

## Effective Interaction between Active Colloids and Fluid Interfaces Induced by Marangoni Flows

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We show theoretically that near a fluid-fluid interface a single active colloidal particle generating, e.g., chemicals or a temperature gradient experiences an effective force of hydrodynamic origin. This force is due to the fluid flow driven by Marangoni stresses induced by the activity of the particle; it decays very slowly with the distance from the interface, and can be attractive or repulsive depending on how the activity modifies the surface tension. We show that, for typical systems, this interaction can dominate the dynamics of the particle as compared to Brownian motion, dispersion forces, or self-phoretic effects. In the attractive case, the interaction promotes the self-assembly of particles into a crystal-like monolayer at the interface.

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Significant attention has been paid lately to micrometer sized particles capable of self-induced motility [1–3]. They are seen as promising candidates for novel techniques in chemical sensing [4] or water treatment [5]. The motion of active colloidal particles has been the subject of numerous experimental [1–3,6,7] and theoretical [8–12] studies. One realization is a particle with a catalytic surface promoting a chemical reaction in the surrounding solution [13]. For an axisymmetric particle lacking fore-and-aft symmetry, the distributions of reactant and product molecules may become nonuniform along its surface and the particle could move due to self-induced phoresis [14]. If the particle is spherically symmetric, it will remain immobile in bulk solution but can be set into motion by the vicinity of walls or other particles (not necessarily active) which break the spherical symmetry [10,13–16].

A relevant case corresponds to the movement of active particles bounded by a fluid-fluid interface. This situation raises new issues, in particular if the reactants or the products have a significant effect on the properties of the fluid interface implying *tensioactivity*. For example, it has been recently predicted that catalytically active, spherical particles which are trapped *at* the interface may be set into motion *along* the interface by Marangoni flows, self-induced via the spatially nonuniform distribution of tensioactive molecules [17–19]. (A similar motility mechanism can originate from thermally induced Marangoni flows if, e.g., the particle contains a metal cap which is heated by a laser beam [20].) Furthermore, self-induced Marangoni flows, combined with a mechanism of triggering spontaneous symmetry breaking, have also been used to develop self-propelled droplets [21–23].

However, another category of experimental situations occurs if the particles are not trapped *at* the interface but may *reside in the vicinity* of the interface or *get near* it

during their motion. In this study we provide theoretical evidence that such catalytically active or locally heated spherical particles, although immobile in bulk, experience a very strong, long-ranged effective force field due to the Marangoni stresses self-induced at the interface. This force of hydrodynamic origin manifests itself at spatial length scales much larger than those of typical wetting forces. It gives rise to a drift of the particle towards or away from the fluid interface, depending only on how the tensioactive agent, i.e., a gradient in chemical concentration or in temperature, affects the interface. This effect dominates any possible self-phoresis or dispersion interactions, and acts on time scales which can be orders of magnitude shorter than those associated with Brownian diffusion. This drift can facilitate particle adsorption towards the interface and therefore has important implications for the self-assembly of particles at fluid-fluid interfaces. We complement the theoretical calculations with a thorough analysis regarding the observability of these phenomena in future experiments.

The model system consists of a spherical colloidal particle with radius  $R$  in front of a flat interface at  $z = 0$  between two immiscible fluids. Fluid 1 (2) occupies the half space  $z > 0$  ( $z < 0$ ) (see Fig. 1). The spherical particle is located in fluid 1; its center is at  $\mathbf{x}_0 = (0, 0, L)$  with  $L > R$  (i.e., the particle does not penetrate through the interface). By virtue of a chemical reaction occurring uniformly over its surface [24], the particle acts as a spherically symmetric source (or sink) of a chemical species  $A$ . We assume that the time scale for diffusion of  $A$  is much shorter than any relevant time scale associated with fluid flows [25]. Therefore, we consider only the stationary state neglecting advection by the ensuing Marangoni flow. Additionally, the number density  $c(\mathbf{x} = \mathbf{r} + z\mathbf{e}_z)$  of species  $A$  [with  $\mathbf{r} = (x, y, 0)$  in the following] is

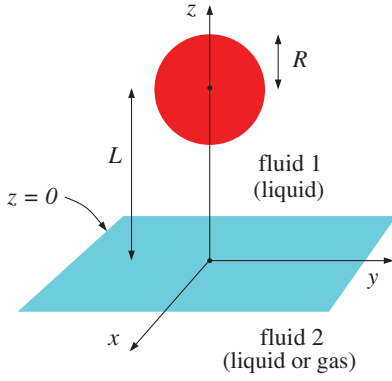


FIG. 1. Coordinates and configuration of the system. The interface between fluid 1 (liquid) and fluid 2 (liquid or gas) is located at  $z = 0$ .

