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ABSTRACT

We generalize power functional theory [Schmidt and Brader, J. Chem. Phys. **138**, 214101 (2013)] to Brownian many-body systems with orientational degrees of freedom. The framework allows the study of active particles in general inhomogeneous and time-dependent nonequilibrium. We prove for steady states that the free power equals half the negative dissipated external work per time, and is hence trivially related to the average forward swim speed of the particles. The variational theory expresses the free power as a functional of the microscopic one-body density and current distribution. Both fields are time-, position- and orientation-dependent, and the total current consists of translational and rotational parts. Minimization of the free power functional with respect to the current(s) yields the physical dynamics of the system. We give a simple approximation for the superadiabatic (above adiabatic) contribution which describes excess dissipation in homogeneous bulk fluids due to drag. In steady states, we evaluate the free power using Brownian dynamics simulations for short-ranged soft repulsive spheres. We describe the necessary sampling strategies and show that the theory provides a good account of the simulation data.

I. INTRODUCTION

The class of active systems covers a wide variety of biological and physical systems. Activity refers to an intrinsic motility of the individual units, such as cells, bacteria or particular colloids. The growing interest in active systems over the last decade is poignantly illustrated by the large number of review articles that address the topic.[1–18] A variety of collective phenomena has been addressed in e.g. systems of microorganisms,[2,19] cells,[20] and bacteria.[6,8,21–30] Further examples include the collective motion of flocks,[1,31–33] school of fish,[34] and opinion formation in social science.[35]

One of the most successful model to describe collective behavior is the Viscek model,[36] and its variations.[37–45] The Viscek model has been used for example to study lane formation,[39,43,46] soft deformable particles,[47] order-disorder transition,[37] and swarming turbulence.[41] A popular application is the description of active nematics. For active rods the collective properties[48,49] and swarming behavior[50] were studied. In active nematics velocity correlations,[51] orientational order and fluctuations have been investigated.[52–54]

There exists a variety of experiments and applications, such as active glasses and gels[55–60] and the collective motion of vibrated polar disks and granular materials.[61–64] Experiments dealing with active colloidal particles use e.g. Janus particles[65,66] or other colloids whose surface suitably is manipulated. Janus particles are built by e.g. by coating one hemisphere of SiO_2 beads with a thin layer of graphite onto one hemisphere.[65] When illuminated with a widened laser, the light is absorbed by the graphite hemisphere. This locally heats up the solvent above the critical temperature, causing a local demixing, which generates a phoretic force that propels the Janus particle. In Ref. 67 a polymer sphere encapsulated most of an anti-ferromagnetic cube, which was then only partially exposed to the solvent. Illumination with blue

light caused the hydrogen peroxide solvent to dissociate and as a result the colloids to self-propel and form clusters. A variety of further propulsion mechanisms is based on chemical gradients.[66,68,69]

A body of work has been based on hydrodynamic frameworks,[7] addressing the behavior of flocks,[70] phase coexistence,[71] pattern formation,[72] confined collective motion,[73] and microorganisms.[2] Furthermore hydrodynamic approaches give the opportunity to study e.g. self-propulsion mechanisms,[74,75] synchronization of anisotropic particles,[76–78] and dynamics near a wall.[79]

Active Brownian particles (ABP) form a simple microscopic model for active matter. These types of particles undergo Brownian motion with a built-in orientation degree of freedom, which itself diffuses freely, and gives the direction of the self-propulsion. In particular spherical particles serve as a minimal model for active colloids. The ABP model is very popular for studying phase separation of active particles, which is based on the particles' motility. This motility-induced phase separation (MIPS) occurs between non-equilibrium steady states in systems with purely repulsive interparticle interactions. The simplicity of the ABP model has led to a thriving number of publications based on computer simulations and theoretical approaches to describe MIPS.[80–103] Recent theoretical developments include a mode coupling theory for ABP,[104–106] a field theory for phase separation,[107] and how swimmer-swimmer correlations affect the collective behavior of active suspensions.[108]

Approaches for describing MIPS were based on continuum theory[95,96] and a hydrodynamic, coarse-graining theory.[82,94] In both approaches the average propulsion speed as a function of bulk density, $v(\rho_b)$, plays an important role. Using continuum theory[95,96] $v(\rho_b)$ is obtained by microscopic estimations of a random walk hindered by collisions. The authors of Refs. 95 and 96 argue that using $v(\rho_b)$ it is possible to construct an effective free energy, which predicts the existence of phase separation. Furthermore they present a formalism which allows the detailed study of phase separation dynamics. The approach introduced in Refs. 82 and 94 is based on the microscopic many-body Smoluchowski equation. The authors derive coarse-grained equations for one tagged particle, in which again an effective (density-dependent) swimming speed $v(\rho_b)$ enters. From the effective hydrodynamics the authors identify an instability region of the homogeneous system, causing a dynamic instability. The fact that this indeed leads to phase separation can be demonstrated by comparison to computer simulations. Recent work showed[109] that $v(\rho_b)$ can also be obtained by Green-Kubo relations, which might in a next step be used as an input for coarse-grained frameworks. Summarizing, both theoretical approaches are able to predict the onset and existence of MIPS (with reasonable approximations), but a detailed description of stable phase coexistence is still missing.

Closely related to MIPS is the clustering of self-propelled particles and active clusters.[28,39,65,66,81,110–118] The kinetics of the formation of the dense phase can be modeled analogously to classical nucleation theory.[119] Supported by simulations, it has been shown that, within this modeling, some properties, such as the location of the binodal and nucleation rates, can be obtained. Another approach is the active phase-field crystal model.[120,121]

Given the number of applications and phenomenological observations, the attempt at unification via formulating thermodynamics and statistical mechanics for active matter seems well justified.[29,84,101,122–132] Under special investigation is the possibility to find an equation of state and a closed form for the pressure in active systems.[89,126,133–139] In particular, in the context of MIPS the surface tension of the gas-liquid interface in systems with phase coexistence has been of significant interest.[140–142] Recent results suggest that the interfacial tension is negative,[140] which violates the physical intuition earned from equilibrium statistical mechanics.

A fundamentally different approach is the effective equilibrium description of ABPs. The basic idea behind this framework is to eliminate the orientational degrees of freedom by integrating them out, resulting in a Langevin equation for non-Markovian dynamics for the translational coordinates.[143] The underlying stochastic process is an Ornstein-Uhlenbeck process. There are two concepts which allow a representation of this process using effective Markovian dynamics: the unified colored noise approximation (UCNA) introduced by Hänggi and Jung,[144,145] and the Fox approximation.[146,147] Applying the former to ABPs allows the study of a variety of problems, e.g. statistical properties such as the velocity distribution of ABPs.[148] Furthermore the UCNA is also used as a first step to formulate a statistical mechanics theory[124] and describe critical phenomena.[149] The Fox approximation is used to study the physics of ABPs.[139,143,150,151] Within this approach an approximated Fokker-Plank equation is derived, whereby one can define an effective interaction between the particles, due to activity.[143] The existence of such an effective interaction has been proposed earlier, because of the similarities between MIPS and equilibrium phase transitions.[152] The application of Fox's approximation[143,150] and the UCNA[153] gave an explicit formulation. Interfacial properties, pressure and tension have been studied as well in both approximations.[139,141,154] Very recent work by Wittmann *et al.*[155,156] nicely shows the equivalence of both approaches and gives insight into previous results. The authors present new findings regarding interaction forces, phase equilibria, structure, and mechanical properties.

