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# Pressure and Phase Equilibria in Interacting Active Brownian Spheres

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We derive a microscopic expression for the mechanical pressure  $P$  in a system of spherical active Brownian particles at density  $\rho$ . Our exact result relates  $P$ , defined as the force per unit area on a bounding wall, to bulk correlation functions evaluated far away from the wall. It shows that (i)  $P(\rho)$  is a state function, independent of the particle-wall interaction; (ii) interactions contribute two terms to  $P$ , one encoding the slow-down that drives motility-induced phase separation, and the other a direct contribution well known for passive systems; (iii)  $P$  is equal in coexisting phases. We discuss the consequences of these results for the motility-induced phase separation of active Brownian particles, and show that the densities at coexistence *do not* satisfy a Maxwell construction on  $P$ .

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Much recent research addresses the statistical physics of active matter, whose constituent particles show autonomous dissipative motion (typically self-propulsion), sustained by an energy supply. Progress has been made in understanding spontaneous flow [1] and phase equilibria in active matter [2–6], but as yet there is no clear thermodynamic framework for these systems. Even the definition of basic thermodynamic variables such as temperature and pressure is problematic. While “effective temperature” is a widely used concept outside equilibrium [7], the discussion of pressure,  $P$ , in active matter has been neglected until recently [8–14]. At first sight, because  $P$  can be defined mechanically as the force per unit area on a confining wall, its computation as a statistical average looks unproblematic. Remarkably though, it was recently shown that for active matter the force on a wall can depend on details of the wall-particle interaction so that  $P$  is not, in general, a state function [15].

Active particles are nonetheless clearly capable of exerting a mechanical pressure  $P$  on their containers. (When immersed in a space-filling solvent, this becomes an *osmotic* pressure [8, 10].) Less clear is how to calculate  $P$ ; several suggestions have been made [9–12] whose inter-relationships are, as yet, uncertain. Recall that for systems in thermal equilibrium, the mechanical and thermodynamic definitions of pressure (force per unit area on a confining wall, and  $-(\partial\mathcal{F}/\partial V)_N$  for  $N$  particles in volume  $V$ , with  $\mathcal{F}$  the Helmholtz free energy) necessarily coincide. Accordingly, various formulae for  $P$  (involving, e.g., the density distribution near a wall [16], or correlators in the bulk [17, 18]) are always equivalent. This ceases to be true, in general, for active particles [11, 15].

In this Letter we adopt the mechanical definition of  $P$ . We first show analytically that  $P$  is a state function, independent of the wall-particle interaction, for one important and well-studied class of systems: spherical active

Brownian particles (ABPs) with isotropic repulsions. By definition, such ABPs undergo overdamped motion in response to a force that combines an arbitrary pair interaction with an external forcing term of constant magnitude along a body axis; this axis rotates by angular diffusion. While not a perfect representation of experiments (particularly in bulk fluids, where self-propulsion is created internally and hydrodynamic torques arise [19]), ABPs have become the mainstay of recent simulation and theoretical studies [3, 5, 6, 20–24]. They provide a benchmark for the statistical physics of active matter, and a simplified model for the experimental many-body dynamics of autophoretic colloidal swimmers, or other active systems, coupled to a momentum reservoir such as a supporting surface [24–29]. (We comment below on the momentum-conserving case.) By generating large amounts of data in systems whose dynamics and interactions are precisely known, ABP simulations are currently better placed than experiments to answer fundamental issues concerning the physics of active pressure, such as those raised in [9, 10].

Our key result exactly relates  $P$  to bulk correlators, powerfully generalizing familiar results for the passive case. The pressure for ABPs is the sum of an ideal-gas contribution and a non-ideal one stemming from interactions. Crucially, the latter results from *two* contributions: one is a standard, ‘direct’ term (the density of pairwise forces acting across a plane), which we call  $P_D$ , while the other, ‘indirect’ term, absent in the passive case, describes the reduction in momentum flux caused by collisional slowdown of the particles. For short-ranged repulsions and high propulsive force,  $P_D$  becomes important only at high densities; the indirect term dominates at intermediate densities and is responsible for motility-induced phase separation (MIPS) [2–4]. The same calculation establishes that, for spherical ABPs (though not in general [15])  $P$  must be equal in all coexisting phases.