assumed to be sufficiently small so that for  $A$  the ideal-gas approximation holds, and thus  $c(\mathbf{x})$  obeys Fick's law for diffusion with constant diffusivity  $D_\alpha$  in fluid  $\alpha$  ( $= 1, 2$ ):

$$\nabla^2 c(\mathbf{x}) = 0, \quad \mathbf{x} \in \text{fluid 1 or 2}, \quad (1a)$$

subject to the boundary conditions [25] that (i) a single reservoir of species  $A$  fixes the number density far away from the particle to be  $c_\alpha^\infty$  in fluid  $\alpha$  ( $= 1, 2$ ), (ii) the discontinuity of  $c(\mathbf{x})$  at the interface, given by  $\lambda := c(\mathbf{r}, z = 0^-)/c(\mathbf{r}, z = 0^+) = c_2^\infty/c_1^\infty$ , is determined, as in equilibrium, by the distinct solubilities of species  $A$  in the two fluids, (iii) the current of species  $A$  along the direction of the interface normal is continuous at the interface [ $D_1(\partial c/\partial z)|_{z=0^+} = D_2(\partial c/\partial z)|_{z=0^-}$ ], and (iv) the current at the surface  $S_p$  of the particle is

$$\mathbf{n} \cdot [-D_1 \nabla c(\mathbf{x})] = \frac{Q}{4\pi R^2}, \quad \mathbf{x} \in S_p, \quad (1b)$$

where  $\mathbf{n}$  is the unit vector normal to  $S_p$  (pointing into fluid 1);  $Q > 0$  ( $Q < 0$ ) is the rate of production (annihilation) of species  $A$ .

The surface tension  $\gamma$  of the fluid interface is assumed to depend on the local number density of species  $A$  and is modeled within the local equilibrium approximation as [25]

$$\gamma(\mathbf{r}) = \gamma_0 - b_0[c(\mathbf{r}, z = 0^+) - c_1^\infty]. \quad (2)$$

Here,  $\gamma_0$  is the surface tension in equilibrium in which the density of  $A$  in fluid 1 is  $c_1^\infty$ ; the effect of local deviations thereof are quantified by the coefficient  $b_0$ , the sign of which depends on the chemical.

This inhomogeneity of the surface tension induces Marangoni stresses which set the fluids into motion. The velocity field  $\mathbf{v}(\mathbf{x})$  can be derived as a solution of the Stokes equations (i.e., in the limit of incompressible flow and negligible inertia):

$$\nabla \cdot \mathbf{v}(\mathbf{x}) = 0, \quad \nabla \cdot \vec{\sigma}(\mathbf{x}) = 0, \quad \mathbf{x} \in \text{fluid 1, 2}, \quad (3a)$$

where  $\vec{\sigma}(\mathbf{x}) = \eta(\mathbf{x})[\nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger] - p(\mathbf{x})\mathcal{I}$  is the stress tensor in the fluid,  $p(\mathbf{x})$  the pressure,  $\eta(\mathbf{x}) = \eta_1$  or  $\eta_2$  the viscosity, and  $\mathcal{I}$  denotes the second-rank identity tensor. The Stokes equations are subject to the following boundary conditions: (i) vanishing velocity at infinity, (ii) no slip flow at the surface of the particle, (iii) at the interface, continuity of the tangential velocity and vanishing of the normal velocity, and (iv) balance between the tangential fluid stresses and the Marangoni stresses induced by the gradient of the surface tension along the interface:

$$(\mathcal{I} - \mathbf{e}_z \mathbf{e}_z) \cdot [\vec{\sigma}|_{z=0^+} - \vec{\sigma}|_{z=0^-}] \cdot \mathbf{e}_z = -\nabla_{\parallel} \gamma. \quad (3b)$$

The tensor  $\mathcal{I} - \mathbf{e}_z \mathbf{e}_z$  provides the projection onto the interfacial plane and  $\nabla_{\parallel} = (\partial_x, \partial_y, 0)$  is the nabla operator within the interfacial plane. (Actually, the interface must deform so that the normal component of the fluid stresses can be balanced by the Laplace pressure. *A posteriori* it turns out [25] that this deformation is typically so small that the flat interface approximation is reliable.)

