Which Interactions Dominate in Active Colloids?

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Active colloids, which self-propel by producing a chemical gradient across their own surface, serve as versatile microengines sparking a huge potential for the creation of functional materials through nonequilibrium self-assembly. Despite a mounting evidence that the same gradients which are used for swimming induce important cross-interactions among active colloids (phoretic interaction), they are still largely ignored in many-body descriptions, perhaps to avoid complexity and a zoo of unknown parameters. Here we derive a simple model for active colloids, suggesting that phoretic interactions are the dominant interaction in typical active colloids (rather than hydrodynamic interactions) and are largely controlled by one genuine parameter - the self-propulsion speed. Unlike present standard models, but in accordance with canonical experiments, the model predicts dynamic clustering as a generic behaviour in active colloids. The present work strongly simplifies descriptions of interacting active colloids and enables exploring phoretic interactions in simple Brownian dynamics simulations and generic field theories.

Introduction Since their first realization at the turn to the 21st century \textsuperscript{12,13} active colloids \textsuperscript{14,15} have experienced a steep evolution. Now, they serve as a versatile platform for designing functional devices and are used, for example, as microengines \textsuperscript{11,16,17} and cargo-carriers \textsuperscript{10,11} aimed to deliver drugs towards cancer cells in the future. These colloids self-propel by catalyzing a chemical reaction on part of their surface, creating a gradient which couples to the surrounding solvent and drives them forward. When many active colloids come together, they self-organize into spectacular patterns, which would be impossible in equilibrium, but offers huge perspectives for the creation of new materials through nonequilibrium self-assembly \textsuperscript{12,19}. A typical pattern, reoccurring in canonical experiments with active Janus colloids, are so-called living clusters which spontaneously emerge at remarkably low densities (area fraction 3 – 10\%) and dynamically split up and reform as time proceeds \textsuperscript{12,22,24}. When trying to understand such collective behaviour in active colloids, we are facing complex setups of motile particles showing multiple competing interactions, such as steric, hydrodynamic and phoretic ones (the latter ones hinge on the cross-action of self-produced chemicals on other colloids).

Therefore, to reduce complexity and allow for descriptions which are simple enough to promote our understanding of the colloids' collective behaviour, yet sufficiently realistic to represent typical experimental observations (such as dynamic clustering) we have to resolve the quest: which interactions dominate in active colloids? - the topic of the present letter. Presently, the most commonly considered models in the field, like the popular Active Brownian particle model \textsuperscript{25,26} and models involving hydrodynamic interactions \textsuperscript{27,28} neglect phoretic interactions altogether, perhaps to avoid complexity and unknown parameters which their description usually brings along. Conversely, recent experiments \textsuperscript{12,16,19,22}, simulations \textsuperscript{20,21}, and theories \textsuperscript{29} suggest a dominant importance of phoretic interactions in active colloids - which, after 15 years of research on active colloids, still leaves us with a conflict calling for clarification.

Here, we show that phoretic interactions dominate over hydrodynamic interactions in typical active colloids (with exceptions being discussed below). As opposed to (biological) microswimmers moving by body-shape deformations \textsuperscript{27,30,38}, hydrodynamic interactions can therefore be approximately neglected for many active colloids at low and moderate density, but not phoretic interactions. As our key result, we derive the Active Attractive Alignment model (AAA model), providing a strongly reduced but essentially realistic description of typical active colloids. In particular, the AAA model reduces phoretic interactions to a simple pair interaction among the colloids whose strength is controlled by one genuine parameter, the self-propulsion speed (or Pécelt number). This allows to include them e.g. in Brownian dynamics simulations, rather than requiring hydrid particle-field descriptions and releases their modeling from the zoo of unknown parameters it usually involves \textsuperscript{39,44}. As opposed to present standard models of active colloids, the AAA model generically predicts dynamic clustering at low density, in agreement with experiments \textsuperscript{12,22,24}. Complementarily to \textsuperscript{29} predominantly exploring repulsive phoretic interactions and delay effects, the present work focuses on attractive phoretic interactions as probably relevant to most existing experiments. Our work should be broadly useful to model active colloids and to design active self-assembly \textsuperscript{16,19,45,46}.

