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## The nonequilibrium glassy dynamics of self-propelled particles†

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We study the glassy dynamics taking place in dense assemblies of athermal active particles that are driven solely by a nonequilibrium self-propulsion mechanism. Active forces are modeled as an Ornstein–Uhlenbeck stochastic process, characterized by a persistence time and an effective temperature, and particles interact *via* a Lennard–Jones potential that yields well-studied glassy behavior in the Brownian limit, which is obtained as the persistence time vanishes. By increasing the persistence time, the system departs more strongly from thermal equilibrium and we provide a comprehensive numerical analysis of the structure and dynamics of the resulting active fluid. Finite persistence times profoundly affect the static structure of the fluid and give rise to nonequilibrium velocity correlations that are absent in thermal systems. Despite these nonequilibrium features, for any value of the persistence time we observe a nonequilibrium glass transition as the effective temperature is decreased. Surprisingly, increasing departure from thermal equilibrium is found to promote (rather than suppress) the glassy dynamics. Overall, our results suggest that with increasing persistence time, microscopic properties of the active fluid change quantitatively, but the general features of the nonequilibrium glassy dynamics observed with decreasing the effective temperature remain qualitatively similar to those of thermal glass-formers.

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### 1. Introduction

Systems composed of self-propelled (active) particles represent a class of out-of-equilibrium fluids that exhibit behavior not observed in other fluid systems. One well-studied example is the phase separation of moderately dense self-propelled fluid systems without attractive interactions.<sup>1–6</sup> It is of fundamental interest to understand differences between these novel types of active fluids and equilibrium fluids. In particular, many groups strive to determine whether and what concepts known from the theory of equilibrium fluids can be used to describe active fluids.<sup>7–14</sup> One of the questions discussed recently is the glassy dynamics and the glass transition in self-propelled fluids, and their comparison to the corresponding phenomena in equilibrium fluids.<sup>15–33</sup>

Part of the motivation to analyze the glassy dynamics of active materials stems from experiments. There is ample experimental evidence that active fluids may exhibit glassy dynamics. Angelini *et al.*<sup>15</sup> found, for instance, that migrating cells exhibited glassy dynamics, such as a diminishing self diffusion coefficient and heterogeneous dynamics, as the cell density increases.

Schötz *et al.*<sup>16</sup> found signatures of glassy dynamics in embryonic tissues. More recently, Garcia *et al.*<sup>17</sup> found an amorphous solidification process in collective motion of a cellular monolayer. At larger scales, ant traffic<sup>18</sup> and ant aggregates<sup>19</sup> were shown to display physical processes reminiscent of glassy solids. To understand the glassy dynamics of cells or ant colonies, it is important to understand what are the general features of glassy dynamics that are preserved when a fluid is driven out of equilibrium, and what new behavior may emerge that does not have an equilibrium counterpart.

On the theoretical front, these experimental investigations are complemented by a large number of simulational studies of glassy dynamics, which have mainly focused on the narrower class of self-propelled particles.<sup>20–26</sup> Henkes *et al.*<sup>20</sup> analyzed how activity and aligning velocity interactions excite vibrational modes in an amorphous solid near jamming. Hard particles with self-propulsion were studied in two and three dimensions, with the main outcome being that the equilibrium glass transition is shifted to larger densities when either the magnitude<sup>22</sup> or the persistence time<sup>23,24</sup> of the self-propulsion is increased. Simulational studies of systems with non-aligning continuous interactions started with Wysocki *et al.*,<sup>6</sup> who simulated a dense system of active particles. Fily *et al.*<sup>5</sup> investigated the phase diagram of a dense active system as a function of density, activity and thermal noise, and identified a glassy phase in the high-density, small self-propulsion speed regime, but the detailed structure and glassy dynamics were not analyzed. More recently,

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Mandal *et al.*<sup>28</sup> introduced an active version of the binary Lennard-Jones mixture (the Kob–Andersen model<sup>34</sup>) that we also study in the present work. They found that, upon increasing the magnitude of the self-propulsion, the long-time dynamics speeds up leading to a disappearance of the glass phase, in apparent agreement with hard sphere studies. We shall present below a very different qualitative picture of the glass transition in systems of self-propelled particles.

On the theoretical side, Berthier and Kurchan<sup>21</sup> used a general spin-glass model to argue that systems that have a glass transition in equilibrium could also exhibit a glass transition when driven away from equilibrium through self-propulsion. They concluded that although the specific features of the transition, such as its location, may change with the nature of the non-thermal driving force, general signatures exhibited by thermal glass-forming systems should still be relevant. Farage and Brader<sup>32</sup> attempted to extend the mode-coupling theory for the glass transition to account for activity, and also concluded that self-propulsion could shift, but not destroy, the glass transition. Very recently, Nandi<sup>33</sup> proposed a phenomenological extension of random-first order transition theory to study active glasses.

In an effort to construct a minimal non-trivial model to understand the competition between activity and glassy dynamics, we<sup>25</sup> started a simulational investigation of a self-propelled version of the Kob–Andersen Lennard-Jones binary mixture that is different from the model proposed in ref. 28 in many aspects. Our goal is to construct a model where self-propulsion is the unique source of motion, which neglects aligning interactions, obeys continuous equations of motions and where the equilibrium limit can be taken continuously. To this end, we consider a system with continuous interactions in which the self-propulsion evolves according to an Ornstein–Uhlenbeck stochastic process. This self-propulsion model was introduced by one of us earlier,<sup>35</sup> as a continuous representation of the Monte-Carlo dynamics proposed in ref. 21. An essentially identical model was independently introduced and studied by Maggi *et al.*<sup>36</sup> The general class of model active systems in which the self-propulsion evolves according to an Ornstein–Uhlenbeck process was recently named Active Ornstein–Uhlenbeck Particles (AOUPs) systems.<sup>14</sup> In this class of systems, the evolution of the self-propulsion is controlled by two parameters. An effective temperature quantifies the strength of the self-propulsion force, and a persistence time controls the duration of the persistent self-propelled motion. One attractive feature of this model is that the departure from thermal equilibrium is characterized by one parameter, the persistence time, because in the limit of vanishing persistence time the system becomes equivalent to an equilibrium thermal system at the temperature equal to the effective temperature. Having a model with one parameter that controls a continuous approach to the equilibrium situation is an attractive feature of the model, which is a feature that is not shared by both active Brownian<sup>37</sup> or run-and-rumble<sup>38</sup> particle models. This is also very different from alternative approaches in which the active force acts in addition to thermal motion, where fluidization of the system by addition of active forces is then automatically guaranteed.<sup>28,32,33</sup>

In a previous short account of our results,<sup>25</sup> we showed that with increasing departure from equilibrium for a range of effective temperatures the dynamics can both speed up or slow down. We showed that this effect can be qualitatively rationalized within a mode-coupling-like theory for the dynamics of active systems (see ref. 26 for a detailed presentation of the theory). We found that the important ingredient of this theory was incorporation of the spatial correlations of the velocities of the individual active particles. These velocity correlations have no equilibrium counterpart. Very recently, the existence and properties of velocity correlations in active particles systems were independently studied by Marconi *et al.*<sup>39</sup>

In the present article, we present a very comprehensive set of numerical results regarding the structure and glassy dynamics of the self-propelled Kob–Andersen Lennard-Jones binary mixture which we introduced earlier.<sup>25</sup> We focus on the dependence of the glassy dynamics on the departure from thermal equilibrium, which we quantify by the value of the persistence time of the self-propulsion. By increasing the persistence time from zero to a finite value we continuously move from an overdamped thermal Brownian system through a moderately non-equilibrium system and then to a strongly non-equilibrium self-propelled system. Along the way, we systematically study changes in the structural and dynamic properties of the system. Overall, our results suggest that activity induces profound changes in the detailed structure of the nonequilibrium fluid, but the glassy dynamics of active particles, despite taking place far from thermal equilibrium,<sup>21,24</sup> is qualitatively similar to that observed in equilibrium fluids. We find that active particles display slow dynamics, complex time dependencies of relaxation functions, and spatially heterogeneous dynamics with only quantitative differences between equilibrium and active systems.

The paper is organized as follows. In Section II we describe the self-propulsion model and the interactions. In Section III we describe how the activity influences the pair structure and find that the structure is sensitive to the persistence time, but weakly dependent on the effective temperature at fixed persistence time. We also discuss equal-time velocity correlations, which are a novel feature of self-propelled systems. In Section IV we classify the slowing down at a fixed persistence time and characterize the glass transition as a function of the effective temperatures. In Section V we examine dynamic heterogeneity. We finish with a summary and the conclusions that can be drawn from this work.

## II. Athermal model for self-propelled particles

We simulate a system of interacting self-propelled particles moving in a viscous medium. The motion of the particles is solely due to an internal driving force that evolves according to the Ornstein–Uhlenbeck process. The absence of thermal noise qualifies our model as being ‘athermal’. This model should be applicable for large enough active particles where thermal

noise is negligible compared to their self-propulsion. The equations of motion for the active particles are

$$\dot{\mathbf{r}}_i = \xi_0^{-1}[\mathbf{F}_i + \mathbf{f}_i], \quad (1)$$

where  $\mathbf{r}_i$  is the position of particle  $i$ , the force  $\mathbf{F}_i = -\sum_{j \neq i} \nabla_i V(r_{ij})$

originates from the interactions,  $\mathbf{f}_i$  is the self-propulsion force, and  $\xi_0$  is the friction coefficient of an isolated particle. Note that by using the single-particle friction coefficient in eqn (1) we neglect hydrodynamic interactions. The equations of motion for self-propulsion force  $\mathbf{f}_i$  are given by

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

where  $\tau_p$  is the persistence time of the self-propulsion and  $\boldsymbol{\eta}_i$  is a Gaussian white noise with zero mean and variance  $\langle \boldsymbol{\eta}_i(t) \boldsymbol{\eta}_j(t') \rangle_{\text{noise}} = 2\xi_0 T_{\text{eff}} \mathbf{I} \delta_{ij} \delta(t - t')$ , where  $\langle \cdots \rangle_{\text{noise}}$  denotes averaging over the noise distribution,  $T_{\text{eff}}$  is a single particle effective temperature, and  $\mathbf{I}$  is the unit tensor. We alert the reader that we changed the equation of motion for the self-propulsion, eqn (2), to simplify notation with respect to our earlier work.<sup>25</sup> This also makes it more consistent with the corresponding equation of motion used by Fodor *et al.*<sup>14</sup>

As we noted above, it follows from eqn (1) that our model neglects hydrodynamic interactions. The rationale for this is the same as for thermal glassy systems. There, hydrodynamic interactions are believed to play only a weak role, because glassy dynamics occurs over long time scales that should be decoupled from hydrodynamics. Of course, this assumption could only be validated by careful experimental studies of dense systems of self-propelled colloidal particles, which by construction include both glassiness and hydrodynamic interactions.

