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## Nonequilibrium glassy dynamics of self-propelled hard disks

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We analyse the collective dynamics of self-propelled particles in the large density regime where passive particles undergo a kinetic arrest to an amorphous glassy state. We capture the competition between self-propulsion and crowding effects using a two-dimensional model of self-propelled hard disks, which we study using Monte-Carlo simulations. Although the activity drives the system far from equilibrium, self-propelled particles undergo a kinetic arrest, which we characterize in detail and compare with its equilibrium counterpart. In particular, the critical density for dynamic arrest continuously shifts to larger density with increasing activity, and the relaxation time is surprisingly well described by an algebraic divergence resulting from the emergence of highly collective dynamics. These results show that dense assemblies of active particles undergo a nonequilibrium glass transition which is profoundly affected by self-propulsion mechanisms.

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The equilibrium physics of dense particle systems is usually understood in the framework of statistical mechanics because it stems from the competition between particle interactions and thermal fluctuations [1]. In particular, phase transitions towards crystalline or amorphous structures are routinely observed at equilibrium [2]. This approach is challenged for particle assemblies that are not uniquely driven by thermal fluctuations, but can also pump energy from their environment to self-propell themselves [3, 4]. Active particles are presently the focus of a large interest, fueled by experimental developments allowing the study of both natural living systems (such as bacteria [5] and cells [6]) and synthetic colloidal [7] and granular [8] particles. It is thus important to understand if and how equilibrium phenomena are affected by this novel type of nonequilibrium driving and dissipation mechanisms.

We study the behaviour of self-propelled particles when steric effects compete with self-propulsion [9–11]. Provided crystallization is suppressed (for instance by size polydispersity), simple fluids at thermal equilibrium display at large density a gradual transformation towards an arrested disordered state [12]. While not vet systematically explored, this situation is of experimental interest for several systems of active particles. For instance, the complex mechanical properties of epithelium tissues result from the influence of self-propulsion mechanisms for close-packed cells [6, 13], while dense bacterial colonies are being studied experimentally [14]. Selfpropelled colloidal and granular assemblies can also be compressed to large densities [15]. On the theoretical side, it was recently suggested that active particles, despite being far from equilibrium, could display kinetic arrest with qualitative analogies, but also strong differences, with the equilibrium glass transition [11]. This suggestion, obtained in the framework of mean-field approaches to driven glassy dynamics, is by no means obvious as slow dynamics is usually fully disrupted by driving

forces, such as a shear flow [16, 17]. Therefore, it is important to study whether the competition between particle scale driving forces and glassy dynamics can yield a nonequilibrium phase transition even in a more realistic situation, which is our central goal.

To this end, we seek a minimal model to study the impact of self-propulsion on the dynamics of dense assemblies of self-propelled particles, allowing us to interpolate smoothly between the well-known (but already complex) equilibrium glassy dynamics, and the driven active case. Therefore, by contrast with detailed numerical study of active matter at moderate densities, our model incorporates active motion following the simplest models of active matter, neglecting for instance hydrodynamic interactions, particle anisotropy or aligning interactions. We work in two spatial dimensions, which is experimentally relevant [8, 13] and typically preferred in earlier studies [4, 9, 18]. To capture crowding effects, we use a 50:50 binary mixture of hard disks with diameter ratio  $\sigma_1/\sigma_2 = 1.4$ , which both suppresses crystallization and displays realistic glassy dynamics at equilibrium. The hard sphere model is also convenient because it does not require the introduction of an energy (or a temperature) scale. Instead it is uniquely controlled, at equilibrium, by the packing fraction,  $\varphi = \pi N (\sigma_1^2 + \sigma_2^2)/(2L^2)$  for N particles in a system of linear size L, using periodic boundary conditions. We express lengthscales in units of  $\sigma_1$ .

