Soft Matter

PAPER

Cite this: *Soft Matter*, 2017, **13**, 7617

Received 1st May 2017, Accepted 27th September 2017

DOI: 10.1039/c7sm00865a

rsc.li/soft-matter-journal

Cooperative behavior of biased probes in crowded interacting systems†

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We study, *via* extensive numerical simulations, dynamics of a crowded mixture of mutually interacting (with a short-range repulsive potential) colloidal particles immersed in a suspending solvent, acting as a heat bath. The mixture consists of a majority component – neutrally buoyant colloids subject to internal stimuli only, and a minority component – biased probes (BPs) also subject to a constant force. In such a system each of the BPs alters the distribution of the colloidal particles in its vicinity, driving their spatial distribution out of equilibrium. This induces effective long-range interactions and multi-tag correlations between the BPs, mediated by an out-of-equilibrium majority component, and prompts the BPs to move collectively assembling in clusters. We analyse the size-distribution of the self-assembling clusters in the steady-state, their specific force–velocity relations and also properties of the effective interactions emerging between the BPs.

1 Introduction

Situations in which a microscopic probe is pulled by an external force through a quiescent medium composed of other microscopic particles are rather common.[1–11] For a theorist, this is a natural framework for testing the Einstein–Stokes relation, fluctuation–dissipation theorems and force–velocity relations in the linear response regime and beyond.[1–4] Viewed from a different perspective, in such a model system one faces a non-trivial non-linear velocity-selection problem: here, the propagation velocity of the probe in the steady-state results from an intricate interplay between the jamming produced ongoingly by the probe and the diffusive smoothening of the inhomogeneity created in the spatial distribution of the particles. In some cases, the resulting velocity even can be a non-monotonic function of the force such that the differential mobility can become negative.[5–7] In experiments, these settings appear in the so-called constant force active micro-rheology (CF AMR), when a magnetic probe is manipulated externally and is moved through a medium by a magnetic field.[12] This experimental technique has been successfully applied for the analysis of the micro-rheological properties of systems in which either the materials cannot be produced in bulk quantities, or local rheological properties are strongly inhomogeneous, as it happens, *e.g.*, living cells,[13] glasses[14,15] or granular media.[16,17] Different aspects of AMR have been extensively reviewed.[18,19]

Systems often studied by CF AMR are mixtures of colloidal particles (CPs).[18–22] Two types of cooperative behaviour emerge in such media: (a) between the biased probe (BP) and the CPs, and (b) between the BPs themselves, if more than one BP is present.

In absence of the BPs, the CPs are subject to the interactions between themselves and to internal stimuli, due to interactions with the solvent molecules. In consequence, the CPs are in thermal equilibrium with the solvent and are homogeneously distributed in space. A single BP alters the environment, driving the entire colloidal suspension out of equilibrium: the CPs spatial distribution attains a steady-state form around a steadily moving BP, characterised by asymmetric density profiles – a dense, traffic jam-like region in front of the BP, which enhances effectively the friction coefficient and makes it dependent on the force, density, diffusion coefficient of the CPs, and a pronounced wake depleted by the CPs which is formed past the BP. These wakes have been studied in detail for lattice gas models,[23–31] in various continuous-space settings[32–37] and also seen in granular media[16] and colloidal experiments.[38–40]

Lattice gas modelling suggests that the density in the traffic jam-like region in front of the BP approaches its unperturbed value as an exponential function of the distance. In contrast, in

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/c7sm00865a

unbounded systems of spatial dimension d, the CPs density variation in the wake of the BP as a function of the distance is described, in the frame of reference moving with the BP, by a power-law dependence with universal exponents[24–29] 3/2 and 2 for $d = 2$ and $d = 3$, respectively. In systems which are bounded in the direction perpendicular to the direction of the force (*e.g.*, in two-dimensional stripes or in three-dimensional capillaries) the power-law decay exists only as an intermediate asymptotic regime, which is then followed by an exponential function of the distance.[30,31] In confined single files, *i.e.*, for $d = 1$, the density of the CPs past the BP does not attain a steady-state form, but the depleted region rather grows in size indefinitely,[23] in proportion to $\sqrt{t}$, t being time.

Appearance of such inhomogeneous density profiles around a steadily moving BP is a manifestation of the entrainment phenomenon (known very well for macroscopic objects) on a microscopic, molecular scale. For $d > 1$, the profiles attain a stationary form meaning that the entrainment is partial, in the sense that upon an encounter with the BP the CPs are temporarily involved in a directed motion and travel alongside the BP for some random time, leaving it afterwards and being replaced by other CPs so that the total amount of the entrained CPs stays constant, on average, in time. In contrast, in single-files in which the mean displacement of the BP grows in proportion to $\sqrt{t}$[23,41,42] and the profiles do not attain a stationary form, entrainment is complete, such that a single BP ultimately involves in a directed motion all the CPs present in the system. This is rather evident for the CPs appearing in front of the BP which form a growing in size traffic jam, and is less evident for the CPs which are behind the BP. Here, the point is that the CPs behind the BP exert a pressure on the right-most CP (closest to the BP) and as soon as the BP moves away leaving a void space behind it, the right-most CP starts to move faster,[43] as $t\ln(t)$, catching up eventually the BP.