The mean-squared displacement of an isolated particle is given by

$$\langle \delta r^2(t) \rangle = \frac{6T_{\text{eff}}}{\xi_0} \left[ \tau_p \left( e^{-t/\tau_p} - 1 \right) + t \right]. \quad (3)$$

For short times,  $t \ll \tau_p$ , the motion is ballistic with  $\langle \delta r^2(t) \rangle \approx 3T_{\text{eff}}\tau_p^{-1}t^2/\xi_0$ , and for long times,  $t \gg \tau_p$ , the motion is diffusive with  $\langle \delta r^2(t) \rangle \approx 6T_{\text{eff}}t/\xi_0$ . At the isolated particle level, the time dependence is thus the same as that of the active Brownian model studied earlier,<sup>20</sup> and we expect that the many-body physics is also very similar.

For a many-particle system, we choose as three independent control parameters, the number density  $\rho$ , the effective temperature  $T_{\text{eff}}$  and the persistence time  $\tau_p$ . For a constant effective temperature, in the limit of vanishing persistence time, the dynamics defined by eqn (1) and (2) approaches overdamped Brownian dynamics at temperature  $T = T_{\text{eff}}$ . Therefore,  $\tau_p$  can be considered as the appropriate measure of the deviation from an equilibrium system undergoing overdamped Brownian motion. For this reason, we compare some results to simulations of the corresponding overdamped Brownian system.<sup>40,41</sup> We note that an approximate mapping has recently been proposed<sup>12</sup> between our model and the standard active Brownian particles model.<sup>37</sup>

We simulate the classic Kob–Andersen 80 : 20 binary mixture at a number density  $\rho = 1.2$  to study the glassy dynamics.

At such large density, the physics of phase separation is irrelevant and we can focus exclusively on the glassy dynamics of a homogeneous active fluid. The particles interact *via* a Lennard-Jones potential

$$V_{\alpha\beta}(r_{nm}) = 4\epsilon_{\alpha\beta} \left[ \left( \frac{\sigma_{\alpha\beta}}{r_{nm}} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r_{nm}} \right)^6 \right], \quad (4)$$

where  $r_{nm} = |\mathbf{r}_n - \mathbf{r}_m|$ . The interaction parameters are  $\epsilon_{\text{BB}} = 0.5\epsilon_{\text{AA}}$ ,  $\epsilon_{\text{AB}} = 1.5\epsilon_{\text{AA}}$ ,  $\sigma_{\text{BB}} = 0.88\sigma_{\text{AA}}$ , and  $\sigma_{\text{AB}} = 0.8\sigma_{\text{AA}}$ . The results will be presented in reduced units where  $\epsilon_{\text{AA}}$  is the unit of energy,  $\sigma_{\text{AA}}$  is the unit of length, and  $\sigma_{\text{AA}}\xi_0/\epsilon_{\text{AA}}$  is the unit of time. Most simulations used systems of 1000 particles with periodic boundary conditions. A few runs that focused on the small wave-vector behavior of certain correlation functions used systems of 27 000 particles. Since the equations of motion allow for a drift of the center of mass, we corrected for this center of mass drift in calculating time correlation functions.

We simulated persistence times  $\tau_p = 2 \times 10^{-4}$ ,  $5 \times 10^{-4}$ ,  $2 \times 10^{-3}$ ,  $1 \times 10^{-2}$ ,  $2 \times 10^{-2}$ ,  $3 \times 10^{-2}$ ,  $5 \times 10^{-2}$ , and  $1 \times 10^{-1}$ . For each persistence time we simulated systems for  $T_{\text{eff}} = 2.2$ , 2.0, 1.8, 1.6, 1.4, 1.2, 1.0, 0.95, 0.9, 0.85, 0.8, 0.75, 0.7, and 0.65. For  $\tau_p = 2 \times 10^{-4}$ ,  $5 \times 10^{-4}$ ,  $1 \times 10^{-2}$ , and  $2 \times 10^{-2}$  we also simulated  $T_{\text{eff}} = 0.6$ , 0.55, 0.5, and 0.47. For  $\tau_p = 3 \times 10^{-2}$  we also simulated  $T_{\text{eff}} = 0.6$ , 0.55, and 0.5. For  $\tau_p = 5 \times 10^{-2}$  and  $\tau_p = 1 \times 10^{-1}$  we also simulated  $T_{\text{eff}} = 1.1$ . This range of temperatures allowed us to study the high temperature liquid-like behavior of the active fluid down to the glassy dynamics regime.

### III. Equal-time correlations of particles' positions and velocities

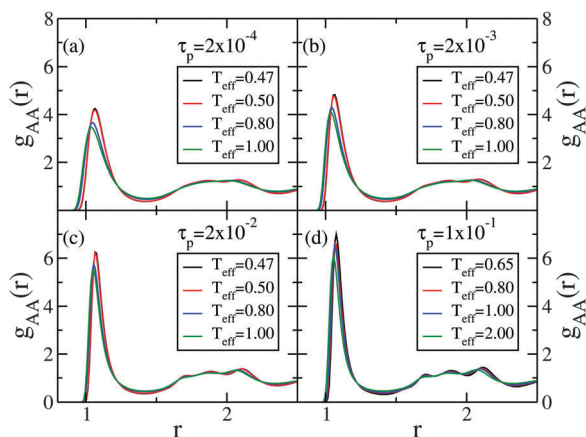
Glassy dynamics is associated with a dramatic increase of the relaxation time with a small change of the control parameter (generally temperature or density) without a dramatic change in the structure. In this section we discuss the changes in the steady-state structure by examining two equal-time two-body distribution functions, the well-known pair distribution function and a novel function characterizing spatial correlations of the velocities of the self-propelled particles.

#### A. Pair correlation functions

The pair distribution function is defined as<sup>42</sup>

$$g_{\alpha\beta}(r) = \frac{V}{N_\alpha N_\beta} \left\langle \sum_n \sum_{m \neq n} \delta(\mathbf{r} - (\mathbf{r}_n - \mathbf{r}_m)) \right\rangle. \quad (5)$$

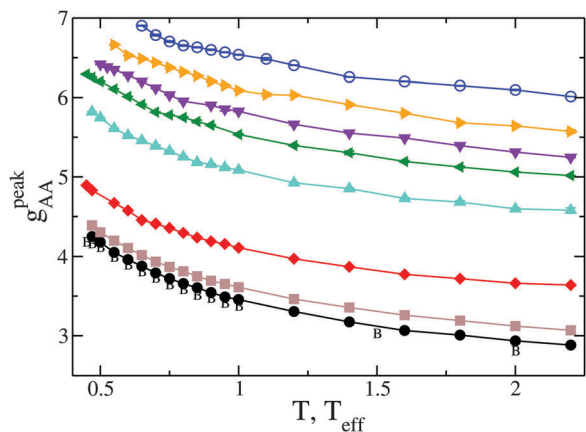
Shown in Fig. 1 is the pair distribution function for the larger particles,  $g_{\text{AA}}(r)$ , for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $2 \times 10^{-3}$ , (c)  $2 \times 10^{-2}$  and (d)  $1 \times 10^{-1}$  for some representative effective temperatures. We note that  $g_{\text{AA}}(r)$  for  $\tau_p = 2 \times 10^{-4}$  is nearly identical to  $g_{\text{AA}}(r)$  obtained for over-damped Brownian dynamics simulations.<sup>40,41</sup> However, for the larger persistence times there is an enhancement of the structure as shown by the increased height of the first peak of  $g_{\text{AA}}(r)$ . Furthermore, for the largest persistence time shown, the second peak begins to split into three peaks.



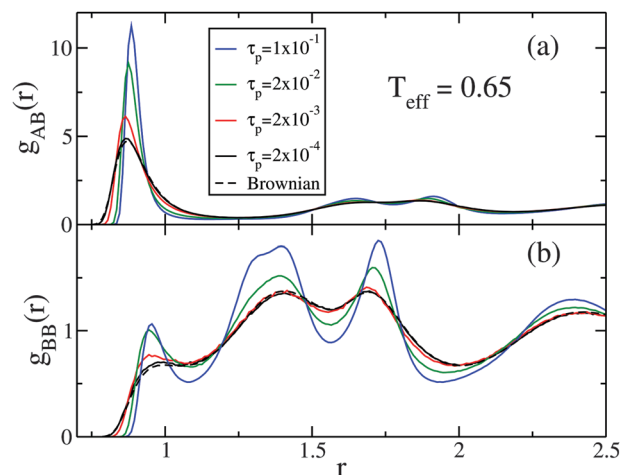
**Fig. 1** The pair distribution function  $g_{AA}(r)$  calculated for the larger, more abundant A particles for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $2 \times 10^{-3}$ , (c)  $2 \times 10^{-2}$  and (d)  $1 \times 10^{-1}$ . For panel (d), we could not equilibrate the system below  $T_{\text{eff}} = 0.65$ , thus we do not show data for  $T_{\text{eff}} = 0.47$ . For all  $\tau_p$ , the weak temperature dependence contrasts with the profound influence of the persistence time on the pair structure.

