Controlling Transport in Mixtures of Interacting Particles using Brownian Motors

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An outstanding open problem in nanoscience is how to control the motion of tiny particles. Ratchetlike devices, inspired by biological motors, have been proposed as a way to achieve this goal. However, the net directed transport is almost suppressed if the diffusing particles are weakly coupled to the underlying spatially asymmetric substrate. Here we show how adding particles of an auxiliary species, that interact with both the primary particles of interest and the substrate, provides a controlled enhancement of the flow for both species. These can move either together or in opposite directions, depending upon the strength of the interaction, and whether it is attractive or repulsive.

Brownian motors [1], rectifying nonequilibrium random walks on asymmetric potentials, have recently been proposed for a variety of applications, including the separation of molecules or small particles [1–4], motion control of flux quanta [5], as well as for transport in biological systems—e.g., through ion channels in cell membranes [6]. However, the techniques proposed thus far to regulate these motions are difficult to implement under most experimental circumstances, since the mobile particles are often weakly coupled to the underlying asymmetric substrate, and the substrate itself is not easily tunable.

Here we show *how to control the intensity and even the direction of the net rectified motion by taking advantage of the interaction between particles.* In particular, for binary mixtures of interacting Brownian particles, the motion of *both* fractions of particles can be rectified, even if *only one*, the "active" species (or B particles), experiences a spatially asymmetry substrate potential. If the "passive" species (or A particles) repels the active one, the two species drift in opposite directions at least when interaction is weaker than a certain threshold. This mechanism for particle-motion control is of potential applicability for the *separation of repelling particles* which are differently coupled to an asymmetric substrate (e.g., having the same charges but different magnetic moments or masses). Moreover, for the case of attracting *A–B* interspecies interaction, the rectified active particles can drag along the passive ones. Such an *indirect or mediated* rectification of the random motion of particles which weakly interact with the asymmetric substrate or funnel can be used to deliver or remove passive ingredients from a cell or an artificial microstructure [6]. In addition, the injection of an appropriate density of passive particles can enhance, *in a controllable manner*, the flux of active particles, and vice versa. Furthermore, interparticle interactions play a crucial role also in ratchet transport of *a single* species. We prove that an attract-

ing (repelling) interaction among identical particles can result in the amplification (inversion) of their net current.

Model.—Let us consider a mixture of two species of pointlike Brownian particles A and B diffusing on one-dimensional periodic substrates described by the potentials $\mathcal{U}_A$ and $\mathcal{U}_B$, respectively. Particles of type A interact with one another as well as with the B particles via potentials $\mathcal{W}_{AA}$ and $\mathcal{W}_{AB} (= \mathcal{W}_{BA})$, while $\mathcal{W}_{BB}$ describes the interaction of B pairs. The environment acts independently on each a particle (the index a denotes the species A or B) through a Gaussian random force with zero mean and intensity proportional to the temperature T. Under these very general assumptions, the Langevin equations describing the overdamped diffusion of the particles are $\eta_a dx_{a,i}/dt = -\partial \mathcal{U}_a/\partial x_{a,i} - \sum_j \partial \mathcal{W}_{aA}(x_{a,i} - x_{A,j})/\partial x_{a,i} - \sum_j \partial \mathcal{W}_{aB}(x_{a,i} - x_{B,j})/\partial x_{a,i} + \sqrt{2\eta_a T}\, \xi_a^{(i)}(t)$, where the Boltzmann constant has been set to one, and the two sums are taken over both constituents A and B. Here $x_{a,i}$ is the position of the ith particle of type a and η_a its viscosity. Thermal equilibrium is established through the autocorrelations of the noise functions $\xi_a^{(i)}(t)$: $\langle \xi_a^{(i)}(t)\xi_a^{(j)}(0)\rangle = \delta_{i,j}\delta(t)$, $\langle \xi_A^{(i)}\xi_B^{(j)}\rangle = 0$.

In the Fokker-Planck equation formalism [7], this set of stochastic differential equations is replaced by a set of partial differential equations for the particle distributions F_A, F_B. In the "mean field" (MF) approximation, where all two-particle joint distributions are approximated by the product of the two relevant one-particle distributions, the time evolution of the a-particle distribution F_a is described by the *nonlinear* Fokker-Planck equation: $\eta_a \partial F_a/\partial t = \partial/\partial x\{F_a(\mathcal{U}_a^{\mathrm{mf}})' + TF_a'\}$, with $\prime \equiv \partial/\partial x$. The mean field potential, $\mathcal{U}_a^{\mathrm{mf}} = \mathcal{U}_a + \int \mathcal{W}_{aA}(x - \tilde{x})F_A(\tilde{x})d\tilde{x} + \int \mathcal{W}_{aB}(x - \tilde{x})F_B(\tilde{x})d\tilde{x}$, "dresses" or renormalizes the substrate potential $\mathcal{U}_a(x)$ to account for the effective interaction of an individual particle, located at x, with the nonuniform distribution of the remaining particles of either species.