The translation velocity  $\mathcal{V}$  of the particle, or equivalently the force  $\mathbf{F}$  exerted by the particle on the fluid, can be inferred from the Lorentz reciprocal theorem [32]. We consider the auxiliary flow field  $\mathbf{v}_{\text{aux}}(\mathbf{x})$ , for the same geometrical setup, corresponding to the translation of a rigid, spherical, *chemically passive* (i.e., without Marangoni stresses) particle in front of a flat fluid interface. This is the solution of Eq. (3a) subject to the same boundary conditions as above but with  $\nabla_{\parallel} \gamma = 0$  in Eq. (3b), a problem studied in Refs. [33–35]. We thus obtain [25]

$$\begin{aligned} \mathbf{F}_{\text{aux}} \cdot \mathcal{V} - \mathcal{V}_{\text{aux}} \cdot \mathbf{F} &= \int_{z=0} d^2 \mathbf{r} \nabla_{\parallel} \gamma(\mathbf{r}) \cdot \mathbf{v}_{\text{aux}}(\mathbf{r}) \\ &= - \oint_{S_p} dS \mathbf{n} \cdot \vec{\sigma}_{\text{aux}}(\mathbf{x}) \cdot \mathbf{u}(\mathbf{x}), \end{aligned} \quad (4)$$

with

$$\mathbf{u}(\mathbf{x}) = \int_{z=0} d^2 \mathbf{r}' \nabla_{\parallel} \gamma(\mathbf{r}') \cdot \mathcal{O}(\mathbf{x} - \mathbf{r}') \quad (5a)$$

in terms of the Oseen tensor,

$$\mathcal{O}(\mathbf{x}) = \frac{1}{8\pi\eta_+ x} \left[ \mathcal{I} + \frac{\mathbf{x}\mathbf{x}}{x^2} \right], \quad \eta_+ := \frac{1}{2}(\eta_1 + \eta_2). \quad (5b)$$

Here,  $\mathbf{u}(\mathbf{x})$  is the Marangoni flow, which would be induced solely by the Marangoni stresses  $\nabla_{\parallel} \gamma(\mathbf{r})$ , i.e., as if the surface of the particle would not impose any boundary condition on the flow [25]. Note that Eq. (4) can be interpreted as a generalization of the Faxén laws [36] for the present problem. For a force-free [ $\mathbf{F} = 0$  in Eq. (4)] spherical particle, the problem exhibits axial symmetry,

which implies that  $\mathcal{V}$  is parallel to  $\mathbf{e}_z$ . Thus, it suffices to solve the auxiliary problem with  $\mathbf{F}_{\text{aux}} \parallel \mathbf{e}_z$ , which allows one to introduce the dimensionless stream function  $\psi_{\text{aux}}(r, z)$ , in terms of which Eq. (4) reduces to [25]

$$\mathcal{V} = -\mathbf{e}_z 2\pi R^2 b_0 \Gamma_z \int_0^\infty dr \frac{\partial c}{\partial r} \Big|_{z=0^+} \frac{\partial \psi_{\text{aux}}}{\partial z} \Big|_{z=0}, \quad (6)$$

where  $\Gamma_z$  is the  $L$ -dependent mobility of a (chemically passive) rigid spherical particle moving normal to the planar fluid interface [35].

The boundary-value problems given by Eqs. (1) and (3), subject to the coupling provided by Eq. (2), can be solved exactly as a series in terms of bipolar coordinates [25]. However, the relevant phenomenology can be highlighted and significant physical intuition can be gained from an approximate closed form valid asymptotically in the limit  $R/L \rightarrow 0$ . We therefore proceed with the latter; its range of validity will be assessed later by comparison with the exact solution (cf. Fig. 3).