We further show that our ideal and indirect terms together form exactly the ‘swim pressure’,  $P_S(\rho)$  at density  $\rho$ , previously defined via a force-moment integral in [9, 10], and moreover that (in 2D)  $P_S$  is simply  $\rho v(0)v(\rho)/(2D_r)$ , where  $v(\rho)$  is the mean propulsive speed of ABPs and  $D_r$  their rotational diffusivity. We interpret this result, and show that (for  $P_D = 0$ ) the mechanical instability,  $dP_S/d\rho = 0$ , coincides exactly with a diffusive one previously found to cause MIPS among particles whose interaction comprises a density-dependent swim speed  $v(\rho)$  [2–4]. We briefly explain why this correspondence does not extend to phase equilibria more generally, deferring a full account to a longer paper [33].

To calculate the pressure in interacting ABPs, we follow [15] and consider the dynamics in the presence of an explicit, conservative wall-particle force  $\mathbf{F}_w$ . For simplicity, we work in 2D, and consider periodic boundary conditions in  $y$  and confining walls parallel to  $\mathbf{e}_y = (0, 1)$ . We start from the standard Langevin dynamics of ABPs with bare speed  $v_0$ , interparticle forces  $\mathbf{F}$  and unit mobility [5, 6, 34]:

$$\begin{aligned}\dot{\mathbf{r}}_i &= v_0 \mathbf{u}(\theta_i) + F_w(x_i) \mathbf{e}_x + \sum_{j \neq i} \mathbf{F}(\mathbf{r}_j - \mathbf{r}_i) + \sqrt{2D_t} \boldsymbol{\eta}_i, \\ \dot{\theta}_i &= \sqrt{2D_r} \xi_i.\end{aligned}\quad (1)$$

Here  $\mathbf{r}_i(t) = (x_i, y_i)$  is the position, and  $\theta_i(t)$  the orientation, of particle  $i$  at time  $t$ ;  $\mathbf{u}(\theta) = (\cos(\theta), \sin(\theta))$ ;  $F_w = \|\mathbf{F}_w\|$  is a force acting along the wall normal  $\mathbf{e}_x = (1, 0)$ ;  $D_t$  is the bare translational diffusivity; and  $\boldsymbol{\eta}_i(t)$  and  $\xi_i(t)$  are zero-mean unit-variance Gaussian white noises with no correlations among particles.

Following standard procedures [2, 3, 35, 36] this leads to an equation for the fluctuating distribution function  $\hat{\psi}(\mathbf{r}, \theta, t)$  whose zeroth, first, and second angular harmonics are the fluctuating particle density  $\hat{\rho} = \int \hat{\psi} d\theta$ ; the  $x$ -polarization  $\hat{\mathcal{P}} = \int \hat{\psi} \cos(\theta) d\theta$ ; and  $\hat{\mathcal{Q}} = \int \hat{\psi} \cos(2\theta) d\theta$ , which encodes nematic order normal to the wall:

$$\begin{aligned}\dot{\hat{\psi}} &= -\nabla \cdot \left( (v_0 \mathbf{u}(\theta) + F_w(x) \mathbf{e}_x + \int \mathbf{F}(\mathbf{r}' - \mathbf{r}) \hat{\rho}(\mathbf{r}') d^2 r') \hat{\psi} \right) \\ &+ D_r \partial_\theta^2 \hat{\psi} + D_t \nabla^2 \hat{\psi} + \nabla \cdot (\sqrt{2D_t} \hat{\psi} \boldsymbol{\eta}) + \partial_\theta (\sqrt{2D_r} \hat{\psi} \xi),\end{aligned}\quad (2)$$

where  $\boldsymbol{\eta}(\mathbf{r}, t)$  and  $\xi(\mathbf{r}, t)$  are  $\delta$ -correlated, zero-mean, and unit-variance, Gaussian white noise fields. In steady-state, the noise-averages  $\rho = \langle \hat{\rho} \rangle$ ,  $\mathcal{P} = \langle \hat{\mathcal{P}} \rangle$ , and  $\mathcal{Q} = \langle \hat{\mathcal{Q}} \rangle$  are, by translational invariance, functions of  $x$  only, as is the wall force  $F_w(x)$  [37]. Integrating (2) over  $\theta$ , and then averaging over noise in steady state gives  $\partial_x J = 0$ , with  $J$  the particle current. For any system with impermeable boundaries,  $J = 0$ . Writing this out explicitly gives:

$$0 = v_0 \mathcal{P} + F_w \rho - D_t \partial_x \rho + I_1(x), \quad (3)$$

$$I_1(x) \equiv \int F_x(\mathbf{r}' - \mathbf{r}) \langle \hat{\rho}(\mathbf{r}') \hat{\rho}(\mathbf{r}) \rangle d^2 r'. \quad (4)$$