In order to study the stochastic transport in such a system, we focus here on the so-called "temperature ratchet" [8], where a steady nonequilibrium dynamics is sustained by the oscillating temperature $T(t) = T_0(1 + \epsilon \cos \omega t)$. Our analytical results do not depend on the precise choice of the interparticle potentials, as long as the interaction range is much shorter than the spatial period of the ratchet potential. Thus, in order to have an analytically tractable model, we discard nonlocal interaction effects by adopting a local (e.g., strongly screened) pairwise interaction $\mathcal{W}_{aA}(x) = g_{aA}\,\delta(x)$ and $\mathcal{W}_{aB}(x) = g_{aB}\,\delta(x)$. Accordingly, the above Fokker-Planck equation becomes

$$\eta_a \frac{\partial F_a}{\partial t} = \frac{\partial}{\partial x}\{F_a(\mathcal{U}_a + g_{aA}F_A + g_{aB}F_B)' + TF_a'\}. \quad (1)$$

Stationary distributions.—Next, we consider low frequency or adiabatic temperature oscillations, where the mixture is close to equilibrium at any dimensionless time $\tau \equiv \omega t$. The particle distribution F_a can be separated into an equilibrium distribution $\phi_a^{(0)}$ and a perturbation ψ_a, namely, $F_a = \phi_a^{(0)} + \psi_a$. The time-dependent modulation ψ_a is responsible for the rectification of the stochastic dynamics of the A and B particles and vanishes for $\omega \to 0$. The instantaneous equilibrium solution $\phi_a^{(0)}$ of Eq. (1) satisfies the identity $\phi_a^{(0)}\mathcal{U}_a' + T(\phi_a^{(0)})' + g_{aA}\phi_a^{(0)}(\phi_A^{(0)})' + g_{aB}\phi_a^{(0)}(\phi_B^{(0)})' = 0$ with periodic boundary conditions $\phi_a^{(0)}(x + l) = \phi_a^{(0)}(x)$. Quite naturally, we have assumed that both substrate potentials $\mathcal{U}_A$ and $\mathcal{U}_B$ have equal unit cell l. Note that if the periods l_a of $\mathcal{U}_a$ are different but still commensurate, i.e., $l_A/l_B = N/M$ with N, M integer, then the analytical results presented below are still valid in the extended ratchet cell Ml_A. The equilibrium distributions $\phi_a^{(0)}$ are (non-Boltzmannian) solutions of the transcendental equations:

$$\phi_a^{(0)} \exp\!\left[\frac{g_{aA}\phi_A^{(0)} + g_{aB}\phi_B^{(0)}}{T}\right] = C_a^{(0)} \exp\!\left(-\frac{\mathcal{U}_a}{T}\right), \quad (2)$$

where the constants $C_a^{(0)}$ are determined by the normalization conditions $\int_0^l \phi_a^{(0)} dx/l = n_a$ and n_a denotes the a-particle density.

The nonequilibrium contribution ψ_a satisfies the equation $\eta_a \omega(\dot{\psi}_a + \dot{T}\partial_T\phi_a^{(0)}) = \partial/\partial x[\psi_a(\mathcal{U}_a^{\text{eff}})' + T_a^{\text{eff}}\psi_a' + g_{AB}\phi_a^{(0)}\psi_b' + g_{aA}\psi_a\psi_A' + g_{aB}\psi_a\psi_B']$, where the overdot denotes a τ derivative and $\partial_T \equiv \partial/\partial T$; $b = B$ if $a = A$ and vice versa. The effective potentials and temperatures in the equation above are

$$\begin{aligned}
\mathcal{U}_a^{\text{eff}} &= \mathcal{U}_a + g_{aA}\phi_A^{(0)} + g_{aB}\phi_B^{(0)}, \\
T_a^{\text{eff}} &= T + g_{aa}\phi_a^{(0)}.
\end{aligned} \quad (3)$$

The renormalization of the diffusion constant (or effective temperature) and the potential experienced by the fraction ψ_a of moving particles accounts for their interaction with the remaining particles of *both* species.

