Phase separation in binary mixtures of active and passive particles

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We study binary mixtures of small active and big passive athermal discs interacting via soft repulsive forces on a frictional substrate. Athermal self-propelled particles are known to phase separate into a dense aggregate and a dilute gas-like phase at fairly low packing fractions. Known as motility induced phase separation (MIPS), this phenomenon governs the behaviour of binary mixtures for small to intermediate size ratios of the particle species. The presence of small active particles induces an effective attraction between the big passive particles due to the activity of small particles. For large concentrations of active particles and large asymmetry complete phase separation occurs. The effective interaction between active and passive particles can be attractive or repulsive at short range depending on the size ratio and volume fractions of the particles. Both lead to clustering of passive particles. The cluster size distribution of passive particles decays as a power law with an exponential cutoff. This distribution tends to a power law as the system approaches a phase transition.

1 Introduction

Phase separation in equilibrium binary mixtures is known to occur even when the interactions are purely repulsive. This has been attributed to an attractive depletion force between large particles in the mixture which is due to the unbalanced osmotic pressure exerted on them by the surrounding small particles. Phase separation in these mixtures is an entropy-driven first order transition and occurs for size ratios greater than 5 in binary mixtures of hard spheres.

Active systems or systems consisting of self propelled particles have been a subject of great interest and research in recent years. It was shown recently that athermal self-propelled particles interacting solely via steric repulsion, phase separate into a dense solid-like phase and a dilute gas phase at very low volume fractions, much before the system reaches close packing. This motility induced phase separation (MIPS) is a phenomenon unique to active systems and has been observed in a number of Brownian dynamics simulations of self-propelled particles. It has also been realised experimentally in a system of synthetic colloids. Activity induced phase separation has also been observed in Brownian dynamics simulations of monodisperse mixtures of active and passive particles. Phase segregation of passive advective particles has been observed in a medium of active polar actomyosin filaments

Small active particles have been found to induce effective interactions between large passive colloidal particles, similar to the depletion interaction in equilibrium mixtures. Known as active depletion, the range, strength, and sign of these interactions are crucially dependent on the shape and size of the colloidal particle. Colloidal rods experience a long-ranged predominantly attractive interaction while colloidal disks feel a repulsive force that is short-ranged in nature and grows in strength with the size ratio of the colloids and active particles.

These two phenomena, depletion induced binary phase separation and motility induced phase separation (MIPS), motivate us to choose as our system a “binary mixture” of active and passive discs. Experimentally, passive particles in active systems have been used as tracers to probe and quantify the dynamics of active particles in active suspensions. The motion of micron-sized passive beads suspended in an active bath was first studied by Xiaolun Wu et al. The effective diffusivities of passive particles was found to be larger than their thermal diffusivities. More recently A. E. Patteson et al. have experimentally shown that, unlike classical results, the diffusivities of passive particles in an active bath is non-monotonic in particle size.

In this paper, we study the phase behaviour of athermal mixtures of active and passive particles. All particles in our system interact via soft repulsive forces. Active particles are self driven and have a self-propulsion velocity $v_1$. Their speed $v_1$ remains constant whereas their direction changes randomly over a time scale $\tau = v_1^{-1}$, the inverse of the rotational diffusion constant. Neither species is subjected to random translational noise and is hence athermal. In addition to this, the two species of particles differ in size (or radius). The mixture is analyzed for various size ratios and compositions.

We address the following questions. What is the effective interaction between the big particles in the mixture? Does the binary mixture phase separate as in equilibrium binary mixtures? What is the effective interaction between the passive and active particles? How does MIPS affect this?

The tendency of active particles to undergo motility induced phase separation governs the phase behaviour of our binary mixture. We find an effective attraction between big passive particles, which leads to a true phase separation for large size ratios, volume fraction of active particles. The effective interaction between active and passive particles can be attractive or repulsive at short range depending on the size ratio and volume fractions of the particles. Both lead to clustering of passive particles. For the former, the cluster size decreases with increasing size ratio while for the latter, it increases with increasing size ratio. The cluster size distribution of passive particles decays as a power law with an exponential cutoff. This distribution tends to a power law as the system approaches a phase transition.