Phoretic motion in external gradients: When exposed to a gradient in an imposed phoretic field $c$, which may represent e.g. a chemical concentration field, the temperature field or an electric potential, colloids move due to phoresis. Here, the gradients in $c$ act on the fluid el-

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emements in the interfacial layer of the colloid and drive a localized solvent flow tangentially to the colloidal surface with a velocity, called slip velocity

$$v_s(r_s) = \mu(r_s) \nabla_c(r_s)$$  \hspace{1cm} (1)$$

Here \(r_s\) is a point immediately above the colloidal surface and \(\nabla_c\) is the projection of the gradient of \(c\) onto the tangential plane of the colloid. The colloid moves opposite to the average surface slip with a velocity \(\mathbf{v} = -\langle v(r_s) \rangle\) where brackets represent the average over the colloidal surface. If the solvent slips asymmetrically over the colloidal surface, the colloid also rotates with a frequency \(\frac{3}{2\pi} \langle \mathbf{v}(r_s) \times \mathbf{n} \rangle\) where \(R, \mathbf{n}\) are the radius and the local surface normal of the colloid. Performing surface integrals, specifically for a Janus colloid with a catalytic hemisphere with uniform surface mobility \(\mu_c\) and a mobility of \(\mu_N\) on the neutral side, yields:

$$\mathbf{v}(r) = -\frac{\mu_c + \mu_N}{3} \nabla_c; \quad \Omega(r) = \frac{3(\mu_c - \mu_N)}{8R} \mathbf{e} \times \nabla_c$$  \hspace{1cm} (2)$$

Here, we evaluate \(c\) at the colloid center \(r\) for simplicity, and have introduced the unit vector \(\mathbf{e}\) pointing from the neutral side to the catalytic cap.

**Self-propulsion**  Autophoretic colloidal microswimmers, or active colloids, self-propel phoretic fields on part of their surface with a local surface production rate \(\sigma(r_s)\). In steady state, we can calculate the self-produced field by solving

$$0 = D_c \nabla^2 c + \int d\mathbf{x}_i \delta(r - \mathbf{r}_i(t) - R\mathbf{n}_i) \sigma(\mathbf{x}_i) - k_d c$$  \hspace{1cm} (3)$$

where \(D_c\) is the diffusion constant of the relevant phoretic field \([28]\), \(k_0\) is the production rate per particle and the sink term \(-k_d c\) represents an effective decay of the relevant phoretic field (chemicals, heat, ions), which may result e.g. from bulk reactions \([29]\) (including fuel recovery \([10]\)), or heat absorption processes. While so-far neglected in most of the literature, Fig. 1 shows that such a sink term should be included when describing phoretic interactions among active colloids as we shall see below. Conversely, self-propulsion, i.e. the phoretic drift of a colloid in its self-produced gradient, depends only on the phoretic field close to the colloid surface, so that we can ignore the decay. Considering Janus colloids which produce chemicals with a local rate \(\sigma = k_0/(2\pi R^2)\) on one hemisphere and \(\sigma = 0\) on the other one, solving Eq. (3) for \(k_d = 0\) and using \([12]\), we find its self-propulsion velocity \([7]\)

$$v_0 = -\frac{k_0(\mu_N + \mu_c)}{16\pi R^2 D_c} \mathbf{p}$$  \hspace{1cm} (4)$$

where \(\mathbf{p}\) is the unit vector pointing from \(r\) into the swimming direction of the test colloid. For symmetry reasons the considered Janus colloids do not show self-rotations.

