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 ρ. 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; and (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.

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 interrelations 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 formulas 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 Refs. [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 nonideal 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 slow-down 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 ρ, previously defined via a force-moment integral in Refs. [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 Ref. [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],

$$\dot{\mathbf{r}}_i = v_0\mathbf{u}(\theta_i) + F_w(x_i)\mathbf{e}_x + \sum_{j \neq i}\mathbf{F}(\mathbf{r}_j - \mathbf{r}_i) + \sqrt{2D_t}\boldsymbol{\eta}_i,$$

$$\dot{\theta}_i = \sqrt{2D_r}\xi_i. \qquad (1)$$

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,

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

where $\boldsymbol{\eta}(\mathbf{r},t)$ and $\xi(\mathbf{r},t)$ are δ-correlated, zero-mean, and unit-variance Gaussian noise fields. In the 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 θ and then

averaging over noise in the 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

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

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

Applying the same procedure to the first angular harmonic gives

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

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

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 nonzero only for $x > 0$. The wall is confining, i.e., $F_w\rho \to 0$ 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

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

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

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

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

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

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),

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

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],

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

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

$$P_I \equiv \frac{v_0}{D_r} I_2(\Lambda) \tag{12}$$

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

$$P = P_0 + P_I + P_D, \tag{13}$$

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 interparticle 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_I 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 $v(\rho) \leq v_0$,

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

This requires the mean particle speed to obey

$$v(\rho) = v_0 + 2I_2/\rho. \tag{15}$$

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 Refs. [9,10],

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

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 the Supplemental Material [39] and confirmed numerically in Fig. 1 for ABP simulations performed as in Refs. [20,21].

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 Ref. [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
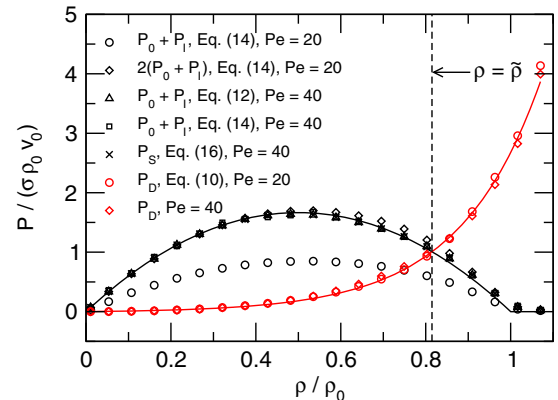

FIG. 1 (color online). Numerical measurements of $P_0 + P_I$, P_S, and P_D in single-phase ABP simulations at Péclet number $\text{Pe} \equiv 3v_0/(D_r\sigma) = 40$, where σ is the particle diameter. Expressions (12), (14), and (16) for $P_0 + P_I$ and P_S show perfect agreement. Also shown are data for $\text{Pe} = 20$, unscaled and rescaled by factor 2. This confirms that $P_S = P_0 + P_I$ is almost linear in Pe; small deviations arise from the Pe dependence of the correlators. In red is P_D for $\text{Pe} = 20, 40$, 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 semiempirical equation of state. ρ_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 the Supplemental Material [39] for details.

swim pressure P_S and extends to finite ρ 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 ρ_0 a near-close-packed density [5,6,20]. P_D should scale as $\sigma \rho v_0 \mathcal{S}(\rho/\rho_0)$, where σ 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 numbers 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 Refs. [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 Refs. [2,3], particles with speed $v(\rho)$ admit an effective bulk free-energy density $f(\rho) = k_BT[\rho(\ln \rho - 1) + \int_0^\rho \ln v(u)du]$. [Interestingly, the equality of P in coexisting phases is equivalent at high Pe and $\rho < \tilde{\rho}$ to the equality of $k_BT \log(\rho v)$, 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 [41], 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 Ref. [38]. (The equivalence will be detailed in Ref. [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 [41], 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 the Supplemental Material [39]; the two distinct routes indeed predict different binodals at high Pe (see Fig. 2) [43]. Each approach suffers its own limitations. That via f (or P_f) appears more accurate, but neglects nonlocal 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 [41]. Nonlocal corrections [44] are again neglected.

At finite Pe where the crossover at $\tilde{\rho}$ is soft, (13) shows how P_I and P_D compete, giving Pe-dependent binodals (see Fig. 2). To test the predictions of the mechanical approach (equivalent to Ref. [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 $P = P(\rho, \text{Pe})$. At each Pe the binodal pressures and densities do lie on this
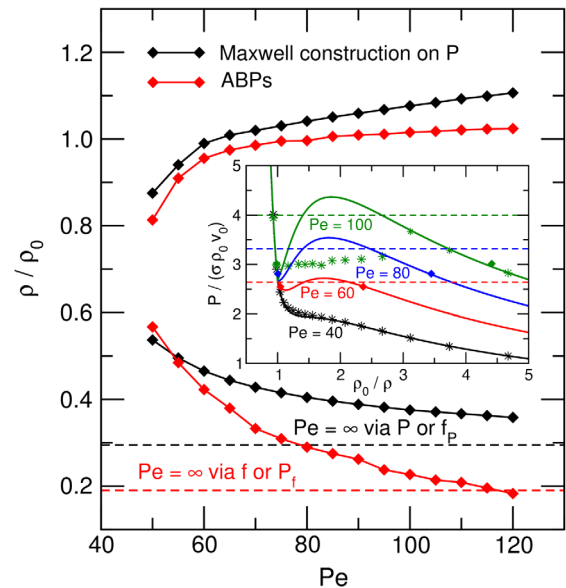

FIG. 2 (color online). Simulated coexistence curves (binodals) for ABPs (red) and those calculated via the Maxwell construction (black) on the mechanical pressure P using the semiempirical equation of state for P_S and P_D fitted from Fig. 1. Dashed lines: predicted high Pe asymptotes for the binodals calculated via f or P_f (lower line) and calculated via P or f_P (upper line). 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 ρ_0/ρ) that are yet to phase separate.

equation of state, validating its semiempirical 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 Ref. [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_I caused by collisional slowing-down of propulsion. We established an exact link between $P_0 + P_I$ 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 instability [2,3,11,20] is fully appropriate in systems whose particles are "programed" to change their dynamics at high density (e.g., via bacterial quorum sensing [45,46]), 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 slow-down 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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that move by rolling on a surface [32]) is exchanging momentum with an external reservoir (the support).

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