This paper is organized as follows: in Sec. we introduce our model of an athermal binary mixture of active and passive par-
The structure and effective interactions in our system are presented in Sec. 3. We present our results on cluster size distributions in Sec. 4 before the final section, Sec. 5, where we discuss our results and conclusions.

2 The model: binary mixture of active and passive discs

Our system consists of a binary mixture of $N_1$ small active particles of radius $\sigma_1$ and $N_2$ big passive particles of radius $\sigma_2$ ($\sigma_2 > \sigma_1$) moving on a two dimensional frictional substrate. Each active particle has a self propulsion speed $v_1$ and its orientation is represented by a unit vector $\hat{v}_i = (\cos \theta_i, \sin \theta_i)$ where $\theta_i$ is the angle that its velocity makes with respect to some reference direction. The motion of active particles is governed by the following Langevin equations:

$$\dot{r}_i = v_1 \hat{v}_i + \mu_1 \sum_{j \neq i} F_{ij},$$

(1)

$$\dot{\theta}_i = \eta \dot{v}_i(t).$$

(2)

Here $\eta \dot{v}_i(t) = 2 \nu r \delta(r - r')$ where $\nu$ is the rotational diffusion constant of active particles $\hat{\sigma}_i$. $\mu_1$ is the mobility and $F_{ij}$ the force acting on each active particle due to interactions with other particles. $\nu^{-1}$ is the timescale over which the orientation of an active particle changes. The equation of motion for passive particles is

$$\dot{r}_i = \mu_2 \sum_{j \neq i} F_{ij},$$

(3)

where $\mu_2$ is the mobility of passive particles and $F_{ij}$ is the force on a passive particle due to interactions with other particles. There is no translational noise in Eqs. (1,3) and hence the particles are athermal. We choose the mobility of each species to be the same i.e., $\mu_1 = \mu_2$.

Particles interact through short ranged soft repulsive forces $F_{ij} = F_{ij} \delta(r_{ij})$ where $F_{ij} = k(\sigma_i + \sigma_j - r_{ij})$ if $r_{ij} < \sigma_i + \sigma_j$ and $F_{ij} = 0$ otherwise; $r_{ij} = |r_i - r_j|$ and $k$ is a constant. $(\mu_2 k)^{-1}$ defines the elastic timescale.

We simulate the system in a square box of size $140 \sigma_1 \times 140 \sigma_2$ with periodic boundary conditions varying the volume fractions $\phi_s = N_s \pi \sigma_s^2 / L^2$ and $\phi_b = N_b \pi \sigma_b^2 / L^2$ of small and big particles respectively. We start with a random homogeneous distribution of small and big particles in the box and with random directions for the velocity of active particles. Equations (1,3) are updated for all particles and one simulation step is counted after a single update for all the particles. The system is defined by the volume fractions $\phi_s$ and $\phi_b$ of the small and big particles respectively, the activity $v_1$ of active particles and the size ratio $(s = \sigma_2 / \sigma_1)$ as defined as the ratio of the radius of a big particle to the radius of a small particle. We scale the activity by $\sigma_1 \mu_1 k$ to make it dimensionless. The scaled activity is denoted by $v_0$. We compute the radial distribution functions (RDFs) between pairs of big particles, and small particles to characterise the structure of the mixture and effective interactions between particles. Pure active systems have been shown to form clusters and phase separate on increasing $\phi_s$ and $v_0$. To estimate the clustering of both particle species we calculate the cluster size distributions (CSD). All data are recorded in the steady state.