**How strong are phoretic interactions?** Besides leading to self-propulsion, the gradients produced by an autophoretic colloid also act in the interfacial layer of all other colloids. Here, they drive a solvent slip over the colloids’ surfaces, which induce a phoretic translation and a rotation. Following Eqs. \([12, 14]\) a colloid at the origin at position \(r\) with

$$\mathbf{v}_p(r) = -\nu \frac{6D_c \pi R v_0}{k_0} \nabla_c; \quad \Omega_p(r) = \mu_r \frac{6D_c \pi R v_0}{k_0} \nabla_c$$  \hspace{1cm} (5)$$

Here, \(\nu = -1\) for swimmers moving with their catalytic cap ahead and \(\nu = 1\) for cap-behind swimmers \([29]\); we have further used \(v_0 = |v_0|\) and have introduced the reduced surface mobility \(\mu_r = (\mu_c - \mu_N)/\mu_c\). Now solving Eq. (3) in far-field, yields the chemical field produced by the colloid at the origin

$$c(r) = \left[ \frac{k_0}{4\pi D_c r^4} + \frac{R}{4(2\pi)^{3/2}} \frac{\hat{p} \cdot \hat{r}}{r^2} + O\left(\frac{1}{r^3}\right) \right] e^{-\kappa r}$$  \hspace{1cm} (6)$$

where \(\kappa = \sqrt{k_d/D_c}\) is an effective inverse screening length; the case \(\kappa = 0\) corresponds to absence of screening. Finally combining Eqs. (3) and (5) yields, in leading order

$$\mathbf{v}_p(r) = -\frac{4\pi R v_0}{3} \frac{\mathbf{e} \times \nabla e^{-\kappa r}}{r}$$  \hspace{1cm} (7)$$

$$\Omega_p(r) = -\frac{3\pi R \mu_r}{2} \hat{p} \times \frac{\mathbf{e} \times \nabla e^{-\kappa r}}{r}$$  \hspace{1cm} (8)$$

Except for \(\kappa, \mu_r\) which we will estimate below and \(\nu = \pm 1\), the prefactors in Eqs. (7,8) only depend on the self-propulsion velocity and the colloidal radius, which are well known in experiments. We can further see from Eq. (7) that colloids at a typical distances of \(\sim 5R\), approach each other (for \(\nu = 1\)) within a few seconds (this is consistent with experiments, e.g. \([22, 13]\); at shorter distances the phoretic translation speed becomes comparable to the self-propulsion speed, i.e. \(v_p \sim v_0\). A typical alignment rate with the chemical gradient produced by an adjacent colloid \((\mathbf{R} = 1\mu m, v_0 \sim 10\mu m/s, \mu_r = 0.15\) \([19]\), is \(|\mathbf{R}| \sim 0.1/s\), i.e. colloids may approach each other due to phoretic translation before turning much. Thus, it is plausible that when forming clusters (see below), they do not show much orientational order \([21]\).

**Comparison with hydrodynamic interactions** In bulk, the hydrodynamic flow field induced by an isotropic, phoretically moving colloid (well beyond its interfacial layer), at a point \(r\) relative to its center is long known \([50]\) as

$$\mathbf{v}(r) = \frac{1}{2} \left( \frac{R}{r} \right)^3 (3\hat{r} \times I) \cdot \mathbf{v}_0$$  \hspace{1cm} (9)$$

Such a potential flow field also occurs frequently for Janus colloids in the literature \([28, 51]\), e.g. for certain self-thermophoretic swimmers (with thin caps) \([52, 55]\), and for half-coated self-electrophoretic \([50, 57]\) and
self-diffusiophoretic \[20\] \[57\] Janus colloids with uniform surface mobility. We estimate the relative strength of phoretic \[7\] and such \(1/r^3\)-hydrodynamic flows \[9\] advecting other colloids (in far field) via a parameter \(m(r) := 8r^3\partial_r(\exp[-\kappa r]/r)/(3R)\). When \(m > 1\) phoretic interactions should dominate over hydrodynamic ones. Without a chemical decay \((\kappa = 0)\) \[12\] \[19\] \[29\] \[41\] we have \(m \gg 1\) at all relevant distances (i.e. beyond the near field regime). For \(\kappa > 0\), hydrodynamic interactions may dominate at very long distances, but not at typical ones. For \(R = 1 \mu m\) colloids at 10% area fraction (average distance 5.6\(\mu m\)) and a screening length of \(\kappa R = 0.25\) (Fig. 1), we find \(m \sim 9\), and even for \(\kappa R \sim 0.5\), we have \(m \sim 4\); higher densities further support the dominance of phoretic interactions.