An out-of-equilibrium environment may mediate long-ranged mutual interactions between intruders, which are non-reciprocal and violate Newton's third law.[44–51] In the typical settings of CF AMR, when more than one BP is present, the micro-structural changes of the environment induce effective interactions between the BPs so that they start to move collectively, minimising the dissipation and increasing their propagation velocity. Formation of string-like clusters of BPs, which reveals an emerging effective attraction, has been first evidenced in numerical simulations[46] studying dynamics of a small fraction of strongly repulsive BPs in a two-dimensional sea of two types of repulsive CPs. The same effect has been seen in three dimensions.[49] Further on, stochastic pairing of the BPs has been observed in a lattice model with two BPs only[27] and for the relative motion of two BPs in a nearly-critical fluid mixture, due to emerging critical Casimir forces.[52] Recently, for an off-lattice model with a high density of BPs it was observed that the latter propagate by forming steadily moving "lanes".[37] This self-organisation phenomenon resembles (albeit it emerges under different physical conditions) spontaneous lane formation in binary mixtures of oppositely charged colloids,[53–61] dusty complex plasmas,[62] and also for such seemingly unrelated systems as pedestrian counter flows.[63]

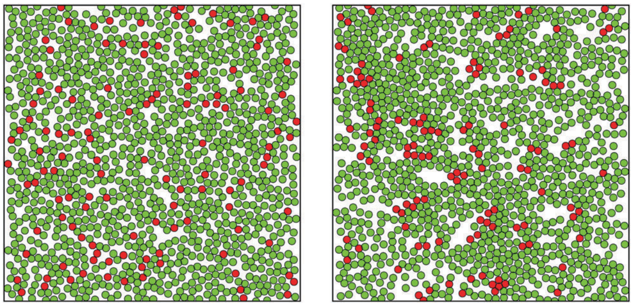

Fig. 1 Two-dimensional box with periodic boundary conditions containing a mixture of interacting particles – the BPs (red circles) and the CPs (green circles). Left: A snapshot of the system when both the BPs and the CPs are subject to the thermal forces only. Right: BPs are subject to a constant force F pointing along the x-axis (see also movie, ESI†).

In this paper we study, *via* extensive numerical simulations, some aspects of dynamical self-organisation in two-dimensional and three-dimensional binary mixtures suspended in an inert solvent (acting as a heat bath held at a temperature T) in typical CF AMR settings. The mixture consists of CPs, which are present at a high density, and BPs, which are present at a low density (see Fig. 1). The situation under study here differs from the previous analysis[46,49] in that the repulsive interactions between the BPs and all the CPs are identical, and also from the case[37] in that the density in our settings is still sufficiently small so that there is not enough of the BPs to build a complete lane spanning the entire system. The CPs are subject to internal stimuli due to interactions with the solvent, and also interact between themselves and with the BPs. The BPs are subject to thermal forces, interact among themselves and with the CPs, and are also exposed to a constant force F oriented in the positive x-direction.

We emphasise that we deliberatively exclude from our modelling the hydrodynamic interactions between the CPs and the BPs, focussing specifically on the interactions between the particles of the driven component which emerge due to a self-produced non-equilibrium environment. This permits us to highlight the effects of the non-reciprocal forces, disentangling them from other possible factors which may affect the dynamical self-organisation phenomenon. At the same time, we note that the hydrodynamic interactions may turn out to be important, especially in three-dimensional systems and for high Péclet numbers.[34,40] In particular, for a related, unbounded system of sedimenting colloids, they are known to produce themselves correlated motions and large-scale dynamic structures.[64] For bounded two- and three-dimensional systems and for small and moderate driving forces studied here, the hydrodynamic interactions may have a less dramatic effect resulting, arguably, in a mere renormalisation of the diffusion coefficient. In any case, of course, the question whether the hydrodynamic interactions will induce qualitative or just quantitative changes is an important issue for further research.

We proceed to show that in the system under study and in absence of the hydrodynamic interactions the BPs spontaneously assemble in "living" string-like clusters, similar to the previously made observations,[46,49] which propagate as one entity. Our aim is to determine, for different values of the applied force, the size-distribution of these clusters in the steady-state, their extensions and propagation velocities, as well as to highlight, for two-dimensional systems a pronounced micro-structuring of the CPs in front of steadily moving BPs. Lastly, also for the two-dimensional case, we quantify the emerging interactions between the BPs, which such a micro-structuring promotes.

The paper is outlined as follows: in Section 2 we introduce basic notations, discuss the computational method and define physical parameters of the system under study. Section 3 presents the results of our simulations. Section 4 concludes our work with a brief recapitulation of our results and some remarks on possible experimental verifications.