We use off-lattice Monte-Carlo simulations to study the glassy dynamics of the model [19]. At equilibrium, an elementary move proceeds as follows. At time t, a particle is chosen at random, say particle i, and a small random displacement  $\vec{\delta_i}(t) = \delta_0 \vec{\xi_i}(t)$  is proposed, where  $\delta_0$  sets the typical amplitude of the moves, and  $\vec{\xi_i}(t)$  is a random vector drawn independently at each step from a unit square centered around the origin with a flat distribution. The move is accepted provided it creates no overlap with another hard disk. In equilibrium conditions, it was established that Monte-Carlo simulations meaningfully and efficiently describe the slow dynamics of glass-formers provided the jump length  $\delta_0$  is adjusted by seeking a compromise between a small value (where the method becomes equivalent to Langevin dynamics) and a large value (creating unphysical non-local moves) [20]. We use  $\delta_0 = 0.1\sigma_1$ , so that  $\delta_0$  does not influence the physics, apart from a trivial rescaling of the time. We have explicitly checked that our results are not qualitatively affected by this choice. Timescales are expressed in Monte-Carlo steps, such that one time unit  $\tau_{MC}$  represents N attempted particle moves. The equilibrium dynamics of the hard disk system is thus characterized by a unique control parameter, the packing fraction  $\varphi$ .

Following previous work [9, 21], we introduce a selfpropulsion mechanism using a persistence timescale,  $\tau$ , defined as a finite timescale governing rotational diffusion so that our model falls into the class of 'apolar active particles', characterized in particular by the absence of any alignment rule. Rotational diffusion is easily implemented in the Monte-Carlo algorithm by generating time correlated random displacements. In practice, we initialize  $\vec{\delta_i}(t=0)$  as before,  $\vec{\delta_i}(0) = \delta_0 \vec{\xi_i}(0)$ , but introduce temporal correlations between successive attempted displacements at times t and t':

$$\vec{\delta_i}(t) = \vec{\delta_i}(t') + \delta_1 \vec{\xi_i}(t), \tag{1}$$

constraining  $|\delta_{i,\alpha}(t)| \leq \delta_0$ , and  $\delta_1 \leq \delta_0$ . As in equilibrium, the particle move is only accepted if it creates no overlap between particles, but the random displacement is updated as in Eq. (1) independently of the acceptance condition, thus generating a fixed persistent time  $\tau$  for the orientation. Equation (1) means that particle displacements have the same amplitude as in equilibrium, but now keep a memory of previous displacements over a finite timescale,  $\tau = (\delta_0/\delta_1)^2$  (expressed in Monte-Carlo time units defined above). Equation (1) represents a discrete-time analog of the Langevin dynamics studied in Refs. [9, 21], which is recovered in the limits  $\delta_0, \delta_1 \to 0$ , keeping the persistence time fixed [19]. Self-propulsion is thus uniquely characterized by  $\tau$ , which reduces, in the dilute limit, to the persistence time of a persistent random walk motion. Equivalently, this control parameter  $\tau/\tau_{MC}$  can be seen as an adimensional rotational Péclet number [22]. Because thermal fluctuations only affect rotational degrees of freedom, the translational Péclet number is not a convenient control parameter in our model [21].

While clearly minimal, the model efficiently captures the competition between steric hindrance (controlled by  $\varphi$ ) and self-propulsion (controlled by  $\tau$ ). We performed extensive simulations in the steady state varying ( $\varphi, \tau$ ) over a broad range, typically using  $N = 10^3$  particles. Our longer simulations last  $10^{10}$  steps. The model is presented more extensively and compared to alternative numerical models in Ref. [19], which shows in particular



FIG. 1: Glassy dynamics for (a,c) equilibrium and (b,d) selfpropelled hard disks with persistence time  $\tau = 10^2$ . Time dependence of the mean-squared displacement, Eq. (2), and the self-intermediate scattering function, Eq. (3) for increasing packing fraction. From left to right in (a, c):  $\varphi = 0.607$ , 0.700, 0.754, 0.773, 0.785, 0.790, 0.795, 0.800, 0.802, and 0.803. From left to right in (b, d):  $\varphi = 0.607$ , 0.700, 0.743, 0.781, 0.806, 0.819, 0.823, 0.825, and 0.828. Note the change of vertical scale between (a) and (b). Two-step, glassy dynamics emerge in both cases, suggesting that self-propelled particles undergo a nonequilibrium glass transition.

that the system remains homogeneous at all densities, in contrast with earlier numerical works [23–27]. Here we concentrate on the large density regime, not explored before.

