Usage Report for Fiscal Year 2021

Project Title: Transport properties of self-propelled micro-swimmers

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1. Background and purpose of the project, relationship of the project with other projects

Brownian motion in convective laminar flows has been attracting recurrent interdisciplinary attention for its potential applications to transport phenomena at both large and small scales. In particular, advection proved to impact our notion of particle diffusion.

An overdamped passive Brownian particle, say a colloidal particle, moving in a periodic array of counter-rotating convection rolls, undergoes normal diffusion with diffusion constant proportional to the square root of free diffusion constant. At high Péclet numbers, advection enhances the spatial diffusion of the suspended particle by dragging it along the flow boundary layers separating the convection rolls. Quite remarkably, despite such an inhomogeneous convective dynamics, the distribution of a single passive Brownian particle inside a convection array is uniform, with average residence time inside a convection roll inverse proportional to free space diffusivity.

Peculiar is the case of noiseless particles, which get trapped inside a single convection roll, where they retrace some closed orbit, depending on their injection point. Advection favors the confinement of noiseless active particles, as well. An active particle is modeled as a particle propelling itself with tunable constant speed, its direction changing upon collision with obstacles and other particles, or because of fluctuations (either of the suspension fluid or the self-propulsion mechanism). On ignoring its rotational fluctuations, a noiseless active particle ends up trapped in a convection roll, unless its speed is raised above at threshold value which is proportional to the fluid advection speed. The combined effects of rotational and thermal fluctuations on an advected active particle have been investigated in Refs [1-2].

The dynamics of a mixture of Brownian particles is another topic of current interest in applied physics, most notably in colloids and aerosols science. In the context of soft matter, it has been observed that clustering of active particles in a stationary suspension fluid can occur even in the absence of particle attraction, because the particles on the cluster surface point mostly inwards. Under the same conditions, clustering of steric-interacting passive particles is ruled out; the particle distribution would be uniform at all times.

We investigate the clustering of a uniform mixture of finite-size particles suspended in a convective laminar flow. Extensive numerical simulations led us to conclude that (i) a mixture of steric-interacting passive particles tend to cluster at the center of the convection rolls, as a combined effect of advection and pair collisions; (ii) particle-particle collisions also allow convection cell crossings by noiseless particles and, therefore, athermal diffusion in convection arrays and turbulent flows at large; (iii) particle clustering in a convection roll is characterized by a variety of spatial structures, including micellar, ring-like and hexatic patterns, all coherently rotating subject to the vorticity of the suspension fluid.

2. Specific usage status of the system -- This fiscal year we used Hokusai supercomputer mainly for the above-mentioned project and produced some interesting results presented in the next section. However, to complete this project more simulation work is needed. In addition to this item, we used super-computer to investigate the more challenging problems of active particle diffusion in convection roll arrays [5].

3. Theoretical and simulation method

Model - By (linear) convection array we mean here a stationary laminar flow with periodic stream function,

$$\psi(x,y) = \frac{U_0 L}{2\pi}\sin\left(\frac{2\pi x}{L}\right)\sin\left(\frac{2\pi y}{L}\right) \quad - - (1)$$

confined between two parallel edges, y = 0 and y = L/2, which act as dynamical reflecting boundaries. The unit cell of the array consists of two counter-rotating convection rolls [Fig.1(a)].

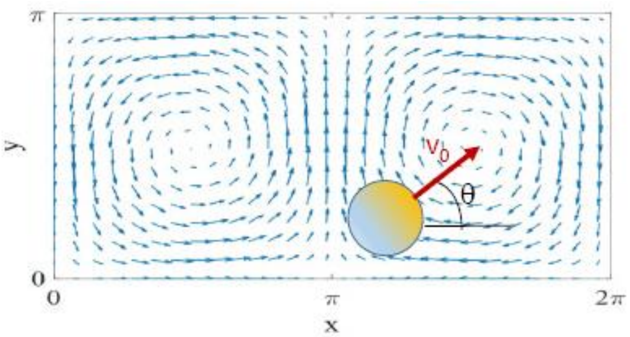

Fig.1: Advection flow field, v_ψ, in a linear convection array with periodic stream function $\psi(x, y)$. The unit cell is delimited by horizontal reflecting walls, y = 0 and y = L/2, and periodic vertical boundaries, x = 0 and x = L, with L = 2π. The self-propulsion velocity $\mathbf{v}_0$ oriented making an angle θ with respect to laboratory x-axis.

