Current fluctuations of interacting active Brownian particles

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We derive the distribution of particle currents for a system of interacting active Brownian particles in the long time limit using large deviation theory and a weighted many body expansion. We find the distribution is non-Gaussian, except in the limit of passive particles. The non-Gaussian fluctuations can be understood from the effective potential the particles experience when conditioned on a given current. This potential suppresses fluctuations of the particles orientations and surrounding density, aligning particles and reducing their effective drag. From the distribution of currents, we compute the diffusion coefficient, which is in excellent agreement with molecular dynamics simulations over a range of self-propulsion velocities and densities. We show that mass transport is Fickian in that the diffusion constant determines the response of a small density gradient, and that nonlinear responses are similarly computable from the density dependence of the current distribution.

Persistent currents are the hallmark of a system driven away from equilibrium. One of the simplest and most fundamental problems of nonequilibrium physics is to predict the structure of the fluctuations of currents around a nonequilibrium steady-state and to decode the microscopic information contained in them. Non-equilibrium fluctuation-dissipation relations [1–7], fluctuation theorems [8–14], and thermodynamic uncertainty relations [15–17] are notable examples of successes towards this end. Much of this progress has been underpinned by the study of large deviation functions (LDFs), which supplies a general framework to compute and characterize fluctuations of extensive observables [18, 19]. The LDF of the current can be viewed as the analog of a free energy, making relationships between fluctuations and response to external perturbations transparent [20–22]. However, the evaluation of LDFs for interacting systems remains challenging. In this letter, we characterize the fluctuations of currents in a system of interacting active Brownian particles (ABPs) and show how these fluctuations encode the response of the system.

ABPs are a simple model of active matter, a class of systems that convert energy from the environment into directed motion. ABPs evolve nonequilibrium steady states as they break detailed balance at the single particle level due to a constant nonconservative driving force. More than just being non-Boltzmann, their steady-states support unique phenomena such as motility induced phase separation [23]. Laboratory realizations of ABPs include cellular biopolymers [24–26], bacteria [27–33], and synthetic colloids [34–38]. We derive the current LDFs for ABPs and validate it with molecular simulation. We find that small current fluctuations are Gaussian, and the associated linear response obeys Fick’s law, as has been shown for noninteracting ABPs [39]. Large current fluctuations are non-Gaussian and the associated nonlinear response results from a change in the particle’s orientational correlations, which we characterize with the effective potential that renders those fluctuations typical.

We consider a collection of $N$ ABPs in two spatial dimensions, whose positions and orientations are denoted $\mathbf{r}^N = \{ \mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N \}$ and $\mathbf{\theta}^N = \{ \theta_1, \theta_2, \ldots, \theta_N \}$, respectively. These dynamical variables are coupled through their equations of motion, which for the position of the $i$th ABP is

$$\dot{\mathbf{r}}_i(t) = \mathbf{F}_i[\mathbf{r}^N(t)] + v_o \mathbf{e}[\mathbf{\theta}(t)] + \eta_i(t), \quad (1)$$

and for its corresponding orientation is

$$\dot{\theta}_i(t) = \eta_\theta(t), \quad (2)$$

where the dot denotes time derivative, $v_o$ is the magnitude of the self-propulsion velocity, $\mathbf{e}[\theta(t)] = \{ \cos(\theta), \sin(\theta) \}$ is the unit vector on a circle. The Gaussian random variables, $\eta_i(t)$, satisfy $\langle \eta_i(t) \rangle = 0$ and $\langle \eta_i(t) \rangle \eta_j(t') = 2D \delta(t - t')$, where $\langle \cdot \rangle$ denotes ensemble average. The particles interact with a pairwise additive force, $\mathbf{F}_i[\mathbf{r}^N] = \sum_j F(r_{ij}) \hat{\mathbf{r}}_{ij}$, where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$. $F(r)$ is assumed to be short-ranged and repulsive, and $\hat{\mathbf{r}}$ denotes unit vector. In simulations of this model, $F(r)$ is derived from a WCA potential [40] with characteristic length and energy scales set to 1. We assume no slip boundary conditions, so that the rotational, $D_\theta$, and translational, $D_t$, diffusion constants are related through, $D_t = 3D_i$, and set $D_i = 1$ which determine a set of dimensionless length and time units.