We start our analysis with a brief description of the glassy dynamics observed when  $\varphi$  increases in the absence of self-propulsion,  $\tau = 0$ . In Fig. 1(a) we show the time dependence of the mean-squared displacement,

$$\langle \Delta r^2(t) \rangle = \langle |\vec{r_j}(t) - \vec{r_j}(0)|^2 \rangle, \qquad (2)$$

where  $\vec{r_j}(t)$  denotes the position of particle j at time tand brackets indicate an ensemble average performed in steady state conditions. The average is specialized to large particles, the slowest component of the binary mixture. While particles diffuse rapidly for moderate packing fractions, diffusion slows down dramatically as  $\varphi$  increases. We cannot observe long-time diffusion in the time window explored by the simulation for  $\varphi > 0.803$ , because it is too slow. Another signature of glassy dynamics is the emergence of the intermediate time plateau in Fig. 1(a), indicating that particle dynamics is essentially a 'caged' motion at intermediate times. This twostep dynamics is confirmed in Fig. 1(c) by the time evolution of the self-intermediate scattering function,

$$F_s(q,t) = \langle e^{i\vec{q}\cdot[\vec{r_j}(t)-\vec{r_j}(0)]} \rangle, \tag{3}$$

which quantifies dynamics occurring over a length  $2\pi/|\vec{q}|$ . We perform a circular average over wavevectors corresponding to the typical interparticle distance,  $|\vec{q}| = 6.2$ , corresponding to the first peak of the structure factor.

Turning to self-propelled particles with  $\tau = 10^2$  in Figs. 1(b,d), we find that dynamics again becomes slower as  $\varphi$  increases, with the development of complex time dependences in both time correlators. Clear differences with the equilibrium situation already emerge for moderate densities and short-times, where active particles move ballistically as a direct result of self-propulsion.

At larger density, the plateau in  $F_s(q,t)$  is less pronounced for self-propelled than equilibrium particles. Mean-squared displacements take lower values at shorttimes, showing that cage dynamics is profoundly affected by the particle activity. While a fast erratic exploration of the cage results from thermal noise, persistent motion is impossible within a cage. Instead, we observe that self-propelled particles transiently 'stick' to the neighbor found in the direction of motion for a duration  $\tau$ , until randomization of the direction of motion allows further displacement. As a result, particles can be fully arrested at short times, reducing  $\langle \Delta r^2(t) \rangle$  in this regime. The cage exploration thus occurs over a broader distribution of times, which produces a complex time dependence of  $F_s(q,t)$  and  $\langle \Delta r^2(t) \rangle$  in the plateau regime. Physically, thermal vibrations are suppressed by the persistent motion and occur over a time  $\tau$  that may become decoupled from the microscopic scale. This observation is crucial, because the equilibrium physics of hard spheres is controlled by entropic forces [1], which are then considerably impacted by self-propulsion. Finally, although less mobile at short times, self-propelled particles diffuse much faster at long times. Diffusive motion is for instance still observed for  $\varphi = 0.823$  and  $\tau = 10^2$ , while it is fully arrested at this density at equilibrium. These observations reveal that the nature of the glass transition is dramatically modified for active particles.

We show in Fig. 2 a displacement map for self-propelled particles with  $\tau = 10^2$  and large density  $\varphi = 0.823$ , measured over a time interval corresponding to structural relaxation (see below for a definition). Clearly, the flow of self-propelled particles at large density is spatially correlated over large distances, and thus displays large scale dynamic heterogeneity [28, 29]. Spatially correlated displacements represent a form of emergent collective motion arising from the competition between self-propulsion and steric effects, which differs qualitatively from earlier observations in active particle systems [30]. The analogy between collective motion and dynamic heterogeneity in epithelium tissues was noted [13].

We extract the long-time self-diffusion constant,  $D_s$ , from its definition,  $D_s \equiv \lim_{t\to\infty} \langle \Delta r^2(t) \rangle / (4t)$ , and report in Fig. 3 the density evolution of  $D_s$  for equilibrium and self-propelled disks. These data confirm that in all cases  $D_s$  decreases sharply upon increasing  $\varphi$ , as it varies by nearly 6 orders of magnitude between the simple fluid at  $\varphi \approx 0.6$  to the dense regime near  $\varphi \approx 0.8 - 0.83$ .



FIG. 2: Displacement map for self-propelled disks with  $\varphi = 0.823$  and  $\tau = 10^2$  over a time  $t \approx 1.5 \cdot 10^7$  corresponding to structural relaxation. It shows the emergence of collective motion correlated over large distance in dense assemblies of active particles.