3 Structure and effective interactions

To get a quantitative and qualitative picture of inter particle interactions and the structure of the binary mixture, we compute the radial distribution functions (RDF) between pairs of particles. The radial distribution function $g(r)$ is a measure of the probability of finding a particle at $r_2$ given a particle at $r_1$. It is calculated by counting the number of particles within a distance $r$ and $r + dr$ from the reference particle where $r = (r_1 - r_2)$. The pair distribution function is defined as

$$g(r) = \frac{1}{< n >} < \sum_{i \neq 0} \delta(r - r_i + r_0) >$$

(4)

where $< n >$ is the average particle density, $r_i$ and $r_0$ are the positions of the particle $i$ and the reference particle. For an isotropic system $g(r) \rightarrow g(r)$ and is called radial distribution function with $r = |r|^{2|\sigma_i|}$. In two dimensions $< n > g(r)d^2r$ gives the number of particles in $d^2r$.

The structure of the mixture in the steady state and effective interactions are characterised by the radial distribution functions: $g_{bb}(r), g_{ss}(r)$ of pairs of big and pairs of small particles respectively and $g_{sb}(r)$ of pairs of small and big particles. Now we discuss the features of these distribution functions as a function of the various parameters characterising the system -- $\phi_s$, the volume fraction of small active particles, $\phi_b$, the volume fraction of big passive particles and $s$ the size ratio of the particles.

3.1 Variation with concentration of passive particles

We begin by studying the effect of increasing the concentration of passive particles in the binary mixture for fixed size ratio and concentration of small particles. Plotted in Fig. 2 (left column) is the radial distribution function of passive particles for different $\phi_b$ at fixed size ratio $s = 10$, activity $v_0 = 0.125$ and $\phi_s = 0.2$. We notice the following main features in $g_{bb}(r)$: (i) for all $\phi_b$, the first peak in $g_{bb}(r)$ occurs at $r = 2\sigma_2$ corresponding to the diam-

Fig. 1 Schematic diagram of our system of a binary mixture of athermal active and passive particles. Big passive particles are in a bath of small active particles.
between the small active and big passive particles is attractive (see $g_{ab}$ in Fig. 3(a)). Since there exists a fixed concentration of active particles, not all passive particles can be surrounded by active particles as their concentration is increased. They hence cluster and the clustering increases with increasing concentration of big particles.

The effective force between two big particles must also depend on the concentration of small particles and the size ratio of particles. To understand this dependence, we first analyse the behaviour of the mixture at different $\phi_{s}$ keeping $\phi_{b}$, $s$ and the activity fixed.

### 3.2 Variation with concentration of active particles

Plotted in Fig. 4(a-d) is the RDF $g_{bb}(r)$ for different $\phi_{b}$ at fixed $\phi_{s} = 0.2$ and activity $v_{0} = 0.125$. For small $\phi_{b} < 0.2$, the first peak occurs at $2\sigma_{2}$ and a second larger peak occurs at $2(\sigma_{1} + \sigma_{2})$ indicating the presence of a layer of small particles around the big particles. A third much smaller peak is seen at $2\sigma_{2} + 2\sigma_{1}$ indicating a second layer of active particles around some of the big passive particles. $g_{bb}(r)$ decays to 1 at large distances. The tendency of active particles to aggregate in regions of low motility (motility induced aggregation) leads to their increased presence around the passive particles. But as we increase $\phi_{b}$, the peak at $2(\sigma_{2} + \sigma_{1})$ diminishes and $g_{bb}(r)$ starts to show signatures of a peak at $4\sigma_{2}$. These are indicative of the beginning of clustering of big particles and the presence of fewer isolated passive particles. The concentration of active particles is now sufficient to lead to a significant attractive interaction between the passive particles and cause clustering. For larger $\phi_{b} = 0.3$, the peak at $2(\sigma_{1} + \sigma_{2})$ disappears and the second peak is located close to but less than

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**Fig. 2** $g_{bb}(r)$ for different $\phi_{b}$ at fixed $\phi_{s} = 0.2$ for size ratio $s = 10$ and activity $v_{0} = 0.125$. Corresponding snapshots are shown in right column.

