Dynamic clustering and chemotactic collapse of self-phoretic active particles

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Recent experiments with self-phoretic particles at low concentrations show a pronounced dynamic clustering [I. Theurkauff et al., Phys. Rev. Lett. 108, 268303 (2012)]. We model this situation by taking into account the translational and rotational diffusiophoretic motion, which the active particles perform in their self-generated chemical field. Our Brownian dynamics simulations show pronounced dynamic clustering only when these two phoretic contributions give rise to competing attractive and repulsive interactions, respectively. We identify two dynamic clustering states and characterize them by power-law-exponential distributions. In case of mere attraction a chemotactic collapse occurs directly from the gas-like into the collapsed state, which we also predict by mapping our Langevin dynamics on the Keller-Segel model for bacterial chemotaxis.

The collective motion of self-propelling objects is a most fascinating subject which has been studied in a variety of systems [1, 2]. At the macroscale, collective patterns occur, for example, in flocks of birds or fish schools [3–5] while at the microscopic scale bacterial cells in an aqueous environment generate intricate motional patterns [6–8]. To understand basic features of structure formation in non-equilibrium, systems with spherical or circular microswimmers are investigated. A number of theoretical and experimental studies have demonstrated that activity of microswimmers alone can result in clustering and phase separation [9–17] due to reduced motility in dense aggregates [9–15]. However, the colloidal density has to be large enough that the characteristic time for a particle to join a cluster becomes comparable to its rotational diffusion time needed to dissolve from it [18]. Other investigations explore the influence of hydrodynamics on collective motion [18–21].

In experiments with dilute suspensions of self-phoretic active Janus colloids, dynamic clustering has been observed [25, 26]. In this novel non-equilibrium phenomenon, particles constantly join and leave clusters which exhibit a very dynamic shape. Since the colloids consume a chemical, they create a non-uniform chemical field around themselves. The chemical gradients initiate diffusiophoresis [27], whereby colloids can attract each other as demonstrated in [20]. The diffusiophoretic mechanism not only provides a novel colloidal interaction, it also serves as a biomimetic version of bacterial chemotaxis [28], where cells identify and swim along chemical gradients. Autochemotactic cells typically conglomerate in large aggregates or even exhibit a chemotactic collapse [2].

Recent theoretical and experimental studies included short-range attraction between active colloids and observed clustering at low colloidal densities [20, 23–25]. Ref. [26] implements diffusiophoresis for concrete surface properties of self-phoretic colloids and indentifies various states such as clumping and asters.

The work presented here has very much been inspired by the experiments of the Lyon group [25]. The diffusiophoretic interaction has a translational and orientational contribution. Using Brownian dynamics simulations, we demonstrate that pronounced dynamic clustering occurs only when these two contributions give rise to competing attractive and repulsive interactions, respectively. We identify two dynamic clustering states and characterize them. Otherwise, in case of mere attraction a chemotactic collapse occurs directly from the gas-like state before pronounced clusters are able to form. We support this result by mapping our Langevin dynamics on the Keller-Segel model for bacterial chemotaxis.

In our model we consider a dilute suspension of N self-phoretic colloids confined in a quadratic box. For example, by adding the chemical H\textsubscript{2}O\textsubscript{2}, they become active due to self-electrophoresis [37] and the colloids move with a swimming velocity \(v_0\). It depends on the concentration \(c\) of the chemical that is a control parameter in the experiments [38, 39]. We assume that \(v_0\) does not change noticeably due to local inhomogeneities of \(c\) and consider it as a system parameter [25, 40]. In addition, the active colloids experience diffusiophoretic forces along chemical gradients, which are generated by the colloids themselves when they consume the chemical. Since the colloids are bipolar, a torque also aligns their directions \(e\) along a chemical gradient. The associated translational and rotational diffusiophoretic velocities are given by [27]:

\[
v = [(\zeta) 1 - (\zeta(3n \otimes n - 1)/2)] \nabla c \quad (1)
\]

\[
\omega = \frac{9}{4a} \langle n \rangle \times \nabla c. \quad (2)
\]

