the short-time ballistic dynamics due to velocity correlations. The effect of these velocity correlations on the short-time dynamics is correctly captured through a mode-coupling-like theory which we developed previously [9, 15].

Furthermore, increasing τ_p results in particles with smaller cages, which is easily seen in the lower plateau height of the mean-squared displacements, and a larger effective radius, which is easily observed in an increase in the peak position of the pair-correlation function $g(r)$ at the same packing fraction. While the system continues to flow with increasing τ_p , there are differences in the structure. However, at fixed τ_p these structural changes are small and a glass transition still exists. The persistence time dependence of the glass transition can be inferred from the decay of the oscillations of $g(r)$ at a fixed volume fraction. A faster decay indicates a larger glass transition packing fraction. This feature can be observed more easily in the evolution of the peak height of the static structure factor $S(q)$ with increasing τ_p . A smaller height of $S(q)$ at fixed packing fraction leads to a larger glass transition packing fraction.

We can gain some insight into the effect of activity by examining the potential of mean force $u(r) = -T_{\text{eff}} \ln[g(r)]$. While it is important to emphasize that, strictly speaking, $u(r)$ only has meaning for equilibrium systems, it does allow us to gain some insight into how to think about the effect of activity on the behavior of systems. The potential of mean force is reminiscent of systems with short-range attractions, commonly called sticky spheres. With increasing τ_p the attractive range decreases, but the effective particle size increases. The former effect is more pronounced at lower effective temperatures where the width of the well of $u(r)$ is very small and the latter effect seems to be the dominant feature at higher effective temperatures. This shows that predicting the effect of increasing departure from equilibrium is not trivial, and depends on the specifics of the studied model.

Our conclusion is reminiscent of previous findings in equilibrium studies of the glass transition. When the pair structure evolves significantly by varying an external parameter, then the qualitative evolution of the location of the glass transition can be correctly inferred from the sole knowledge of pair density correlations. Good examples are the effect of increasing the adhesion strength in adhesive hard spheres [46], or increasing the density in ultra-soft colloidal particles [47]. However, in order to quantitatively predict the location of the glass transition, it is well-established that two-point density functions are insufficient at equilibrium [48], and that higher-order correlations become relevant [21]. This cautionary remark presumably also applies to active forces, and these more complicated correlations should also be studied in the present nonequilibrium context.

Overall, our work demonstrates that nonequilibrium glass transitions appear robustly in dense active materials, and that much remains to be done at the theoretical level to derive predictive theories, even for very simple models of active particles. Another promising line of research concerns the physical properties of the glass phase, where the influence of the nonequilibrium self-propulsion dynamics on the properties of the glass has not yet been carefully explored.

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References

- [1] Henkes S, Fily Y and Marchetti M C 2011 *Phys. Rev. E* **84** 040301
- [2] Angelini T E, Hannezo E, Trepas X, Marques M, Fredberg J J and Weitz D A 2011 *Proc. Natl Acad. Sci.* **108** 4714
- [3] Berthier L and Kurchan J 2013 *Nat. Phys.* **9** 310
- [4] Ni R, Cohen Stuart M A and Dijkstra M 2013 *Nat. Commun.* **4** 2704
- [5] Berthier L 2014 *Phys. Rev. Lett.* **112** 220602
- [6] Wysocki A, Winkler R G and Gompper G 2014 *Europhys. Lett.* **105** 48004
- [7] Fily Y, Henkes S and Marchetti M C 2014 *Soft Matter* **10** 2132
- [8] Pilkievicz K R and Eaves J D 2014 *Soft Matter* **10** 7495
- [9] Szamel G, Flenner E and Berthier L 2015 *Phys. Rev. E* **91** 062304
- [10] Mandal R, Bhuyan P J, Rao M and Dasgupta C 2016 *Soft Matter* **12** 6268
- [11] Flenner E, Szamel G and Berthier L 2016 *Soft Matter* **12** 7136
- [12] Bi D, Yang X, Marchetti M C and Manning M L 2016 *Phys. Rev X* **6** 021011
- [13] Mandal R, Bhuyan P J, Chaudhuri P, Rao M and Dasgupta C 2017 *Phys. Rev. E* **96** 042605
- [14] Ding H, Jiang H and Hou Z 2017 *Phys. Rev. E* **95** 052608
- [15] Szamel G 2016 *Phys. Rev. E* **93** 012603
- [16] Feng M and Hou Z 2017 *Soft Matter* **13** 4464