In this work we present a theory for active Brownian particles with orientational degrees of freedom. Our approach is based on the recently developed power functional theory (PFT).[157] This framework allows the description of many-body systems that follow over-damped Brownian dynamics (BD). These are described by a unique power functional $R_t[\rho, \mathbf{J}]$ of the one-body density distribution ρ and the one-body current distribution $\mathbf{J}$, which both depend on position $\mathbf{r}$ and time t.[157] Minimizing the power functional with respect to the current gives the physical time evolution of the system. Therefore R_t can be regarded as an analogue to the free energy functional in equilibrium statistical physics. One central significance of the power functional is that its derivative determines the forces acting in overdamped systems. Therein lies the second analogy to equilibrium systems, where the thermodynamic

potentials appear also as abstract quantities that are only detectable through their derivatives. The theory is generally formulated in the Smoluchowski picture, starting from the many-body probability distribution Φ.

Dynamical density functional theory (DDFT) is a widely used approach, e.g. for studying sheared systems,[158,159] spinodal decomposition,[160] and systems with orientational degrees of freedom.[161] DDFT can be viewed as an extension to density functional theory (DFT) to nonequilibrium systems.[162,163] However, in contrast to DDFT, PFT is formally exact in the sense that no adiabatic assumption is involved in order to describe the time evolution of the density profile. Hence PFT goes beyond the DDFT description and allows the study of additional forces in the system that are not contained in the adiabatic construction. These forces are called superadiabatic forces and are accessible via computer simulations.[164–166] Superadiabatic forces, and hence PFT, may serve therefore as a tool to describe nonequilibrium phenomena which are not fully understood, such as the laning transition in colloidal systems,[167,168] where DDFT only fits when adding phenomenological terms.[169,170] Moreover PFT potentially gives opportunities to describe a whole class of nonequilibrium system from first principles in a general and unified way. Furthermore PFT was also formulated for quantum[171] and classical Hamiltonian[172] many-body systems. For an overdamped Brownian system, nonequilibrium Ornstein-Zernike equations were formulated for two-body dynamic correlation functions.[173,174] Much recent progress has been made in the development of PFT approximations for simple fluids[175,176] and in corresponding computer simulation techniques.[177,178,180]

Recently PFT has been used for the description of ABPs[179] by taking the orientational degree of freedom of the particles into account, which can be viewed as a generalization of the PFT for mixtures.[181] Here we give a complete account of the theory and present further comparisons to computer simulation results. We also derive several exact sum rules for nonequilibrium steady states, Eqs. (43), (44), and (49) below.

The paper is organized as follows: In Sec. II we formulate power functional theory for active particles. We start from the microscopic (Smoluchowski) many-body description of the active system and show the representation of PFT on the one-body level in Sec. II A. We then focus on steady states and derive an exact non-equilibrium sum rule for the splitting into internal and external contributions to the free power in Sec. II B. We give an approximation for the excess dissipation in Sec. II C and formulate the Langevin dynamics of the ABP model in Sec. III. Furthermore we give details about the Brownian dynamics computer simulations, and the external power in the Langevin description is presented. In Sec. IV we present our results obtained by simulations and theory. We conclude in Sec. V.

II. POWER FUNCTIONAL THEORY

A. General framework

We consider N active particles with position coordinates $\{\mathbf{r}_1, \ldots, \mathbf{r}_N\} \equiv \mathbf{r}^N$ in a d-dimensional space, and orientations $\{\boldsymbol{\omega}_1, \ldots, \boldsymbol{\omega}_N\} \equiv \boldsymbol{\omega}^N$, where particle i at position $\mathbf{r}_i$ swims with speed s in direction $\boldsymbol{\omega}_i$, with $|\boldsymbol{\omega}_i| = 1$. We consider possibly anisotropic inter-particle interaction potentials $u(\mathbf{r}^N, \boldsymbol{\omega}^N)$. The Smoluchowski equation for the time-dependent probability distribution $\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t)$ of an ensemble of such systems is

$$\frac{\partial}{\partial t}\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t) = -\sum_i (\nabla_i \cdot \hat{\mathbf{v}}_i + \nabla_i^{\omega} \cdot \hat{\mathbf{v}}_i^{\omega})\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{1}$$

where ∇_i is the derivative with respect to $\mathbf{r}_i$, and ∇_i^{ω} is the derivative with respect to $\boldsymbol{\omega}_i$ (acting on the unit sphere); $\hat{\mathbf{v}}_i$ and $\hat{\mathbf{v}}_i^{\omega}$ are the translational velocity and rotational velocity operators, respectively. The former is given by

$$\gamma\hat{\mathbf{v}}_i = -(\nabla_i u) - (\nabla_i v_i^{\mathrm{ext}}) + \mathbf{X}_i + \gamma s\boldsymbol{\omega}_i - k_B T\nabla_i, \tag{2}$$

where $u(\mathbf{r}^N, \boldsymbol{\omega}^N)$ is the interparticle interaction potential, $v^{\mathrm{ext}}(\mathbf{r}, \boldsymbol{\omega}, t)$ is a position-, orientation-, and time-dependent external potential, $\mathbf{X}(\mathbf{r}, \boldsymbol{\omega}, t)$ is an external non-conservative force; we use $v_i^{\mathrm{ext}} = v^{\mathrm{ext}}(\mathbf{r}_i, \boldsymbol{\omega}_i, t)$ and $\mathbf{X}_i = \mathbf{X}(\mathbf{r}_i, \boldsymbol{\omega}_i, t)$ as a shorthand notation; γ is the friction coefficient for translational motion, s = constant is the swimming speed of an isolated particle, k_B is the Boltzmann constant and T is absolute temperature. The vector fields $\nabla_i u$ and $\nabla_i v_i^{\mathrm{ext}}$ act via multiplication in (1); only the thermal diffusive term [last contribution in (2)] acts via differentiation. Having the nonconservative force field $\mathbf{X}(\mathbf{r}, \boldsymbol{\omega}, t)$ in (2) can be useful for modeling e.g. shear flow.[175,176]

The rotational velocity operator is given by

$$\gamma^{\omega}\hat{\mathbf{v}}_i^{\omega} = -(\nabla_i^{\omega} u) - (\nabla_i^{\omega} v_i^{\mathrm{ext}}) + \mathbf{X}_i^{\omega} - k_B T\nabla_i^{\omega}, \tag{3}$$

where γ^{ω} is the rotational friction coefficient, and $\mathbf{X}_i^{\omega} \equiv \mathbf{X}^{\omega}(\mathbf{r}_i, \boldsymbol{\omega}_i, t)$, where $\mathbf{X}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t)$ is a non-conservative external torque field.

Following the procedure of Ref. 157, we introduce variational fields $\tilde{\mathbf{v}}^N \equiv \{\tilde{\mathbf{v}}_1, \ldots \tilde{\mathbf{v}}_N\}$ and $\tilde{\mathbf{v}}^{\omega N} \equiv \{\tilde{\mathbf{v}}_1^{\omega}, \ldots \tilde{\mathbf{v}}_N^{\omega}\}$ for the translational and rotational velocity, respectively. Each variational field is a function in configuration space, i.e. $\tilde{\mathbf{v}}_i \equiv \tilde{\mathbf{v}}_i(\mathbf{r}^N, \boldsymbol{\omega}^N, t)$ and $\tilde{\mathbf{v}}_i^{\omega} \equiv \tilde{\mathbf{v}}_i^{\omega}(\mathbf{r}^N, \boldsymbol{\omega}^N, t)$ for all $i = 1, \ldots, N$.

