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# Theory for the Anomalous Phase Behavior of Inertial Active Brownian Particles

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In contrast to equilibrium systems, inertia can profoundly impact the phase behavior of active systems. This has been made particularly evident in recent years, with motility-induced phase separation (MIPS) exhibiting several intriguing dependencies on translational inertia. Here we report extensive simulations characterizing the phase behavior of inertial active matter and develop a mechanical theory for the complete phase diagram without appealing to equilibrium notions. Our theory qualitatively captures all aspects of liquid-gas coexistence, including the critical value of inertia above which MIPS ceases. Notably, our findings highlight that particle softness, and not inertia, is responsible for the MIPS reentrance effect at the center of a proposed active refrigeration cycle.

**Introduction.**— The driving forces for many of the intriguing patterns and phases formed by nonequilibrium systems can entirely differ from those of equilibrium systems. Perhaps the most notable example of this is single-component, purely-repulsive active particles which phase separate into two disordered phases despite the absence of attractive interactions [1–4]. This “motility-induced phase separation” [1, 4] (MIPS) has attracted much attention and has served as a model nonequilibrium transition used to generalize thermodynamic arguments. While the primary axes of the phase diagram that have been interrogated for active particles are density and measures of “activity”, all nonequilibrium systems inherently have an additional axis as particle inertia can impact the phase behavior [5–15]. This stands in contrast with equilibrium systems where, absent the influence of gravity, particle inertia has no impact on phase behavior and pattern formation.

While most studies of active systems have focused on the overdamped (i.e., inertia-free) limit, Mandal *et al.* highlighted the significant role of inertia in the phase behavior of two-dimensional (2D) repulsive active disks [12]. There, the authors reported that, for finite inertia and for certain fixed density, increasing activity can result in moving from a stable homogeneous system to MIPS and finally *back* to a stable homogeneous system at *large* activities. This unexpected reentrance effect, along with the observation that the phases have different kinetic temperatures [12, 14], was used to propose a refrigeration cycle powered by inertial active matter [15].

A number of compelling arguments have been offered to explain some of these intriguing observations [10–12]; however a theory for the complete inertial dependence of the phase diagram of active matter remains an outstanding challenge. A mechanical approach to constructing liquid-gas binodals has been proposed that, crucially, makes no appeals to equilibrium ideas [16–18]. We are thus at an exciting juncture at which many of the anomalous aspects of active phase diagrams may now receive a detailed theoretical treatment.

In this Letter, we develop a nonequilibrium theory for liquid-gas transition of inertial active systems as a function of density, activity, and inertia. Using extensive Brownian dynamics simulations of three-dimensional (3D) active spheres, we numerically determine the phase diagram and mechanical

equations of state as a function of activity, density, and inertia. We analytically derive the nonequilibrium coexistence criteria for active inertial particles *solely in terms of bulk equations* and use our simulations to construct empirical expressions for these state functions. The resulting theoretical phase diagram qualitatively captures all aspects of the phase diagram, including the increasing (and diverging) critical activity and the non-monotonicity of the kinetic temperature difference between the phases with increasing inertia. We find that the previously observed reentrance effect is not present for active particles in the hard-sphere limit and is in fact driven by the *softness* of the interparticle potential rather than particle inertia. More generally, we hope that the framework used in this study can be used to systematically probe the origins and build an intuition for the phase behavior of driven systems as a function of less familiar dynamical control parameters, including inertia.

**Model system.**— We consider the simplest model that captures the inertial effects on MIPS: 3D athermal active spheres [19] with underdamped translational dynamics. Each of the  $N$  particles experiences three translational forces: a drag force  $-\zeta\dot{\mathbf{x}}$  proportional to the particle velocity  $\dot{\mathbf{x}}$  and translational drag coefficient  $\zeta$ , a conservative interparticle force  $\mathbf{F}^C[\mathbf{x}^N]$ , where  $\mathbf{x}^N$  represents all particle positions, and an active force  $\mathbf{F}^A = \zeta U_0 \mathbf{q}$  where  $U_0$  is the intrinsic active speed. Stochastic Brownian translational forces, which simply attenuate the influence of activity, are neglected. The particle orientation  $\mathbf{q}$  independently follow diffusive dynamics  $\dot{\mathbf{q}} = \boldsymbol{\Omega} \times \mathbf{q}$  where the stochastic angular velocity has mean  $\mathbf{0}$  and variance  $\langle \boldsymbol{\Omega}(t) \boldsymbol{\Omega}(0) \rangle = 2/\tau_R \delta(t) \mathbf{I}$  and  $\tau_R$  represents characteristic reorientation time (or inverse rotational diffusion). Here, we consider only the effects of translational inertia (neglecting rotational inertia) to facilitate a comparison with existing studies [10, 12, 15, 20].