Here the slip-velocity coefficient \(\zeta\) depends on the interaction of the chemical with the colloid’s surface, \(\langle \ldots \rangle\) is an average over the surface with local normal vector \(n\), and \(a\) is the colloid radius. The quadrupolar term in Eq. (1) vanishes for half-coated colloids, whereas the angular velocity \(\omega\) with \(\langle n \rangle \propto e\) is non-zero since the strength of the chemical-surface interaction is different for the two halves of the Janus colloids. Using \(\dot{e} = \omega \times e\) and \((e_i \times \nabla c) \times e_i = (1 - e_i \otimes e_i) \nabla c\), we obtain a system of two Langevin equations describing the two-dimensional position \(\mathbf{r}_i\) and direction \(e_i\) of the \(i\)th colloid in the over-
Thermal and intrinsic fluctuations enter the equations by translational ($\xi_t$) and rotational ($\mu_t$) noise with zero mean and correlations $\langle \xi_t(0) \xi_t(t) \rangle = 2 D_t \delta(t-t')$ and $\langle \mu_t(0) \mu_t(t) \rangle = 2 D_{rot} \delta(t-t')$. We model the active particles as hard spheres; so, whenever we start to overlap, we separate them along the line connecting their centers. The chemical field diffuses and has sinks at the positions of the particles since they consume the chemical field [11]:

$$c(r) = D_c \nabla^2 c - k \sum_{i=1}^N \delta(r-r_i) ,$$

(5)

In experiments, active colloids settle on a surface and swim in two dimensions while the chemical field diffuses freely in half-space. Implementing zero flux at the surface by image colloids does not change the form of Eq. (5). The same holds when we integrate the concentration over a layer with thickness twice the colloid radius $a$ to make the relevant concentration field two-dimensional.

Within clusters of active colloids, the concentration field cannot freely diffuse. So, whenever a colloid is surrounded by six closely packed neighbours, we introduce for it a screened chemical field, $\exp[-(r-\xi)/\xi/r]$, with the screening length $\xi = 2a(1+\epsilon)$. In our simulations we typically take $\epsilon = 0.3$ and have checked that our results do not change when $\epsilon$ is changed by 50%. We rescale time and length by $t_r = 1/(2D_{rot})$ and $l_r = \sqrt{D_t/D_{rot}} = 2.33a$ respectively. For historical reasons this is above the experimental value of 1.79a [25], while the thermal value is 1.15a. Again, the following results do not change drastically, if we adjust the value. The noise intensities for rescaled $\xi_t$ and $\mu_t$ become one and we are left with three essential parameters: the Peclet number $Pe = v_0/(2\sqrt{D_t D_{rot}})$ and reduced translational as well as rotational diffusiophoretic parameters $\zeta_{tr} k/(8\pi D_t D_c) \to \zeta_{tr}$ and $\zeta_{rot} k/(8\pi D_c \sqrt{D_{tr} D_{rot}}) \to \zeta_{rot}$, respectively, for which we keep the same symbols.

Note that in our model each active colloid creates a chemical sink. So, for $\zeta_{tr} > 0$ and $\zeta_{rot} > 0$ it pulls and rotates other colloids towards itself, which means an effective attraction. At boundaries of the simulation box, we let colloids move with a randomly chosen direction back into the box. Following experiments [25], we investigate dilute systems with an area fraction of $\sigma = 5\%$ and simulate a total of 800 particles. We have checked that a larger number does not change our results as long as the area fraction is maintained. Finally, we choose the swimming velocity $v_0$ in the range $2 - 4.5 \mu m/s$ as in experiments [25] corresponding to Peclet numbers $Pe = 10 - 22$. 