While it is often observed that the second peak of  $g_{AA}(r)$  begins to split into two peaks for the thermal Kob–Andersen system at low temperatures, the splitting into three peaks is typically not observed. These results suggest that the self-propulsion can promote the presence of structures that are not favored in the thermal system, indicating that the local structure of the active fluid is presumably very hard to predict from the sole knowledge of the pair potential. Importantly, the structure does not significantly change with decreasing  $T_{\text{eff}}$ . In particular, we see no evidence of crystallization or phase separation between the two species for any of the simulations.

To quantify the change in structure we calculated the height of the first peak of  $g_{AA}(r)$ ,  $g_{AA}^{\text{peak}}$ , as a function of  $T_{\text{eff}}$  for each persistence time. The results are shown in Fig. 2. For a fixed persistence time,  $g_{AA}^{\text{peak}}$  increases as  $T_{\text{eff}}$  decreases. At fixed  $T_{\text{eff}}$ ,  $g_{AA}^{\text{peak}}$  increases with increasing persistence time, which demonstrates



**Fig. 2** The height of the first peak of  $g_{AA}(r)$  as a function of the effective temperature  $T_{\text{eff}}$  for  $\tau_p = 2 \times 10^{-4}$ ,  $5 \times 10^{-4}$ ,  $2 \times 10^{-3}$ ,  $1 \times 10^{-2}$ ,  $2 \times 10^{-2}$ ,  $3 \times 10^{-2}$ ,  $5 \times 10^{-2}$ , and  $1 \times 10^{-1}$  listed from bottom to top. The results for over-damped Brownian dynamics from ref. 40 is shown using 'B' symbols.



**Fig. 3** The pair distribution functions  $g_{AB}(r)$  and  $g_{BB}(r)$  at  $T_{\text{eff}} = 0.65$  for  $\tau_p = 2 \times 10^{-4}$ ,  $2 \times 10^{-3}$ ,  $2 \times 10^{-2}$ , and  $1 \times 10^{-1}$ . The results for over-damped Brownian dynamics from ref. 40 are shown as dashed lines. Increasing the persistence time modifies strongly the local structure of the fluid.

that the structure is enhanced with increasing persistence times. We note that the influence of the persistence time on the peak is much more pronounced than the influence of the effective temperature. This means that, for a given persistence time, the local structure changes weakly as the nonequilibrium glass transition temperature is approached, which is reminiscent of the equilibrium glass phenomenology. We note, for later reference, that the relative change of the peak height as temperature decreases is actually weaker for the largest persistence time than it is for the equilibrium model.

Finally, we examined distribution functions involving the minority component,  $g_{AB}(r)$  and  $g_{BB}(r)$ , at a fixed  $T_{\text{eff}} = 0.65$  for  $\tau_p = 2 \times 10^{-4}$ ,  $2 \times 10^{-3}$ ,  $2 \times 10^{-2}$ ,  $1 \times 10^{-1}$  and for over-damped Brownian dynamics. These functions are shown in Fig. 3. Note that  $T_{\text{eff}} = 0.65$  is the lowest effective temperature where we have equilibrated simulations for  $\tau_p = 1 \times 10^{-1}$ .

We find that with increasing persistence time the height of the first peak of  $g_{AB}(r)$  in Fig. 3(a) increases and the splitting of the second peak becomes more pronounced. Furthermore, the width of the first peak decreases, which suggests an increased ordering. The enhanced structure is more pronounced when one examines  $g_{BB}(r)$ , which is shown in Fig. 3(b). For  $g_{BB}(r)$  there is a pronounced increase of the first four peaks. Notably, the first peak is generally a non-distinct shoulder for this model in the Brownian limit. It becomes much more pronounced with the increasing  $\tau_p$ . Recall that  $\sigma_{BB} = 0.88$ , thus this peak corresponds to an increased probability of finding another B particle in the first neighbor shell of a B particle.

An interesting question, which is beyond the scope of this work, is how the self-propulsion influences the relative populations of locally favored structures<sup>43</sup> of the system. Answering this question could lead to a new route to make desired structures more favorable in dense liquids. What we do observe here is that increasing persistence times of the self-propulsion changes the local structure of a dense binary fluid in a highly non-trivial manner. This implies that the active Lennard-Jones fluid driven

by nonequilibrium self-propulsion forces behaves as a dense fluid with thermodynamic properties that are quantitatively distinct from the original equilibrium model, and it becomes a 'different' fluid.

## B. Velocity correlations

Most theories of equilibrium fluid dynamics use as input the pair distribution function or quantities that can be calculated from this function.<sup>42,44</sup> While deriving a theory for the dynamics of active systems,<sup>25,26</sup> we discovered that the active fluid dynamics is also influenced by correlations of velocities of individual active particles. This influence appears in two places. First, the short-time dynamics can be analyzed exactly and one can show that it is determined by a pair correlation function of the particles' velocities.<sup>26</sup> Second, within an approximate theory, which is similar in spirit to the well-known theory for the dynamics of glass forming fluids, the mode-coupling theory,<sup>45</sup> the long-time dynamics is influenced by the same correlation function.<sup>26</sup> We emphasize that this correlation function does not have an equilibrium analogue. It becomes structure-less in the limit of vanishing persistence time, *i.e.* when our system becomes equivalent to an equilibrium (thermal) system. We should mention that the existence and importance of velocity correlations in active particles systems were also recently stressed by Marconi *et al.*<sup>39</sup>

Our theoretical analysis, therefore, leads us to the following correlation function of the particle velocities,

$$\omega_{\parallel}(q) = \frac{1}{N\xi_0^2} \left\langle \left| \hat{\mathbf{q}} \cdot \sum_i (\mathbf{F}_i + \mathbf{f}_i) e^{-i\mathbf{q} \cdot \mathbf{r}_i} \right|^2 \right\rangle. \quad (6)$$

In eqn (6),  $\xi_0^{-1}(\mathbf{F}_i + \mathbf{f}_i)$  is the velocity of particle  $i$ , see eqn (1), and thus  $\omega_{\parallel}(q)$  quantifies longitudinal spatial correlations of the velocities of the individual particles.

It turns out that both the wave-vector dependence of  $\omega_{\parallel}(q)$  and its overall magnitude change with our two control parameters,  $T_{\text{eff}}$  and  $\tau_p$ . For this reason, we first show the changes in the wave-vector dependence by presenting velocity correlations normalized by their infinite wave-vector limit,  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$ , and then we show the changes in the overall magnitude by presenting the infinite wave-vector limit,  $\omega_{\parallel}(\infty)$ . We note that  $\omega_{\parallel}(\infty)$  is related to the average magnitude of the velocity,

$$\omega_{\parallel}(\infty) = \frac{1}{3N\xi_0^2} \left\langle \sum_i (\mathbf{F}_i + \mathbf{f}_i)^2 \right\rangle. \quad (7)$$

In Fig. 4(a) we show  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  for  $\tau_p = 1 \times 10^{-1}$  at  $T_{\text{eff}} = 2.0, 1.0, 0.8,$  and  $0.65$ . We find that, possibly except at the smallest wave-vectors, there is very little change in  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  with decreasing  $T_{\text{eff}}$  at fixed  $\tau_p$ , and we find this to be the case for every  $\tau_p$ . However, as shown in Fig. 4(b), there is a large change in  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  with  $\tau_p$  at constant  $T_{\text{eff}} = 1.4$ . Thus, the increase of the velocity correlations is a consequence of the increased persistence time, but this structural measure also does not change significantly with decreasing effective temperature. This trend is similar to the observation made for the pair distribution function.

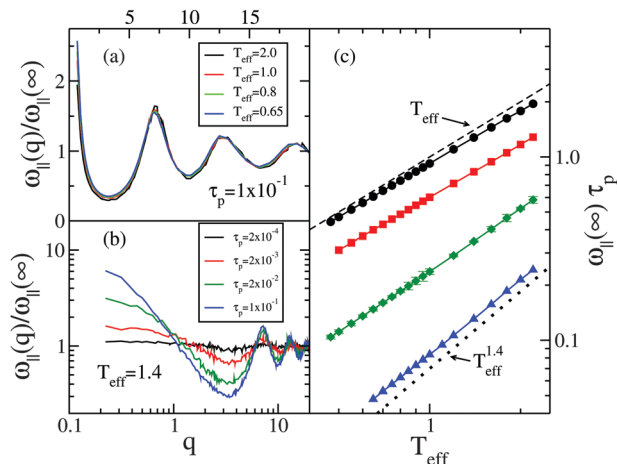


Fig. 4 (a) Velocity correlations as measured by  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  for  $T_{\text{eff}} = 2.0, 1.0, 0.8,$  and  $0.65$  for  $\tau_p = 1 \times 10^{-1}$ . (b) Velocity correlations as measured by  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  for  $\tau_p = 2 \times 10^{-4}, 2 \times 10^{-3}, 2 \times 10^{-2},$  and  $1 \times 10^{-1}$  at a fixed  $T_{\text{eff}} = 1.4$ . (c)  $\omega_{\parallel}(\infty)\tau_p$  for  $\tau_p = 2 \times 10^{-4}, 2 \times 10^{-3}, 2 \times 10^{-2},$  and  $1 \times 10^{-1}$  as a function of  $T_{\text{eff}}$ .

We note two different aspects of the increasing velocity correlations. First,  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  develops finite wave-vector oscillatory structure, which, roughly speaking, follows that of the static structure factor. Second,  $\omega_{\parallel}(q)/\omega_{\parallel}(\infty)$  develops a peak at zero wave-vector, which indicates a growing velocity correlation length. A detailed investigation of this latter feature is left for a future study. Here we only note that at  $T_{\text{eff}} = 1.4$  and  $\tau_p = 1 \times 10^{-1}$ , which corresponds to a liquid state, this length reaches the value of about 2.6 particle diameters (recall that this correlation length scale is not even defined for Brownian particles). While still modest, such a correlation length is longer than any other structural length in an equilibrium fluid with similar dynamics. Thus, increasing the persistence time induces the emergence of non-trivial spatial correlations in the active fluid that are fully nonequilibrium in nature.