We consider a uniform planar mixture of N hard disks per unit cell. We assume that for very short distances they repel each other via an effective potential function modeled by the truncated shifted Lennard-Jones function,

$$\begin{aligned} V_{ij} &= 4\epsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right], \text{ if } r_{ij} \le r_m \\ &= 0 \text{ otherwise}, \end{aligned}$$

with $r_m = 2^{1/6}\sigma$ and σ representing the particle "diameter". The overdamped dynamics of the i-th particle is then formulated by means of two translational and one rotational Langevin equation (LE),

$$\begin{aligned} \dot{\mathbf{r}}_i &= \mathbf{v}_{LJ,i} + \mathbf{v}_{\psi,i} + \mathbf{v}_{0,i} + \sqrt{D_0}\,\boldsymbol{\xi}_i(t) \\ \dot{\theta}_i &= (\alpha/2)\,\nabla\times\mathbf{v}_{\psi,i} + \sqrt{D_\theta}\,\xi_{\theta,i}(t), \end{aligned} \quad - - - - (2)$$

where $\mathbf{r}_i = (x_i, y_i)$, $\mathbf{v}_{\psi,i}$ is the corresponding advection velocity introduced above, and $\mathbf{v}_{LJ,i}$ is the collisional term due to pair repulsion. In the case of an active particle, its self-propulsion vector, $\mathbf{v}_{0,i} = v_0(\cos\theta_i, \sin\theta_i)$, has constant modulus, v_0, and is oriented at an angle θ_i with respect to the array axis.

The translational (thermal) noises in the x- and y-directions, $\xi_{x,i}(t)$, $\xi_{y,i}(t)$ and the rotational noise, $\xi_{\theta,i}(t)$, are stationary, independent, delta-correlated Gaussian noises,

$$\begin{aligned} &\left\langle \xi_i^q(t) \right\rangle = 0 \\ &\left\langle \xi_i^q(t)\xi_j^{q'}(0) \right\rangle = 2\delta_{ij}\delta_{qq'}\delta(t) \end{aligned}$$

Where q or q' = {*x, y, θ*}. D_0 and D_θ are the respective noise strengths, which for generality we assume to be unrelated.

To avoid uncontrolled hydrodynamic effects, the particle is taken to be point like. Other effects due to its actual geometry and chemical-physical characteristics are encoded in the model dynamical parameters. The reciprocal of D_θ coincides with the angular persistence (or correlation) time. The flow shear exerts a torque on the particle proportional to the local fluid vorticity. For simplicity, we adopt second law, which, for an ideal no-stick spherical particle, yields α=1.

Simulation method - We numerically integrated Eqs. (2) using a standard Milstein algorithm to obtain diffusivity, mobility, and probability density function. The numerical integration was performed using a very short time step, 10^{-3} -10^{-4} to ensure numerical stability. At t = 0, the particles were uniformly distributed in cell of convection roll with random orientation. The data points reported in the figures shown here, have been obtained by ensemble averaging over a minimum of 10,000 trajectories. We study how cluster are formed under different conditions. We also studied cluster formation dynamics in both thermal and athermal condition.

Beside that we explore diffusive of sterically interacting particles. First, we calculate mean square displacement then we extracted value of diffusion constant from there. Caution was exerted when computing the asymptotic diffusion constant,

$$D = \lim_{t\to\infty} \frac{\langle [x(t)-x(0)]^2 \rangle}{2t}$$

Under athermal condition or low values of the noise strengths, D_0 and D_θ, the transients of the diffusion process grow exceedingly long. For asymptotically large running times, our estimates of diffusivity and clustering properties are independent of the starting point $(x_i(0),y_i(0))$.

4. Results

Particle clustering -- The clustering of an initially uniform mixture of hard disks advected in a linear convection array is illustrated in Fig. 2 for increasing values of its density. After an appropriately long running time, $t = 10^5$, we observed that the particles tend to drift toward the center of the convection rolls. In the process, they form intriguing structures resembling micellar patterns at low N, and compact clusters at high N. Such clusters consist of a lattice core surrounded by ring-like formations and a dilute gas of strongly advected particles in the vicinity of the cell boundaries. All emerging patterns rotate inside the relevant convection roll subjected to the stream vorticity.