2 Physical parameters and computational method

To emphasise collective behaviour of the BPs, we choose purely repulsive interactions between all the particles, described by the Weeks–Chandler–Andersen potential,[65]

$$U(r) = \begin{cases} \varepsilon\left[(\sigma/r)^{12} - 2(\sigma/r)^{6} + 1\right], & r < r_c \\ 0, & r \geq r_c, \end{cases} \qquad (1)$$

which is a shifted Lennard-Jones potential truncated at $r_c = \sigma$, so that $U(r_c) = 0$ and $\mathrm{d}U(r)/\mathrm{d}r|_{r=r_c} = 0$. We identify in what follows σ with the diameter of particles and define the radius of a particle as $R = \sigma/2$. Note, however, that the potential is soft and permits the particles to approach each other to separations less than σ. The possibility of such an approach is controlled by $\varepsilon \geq 0$ which sets the amplitude of repulsion at $r = \sigma$. Such a potential cannot cause any self-organisation of the BPs, but will tend to suppress it.

We suppose that the dynamics are Newtonian subject to random thermal forces generated by the solvent. Then, the equations of motion for the particles trajectories $\mathbf{r}_{i_k}(t)$ obey

$$\begin{aligned} m\ddot{\mathbf{r}}_{i_k}(t) &= -\sum_{j_k \neq i_k} \vec{\nabla} U\left(\mathbf{r}_{i_k}(t) - \mathbf{r}_{j_k}(t)\right) \\ &\quad - \xi\dot{\mathbf{r}}_{i_k}(t) + \vec{\zeta}_{i_k}(t) + \vec{F}\delta_{k,2}, \end{aligned} \qquad (2)$$

where the index $k = 1, 2$ (with 1 corresponding to the CPs and 2 – to the BPs), the index i_k labels the particles of the same species, $i_k = 1,2,\ldots,N_k$, and the sum with the subscript $j_k \neq i_k$ extends over all j_k excluding $j_k = i_k$. Next, m is the mass of a particle (the same for both types of particles), the dot denotes the derivative with respect to time t, and ξ is the friction coefficient related to the viscosity η of the suspending solvent *via* the single-particle Stokes relation $\xi = 6\pi\eta R$.

Further on, $\delta_{k,2}$ is the Kronecker-delta, such that $\delta_{1,2} = 0$ and $\delta_{2,2} = 1$. The force $\vec{F} = (F,0)$ acts on the BPs only, while $\vec{\zeta}_{i_k}(t)$ are random thermal forces which mimic interactions with the solvent. We suppose that the components of $\vec{\zeta}_{i_k}(t) = \left(\zeta^X_{i_k}(t), \zeta^Y_{i_k}(t)\right)$ in two-dimensions and of $\vec{\zeta}_{i_k}(t) = \left(\zeta^X_{i_k}(t), \zeta^Y_{i_k}(t), \zeta^Z_{i_k}(t)\right)$ in three-dimensional systems have a Gaussian distribution with zero mean and a finite variance, and

$$\overline{\zeta^{\alpha}_{i_k}(t)\zeta^{\beta}_{i'_{k'}}(t')} = 2k_\mathrm{B}T\xi\delta(t-t')\delta_{\alpha,\beta}\delta_{k,k'}\delta_{i,i'}, \qquad (3)$$

where the horizontal bar denotes averages over thermal histories, α, $\beta = X$, Y and Z, and k_B is the Boltzmann constant.

The parameters entering eqn (2) and (3) are chosen such that we mimic a physical situation corresponding to having a mixture of two-types of micrometer-sized (*i.e.*, with radius $R \sim 1$ μm and mass $m_c \sim 4 \times 10^{-15}$ kg) colloids in an aqueous solution (viscosity $\eta \sim 10^{-3}$ Pa s) at a room temperature ($k_\mathrm{B}T \sim 4 \times 10^{-21}$ J). This gives the diffusion coefficient D_c of the colloid $D_c \sim 0.2$ μm^2 s^{-1}, so that the corresponding diffusion time $\tau_d = R^2/D_c \sim 5$ s. In what follows, we use a dimensionless time $\tau = t/\tau_d$ and the length scales are measured in units of σ. Accordingly, the force F gets renormalised to give a dimensionless force $f = 0.5RF/(k_\mathrm{B}T) = 0.5\mathrm{Pe}_f$, Pe_f being the Péclet number for CF AMR.[18,19] We note that for such a choice of the parameters and dimensionless variables, the numerical coefficient before the dimensionless inertial term in eqn (2) appears to be very small ($\sim 10^{-6}$), while the numerical coefficient before the first term in the second line in eqn (2) is much larger (~ 10). We nonetheless do not drop the inertial term, since keeping it permits us to resort to the well-justified Velocity Verlet integration algorithm, which provides a simultaneous access to both instantaneous positions and the velocities of the colloids.