We define a generator that depends on the trial fields via

$$\mathcal{R}_t = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \left[\gamma\left(\frac{\tilde{\mathbf{v}}_i^2}{2} - \tilde{\mathbf{v}}_i \cdot \hat{\mathbf{v}}_i\right) + \gamma^{\omega}\left(\frac{(\tilde{\mathbf{v}}_i^{\omega})^2}{2} - \tilde{\mathbf{v}}_i^{\omega} \cdot \hat{\mathbf{v}}_i^{\omega}\right)\right] \times \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t). \tag{4}$$

Due to its quadratic structure, the generator is instantaneously (i.e. at fixed time t and fixed distribution Φ) minimized by the true value of each of the trial fields, and hence when evaluated at the minimum

$$\frac{\delta\mathcal{R}_t}{\delta\tilde{\mathbf{v}}_i} = 0, \tag{5}$$

$$\frac{\delta\mathcal{R}_t}{\delta\tilde{\mathbf{v}}_i^{\omega}} = 0, \tag{6}$$

which implies that

$$\tilde{\mathbf{v}}_i\Phi = \hat{\mathbf{v}}_i\Phi, \tag{7}$$
$$\tilde{\mathbf{v}}_i^{\omega}\Phi = \hat{\mathbf{v}}_i^{\omega}\Phi \tag{8}$$

at the given time t. Hence the trial fields at the minimum of the functional can "stand in" for the action of the corresponding operators. Clearly the minimum corresponds to the physical dynamics, as the trial fields possess the "correct" values that determine the actual time evolution. Here the translational contribution to the power functional takes on the value

$$\mathcal{R}_t^{0,\mathrm{trans}} = -\frac{\gamma}{2}\int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \tilde{\mathbf{v}}_i^2 \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{9}$$

where the superscript 0 indicates the value at the minimum. The total value of $\mathcal{R}_t$ at the minimum consists of translational and rotational contributions:

$$\mathcal{R}_t^0 = -\frac{1}{2}\int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \left(\gamma \tilde{\mathbf{v}}_i^2 + \gamma^{\omega}(\tilde{\mathbf{v}}_i^{\omega})^2\right)\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{10}$$

[As an aside, this formulation is analogous to the one given in Ref. 157 for spheres. Note that the operator expression $\hat{\mathbf{v}}_i\Phi$ in (4) is analogous to that in (5) of Ref. 157 by observing that $\hat{\mathbf{v}}_i\Phi = \gamma^{-1}\mathbf{F}_i^{\mathrm{tot}}\Phi$, with the configuration space expression $\mathbf{F}_i^{\mathrm{tot}}$ for the total force acting on particle i.]

We can further use $\mathcal{R}_t$ as a generator for the one-body fields of interest, via functional differentiation,[157]

$$\frac{\delta \mathcal{R}_t}{\delta \mathbf{X}(\mathbf{r}, \boldsymbol{\omega}, t)} = \mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t), \tag{11}$$
$$\frac{\delta \mathcal{R}_t}{\delta \mathbf{X}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t)} = \mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t), \tag{12}$$

where the left hand side is evaluated at the physical dynamics, cf. (7) and (8), after the derivative has been taken. Here the translational and rotational one-body currents are defined, respectively, via

$$\mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t) = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\delta^{\omega}(\boldsymbol{\omega} - \boldsymbol{\omega}_i)\hat{\mathbf{v}}_i\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{13}$$

$$\mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t) = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\delta^{\omega}(\boldsymbol{\omega} - \boldsymbol{\omega}_i)\hat{\mathbf{v}}_i^{\omega}\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{14}$$

where $\delta(\cdot)$ indicates the d-dimensional Dirac distribution and $\delta^{\omega}(\cdot)$ is the Dirac distribution on the unit sphere.

The one-body currents are related to the temporal change of the one-body density via the continuity equation

$$\frac{\partial \rho(\mathbf{r}, \boldsymbol{\omega}, t)}{\partial t} = -\nabla \cdot \mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t) - \nabla^{\omega} \cdot \mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t), \tag{15}$$

as can be shown from integrating (1) over the degrees of freedom of N − 1 swimmers. Here the one-body density distribution is defined via

$$\rho(\mathbf{r}, \boldsymbol{\omega}, t) = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\delta^{\omega}(\boldsymbol{\omega} - \boldsymbol{\omega}_i)\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t). \tag{16}$$

In order to connect the many-body theory with the one-body level, we perform a constrained Levy search[182,183] as

$$\mathrm{R}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}] = \min_{\tilde{\mathbf{v}}^N, \tilde{\mathbf{v}}^{\omega,N} \to \rho, \mathbf{J}, \mathbf{J}^{\omega}} \mathcal{R}_t, \tag{17}$$

where the constraints are obtained by replacing the operators on the right hand sides of Eqs. (13) and (14) by their respective trial fields, i.e.

$$\mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t) = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\delta^{\omega}(\boldsymbol{\omega} - \boldsymbol{\omega}_i)\tilde{\mathbf{v}}_i\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{18}$$

$$\mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t) = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\delta^{\omega}(\boldsymbol{\omega} - \boldsymbol{\omega}_i)\tilde{\mathbf{v}}_i^{\omega}\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t). \tag{19}$$

The equalities (18) and (19) represent constraints on the trial velocities, as the left hand side is considered to be prescribed. In equilibrium systems, the method of constrained search provides an alternative to the more familiar Mermin-Evans foundation of density functional theory.[162,184] The advantages of the Levy method are that no Legendre transform is required and that the intrinsic free energy functional is given as an explicit (many-body) expression.

As a consequence of the constrained search (17), the variational principle is now elevated to the one-body level, such that both

$$\frac{\delta \mathrm{R}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}]}{\delta \mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t)} = 0, \tag{20}$$
$$\frac{\delta \mathrm{R}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}]}{\delta \mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t)} = 0, \tag{21}$$

hold at the minimum of the functional. Here the (partial) functional derivatives are performed at fixed $\rho(\mathbf{r}, \boldsymbol{\omega}, t)$. As we will show below, (20) and (21) constitute a force balance and a torque balance equation, which together with the continuity equation (15), completely determine the dynamics. One advantage of this setup is that the different contributions to the total force and total torque can be systematically formulated. In particular, one can identify the genuine nonequilibrium contributions as being superadiabatic, i.e. above free energy contributions, as we will show in the following.

The structure laid out so far implies that the intrinsic and external contributions to the total power functional can be separated according to

$$\mathrm{R}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}] = \mathrm{W}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}] - \mathrm{X}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}] \tag{22}$$

where $\mathrm{W}_t[\rho, \mathbf{J}, \mathbf{J}^{\omega}]$ is an intrinsic contribution, solely dependent on the interparticle interactions $u(\mathbf{r}^N, \boldsymbol{\omega}^N)$. The external power is generated from the external forces and torques (both of which act local in time and space) according to

$$X_t[\rho,\mathbf{J},\mathbf{J}^\omega] = \int d\mathbf{r}d\omega \left[\mathbf{J}\cdot(-\nabla v^{\text{ext}} + \mathbf{X} + \gamma s\boldsymbol{\omega}) + \mathbf{J}^\omega\cdot(-\nabla^\omega v^{\text{ext}} + \mathbf{X}^\omega)\right]. \quad (23)$$

We next split the intrinsic contribution in (22) into ideal and excess (over ideal) parts

$$W_t = W_t^{\text{id}} + W_t^{\text{exc}}, \quad (24)$$

where the intrinsic ideal (i.e. in a system with no internal interactions) power functional is given by

$$W_t^{\text{id}}[\rho,\mathbf{J},\mathbf{J}^\omega] = \int d\mathbf{r}d\omega\left[\frac{\gamma\mathbf{J}^2+\gamma^\omega(\mathbf{J}^\omega)^2}{2\rho} + k_BT\left(\mathbf{J}\cdot\nabla + \mathbf{J}^\omega\cdot\nabla^\omega\right)\ln\rho\right]. \quad (25)$$