We only investigated velocity correlations in the reciprocal space, but in future research it would be interesting to complement these by calculating correlation functions in real space. On the basis of past experience with correlation functions describing the dynamic heterogeneity (see Section V), we expect that the velocity correlation function in real space will exhibit oscillations mirroring those of the pair distribution function, with a large-distance tail reflecting the growth of the velocity correlation length.

In Fig. 4(c) we present the dependence of the overall magnitude of  $\omega_{\parallel}(q)$  on  $T_{\text{eff}}$  and  $\tau_p$  by plotting  $\tau_p\omega_{\parallel}(\infty)$ . We found<sup>26</sup> that this quantity is a measure of the intermediate time dynamics (under the assumption that there is a separation of time scales between short-time ballistic motion and the long-time motion heavily influenced by the time-dependent internal friction). We show the dependence of  $\tau_p\omega_{\parallel}(\infty)$  on  $T_{\text{eff}}$  for  $\tau_p = 2 \times 10^{-4}, 2 \times 10^{-3}, 2 \times 10^{-2}, 1 \times 10^{-1}$  listed from top to bottom. Since at a given effective temperature  $T_{\text{eff}}$  the strength of the self-propulsion force is  $|\mathbf{f}| \sim \sqrt{T_{\text{eff}}/\tau_p}$ , an increase of the  $\tau_p$  at fixed  $T_{\text{eff}}$  will result in an overall decrease of  $\omega_{\parallel}(\infty)$ . If the self-propulsion

force dominates the velocity correlations, one would expect that  $\tau_p \omega_{\parallel}(\infty) \sim F^2$ , thus  $\tau_p \omega_{\parallel}(\infty) \sim T_{\text{eff}}$ , and we find that there is an approximate linear decrease of  $\tau_p \omega_{\parallel}(\infty)$  with  $T_{\text{eff}}$  for  $\tau_p = 2 \times 10^{-4}$ . However,  $\tau_p \omega_{\parallel}(\infty)$  no longer decreases linearly with decreasing  $T_{\text{eff}}$  for larger  $\tau_p$ , and  $\tau_p \omega_{\parallel}(\infty) \sim T_{\text{eff}}^{1.4}$  for  $\tau_p = 1 \times 10^{-1}$ . Therefore, there is an increased influence of the particle interactions with increasing persistence time, which is consistent with findings of Marconi *et al.*<sup>39</sup> Physically, this implies that the intermediate-time dynamics slows down more rapidly with decreasing effective temperature for large persistence times than for small ones.

While both we<sup>25,26</sup> and Marconi *et al.*<sup>39</sup> realized the importance of velocity correlations through theoretical considerations, equal-time velocity correlations have also been noticed and analyzed in experiments. Garcia *et al.*<sup>17</sup> monitored equal-time velocity correlations and extracted a velocity correlation length. The maximum value reached by this correlation length was about 7 cell sizes. While the experimental system analyzed by Garcia *et al.* is quite complex and different from the simple model system considered here, the presence of spatially extended velocity correlations in both systems suggests that they are a generic feature of out-of-equilibrium assemblies of self-propelled particles. It would be very interesting to measure and analyze these equal-time velocity correlations also in experimental active systems which are closer to our model system, such as for instance the self-propelled grains of Deseigne *et al.*<sup>46</sup> or active Janus colloidal systems of the Lyon<sup>47</sup> and Stuttgart<sup>48</sup> groups.

## IV. Dynamic slowing down and the glass transition

One fundamental feature of glassy dynamics is the dramatic slowing down of the dynamics accompanied by little change in the two-body static structure. In the previous section we found little change in the structure with decreasing the effective temperature  $T_{\text{eff}}$  at fixed persistence time  $\tau_p$ . In this section we examine the slowing down of the dynamics upon changing the control parameters in the same way, *i.e.* with decreasing the effective temperature at fixed persistence time.

### A. Self-intermediate scattering function

To examine the dynamics we study the self-intermediate scattering function for the A particles

$$F_s(k; t) = \frac{1}{N_A} \left\langle \sum_{n=1}^{N_A} e^{i\mathbf{k} \cdot (\mathbf{r}_n(t) - \mathbf{r}_n(0))} \right\rangle. \quad (8)$$

While we only present results for the more abundant, larger A particles, similar conclusions are drawn if one examines the dynamics of the smaller B particles. We choose  $k = 7.2$ , which is around the first peak of the partial static structure factor calculated for the A particles. Shown in Fig. 5 is  $F_s(k; t)$  for representative values of the persistence time. While we observed little change in the structure with decreasing  $T_{\text{eff}}$  at fixed  $\tau_p$ , we do see a dramatic slowing down of the dynamics, in the sense that the relaxation becomes very slow at low temperatures.

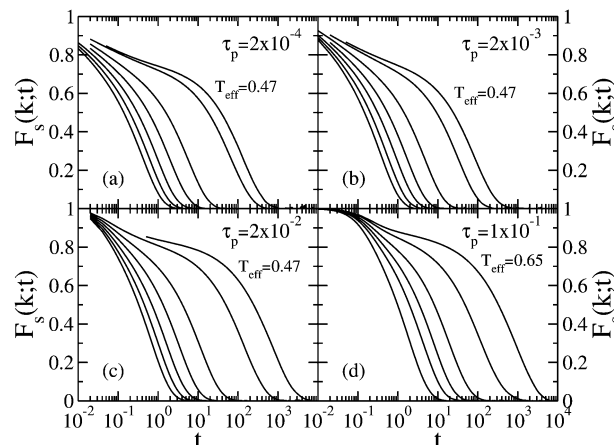


Fig. 5 The self-intermediate scattering function  $F_s(k; t)$  from  $T_{\text{eff}} = 2.2$  down to the lowest temperature we could equilibrate for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $\tau_p = 2 \times 10^{-3}$ , (c)  $\tau_p = 2 \times 10^{-2}$ , and (d)  $\tau_p = 1 \times 10^{-2}$ . For panels (a–c)  $T_{\text{eff}} = 1.0, 0.9, 0.8, 0.7, 0.6, 0.5, 0.47$  are shown (left to right). In panel (d)  $T_{\text{eff}} = 1.1, 1.0, 0.9, 0.8, 0.7, 0.65$  are shown (left to right). In all cases, nonequilibrium glassy dynamics is observed as  $T_{\text{eff}}$  decreases.

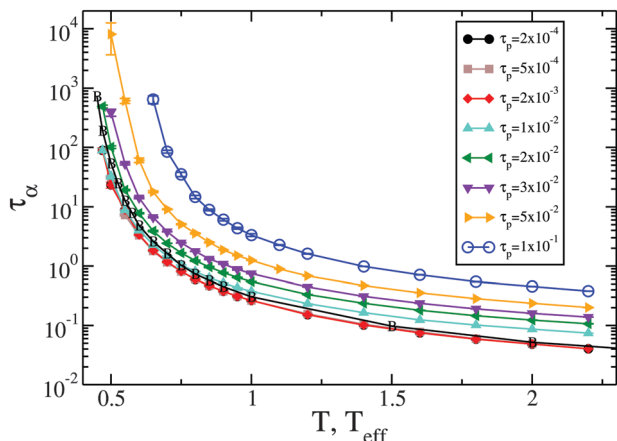
Also, as is commonly observed in equilibrium supercooled liquids, the functional form of the time decay of the correlation functions is relatively independent of temperature, and is well-described by a stretched exponential form  $F(k; t) \approx A \exp[-(t/\tau)^\beta]$ . Our fits to the stretched exponential form indicate that  $\beta$  decreases modestly with increasing the degree of supercooling from about 0.87 to 0.7, for a given persistence time. Importantly, this evolution of  $\beta$  with decreasing the effective temperature appears to be independent of the persistence time, as the same trend is observed for all studied persistence times.

Therefore, in the following we mostly focus on the relaxation time  $\tau_\alpha$  of  $F_s(k; t)$  which we define as when  $F_s(k; \tau_\alpha) = e^{-1}$ .

### B. Relaxation times

Shown in Fig. 6 is the  $\alpha$  relaxation time,  $\tau_\alpha$ , as a function of the effective temperature,  $T_{\text{eff}}$ , for all the persistence times studied in this work. Also shown with ‘B’ symbols is  $\tau_\alpha$  for overdamped Brownian dynamics simulations of the same system.<sup>40,41</sup> Note that at small  $\tau_p$  and a fixed  $T_{\text{eff}}$ , the active system relaxes faster than the thermal Brownian system at  $T = T_{\text{eff}}$ . The relaxation time becomes larger than that of the over-damped Brownian system for  $\tau_p \geq 2 \times 10^{-2}$ . This non-monotonic dependence of the relaxation time on the persistence time was reported previously, and a mode-coupling like theory was developed that also predicted a non-monotonic change of the relaxation time with the persistence time.<sup>25,26</sup> The physics behind the non-monotonic behavior is a competition between increasing structure in the velocity correlations (speeding up the dynamics) and an increase in the local structure (slowing down the dynamics) as  $\tau_p$  increases.

Below we shall analyze in great detail the dramatic increase of  $\tau_\alpha$  with decreasing effective temperature. First, it is useful to notice that the dynamics in the high effective temperature liquid also bears a non-trivial dependence on the persistence time. For  $T_{\text{eff}} = 2.2$ , for instance, we observe that  $\tau_\alpha$  increases by



**Fig. 6** The dependence of the relaxation time  $\tau_\alpha$  on  $T_{\text{eff}}$  for all the persistence times studied in this work. The results for over-damped Brownian dynamics from ref. 40 is shown using 'B' symbols. Note that on the scale of the figure the results for the three shortest persistence times are almost identical, whereas larger persistence times seem to promote, rather than suppress, the glassy dynamics.

about one order of magnitude between Brownian dynamics and our largest simulated  $\tau_p$ . This change mirrors the increase in  $\omega_{\parallel}(\infty)\tau_p$  reported in Fig. 4(c). This is reasonable as  $\omega_{\parallel}(\infty)\tau_p$  controls the intermediate-time dynamics, and thus it also affects the structural relaxation time in the high-temperature fluid, where glassy effects at long times are absent.