The inclusion of inertia introduces the momentum relaxation time,  $\tau_M \equiv m/\zeta$  where  $m$  is the particle mass. The interparticle force  $\mathbf{F}^C[\mathbf{x}_N; \sigma, \varepsilon]$  is taken from a Weeks-Chandler-Anderson (WCA) potential, which is characterized by a Lennard-Jones diameter  $\sigma$  and energy  $\varepsilon$  [21]. Taking  $\zeta U_0$ ,  $\sigma$ , and  $\tau_R$  to be the characteristic units of force, length, and time,

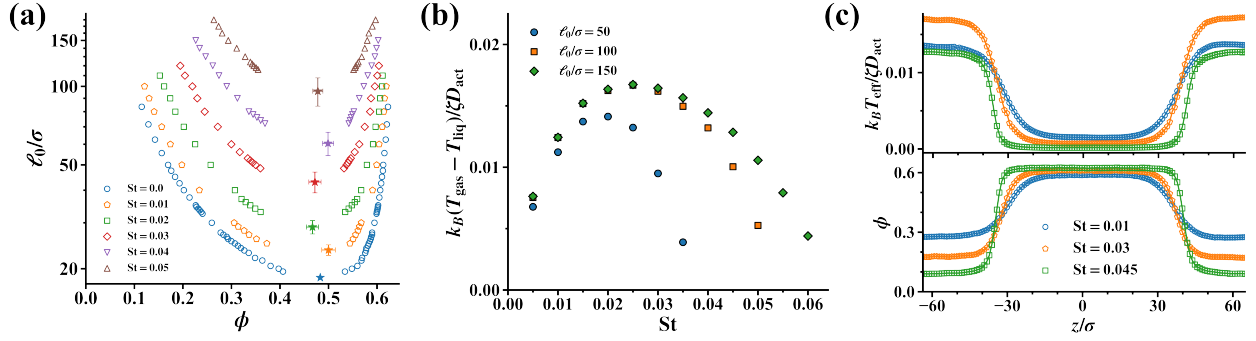


FIG. 1. Simulation results for: (a) binodals of 3D ABPs with  $St = 0.0$  data obtained from Ref. [19] (data near  $\phi = 0.5$  are absent due to challenges in obtaining binodal data close to the critical point); (b) temperature difference between coexisting phases (where  $D_{\text{act}} \equiv U_0 \ell_0/6$ ); and (c) density and kinetic temperature spatial profiles with  $\ell_0/\sigma = 150.0$  (lines are a guide to the eye).

respectively, the dimensionless Langevin equation follows as:

$$St \ddot{\mathbf{x}} = -\dot{\mathbf{x}} + \frac{\ell_0}{\sigma} \left( \mathbf{q} + \bar{\mathbf{F}}^C[\bar{\mathbf{x}}^N; \mathcal{S}] \right) \quad (1)$$

where we have defined the Stokes number  $St \equiv \tau_M/\tau_R$  and intrinsic “run length” (activity)  $\ell_0 \equiv U_0 \tau_R$ . The dimensionless force  $\bar{\mathbf{F}}^C$  depends on the reduced positions  $\bar{\mathbf{x}}^N$  and is fully characterized by the “stiffness” parameter  $\mathcal{S} \equiv \varepsilon/(\zeta U_0 \sigma)$  [19]. For overdamped particles, as  $\mathcal{S} \rightarrow \infty$ , the finite amplitude of the active force and absence of stochastic translational Brownian forces strictly prevents particles from overlapping within a center-to-center distance of  $D = 2^{1/6} \sigma$  [19]. The particles behave precisely as hard spheres in this limit, despite the use of a continuous potential. The system state is fully described by four dimensionless parameters: the volume fraction ( $\phi \equiv N \pi D^3/6V$ ),  $\ell_0/\sigma$ ,  $St$ , and  $\mathcal{S}$ .