**Fig. 3** $g_{bb}(r)$ for different (a) $\phi_{b}$ at fixed $\phi_{s} = 0.2$ for size ratio $s = 10$ and (b) for different $s$ for fixed $\phi_{b} = 0.4$, $\phi_{s} = 0.3$, activity $v_{0} = 0.125$. 
shows the formation of dense clusters of passive particles and the absence of isolated ones. At very large $\phi_s$, the second peak is very clearly located at $2\sqrt{3}\sigma_2$, indicating hexagonal close packed clusters of passive particles which is also clear from the corresponding snapshot (see Fig. 3(d)). Also $g_{bb}(r)$ decreases below 1 for large $r$. This is an indication of complete phase separation.

Shown in Fig. 5(a), are the RDFs between pairs of active particles $g_{sb}(r)$ for different $\phi_s$. The first peak occurs at $2\sigma_1$ and second peak at $4\sigma_1$. The second peak has a structure. A small peak appears at $r = 2\sqrt{3}\sigma_1$. This small peak is more prominent for large $\phi_s$ and indicates hexagonal closed packed (HCP) structures in the cluster of small particles. Broadening of the peaks indicates squeezing of small particles for large $\phi_s$. Fig. 5(b) shows the corresponding $g_{sb}(r)$. As the concentration $\phi_s$ increases the attractive interaction at short range between active and passive particles diminishes. At $\phi_s = 0.5$, it is completely absent and the interaction at ($\sigma_1 + \sigma_2$) is repulsive.

### 3.3 Variation with size of passive particles

The force on a passive particle depends crucially on the size ratio of particles in our system. We have chosen the mobility of a passive particle to be a constant independent of its size. The momentum imparted to a passive particle depends on its size because of the number of active particles that can strike it over a short duration of time. To understand better the role of size on the effective interaction between passive particles in our system, we vary the size of the passive particle keeping all other parameters fixed in our system. The pair distribution functions $g_{bb}(r)$ for different size ratios $s = 4, 6, 8$ and $10$ are plotted in Fig. 5(left column) for fixed $\phi_b = 0.3$ and $\phi_s = 0.4$ and activity $v_0 = 0.125$ of small particles. The size of passive particles is changed keeping the size of active particles fixed. For small $s \leq 4$, we find pairs and groups of 3 passive particles clustered together in the surrounding medium of active particles. The first peak in $g_{bb}(r)$ at $r = 2\sigma_2$ is the dominant peak and subsequent peaks (decreasing in magnitude) are observed at a separation of $2\sigma_1$ from this one. These are indicative of layers of small particles around some of the passive particles. For slightly larger $s$, the passive particles are isolated (from each other) and surrounded by layers of active particles. The peak at $r = 2(\sigma_2 + \sigma_1)$ is now the dominant one indicating that a layer of active particles around a passive particle is the most favored configuration (see Fig 5b)). Higher order peaks are observed at a separation of $2\sigma_1$, as for smaller $s$, but they are more pronounced here $30$. In both the cases, the dynamics and structure of passive particles is dictated by the active particles and predominantly by their tendency to aggregate. Here again, passive particles act as regions of low mobility and facilitate motility induced aggregation of active particles. As the asym-
metric in size increases, the second peak appears close to \( r = 4\sigma_2 \). This is indicative of clustering of passive particles. For large size ratios \( s \geq 7 \), the effective attractive interaction between passive particles induced by the active particles is significant. This is so for large size ratios because the number of active particles interacting and transferring momentum to a passive particle in a short duration of time is larger for a larger passive particle. The peaks at intermediate distances, seen at smaller \( s \), disappear implying that there are almost no isolated passive particles. For size ratio \( s = 10 \), a strong clustering of passive particles leads to complete phase separation. The first peak occurs at \( 2\sigma_2 \), the diameter of the passive particle, and the second peak appears at \( 2\sqrt{3}\sigma_2 \). This indicates that the particles are hexagonally close packed inside the cluster. The corresponding snapshot in Fig.6(d) also reveals the same. \( g_{bb}(r) \) tends to a value < 1.0 at large \( r \) indicating complete phase separation. Fig.5 (right column) shows snapshots for different size ratios at fixed \( \phi_b = 0.3 \) and \( \phi_r = 0.4 \) and activity \( v_0 = 0.125 \) of small particles.