Net flows of A and B particles.—The basic mechanism behind the flow rectification induced by temperature oscillations is well illustrated by a simplified model where one subsystem (say, the B particles) is subject to the asymmetric ratchet potential $\mathcal{U}_B$, with one minimum per unit cell, while the other one (the A particles) is not, $\mathcal{U}_A \equiv 0$. This may happen in a mixture of neutral and charged (magnetic and nonmagnetic, heavy and light) particles with the ratchet potential being produced by an electrical (magnetic, gravitational) field. Our goal is thus to control the transport of A particles through their interaction with the B particles.

We now discuss the case when the interaction among particles of the same type is repulsive, i.e., $g_{AA} > 0$, $g_{BB} > 0$. The B particles condense naturally at the minima of the asymmetric substrate potential [see Figs. 1(a) and 1(b), bottom panels]. In addition, if the B particles repel (attract) the A particles, then the latter ones will accumulate in the regions where the density of the B particles is minimum (maximum), that is near the maxima (minima) of the substrate potential $\mathcal{U}_B$. Therefore, the effective potential $\mathcal{U}_A^{\text{eff}}$ for the A particles has opposite spatial asymmetry with respect to $\mathcal{U}_B$, if A and B repel one another, $g_{AB} > 0$, and has the same asymmetry for $g_{AB} < 0$ [see Figs. 1(a) and 1(b), top panels]. (By definition, $\mathcal{U}_B$ and $-\mathcal{U}_B$ are said to have opposite asymmetry). During the lower temperature half cycle, all particles are confined tighter around the minima of the relevant effective potential, while, during the higher temperature half cycle, the particles of both species diffuse more easily out of the $\mathcal{U}_A^{\text{eff}}$, $\mathcal{U}_B^{\text{eff}}$ potential wells. Moreover, if a minimum of the potential $\mathcal{U}_a^{\text{eff}}$ is closer to an adjacent maximum on the right than on the left side, then the fluctuating a particles diffuse with higher probability to the right than to the left during one full temperature oscillation. As the asymmetry of the ratchet potentials $\mathcal{U}_A^{\text{eff}}$ and $\mathcal{U}_B^{\text{eff}}$ for repelling A and B particles is opposite, so is the orientation of their currents. On the contrary, two attracting A and B species drift in the same direction. This implies that the transport of both particle species can be effectively and *separately controlled* by regulating their densities n_A and n_B *without* the need of any *tunable* asymmetric potential substrate.

As the interaction among particles of the same type was assumed to be repulsive, the potential wells of $\mathcal{U}_B^{\text{eff}}$ ($\mathcal{U}_A^{\text{eff}}$) tend to flatten out when increasing the corresponding density n_B (n_A). This results in the decay of the associated ratchet current J_B (J_A). In contrast, the ratchet asymmetry and current of one species may be enhanced by increasing the density of the other mixture component. For instance, in the case of A–B attractive forces, the A particles tend to concentrate in the regions of higher B densities, that is around the minima of $\mathcal{U}_B$, thus attracting even more B particles and making the $\mathcal{U}_B^{\text{eff}}$ potential wells deeper. In short, one can enhance the transport of the "target"
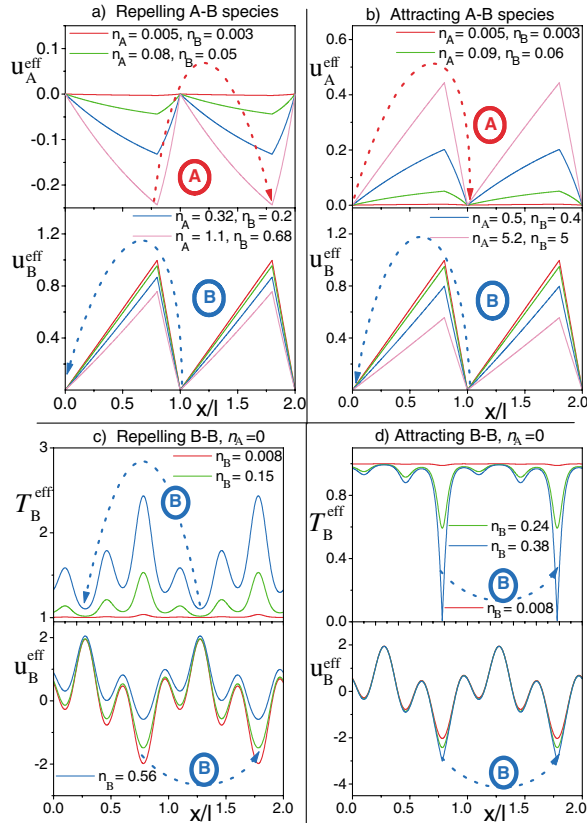
FIG. 1 (color). Panels (a), (b): Spatial dependence of the effective potentials $\mathcal{U}_A^{\text{eff}}$, $\mathcal{U}_B^{\text{eff}}$ at different densities of the A and B particles. In both panels, particles of the same type repel one another; the interaction between particles of different species is repulsive in (a) and attractive in (b). There is no "bare" potential for the A particles, $\mathcal{U}_A \equiv 0$, whereas the ratchet potential $\mathcal{U}_B(x)$ is piecewise linear: $\mathcal{U}_B = \mathcal{U}^{\text{linear}}$, where $\mathcal{U}^{\text{linear}}(0 < x/l < l_1/l) \equiv Qx/l_1$ and $\mathcal{U}^{\text{linear}}(l_1/l < x/l < 1) \equiv Q(l - x)/(l - l_1)$, and here $Q = 1$, $l_1 = 0.8$. The other coupling parameters are $g_{AA} = g_{BB} = |g_{AB}| = 1$ and $T = 1$. Panels (c),(d): Spatial dependence of the effective temperature T_B^{eff} and potential $\mathcal{U}_B^{\text{eff}}$ for an assembly of repelling (c) and attracting (d) identical particles of the B type. Here the substrate ratchet potential is $\mathcal{U}_B = \mathcal{U}^{\sin} \equiv \sin(2\pi x/l) + a_1 \sin(4\pi x/l - p_1) + a_2 \sin(6\pi x/l - p_2)$ with $a_1 = 0.1$, $a_2 = -1$, $p_1 = 0$, and $p_2 = 0.6$. Moreover, $g_{BB} = 1$ in (c), -0.3 in (d), for $T = 1$.