To the lowest order in  $R/L$ , the solution of Eq. (1) for the given boundary conditions can be obtained using the method of images in terms of monopoles located at  $\mathbf{x}_0 = (0, 0, L)$  and  $\mathbf{x}_0^* = (0, 0, -L)$ . In fluid 1 ( $z > 0$ ) one has

$$c(\mathbf{x}) = c_1^\infty + \frac{Q}{4\pi D_1} \left[ \frac{1}{|\mathbf{x} - \mathbf{x}_0|} + \frac{D_1 - \lambda D_2}{D_1 + \lambda D_2} \frac{1}{|\mathbf{x} - \mathbf{x}_0^*|} \right]. \quad (7)$$

Accordingly, the Marangoni flow (illustrated in Fig. 2) is given by Eqs. (2) and (5a) as  $\mathbf{u}(\mathbf{x}) = \mathbf{e}_z u_z(r, z) + \mathbf{e}_r u_r(r, z)$  with [25]

$$u_z(r, z) = -\frac{Qb_0}{16\pi D_+ \eta_+} \frac{z(|z| + L)}{[r^2 + (|z| + L)^2]^{3/2}}, \quad (8a)$$

$$u_r(r, z) = \frac{Qb_0}{16\pi D_+ \eta_+ r} \left[ 1 - \frac{r^2 L + (|z| + L)^3}{[r^2 + (|z| + L)^2]^{3/2}} \right], \quad (8b)$$

and  $D_+ := (D_1 + \lambda D_2)/2$ . The integral over  $S_p$  in the second line of Eq. (4) can be evaluated by expanding the Marangoni flow in terms of a Taylor series about the particle center so that asymptotically  $\mathbf{u}(\mathbf{x}) \approx \mathbf{u}(\mathbf{x}_0)$  for  $R/L \rightarrow 0$ . Since  $\oint_{S_p} dS \mathbf{n} \cdot \vec{\sigma}_{\text{aux}}(\mathbf{x}) = -\mathbf{F}_{\text{aux}}$ , and  $\mathbf{F}_{\text{aux}}$  can be chosen arbitrarily, one concludes that a force-free ( $\mathbf{F} = 0$ ) active particle at a distance  $L$  from the interface is carried by the flow with velocity

$$\mathcal{V}(L) \approx \mathbf{u}(\mathbf{x}_0) \approx -\mathbf{e}_z \frac{Qb_0}{64\pi D_+ \eta_+ L} \quad (9)$$

due to the self-induced Marangoni stresses. Since  $\mathcal{V} \parallel \mathbf{e}_z$ , this implies a time dependence of  $L$ .

Equation (9) captures the essence of all the relevant phenomenology. (i) For  $b_0 > 0$  (i.e., the generic surfactant case) the particle drifts towards or away from the interface

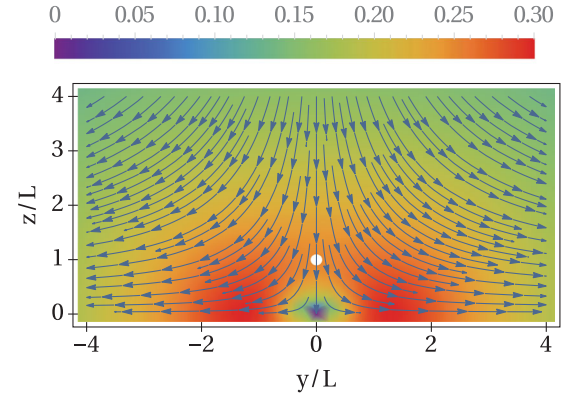


FIG. 2. Vertical cut through the Marangoni flow  $\mathbf{u}(\mathbf{x})$  in the limit  $R/L \rightarrow 0$  [Eq. (8)]. The streamlines follow the direction of the vector field (assuming  $Qb_0 > 0$ ), while the color code corresponds to  $|\mathbf{u}(\mathbf{x})|$  in units of  $|Qb_0|/(16\pi D_+ \eta_+ L)$ . The center of the particle (white dot) is at  $y = 0$ ,  $z/L = 1$ . The three-dimensional flow field is obtained by rotation around the  $z$  axis and mirror reflection with respect to the interfacial plane  $z = 0$ . (This flow is driven by the stress located at the interface, not by the particle.)

if it is a source ( $Q > 0$ ) or a sink ( $Q < 0$ ) of species  $A$ , respectively. For  $b_0 < 0$ , the behavior is reversed. (ii) The slow  $1/L$  decay of  $\mathcal{V}$  is tantamount to a long-ranged interaction with the fluid interface. The associated phenomenology can dominate the influence of dispersion forces between the particle and the interface, which decay  $\sim 1/L^4$  at best [37], and also the motion due to Brownian diffusion alone.