Here the right hand side can be split into a sum

$$W_t^{\text{id}}[\rho,\mathbf{J},\mathbf{J}^\omega] = P_t^{\text{id}}[\rho,\mathbf{J},\mathbf{J}^\omega] + \dot{F}_{\text{id}}[\rho], \quad (26)$$

with contributions due to dissipation, P_t^{id}, and adiabatic (reversible) ideal free energy changes, $\dot{F}_{\text{id}}[\rho]$. Here

$$F_{\text{id}}[\rho] = k_BT\int d\mathbf{r}d\omega\rho(\mathbf{r},\boldsymbol{\omega})(\ln(\rho(\mathbf{r},\boldsymbol{\omega})\Lambda^d) - 1) \quad (27)$$

is the intrinsic Helmholtz free energy functional of an ideal gas of uniaxial rotators in d spatial dimensions; Λ is the (irrelevant) thermal de Broglie wavelength, and the overdot in (26) indicates a derivative with respect to time. Explicitly, the dissipative and reversible ideal intrinsic contributions are given by

$$P_t^{\text{id}}[\rho,\mathbf{J},\mathbf{J}^\omega] = \int d\mathbf{r}d\omega\,\frac{\gamma\mathbf{J}^2+\gamma^\omega(\mathbf{J}^\omega)^2}{2\rho}, \quad (28)$$

$$\dot{F}_{\text{id}}[\rho]/(k_BT) = \int d\mathbf{r}d\omega(\mathbf{J}\cdot\nabla + \mathbf{J}^\omega\cdot\nabla^\omega)\ln(\rho\Lambda^d), \quad (29)$$

where (29) is obtained from (27) via the chain rule, replacing the partial time derivative of the density distribution $\dot{\rho}$ via the continuity equation (15), and integration by parts in both position and orientation.

The functional derivatives of the ideal intrinsic contribution (25) are then obtained as

$$\frac{\delta W_t^{\text{id}}}{\delta\mathbf{J}(\mathbf{r},\boldsymbol{\omega},t)} = \frac{\gamma\mathbf{J}(\mathbf{r},\boldsymbol{\omega},t)}{\rho(\mathbf{r},\boldsymbol{\omega},t)} + k_BT\nabla\ln\rho(\mathbf{r},\boldsymbol{\omega},t), \quad (30)$$

$$\frac{\delta W_t^{\text{id}}}{\delta\mathbf{J}^\omega(\mathbf{r},\boldsymbol{\omega},t)} = \frac{\gamma^\omega\mathbf{J}^\omega(\mathbf{r},\boldsymbol{\omega},t)}{\rho(\mathbf{r},\boldsymbol{\omega},t)} + k_BT\nabla^\omega\ln\rho(\mathbf{r},\boldsymbol{\omega},t), \quad (31)$$

where the arguments of $W_t^{\text{id}}[\rho,\mathbf{J},\mathbf{J}^\omega]$ have been omitted for clarity. The variational principle (20) and (21) can be then cast into the form of a force balance and a torque balance equation, which are given, respectively, by

$$\frac{\gamma\mathbf{J}(\mathbf{r},\boldsymbol{\omega},t)}{\rho(\mathbf{r},\boldsymbol{\omega},t)} = \gamma s\boldsymbol{\omega} - k_BT\nabla\ln\rho(\mathbf{r},\boldsymbol{\omega},t) - \frac{\delta W_t^{\text{exc}}}{\delta\mathbf{J}(\mathbf{r},\boldsymbol{\omega},t)} - \nabla v^{\text{ext}}(\mathbf{r},\boldsymbol{\omega},t) + \mathbf{X}(\mathbf{r},\boldsymbol{\omega},t), \quad (32)$$

$$\frac{\gamma^\omega\mathbf{J}^\omega(\mathbf{r},\boldsymbol{\omega},t)}{\rho(\mathbf{r},\boldsymbol{\omega},t)} = -k_BT\nabla^\omega\ln\rho(\mathbf{r},\boldsymbol{\omega},t) - \frac{\delta W_t^{\text{exc}}}{\delta\mathbf{J}^\omega(\mathbf{r},\boldsymbol{\omega},t)} - \nabla^\omega v^{\text{ext}}(\mathbf{r},\boldsymbol{\omega},t) + \mathbf{X}^\omega(\mathbf{r},\boldsymbol{\omega},t). \quad (33)$$

In order to describe the contribution due to internal interactions, we assume a splitting of the intrinsic excess functional into adiabatic and superadiabatic (above "adiabatic," i.e. equilibrium) contributions,

$$W_t^{\text{exc}}[\rho,\mathbf{J},\mathbf{J}^\omega] = \dot{F}^{\text{exc}}[\rho] + P_t^{\text{exc}}[\rho,\mathbf{J},\mathbf{J}^\omega], \quad (34)$$

where the total time derivative of the intrinsic excess (over ideal gas) Helmholtz free energy functional $F^{\text{exc}}[\rho]$, which is due to the intrinsic interaction potential $u(\mathbf{r}^N,\boldsymbol{\omega}^N)$, is

$$\dot{F}_{\text{exc}}[\rho] = \int d\mathbf{r}d\omega\left(\mathbf{J}(\mathbf{r},\boldsymbol{\omega},t)\cdot\nabla + \mathbf{J}^\omega(\mathbf{r},\boldsymbol{\omega},t)\cdot\nabla^\omega\right)\frac{\delta F_{\text{exc}}[\rho]}{\delta\rho(\mathbf{r},\boldsymbol{\omega},t)}, \quad (35)$$

and $P_t^{\text{exc}}[\rho,\mathbf{J},\mathbf{J}^\omega]$ is the superadiabatic contribution, which also originates from the internal interactions, and describes the difference to the equilibrium physics. $P_t^{\text{exc}}[\rho,\mathbf{J},\mathbf{J}^\omega]$ depends in general non-locally in time and in space on its (functional) arguments. The functional derivative in (35) is taken with respect to an equilibrium density distribution $\rho_{\text{eq}}(\mathbf{r},\boldsymbol{\omega})$ (which is independent of time), as is appropriate for the equilibrium excess free energy functional $F[\rho_{\text{eq}}]$. This functional derivative is then evaluated at the time-dependent density, i.e. at $\rho_{\text{eq}}(\mathbf{r},\boldsymbol{\omega}) = \rho(\mathbf{r},\boldsymbol{\omega},t)$. Note that this identity defines the adiabatic state.[164] Similar to the derivation of (29), one obtains (35) from applying the chain rule, replacing $\dot{\rho}$ via (15) and then "reversing" the action of the nabla operators by partial integration, i.e. building the adjoint operator. We re-emphasize that the occurrence of the dependence on the instantaneous density distribution, namely in the adiabatic contributions $\dot{F}_{\text{id}}$ and $\dot{F}_{\text{exc}}$ does not constitute an approximation, as all further nonequilibrium effects are contained in the superadiabatic terms P_t^{id} and P_t^{exc}.

Inserting the splitting (24), (25), (28), (29), and (34) into (22) yields a decomposition which is identical to the case of systems with only translational degrees of freedom:[157]

$$R_t = P_t^{\text{id}} + P_t^{\text{exc}} + \dot{F}_{\text{id}} + \dot{F}_{\text{exc}} - X_t. \quad (36)$$

Evaluating the functional at the physical time evolution yields

$$\mathcal{R}_t^0 = R_t^0 \equiv R_t[\rho,\mathbf{J},\mathbf{J}^\omega], \quad (37)$$

where $\mathcal{R}_t^0$ is defined via (10).