The time integrated current for particle $i$ is defined as

$$\mathbf{J}_i = \frac{1}{t} \int_0^t dt' \mathbf{r}_i(t') = \frac{\mathbf{r}_i(t) - \mathbf{r}_i(0)}{t}, \quad (3)$$

where the observation time, $t$, is assumed to be large. The total current for all $N$ particles in the system is $\mathbf{J}^N = \{ \mathbf{J}_1, \ldots, \mathbf{J}_N \}$. To characterize the statistics of $\mathbf{J}^N$...
in the long time limit, we aim to compute its LDFs. We define a generating function,
\[ \hat{P}(\lambda, r^N, \theta^N, t) = \int dJ^N P(r^N, \theta^N, J^N, t) e^{\lambda \cdot J^N} \]  
(4)
with \( P(r^N, \theta^N, J^N, t) \) being the joint distribution of observing all of the particles in a particular position, orientation, and total integrated current, at time \( t \). The vector \( \lambda \) is conjugate to the current vector and exponentially reweights \( P(r^N, \theta^N, J^N, t) \).

The time evolution of the generating function is given by
\[ \frac{\partial \hat{P}(\lambda, r^N, \theta^N, t)}{\partial t} = L^N_\lambda \hat{P}(\lambda, r^N, \theta^N, t) \]  
(5)
that defines the Lebowitz-Spohn operator \([19, 41, 42]\). This operator has two pieces, \( L^N_\lambda = L^N_0 + \Delta L^N_\lambda \), where
\[ L^N_0 = \sum_{i=1}^{N} (F_i [r^N(t)] + v_o e [\theta_i(t)] + D_i \nabla_i) \cdot \nabla_i + D_i \partial^2_{\theta_i} \]  
(6)
is conservative and whose adjoint gives the Fokker Planck operator, and the piece dependent on \( \lambda \)
\[ \Delta L^N_\lambda = \sum_{i=1}^{N} (F_i [r^N(t)] + v_o e [\theta_i(t)] + 2 D_i \nabla_i + D_i \lambda) \cdot \lambda \]  
(7)
does not conserve probability. The spectrum of \( L^N_\lambda \) is generally complex, but its largest eigenvalue is guaranteed to be real, and whose dependence on \( \lambda \) yields the cumulant generating function (CGF) for the current.

Within this framework we can naturally describe two limiting cases. First, we can consider the statistics of the total system current defined as the sum over the individual particle currents, by setting \( \lambda = \lambda \cdot 1 \) where \( \lambda \) is a scalar parameter and \( 1 \) the identity. However, this case is trivial because the sum of the interparticle force in Eq. 7 vanishes, decoupling the equation into a sum of \( N \) independent equations. In this case, the total current CGF is equivalent to \( N \) times the CGF for a single ABP. Alternatively, we can consider the current statistics of a single tagged ABP, subject to the interactions of the surrounding particles. This is done by setting \( \lambda \) to be a vector with a single nonzero element, \( \lambda = \{0, 0, 0, \cdots, \lambda, \cdots, 0, 0, 0\} \). This second case contains the first in the limit of low density, and provides additional information on the dependence of current fluctuations on interactions. In the following we will consider the second definition.