Applying the same procedure to the first angular har-

monic gives

$$D_r \mathcal{P} = -\partial_x \left[ \frac{v_0}{2} (\rho + \mathcal{Q}) + F_w \mathcal{P} - D_t \partial_x \mathcal{P} + I_2(x) \right], \quad (5)$$

$$I_2(x) \equiv \int F_x(\mathbf{r}' - \mathbf{r}) \langle \hat{\rho}(\mathbf{r}') \hat{\mathcal{P}}(\mathbf{r}) \rangle d^2 r'. \quad (6)$$

Note that the integrals  $I_1$  and  $I_2$  defined in (4) and (6) are, by translational invariance, functions only of  $x$ .

The mechanical pressure on the wall is the spatial integral of the force density exerted upon it by the particles. The wall force obeys  $F_w = -\partial_x U_w$  where an origin is chosen so that  $U_w$  is non-zero only for  $x > 0$ . The wall is confining, i.e.  $F_w \rho \rightarrow 0$  for  $x \gg 0$ , whereas  $x = \Lambda \ll 0$  denotes any plane in the bulk of the fluid, far from the wall. By Newton’s third law, the pressure is then

$$P = - \int_\Lambda^\infty F_w(x) \rho(x) dx, \quad (7)$$

In (7) we now use (3) to set  $-F_w \rho = v_0 \mathcal{P} - D_t \partial_x \rho + I_1$ :

$$P = v_0 \int_\Lambda^\infty \mathcal{P}(x) dx + D_t \rho(\Lambda) + \int_\Lambda^\infty I_1(x) dx. \quad (8)$$

We next use (5), in which  $\mathcal{P}$  and  $\mathcal{Q}$  vanish in the bulk and all terms vanish at infinity, to evaluate  $\int \mathcal{P} dx$ , giving:

$$P = \frac{v_0}{D_r} \left( \frac{v_0}{2} \rho(\Lambda) + I_2(\Lambda) \right) + D_t \rho(\Lambda) + \int_\Lambda^\infty I_1(x) dx. \quad (9)$$

Using Newton’s third law, the final integral in (9) takes a familiar form, describing the density of pair forces acting across some plane through the bulk (far from any wall):

$$\int_{x>\Lambda} dx \int_{x'<\Lambda} d^2 r' F_x(\mathbf{r}' - \mathbf{r}) \langle \hat{\rho}(\mathbf{r}') \hat{\rho}(\mathbf{r}) \rangle \equiv P_D. \quad (10)$$

Thus in the passive limit ( $v_0 = 0$ ) we recover in  $P_D$  the standard interaction part in the pressure [18]. We call  $P_D$  the ‘‘direct’’ contribution; it is affected by activity only through changes to the correlator. Activity also enters (via  $v_0$ ) the well-known ideal pressure term [9, 10, 13, 15]:

$$P_0 \equiv \left( D_t + \frac{v_0^2}{2D_r} \right) \rho(\Lambda). \quad (11)$$

Having set friction to unity in (1),  $D_t = k_B T$ , so that within  $P_0$  (only) activity looks like a temperature shift.

Most strikingly, activity in combination with interactions also brings an ‘‘indirect’’ pressure contribution

$$P_1 \equiv \frac{v_0}{D_r} I_2(\Lambda) \quad (12)$$

with no passive counterpart. Here  $I_2(\Lambda)$  is again a wall-independent quantity, evaluated on *any* bulk plane  $x = \Lambda \ll 0$ . We discuss this term further below.

Our exact result for mechanical pressure is finally

$$P = P_0 + P_1 + P_D \quad (13)$$

with these three terms defined by (11), (12), and (10), respectively.  $P$  is thus for interacting ABPs a state function, calculable solely from bulk correlations and independent of the particle-wall force  $F_w(x)$ . Because the same boundary force can be calculated using *any* bulk plane  $x = \Lambda$ , it follows that, should the system undergo phase separation,  $P$  is the same in all coexisting phases [37]. This proves for ABPs an assumption that, while plausible [10, 38], is not obvious, and indeed can fail for particles interacting via a density-dependent swim speed rather than direct interparticle forces [15].