The structure of our binary mixture can be largely understood from the tendency of active particles to phase separate into a dense solid-like phase and a dilute gas-like phase. This phenomenon, known as motility induced phase separation, results from the growth of a small fluctuation in the local density of active particles. \( \sigma^2 \). Particles in this dense region are slower than those elsewhere because of their increased density (or enhanced crowding). Particles in the vicinity of this region slow down as they approach it, again because of crowding, and become a part of it on slowing down sufficiently. This positive feedback causes the fluctuation to grow and result in a dense macroscopic aggregate of active particles. Particles far away from this region remain in a dilute phase so the result is a phase separated system. In our binary mixture, the dynamics of passive particles comes largely from their interactions with active particles. For a large range of parameters, passive particles act as regions of low motility and seed MIPS, just as a small region of increased density does in pure active systems. Active particles in the mixture are hence attracted to passive particles and accumulate around them. This effective attractive interaction can be deduced from the \( g_{bb} \) plotted in Fig.5(b). The majority of passive particles are hence embedded inside aggregates of active particles. Depending on the concentration of active particles and size ratio, single or small clusters of passive particles are formed. The cluster size decreases on increasing the size of passive particles (for a fixed concentration of active particles) so that the (active-passive particle) interface length is more or less constant. \( g_{bb} \) has peaks at \( 2(\sigma_1 + \sigma_2) \) and successively at a separation of \( 2\sigma_1 \). An effective attractive interaction between passive particles exists.

For large \( \phi_r \) and sufficiently large size ratio \( s \), the passive particles are faster as they receive more momentum from the active particles (see Fig.7). This is due to the increased number of collisions with active particles for larger passive particles. Passive particles no longer seed MIPS as they are not slow and in fact, their effective interaction with active particles is now repulsive at short range. This is seen in the plots for \( g_{bb} \) in Fig.3(b). In this parameter range, the passive particles phase separate or form large clusters to stay away from active particles. This kind of behaviour can be seen in Fig.6(d), 8(d).

## 4 Cluster size distributions

We calculate the cluster size distributions (CSD) of both active and passive particles for different volume fractions and size ratios corresponding to the parameters in Figs.8. Plots in the left column of Fig.8 are CSDs of passive particles while those in the right column are CSDs for active particles in the binary mixture. We begin by analysing the effect of varying the volume fraction of passive particles, \( \phi_b \) in the mixture. Fig.8(a) shows the CSD of passive particles \( p_b(n) \) for different \( \phi_b \) at fixed activity \( v_0 = 0.125 \), size ratio \( s = 10 \) and \( \phi_r = 0.2 \). For small volume fractions of passive particles, \( \phi_b < 0.3 \), \( p_b(n)/p_b(1) \) has the exponential form \( \exp(-n/n_0) \) as observed in the thermal case. There is no clustering at these volume fractions. For intermediate volume fractions \( 0.3 < \phi_b < 0.5 \), \( p_b(n)/p_b(1) \) shows a power law decay with exponential cutoff at large \( n \), i.e., \( p_b(n)/p_b(1) \approx \alpha \exp(-n/n_0) \) for \( \phi_b = 0.3 \) and 0.4 . We find \( (\alpha, n_0) = (0.5, 2) \) and \( (0.65, 4.2) \) respectively. As
we further increase $\phi_0 > 0.4$, the distribution is predominantly a power law with a sudden decay for large $n$. Here the average cluster size is very large (of the order of the system size).

Correspondingly, the CSDs of active particles fits to the form $p_a(n)/p_a(1) \simeq \frac{1}{n^\alpha} \exp(-n/n_0)$, a power law at small $n$ followed by an exponential decay at large $n$, for volume fractions $\phi_0 \leq 0.3$. For larger $\phi_0$, the distribution is predominantly a power law. Shown in Fig. 8(b) is a power law fit $1/n^{1.49}$ for $\phi_0 = 0.4$.