In order to calculate the CGF for a tagged particle, we first introduce a weighted many body expansion that follows from a BBGKY-like hierarchy \([43]\). Specifically, we define an \( n \)-particle reduced generating function
\[ \hat{P}^{(n)}(\lambda, r^n, \theta^n, t) = \frac{N!}{(N-n)!} \int d\theta^{(N-n)} d\theta^{(N-n)} \hat{P}(\lambda, r^N, \theta^N, t), \]  
(8)
which, when introduced into Eqs. 5-7, will result in a set of coupled evolution equations for different \( \hat{P}^{(n)} \)'s. We can close the single particle equation with the two-particle generating function, decomposed as
\[ g_\lambda(r, \theta, r', \theta', t) = \frac{\hat{P}^{(2)}(\lambda, r, \theta, r', \theta', t)}{\hat{P}^{(1)}(\lambda, r, \theta, t) \hat{P}^{(1)}(\lambda, r', \theta', t)} \]  
(9)
where \( g_\lambda(r, \theta, r', \theta', t) \) is the pair distribution function conditioned on a given current through \( \lambda \) \([44, 45]\). This function can be simplified when the system is in a homogeneous steady-state and assuming that it does not depend on the difference in orientations between particles. In this limit, \( g_\lambda(r, \theta, r', \theta', t) \approx g_\lambda(r, \phi) \), where \( \phi \) is the angle of the displacement vector of two particles relative to the orientation of the particle at the origin. This closure to the many-body hierarchy was introduced previously for the case of \( \lambda = 0 \) \([46-49]\).

The equation of motion for the single particle generating function will depend on the average interparticle force. We can decompose this force into components in the parallel and perpendicular direction of the self-propulsion, however this will result in an average force that depends on both the relative angle between the interparticle displacement vector and the tagged particle’s orientation. Following Speck et al.\([46]\) if we approximate the component perpendicular to the orientation as that parallel to the surface of the particle, this will uncouple these two terms allowing for the expansion to be closed. This approximation to the perpendicular force is exact in the limit of passive particles, where there are no orientational correlations, and is numerically accurate when the parallel component is much larger than the perpendicular component, as occurs for \( v_o > 1 \).

As we consider only homogeneous systems, for notational simplicity and without loss of generality we restrict our attention to currents in just the \( x \) direction. Under these assumptions, we obtain the evolution operator for the single particle generating function for currents,
\[ L_\lambda = V_\lambda(\rho) \cos(\theta) (\partial_x + \lambda) + D_\lambda(\rho)(\partial_x + \lambda)^2 + D_\lambda \partial^2_\theta, \]  
(10)
that has the same drift-diffusion form as an independent ABP, but with renormalized effective propulsion speed, \( V_\lambda(\rho) \), and translational diffusion constant, \( D_\lambda(\rho) \), where \( \rho \) is the local density, which in the homogeneous assumption is taken as the bulk density. The adjoint of the operator in Eq. 10 evaluated at \( \lambda = 0 \) yields the propagator for the single particle density. Both \( V_\lambda(\rho) \) and \( D_\lambda(\rho) \) in principle depend on \( \rho \), \( v_o \), and \( \lambda \), through the state-dependent pair correlation function. Within this force decomposition, \( D_\lambda(\rho) \) is the diffusion coefficient for a system of interacting passive Brownian particles and we have found that for the conditions we study, \( D_\lambda(\rho) \) can be approximated by the mean field form, \( D_\lambda(\rho) \approx D_\lambda(1 - \rho) \) \([48]\). The effective propulsion speed takes the form \( V_\lambda(\rho) = v_o - \rho \xi_\lambda(\rho) \) where \( \rho \xi_\lambda(\rho) \) is an
effective drag. This drag is given by an integral over the interparticle force

$$\zeta_\lambda (\rho) = \int_0^\infty \! dr \int_0^{2\pi} \! d\phi \, r \cos(\phi) g_\lambda (r, \phi) F(r)$$

(weighted by the pair distribution function. This coefficient describes the decrease in the effective velocity of a tagged particle due to the increased density of impenetrable particles in the direction of self-propulsion\[46]. In the following, we take \(\rho \zeta_\lambda (\rho)\) as input for our evaluation of the CGF, though simple approximations to \(\zeta_\lambda (\rho)\) exist in specific limits \[50\].