Generally, the far field flow of Janus colloids with a nonuniform surface mobility shows also a \(1/r^2\)-contribution \[9\] \[58\] \[59\], although often with a small coefficient. Remarkably, \[59\] has recently quantified the flow induced by autophoresis Janus colloids by mapping to a squirmer model, unveiling a radial component of strength \(v(r) \sim |\mu_r|(R/r)^2v_0\). A comparison with Eq. \([7]\) suggests that unscreened \((\alpha = 0)\) and moderately screened \((\alpha = 0.25)\) phoretic interactions still dominate over hydrodynamic ones by about a factor of \(1/\mu_r \sim 3-20\) \((\sim 2-6\), at 10% area fraction) \[49\]. Besides this strength-comparison, the dominance of phoretic interactions in active suspensions receives further support (i) from the fact that they are isotropic (in leading order), whereas anisotropic hydrodynamic flows should mutually cancel to some extent and (ii) from the additional phoretic alignment, Eq. \([8]\), which on its own can destabilize the uniform phase at low density \[29\] and is particularly important when \(\mu_r\) is large, i.e. when hydrodynamic \(1/r^2\)-flows are significant. These findings agree with microscopic simulations of Janus colloids showing clustering at low density due to phoretic interactions, but not without \[20\]. Conversely to the discussed cases which we consider as typical, hydrodynamic interactions may of course still be important (or dominant) e.g. in Janus colloids with strong effective screening, strongly nonuniform surface mobility, and at high density, where hydrodynamic \[60\] \[61\] and other near-field interactions become relevant.

The Active Attractive Aligning Model To describe the collective behaviour of \(N\) active colloids, we first consider the Active Brownian particle model for colloids confined to quasi-2D. Using \(x_n = R\) and \(\bar{t}_u = 1/D_t\) as space and time units, where \(D_t\) is the translational diffusion constant, and introducing the Péclét number \(\text{Pe} = v_0/(D_tR)\) this model reads (in dimensionless units)

\[
\dot{x}_i = \text{Pe} \ p_i + f_i(x_i); \quad \dot{\theta}_i = \sqrt{2\eta_i}(t) \tag{10}
\]

Eqs. \([10]\) describe particles which sterically repel each other (here represented by dimensionless forces \(\mathbf{f}_i\) preventing particles to overlap at short distances) and self-propel with a velocity \(v_0\) in directions \(\mathbf{p}_i = (\cos \theta_i, \sin \theta_i)\) \((i = 1..N)\) which change due to rotational Brownian diffusion; here \(\eta_i\) represents Gaussian white noise with zero mean and unit variance. Following Eq. \([7,8]\), we can now account for phoretic far-field interactions leading to the “Active Attractive Aligning Model”, or AAA model which we define as (see below for a 3D variant):

\[
\dot{x}_i = \text{Pe} \ p_i - \frac{4\text{Pe} \mu}{3} \nabla u + f_i(x_i)
\]

\[
\dot{\theta}_i = -\frac{3\text{Pe} \mu}{2} p_i \times \nabla u + \sqrt{2\eta_i}(t) \tag{11}
\]