*Simulated phase diagram.*— We use Brownian dynamics simulations to construct the phase diagram with simulation details provided in the Supplemental Material (SM) [22]. We vary density, activity, and inertia while fixing  $\mathcal{S} = 50$  [23]. All simulations were conducted with a minimum of 93000 particles using HOOMD-blue [22, 24]. The resulting binodal curves are presented in the activity-volume fraction plane for several different values of inertia, shown in Fig. 1(a). With increasing inertia, the critical activity monotonically increases, while the critical volume fraction displays no statistically significant dependence on inertia. Here, the critical points are determined by fitting the binodal data nearest to the critical point to power-law critical scaling [22]. While a comprehensive critical scaling analysis will be required to investigate the critical phenomena of inertial ABPs [25–32], the present results make clear that, for fixed stiffness, increasing translational inertia monotonically broadens the single-phase region, shifting the binodal to higher activities.

An interesting peculiarity of inertial MIPS is that the kinetic temperature – defined as  $k_B T_{\text{eff}} = m \langle \dot{\mathbf{x}}^2 \rangle / 3$  in 3D – is distinct between the coexisting phases [12–15]. The temperature differences between coexisting gas and liquid phases for different  $St$  and  $\ell_0/\sigma$  are displayed in Fig. 1(b) and were ex-

tracted from the spatial kinetic temperature profiles (along the axis,  $z$ , normal to the interface) shown in Fig. 1(c). For each activity, the inertial dependence of this temperature difference is nonmonotonic and concave. We can understand the origins of this nonmonotonicity by considering that the kinetic temperature is a decreasing function of density and increasing function of inertia [10, 22]. For a fixed activity, increasing inertia initially increases the temperature difference but, eventually, increasing inertia begins to narrow the density difference between the phases as the critical activity shifts to higher values [see Fig. 1(a)] with these competing effects giving rise to the nonmonotonicity.

*Theory of inertial MIPS.*— In the absence of a variational principle, mechanics is a natural choice for constructing nonequilibrium phase diagrams. The mechanical theory of nonequilibrium fluid-fluid coexistence was recently presented in Ref. [18]. There, a procedure for obtaining the coexistence criteria which makes no appeals to equilibrium notions was outlined. The criteria that emerge from this mechanical framework can be expressed in the form of a generalized equal-area Maxwell construction. Crucially, while the integration variable in this construction is identical for *all equilibrium systems*, out of equilibrium this variable is *system specific* and must be determined from a detailed statistical mechanical treatment.

A statistical mechanical description of interacting ABPs was provided for overdamped active particles in Ref. [18]. Introducing particle inertia results in  $3N$  additional degrees of freedom that, unlike equilibrium systems, will have a non-trivial distribution. Beginning from our microscopic equation-of-motion [Eq. (1)], the probability density  $f_N(\mathbf{\Gamma}; t)$  of finding the system in a microstate  $\mathbf{\Gamma} = (\mathbf{x}^N, \dot{\mathbf{x}}^N, \mathbf{q}^N)$  satisfies  $\partial f_N / \partial t = \mathcal{L} f_N$ , where  $\mathcal{L}$  is a dynamical operator specific to the equation-of-motion [22]. In the case of phase separation the order parameter is the number density field  $\rho(\mathbf{x}; t) = \langle \hat{\rho}(\mathbf{x}) \rangle = \langle \sum_{\alpha=1}^N \delta(\mathbf{x} - \mathbf{x}_\alpha) \rangle$  with its evolution equation given by  $\partial \rho / \partial t = \int_\gamma \hat{\rho} \mathcal{L} f_N d\mathbf{\Gamma}$ , where  $\langle \dots \rangle = \int (\dots) f_N(\mathbf{\Gamma}, t) d\mathbf{\Gamma}$  is the ensemble average and  $\gamma$  denotes the phase-space volume.