Notably, although ABPs exchange momentum with a reservoir, (1) also describes particles swimming through a momentum-conserving bulk fluid, in an approximation where inter-particle and particle-wall hydrodynamic interactions are both neglected. So long as the wall interacts *solely* with the swimmers, our results above continue to apply to what is now the *osmotic* pressure.

The physics of the indirect contribution  $P_1$  is that interactions between ABPs reduce their motility as the density increases. The ideal pressure term  $P_0$  normally represents the flux of momentum through a bulk plane carried by particles that *move* across it (as opposed to those that *interact* across it) [17]. In our overdamped system one should replace in the preceding sentence ‘momentum’ with ‘propulsive force’ (plus a random force associated with  $D_t$ ). Per particle, the propulsive force is density-independent, but the rate of crossing the plane is not. Accordingly we expect the factor  $v_0^2$  in (11) to be modified by interactions, with one factor  $v_0$  (force or momentum) unaltered, but the other (speed) replaced by a density-dependent contribution  $v(\rho) \leq v_0$ :

$$P_0 + P_1 = \left( D_t + \frac{v_0 v(\rho)}{2D_r} \right) \rho. \quad (14)$$

This requires the mean particle speed to obey

$$v(\rho) = v_0 + 2I_2/\rho. \quad (15)$$

Remarkably, (14) and (15) are *exact* results, where (15) is found from the mean speed of particle  $i$  in bulk,  $v = v_0 + \langle \mathbf{u}(\theta_i) \cdot \sum_{j \neq i} \mathbf{F}(\mathbf{r}_j - \mathbf{r}_i) \rangle$ . To see why this average involves  $I_2$ , note that the system is isotropic in bulk, so  $x$  and  $y$  can be interchanged in  $I_2(x)$ , and that  $\cos(\theta) \equiv \mathbf{u} \cdot \mathbf{e}_x$ . Relation (6) then links  $v$  to  $I_2$  via the  $\langle \hat{\rho} \hat{\mathcal{P}} \rangle$  correlator, which describes the imbalance of forces acting on an ABP from neighbors in front and behind.

Furthermore, the self-propulsive term in (14) is exactly the ‘swim pressure’  $P_S$  of [9, 10]:

$$\frac{v_0 v(\rho)}{2D_r} \rho = P_S \equiv \frac{\rho}{2} \langle \mathbf{r} \cdot \mathbf{F}^a \rangle \quad (16)$$

with  $\mathbf{F}^a = v_0 \mathbf{u}$  a particle’s propulsive force and  $\mathbf{r}$  its position. (The particle mobility  $v_0/F^a = 1$  in our units.) The equivalence of (12), (14), and (16) is proven analytically in [39] and confirmed numerically in Fig. 1 for ABP simulations performed as in [20, 21].

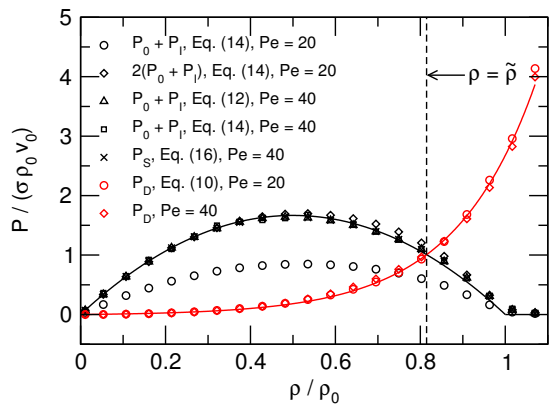


Figure 1. Numerical measurements of  $P_0 + P_1$ ,  $P_S$ , and  $P_D$  in single-phase ABP simulations at Péclet number  $Pe \equiv 3v_0/(D_r\sigma) = 40$ , where  $\sigma$  is the particle diameter. Expressions (12), (14), and (16) for  $P_0 + P_1$  and  $P_S$  show perfect agreement. Also shown is data for  $Pe = 20$ , unscaled and rescaled by factor 2. This confirms that  $P_S = P_0 + P_1$  is almost linear in  $Pe$ ; small deviations arise from  $Pe$ -dependence of the correlators. In red is  $P_D$  for  $Pe = 20, 40$ , with no rescaling.  $Pe$  was varied using  $D_r$ , at fixed  $v_0$  and with  $D_t = D_r\sigma^2/3$ . Solid lines are fits to piecewise parabolic ( $P_S$ ) and exponential ( $P_D$ ) functions used in the semi-empirical equation of state.  $\rho_0$  is a near-close-packed density at which  $v(\rho)$  vanishes and  $\tilde{\rho}$  is the threshold density above which  $P_D > P_S$ . See [39] for details.