The argument can be quantified by introducing the diffusion coefficient  $D_p := k_B T / (6\pi \eta_+ R)$  of the particle in a medium of viscosity  $\eta_+$  at temperature  $T$ . Equation (9) leads to the Peclet number of the particle

$$\text{Pe}(L) := \frac{R|\mathcal{V}(L)|}{D_p} = |q| \frac{R}{L}, \quad q := \frac{3Qb_0 R}{32D_+ k_B T}. \quad (10)$$

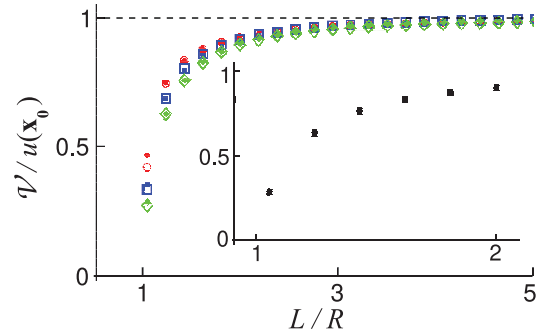


FIG. 3. The ratio  $\mathcal{V}/u(\mathbf{x}_0)$  [Eqs. (6) and (9)] as a function of  $L/R$  for  $\lambda D_2/D_1 = 0.1, 1, 10$  (circle, square, diamond) and  $\eta_2/\eta_1 = 0.1, 10$  (open, filled). The inset provides an enlarged view of the range  $L/R \lesssim 2$  for  $\lambda D_2/D_1 = 10$  and  $\eta_2/\eta_1 = 0.02, 0.1, 1, 10, 50$  (circle, square, diamond, triangle, times); at that scale, the data are indistinguishable. The dashed line indicates the approximation provided by Eq. (9).

The dominance of drift means  $\text{Pe}(L) \gtrsim 1$ . Thus, one estimates the distance  $L_{\text{max}}$  from the interface, beyond which the motion of the particle is controlled by diffusion rather than by drift, as  $\text{Pe}(L_{\text{max}}) = 1$  so that  $L_{\text{max}} = |q|R$ . Focusing on the case  $b_0 > 0$ , one can determine the time  $t_{\text{drift}}$  it takes the particle to reach the interface starting (for  $Q > 0$ ) from a given distance  $L_0$  (or, for  $Q < 0$ , to reach a given distance  $L_0$  starting from near the interface) via straightforward integration of the equation of motion  $dL/dt = \mathbf{e}_z \cdot \mathcal{V}$  (within the overdamped regime [25]). This renders the drift time  $t_{\text{drift}} = t_{\text{diff}}/(2|q|)$  in terms of the time of diffusion  $t_{\text{diff}} := L_0^2/D_p$  over the same distance  $L_0$ . Therefore, for large values of  $|q|$  the drift caused by the Marangoni flow, rather than diffusion, dominates the dynamics of the particle.

In order to estimate the magnitude of  $q$ , we shall use the experimental result that typically  $b_0$  can take values in the range from  $b_0 \sim -10^{-3} \text{ N}/(\text{m} \times \text{M})$  (M denotes mol/liter) for simple inorganic salts in water [38], up to  $b_0 \sim 10^2 \text{ N}/(\text{m} \times \text{M})$  for dilute solutions of surfactants (i.e., far from their critical micelle concentrations) [39]. We consider two distinct setups of potential experimental relevance.

(i) The particle is a source. The chemical species  $A$  is molecular oxygen liberated from peroxide in aqueous solution by a platinum-covered particle. For the experimental conditions described in Ref. [40], one has  $Q/(4\pi R^2) \approx 10^{-3} \text{ mol}/(\text{s} \times \text{m}^2)$  [compare Eq. (1b)]. At room temperature (300 K) and for  $R \approx 1 \mu\text{m}$ , Eq. (10) leads to  $|q| \sim 3 \times 10^{-4} \times [D_+ / (\text{m}^2 \times \text{s}^{-1})]^{-1} [|b_0| / (\text{N} \times \text{m}^{-1} \times \text{M}^{-1})]$ . For an air (fluid 2)-water (fluid 1) interface [41], diffusion in the gas phase dominates (typically  $D_2 \approx 10^{-5} \text{ m}^2/\text{s}$  and  $D_1 \approx 10^{-9} \text{ m}^2/\text{s}$  [42]), and  $D_+ \approx \lambda D_2/2 \approx 10^{-4} \text{ m}^2/\text{s}$  (since  $\lambda \sim 10\text{--}100$  for oxygen and air-water interface [43]), while  $b_0$  is in the lower range of values [40]. Thus  $L_{\text{max}}/R = |q| \sim 10^{-2}$ , which explains the lack of reports of such effects for experimental setups as in Ref. [40]. However, for a liquid-liquid interface (e.g., water-decane), one has  $D_+ \approx D_1 \approx D_2 \approx 10^{-9} \text{ m}^2/\text{s}$  (one expects  $\lambda \lesssim 1$  [42]) and thus  $|q| \gtrsim 10^2$  across the range of values  $b_0$  noted above. Therefore, for the same experimental setups of active colloids, but which involve liquid-liquid interfaces instead of liquid-gas ones, we predict that the effective interactions discussed here dominate. The same conclusion holds for liquid-gas interfaces but with a reaction product with very low solubility in the gas phase (i.e.,  $\lambda \ll 1$ ).