We now simplify our formally exact expressions using the

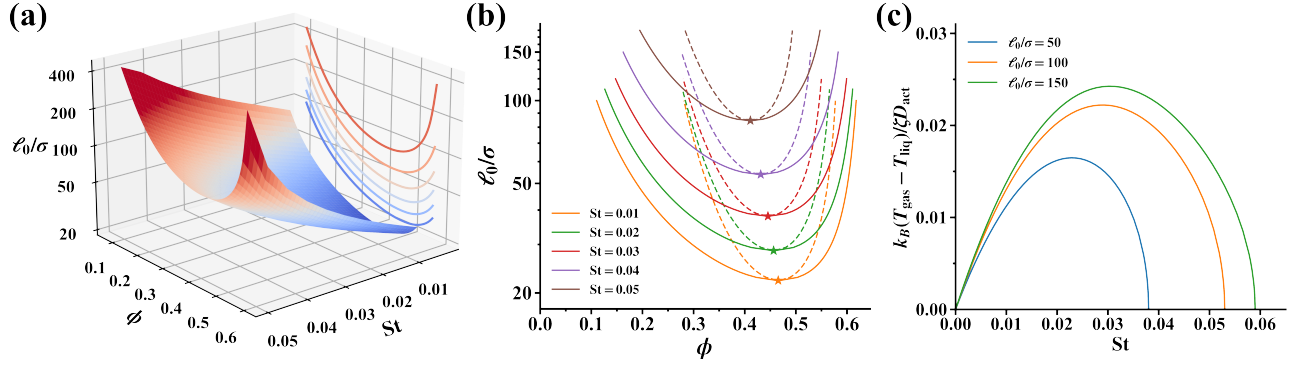


FIG. 2. Theoretically predicted: (a) binodals for 3D ABPs in the continuous three-dimensional parameter space; (b) binodals (solid lines) and spinodals (dashed lines) for several  $St$  (stars denote critical points); and (c) kinetic temperature difference between coexisting phases.

conditions of stationary phase coexistence with a planar interface and taking  $z$  to be the direction normal to the interface. Solving for the density profile of a quasi one-dimensional flux-free steady state requires solving the static mechanical balance,  $d\Sigma_{zz}/dz = 0$  [18, 33], where  $\Sigma$  is the *dynamic stress* tensor which includes both the effects of true stresses and body forces [10, 34]. Straightforward integration of the mechanical balance under these conditions reveals that the dynamic stress must be spatially uniform, with  $\Sigma_{zz} = C$ . When the dynamic stress can be expressed as:

$$-\Sigma_{zz} = \mathcal{P}(\rho) - a(\rho) \frac{d^2 \rho}{dz^2} - b(\rho) \left( \frac{d\rho}{dz} \right)^2, \quad (2)$$

where  $\mathcal{P}$  is the bulk dynamic pressure and  $a$  and  $b$  represent interfacial coefficients, the condition of uniform stress results in the following nonequilibrium coexistence criteria [10]:

$$\mathcal{P}(\rho_{\text{liq}}) = \mathcal{P}(\rho_{\text{gas}}) = \mathcal{P}^{\text{coexist}}, \quad (3a)$$

$$\int_{\rho_{\text{gas}}}^{\rho_{\text{liq}}} [\mathcal{P}(\rho) - \mathcal{P}^{\text{coexist}}] E(\rho) d\rho = 0, \quad (3b)$$

where

$$E(\rho) = \frac{1}{a(\rho)} \exp \left( 2 \int \frac{b(\rho)}{a(\rho)} d\rho \right). \quad (3c)$$

The second criterion (3b), is a *weighted-area* Maxwell construction in the  $\mathcal{P} - \rho$  plane where  $E(\rho)$  is a weighting factor [Eq. (3c)] defined such that  $\rho(z)$  does not appear in the criterion: only bulk equations of state ( $\mathcal{P}$ ,  $a$ , and  $b$ ) are needed to now find the binodal densities [35].