Most remarkably, the disks in the cluster core rotate like a solid with angular frequency of the order of advection vorticity. Therefore, the outer core disks are forced to rotate with tangential velocity larger than the advection velocity corresponding to their distance from the roll's center. This sets an N-dependent upper bound to the size of the cluster core, that is, to the extension of the solid phase. Indeed, additional disks attracted to the cluster rearrange themselves into distinct ring-like files of increasing radius, each rotating with the corresponding advection speed. Such annular patterns emerge only at rather large values of the packing fraction, i.e., $\varphi > 0.2$. The remaining disks in the boundary flow layers are swept away by advection with maximum speed close to U_0; being their local density quite low, the rate of their collisions is not high enough to cause any further significant spatial rearrangement; hence the appearance of the disordered phase displayed in the snapshots of Fig. 2.

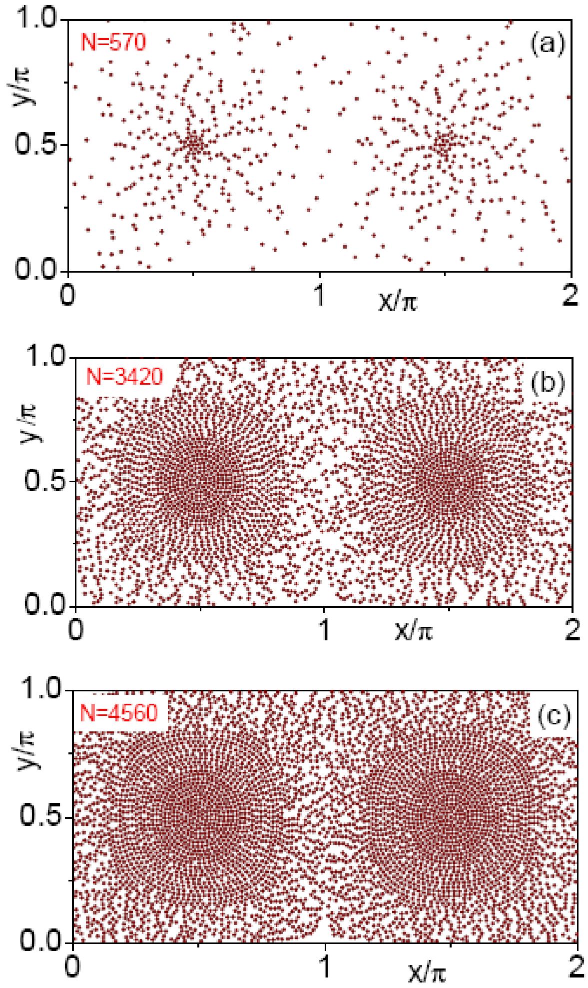

Fig.2: Clustering of a mixture of noiseless passive particles, after a running time of $t = 10^5$. At $t = 0$ the mixture was uniformly distributed with N particles per unit cell of Fig. 1. Other simulation parameters are: $\sigma = 0.05$, $L = 2\pi$ and $U_0 = 1$.

The clustering process of an advected mixture is the result of the inter-particle collisions. Individual noiseless passive particles follow closed orbits with size depending on their injection point. Two such particles tracing parallel orbits separated by less than their diameter, σ, will eventually collide, with the faster outer particle overtaking the slower inner one. As a result, the latter is kicked toward the center of the convection roll, thus initiating a cascaded clustering mechanism. The micellar patterns of Figs. 2(a) are a sort of optical effect due to the free advection of the steric-interacting disks. The spatial ramifications displayed there are no stable structures, but rather time evolving patterns captured by our still snapshots.

Clustering in perturbed mixtures- Now we investigate the robustness of the clustering mechanism under a variety of perturbations, either internal or external.