particles by adding a certain amount of auxiliary particles.

All these properties can be reproduced using a standard perturbation technique [7] which can be generalized to the set of nonlinear equations (1) [9]. The ensuing drift velocities $V_a = \langle J_a \rangle / n_a$ are functions of both particle densities, n_A and n_B (Fig. 2). Both drift velocities are of the same order of magnitude; V_A and V_B have the same sign for attracting species [Fig. 2(a), panels I, II] and opposite sign for repelling A and B particles [Fig. 2(b), I, II]. To show more clearly the dependence of the ratchet transport of one species on the density of its own constituents as well as on the density of the other species in
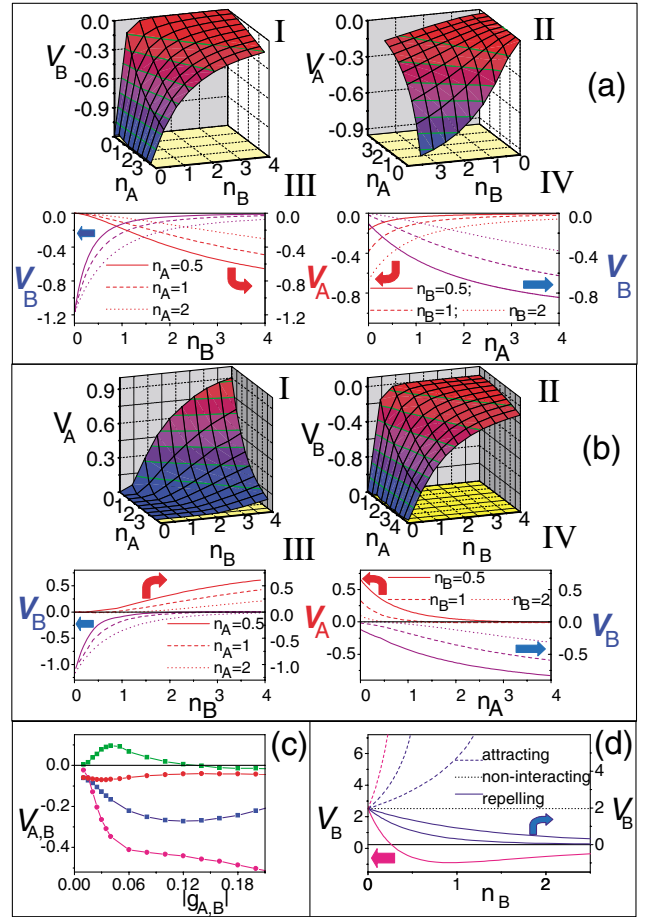