Using these definitions, we are able to solve for the CGF for this effective single particle description of the system by the largest eigenvalue of the equation \(\lambda \nu_\lambda = \psi(\lambda) \nu_\lambda\), with \(\psi(\lambda)\) being the CGF and \(\nu_\lambda\) its corresponding right eigenvector. The solution is given by the zeroth characteristic function of Mathieu’s equation \[51\], with a representation for small \(\lambda\) given by the expansion,

$$\psi(\lambda) = D_t (\rho) \lambda^2 + D_r \left[ z_\lambda^2 (\rho) \right] - \frac{7 z_\lambda^4 (\rho)}{32} + \frac{29 z_\lambda^8 (\rho)}{144} + O(\lambda^8)$$

with \(z_\lambda (\rho) = V_\lambda (\rho) \lambda / D_r\). The CGF is symmetric about \(\lambda = 0\) as a consequence of spatial inversion symmetry and retains all even powers, alternating in sign. For passive particles \(z_\lambda (\rho) = 0\), and \(\psi(\lambda)\) reduces to that for Brownian motion with an effective diffusion constant, \(D_\xi (\rho)\). In the limit that the particles are non-interacting, or \(\rho \to 0\), our results reduce to those obtained previously \[52\], and additionally gives the CGF for the center of mass motion.

Given the CGF, the rate function for current fluctuations can be computed from the Legendre-Fenchel transform, \(I(J) = \max_\lambda [\lambda J - \psi(\lambda)]\), where \(I(J)\) is minus the logarithm of the probability of \(J\) divided by the observation time. Figure 1 shows the rate functions computed from simulation and from \(\psi(\lambda)\). For a variety of different \(\rho\)’s and \(v_o\)’s we find quantitative agreement between the analytical result and the simulations. Small fluctuations around \(J = 0\) are Gaussian as expected, but larger fluctuations are markedly non-Gaussian, revealing fluctuations that are more rare than anticipated from the time intensive variance, \(t(J^2)\). The deviations from Gaussian behavior become more distinct with increasing \(v_o\), as highlighted in Fig. 1 by scaling the current by \(\sqrt{t(J^2)}\). Within the scale presented, increasing \(\rho\) with fixed \(v_o\) does not affect the shape of the distribution more than changing the scale of fluctuations through the variance.

We can gain insight into the shape of \(I(J)\) by constructing an auxiliary process that generates the same ensemble of trajectories in the long time limit as the original model conditioned on a given current \[18\]. The auxiliary process is a transformation of the Lebowitz-Spohn operator,

$$\mathcal{L}_\lambda = \nu_\lambda (\theta)^{-1} L_\lambda \nu_\lambda (\theta) - \psi(\lambda)$$

$$= L_0 + 2D_t(\rho) \lambda \partial_x + 2D_r \partial_t \ln \nu_\lambda (\theta) \partial_\theta$$

which leaves the diffusion terms unmodified, but adds additional drift terms in that restore normalization. The auxiliary process for a tagged particle is

$$\dot{r}(t) = F [r^N(t)] + v_o e [\theta (t)] + 2D_t(\rho) \lambda \dot{x} + \eta_t (t)$$

where the added force is a constant proportional to \(\lambda\) in the \(x\) direction. The equation of motion for the orientation includes a force, \(F_\lambda (\theta) = 2D_r \partial_\theta \ln \nu_\lambda (\theta)\), which for small \(\lambda\) is