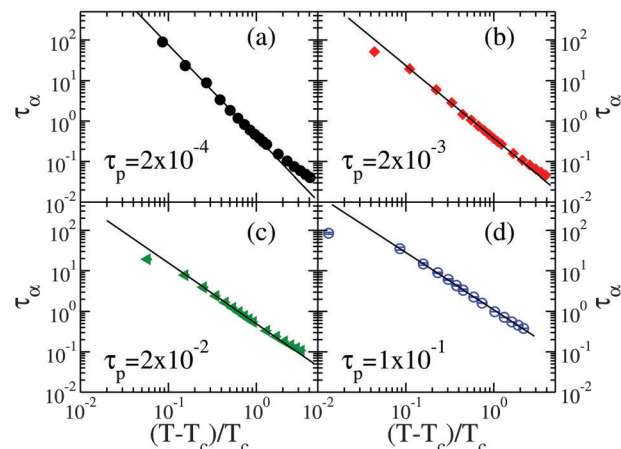
In contrast to the small changes in structure with decreasing effective temperature shown in the previous section, Section III, there is a very large increase in the relaxation time with a small decrease of the effective temperature. We note that changing  $\tau_p$  can dramatically change the structure, and thus it makes sense to examine the glass transition at a fixed  $\tau_p$  and using  $T_{\text{eff}}$  as the control parameter.

Finally, we note that increasing departure from the thermal equilibrium does not 'fluidize' the glassy system under study, and neither does the glass transition disappear completely, contrary to previous reports.<sup>28,33</sup> The difference stems from the fact that in the model of ref. 28, active forces act in addition to Brownian noise, so that the bath temperature is in fact not the correct control parameter to describe the glassy dynamics.

### C. Mode-coupling analysis of relaxation times

One common way to examine glassy dynamics in thermal systems is to investigate power-law fits of the relaxation time inspired by the mode-coupling theory.<sup>45</sup> These fits result in the so-called mode-coupling transition temperature  $T^c$ , which is interpreted as a crossover between a moderately supercooled regime where mode-coupling theory is applicable and a strongly supercooled regime where the dynamics is thought to be dominated by somewhat vaguely defined hopping events. The development of a mode-coupling-like dynamic theory for active systems<sup>21,25,26</sup> suggests that similar analysis could be performed for active glassy dynamics.

We examine a mode-coupling like regime by fitting  $\tau_\alpha = a(T_{\text{eff}}/T_{\text{eff}}^c - 1)^{-7}$  for  $T_{\text{eff}}$  lower than the onset temperature,

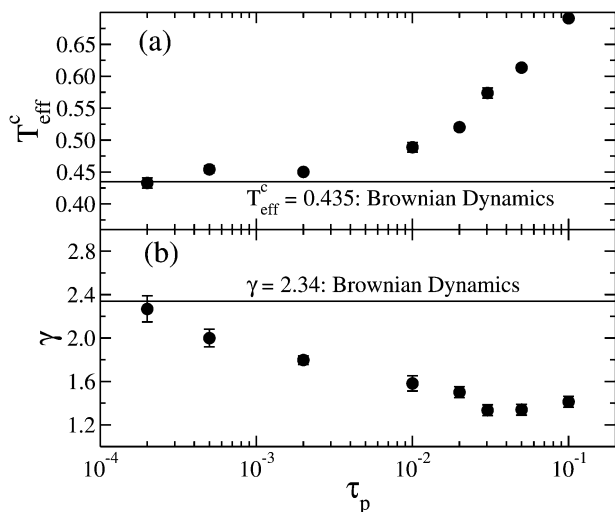


**Fig. 7** Mode-coupling-like power law fits  $\tau_\alpha \propto (T_{\text{eff}} - T_{\text{eff}}^c)^{-7}$  for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $2 \times 10^{-3}$ , (c)  $2 \times 10^{-2}$ , and (d)  $1 \times 10^{-1}$ . In all cases, there is a modest intermediate time regime where the power-law fit holds, as commonly found in equilibrium supercooled liquids.

which we define as the highest temperature where  $\tau_\alpha$  deviates from the high temperature Arrhenius behavior (discussed further below). Shown in Fig. 7 are the fits for four representative persistence times. For each persistence time the mode-coupling like fits provide a reasonable description of the data for around two decades of slowing down. Like for thermal systems, there are deviations from the mode-coupling behavior at the lowest effective temperatures that we could simulate. Notice that the quality (or poorness) of such power law fit does not seem to be very different between Brownian and self-propelled dynamics. A different result was obtained for self-propelled disks where the quality of the fits seemed to improve with increasing the persistence time.<sup>23</sup> The present result for the binary Lennard-Jones model then provides a counter-example suggesting that the finding for the hard disk system is not universal.

Shown in Fig. 8(a) are the mode-coupling effective temperatures,  $T_{\text{eff}}^c$ , resulting from power-law fits as a function of the persistence time  $\tau_p$ . For small  $\tau_p$ ,  $T_{\text{eff}}^c$  is close to the value of  $T^c \approx 0.435$  found in previous studies of the Kob-Andersen system undergoing Newtonian<sup>34</sup> or Brownian dynamics.<sup>40</sup> For larger  $\tau_p$ , the mode-coupling effective temperature increases monotonically with increasing persistence time.

Our somewhat surprising finding is that moving away from equilibrium by increasing the persistence time promotes the glassy dynamics, which occurs at larger values of the effective temperatures. Therefore, the self-propelled Lennard-Jones system appears more glassy than the Brownian version with the same interaction potential. This finding was not fully expected as previous studies using hard potentials have reported that the glassy dynamics is pushed to larger densities.<sup>22,23</sup> The present result then suggests that, again, the finding for hard particles is not universal and the opposite situation is in fact possible. This was suggested theoretically on general grounds in ref. 21. Physically these findings show that the manner in which the glass transition shifts when departing from thermal equilibrium stems from a non-trivial combination of how the local



**Fig. 8** Mode-coupling effective temperature  $T_{\text{eff}}^c$  and scaling exponent  $\gamma$  obtained from power-law fits to  $\tau_\alpha \propto (T_{\text{eff}} - T_{\text{eff}}^c)^{-\gamma}$  as a function of  $\tau_p$ . The horizontal lines are the results for the over-damped Brownian dynamics simulations taken from ref. 40. The increase of  $T_{\text{eff}}^c$  with  $\tau_p$  directly indicates that self-propulsion promotes glassy dynamics, whereas the strong change in  $\gamma$  is correlated to the strong change in the local structure observed at large persistence times.

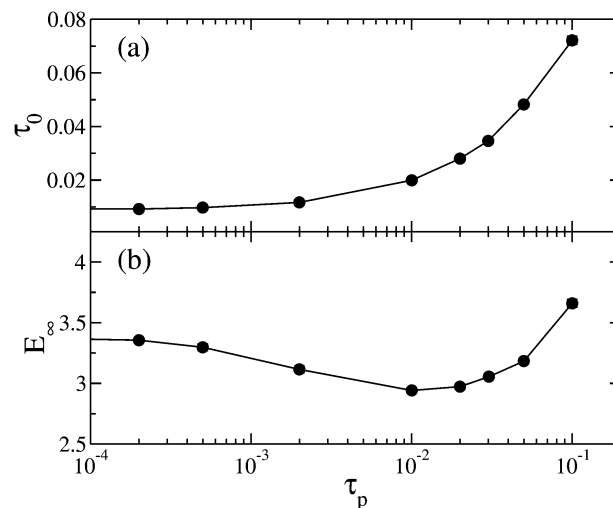
structure and velocity correlations are affected by the self-propulsion. No obvious qualitative guess can be made and details of the original Brownian system matter.

Shown in Fig. 8(b) is the mode-coupling exponent  $\gamma$  as a function of  $\tau_p$ . For  $\tau_p = 2 \times 10^{-4}$ ,  $\gamma$  is equal to the value obtained from Brownian dynamics simulations.<sup>40</sup> With increasing  $\tau_p$  there is an initial decrease in  $\gamma$  until  $\tau_p \approx 3 \times 10^{-2}$ , then  $\gamma$  is approximately constant for the largest persistence times studied in this work. In the equilibrium theory, the exponent  $\gamma$  follows, in a non-trivial way, from the static structure of the fluid.<sup>45</sup> The situation is unclear for the nonequilibrium dynamics but the evolution of  $\gamma$  appears qualitatively consistent with the dramatic change in the local structure observed in the pair correlation function shown in Fig. 1.

#### D. Fragility and activated dynamics

A different examination of the glassy dynamics focuses on the departures from the high temperature Arrhenius behavior (see, e.g. ref. 49). To this end we start by fitting the relaxation time to the Arrhenius-like formula,  $\tau_\alpha = \tau_0 \exp[E_\infty/T_{\text{eff}}]$ , for  $T_{\text{eff}} \geq 1.2$  and fixed  $\tau_p$  to obtain the time constant  $\tau_0$  and the activation energy  $E_\infty$ , see Fig. 9, at each persistence time. We see that  $\tau_0$  grows monotonically with increasing persistence time in a manner which is again reminiscent of the changes observed in  $\tau_p \omega_1(\infty)$  in Fig. 4(c) and of the high-temperature liquid dynamics in Fig. 6.

In contrast, the effective activation energy  $E_\infty$  initially decreases with increasing persistence time and then increases at higher persistence times. The minimum in  $E_\infty$  is around  $\tau_p = 1 \times 10^{-2}$ . This non-monotonic dependence of  $E_\infty$  is mirrored by the non-monotonic dependence of  $\tau_\alpha$  versus  $\tau_p$  identified in ref. 25 and already discussed above. Note that

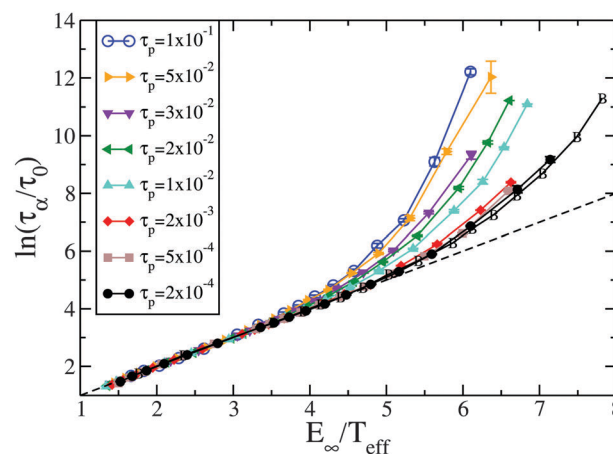


**Fig. 9** The time constant  $\tau_0$  and the activation energy  $E_\infty$  resulting from the Arrhenius-like fit  $\tau_\alpha = \tau_0 \exp(E_\infty/T_{\text{eff}})$ . The fits were performed in the high-temperature regime, i.e. for  $T_{\text{eff}} \geq 1.2$ , for each persistence time.