FIG. 2 (color). The net velocities V_A and V_B in units of $\tilde{V} = 10^{-4} \eta a^2 \omega^2 l^3 / (2T_0)$ with $\eta_A = \eta_B = \eta$, as a function of the particle densities n_A and n_B for attractive (a) and repulsive (b) A-B interactions; the interaction among particles of the same type is repulsive in both cases; $\mathcal{U}_A = 0$, $\mathcal{U}_B = \mathcal{U}^{\text{linear}}$, parameters are the same as in Fig. 1. The 3D panels I and II in (a) and (b) display the surfaces $V_{B,A}(n_A, n_B)$, a few sectional views of which are shown in III (fixed n_A, varying n_B) and IV (fixed n_B, varying n_A). (c) $V_{B,A}$ versus the interspecies interaction g_{AB} obtained through molecular dynamics for the parameter values in [10]. (d) The net velocity V_B of attracting and repelling identical B particles ($n_A = 0$) for the ratchet potentials $\mathcal{U}_B = \mathcal{U}^{\sin}$ (see Fig. 1) with parameters $a_1 = 0.1$, $a_2 = 0$, $p_1 = 0.45$, and $p_2 = 0$ (blue curves), and $a_1 = 0.1$, $a_2 = -1$, $p_1 = 0$, and $p_2 = 0.6$ (magenta curves). Interaction constant takes $g_{BB}/T = \pm 0.5$, ± 0.2 (blue curves) and $g_{BB}/T = \pm 0.3$ (magenta curves).

the mixture, sectional views of the 3D surfaces $V_a(n_A, n_B)$ are displayed in panels III and IV of Figs. 2(a) and 2(b). The behaviors shown are perfectly consistent with our earlier predictions based on simple effective potential arguments.

To assess the validity of the rectification mechanism above, we performed molecular dynamics simulations for a mixture of active B and passive A particles subject to a pulsated temperature [10] (so as to enhance the rectified flows). Consistent with our predictions, attracting active and passive particles move along in the same direction

[Fig. 2(c), blue and magenta dots]; moreover, both species speed up as the coupling constant $|g_{AB}|$ increases. Vice versa, two repelling species drift in the opposite direction [Fig. 2(c), red and green dots] as expected in our analysis for sufficiently small values of g_{AB}. However, on further increasing g_{AB}, the net velocity of the passive species inverts its direction after hitting a maximum. The physical origin of such a nonperturbative effect can be traced back to the pushing (dragging) exerted by the ratcheted B particles on the otherwise freely moving A particles. This result is still consistent with the overall goal of using the B particles to control the As.

Interacting particles of one species.—Now we consider a system consisting of particles of one type only (here, B particles). Accordingly, we take the limit $n_A \rightarrow 0$ in the analytical results of Eqs. (2) and (3) . The effective temperature and potential for repelling ($g_{BB} > 0$) and attracting ($g_{BB} < 0$) B particles are shown in Fig. 1(c) and 1(d). Repelling particles tend to expel one another from the wells of the effective potential, thus lowering its confining barriers. This evidently suppresses the drift velocity V_B. Moreover, the minima of the effective temperature function $T_B^{\text{eff}}(x)$ coincides with the maxima of the effective potential $\mathcal{U}_B^{\text{eff}}(x)$; the two functions have now opposite asymmetry. As the nonequilibrium contribution ψ_B of the distribution F_B tends to peak around the potential and temperature minima, there exist two competing mechanisms in the dynamics of the B particles, determined, respectively, by the gradient of the effective potential and of the effective temperature. The particles condensed around the minima of the effective potential tend to jump to the right, while the "nonequilibrium" particles localized close to the temperature minima jump in average to the left [schematically shown in Fig. 1(c) by the dashed arrows]. The competition between these two opposite drives may result in the current inversions of Fig. 2(d) (magenta curve). The drift velocity V_B for an assembly of repelling particles is plotted in Fig. 2(d) (solid curves) for different choices of the substrate $\mathcal{U}_B(x)$.