$$\dot{\theta}(t) = -2V_\lambda (\rho) \lambda \sin(\theta) + \eta_\theta (t)$$
Derivatives of $\psi(\lambda)$ provide the cumulants of $J$, $d^n\psi(\lambda)/d\lambda^n = C^n_0$, with the first, $C^1_0 = \langle J \rangle_\lambda$, yielding the average current, and the second, $C^2_2 = t\langle (J - \langle J \rangle_\lambda)^2 \rangle_\lambda$, its variance. When evaluated at $\lambda = 0$, these are cumulants of the original model, but for $\lambda \neq 0$, these report on rare fluctuations into the tails of $I(J)$. Shown in Figs. 2c) and d), are the average currents computed at finite $\lambda$, from the exact solution of the eigenvalue equation and from evaluating Eq. 3 directly from simulations of the auxiliary process defined in Eqs. 14 and 15. For small $\lambda$, the current increases linearly from 0 with a slope set by variance at $\lambda = 0$, as is expected from linear response. For large $\lambda$, the system exhibits nonlinear response, manifesting the non-Gaussian current fluctuations. In this limit, the slope decreases dramatically. The asymptotic limit of this secondary response is given by an offset of $v_o$ and slope that depends only on the $D_1(\rho)$. The origin of this dependence is clear from the auxiliary process. For large $|\lambda|$, $V_\lambda(\rho) \to v_o$, and the force on the orientation suppresses angular fluctuations. Analysis of this limit provides an asymptotic form for $\psi(\lambda)$,

$$\psi(\lambda) = D_1(\rho)\lambda^2 \pm v_o\lambda, \quad \lambda \to \pm \infty,$$

which shows that the tails of the $I(J)$ are given by an effective Gaussian with mean, $v_o$, a much smaller variance than at $\lambda = 0$. We note a simple effective temperature mapping between Brownian particles and ABPs would not explain the observed non-Gaussian fluctuations or concomitant secondary response [56–59]. Further, this qualitative behavior of two different effective diffusion constants for small and large fluctuations has been observed recently in active biopolymers [60, 61].

We conclude with a discussion of the second cumulant,

$$C^2_0(\rho) = 2D_1(\rho) + \frac{V_\lambda^2(\rho)}{D_r} \equiv 2D(\rho),$$

that we define as a twice a collective diffusion constant, $D(\rho)$. In Fig. 3, numerical results obtained from simulations of the mean-squared displacement are plotted in excellent agreement with predictions from Eq. 17. This form of the diffusion coefficient has been shown to agree with simulations previously, and was derived by a moment expansion of the joint position and orientation distribution [23, 62–64]. This density dependence of $D(\rho)$ was shown by others [46, 47, 65] to correctly predict the spinodal instability signaling the onset of motility induced phase separation [50].

The current fluctuations encoded by $D(\rho)$ provide the response of a hydrodynamic current, $J_\rho$, generated from a slowly varying spatial density, $\rho(x)$. From the Kramer’s Moyal expansion [60], $J_\rho$ can be generally expressed as a gradient expansion

$$J_\rho = -\sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \partial_x^{-1} M^n[\rho(x)]\rho(x) \quad (18)$$
where $M^n[\rho(x)]$ is the local density-dependent $n$th moment of the current, $\langle (J - \langle J \rangle)^n \rangle$. To first order, the mass current is linear in the density gradient and is given by Fick’s law, $J_\rho \approx - D(\rho) \frac{\partial \rho}{\partial x}$, where $D(\rho)$ is the proportionality constant relating the current to the gradient resulting from identifying the second moment with the second cumulant. Since for small average currents we have $\langle J \rangle_\lambda \approx 2D(\rho)\lambda$, which shows that at linear response, $\lambda$ can be related to an affinity for this nonequilibrium system. We have computed $D(\rho)$ from $-J_\rho/(\partial \rho/\partial x)$ by simulating an open channel in contact with two reservoirs. As shown in Fig. 3, we find good agreement with $D(\rho)$ computed in this way and from $\psi(\lambda)$. From $\psi(\lambda)$, we have access to all moments of $J$, and together with its $\rho$ dependence this framework allows us to quantify higher order responses [50] that are not naturally considered in standard field theoretic treatments of ABPs [46–48, 57, 62].