In summary, (36) constitutes a splitting into intrinsic ($P_t^{\text{id}} + P_t^{\text{exc}} + \dot{F}_{\text{id}} + \dot{F}_{\text{exc}}$) and external contributions ($-X_t$). Crucially, all intrinsic terms are independent of the external force and torque fields, and depend only on the one-body density distribution and on the one-body current distribution. The non-interacting motion is described by the ideal terms ($P_t^{\text{id}} + \dot{F}_{\text{id}}$); together with the external power, these generate the free

drift-diffusion equation. The contributions due to internal interactions are contained in the excess parts ($P_t^{exc}+\dot{F}_{exc}$). Here $\dot{F}_{exc}$ arises from adiabatic free energy changes and P_t^{exc} contains the genuine nonequilibrium (superadiabatic) effects due to the motion in the system.

B. Steady state sum rules

The formulation in Sec. II A is general and applies to arbitrary time-dependent situations. We will henceforth consider steady states, i.e. where the one-body distributions carry no time dependence. This still includes cases with flow, $\mathbf{J}, \mathbf{J}^{\omega} \neq 0$, as long as the currents are time-independent and divergence-free, such that $\dot{\rho} = 0$ follows from (15). We split the value of the free power (37) into a contribution from internal forces, I_t, and an external contribution, X_t, according to

$$R_t^0 = -I_t/2 - X_t/2. \tag{38}$$

Here the external part of the free power is determined by (23) at the physical dynamics. On the many-body level, this is given by

$$X_t = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i (\mathbf{f}_i^{ext} \cdot \hat{\mathbf{v}}_i + \mathbf{f}_i^{\omega,ext} \cdot \hat{\mathbf{v}}_i^{\omega}) \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t), \tag{39}$$

where $\mathbf{f}_i^{ext} = -(\nabla_i v_i^{ext}) + \mathbf{X}_i + \gamma s \boldsymbol{\omega}$ is the external force and $\mathbf{f}_i^{\omega,ext} = -(\nabla_i^{\omega} v_i^{ext}) + \mathbf{X}_i^{\omega}$ is the external torque. The internal part in (38) is given by

$$\begin{aligned} I_t = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \sum_i \Big\{ \Big[-\nabla_i u(\mathbf{r}^N, \boldsymbol{\omega}^N) - k_B T \nabla_i \ln \Phi \Big] \cdot \hat{\mathbf{v}}_i \\ + \Big[-\nabla_i^{\omega} u(\mathbf{r}^N, \boldsymbol{\omega}^N) - k_B T \nabla_i^{\omega} \ln \Phi \Big] \cdot \hat{\mathbf{v}}_i^{\omega} \Big\} \Phi, \end{aligned} \tag{40}$$

where the arguments of $\Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t)$ have been left away for clarity. Here the operators ∇_i and ∇_i^{ω} only act inside of the brackets. Integration by parts and rearranging gives

$$\begin{aligned} I_t = \int d\mathbf{r}^N d\boldsymbol{\omega}^N \Big[u(\mathbf{r}^N, \boldsymbol{\omega}^N) + k_B T \ln \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t) \Big] \\ \times \sum_i \Big(\nabla_i \cdot \hat{\mathbf{v}}_i + \nabla_i^{\omega} \cdot \hat{\mathbf{v}}_i^{\omega} \Big) \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t). \end{aligned} \tag{41}$$

Using the Smoluchowski equation (1) in order to replace the sum allows us to obtain

$$I_t = -\int d\mathbf{r}^N d\boldsymbol{\omega}^N \Big[u(\mathbf{r}^N, \boldsymbol{\omega}^N) + k_B T \ln \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t) \Big] \frac{\partial}{\partial t} \Phi(\mathbf{r}^N, \boldsymbol{\omega}^N, t). \tag{42}$$

In steady state, the partial time derivative of Φ vanishes and hence we can conclude that

$$I_t = 0. \tag{43}$$

Using the splitting (38) the free power in steady state is thus trivially related to the external contribution,

$$R_t^0 = -X_t/2. \tag{44}$$

We next seek to exploit the availability of the two splittings of R_t, given by (36) and (38), in steady state. We start by rewriting (28) as

$$P_t^{id} = \frac{1}{2} \int d\mathbf{r} d\boldsymbol{\omega} \left(\mathbf{J} \cdot \frac{\gamma \mathbf{J}}{\rho} + \mathbf{J}^{\omega} \cdot \frac{\gamma^{\omega} \mathbf{J}^{\omega}}{\rho} \right). \tag{45}$$

Inserting the force balance equations (32) and (33) and rearranging yields

$$\begin{aligned} P_t^{id} = \frac{1}{2} \int d\mathbf{r} d\boldsymbol{\omega} \Big[\mathbf{J} \cdot \left(\gamma s \boldsymbol{\omega} - \nabla v^{ext} + \mathbf{X} - k_B T \nabla \ln \rho \right) \\ + \mathbf{J}^{\omega} \cdot \left(-\nabla^{\omega} v^{ext} + \mathbf{X}^{\omega} - k_B T \nabla^{\omega} \ln \rho \right) \Big] \\ - \frac{1}{2} \int d\mathbf{r} d\boldsymbol{\omega} \left(\mathbf{J} \cdot \frac{\delta W_t^{exc}}{\delta \mathbf{J}} + \mathbf{J}^{\omega} \cdot \frac{\delta W_t^{exc}}{\delta \mathbf{J}^{\omega}} \right). \end{aligned} \tag{46}$$

Integration by parts of the terms $\mathbf{J} \cdot k_B T \nabla \ln \rho$ and $\mathbf{J}^{\omega} \cdot k_B T \nabla^{\omega} \ln \rho$ and using the continuity equation (15), gives the time derivative of the density. As $\dot{\rho} = 0$ in steady state, the first integral in (46) reduces to the external power X_t, cf. (23). The second integral in (46) is determined by the superadiabatic contribution P_t^{exc}, as (34) reduces to $W_t^{exc} = P_t^{exc}$ in steady state, because then $\dot{F}_{exc} = 0$. Thus the free power in steady state is given by inserting (46) into (36), which yields upon observing that $\dot{F}_{id} = 0$ the result

$$R_t^0 = P_t^{exc} - \frac{X_t}{2} - \frac{1}{2} \int d\mathbf{r} d\boldsymbol{\omega} \left(\mathbf{J} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}} + \mathbf{J}^{\omega} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}^{\omega}} \right). \tag{47}$$

Comparing to (38) yields

$$I_t = -2P_t^{exc} + \int d\mathbf{r} d\boldsymbol{\omega} \left(\mathbf{J} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}} + \mathbf{J}^{\omega} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}^{\omega}} \right). \tag{48}$$

Using (43) the value of the superadiabatic functional in steady state is determined by the superadiabatic force, $\delta P_t^{exc}/\delta \mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t)$, and by the superadiabatic torque, $\delta P_t^{exc}/\delta \mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t)$, via

$$P_t^{exc} = \frac{1}{2} \int d\mathbf{r} d\boldsymbol{\omega} \left(\mathbf{J} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}} + \mathbf{J}^{\omega} \cdot \frac{\delta P_t^{exc}}{\delta \mathbf{J}^{\omega}} \right), \tag{49}$$

where all quantities are evaluated at the physical dynamics.

In summary, we have derived the following non-trivial identities (i.e. "sum rules") (43) and (44), as well as (47) and (49).

C. Excess dissipation functional

We next assume a model form for the excess dissipation functional which is instantaneous in time (i.e. Markovian) and given by

$$P_t^{exc} = \frac{\gamma}{2} \int d1 d2 \, \rho(1) \rho(2) \left(\frac{\mathbf{J}(1)}{\rho(1)} - \frac{\mathbf{J}(2)}{\rho(2)} \right)^2 M(1,2), \tag{50}$$

where the roman numerals refer to one position and one orientation, i.e. $1 \equiv \mathbf{r}, \boldsymbol{\omega}$ and $2 \equiv \mathbf{r}', \boldsymbol{\omega}'$, and $M(1, 2)$ is a convolution kernel, which depends on the differences $\mathbf{r} - \mathbf{r}'$ and $\boldsymbol{\omega} - \boldsymbol{\omega}'$. The form (50) was first proposed in Ref. 179. It constitutes a power

series up to second order in the velocity difference at two space points. Physically it models friction (or "drag") effects that occur in nonequilibrium due to the internal interactions in the system.