\[ \mathbf{r}_i = v_0 \mathbf{e}_i - \zeta_{tr} \nabla c(\mathbf{r}_i) + \xi_t, \]
\[ \mathbf{e}_i = -\zeta_{tr}(1 - \mathbf{e}_i \otimes \mathbf{e}_i) \nabla c(\mathbf{r}_i) + \mu_t \times \mathbf{e}_i . \]
\[ \zeta_{tr} = 15.4, N_c = 6.3 \]
\[ \zeta_{tr} = 0, N_c = 3.26 \]
\[ \zeta_{tr} = 17.9, N_c = 22 \]
\[ \zeta_{tr} = 23.2, N_c = N \]

In Fig. 1 we illustrate typical particle configurations for increasing $\zeta_{trans}$ at $\zeta_{rot} = -0.38$ and $Pe = 19$. Top left: Gas-like state, top right: dynamic clustering 1, bottom left: dynamic clustering 2, bottom right: collapsed state. $N_c$ is the mean cluster size.

![FIG. 1. Snapshots of colloid configurations for increasing $\zeta_{trans}$ at $\zeta_{rot} = -0.38$ and $Pe = 19$. Top left: Gas-like state, top right: dynamic clustering 1, bottom left: dynamic clustering 2, bottom right: collapsed state. $N_c$ is the mean cluster size.](image)

![FIG. 2. State diagram: $\zeta_{tr}$ versus $\zeta_{rot}$ at $Pe = 19$. The mean cluster size $N_c$ is color coded.](image)
(see movie 1 and 2 in the supplemental material). In Fig. 2, we plot a state diagram in the phoretic parameters $\zeta_{\text{tr}}$, $\zeta_{\text{rot}}$, where we also color-code the mean cluster size $N_c$. At $\zeta_{\text{rot}} = 0$ the maximal dynamic cluster size just before the collapse is $N_c \approx 5$. When we turn on rotational diffusiophoresis with $\zeta_{\text{rot}} > 0$, the swimming direction $e$ of a free active colloid points towards a cluster (chemical sink) which further supports the formation of one cluster. This explains the fact that in Fig. 2 the collapse occurs for smaller $\zeta_{\text{tr}}$ when $\zeta_{\text{rot}}$ increases. The mean cluster size just before the collapse decreases and dynamic clustering is hardly visible. In contrast, at $\zeta_{\text{rot}} < 0$ active particles rotate away from chemical sinks and thus an effective repulsion is introduced. Once active colloids join a cluster, their swimming direction rotates outwards and the colloids can leave the cluster again if the translational phoretic attraction is not too large. This balance of effective phoretic attraction and repulsion is the cause for pronounced dynamical clustering with large cluster sizes. Interestingly, the state diagram in Fig. 2 indicates two clustering states: one where cluster sizes up to 6.5 are observed (see snapshot top, right in Fig. 1) and a second clustering state where much larger dynamic clusters occur (see snapshot bottom, left in Fig. 1). We will characterize these two states in the next paragraph. In a chemotaxis model for bacteria, the chemotactic collapse has been rationalized by Keller and Segel [29]. Below, we will demonstrate that our model can be mapped on the Keller-Segel equation, which qualitatively explains the nearly straight transition line in the state diagram between the gas-like and the collapsed state.