While our focus has been on ABPs, the framework we have presented is general and allows for the quantification of current fluctuations, and the calculation of transport coefficients for continuous interacting systems. For ABPs, we found that large current fluctuations near the mean are not representative of rarer fluctuations which are restricted as a result of coherent active movement. These specific results are consistent with deviations from Gaussian behavior that have been reported in recent experimental studies of active colloids [67, 68]. Furthermore, there are a number biological systems are modeled by ABPs, such as cellular biopolymers that exhibit two types of transport characterized by two different diffusion constants for small and large fluctuations [60, 61]. Our analysis solution may help understand these observations. Finally, while we have focused on current fluctuations, our development of the weighted many body expansion provides a way to calculate the CGF of other relevant quantities for nonequilibriums systems such as activity [45, 55, 69–72], entropy production [73–76] and density that are currently difficult to estimate.

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Supplementary Information

SIMULATIONS DETAILS

In all the simulations, the system of ABPs interacted through the WCA potential[1] given by

\[ U(r) = \begin{cases} 
4\epsilon \left( \left( \frac{\sigma}{r} \right)^{12} + \left( \frac{\sigma}{r} \right)^{6} \right) + \epsilon & r < 2^{1/6}\sigma \\
0 & r \geq 2^{1/6}\sigma 
\end{cases} \]

where we set the energy scale, \( \epsilon \), and lengthscale, \( \sigma \), to be 1. From this potential the force is given by \( F(r) = -\nabla U(r) \). All simulation results are represented in Lennard Jones units with times in units of \( \tau_{\text{LJ}} = \sigma^2/(D_{\text{LJ}}) \) and currents, \( J \), in units of \( \sigma/\tau_{\text{LJ}} \). Our simulations are in two-dimensions with a domain of 100\( \sigma \) x 100\( \sigma \) and periodic boundary conditions. We used particle numbers of \( N = 1000, 3000, \) and 5000, which corresponds to densities of \( \rho = 0.1, 0.3, \) and 0.5. We used a second order Stochastic Runge Kutta algorithm[2] with a timestep of \( \delta t = 10^{-5}\tau_{\text{LJ}} \). All data presented were computed with 2-3 independent simulations, with a total simulation time between 500-5000 \( \tau_{\text{LJ}} \), including 200 \( \tau_{\text{LJ}} \) of equilibration.

The current was estimated by

\[ J_i = \frac{r_i(\Delta t) - r_i(0)}{\Delta t} \]  \hspace{1cm} (1)

which is the total displacement of particle \( i \) over the observation time, \( \Delta t \). The observation time is chosen to be in the long time limit where all current distributions for that time and larger all collapse onto the same distribution. We find \( \Delta t = 10\tau_{\text{LJ}} \) for all conditions studied. The numerical distributions in Fig. 1 of the main text were obtained by taking the bin edges of the histogram of currents. The analytical results were calculated by taking the numerical Legendre transform of the CGF, \( \psi(\lambda) \). We calculate the CGF using a Fourier-Bloch decomposition of the Lebowitz-Spohn operator. We find that using a basis of 25 cosine functions is sufficient to converge \( \psi(\lambda) \) for all conditions studied.[3]

In Fig. 2a), we show the calculation of the effective force given by \( F_\lambda = 2D_{\rho} \partial_\rho \ln \nu_\lambda(\theta) \), where \( \nu_\lambda(\theta) \) is the right eigenvector of the CGF. This effective force is fit to a sum of sines to get a functional form and then used in the equations of motion to generate an auxiliary dynamics that realize the large currents [4]. We integrate these auxiliary equations of motion and measure the current according to Eq. 1. The average currents from simulations are compared with the derivative with respect to \( \lambda \) of the CGF, \( \psi(\lambda) \). The diffusion coefficient is estimated by measuring the mean-squared displacement in the diffusive regime:

\[ D(\rho) = \frac{1}{4N} \sum_{i=1}^{N} \frac{|r_i(\Delta t) - r_i(0)|^2}{\Delta t} \]  \hspace{1cm} (2)

with \( \Delta t = 10\tau_{\text{LJ}} \).