FIG. 3: Density dependence of the diffusion constant for different persistence time. Inset: The critical density  $\varphi_c$  obtained from Eq. (4), increases continuously with  $\tau$ .

Increasing  $\tau$  has two opposite effects, as demonstrated by the non-monotonic evolution of  $D_s$  with  $\tau$  at fixed  $\varphi$ . First, increasing  $\tau$  slows down diffusion as particles need to wait at least a timescale  $\tau$  to see their orientation diffuse significantly. This effect dominates at moderate densities, where  $D_s$  decreases with increasing  $\tau$ , see Fig. 3. However, self-propulsion has a less trivial effect at large  $\varphi$ , where it accelerates the dynamics dramatically. For  $\varphi = 0.8$ ,  $D_s$  increases by 3 orders of magnitude between  $\tau = 0$  (equilibrium) and  $\tau = 10$ . Such an acceleration of the dynamics could result from the complete disappearance of the glass transition (as for shear flow [31]), but the data in Fig. 3 suggest a different scenario. Although dramatically affected, the density dependence of the diffusion constant for  $\tau > 0$  remains very sharp, indicating that diffusion will cease above a density which remains well-defined. In other words, our simple model of self-propelled hard disks displays a nonequilibrium form of dynamic arrest, despite the presence of driving forces with finite amplitude. This finding is fully consistent with the theoretical suggestion in Ref. [11].

We quantify the effect of the self-propulsion on the location of the glass transition by extracting a critical density  $\varphi_c$  using a power law description:

$$D_s \sim (\varphi_c - \varphi)^{\gamma},$$
 (4)

 $D_s$ 

 $10^{\circ}$ 

where the exponent  $\gamma$  and the critical density  $\varphi_c$  might depend on  $\tau$ . Equation (4) is inspired by equilibrium studies of the glass transition [32], and can be derived in the framework of mode-coupling approaches [11]. The evolution with  $\tau$  of the fitted  $\varphi_c$  shown in Fig. 3 shows that it increases continuously, departing from its equilibrium value as soon as a finite persistence time  $\tau > 0$  is introduced. This confirms that the 'reentrant' evolution of the diffusion constant with  $\tau$  results from the competition between a growing  $\varphi_c$  (which accelerates dynamics at constant  $\varphi$ ) and suppressed short-time vibrations (which slows down dynamics). The shift of  $\varphi_c$  with  $\tau$ , although small in absolute value, in fact represents a spectacular effect. With thermal fluctuations, it is not possible to observe structural relaxation for  $\varphi \approx 0.83$ , which is instead observed when  $\tau > 10$ . This implies that by breaking detailed balance and going out of equilibrium, the system discovers dynamical pathways that are essentially closed at equilibrium.

A tentative analogy with equilibrium systems suggests a physical explanation to the observed shift of the glass transition density with activity. Because hard disks cannot cross, self-propulsion then generates an 'effective' attractive force between particles moving towards one another [33]. Equilibrium studies of adhesive hard spheres showed that the glass transition density increases with the strength of the attraction [34, 35], because the equilibrium structure at short lengthscale is modified. Although structural changes occur in our system, it remains to be understood whether a mapping from self-propelled hard spheres to equilibrium adhesive particles is meaningful [33].

The relaxation dynamics in self-propelled hard disks seems however fundamentally distinct from the equilibrium case. In the hard sphere fluid, the onset of dynamic slowdown is described by a mode-coupling regime where Eq.(4) holds, followed by a crossover to another regime controlled by activated relaxation events between low-lying metastable states [12, 37]. Therefore, introducing self-propulsion could affect the relevance of such activated dynamical processes. In Fig. 4 we confirm that the domain of validity of the power law in Eq. (4) increases from 2 to 4 decades between equilibrium and selfpropelled particles with  $\tau > 10$ . This suggests that meanfield, mode-coupling types of approaches might represent



 $10^{-2}$ 

 $\varphi_c - \varphi$ 

FIG. 4: Critical representation of the diffusion constant showing that the range of validity of Eq. (4) increases with  $\tau$  from about 2 to 4 decades of slowing down.

a valuable theoretical starting point to describe the microscopic dynamics of dense assemblies of active particles [10, 11].

In conclusion, we found that self-propelled particles undergo a nonequilibrium form of a glass transition at large density that is distinct from its equilibrium counterpart, and characterized by the emergence of a new form of collective motion directly resulting from the interplay between activity and steric effects.

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