Here we concentrate on the response of the system under study to an external dimensionless force f of a small and intermediate amplitude, $0 \leq f \leq 10$, (so that the Péclet number is in the range $0 \leq \mathrm{Pe}_f \leq 20$), focussing solely on the case of an ambient temperature and on the case when the area (for two dimensions) and the volume (for three dimensions) fractions occupied by all the particles is slightly less than 0.6. In two-dimensional systems, our simulations are performed in a 40×40 (in units of σ) box with periodic boundary conditions comprising $N_1 = 1100$ CPs and $N_2 = 122$ BPs. In three dimensions, the simulation box has size $20 \times 20 \times 20$ with $N_1 = 8251$ CPs and $N_2 = 916$ BPs. Therefore, in both cases the area fraction $\phi_2 = \pi N_1/(4 \times 40^2)$ and the volume fraction $\phi_3 = \pi N_1/(6 \times 20^3)$ occupied by the neutrally buoyant component is slightly less than 0.54. In two-dimensions, such an area fraction is significantly below where the system could become hexatic or glassy and should behave like a simple fluid, while for the three-dimensional systems the volume fraction is slightly above the freezing transition point for a mixture of hard-core particles.[66,67] We recall, however, that in our study the potential is soft such that both the area and volume fractions can be effectively less than the mentioned above values. Moreover, any self-ordering of the neutrally buoyant component can be destroyed due to the presence of a biased component.

Further on, after introducing the particles into a box, we let the system equilibrate for a fairly long period of time ($\tau = 2 \times 10^5$) at $f = 0$, when the BPs are identical to the CPs, and then switch on the force $f > 0$ acting on the BPs only. We do not observe any onset of crystallisation in the three-dimensional systems during the equilibration stage. We let then the system evolve at a fixed non-zero f till $\tau = 10^6$, recording periodically all the parameters of interest. Behaviour at different temperatures, different densities of the CPs and the BPs and for larger forces will be reported elsewhere.

3 Results

For two-dimensional systems, typical behaviour is illustrated in a movie (ESI†),‡ showing an evolution of the system under an external force $f = 2$ ($\mathrm{Pe}_f = 4$). In general, we observe that for both two- and three-dimensional systems, the BPs spontaneously agglomerate into clusters – a sort of living "trains", which propagate as one entity leaving a trail of clear solvent behind. This trail extends over quite a few particles diameters and closes completely within a few diffusion times τ_D, due to diffusive mixing and due to overlapping with the condensed regions in front of the trailing BPs. In consequence, we expect that even for relatively small systems considered here the finite-size effects do not matter. We note, as well, that the formation of such clusters is in agreement with the earlier observed behaviour for two-dimensional systems.[46,49]

The trains of the BPs interact with the CPs which sometimes leads to the breakup of the clusters, which can then either recombine again or travel further separately, and also grow in size by accretion of isolated BPs or of smaller trains of BPs drifting towards the bigger ones. This is reminiscent, in a way, of the behaviour of the so-called living polymers,[68] although our system differs from the latter one in two important aspects: (a) all the trains move steadily in the direction of the applied force, and (b) clustering is prompted not by any sort of a chemical bonding, but rather by the forces mediated by the environment, which is brought out-of-equilibrium by the BPs themselves. Similar to living polymers, the size distribution of the trains of the BPs attains a steady-state form as time progresses, which we discuss below. Typical relaxation times are of order of $10^2\tau_D$.

3.1 Cluster-size distributions and spatial extensions of clusters

We define clusters of BPs as a collection of "connected" particles, where any two particles are called connected once the distance between their centres is σ or smaller; we note this definition is somewhat arbitrary. In Fig. 2 we plot, for both two- and three-dimensional systems, the (normalised on the number of the BPs) steady-state size distribution n_s for the number of particles s in a cluster at different values of the force f. At $f = 0$, the cluster size distribution decays exponentially with a steep slope (related to the inverse concentration of the BPs). Upon a gradual increase of f up to $f = 1$ (corresponding to $\mathrm{Pe}_f = 2$), we observe that the distribution gets shifted upwards, attaining an exponential form with the characteristic length which is clearly dependent on the value of f. In this regime, we expect that the mixture of the CPs is still close to equilibrium and the break up of large clusters is dominated by diffusion. For intermediate f, for both two- and three-dimensional systems, there is an initial rapid decay of n_s followed by a slower decay that is again exponential, but with a slower decay rate. In fact, for two-dimensional systems the decay rates for $f = 4$ and $f = 10$ do not differ much, so that we may expect that for $f = 10$ we observe some asymptotic form. For three-dimensional system with $1 \leq f \leq 10$, the variation of the slope is even less pronounced than in two-dimensions.