In order to compute the diffusion coefficient from an imposed density gradient, a long channel connecting two large reservoirs was constructed out of static WCA particles. The dimensions of each reservoir was 100 by 150 in the \( x \) and \( y \) direction, and the channel was 200 by 100, as determined by an insensitivity of the computed diffusion constant to the system geometry. An initial density gradient was generated by initializing the reservoirs with densities \( \pm 5\% \) of the mean target density in the channel, which in all cases was well within the linear response regime. After an initial transient period, a steady-state current was evaluated by counting the number of particles transfer from one reservoir to the other per time per width of the channel. Statistics were accumulated over a simulation time where the current was linearly proportional to time, typically 3000 \( \tau_{\text{LJ}} \). The gradient was measured away from the entrance of the channel and the diffusion constant was computed by the ratio of the gradient to the flux.

CALCULATION OF \( \zeta_\lambda(\rho) \)

To evaluate \( \zeta_\lambda(\rho) \) we use molecular simulations and a generalized variational principle[5]. To do this, we first run the auxiliary dynamics with an approximate effective potential derived with \( \zeta_0(\rho) \) for \( \lambda \) values ranging from 0 to 20. Then, we recalculate \( \zeta_\lambda(\rho) \) using an approximate \( g_\lambda(r, \phi) \) computed from these auxiliary dynamics and Eq. 11. We fit
FIG. 1. Dependence of the pair distribution function on $\lambda$. Pair distribution function for $\lambda = 0$ (left), and $\lambda = 20$ (right) for $v_o = 10$ and $\rho = 0.5$.

the $\zeta_\lambda(\rho)$ as a function of $\lambda$, which is then incorporated into the Lebowitz-Spohn operator (Eq. 10 in the main text). This operator is re-diagonalized and its largest eigenvector is used to obtain a better estimate of the effective potential. This process is iterated until $\zeta_\lambda(\rho)$ is self-consistent, typically requiring only 2 or 3 iterations. This condition does not ensure an optimal estimate of the CGF, as the Lebowitz-Spohn operator is not Hermitian, but it does enforce stationarity.

The drag coefficient has a number of known limiting forms. By construction, for independent particles or $\rho \to 0$, the drag $\rho \zeta_\lambda(\rho) \to 0$. We find that for all $v_o$, this approach is linear in $\rho$. Similarly, for $v_o \to 0$, the system becomes isotropic and $\zeta_\lambda(\rho) \to 0$. For large $v_o$, $\zeta_0(\rho) \approx v_o/\rho^*$ where $\rho^* \approx 1.2$, corresponding to a effective closed-packing density. We find that for $v_o > 30$, this approximation is within 1%, but even for $v_o \approx 5$, this form is within 10% of the computed value.

COMPARISON OF PAIR DISTRIBUTION FUNCTION AT FINITE $\lambda$

Shown in Fig. 1 are the pair distribution functions, $g_\lambda(r, \phi)$, computed at $\lambda = 0$ and $\lambda = 20$ for $v_o = 10$ and $\rho = 0.5$. At $\lambda = 0$, $g_0(r, \phi)$ is anisotropic, meaning that there are more particles in front of a tagged particle and less behind. This imbalance and the bow wave structure of the correlations result in a nonzero value of $\zeta_\lambda(\rho)$. This is the same coefficient that has been calculated previously and describes the linear decrease of the hydrodynamic velocity with density due to interactions [6–8]. At large $\lambda$, $g_\lambda(r, \phi)$ becomes uniform in $\phi$ and accordingly $\zeta_\lambda(\rho) = 0$. In addition, there is a reduction in the intensity of the peaks, most significantly in the first peak that drops from a value of 4.2 to 1.5. These correlations at large $\lambda$ are like those of a low density system of passive Brownian particles, which have no angular dependence, despite the significant net particle drift in one direction.