Thus for  $D_t = 0$ , (13) may alternatively be rewritten as  $P = P_S + P_D$  [9, 10]. Together, our results confirm that  $P_S$ , defined in bulk via (16), determines (with  $P_D$ ) the force acting on a confining wall. This was checked numerically in [9] but is not automatic [15]. Moreover, our work gives via (14) an exact kinetic expression for  $P_S$  with a clear and simple physical interpretation in terms of the transport of propulsive forces. This illuminates the nature of the swim pressure  $P_S$  and extends to finite  $\rho$  the limiting result  $P_S = P_0$  [9, 10].

The connections made above are our central findings; they extend statistical thermodynamics concepts from equilibrium far into ABP physics. Before concluding, we ask how far these ideas extend to phase equilibria.

In the following we ignore for simplicity the  $D_t$  term (negligible in most cases [3, 5, 20, 34]). Then, assuming short-range repulsions, we have  $P_S = \rho v_0 v(\rho)/(2D_r)$ , with  $v(\rho) \simeq v_0(1 - \rho/\rho_0)$  and  $\rho_0$  a near-close-packed density [5, 6, 20].  $P_D$  should scale as  $\sigma \rho v_0 \mathcal{S}(\rho/\rho_0)$ , where  $\sigma$  is the particle diameter and the function  $\mathcal{S}$  diverges at close packing; here the factor  $v_0$  is because propulsive forces oppose repulsive ones, setting their scale [10]. Figure 1 shows that both the approximate expression for  $P_S$  (with a fitted  $\rho_0 \simeq 1.19$  roughly independent of  $Pe$ ), and the scaling of  $P_D$ , hold remarkably well. Defining a threshold value  $\tilde{\rho}$  by  $P_S(\tilde{\rho}) = P_D(\tilde{\rho})$  (see Fig. 1), it follows that at large enough Péclet number,  $Pe = 3v_0/(D_r\sigma)$ ,  $P_S$  dominates completely for  $\rho < \tilde{\rho}$ , with  $P_D$  serving *only* to prevent the density from moving above the  $\tilde{\rho}$  cutoff.

When  $\rho < \tilde{\rho}$ ,  $P_D$  is negligible; the criterion  $P'_S(\rho) < 0$ , used in [10, 38] to identify a mechanical instability, is then via (16) *identical* to the spinodal criterion  $(\rho v)' < 0$  used to predict MIPS in systems whose sole physics is a density-dependent speed  $v(\rho)$  [2, 3]. Thus, for ABPs at large  $Pe$ , the mechanical theory reproduces one result of a long-established mapping between MIPS and equilibrium colloids with attractive forces [2, 3].

We next address the binodal densities of coexisting phases. According to [2, 3], particles with speed  $v(\rho)$  admit an effective bulk free-energy density  $f(\rho) = k_B T [\rho(\ln \rho - 1) + \int_0^\rho \ln v(u) du]$ . (Interestingly, the equality of  $P$  in coexisting phases is equivalent at high  $Pe$  and  $\rho < \tilde{\rho}$  to the equality of  $k_B T \log(\rho v)$ , which is the chemical potential in this ‘thermodynamic’ theory [2, 4].) The binodals are then found using a common tangent construction (CTC, i.e., global minimization) on  $f$ , or equivalently an equal-area Maxwell construction (MC) on an effective *thermodynamic* pressure  $P_f = \rho f' - f$ , which differs from  $P$  [11]. Formally,  $f$  is a local approximation to a large-deviation functional [40], whose nonlocal terms can (in contrast to equilibrium systems) alter the CTC or MC [11, 20]; we return to this issue below.

An appealing alternative is to apply the MC to the mechanical pressure  $P$  itself; this was, in different language, proposed in [38]. (The equivalence will be detailed in [33].) It amounts to constructing an effective free-energy density  $f_P(\rho) \neq f$ , defined via  $P = \rho f'_P - f_P$ , and using the CTC on  $f_P$ . However,  $f_P$  has no clear link to any large deviation functional [40]; and since it differs from  $f$ , these approaches *generically predict different binodals*.