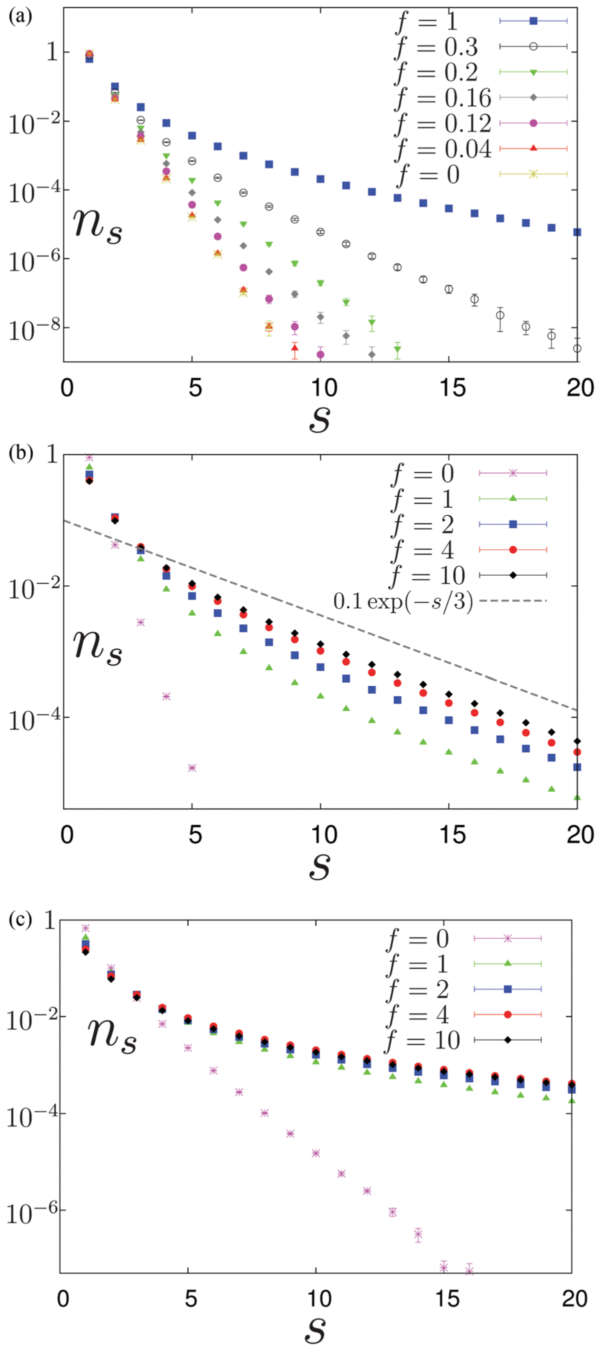

Fig. 2 The cluster-size distribution n_s of the BPs for different values of f. Upper panel: Two-dimensional systems with small forces ($\mathrm{Pe}_f = 0.08$, 0.24, 0.32, 0.4, 0.6 and 2). Middle panel: Two-dimensional systems with intermediate values of f ($\mathrm{Pe}_f = 2$, 4, 8 and 20). Lower panel: Three-dimensional systems with intermediate values of f ($\mathrm{Pe}_f = 2$, 4, 8 and 20).

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‡ The equilibration time in the movie is $\tau = 30$ and is substantially shorter than the analogous time used in numerical simulations.

We focus next on the mean spatial extensions R_x and R_y of the clusters along the x- and y-axes in two-dimensional systems. R_x is given by

$$R_x = \left(\frac{1}{s}\left\langle \sum_{j=1}^{s} \left(x_j - x^c\right)^2 \right\rangle\right)^{1/2}, \qquad (4)$$

where x_j and x^c are instantaneous positions along the x-axis of a given BP within a cluster, and of the centre of mass of this cluster, respectively. A similar definition is used for R_y. In Fig. 3 we present plots of R_x and R_y as functions of s, which show that the mean extensions of the clusters do grow with s but this growth cannot be characterised by a power-law with a unique exponent z – for small clusters z is close to one but for larger ones z is close to 1/2. Interestingly enough, neither R_x nor R_y show any appreciable dependence on f. The behaviour of mean spatial extensions of the clusters in three-dimensional systems is very similar, and we do not present the corresponding plots.

3.2 Clusters' velocities

In Fig. 4, for both two- and three dimensional systems, we plot our numerical results for the steady-state averaged velocity v of a cluster along the x-axis as a function of s for different values of the force f. We observe that for f = 1, 2 and 4 the reduced velocity v/f (the mobility) is a monotonically growing function of s, and apparently plateaus becoming s-independent for larger values of s. This behaviour is consistent with the earlier observations[46,49] (apart from the onset of saturation clearly visible in our simulations) and also the analysis[27] of the dynamics of a pair of BPs in a lattice gas of unbiased hard-core particles. We note that for two-dimensional systems v/f is somewhat larger, but otherwise there is no apparent difference in the behaviour observed for two- and three-dimensional systems. For the largest force (*i.e.*, for $f = 10$) v/f exhibits some non-monotonic behaviour characterised by a dip at $s = 2$ followed then by a monotonic growth with s and an apparent saturation for larger s. In three-dimensional systems such a dip is absent and v/f is a monotonic function of f.