To use this nonequilibrium coexistence framework we must: (i) formally expand our exact expression of the dynamic stress to match the form of Eq. (2); and (ii) construct equations of state for the terms appearing in the expansion [36]. The dynamic stress for interacting inertial active particles is found to have three distinct contributions arising from particle interactions, the active force, and the kinetic energy of the particles [22]. Discarding higher-order gradient terms and gradient terms with coefficients that are independent or weakly dependent on activity, we are able to cast our dynamic stress in the

desired form, finding (in 3D) [22]:

$$\mathcal{P}(\rho) = p_{\text{int}} + p_{\text{act}} + p_{\text{k}}, \quad (4a)$$

$$a(\rho) = \frac{\ell_0^2}{20(1+6St)(1+2St)} \left[ \frac{6St}{1+St} + 1 \right] \bar{U}^2 \frac{\partial p_{\text{int}}}{\partial \rho}, \quad (4b)$$

$$b(\rho) = \frac{\ell_0^2}{20(1+6St)(1+2St)} \left[ \frac{6St}{1+St} \frac{\partial}{\partial \rho} \left( \bar{U}^2 \frac{\partial p_{\text{int}}}{\partial \rho} \right) + \bar{U} \frac{\partial}{\partial \rho} \left( \bar{U} \frac{\partial p_{\text{int}}}{\partial \rho} \right) \right], \quad (4c)$$

where  $\bar{U} \equiv (p_{\text{act}} + p_{\text{k}})/(\zeta D^{\text{act}} \rho)$  is a dimensionless active speed with  $\bar{U} \in [0, 1]$ . We thus identify the weighting factor:

$$E(\rho) = \bar{U}^{12St/(1+7St)} \frac{\partial p_{\text{int}}}{\partial \rho}, \quad (4d)$$

and can now formally construct the liquid-gas binodal for inertial active particles *without appealing to equilibrium notions*. We directly fit our equations of state ( $p_{\text{int}}$ ,  $p_{\text{act}}$ ,  $p_{\text{k}}$ ) to homogeneous fluid simulation data while ensuring that all known limits are enforced. The resulting analytical forms of  $p_{\text{k}}$ ,  $p_{\text{act}}$ , and  $p_{\text{int}}$  with respect to  $(\phi, \ell_0/\sigma, St)$  are provided in the SM [22]. Moreover, as detailed in the SM [22], several closures and approximations were made to derive Eqs. (4a)-(4d). While these assumptions may affect the quantitative accuracy of the coexistence criteria, our goal is to qualitatively understand how inertia alters these criteria compared to overdamped ABPs. We believe that the key finding—that inertia has a highly nonlinear effect on the coexistence criteria—is likely robust.

Employing our coexistence criteria and equations of state, we construct the three-dimensional liquid-gas binodal surface for inertial ABPs across the entire parameter space  $(\ell_0/\sigma, \phi, St)$  [see Fig. 2(a)]. Slices of the binodal for several values of inertia are presented in Fig. 2(b) alongside the spinodal, the latter of which was obtained from a mechanical linear stability analysis [22]. The theoretical binodal correctly captures the trend of an increasing critical activity and, intriguingly, predicts a systematic reduction in the critical density with increasing inertia. As presented in Fig. S9 [22],

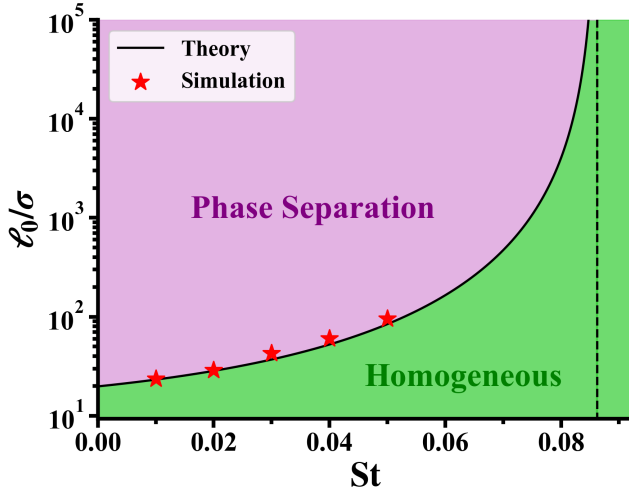


FIG. 3. Theoretically predicted critical activity with  $St$ . Stars denote simulation results. The vertical dashed line is located at  $St = 0.086$ .

the binodal of inertial ABPs can be well approximated by the overdamped theory *despite* the nonlinear dependence of the coexistence criteria on inertia. This nonlinear dependence is not sufficient to quantitatively alter binodal from the overdamped prediction precisely because MIPS occurs at relatively small  $St$  and the coexistence theory for inertial ABPs reduces to that of Ref. [18] as  $St \rightarrow 0$ . We can also predict the kinetic temperature difference between coexisting phases as shown in Fig. 2(c), correctly capturing the qualitative trend of a nonmonotonic inertia dependence found in our simulations.