$E_\infty$  characterizes the dynamics of the high-temperature active fluid, which exhibits a non-monotonic evolution with  $\tau_p$ .

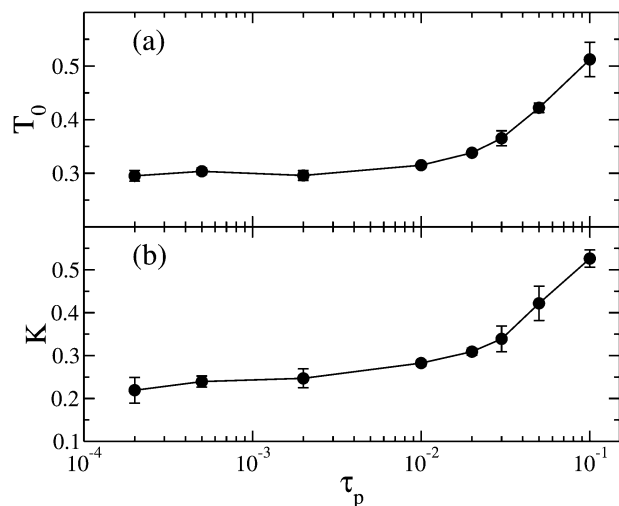
In Fig. 10, we show that, once the high-temperature Arrhenius regime has been scaled out, the glassy dynamics evolves monotonically with the persistence time. To this end, we plot  $\ln(\tau_\alpha/\tau_0)$  versus  $E_\infty/T_{\text{eff}}$ . When rescaled by the high temperature behavior of the active liquid, we now find that the slowing down, compared to the high temperature dynamics, monotonically increases with increasing  $\tau_p$ . Furthermore, the liquid dynamics appear to be more fragile<sup>50</sup> with increasing  $\tau_p$ , in the sense that the temperature dependence in the glassy regime becomes much steeper for larger persistence times.

To quantify the kinetic fragility and examine the glass transition temperature, we fit the relaxation time to a Vogel-Fulcher-like equation,  $\ln(\tau_\alpha) = a + 1/[K(T_{\text{eff}}/T_0 - 1)]$ , for  $T_{\text{eff}} \leq 0.85$ . We find



**Fig. 10** Relaxation times rescaled by the high-temperature Arrhenius fit  $\tau_\alpha = \tau_0 \exp(E_\infty/T_{\text{eff}})$  for all the persistence times studied in this work. A smooth monotonic evolution of the glassy dynamics with  $\tau_p$  is observed, where more persistent particles display steeper temperature dependence, i.e. they become more fragile. The Brownian results are shown with 'B' symbols.





**Fig. 11** (a) The glass transition temperature  $T_0$  and (b) the fragility parameter  $K$  obtained from Vogel–Fulcher-like fits  $\ln(\tau_\alpha) = a + 1/[K(T_{\text{eff}}/T_0 - 1)]$  as a function of persistence time. Both the glass transition temperature  $T_0$  and the fragility parameter  $K$  monotonically increase with increasing the persistence time.

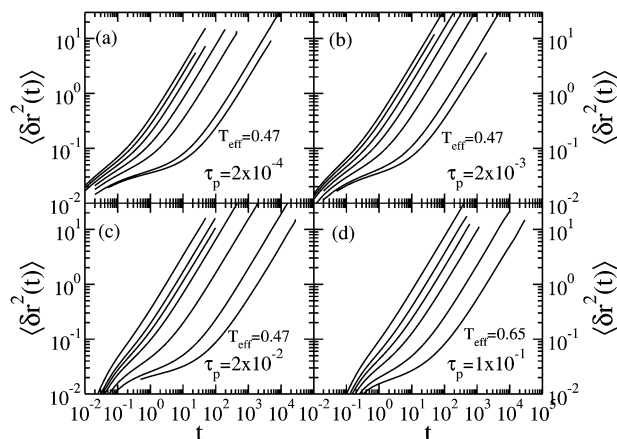
that the glass transition temperature  $T_0$  is approximately constant with increasing  $\tau_p$  until  $\tau_p \geq 2 \times 10^{-2}$ , see Fig. 11(a). Note that this is about the same persistence time in which we observe a slowing down compared to the overdamped Brownian dynamics simulation. For smaller  $\tau_p$  the transition temperature is close to the value known from equilibrium studies of the model. For larger  $\tau_p$  the glass transition temperature  $T_0$  increases up to  $T_0 = 0.51 \pm 0.03$  for  $\tau_p = 1 \times 10^{-1}$ , which again shows that persistent motion promotes glassy dynamics in the present system.

The fragility parameter  $K$  is larger for systems that are more fragile. As suggested by the dependence on the effective temperature of  $\tau_\alpha$  in Fig. 10, the fragility increases with increasing  $\tau_p$ , see Fig. 11(b). Again, this result contrasts strongly with the results in ref. 28.

A deep understanding of the kinetic fragility for equilibrium supercooled liquids is not available,<sup>50</sup> therefore it is difficult to interpret the evolution of  $K$  for the present nonequilibrium situation. The monotonic evolution of  $K$  with  $\tau_p$  again confirms the smooth evolution of glassy dynamics with the degree of departure from equilibrium. A large change in fragility is also consistent with the finding that self-propulsion dramatically changes the structure of the fluid, and the above conclusion that self-propulsion produces a ‘different’ liquid whose glassy dynamics only differs in its details (such as kinetic fragility and glass temperature), as compared to typical equilibrium supercooled liquids.

### E. Mean-squared displacements and Stokes–Einstein relation

We now examine the mean-squared displacement  $\langle \delta r^2(t) \rangle = N^{-1} \left\langle \sum_i |\mathbf{r}_i(t) - \mathbf{r}_i(0)|^2 \right\rangle$ , which is shown in Fig. 12 for various  $\tau_p$  values. At short times we see the ballistic motion that results from the finite persistence time of the self-propulsion. This regime is followed by a crossover to diffusive motion at long



**Fig. 12** The mean-squared displacement  $\langle \delta r^2(t) \rangle$  for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $\tau_p = 2 \times 10^{-3}$ , (c)  $\tau_p = 2 \times 10^{-2}$ , and (d)  $\tau_p = 1 \times 10^{-1}$ . For panels (a–c)  $T_{\text{eff}} = 1.0, 0.9, 0.8, 0.7, 0.6, 0.5, 0.47$  are shown (left to right). In panel (d)  $T_{\text{eff}} = 1.1, 1.0, 0.9, 0.8, 0.7, 0.65$  are shown (left to right).

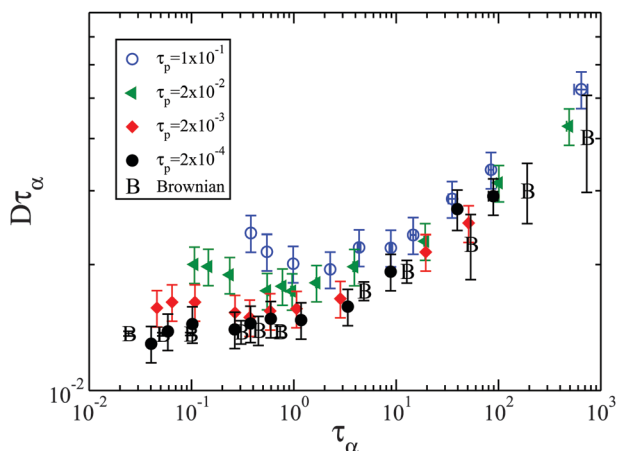
times, which defines the self-diffusion coefficient  $D$  of the model. The data in Fig. 12 reveal that  $D$  decreases dramatically as the temperature is decreased, which is another well-known characteristic signature of the glass transition.<sup>50</sup>

Between the ballistic motion and diffusive motion a plateau emerges, which indicates a strongly sub-diffusive regime whose duration increases rapidly as the temperature decreases. For  $\tau_p = 2 \times 10^{-4}$  this plateau is similar to what is found for Brownian dynamics. For increasing  $\tau_p$  the plateau is less pronounced and appears to occur at smaller values of  $\langle \delta r^2(t) \rangle$  for increasing  $\tau_p$ . To quantify this observation we defined the plateau in  $\langle \delta r^2(t) \rangle$  as the value of  $\langle \delta r^2(t) \rangle$  at the inflection point in the plot of  $\ln(\langle \delta r^2(t) \rangle)$  versus  $\ln(t)$  at  $T_{\text{eff}} = 0.65$ . We found that the plateau was around 0.035 for  $\tau_p = 2 \times 10^{-4}$  and was approximately constant until  $\tau_p = 1 \times 10^{-2}$  where it decreased to 0.0164 at  $\tau_p = 1 \times 10^{-1}$ .

In equilibrium systems at temperatures above the onset of supercooling it is generally found that the Stokes–Einstein relation  $D \sim \tau_\alpha^{-1}$  holds. Therefore,  $D\tau_\alpha$  is approximately constant for high temperature liquids. The Stokes–Einstein relation has been found to be violated in supercooled liquids, resulting in a growth of  $D\tau_\alpha$  below the onset temperature.<sup>51,52</sup> We examined the Stokes–Einstein relation for our model of self-propelled particles. Recall that as  $\tau_p$  goes to zero for a fixed effective temperature, the dynamics becomes identical to over-damped Brownian dynamics.