Next, we notice that v/f shows an apparently non-Stokesian behavior: even for clusters comprising just a single BP the reduced velocity, (which has to be an f-independent constant in the Stokesian regime), varies appreciably with f. The reason for such a departure can be two-fold: first, it can be due to effective interactions between different BPs; second, the values of the force f can be already too large to ensure the validity of the linear response relation.[49] We performed simulations with a single BP in a bath of CPs in two dimensions and determined the force–velocity relation for this case. The latter is presented in the lower inset in the upper panel in Fig. 4 and shows that, indeed, v/f starts to deviate from a constant for f below f = 1.

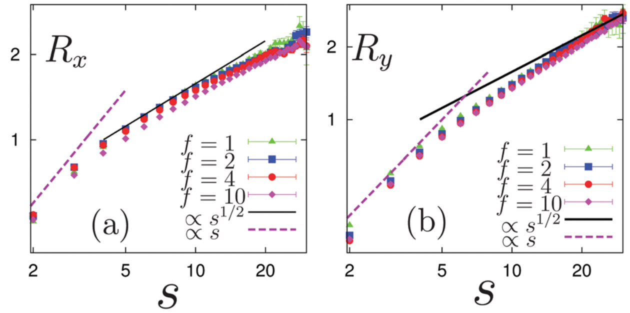

Fig. 3 Two-dimensional systems. Mean spatial extensions R_x (panel (a)) and R_y (panel (b)) of a cluster as functions of s for different values of the force f: f = 1, 2, 4 and 10 (Pe_f = 2, 4, 8 and 20, respectively). Solid line and dashed line indicate the slopes 1/2 and 1, respectively.

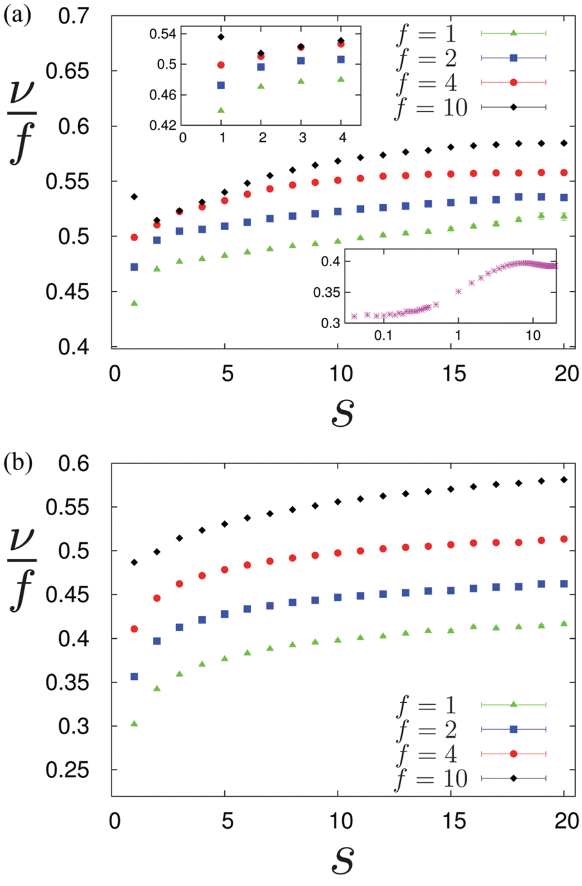

Fig. 4 Reduced velocity v/f of a cluster *versus* s for different values of f: f = 1, 2, 4 and 10 (Pe_f = 2, 4, 8 and 20). Upper panel: Two-dimensional systems. The upper inset: v/f *vs.* s for s = 1, 2, 3 and 4. The lower inset: v/f *vs.* f for the situation with just a single BP moving in a mixture of CPs. Lower panel: Three-dimensional systems.

3.3 Inhomogeneous distribution of the majority component around a pair of steadily moving BPs and effective probe–probe interactions

We finally turn to the analysis of (a) the inhomogeneous distribution of the CPs in presence of two steadily moving BPs, and (b) effective interactions between these two BPs emerging due to such an out-of-equilibrium environment in two-dimensional systems.

To this end, we perform simulations with a pair of BPs, kept in a fixed configuration (characterised by the fixed mutual centre-to-centre distance r, measured in units of σ, and the fixed polar angle ϕ, see Fig. 5) and dragged through a suspension of CPs by a constant force. In simulations, we have a box of size 40 × 40 with periodic boundary conditions and the number of the CPs is $N_1 = 1220$, such that the area fraction occupied by all the particles is still equal to 0.6.