(a) External bias in clustering - Let us start supposing that all particles move under the action of an external uniform bias represented by the additional term, to be inserted on the r.h.s. of the first LE (2). In Fig. 3 we display the mixture snapshots corresponding to those of Fig. 2, i.e., obtained under the same simulation conditions, except for a transverse bias with g = 0.4. The dynamics of a single particle in a biased convection array has already been studied in detail [3,4]. We know that, as the bias points downwards, a single particle is pushed against the bottom of the array, where opposed horizontal advective flows drag it toward the base of the upward flows – one is centered in the middle to the simulation unit cell, x = L/2, in Fig. 1. In that region the stream velocity, $\mathbf{v}_\psi(x, y)$, is quite low, so that the particle sojourns there long, before being re-injected in the upward boundary flow delimiting the counter rotating convection rolls of the unit cell. The response of a mixture of noiseless passive particles to the combined action of transverse bias and advection is reminiscent of that

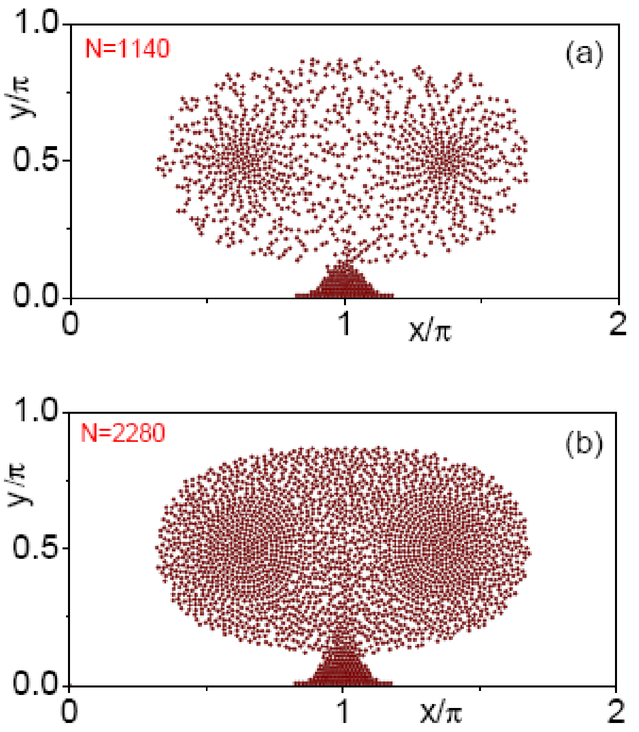

Fig.3: Clustering of a mixture of noiseless passive particles, in the presence of a transverse bias with g = 0.4 after a running time of $t = 10^5$. At t = 0 the mixture was uniformly distributed with N particles per unit cell of Fig. 1. All other simulation parameters are as in Fig. 2.

mechanism. In Fig. 3, the particle pile-up at the base of the upward flow is apparent. However, a substantial fraction of the mixture accumulates in the mid section of the unit cell, along the array axis; hence the resulting tree-like structures displayed there. This means that the particles swept upwards by the ascending flow get momentarily trapped by the convection rolls, where pair collisions favours their clustering. To a closer inspection, the tree canopy consists of two distinct clusters both centered at the same height, y = L/4, but coming closer to one another as N increases. Moreover, such clusters exhibit the same structures as the unbiased case with micellar patterns at low density, and hexatic lattice and ring-like formations at higher densities. At high mixture densities canopy and trunk of the tree structure tend to merge. Most remarkably, the lattice structure of its base becomes unstable, and a melting process sets in, as an effect of the exceeding blocking action exerted by the canopy particles against the upward advective flow. This mechanism was not observed in the unbiased mixtures, no matter what N. Finally, we confirm that, not surprisingly, there is a continuous, though slow, dynamical particle exchange between tree's canopy and trunk.

Impact of thermal noise in clustering - Thermal noise has a destructive effect on the clusters of passive unbiased particles, g = 0. Our simulation results show that the micellar patterns of panel (a) in the noiseless regime with apparently disappears in panel even at relatively low noise levels, i.e., already at high Péclet numbers. This result is consistent with the notion that, under periodic boundary conditions, a single Brownian particle approaches a uniform spatial distribution inside the unit cells of a periodic convection array Hard-core collisions seem not to alter much this property. Vice versa, a biased mixture tends to preserve its tree-like cluster structure. Adding thermal noise simply blurs the patterns previously detected in noiseless clusters. We also notice that the advected particles keep accumulating in the mid section of the simulation unit cell. Accordingly, escape from it is severely suppressed, and so is thermal diffusion along the array.

Effect of self-propulsion in clustering - Fily and Marchetti [5] proved that athermal active particles tend to cluster under the sheer effect of self-propulsion. By varying the mixture density, a rich phenomenology emerges, including homogeneous, separated and frozen phases. Our simulation showed that advection can play a role similar to self-propulsion by inducing particle clustering also in the absence of attractive pair interactions. Now a question rises about the combined effect of advection and self-propulsion. One might guess that self-propulsion works against the collisional clustering of advected passive particles.