Next we study the effect of the concentration of active particles on the clustering of big passive particles. In Fig. 8(c) we plot the $p_b(n)/p_b(1)$ for fixed $\phi_0 = 0.2$ and for different $\phi_0$ (values indicated in the plot). For very small $\phi_0 \lesssim 0.2$, $p_b(n)/p_b(1)$ decays exponentially with $n$, which represents the homogeneous state or no clustering. For larger $\phi_0 = 0.3$, $p_b(n)/p_b(1) \simeq \frac{1}{n^\alpha}$, with power $\alpha = 1.47$. At very large $n$, the distribution drops suddenly. The CSDs for larger $\phi_0$ cannot be fitted to any of these forms.

The corresponding CSDs for active particles are plotted in Fig. 8(d). For small $\phi_0$, the distribution is a power law for small $n$ followed by an exponential drop at large $n$, $p_a(n)/p_a(1) \simeq \frac{1}{n^\alpha} \exp(-n/n_0)$ as before with the power $\alpha = 1.45, 1.47$ respectively for $\phi_0 = 0.1, 0.2$. For larger volume fractions, the CSDs very nearly obey a power law but with peaks at large $n$ indicating the formation of large aggregates. This indicates a phase separated state. The distribution shifts to the left on increasing $\phi_0$ while the height of the peak at large $n$ increases. This implies that small clusters become less probable as the formation of one large aggregate is favoured more and more as $\phi_0$ increases. Such a distribution agrees well with the predictions of [2].

Fig. 8(e) depicts the $p_b(n)/p_b(1)$ for fixed $\phi_0 = 0.4$, $\phi_0 = 0.3$ for different sizes of passive particles, keeping the size of active particles fixed at $\sigma_1 = 0.1$. As the size ratio increases from $s = 4$ to $s = 6$, clustering of passive particles decreases. The mean cluster size drops from 4 at $s = 4$ to 2 at $s = 6$. As before the distributions fit well to a power law with an exponential cut off, with the power $\approx 1.5$. For $s = 8, 10$, the CSDs do not fit to any known functional forms, however it is clear from the distributions that the clustering increases with increasing $s$. The absence of a functional form could be due to the fact that the number of passive particles in our system is small and at these parameter values the system is phase separated. The CSDs for active particles for the same parameters, shown in Fig. 8(f), nearly obey a power law but with peaks at large $n$ indicating that they are phase separated. This is also clear from the snapshots in Fig. 4 where dense aggregates of small particles can be seen.

We summarise our results on the CSDs of active and passive particles. At very low concentrations or volume fractions, the CSD resembles that at equilibrium and has an exponential form $\exp(-n/n_0)$. At intermediate concentrations, the distributions are well described by a power law with an exponential cutoff $n_0$, $\frac{1}{n^\alpha} \exp(-n/n_0)$. As the system approaches the transition to phase separate, the CSD is very nearly a power law $1/n^\alpha$ but with a sudden drop at large cluster sizes. The power $\alpha \to 1.5$ as the phase
transition is approached. In the phase separated state, the CSD is very nearly a power law but with a peak at large $n$ indicating the formation of large aggregates. Small clusters become less probable and the formation of one large aggregate is favoured more and more as moves deeper into the phase separated regime.

5 Conclusions

We have studied the dynamics of binary mixtures of small active and big passive athermal particles, interacting via soft repulsive forces. The motility of small active particles induces an effective attraction between big passive particles. This attraction leads to a true phase separation for large size ratios, volume fraction of active particles. For small volume fractions of the active particles and size ratios, passive particles being slower seed MIPS. The effective interaction between active and passive particles can be attractive or repulsive at short range depending on the size ratio and volume fractions of the particles. Both lead to clustering of passive particles. For the former, the cluster size decreases with increasing size ratio while for the latter, it increases with increasing size ratio. At very low concentrations or volume fractions, the CSD of passive particles resembles that at equilibrium and has an exponential form $\exp(-n/n_0)$. The cluster size distribution of passive particles decays as a power law with an exponential cutoff. This distribution tends to a power law as the system approaches a phase transition.

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