The fixed orientation of two BPs with respect to each other is maintained by compensating the forces exerted on the BPs by the neutrally buoyant component and internal stimuli in the following way: at each moment of time we record the instantaneous position $\mathbf{r}_j(t)$ and the instantaneous velocity $\dot{\mathbf{r}}_j(t)$ of each of the BPs, $j = 1,2$, and calculate numerically the instantaneous value of the force exerted on a given BP by the second BP, all the CPs, the solvent and the external force, (which is defined in the right-hand-side of eqn (2)), such that, explicitly

$$\begin{aligned}\vec{F}_1 = & -\sum_{i_1} \vec{\nabla} U\big(\mathbf{r}_1(t) - \mathbf{r}_{i_1}(t)\big) - \vec{\nabla} U(\mathbf{r}_1(t) - \mathbf{r}_2(t)) \\ & - \xi \dot{\mathbf{r}}_1(t) + \vec{\zeta}_1(t) + \vec{F},\end{aligned} \tag{5}$$

and

$$\begin{aligned}\vec{F}_2 = & -\sum_{i_1} \vec{\nabla} U\big(\mathbf{r}_2(t) - \mathbf{r}_{i_1}(t)\big) - \vec{\nabla} U(\mathbf{r}_2(t) - \mathbf{r}_1(t)) \\ & - \xi \dot{\mathbf{r}}_2(t) + \vec{\zeta}_2(t) + \vec{F}.\end{aligned} \tag{6}$$

We define next the effective force per particle, $\Delta\vec{F} = (\vec{F}_1 + \vec{F}_2)/2$, exerted on the centre of mass of the pair of BPs and also the difference $\delta\vec{F} = (\vec{F}_1 - \vec{F}_2)/2$ of the two forces in eqn (5) and (6). Consequently, we have $\vec{F}_1 = \Delta\vec{F} + \delta\vec{F}$ and $\vec{F}_2 = \Delta\vec{F} - \delta\vec{F}$. Then, in simulations we replace $\vec{F}_1$ and $\vec{F}_2$ by $\vec{F}$, which operation naturally preserves the distance between the two BPs and their mutual orientation. From the formal viewpoint, such a procedure corresponds to "connecting" these two BPs by an inextensible and non-rotatable holonomic bond, which acts with the force $\delta\vec{F}$ on the particle #1 and with the force $-\delta\vec{F}$ on the particle #2 (see Fig. 5).

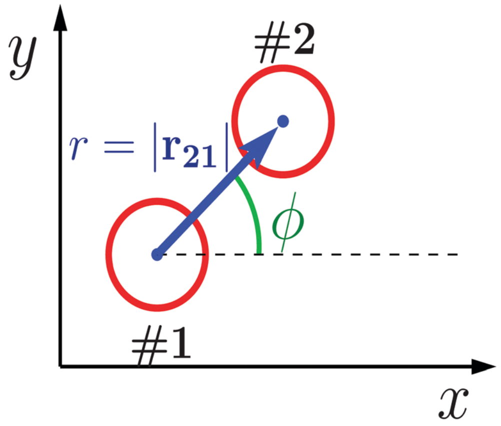

Fig. 5 Configuration of a fixed pair of BPs (#1 is the trailing BP and #2 is the leading one) dragged through a two-dimensional mixture of CPs.

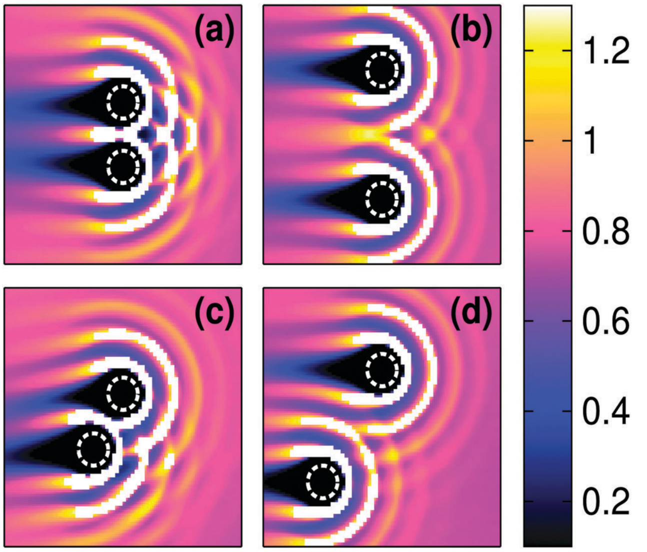

Fig. 6 Distribution of the CPs around a pair of steadily moving BPs (white dashed circles), separated by distance $r = 2$ (panels (a) and (c)) and $r = 4$ (panels (b) and (d)) with polar angles $\phi = \pi/2$ (panels (a) and (b)) and $\phi = \pi/3$ (panels (c) and (d)), for $f = 5$ ($\mathrm{Pe}_f = 10$).

In Fig. 6 we present a density plot of the stationary distribution of the CPs around a pair of steadily moving BPs, separated by distances $r = 2$ and $r = 4$ with the polar angles $\phi = \pi/3$ and $\phi = \pi/2$, for $f = 5$ ($\mathrm{Pe}_f = 10$). The frame of reference is steadily moving with the BP. We observe that the profiles are strongly asymmetric showing that the CPs crowd in front of the BPs and are depleted in the pronounced wakes of the BPs. Qualitatively, they look quite similar to the profiles observed in the lattice gas model,[27] and also in the numerical[44] and experimental[39] analyses of micro-structuring of the flowing CPs in presence of two fixed BPs. We show that also in our settings when the BPs are steadily moving under an external force while the CPs form a quiescent environment, the latter exhibit a very pronounced structuring with a peculiar interference of the high density rims.