Our theory predicts that the critical activity diverges as  $St \rightarrow 0.086$  (see Fig. 3). For  $St$  larger than this incredibly small value of inertia, MIPS is entirely suppressed for all activities and densities. While the precise value of this critical  $St$  depends on the specific forms of equations of state, its existence is consistent with our simulations, which also aligns with earlier findings [10–12]. Moreover, Fig. 3 makes clear that there is no value of inertia where increasing activity can result in a phase-separated system to homogenize, the reentrance effect reported in Ref. [12] is absent.

We can understand the inertia-induced stability of active fluids by considering the stability criteria for phase separation. A homogeneous fluid with a dynamic pressure that decreases with increasing density (resulting in a van der Waals loop),  $\partial\mathcal{P}/\partial\rho < 0$ , is unstable with respect to linear density perturbations [22]. In the absence of inertia, the dynamic pressure of active fluids with purely repulsive interactions consists of  $p_{\text{int}}$  and  $p_{\text{act}}$ , the latter of which can exhibit a nonmonotonic density dependence and induce MIPS at high activities. While inertia also generates  $p_k$ , straightforward energy conservation arguments result in the relation  $p_k/p_{\text{act}} = 2St$  in 3D [10]: *both* the kinetic and active pressures can destabilize active fluids.

Figure 4(a) displays the density dependence of our dynamic pressure as a function of inertia for a fixed activity. Points on the curves correspond to data obtained from simulations of ho-

ogeneous fluids (the absence of data is indicative of regions where MIPS quickly occurs). At the lowest value of inertia, a prominent van der Waals loop exists in  $\mathcal{P}$  signifying a fluid that is unstable for a broad range of densities [37]. With increasing inertia, the van der Waals loop rapidly diminishes and is eventually eliminated entirely. We can see that the increased stability arises from the reduced magnitude of the contributions of  $p_k + p_{\text{act}}$  relative to those of  $p_{\text{int}}$ . The increasing interaction pressure with inertia reflects the enhanced probability of interparticle contacts arising from the sluggish particle dynamics: particles collide and remain in close proximity for a longer duration. These inertia-enhanced collisions reduce the effective speed of the particles and thus the active pressure. In contrast, the kinetic pressure continues to increase with inertia for all densities. This increase is not enough to compensate for the reduced active pressure and increased interaction pressure, resulting in a closing of the van der Waals loop with increasing inertia and stable homogeneous active fluids.

*Origins of reentrant MIPS.*— Mandal *et al.* [12] first observed a reentrance effect, in which a crossover from a homogeneous system to MIPS and back to a homogeneous system was observed with increasing activity for a certain range of inertia. The origins of this observation (and its connection to particle inertia) have remained, to the best of our knowledge, unclear. We now identify the exact origin of this reentrance effect. Often, varying the activity of ABPs is done by increasing the active force while keeping all other parameters fixed (including the interaction parameters). As a result, while the dimensionless activity  $\ell_0/\sigma$  increases with increasing active force the particle stiffness  $\mathcal{S}$  was simultaneously inversely decreasing: the particles become effectively softer. This increasing softness allows particles to more closely approach each other and achieve higher densities. The origins of this reentrance effect is thus not due to inertia, but rather, a consequence of the simultaneous variation of both activity and interparticle stiffness. In other words, the basic physics rooted in the reentrance of MIPS is the increasingly unhindered particle motion due to the diminished role of interparticle repulsion with decreasing stiffness. The slowing down of particles due to particle interactions is a crucial ingredient in MIPS and the reduction of this effect with increasing softness is the origin of MIPS reentrance.