To confirm this, we turn to the large  $Pe$  limit; here, for ABPs with  $v(\rho) = v_0(1 - \rho/\rho_0)$  and  $\tilde{\rho} = \rho_0$ , we can explicitly construct  $f(\rho)$  (and hence  $P_f(\rho)$ ) alongside  $P(\rho)$  (and hence  $f_P(\rho)$ ), using our hard-cutoff approximation (i.e., a constraint  $\rho < \tilde{\rho}$ ). All four functions are plotted in [39]; the two distinct routes indeed predict different binodals at high  $Pe$  (see Fig. 2) [42]. Each approach suffers its own limitations. That via  $f$  (or  $P_f$ ) appears more accurate, but *neglects non-local terms* that can alter the binodals: although  $f'(\rho)$  remains equal in coexisting phases,  $P_f$  is not equal once those terms are included [11]. The most serious drawback of this approach, currently, is that it cannot address finite  $Pe$ , where  $P_D$  no longer creates a sharp cutoff. Meanwhile the ‘mechanical’ route captures the equality of  $P$  in coexisting phases but unjustifiably *assumes the MC on  $P$* , asserting in effect that  $f_P$ , and not  $f$ , is the effective free energy [40]. Nonlocal corrections [43] are again neglected.

At finite  $Pe$  where the crossover at  $\tilde{\rho}$  is soft, (13) shows how  $P_1$  and  $P_D$  compete, giving  $Pe$ -dependent binodals (see Fig. 2). To test the predictions of the mechanical approach (equivalent to [38]), we set  $P_D = \sigma \rho v_0 \mathcal{S}(\rho/\rho_0)$  as above, finding the function  $\mathcal{S}$  by numerics on single-phase systems at modest  $Pe$  (see Fig. 1). Adding this to  $P_S$  (assuming  $P_S \propto Pe$  scaling) gives  $P = P(\rho, Pe)$ . At each  $Pe$  the binodal pressures and densities do lie

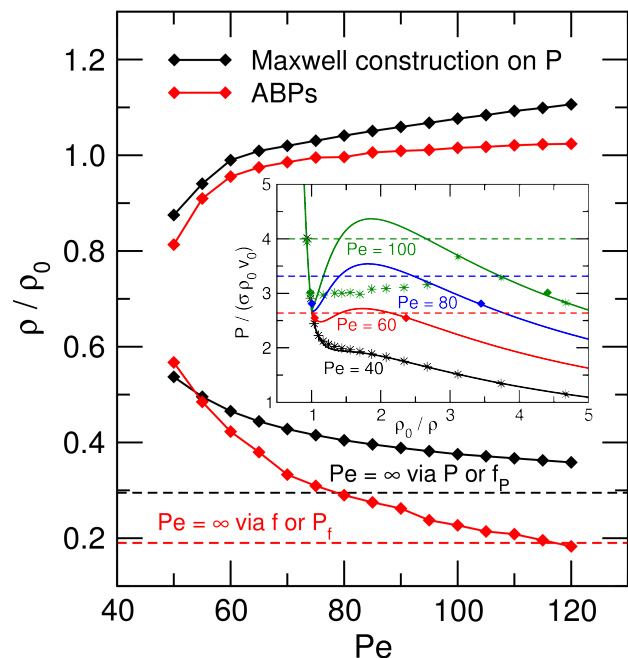


Figure 2. Simulated coexistence curves (binodals) for ABPs (red), and those calculated via the Maxwell construction (black) on the mechanical pressure  $P$  using the semi-empirical equation of state for  $P_S$  and  $P_D$  fitted from Fig. 1. Dashed lines: predicted high  $Pe$  asymptotes for the binodals calculated via  $f$  or  $P_f$  (lower), and calculated via  $P$  or  $f_P$  (upper). Inset: measured binodal pressures and densities (diamonds) fall on the equation-of-state curves but do not match the MC values (horizontal dashed lines). Stars show the  $P(\rho)$  relation across the full density range from simulations at  $Pe = 40$  and  $Pe = 100$ . The latter includes two metastable states at low density (high  $\rho_0/\rho$ ) that are yet to phase separate.

on this equation of state, validating its semi-empirical form; but they do not obey the Maxwell construction on  $P$ , which must therefore be rejected (see Fig. 2, inset). We conclude that, despite our work and that of [38], no complete theory of phase equilibria in ABPs yet exists.