Effective interactions between two BPs, mediated by the CPs, are quantified by calculating numerically the projection f_{12} of the force exerted by the leading BP on the trailing one on their line of centres, which is defined as $f_{12} = \delta\vec{F}\cdot\mathbf{r}_{21}$ (see eqn (5) and (6)). In Fig. 7 we plot this force as a function of the mutual separation r for different values of the angle ϕ. First, we notice that this force can be negative (repulsion) or positive (attraction), depending on the mutual orientation of the BPs, defined by the polar angle ϕ and the distance r between their centres. For $\phi = 0$, $\pi/6$, $\pi/4$ and $\pi/3$ the interactions are attractive at the closest approach distance, while for $\phi = \pi/2$ the force f_{12} is very close to zero. For $\phi = 0$, when two BPs are on the same line, the interactions are always attractive and long-ranged. This is quite evident, because here the trailing BP travels in a depleted wake of the leading BP and easily catches it, which translates into an effective attraction. We note parenthetically that a response of the mixture of CPs on such a driving "in tandem" of a pair of a leading and a trailing BPs in a fixed configuration with $\phi = 0$ has been studied experimentally[40] for a variety of values of the separation distance r. The behaviour observed for other polar angles for which f_{12} changes the sign with r, and decays more rapidly. Second, interestingly enough, the amplitude of the force f_{12} appears to depend on the external force f acting on the BPs: it is substantially larger for $f = 10$ that for

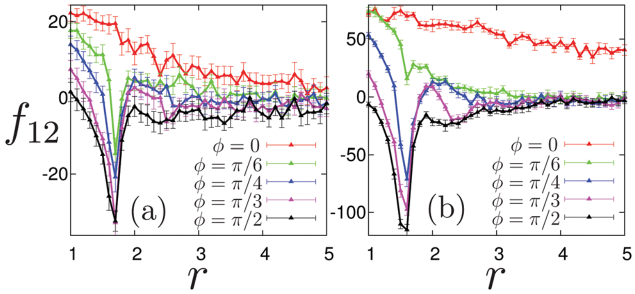

Fig. 7 Force f_{12} exerted by the leading BP (#2 in Fig. 5) on the trailing one (#1 in Fig. 5) in a pair, as a function of the distance r between the centers of the BPs for different polar angles ϕ. (a) $f = 2.5$. (b) $f = 10$. The error bars show how the values of f_{12} vary for different runs. $f_{12} > 0$ corresponds to attractive forces between the pair of BPs. Note that we consider $r \geq 1$ (*i.e.*, the separation distances larger than σ). For $r < 1$ the force f_{12} becomes negative due to the repulsive interactions between all the particles, eqn (1).

$f = 2.5$. In a way, this parallels the observation[39] that the effective force between two BPs, held with an optical trap in a flowing suspension of the CPs, increases with increasing flow velocity. Lastly, we observe that for $\phi = \pi/6$ and $r \approx 1.5$, the sign of f_{12} depends on f – for $f = 2.5$ the two BPs repel each other, while for $f = 10$ the interactions are always attractive.

4 Conclusions

To recap, we studied here the cooperative phenomena emerging in a binary mixture of colloidal particles, with identical repulsive interactions, in typical settings of the constant-force active micro-rheology in two- and three-dimensional systems. The mixture consists of a majority component – neutrally buoyant colloids subject to internal stimuli only, and a minority component – colloidal probes also subject to a constant force. We have shown that in such a system each of the probes alters the distribution of the majority component in its vicinity, resulting in a significant structuring and driving its spatial distribution out of equilibrium. The latter circumstance is responsible for the emergence of effectively attractive interactions between the repulsive probes and prompts them to move collectively assembling in clusters. We analysed the size-distribution of the self-assembling clusters, their specific force–velocity relations and extensions, and also properties of the effective interactions emerging between the probes.

We note finally that a difficulty in experimentally testing this work is that while prior constant force experiments used magnetic particles as the biased probes, magnetic particles are attracted to each other through a magnetic dipole force. A more suitable experimental realisation would be to use non-density matched probes in a system of otherwise neutrally buoyant colloidal component.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

OAV, CMM and GO acknowledge a partial support from the ONRG Grant No. N62909-15-1-C076 and also wish to thank for warm hospitality and financial support the Institute for Mathematical Sciences of the National University of Singapore, where some part of this work has been done. The research of OB was supported by ERC grant FPTOpt-277998. CMM acknowledges the support from the Spanish MICINN grant MTM2015-63914-P. ERW was supported by the National Science Foundation (DMR-1609763).

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