To examine the Stokes–Einstein relation we first calculated the diffusion coefficient using  $D = \lim_{t \rightarrow \infty} \langle \delta r^2(t) \rangle / (6t)$ . Shown in Fig. 13 is the evolution of  $D\tau_\alpha$  versus  $\tau_\alpha$  for several  $\tau_p$  and effective temperatures. The main observation is that the product  $D\tau_\alpha$  changes by less than one decade for all systems, which indicates that the decrease of the diffusion and the increase of the relaxation time are very strongly correlated.

We do see, however, deviations from the Stokes–Einstein relation. For small values of the persistence times the Stokes–Einstein relation is approximately valid at high effective temperatures, in other words for small  $\tau_\alpha$ . However, for the largest  $\tau_p$  we do not find a clear region of effective temperatures where



**Fig. 13** Examination of the Stokes–Einstein relation,  $D\tau_\alpha \sim \text{const}$ . For  $\tau_p = 2 \times 10^{-4}$  and  $2 \times 10^{-3}$   $D\tau_\alpha$  is approximately constant for small  $\tau_\alpha$ , which corresponds to high  $T_{\text{eff}}$ . However, for long persistence times  $\tau_p$  the Stokes–Einstein relation is not valid for any temperature range. In the glassy regime, a similar degree of Stokes–Einstein decoupling is observed for all  $\tau_p$  values.

the Stokes–Einstein relation is valid. Instead, we find a minimum in the plot of  $D\tau_\alpha$  versus  $\tau_\alpha$  for  $\tau_p = 1 \times 10^{-1}$ . At large  $\tau_\alpha$ , it appears that  $D\tau_\alpha$  for all systems approximately follow the same master-curve. This suggests that despite small differences in the high temperature liquid, the glassy dynamics of both Brownian and strongly self-propelled systems shows a similar degree of Stokes–Einstein decoupling. In particular, the (weak) correlation discussed for equilibrium liquids between kinetic fragility and Stokes–Einstein decoupling<sup>52</sup> is not observed here.

## V. Dynamic heterogeneity

An extensively studied feature of the dynamics of supercooled liquids is the emergence of spatially heterogeneous dynamics.<sup>50–52</sup> In high temperature simple fluids, particle displacements are nearly Gaussian, but in supercooled liquids it has been observed that there are subsets of particles whose displacements are either much greater than (fast particles) or much smaller than (slow particles) what would be expected from a Gaussian distribution of particle displacements. Furthermore, the fast and slow particles form transient clusters whose maximum characteristic size increases with decreasing temperature.

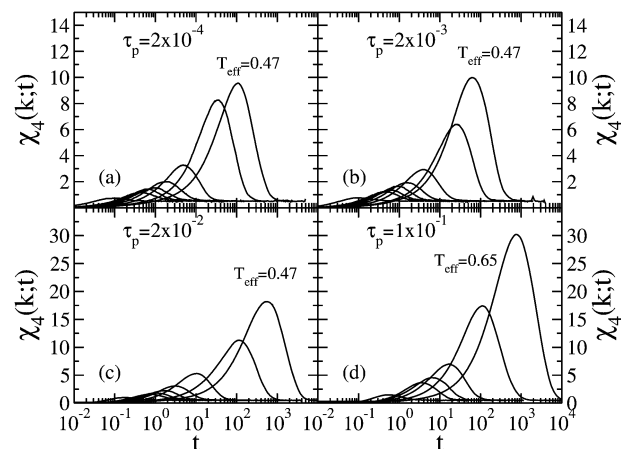
In this section we investigate whether the self-propulsion influences the dynamic heterogeneity. To quantify such dynamic heterogeneity we monitor the fluctuations of the real part of the microscopic self-intermediate scattering function

$$\text{Re}\hat{F}_s^n(k;t) = \cos(\mathbf{k} \cdot (\mathbf{r}_n(t) - \mathbf{r}_n(0))) \quad (9)$$

of the larger A particles. We define the dynamic susceptibility<sup>51,53</sup>

$$\chi_4(k;t) = \frac{1}{N_A} \left[ \left\langle \left( \sum_{n=1}^{N_A} \text{Re}\hat{F}_s^n(k;t) \right)^2 \right\rangle - \left\langle \sum_{n=1}^{N_A} \text{Re}\hat{F}_s^n(k;t) \right\rangle^2 \right], \quad (10)$$

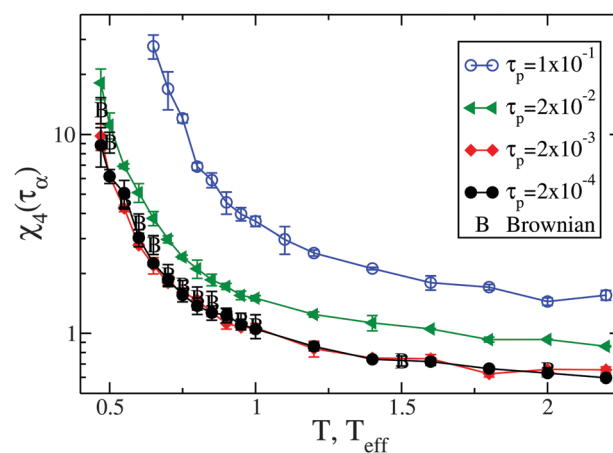
where we fix  $k = 7.2$  as for the self-intermediate scattering function.



**Fig. 14** The dynamic susceptibility  $\chi_4(k;t)$  for (a)  $\tau_p = 2 \times 10^{-4}$ , (b)  $2 \times 10^{-3}$ , (c)  $2 \times 10^{-2}$ , and (d)  $1 \times 10^{-1}$ . For panels (a–c)  $T_{\text{eff}} = 1.0, 0.9, 0.8, 0.7, 0.6, 0.5, 0.47$  are shown. In panel (d)  $T_{\text{eff}} = 1.1, 1.0, 0.9, 0.8, 0.7, 0.65$  are shown. The peak positions are around  $\tau_\alpha$ , and the peak height grows with decreasing  $T_{\text{eff}}$  for a fixed  $\tau_p$ . For a fixed  $T_{\text{eff}}$  the peak height grows with increasing  $\tau_p$ . Notice the change of scale on the y axis for  $\tau_p = 2 \times 10^{-2}$  and  $\tau_p = 1 \times 10^{-1}$ .

Shown in Fig. 14 is the dynamic susceptibility  $\chi_4(k;t)$  for different effective temperatures for the same persistence times as shown in Fig. 5. We find that the susceptibility grows with time, peaks at a time around  $\tau_\alpha$ , and finally decays. With decreasing  $T_{\text{eff}}$  at constant  $\tau_p$ , the dynamic susceptibility increases, in a way qualitatively similar to that in thermal systems.<sup>53</sup> This observation shows that the glassy dynamics observed for self-propelled particles does not appear very distinct from the equilibrium glassy dynamics.

Shown in Fig. 15 is the value of the dynamic susceptibility at the  $\alpha$ -relaxation time,  $\chi_4(\tau_\alpha) \equiv \chi_4(k;t = \tau_\alpha)$ , as a function of the effective temperature,  $T_{\text{eff}}$ , for the same  $\tau_p$  values. The effective temperature dependence of  $\chi_4(\tau_\alpha)$  shows similar trend as  $\tau_\alpha$ . There is a modest change for  $T_{\text{eff}} \geq 1.2$ , but there is a sudden increase in  $\chi_4(\tau_\alpha)$  with decreasing  $T_{\text{eff}}$ , and this increases



**Fig. 15** The increasing value of the four-point susceptibility  $\chi_4(\tau_\alpha)$  at the  $\alpha$ -relaxation time  $\tau_\alpha$  for different  $\tau_p$  mirrors the increase of the relaxation time in Fig. 5.

mirrors the increases in  $\tau_\alpha$  at a fixed effective temperature. This simply reveals that slow dynamics is accompanied by the growth of the extent of spatially heterogeneous dynamics in self-propelled particle systems.

To examine if the increase in  $\chi_4(\tau_\alpha)$  is solely a reflection of the increase in  $\tau_\alpha$ , we replot  $\chi_4(\tau_\alpha)$  versus  $\tau_\alpha$  on a log-log scale in Fig. 16. To build this plot, we also rescaled  $\chi_4(\tau_\alpha)$  and  $\tau_\alpha$  by their high-temperature values, denoted  $\chi_4(\tau_\alpha^s)$  and  $\tau_\alpha^s$ , which we measure for convenience at the highest studied effective temperature,  $T_{\text{eff}} = 2.2$ . This rescaling is a simple way to scale out once again the high-temperature behavior in the liquid, in order to focus our attention on the slowing down and the increase of dynamic heterogeneity resulting from the glassy dynamics itself. Whereas the  $\tau_p$ -dependence of  $\tau_\alpha^s$  was already discussed above, we attribute the increase of  $\chi_4$  with  $\tau_p$  in the liquid to the related increase of the spatial extent of the velocity correlations that was shown in Fig. 4(b). This observation suggests that already in the high effective temperature active liquid the motion is spatially correlated to some extent. Preliminary results suggest that the spatial extent of the high effective temperature dynamic correlations increases with increasing persistence time but a more detailed investigation of this phenomenon is left for a future study.

After rescaling of the high-temperature physics, the dependence of  $\chi_4(\tau_\alpha)$  on  $\tau_\alpha$  changes extremely weakly with the persistence time  $\tau_p$ , see Fig. 16. This observation is consistent with the idea that the glassy dynamics in and out of equilibrium are qualitatively very similar.