Here, \(\nabla u = \sum_{j=1}^{N} \nabla x_j e^{-a x_{ij}}\) with \(x_{ij} = |x_i - x_j|\) and \(a \times b = a_1 b_2 - a_2 b_1\) for 2D vectors \(\mathbf{a}, \mathbf{b}\) and where we have introduced a screening number \(\alpha = R \sqrt{k_B T / D_c}\). In our simple derivation, we have identified phoretic translations and rotations of the colloids with formally identical expressions representing reciprocal interaction forces (attractive Yukawa interactions for \(\nu = 1\); Coulomb for \(\alpha = 0\)) and (nonreciprocal) torques aligning the self-propulsion direction of the colloids, towards \((\mu_r > 0,\) positive taxis\)) or away \((\mu_r < 0,\) negative taxis\)) from regions of high particle density. Remarkably, for a given screening number (realistic values might be \(\alpha \lesssim 0.25 - 0.65\), Fig. 1), the strength of the phoretic interactions is largely determined by one genuine parameter - the Péclét number - constituting a collapse of parameter space.

Properties of the AAA model (i) For \(\mu_r = 0, \nu = 1\); the AAA model reduces to active Brownian particles with attractions; however, as opposed to corresponding phenomenological models \[63\] \[66\] \[73\], the AAA model explicitly relates the interaction strength to the Péclét number. (ii) When the colloids respond with a significant delay to their self-produced fields, which can happen even for very large \(D_r\), \[29\], the AAA model becomes invalid; presumably this is relevant mainly for repulsive phoretic interactions \((\nu < 1\) or \(\mu_r < 0)\) \[29\]. (iii) The Yukawa interactions in Eqs. \([11]\) are reciprocal only when considering identical colloids. Mixtures of nonidentical Janus colloids, active-passive mixtures or of uniformly coated colloids lead to nonreciprocal interactions inducing a net motion of pairs \[18\] \[55\] \[67\]. For example, passive particles can be included in the AAA model via \(\mathbf{f}_i = -(4/3)\mu \nu \text{Pe} \nabla x_i \eta(x_i)\) where \(\text{Pe}\) is the Péclét number of the active colloids and \(\mu = 2\mu_P/(|\mu_N + \mu_C|)\) with \(\mu_P\) being the surface mobility of the (isotropic) passive colloid. (iv) For single-species isotropically coated colloids \((v_0 = 0)\) the AAA model reduces to the hard-core Yukawa model (when accounting for translational diffusion). Thus, chemically active colloids can be used to realize the (attractive or repulsive) hard-core Yukawa model, which has been widely used to describe effective interactions between charged colloids \[68\] \[69\], globular proteins \[70\] and fullerenes \[71\]. (v) Generalizations of the AAA model to 3D are straightforward; here the orientational dynamics follows \(\dot{x}_i = -(3/2)\text{Pe} \mu_r (I - I \mathbf{p}_i) \nabla u + \sqrt{2\eta_i} \times \mathbf{p}_i\), where \(\mathbf{p}_i\) is the 3D unit vector representing the swimming direction of particle \(i\) and \(\eta_i\) represents Gaussian white noise of zero mean and unit variance.
Dynamic Clustering in the AAA Model

The AAA model generically leads to dynamic clustering at low density. We show this in Brownian dynamics simulations (Fig. 1) at Pe = 100 and $\alpha = 0.25$: (i) Without alignment ($\mu_r = 0$) clusters dynamically emerge, break up and move through space, as in canonical experiments [12, 22–24]. For an area fraction of $\phi = 5\%$, these clusters do not grow beyond a certain size (red line in Fig. 1 F); conversely, for $\phi = 10\%$ once a cluster has reached a certain size (Fig. 1 B), it does not break any further and grows (panel E, green line). The cluster size distribution (not shown) is approximately algebraic at small sizes and decays exponentially at larger sizes, also as in experiments [24]. (ii) Similarly for $\mu_r = -1$ (strong negative taxis) we also find dynamic clusters (panel C); here negative taxis stabilizes the dynamic cluster phase and clusters do not grow at late times, even for $\phi = 0.1$ (black curve in F) and for $\phi = 0.2$ (not shown). This combination of attractive translation combined with negative taxis resembles [10]. (iii) For $\mu_r = 1$ where particles strongly align towards regions of high density (positive taxis), we find rigid clusters (panel D) which coalesce and form one macrocluster at late times.