For a bulk steady state the number density becomes a constant, $\rho(\mathbf{r}, \boldsymbol{\omega}, t) = \rho_b$, and the current reduces to $\mathbf{J}(\mathbf{r}, \boldsymbol{\omega}, t) = J_b\boldsymbol{\omega}$, where J_b = constant. Furthermore $\mathbf{J}^{\omega}(\mathbf{r}, \boldsymbol{\omega}, t) = 0$, and $\dot{F} = 0$ due to the steady state condition. The power functional, (36), then reduces to

$$R_t = P_t^{id} - \gamma s \int d1 \mathbf{J}(1)\cdot\boldsymbol{\omega} + P_t^{exc} \tag{51}$$

$$= \frac{\gamma}{2}\int d1 \frac{\mathbf{J}(1)^2}{\rho(1)} - \gamma s \int d1 \mathbf{J}(1)\cdot\boldsymbol{\omega} + \frac{\gamma}{2}\int d1d2\, \rho(1)\rho(2)\left(\frac{\mathbf{J}(1)}{\rho(1)} - \frac{\mathbf{J}(2)}{\rho(2)}\right)^2 M(1,2) \tag{52}$$

which can be simplified to

$$\frac{R_t(J_b)}{4\pi V} = \frac{\gamma J_b^2}{2\rho_b} + \frac{\gamma M_0}{2} J_b^2 - \gamma s J_b, \tag{53}$$

where

$$M_0 = \int d2(\boldsymbol{\omega} - \boldsymbol{\omega}')^2 M(1,2) \tag{54}$$

is a density-dependent parameter, which is independent of J_b. In (53) J_b acts as a variational parameter. In order to apply the fundamental variational principle (20), we thus minimize (53) with respect to J_b,

$$\frac{\partial R_t}{\partial J_b} = \gamma \frac{J_b}{\rho_b} + \gamma M_0 J_b - \gamma s = 0. \tag{55}$$

As a result we obtain the relationship $J_b - s\rho_b = -M_0 J_b \rho_b$, from which the bulk current can be obtained as

$$J_b = \frac{s\rho_b}{1 + M_0\rho_b}. \tag{56}$$

Rearranging yields $M_0 = (s\rho_b - J_b)/\rho_b J_b$. Inserting this result in (53) leads to

$$\frac{R_t^0}{4\pi V} = -\frac{\gamma s}{2} J_b. \tag{57}$$

where R_t^0 denotes (as before) the value of the power functional at the minimum.

The ansatz for the currents, $\mathbf{J} = J_b\boldsymbol{\omega}$, $\mathbf{J}^{\omega} = 0$, allows the calculation of the external power X_t, and the internal power I_t. The external power (23) reduces to $X_t = \gamma s \int d1 \mathbf{J}(1)\cdot\boldsymbol{\omega}$, in the absence of further external forces (besides swimming) and torques. As $\boldsymbol{\omega}$ is a unit vector one obtains

$$\frac{X_t}{4\pi V} = \gamma s J_b. \tag{58}$$

Hence from comparing (57) and (58) we explicitly verify that $R_t^0 = -X_t/2$ holds, i.e. the present approximation for the excess dissipation functional respects the sum rule (44).

Evaluating the internal power (48) yields

$$\frac{I_t}{4\pi V} = -\gamma M_0 J_b^2 + \gamma M_0 \mathbf{J}_b^2 = 0, \tag{59}$$

and therefore the exact sum rule (43), $I_t = 0$, is also satisfied within the current approximation.

By inserting (56) in (57) we obtain

$$\frac{R_t^0}{4\pi V} = -\frac{\gamma s^2 \rho_b}{2(1 + M_0\rho_b)}. \tag{60}$$

It remains to specify the kernel, M(1, 2), in order to arrive at a closed theory. We assume that the kernel M(1, 2) and hence its moment M_0, (54), increase with density. We choose the simple functional form[179]

$$M_0 = \frac{1}{\rho_0 - \rho_b} + \frac{c_0 \rho_b^m}{\rho_0^{m+1}}, \tag{61}$$

where m is a positive integer and ρ_0 and c_0 are constants. Here the value of M_0 increases with increasing bulk density in order to model the increased collective friction effects that tend to slow down the dynamics. We show next that this form generates a decrease of average propulsion speed with increasing bulk density. For $\rho_b \to \rho_0$ jamming occurs (such that the bulk density is restricted to the interval $0 \le \rho_b \le \rho_0$) and hence $M_0 \to \infty$. Insertion into Eq. (56) yields

$$\frac{J_b}{s\rho_b} = \frac{1 - x}{1 + c_0 x^{m+1}(1 - x)}, \tag{62}$$

where $x = \rho_b/\rho_0$ is a scaled density. For the special case $c_0 = 0$ the well-known simple linear decrease of the forward speed with increasing bulk density is recovered.[86,95,136]

Within the approximation (50) the free power per volume becomes

$$\frac{R_t^0}{4\pi V} = \frac{\gamma s^2 \rho_b}{2} \frac{x - 1}{1 + c_0 x^{m+1}(1 - x)}, \tag{63}$$

which yields the free power per particle via division by $\rho_b = N/V$ as

$$\frac{R_t^0}{4\pi N} = \frac{\gamma s^2}{2} \frac{x - 1}{1 + c_0 x^{m+1}(1 - x)}. \tag{64}$$

We will show below that this simple functional form can provide a reliable account of the Brownian dynamics simulation data.

III. BROWNIAN DYNAMICS SIMULATIONS

The simulated system consists of N = 5000 spherical Brownian particles in two dimensions. The dynamics of the particles are represented by the over-damped Langevin equations

$$\dot{\mathbf{r}}_i(t) = s\boldsymbol{\omega}_i(t) + \gamma^{-1}\mathbf{f}_i^{int}(\mathbf{r}^N) + \boldsymbol{\xi}_i(t), \tag{65}$$

$$\dot{\varphi}_i(t) = \eta_i, \tag{66}$$

where, as before, $i = 1, \ldots, N$ labels the particles. The self-propulsion of each particle is along its unit orientation vector $\boldsymbol{\omega}_i(t) = (\sin\varphi_i(t), \cos\varphi_i(t))$ with the (free) swim speed s. The angle $\varphi_i(t)$ then describes the orientation of particle i at time t. The stochastic vector $\boldsymbol{\xi}_i$ and the stochastic scalar η_i are Gaussian distributed with zero mean and auto-correlations

$$\langle \boldsymbol{\xi}_i(t)\boldsymbol{\xi}_j(t')\rangle = 2D_{\text{trans}}\mathbf{1}\delta_{ij}\delta(t-t'), \tag{67}$$

$$\langle \eta_i(t)\eta_j(t')\rangle = 2D_{\text{rot}}\delta_{ij}\delta(t-t'), \tag{68}$$

where $D_{\text{trans}} = k_B T/\gamma$ and $D_{\text{rot}} = k_B T/\gamma^{\omega}$ are the translational and the rotational diffusion coefficients, respectively, and $\mathbf{1}$ is the 2×2 unit matrix. The inter-particle interaction force $\mathbf{f}_i^{\text{int}}(\mathbf{r}^N)$ is generated from the potential $u(\mathbf{r}^N)$ by $\mathbf{f}_i^{\text{int}}(\mathbf{r}^N) = -\nabla_i u(\mathbf{r}^N)$. Here we restrict ourselves to spherically symmetric pair interactions $u(\mathbf{r}^N) = \sum_{i,j,i<j}\phi(r_{ij})$, leading to forces of the form

$$\mathbf{f}_i^{\text{int}}(\mathbf{r}^N) = -\nabla_i \sum_{i,j\neq i}\phi(r_{ij}), \tag{69}$$

where $r_{ij} = |\mathbf{r}_i(t) - \mathbf{r}_j(t)|$. We use the Weeks-Chandler-Anderson (WCA)[95,185] pair potential, which is a Lennard-Jones potential that is cut at its minimum and shifted so it remains continuous:

$$\phi(r_{ij}) = 4\epsilon\left[\left(\sigma/r_{ij}\right)^{12} - \left(\sigma/r_{ij}\right)^{6}\right] + \epsilon, \text{ for } r_{ij} < 2^{1/6}\sigma. \tag{70}$$

Then (69) creates a purely repulsive interparticle interaction force and avoids artifacts at the cut-off.