- [17] Liluashvili A, Onody J and Voigtmann T 2017 arXiv:1707.07373v1
- [18] Mari R, Krzakala F and Kurchan J 2009 *Phys. Rev. Lett.* **103** 025701
- [19] Ikeda A, Berthier L and Sollich P 2012 *Phys. Rev. Lett.* **109** 018301
- [20] OHern C S, Langer S A, Liu A J and Nagel S R 2002 *Phys. Rev. Lett.* **88** 075507
- [21] Berthier L and Biroli G 2011 *Rev. Mod. Phys.* **83** 587
- [22] Berthier L et al (ed) 2011 *Dynamical Heterogeneities in Glasses, Colloids, and Granular Media* (New York: Oxford University Press)
- [23] Levis D and Berthier L 2015 *Europhys. Lett.* **111** 60006
- [24] Preisler Z and Dijkstra M 2016 *Soft Matter* **12** 6043
- [25] Ginot F, Theurkauff I, Levis D, Ybert C, Bocquet L, Berthier L and Cottin-Bizonne C 2015 *Phys. Rev. X* **5** 011004
- [26] Buttinoni I, Bialké J, Kümmel F, Löwen H, Bechinger C and Speck T 2013 *Phys. Rev. Lett.* **110** 238301
- [27] Deseigne J, Dauchot O and Chaté H 2010 *Phys. Rev. Lett.* **105** 098001
- [28] Schötz E-M, Lanio M, Talbot J A and Manning M L 2013 *J. R. Soc. Interface* **10** 20130726
- [29] Garcia S, Hannezo E, Elgeti J, Joanny J-F, Silberzan P and Gov N S 2015 *Proc. Natl Acad. Sci.* **112** 15314
- [30] Gravish N, Gold G, Zangwill A, Goodisman M A D and Goldman D I 2015 *Soft Matter* **11** 6552
- [31] Tennenbaum M, Liu Z, Hu D and Fernandez-Nieves A 2016 *Nat. Mater.* **15** 54
- [32] Weeks J D, Chandler D and Andersen H C 1971 *J. Chem. Phys.* **54** 5237
- [33] Berthier L and Tarjus G 2009 *Phys. Rev. Lett.* **103** 170601
- [34] Fodor E, Nardini C, Cates M E, Tailleur J, Visco P and van Wijland F 2016 *Phys. Rev. Lett.* **117** 038103
- [35] Szamel G 2014 *Phys. Rev E* **90** 012111
- [36] Maggi C, Marconi U M B, Gnan N and Di Leonardo R 2015 *Sci. Rep.* **5** 10742
- [37] Szamel G 2017 *Europhys. Lett.* **117** 50010
- [38] Hansen J-P and McDonald I R 2012 *Theory of Simple Liquids* (Amsterdam: Elsevier)
- [39] Farage T F F, Krinninger P and Brader J M 2015 *Phys. Rev. E* **91** 042310
- [40] Wittmann R, Maggi C, Sharma A, Scacchi A, Brader J M and Marini Bettolo Marconi U 2017 arXiv:1701.09032v1
- [41] Wittmann R, Marini Bettolo Marconi U, Maggi C and Brader J M 2017 arXiv:1702.00337v1
- [42] Rein M and Speck T 2016 *Eur. Phys. J. E* **39** 84
- [43] Härtel A, Richard D and Speck T 2017 arXiv:1708.01115
- [44] Fuchs M and Cates M E 2009 *J. Rheol.* **53** 957
- [45] Levis D and Berthier L 2014 *Phys. Rev. E* **89** 062301
- [46] Pham K N, Puertas A M, Bergenholtz J, Egelhaaf S U, Moussaid A, Pusey P N, Schofield A B, Cates M E, Fuchs M and Poon W C K 2002 *Science* **296** 104
- [47] Berthier L, Moreno A J and Szamel G 2010 *Phys. Rev. E* **82** 060501
- [48] Berthier L and Tarjus G 2010 *Phys. Rev. E* **82** 031502
- [49] Dougan N, Crowther P, Royall C P and Turci F 2016 *J. Stat. Mech.* **16** 124001