RELATION BETWEEN $\lambda$ AND AFFINITY

If we include the density dependence of the effective diffusion constant, then we arrive at an expression for the hydrodynamic current,

$$ J_\rho \approx -\left[ \frac{\rho V_0(\rho)}{D_r} \frac{\partial V_0(\rho)}{\partial \rho} + D(\rho) \right] \frac{\partial \rho}{\partial x} \quad (3) $$

which includes an extra term omitted in the expression in the main text for Fick’s law. We have neglected the density dependence of $D_t(\rho)$, because it is small compared $V_0(\rho)$. The first term on the right is negative, so this expression provides a prediction for phase separation used previously[6–10]. We can define a density dependent effective chemical potential, $\mu_0(\rho)$, by rewriting the hydrodynamic current as

$$ J_\rho = -D(\rho)\rho \frac{d\mu_0(\rho)}{dx} \quad (4) $$

where

$$ \mu_0(\rho) = \ln(\rho V_0^2(\rho)) \quad (5) $$
which acts an affinity driving the hydrodynamic current. By taking the first derivative of the CGF with respect to \( \lambda \), the average current for small \( \lambda \) is

\[
\langle J \rangle_\lambda = 2D(\rho)\lambda + O(\lambda^3)
\]

so within the linear response regime, \( \lambda \) is related to an affinity by

\[
\lambda = -\frac{\rho}{2} \frac{d\mu_0(\rho)}{dx}
\]

and is the same result that could be obtained from the Gallavotti Cohen symmetry of the CGF. More generally, away from linear response, \( \lambda \) and the affinity are still related, but this relation is more complicated\[11\].

**NONLINEAR HYDRODYNAMIC TRANSPORT**

We can continue the Kramers Moyal expansion to the next non-zero term, which would include the fourth moment, \( M^4 = C^4_0 - 3(C^2_1)^2 \). To this order, the hydrodynamic current would be given by

\[
J_\rho = -D(\rho)\rho \left( \frac{d\mu_0(\rho)}{dx} + \frac{d\mu_1(\rho)}{dx} \right)
\]

which includes an additional contribution to the effective chemical potential, \( \mu_1(\rho) \) given by

\[
\mu_1(\rho) = \alpha(\rho) \left( \frac{d\rho}{dx} \right)^2 + \beta(\rho) \frac{d^2\rho}{dx^2},
\]

with \( \alpha(\rho) \) and \( \beta(\rho) \) being coefficients that depend on the density, \( \nu_0 \), \( D_t \) and \( D_r \). Keeping the first order in the derivatives of the density we get

\[
\alpha(\rho)D(\rho)\rho = \frac{(-7 + 4D_r)}{4D_r^3} \left[ V^3_0(\rho) \frac{\partial V_0}{\partial \rho} + \frac{3V^2_0(\rho)\rho}{2} \left| \frac{\partial V_0}{\partial \rho} \right|^2 \right]
\]

\[
+ \frac{V_0(\rho)}{Dr} \left[ \frac{2V_0(\rho)}{Dr} + \frac{\rho}{Dr} \frac{\partial V_0}{\partial \rho} \right]
\]

\[
\beta(\rho)D(\rho)\rho = \frac{1}{2} + \frac{\partial D(\rho)}{\partial \rho} + \frac{(-7 + 4D_r)}{8D_r^3} \left[ \frac{V^2_0(\rho)}{4} + \frac{\rho V^3_0(\rho) \partial V_0}{8} \right]
\]

which shows that our framework allows us to quantify higher order response with the next order response containing non-gradient terms that would give rise to interfacial energy-like terms necessary to stabilize two phases, \( d^2\rho/dx^2 \) and \( |d\rho/dx|^2 \)[6, 7, 12].

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