A common measure for the activity of the particles is the Peclet number, which is defined as (see e.g. Ref. 179)

$$\text{Pe} = \frac{3s}{D_{\text{rot}}\sigma}. \tag{71}$$

This dimensionless number relates the two quantities that characterize the activity of the particles, namely s and D_{rot}.

The discretized dynamics proceed according to the Euler algorithm

$$\mathbf{r}_i(t+\Delta t) = \mathbf{r}_i(t) + \dot{\mathbf{r}}_i(t)\Delta t, \tag{72}$$

$$\boldsymbol{\omega}_i(t+\Delta t) = \boldsymbol{\omega}_i(t) + \dot{\boldsymbol{\omega}}_i(t)\Delta t, \tag{73}$$

where $\dot{\mathbf{r}}_i(t)$ and $\dot{\boldsymbol{\omega}}_i(t)$ are given by Eqs. (65) and (66), respectively. (Note that the noise term carries additional dependence on Δt.) Additionally the orientation vector $\boldsymbol{\omega}_i(t)$ is normalized to unit length at each time step to avoid effects on the propulsion speed.

We carry out Brownian dynamics (BD) simulations in 2D with a fixed time step of $\Delta t/\tau_0 = 10^{-5}$, with the natural time unit $\tau_0 = \sigma^2\gamma/\epsilon$. The fundamental units of the system are σ, γ and ϵ, where σ represents the size of the repulsive (LJ) core and ϵ its energy scale. All simulations are performed at a fixed ratio between the rotational diffusion constant and the translation diffusion constant, i.e. the thermal value $D_{\text{rot}}/D_{\text{trans}} = 3\sigma^{-2}$, and a fixed propulsion speed of $s\tau_0/\sigma = 24$. The particles are placed in a square, periodic box with side length $L = (N/\rho_b)^{1/2}$, where $\rho_b = N/V$, with V being the (two-dimensional) volume of the simulation box. We investigate the properties of the system as a function of the bulk density ρ_b, and of the temperature T. We carried out simulations with bulk densities in the range of $\rho_b\sigma^2 = 0.1$ to 1.2 in steps of 0.1. In addition we performed simulations with $\rho_b\sigma^2 = 0.01$ and $\rho_b\sigma^2 = 0.05$. For each density we consider the temperatures of $k_B T/\epsilon = 0.1, 0.2, 0.3, 0.4, 0.6$. We let the system reach the steady state for $n_{\text{equi}} = 10^7$ integration steps, followed by $n_{\text{sample}} = 10^8$ steps in which the sampled data are collected.

With these choices of parameters, the Peclet number (71) can be re-expressed as

$$\text{Pe} = \frac{s\gamma\sigma}{k_B T}, \tag{74}$$

and the parameters given above amount to the values Pe = 40, 60, 80, 120, 240 upon decreasing the scaled temperature $k_B T/\epsilon$.

All results presented below are averages over time and over particles and are calculated according to $\langle \mathcal{A}_i(j)\rangle = \frac{1}{N}\sum_{i=1}^{N}\frac{1}{n_{\text{sample}}}\sum_{j=1}^{n_{\text{sample}}}\mathcal{A}_i(j)$, where $\mathcal{A}_i(j)$ stands for an arbitrary (sampled) quantity for particle i at each discrete time point j. Hence this corresponds to the configuration space average.

An important example is the external power, given by

$$X_t = \left\langle \sum_i \mathbf{f}_i^{\text{ext}}(t)\cdot\mathbf{v}_i(t)\right\rangle, \tag{75}$$

where the external force is $\mathbf{f}_i^{\text{ext}} = \gamma s\boldsymbol{\omega}_i$ and the velocity $\mathbf{v}_i$ is the velocity of the particles.

IV. RESULTS

Figure 1 shows the external power X_t as a function of density for different temperatures. For temperatures $k_B T/\epsilon > 0.4$ the external power is practically linear in ρ_b, where the value for $\rho_b \to 0$ is purely determined by the external forces, i.e. collision between the particles occur very unlikely. The practically linear decrease of the velocity with increasing bulk density is a well-known phenomenon.[86,95,136] When decreasing the temperature, $k_B T/\epsilon \lesssim 0.4$, the external power develops a dip, i.e., a deviation from the linear shape. This region, where the external power is smaller than expected from a linear dependence on the bulk density, covers the range of $0.4 \lesssim \rho_b\sigma^2 \lesssim 1.2$. Hence for those set of parameters

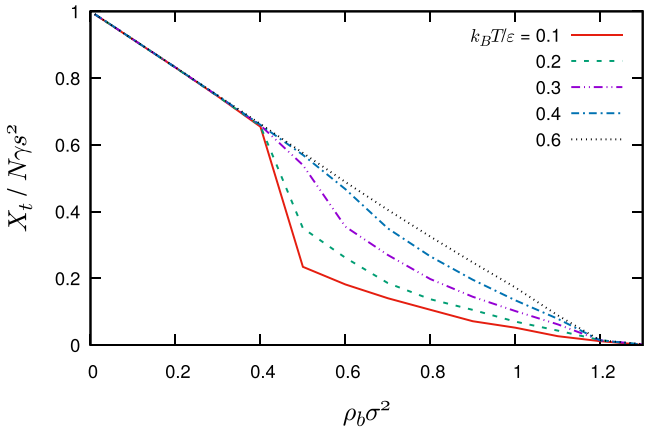

FIG. 1. Scaled external power X_t as a function of density. Temperatures are shown from $k_B T/\epsilon = 0$ to 1 as indicated by colors and line styles.

less external power needs to be provided for the dynamics of the system. As $R_t = -X_t/2$ in steady state this dip will develop also in the free power.