The dynamics of an active particle suspended in a linear convection array has been numerically investigated both in the presence and absence of bias. For g = 0, an active particle with $v_0 > 0$ moves symmetrically against the upper and lower array walls; the boundary advection flows tend to drag it toward the base of the upward (downward) flows on the lower (upper) wall. For g > 0, the particle resides mostly at the base of the upward flow, which then re-injects it into either unit-cell convection roll, thus producing a sort of "fountain" effect. Thus, in contrast with previous study [5] advected active particles are not subject to clustering. However, the small, short-lived particle aggregates randomly forming in the counter-rotating convection rolls point to a residual manifestation of clustering by pair collisions.

5. Conclusion

We have investigated the role of pair collisions in the diffusion of a mixture of colloidal particles suspended in a linear convection array. Finite-size particles tend to cluster toward the center of the convection rolls even in the absence of particle attraction; while advection favors clustering, self-propulsion and thermal noise oppose it. In the present paper we focused of the interplay of advection and pair collisions in a quasi-1D geometry to stress the novelty of our results. A number of related issues were left for future work, like (i) the clustering of binary mixtures of active and passive particles; (ii) their diffusion in 2D convection arrays; (iii) the role of hydrodynamical inter-particle interactions at higher densities; and (iv) mixture clustering in more complex convective and turbulent flows. These are all questions of practical interest, which require an extensive simulation effort. We hope that direct experimental observations can help us devise the best numerical approaches to save time and computational resources.

6. Schedule and prospect for the future

In the next fiscal year, we plan to explore the following issues which emerged during the research work of the current fiscal year.

(i) Our simulation results show that the passive particles in convection roll (Fig.1) drift to the center of the counter rotating rolls and form clusters. Preliminary studies show that structure of the cluster depends on packing fraction, may be micellar like or compact clusters. However, detailed studies are required to understand physical properties of the aggregate. To be specific: how does cluster size and its density depend on packing fraction both in the presence and absence of a gravitational force field? Details of collision dynamics and its impact on the time-scale of cluster formation from a uniform initial distribution.

Further, self-propelled particles in the roll preferably accumulated around the center of ascending and descending flows. What will be the stationary structure of an active-passive binary mixture when allowed to settle down starting from a uniform distribution. Here, we are interested in both the mechanism controlling the time-scale of the clustering process and the mixture stationary state.

(ii) We also plan to explore transport properties of active-passive binary mixture in 2D convection arrays. Our preliminary results show that steric-interacting passive particles can cross cells even under athermal conditions. It motivates us to explore in detail the dynamical features of the mobile phase. We expect the transport properties of the binary mixture could be interesting as well potentially important for nano-technological applications.

To address the above-mentioned issues, densities of different phases, radial distribution function, diffusivity and mobility of the mobile phases will be numerically calculated based on the method described in the section 2.

Currently, I have a "Quick Use" user account and we would like to get extension of computation facilities for next usage term (up to 31st March 2023) under the same user category.

7. References

[1] P. K. Ghosh, D. Debnath, Y. Li and F. Marchesoni, Soft Matter, 2021, 17, 2256–2264.

[2] Y. Li, L. Li, F. Marchesoni, D. Debnath and P. K. Ghosh, Phys. Rev. Research, 2020, 2, 013250.

[3] Y. Li, Q. Yin, F. Marchesoni, T. Debnath and P. K. Ghosh, Phys. Rev. E, 2021, 103, L030106.

[4] Y. Li, P. K. Ghosh and F. Marchesoni, Phys. Rev. Research, 2021, 3, L032065.

[5] Y. Fily and M. C. Marchetti, Phys. Rev. Lett., 2012, 108, 235702.

Usage Report for Fiscal Year 2021

Fiscal Year 2021 List of Publications Resulting from the Use of the supercomputer

P. K. Ghosh, F.Marchesoni,Y. Li and F. Nori, Active particle diffusion in convection roll arrays, Physical Chemistry Chemical Physics, volume - 23, issue 20, pages 11944 - 11953, 10th May 2021.

[Paper accepted by a journal]

None

[Conference Proceedings]

None

[Oral presentation]

None

[Poster presentation]

None

[Others (Book, Press release, etc.)]

None