To further quantify the two dynamic clustering states 1 and 2, we determine the cluster-size distribution $P(n)$. In Fig. 3, we plot it for fixed rotational phoretic parameter $\zeta_{\text{rot}} = -0.38$ and increasing translational coupling $\zeta_{\text{tr}}$. For pure steric interaction ($\zeta_{\text{tr}} = 0$, blue curve), an exponential decay is predominant. Closer to the transition line between dynamic clustering states 1 and 2 in Fig. 2, the distribution follows a power law at small $n$, before it falls off exponentially (orange and red curves in Fig. 3). Indeed, we can fit our results by $P(n) = c_1 n^{-\beta_1} \exp(-n/n_0)$ with exponent $\beta_1 = 2.1 \pm 0.1$, which gradually decreases for more negative $\zeta_{\text{rot}}$. With further increase of $\zeta_{\text{tr}}$ we observe that the distribution $P(n)$ develops an inflection point (green curves) and we have to use a sum of two power-law-exponential curves to fit our distributions, $p(n) = c_1 n^{-\beta_1} \exp(-n/n_0) + c_2 n^{-\beta_2} \exp(-n/n_2)$ with $\beta_1 = 2.1 \pm 0.2$ and $\beta_2 \approx 1.5$. This defines the dynamic clustering state 2, where very large clusters coexist with smaller ones and individual particles. The transition in the cluster-size distribution is observed for all negative $\zeta_{\text{rot}}$. Typically, the mean cluster size $N_c$ increases strongly in state 2 as indicated in the inset of Fig. 3. In contrast, for $\zeta_{\text{rot}} > 0$ the system exhibits the collapse to a single cluster before large dynamic clusters can appear.

Similar forms of the cluster-size distributions including the transition indicated by the occurrence of an inflection point has been observed in experiments on gliding bacteria [7]. However, in this work bacterial density was varied in a system with pure hard-core interactions causing nematic alignment. In contrast, in our case we vary the strength of the diffusiophoretic coupling ultimately leading to the collapsed state which has not been observed in Ref. [7].

The experiments with diffusiophoretic coupling showed a linear scaling of the mean cluster size with $\Pe$: $N_c \sim \Pe$. It appears counterintuitive that at low area fractions of $\sigma = 5\%$ faster colloids generate larger clusters and indeed the simulations of our model show the contrary behavior. In Fig. 4, we plot a state diagram in $\zeta_{\text{tr}}$ versus $\Pe$ while $\zeta_{\text{rot}}$ is kept constant. Clearly, for constant


FIG. 5. Mean cluster size \( N_c \) versus \( \text{Pe} \) for different lines in the full parameter space defined via a parametrization with \( x \in [0, 1] \). We vary \( \text{Pe} \) as in experiments of [25]. \( \text{Pe} = 9.5 + 11.5x \), choose \( \zeta_{tr} = 4.8 + 16.6x \) and \( \zeta_{rot} = -0.16 - \zeta_0 x \), where the parameter \( \zeta_0 \) defines the different graphs. The transition between clustering states 1 and 2 roughly occurs at the intersection of the two straight lines.

\( \zeta_{tr} \) large clusters disappear with increasing \( \text{Pe} \). However, large activity \( \text{Pe} \) is necessary for observing dynamic clustering. At small \( \text{Pe} \) only small cluster sizes occur with increasing \( \zeta_{tr} \), while at sufficiently large \( \text{Pe} \) we observe both dynamic clustering states.

In the experiments of Ref. [25] colloidal activity increases with the concentration \( c_0 \) of the activating chemical \( \text{H}_2\text{O}_2 \). According to [39], the swimming velocity scales as \( v_0 \sim c_0 \propto \text{Pe} \). The concentration \( c \) also couples to our reparametrized diffusiophoretic parameters \( \zeta_{tr} \) and \( \zeta_{rot} \) through the reaction rate \( k \) introduced in Eq. (5). In the following, we assume Michaelis-Menten kinetics for the reaction rate in the linear regime well before saturation, \( k \sim c_0 \), and thus find \( \zeta_{tr} \sim c_0 \sim \zeta_{rot} \). So, the linear dependence of the parameters \( \text{Pe}, \zeta_{tr}, \) and \( \zeta_{rot} \) on \( c_0 \) defines a line in the parameter space. In this space the dynamic clustering states 1 and 2 are separated by a plane. We choose different lines which always hit the transition plane and plot in Fig. 5 the mean cluster size \( N_c \) versus \( \text{Pe} \) along the lines. The blue and purple curves show the strong increase of \( N_c \) when the clustering state 2 is entered, since the respective lines hit the transition plane under angles closer to 90°. Making this angle smaller, the increase is more modest. In particular, the green graph shows an almost linear increase of \( N_c \) in the \( \text{Pe} \) range from 10 to 20. So, Fig. 5 demonstrates that the relation between \( N_c \) and swimming velocity does not have a simple dependence.