Fixing  $\mathcal{S}$  while varying  $\ell_0/\sigma$  is precisely why the reentrance effect was not observed in our work – its origins are rooted in  $\mathcal{S}$  rather than  $St$ . We have explicitly verified that a variable  $\mathcal{S}$  is required for a reentrant MIPS using both 2D and 3D inertial ABPs. Notably, in *overdamped* 2D ABPs, the reentrance effect persists when  $\mathcal{S}$  decreases with increasing  $\ell_0/\sigma$ , reinforcing that a variable  $\mathcal{S}$  drives this effect [22, 38]. Indeed, previous work has reported the reentrance of MIPS in overdamped self-propelled repulsive disks [38] and others have speculated that the observed reentrant effect is an effect of variable particle stiffness [39]. Reentrance appears to be found in all repulsive systems exhibiting MIPS if  $\mathcal{S}$  is varied but is entirely absent when  $\mathcal{S}$  is held constant.

*Conclusions.*— In this work, we have developed a theory for

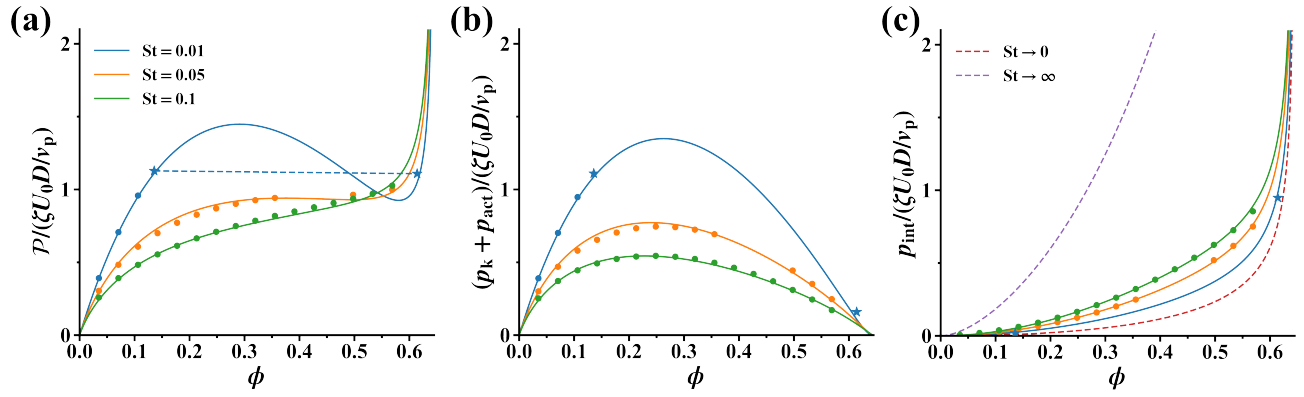


FIG. 4. Inertia dependence of: (a)  $\mathcal{P}$ ; (b)  $p_k + p_{\text{act}}$ ; and (c)  $p_{\text{int}}$  for  $\ell_0/\sigma = 85.0$ . Solid lines indicate fits while symbols denote simulation results. In (a), the dashed line and stars respectively denote  $\mathcal{P}^{\text{coexist}}$  and the binodal densities from simulations with  $St = 0.01$ . In (c), the red and purple dashed lines respectively denote the predicted asymptotic forms of  $p_{\text{int}}$  as  $St \rightarrow 0$  and  $St \rightarrow \infty$ .  $v_p \equiv \pi D^3/6$  is the particle volume.

the full phase diagram of active Brownian spheres as a function of activity, translational inertia, and density. Our theory is able to reproduce all qualitative trends observed in our simulations while also offering an intuitive picture for why inertia eliminates MIPS (particles collide more and move less). Moreover, we find the reentrance effect that forms the basis of a recently proposed active refrigeration cycle, has origins that are rooted in particle interaction softness. As inertia can play a prominent role in the dynamics of both living and artificial active systems, it is our hope that the theory presented in this work could be useful to understand various experimental phenomena exhibited by these diverse systems [40–46]. For example, from the experimental parameters in Ref. [41], we determined a Stokes number  $St = 0.0184$  for phase-separating rotating robots, which is just below the critical value where MIPS is expected to break down and thus consistent with our theory. More generally, we hope that the framework used in this work can be extended to interrogate additional axes of the active phase diagram, including rotational inertia [47–53], and include forms of coexistence with order parameters beyond the density.

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- derive the coexistence criteria, and the stability analysis for inertial ABPs.
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