In summary, we have given in (10)-(13) an exact expression for the mechanical pressure  $P$  of active Brownian spheres. This relates  $P$  directly to bulk correlation functions and shows it to be a state function, independent of the wall interaction, something not true for all active systems [15]. As well as an ideal term  $P_0$ , and a direct interaction term  $P_D$ , there is an indirect term  $P_1$  caused by collisional slowing down of propulsion. We established an exact link between  $P_0 + P_1$  and the so called ‘swim pressure’ [10], allowing a clearer interpretation of that quantity. We showed that when MIPS arises in the regime of high  $Pe = 3v_0/(D_r\sigma)$ , the mechanical ( $P' < 0$  [10]) and diffusive ( $f'' < 0$  [2, 3]) instabilities coincide. That equivalence does not extend to the calculation of coexistence curves, for reasons we have explained. For simplicity we have worked in 2D; generalization of our results to 3D is straightforward [33] but notationally cumbersome.

The established description of MIPS as a diffusive in-

stability [2, 3, 11, 20] is fully appropriate in systems whose particles are ‘programmed’ to change their dynamics at high density (e.g., via bacterial quorum sensing [44, 45]), but it is not yet clear whether the same theory, or one based primarily on the mechanical pressure  $P$ , is better founded for finite-Pe phase equilibria in ABPs whose slowdown is collisional. Meanwhile, our exact results for  $P$  in these systems add significantly to our growing understanding of how statistical thermodynamic concepts can, and cannot, be applied in active materials.