To gain some more insight into the transition to the collapsed state, we write down a Smoluchowski equation for the full spatial and orientational probability distribution. We determine its orientational moments and derive from them an equation for the spatial colloidal density \( P(\mathbf{r}, t) \) coupled to the chemical field \( c(\mathbf{r}, t) \), similar to the approach in Ref. [43]:

\[
\dot{P} = \zeta_{eff} \nabla \cdot (P \nabla c) + \frac{D_{eff}}{2} \nabla^2 P + O((|\nabla c|^2). \tag{6}
\]

Details of the derivation are given in the supplemental material. Equation (6) is reminiscent of one relation of the Keller-Segel model [29] used to describe the chemotaxis of bacteria but here with effective chemotactic and diffusion constants: \( \zeta_{eff} = \zeta_{tr} + \zeta_{rot} \sigma_0 \frac{D_{eff}}{D_{eff}} \) and \( D_{eff} = D_{tr} + \frac{\sigma_0^2}{D_{rot}} \). In two dimensions the Keller-Segel equation exhibits an instability of the uniform state towards a chemotactic collapse when its parameters satisfy \( \zeta_{rot} \frac{\sigma_0^2}{D_{eff}} > b \), where \( \sigma \) is area fraction of the colloids and \( b \) a constant which depends on the geometry of the system [42, 45]. In our unitless parameters the condition becomes \( \frac{\sigma_0^2 (\zeta_{tr} + \zeta_{rot} \text{Pe})}{1 + 2 \text{Pe}^2} > b \). For constant \( \text{Pe} \), this agrees nicely with the nearly straight transition line in Fig. 2 between the gas-like and the collapsed state at \( \zeta_{rot} > 0 \) which extents to the line separating clustering states 1 and 2. However, the condition does not reproduce the straight transition line in Fig. 1. The reason is that at \( \zeta_{rot} < 0 \) pronounced clustering occurs before the collapse takes place. Clustering, however, cannot be described by the Keller-Segel equation since we neglect direct two-particle interactions during its derivation. To test the collapse condition in regions where dynamic clustering is absent, we determine the transition line \( \zeta_{tr} \) versus \( \text{Pe} \) for positive \( \zeta_{rot} \) and indeed find a quadratic dependence in \( \text{Pe} \) as Fig. 1 in the supplemental material shows. The inset reproduces the predicted linear dependence between \( \zeta_{rot} \) and \( \text{Pe} \) for \( \zeta_{tr} = 0 \). On the other hand, the straight transition line between clustering states 1 and 2 in Fig. 4 follows directly from Eq. (3). At \( \zeta_{rot} < 0 \), the particles at the rim of a cluster point away from it. So, large clusters form when swimming away from the cluster and attraction to the cluster by translational diffusiophoresis balance each other, which means \( v_0 \propto |\zeta_{tr} \nabla c| \).

Self-phoretic active colloids mediate diffusiophoretic interactions between each other. They consist of a translational and an orientational part which together with the active swimming can act either attractively or repulsively. When they are both attractive, the colloids show a transition from the gas-like to a collapsed state reminiscent of a chemotactic collapse in bacterial systems as our mapping on the Keller-Segel model demonstrates. When translational and rotational diffusiophoresis generate counteracting attraction and repulsion, two dynamic clustering states with characteristic cluster size distributions are stabilized very similar to the dynamic clustering observed in the experiments of [25].

The present system mimics chemotaxis in bacterial colonies without relying on a complex signalling pathway necessary in cells. Thereby, it may help to explore chemotactic structure formation and design novel dynamic patterns in bacterial colonies [10].

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