Figure 2(a) shows simulation results for the scaled free power per particle obtained from the external power, i.e. using the sum rule (44) and the simulation results obtained via (75). The free power is the central object in PFT and occurs in the theory as a consequence of general considerations for nonequilibrium systems. It is thus quite surprising that for the present case of active Brownian particles the free power is directly related to the average propulsion speed per particle $v(\rho_b)$, which appears as an important quantity in existing hydrodynamic theories.[9,94–96] The average self-propulsion speed is defined as the part of velocity of a particle in the direction of its orientation, averaged over the whole system,[9,82,93,95]

$$v = \frac{1}{N}\left\langle \sum_i \mathbf{v}_i(t)\cdot\boldsymbol{\omega}_i(t)\right\rangle, \tag{76}$$

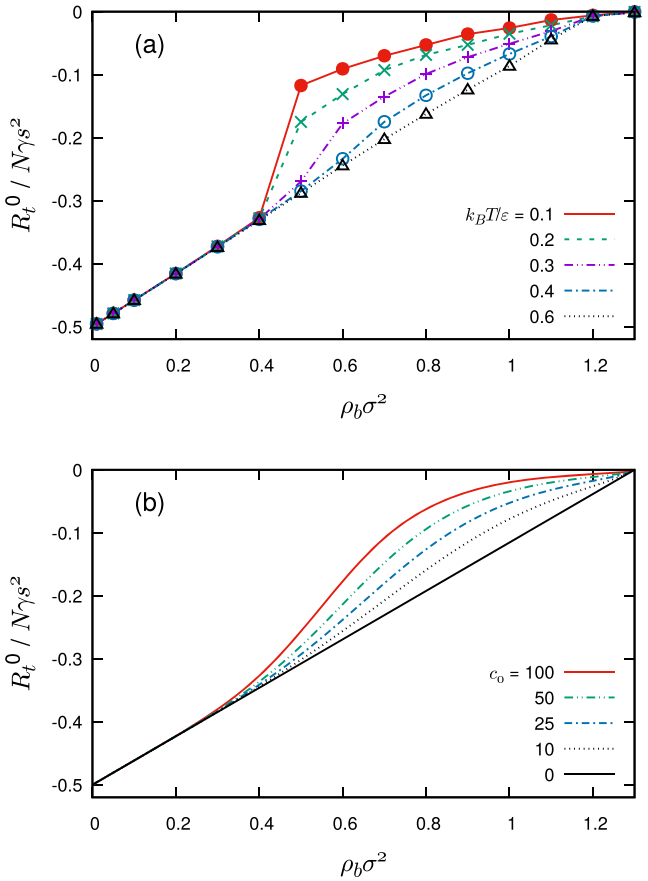

FIG. 2. (a) Scaled free power $R_t^0/(Ns^2\gamma)$ obtained from BD simulations via sampling X_t as a function of density. Temperatures are shown from $k_BT/\epsilon = 0$ to 1 as indicated by colors and line styles. (b) Theoretical results corresponding to (a), as given by (64). The jamming density is fixed to $\rho_0\sigma^2 = 1.3$, $m = 5$ and values of c_0 are 0–100 as indicated.

where $\mathbf{v}_i(t)$ is the velocity of particle i, which we implement as a central derivative of the position: $\mathbf{v}_i(t) = (\mathbf{r}_i(t+\Delta t) - \mathbf{r}_i(t-\Delta t))/(2\Delta t)$.[164,179,180] Therefore interparticle interactions change the velocity of the particles depending on the value of the bulk density. Thus v is density-dependent, $v(\rho_b)$. In the low-density limit, $\rho_b \to 0$, interparticle forces play no role. Hence $v(\rho_b \to 0) = \langle\sum_i(s\boldsymbol{\omega}_i(t) + \boldsymbol{\xi}_i(t))\cdot\boldsymbol{\omega}_i(t)\rangle/N = s$.

Multiplying both sides of (76) by γs yields

$$\gamma s v(\rho_b) = \frac{1}{N}\left\langle \sum_i \mathbf{v}_i(t)\cdot(\gamma s\boldsymbol{\omega}_i(t))\right\rangle. \tag{77}$$

The term between the angle brackets in (77) is X_t. Thus $v(\rho_b)$ is proportional to X_t as

$$X_t = N\gamma s v(\rho_b). \tag{78}$$

We have studied systematically the dependence of the external, and hence the free, power on temperature. Note that according to (74) this is analogous to varying Pe.[95] Similar to the external power, the free power increases linearly with increasing density for $k_BT/\epsilon > 0.4$, because with increasing density the dissipation per particle decreases due to collisions. This leads to a linear increase in the free power until the dynamics arrest and jamming occurs at ρ_0. Within the theory of Sec. II C, we can trace the near-linear behaviour to the specific form of the moment M_0, as given by (61). Clearly, this provides a good account of the simulation data. However, its status is that of an ad hoc assumption at present. Clearly, having a more microscopic justification constitutes a worthwhile task for future work.

For temperatures $k_BT/\epsilon \lesssim 0.4$ the increase of R_t^0 is nonlinear. While hardly any effect for low densities is observed, a significant hump develops for $\rho_b\sigma^2 \gtrsim 0.4$. The physical reason for this hump is an additional dissipation process, which increases the free power per particle even more than the excluded volume of the surrounding particles. We interpret this as a local clustering of the particles, as clustered particles dissipate less power to the solvent, compared to free particles. Whether the presence of a hump is related to the onset of phase separation is an open question.[179] A detailed discussion of motility induced phase separation is beyond the scope of this work. However, a body of work on this topic exists.[80–102,179] Comparing phase diagrams of different authors, see e.g. Fig. 6 in Refs. 94 and 96, shows that the onset of phase separation occurs in the same range as the hump in R_t^0 develops.

In Sec. II we defined an excess dissipation functional (50) and we showed that with a simple approximation an analytic expression for R_t^0 can be obtained, cf. (64). Numerical results for the scaled free power per particle, obtained using this expression, are shown in Fig. 2(b) as a function of ρ_b for different values of the parameter c_0. In order to fit the simulation results, we fixed the exponent in (64) to $m = 5$, the jamming density to $\rho_0\sigma^2 = 1.3$, and increased c_0 from zero to 100. This corresponds to decreasing the temperature in simulations, as a comparison with Fig. 2(a) reveals. Investigating in more detail the exact functional dependence of c_0 on temperature remains a task for future studies. This could involve

a microscopic derivation of the moment, M_0, of the correlation kernel, M(1, 2). Nevertheless, we expect c_0 to decrease to zero with increasing temperature, because thermal motion tends to break up clusters and thus works against phase separation.[179] Comparing Figs. 2(a) and 2(b) it is evident that our approximate form for the excess dissipation functional, (50), can reproduce the simulation results quite well, although the approximation for P_t^{exc} describes single-phase fluids, while the simulation results show MIPS. Hence PFT potentially serves as a theoretical tool to describe MIPS. This is an open problem for future work.

V. CONCLUSIONS

In this work we have investigated the steady state properties of active Brownian particles by applying PFT,[157] suitably generalized to deal with orientational degrees of freedom. We have obtained the value of the power functional, via sampling of the external power, directly from active Brownian dynamics simulations. A possibility to sample the free power directly is proposed elsewhere.[179] We leave a detailed formulation of the free power sampling for future work.

For the system under consideration the free power is straightforwardly related to the average forward swim speed of the particles. We have shown that the free power, and hence the average forward swim speed, exhibit a departure from linearity when dissipative clusters form in the system. However, the focus here is on clarifying details of the power functional approach for active particles and simulation method, rather than the analysis of cluster formation[119–121] and nonequilibrium steady states.[82,93,95] Using the constrained search method, we generate from the many-body starting point of PFT a one-body variational theory, for which the trial fields are the density, translational current and rotational current, respectively. A simple approximation to the dissipation functional generates an analytic variational theory. The one-body theory enables an intuitive physical interpretation of the simulation data, although some model input, most notably $c_0(T)$, still remained to be elucidated on the microscopic level. A possible way to achieve this is via studying dynamic two-body (van Hove) correlation functions, either via the nonequilibrium Ornstein-Zernike route,[173,174] or via the dynamical test particle limit[186,187] for which PFT provides an in-principle exact implementation.[181] Note that even within the dynamic density functional approximation, the dynamic test particle limit constitutes a valuable computational tool.[188,189] Furthermore the occurrence of phase separation is not yet included in the approximation presented here, and requires further work.[190] Also, a more microscopic treatment would require that bulk and interfacial contributions are taken into account separately. Obtaining microscopically based approximations to the excess free power functional and applying those to relevant situations, as e.g. the influence of gravity,[191] are useful future steps. Furthermore we showed that the internal power vanishes in steady state. The free power is thus purely determined by the external power, i.e., the self-propulsion of the particles.

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