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- [1] M. C. Marchetti, J.-F. Joanny, S. Ramaswamy, T. B. Liverpool, J. Prost, M. Rao, and R. A. Simha, *Rev. Mod. Phys.* **85**, 1143 (2013).
- [2] J. Tailleur and M. E. Cates, *Phys. Rev. Lett.* **100**, 218103 (2008)
- [3] M. E. Cates and J. Tailleur, *Europhys. Lett.* **101**, 20010 (2013)
- [4] M. E. Cates and J. Tailleur, *Ann. Rev. Cond. Matt. Phys.*, in press; arXiv:1406.3533.
- [5] Y. Fily and M. C. Marchetti, *Phys. Rev. Lett.* **108**, 235702 (2012)
- [6] G. S. Redner, M. F. Hagan and A. Baskaran, *Phys. Rev. Lett.* **110**, 055701 (2013)
- [7] L. F. Cugliandolo, *J. Phys. A* **44**, 3001 (2011)
- [8] T. W. Lion and R. J. Allen, *Europhys. Lett.* **106**, 34003 (2014); *J. Chem. Phys.* **137**, 244911 (2014)
- [9] X. B. Yang, L. M. Manning and M. C. Marchetti, *Soft Matter* **10**, 6477 (2014)
- [10] S. C. Takatori, W. Yan and J. F. Brady, *Phys. Rev. Lett.* **113**, 028103 (2014)
- [11] R. Wittkowski, A. Tiribocchi, J. Stenhammar, R. J. Allen, D. Marenduzzo and M. E. Cates, *Nature Commun.* **5**, 4351 (2014)
- [12] F. Ginot, I. Theurkauff, D. Levis, C. Ybert, L. Bocquet, L. Berthier, C. Cotton-Bizonne, *Phys. Rev. X*, in press, arXiv:1411.7175
- [13] S. A. Mallory, A. Saric, C. Valeriani, A. Cacciuto. *Phys. Rev. E* **89**, 052303 (2014)
- [14] R. Ni, M. A. Cohen-Stuart, P. G. Bolhuis, arxiv:1403.1533 (2014)
- [15] A. P. Solon, Y. Fily, A. Baskaran, M. E. Cates, Y. Kafri, M. Kardar and J. Tailleur, arXiv:1412.3952 (2014)
- [16] J. R. Henderson, *Statistical Mechanical Sum Rules*, in Fundamentals of Inhomogeneous Fluids, D. Henderson, Ed., Marcel Dekker, New York (1992)
- [17] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids*, Oxford University Press, Oxford (1987)
- [18] M. Doi, *Soft Matter Physics*, Oxford University Press, Oxford (2013)
- [19] R. Matas-Navarro, R. Golestanian, T. B. Liverpool and S. M. Fielding, *Phys. Rev. E* **90**, 032304 (2014); A. Zoettl and H. Stark, *Phys. Rev. Lett.* **112**, 118101 (2014)
- [20] J. Stenhammar, A. Tiribocchi, R. J. Allen, D. Marenduzzo and M. E. Cates, *Phys. Rev. Lett.* **111**, 147502 (2013)
- [21] J. Stenhammar, D. Marenduzzo, R. J. Allen and M. E. Cates, *Soft Matter* **10**, 1489 (2014)
- [22] T. Speck, J. Bialké, A. M. Menzel and H. Löwen, *Phys. Rev. Lett.* **112**, 218304 (2014)
- [23] A. Wysocki, R. G. Winkler and G. Gompper, *Europhys. Lett.* **105**, 48004 (2014)
- [24] I. Buttinoni, J. Bialké, F. Kümmel, H. Löwen, C. Bechinger and T. Speck, *Phys. Rev. Lett.* **110**, 238301 (2013)
- [25] J. R. Howse, R. A. L. Jones, A. J. Ryan, T. Gough, R. Vafabakhsh and R. Golestanian, *Phys. Rev. Lett.* **99**, 048102 (2007)
- [26] J. Palacci, S. Sacanna, A. P. Stenberg, D. J. Pine and P. M. Chaikin, *Science* **339**, 936 (2013)
- [27] I. Theurkauff, C. Cottin-Bizonne, J. Palacci, C. Ybert and L. Bocquet, *Phys. Rev. Lett.* **108**, 268303 (2012)
- [28] S. Thutupalli, R. Seeman and S. Herminghaus, *New J. Phys.* **13**, 073021 (2011)
- [29] Any self-propelled entity whose motility depends on frictional contact with a support (such as human walking, cell crawling [30], vibrated granular materials [31], or colloids that move by rolling on a surface [32]) is exchanging momentum with an external reservoir (the support).
- [30] E. Tjhung, D. Marenduzzo and M. E. Cates, *Proc. Nat. Acad. Sci. USA* **109**, 12381 (2012)
- [31] J. Deseigne, O. Dauchot, H. Chaté, *Phys. Rev. Lett.* **105**, 098001 (2010); V. Narayan, S. Ramaswamy, N. Menon, *Science* **317**, 105 (2007)
- [32] A. Bricard, J. B. Caussin, N. Desreumaux, O. Dauchot, and D. Bartolo, *Nature* **503**, 95 (2013)
- [33] A. P. Solon *et al.* In preparation.
- [34] Y. Fily, S. Henkes and M. C. Marchetti, *Soft Matter* **10**, 2132 (2014)
- [35] F. D. C. Farrell, J. Tailleur, D. Marenduzzo, M. C. Marchetti, *Phys. Rev. Lett.* **108**, 248101 (2012)
- [36] D. S. Dean, *J. Phys. A. Math. Gen.* **29**, L613 (1996)
- [37] We assume, without loss of generality, that translational invariance in  $y$  is maintained even if the system undergoes phase separation into two or more isotropic phases.
- [38] S. C. Takatori and J. F. Brady, arXiv:1411.5776 (2014)
- [39] Supplemental Material is available at [URL will be provided by the publisher].
- [40] The large deviation functional (LDF, or effective free energy)  $\mathcal{F}[\hat{\rho}(\mathbf{r})]/(Vk_B T)$  for the fluctuating density  $\hat{\rho}$  in a nonequilibrium system is defined as  $-\ln(\text{Pr}[\hat{\rho}(\mathbf{r})])/V$

where  $\text{Pr}$  is the steady-state probability distribution [41].

In [2, 3], it is shown that  $\int f(\hat{\rho})d^d r/(Vk_B T)$  is, within the local approximation, the LDF for a system of particles with a density-dependent swim speed  $v(\rho)$ .

- [41] R. S. Ellis, *Entropy, Large Deviations and Statistical Mechanics*, Springer Verlag, Berlin (1985)
- [42] An additional simulation at  $\text{Pe} = 500$  gave a lower binodal value,  $\rho/\rho_0 \simeq 0.08$ . This may be due to the non-local

gradient terms identified in [11].

- [43] C. Y. D. Lu, P. D. Olmsted and R. C. Ball, *Phys. Rev. Lett.* **84**, 642 (2000)
- [44] M. B. Miller and B. L. Bassler, *Ann. Rev. Microbiol.* **55**, 165 (2001)
- [45] M. E. Cates, *Rep. Prog. Phys.* **75**, 042601 (2012)
- [46] The following is cited in Supplemental Material [39]: S. J. Plimpton, *J. Comp. Phys.* **117**, 1 (1995);