The case of an assembly of attracting B particles, $g_{BB} < 0$, is also remarkable. The effective potential $\mathcal{U}_B^{\text{eff}}$ and temperature T_B^{eff} have the same asymmetry, as the position of their maxima and minima coincide [Fig. 1(d)]. Moreover, particles condensing around the minima of $\mathcal{U}_B$ attract more particles, thus making the effective potential wells deeper; hence, the effective asymmetry increases and so does the rectification power of the system. This prediction agrees well with our calculations based on the adiabatic approach: The net velocity V_B drastically increases with increasing the particle density n_B. However, our perturbation technique becomes untenable for all frequencies as soon as the particle density approaches the critical density $n_B = n_{\text{crit}}(T)$, corresponding to a zero effective temperature at the minimum x_0 of the substrate potential $\mathcal{U}_B$; that is, $T_B^{\text{eff}}(x_0) = T + g_{BB}\phi_B^{(0)}(x_0) = 0$. For $n_B > n_{\text{crit}}$, there is no solution $\phi_B^{(0)}(x)$ of Eq. (2) for the equilibrium B-particle distribution. Analytical evidence indicates that the excess particles condense in the liquid phase at the minima of $\mathcal{U}_B$. As the density of the mobile particles is independent of n_B for $n_B > n_{\text{crit}}(T)$, the current J_B saturates or, correspondingly, the drift velocity at large n_B decays.

Conclusions.—The currents of two species of interacting particles can be *separately* controlled by varying their densities. This is potentially useful for enhancing and regulating the transport through, e.g., synthetic ion channels [6], by adding auxiliary species, and for the separation of repelling particles [4], e.g., different isotopes. Moreover, the opposite motion of repelling particles found here could be related to the great selectivity of ion channels which, for instance, push potassium K^+ out while retaining sodium Na^+. Also, the "indirect" rectification of stochastic motion allows us to deliver target particles, which cannot directly feel any asymmetric potential, by using intermediate particles which interact with both the target ones and the substrate.

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[1] P. Reimann, Phys. Rep. **361**, 57 (2002); R. D. Astumian and P. Hänggi, Phys. Today **55**, No. 11, 33 (2002); R. D. Astumian, Science **276**, 917 (1997); F. Jülicher, A. Ajdari, and J. Prost, Rev. Mod. Phys. **69**, 1269 (1997).

[2] J. Rousselet *et al.*, Nature (London) **370**, 446 (1994).

[3] C. Keller, F. Marquardt, and C. Bruder, Phys. Rev. E **65**, 041927 (2002), and references therein.

[4] P. T. Korda *et al.* Phys. Rev. Lett. **89**, 128301 (2002).

[5] J. Wambaugh *et al.*, Phys. Rev. Lett. **83**, 5106 (1999); C. J. Olson *et al.*,*ibid.* **87**, 177002 (2001); C. S. Lee *et al.*, Nature (London) **400**, 337 (1999); S. Savel'ev and F. Nori, Nature Mater. **1**, 179 (2002).

[6] Y. Jiang *et al.*, Nature (London) **417**, 515 (2002); Z. Siwy and A. Fuliński, Phys. Rev. Lett. **89**, 198103 (2002).

[7] H. Risken, *The Fokker-Planck Equation* (Springer, New York, 1984).

[8] P. Reimann *et al.*, Phys. Lett. A **215**, 26 (1996).

[9] The perturbation terms $\psi_a(x)$ of the distribution functions $F_a(x)$ for both species were expanded in powers of ω, $\psi_a = \sum_{m=1}^{\infty}(\eta_a\omega)^m\phi_a^{(m)}$, with time and space periodic conditions $\phi_a^{(m)}(x + l, \tau) = \phi_a^{(m)}(x, \tau + 2\pi) = \phi_a^{(m)}(x, \tau)$ and normalization $\int_0^l \phi_a^{(m)}dx = 0$. To second order, we get the net currents for both species.

[10] The interaction potentials used for the simulations are $\mathcal{W}_{ab}(x) = g_{ab}(\lambda - |x|)/\lambda^2$ for $0 < |x| < \lambda$, and 0 for $|x| > \lambda$, with $\lambda = 0.1l$, a and b denote either A or B. The periodic temperature function $T(t)$ adopted here is $T(t) = 0.3$ for $0 \leq t < 0.9(2\pi/\omega)$ and $T(t) = 40$ for $0.9(2\pi/\omega) \leq t < (2\pi/\omega)$ with $\omega = 4\pi$. The substrate force acting on each active B particle is $f_s(x_B) = -Q/l_1$ for $0 \leq x_B < l_1$ and $Q/(l - l_1)$ for $l_1 \leq x_B < l$ with $Q = 1$, $l = 1$, and $l_1 = 0.9$. Other simulation parameters are $g_{AA} = g_{BB} = 0.005$, $n_A = 50$, and $n_B = 3$. The MF approximation is valid when $n_A\lambda$ and $n_B\lambda \gg 1$.