Note that the clusters seen in cases (i),(ii) differ from those occurring as a precursor of motility-induced phase separation [23, 26, 72–77] in the (repulsive) Active Brownian particle (ABP) model [23, 26, 72–77]. The ABP model only leads to very small and short lived clusters at low area fractions; here the cluster size distribution decays exponentially with the number of particles in the cluster (unless we are at area fractions of $\gtrsim 30\%$ close to the transition to motility induced phase separation). In contrast, both in experiments and in the AAA model, we see significant clusters at low area fractions (3–10%), with a cluster size distribution decaying rather algebraically with size over a significant interval of cluster sizes [12, 22–24].

Conclusions

Our results suggest that phoretic interactions dominate in typical autophoretic colloids (and active-passive mixtures) whereas hydrodynamic interactions can be approximately neglected at low and moderate density. Janus colloids featuring a strongly asymmetric cap geometry or surface mobility or experiencing strong effective screening might serve as exceptions. Our key result, the AAA model, enables a much simplified description of phoretic far-field interactions and allows including them in Brownian dynamics simulations. The model determines the strength of phoretic interactions largely via one genuine parameter, the swimming speed (Péclet number), and does not require multiple unknown parameters as in most previous descriptions. The AAA model offers the to-date simplest microscopic model of active colloids agreeing with canonical experiments showing dynamic clustering and should be useful e.g. to design active self-assembly.
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[48] For self-diffusiophoretic swimmers we understand \( \mu \) as the sum of fuel and reaction-product species and \( D_\text{e} \) as an effective diffusion coefficient of the combined field.
[49] Consider a solute of neutral, dipolar molecules (water, \( H_2O_2 \)) featuring excluded volume and dipolar interactions with a colloidal surface. Excluded volume interactions should not depend much on the surface material favoring \( \mu_\text{r} = 0 \) for Janus particles. The surface mobility of a colloid due to dipolar interactions reads \( \mu \approx \frac{16\xi_\text{c}^2}{5\eta} \xi^2 + O(\xi^4) \) where \( \xi = \tanh[Z\zeta/(4kT)] \) with \( \zeta \) being the zeta potential, \( \mu_\text{D} \) the solute dipole moment and \( Z \) the valence of the support electrolyte (we assume \( Z = 1 \); \( Z > 1 \) reduces |\( \mu_\text{r} |\)). For Janus colloids...
with cap (C) and neutral side (N), this yields $|\mu_r| = |(\mu_C - \mu_N)/(\mu_C + \mu_N)| \approx |(\xi_C^2 - \xi_N^2)/(\xi_C^2 + \xi_N^2)|$. Measurements for typical coating materials: $\zeta \approx -64\text{mV}$ both for isotropic $2R = 1.7\mu\text{m}$ polystyrene (PS) and $1\mu\text{m}$ gold spheres in $5\%\text{H}_2\text{O}_2$ solution [78]; $\zeta \approx -52\text{mV}, -67\text{mV}$ for polystyrene and silica colloids (sizes $1-3\mu\text{m}$) respectively in $0.01\text{M KCl}$ solution [10] and for $2R = 1\mu\text{m}$ spheres in water $\zeta \approx -50\text{mV}$ (PS,SiO$_2$), and $\zeta \approx -63\text{mV}$ (TiO$_2$) [79]. Direct measurements for the two halves of a $2R = 4.8\mu\text{m}$ PS-Pt Janus colloid in water (and in $5\%, 10\%\text{H}_2\text{O}_2$) yield $\zeta \approx -95\text{mV}(-105\text{mV}, -110\text{mV})$ for the PS side and $\zeta \approx -80\text{mV}(-70\text{mV}, -70\text{mV})$ for the Pt. Based on these values we estimate $|\mu_r| \approx 0.05 - 0.3$ for typical Janus swimmers (or less if excluded volume interactions dominate). Similarly, for electrophoretic Janus swimmers $\mu_r$ also hinges on differences in $\zeta$.

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