In detail,  $\chi_4(\tau_\alpha)$  first increases rather rapidly with increasing  $\tau_\alpha$  in the initial regime of slowing down corresponding to the fitted mode-coupling regime in Fig. 7. For even larger relaxation times, the four-point susceptibility reaches a regime where the growth with  $\tau_\alpha$  is much less pronounced, as observed in thermal systems.<sup>51,54</sup> In the mode-coupling regime, one expects<sup>55</sup>  $\chi_4 \sim (T_{\text{eff}} - T_{\text{eff}}^c)^{-1}$ . By combining this behavior to the power law

description of the relaxation time, one then expects that  $\chi_4 \sim \tau_\alpha^{1/\gamma}$  in the regime approximately described by mode-coupling theory inspired fits. Since the exponent  $\gamma$  in Fig. 8(b) decreases from  $\gamma = 2.4$  to  $\gamma = 1.4$  when increasing  $\tau_p$ , we also add these two extreme cases as guides to the eye in Fig. 16. While not terribly convincing, these power laws appear to account qualitatively correctly for the weak dependence on  $\tau_p$  of the various series of data shown in Fig. 16.

Physically, the steady growth of  $\chi_4$  in Fig. 16 as the relaxation time  $\tau_\alpha$  increases suggests that the relaxation dynamics becomes spatially correlated over a dynamic correlation length that also increases as the dynamics slows down. The growth of dynamic correlations as the temperature decreases should be contrasted with a weak temperature dependence reported in Fig. 4(a) for the velocity correlation length, which seems to indicate that these two quantities are unrelated. Quantifying this statement would require the numerical determination of both spatial correlation lengthscales, a task that we left for a future study.

We finish this section by noting that we examined mobility correlations by studying a dynamic susceptibility that in thermal systems depends explicitly on the simulation ensemble.<sup>51,56</sup> It should also be expected that the four-point susceptibility studied in this work would be different if we allowed for fluctuations of conserved quantities, for example if we allowed for volume fluctuations.<sup>53,57,58</sup> However, unlike for an equilibrium system, it is unclear how to account for fluctuations of conserved quantities in the nonequilibrium active systems. Studies of four-point structure factors for large systems may provide insight into the correlated dynamics of dense self-propelled systems since the large system size would allow one to obtain the full dynamic susceptibility by extrapolating the small  $q$  values of the four-point structure factor.<sup>53</sup> This investigation is beyond the scope of the present work.

## VI. Summary and conclusions

We studied the glassy dynamics of the Kob-Andersen binary mixture of self-propelled particles. The self-propulsion force follows the Ornstein-Uhlenbeck stochastic process, and there is no thermal noise. Due to the lack of thermal noise, the dynamics can be characterized by a persistence time of the random force  $\tau_p$ , and an effective temperature  $T_{\text{eff}}$  that is related to the long time diffusion coefficient of an isolated particle.

We found that the local structure of the active fluid, as seen *via* the pair correlation functions, is significantly enhanced as one increases the persistence time  $\tau_p$ . The enhancement can be examined through the height of the first peak of the pair correlation function of the larger, more abundant A particles,  $g_{AA}(r)$ . This peak height for the largest persistence time studied in this work,  $\tau_p = 1 \times 10^{-1}$ , is larger at  $T_{\text{eff}} = 2.0$ , in the fluid phase, than for the smallest persistence time  $\tau_p = 2 \times 10^{-4}$  at  $T_{\text{eff}} = 0.47$ , in the glassy-dynamics regime. Furthermore, new features in the pair correlation functions, *e.g.* the splitting of the second peak of  $g_{AA}(r)$  into three sub-peaks, are observed in the self-propelled system with large persistence times. These features

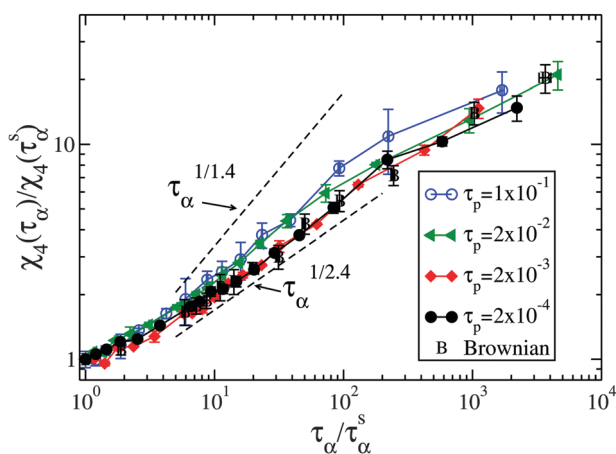


Fig. 16 Evolution of the four-point susceptibility  $\chi_4(\tau_\alpha)$  with  $\tau_\alpha$  for various persistence times and for over-damped Brownian dynamics. Both axes are rescaled by their values at the high effective temperature value  $T_{\text{eff}} = 2.2$ . The remaining dependence on  $\tau_p$  is qualitatively accounted for by the different values of the exponent  $\gamma$  describing the intermediate mode-coupling regime.

suggests that the self-propulsion can favor structures that are not present in the equilibrium system. Therefore, the local structure of the self-propelled particles departs very strongly from that of particles in thermal equilibrium.

More pronounced correlations of self-propelled particles' positions revealed by the pair distribution function are accompanied by correlations of the velocities of the particles. These correlations are absent in equilibrium and they increase significantly with increasing persistence time. The velocity correlations influence the short-time dynamics of the active system. Within our mode-coupling-like theory,<sup>25,26</sup> they also influence the long-time glassy dynamics.

Importantly, we do not see a significant change in the structure with decreasing the effective temperature at a fixed persistence time. Therefore, it makes sense to study the glassy dynamics of systems with fixed persistence time and decreasing effective temperature. When the persistence time goes to zero the system behaves as an overdamped Brownian system at a temperature equal to the effective temperature. We find that for a small increase of the persistence time, the dynamics initially speeds up and then it slows down for larger persistence times at every effective temperature. However, if we fit the high effective temperature results to an Arrhenius law and examine the slowdown as deviations from this high temperature Arrhenius behavior, we find that the slowdown of the dynamics increases monotonically as the persistence time increases. Additionally, we find that fits to a Vogel–Fulcher form of the relaxation time *versus* the effective temperature also lead to a monotonic increase of both the glass transition temperature and the fragility parameter with increasing persistence time. Fits to a mode-coupling theory power law also result in a monotonic increase of the mode-coupling transition temperature.

Overall, these findings suggest that once the (non-trivial and far-from-equilibrium) physics of the high-temperature fluid is scaled out, the resulting glassy dynamics evolves rather smoothly with an increase of the persistence time and does not show any striking qualitative feature that is not also typically observed in standard models for equilibrium supercooled liquids. A remarkable conclusion is that for the present model, the increase of the persistence time promotes glassy behavior, rather than suppresses it. These results, therefore, show that the idea<sup>28,33</sup> that self-propulsion can be universally interpreted as a driving force acting against, and potentially suppressing, the equilibrium glassy dynamics is not correct. We conclude instead that the interplay between self-propulsion and glassy dynamics is much more subtle<sup>21,25,26</sup> and the outcome cannot easily be predicted on phenomenological grounds. Whereas for hard particles, the glass transition is depressed from its equilibrium limit,<sup>22,23</sup> the opposite behavior is found for the present Lennard-Jones model. Capturing these features in an accurate microscopic theory represents an important challenge for future work.

We also found that the Stokes–Einstein relation  $D \sim \tau_\alpha^{-1}$  was valid for small persistence times at effective temperatures above the onset of supercooling. However, for the largest persistence times studied in this work, the Stokes–Einstein relation is never valid. At higher effective temperatures  $D\tau_\alpha$  decreases,

reaches a minimum, then begins to increase for effective temperatures corresponding to the supercooled liquid. However, in the glassy regime corresponding to large relaxation times it appears that  $D\tau_\alpha$  follows a master-curve, which provides additional support to the idea that the glassy dynamics of the supercooled self-propelled liquid shares similar characteristics to the dynamics of equilibrium supercooled liquids.

We finished the study of glassy dynamics by examining correlations in the relaxation of the individual particles, *i.e.* dynamic heterogeneity. To this end we analyzed a dynamic susceptibility  $\chi_A(k;t)$ , which is a measure of the fluctuations of the self-intermediate scattering function. Since our system is at constant density and concentration, our dynamic susceptibility should be considered a lower bound to the dynamic susceptibility that allows for all the fluctuations in the system.<sup>53,56–58</sup> It is unclear how other fluctuations would contribute to the dynamic susceptibility and future work should analyze this issue in more detail. One possibility is to employ large-scale simulations to measure the low wave-vector behavior of a four-point dynamic structure factor. Again, the present work leads us to conclude that when the high temperature regime is scaled out, the relation between  $\chi_A(\tau_\alpha)$  and  $\tau_\alpha$  appears rather insensitive to the persistence time.

In conclusion, the detailed numerical results presented here for the nonequilibrium glassy dynamics of self-propelled particles suggest that the structure and dynamics of a ‘simple’ (*i.e.* non-glassy) dense active fluid change profoundly with increasing departure from thermal equilibrium, quantified by the persistence time of the self-propulsion. With increasing persistence time correlations of the particles' positions become more pronounced and spatial correlations of the velocities develop. The dynamics initially speed up but at longer persistence time they slow down and dynamic correlations develop mirroring the correlations of equal time velocities. These changes in the normal fluid structure and dynamics become more pronounced as the effective temperature decreases and the glassy regime is entered. However, while the details of the slowing down depend quantitatively on the persistence time, the overall picture of glassy dynamics is largely similar for different persistence times. In other words, the evolution of the glassy dynamics with decreasing effective temperature in active systems is qualitatively similar to the evolution of the dynamics in thermal glass-forming systems with decreasing the temperature.

This broad conclusion implies that self-propelled particles undergo a glass transition at low effective temperatures or large densities accompanied by a complex time dependence of time correlation functions, locally-caged particle dynamics, and spatially heterogeneous dynamics very much as in thermal equilibrium. Despite the local injection of energy<sup>14,35</sup> and violations of equilibrium fluctuation–dissipation relations<sup>24</sup> the overall phenomenon studied here is best described as a nonequilibrium glass transition.<sup>21</sup> While this conclusion is broadly consistent with the experimental reports of glassy dynamics in active materials, it remains to be understood whether the present model of self-propelled particles and the generic concept of a nonequilibrium glass transition are sufficient to account for

experimental observations. We hope that future experiments using, for instance, self-propelled grains or Janus colloidal particles will clarify this issue.

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