(ii) The particle is a sink; i.e., the tensioactive species  $A$  is absorbed completely by the particle. One can infer  $Q$  from the diffusion-limited regime in which the surface of the particle acts as an absorbing boundary so that  $c(\mathbf{x} \in S_p) = 0$ , which with Eq. (7) provides the estimate  $Q \approx -4\pi D_1 R c_1^\infty$  as  $R/L \rightarrow 0$ . With Eq. (10) one arrives at  $|q| \sim 3 \times 10^8 \times (D_1/D_+) (R/\mu\text{m})^2 (c_1^\infty/\text{M}) [|b_0| / (\text{N} \times \text{m}^{-1} \times \text{M}^{-1})]$ . Therefore,  $L_{\text{max}}/R = |q|$  can indeed be

large for colloidal particles even if species  $A$  is only weakly tensioactive (i.e.,  $|b_0|$  small) and even for liquid-gas interfaces ( $D_1/D_+ \ll 1$ ).

The availability of an exact series representation for  $\mathcal{V}$  as given by Eq. (6) allows us to assess the range of validity of the asymptotic approximation [Eq. (9)] discussed above. For several values of the viscosity and diffusivity ratios [25], Fig. 3 shows  $\mathcal{V}/u(\mathbf{x}_0)$  as a function of the separation  $L/R$ . It turns out that  $u(\mathbf{x}_0)$  provides a reliable approximation (less than 10% relative error) of the exact solution down to separations  $L/R \approx 2$ , i.e., covering most of the range within which the model is relevant. Furthermore, the deviations from  $u(\mathbf{x}_0)$  depend very weakly on the ratios  $\eta_2/\eta_1$  and  $\lambda D_2/D_1$ .

These results, yet to be explored experimentally, have several implications, which we highlight in conclusion. First, as noted in the Introduction, if the particle maintains a temperature gradient, e.g., through local heating, the very same equations hold with the temperature playing the role of the number density  $c(\mathbf{x})$ . Therefore, all the phenomenology discussed above extends to this case, too. Second, sufficiently close to the interface the drift due to the induced Marangoni flows can dominate even self-phoretic motion. For instance, a Janus particle of size  $R = 1 \mu\text{m}$  (with  $D_p \sim 10^{-13} \text{ m}^2/\text{s}$  in water) and self-propelling with a typical velocity of  $\sim 1 \mu\text{m}/\text{s}$  [1] has a Peclet number  $\text{Pe}_{\text{phor}} \approx 10$ , which, e.g., at distances  $L/R < 10$  is smaller than  $\text{Pe}(L) = (R/L)|q|$  if  $|q| \gtrsim 10^2$  [see Eq. (10)]. Third, based on the single-particle phenomenology studied here one can infer potentially significant collective effects. Consider, for example, a dilute suspension of active particles which are driven towards the interface by the Marangoni stresses and in addition experience a short-ranged repulsion by the interface (e.g., due to electrostatic double layer interactions). Then the particles are expected to reside near the interface while experiencing a mutual long-ranged lateral repulsion as each particle is carried by the Marangoni flows induced by the others [see the flow lines in Fig. 2 and Eq. (8b)], which tells that  $u_r$  exhibits also a slow in-plane decay  $\sim 1/r$ . Therefore, near the interface and in the presence of lateral boundaries self-organized crystal-like monolayers could be reversibly assembled and “dissolved” by simply turning on and off the activity of the particles. Finally, we note that this effective lateral pair interaction violates the action-reaction principle because nonidentical particles (e.g., due to size polydispersity, different production rates  $Q$ , or a heterogeneous coverage of the surface) create Marangoni flows of different strength. As for other systems in which such violations occur [44], this feature can be expected to give rise to a complex collective behavior and to a rich, barely explored phenomenology.

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