Self-Organized Critical Coexistence Phase in Repulsive Active Particles

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We revisit motility-induced phase separation in two models of active particles interacting by pairwise repulsion. We show that the resulting dense phase contains gas bubbles distributed algebraically up to a typically large cutoff scale. At large enough system size and/or global density, all the gas may be contained inside the bubbles, at which point the system is microphase-separated with a finite cut-off bubble scale. We observe that the ordering is anomalous, with different dynamics for the coarsening of the dense phase and of the gas bubbles. This phenomenology is reproduced by a “reduced bubble model” that implements the basic idea of reverse Ostwald ripening put forward in [Phys. Rev. X 8, 031080 (2018)].

Self-propelled particles interacting solely with steric repulsion are well known to be able to spontaneously separate into a macroscopic dense cluster and a residual gas, in spite of the absence of explicit attraction forces. This motility-induced phase separation (MIPS) [1] of active particles has become a cornerstone of the physics of dry active matter (in which the fluid surrounding particles is neglected). As such, it has driven many theoretical works [2-12] as well as countless numerical studies (see, e.g. [8-15] to name a few prominent ones). The motility reduction resulting from persistent collisions, which leads to MIPS, is a generic ingredient encountered both in living and synthetic active matter [16-19].

There is indeed mounting evidence that more complex physics is at play in systems of repulsive disks. In particular, the surface tension between the dense phase and the gas, defined via the Laplace law, has been measured to be negative [20-22,23], triggering a spate of controversy [24]. Very recently, Caporusso et al [25] have shown more clearly that in systems with hard-core interactions, the dense phase is made of hexatic subdomains and interstitial gas regions.

In this Letter we show, within two standard particle models displaying MIPS, that not only the dense phase is endowed with bubbles, but also that these are distributed algebraically up to some cutoff scale that we observe to grow with system size. Finite-size scaling based on this observation suggests that, as system size increases, more and more of the gas is contained in bubbles. At large densities, we are able to observe the vanishing of the macroscopic gas reservoir, and the system is then microphase-separated with bubbles of all sizes up to a maximal bubble size that depends on the average density. Moreover, the coarsening of bubbles is anomalous with the typical length scale growing as $\rho^{0.22}$. We elucidate the basic mechanisms at play, and show, within a reduced model implementing reversed Ostwald ripening for gas bubbles, that they indeed lead to a self-organized critical dynamics.

**Self-organized critical phase coexistence.** We first consider the paradigmatic active brownian particles (ABPs) introduced in [8]. Self-propelled by a force of constant magnitude $F_0$ along its internal polarity $\mathbf{u}_i = (\cos \theta_i, \sin \theta_i)$, particle $i$ evolves according to the overdamped Langevin equations governing its position $\mathbf{r}_i$ and polar angle $\theta_i$:

$$\dot{\mathbf{r}}_i = \mu_i (F_0 \mathbf{u}_i + \mathbf{F}_i) + \eta_i; \quad \dot{\theta}_i = \eta_i$$

where $\mathbf{F}_i = -\sum_{j \neq i} \nabla V(\mathbf{r}_i - \mathbf{r}_j)$ is the force exerted on particle $i$ by the other particles. We choose the pair potential to be a short-range harmonic repulsion $V(r) = \frac{k}{2}(\sigma - r)^2$ if $r < \sigma$ and 0 otherwise with $k$ the
repulsive strength and $\sigma = 1$ the interaction radius. In contrast to previous studies, we allow the mobility tensor $\mathbf{\mu}$, and the translational noise $\mathbf{\eta}$ to be anisotropic, as expected generically for active particles: $\mathbf{\mu}_i = \mu_1 \mathbf{u}_i + \mu_\perp (1 - \mathbf{u}_i \cdot \mathbf{u}_i) \mathbf{z}$, $\mathbf{\eta}_i = \sqrt{2\tau \mu_2 \xi_i^1} \mathbf{u}_i + \sqrt{2\tau \mu_\perp \xi_i^1} (\mathbf{u}_i \times \mathbf{z})$, and $\eta_i = \sqrt{2\tau \mu_\theta \xi_i^2}$ with $\tau$ a parameter controlling the noise strength, $\mathbf{z}$ the unit vector perpendicular to the plane of motion, and the $\xi_i$'s Gaussian white noises with unit variance.

All simulations below are of large two-dimensional domains with periodic boundary conditions. Numerical details are given in [26]. At phase coexistence, we observe, inside the macroscopic dense domain, persistent bubbles with a range of sizes, surrounded by a liquid. Bubbles are more prominent when the mobility is anisotropic [27].

Typical snapshots for $\mu_\parallel = 4 \mu_\perp$ and $\mu_\theta = 6 \mu_\perp$ are shown in Fig. 1(a,b) for systems only differing by their size. Clearly, doubling system size increases the size of the bubbles. Fig. 1(c) shows, at different system sizes, $n(a)$, the average number of bubbles of area $a$, normalized by $S_\ell$, the total area of the liquid in which bubbles live. The distributions collapse on an increasing range, span several orders of magnitude, and decay approximately as a power law $n(a) \sim a^{-\alpha}$ with $\alpha \approx 1.75$ terminated by a cutoff that increases with system size.

The ABP simulations reported above only show a rather short scaling range. Numerically, the main limitation is not so much system size than the huge times needed to obtain clean averages [28]. We thus implemented an active lattice gas [29,33] on an hexagonal lattice [33], particles carrying an internal polarity pointing to one of the 6 lattice directions attempt to perform one of 3 moves (see [26] for details). (i) With a rate $r_P$ they perform a ‘self-propelled’ jump to the nearest site along their internal polarity direction. (ii) They undergo spatial diffusion to any neighboring site at rate $r_D$, and (iii) rotational diffusion (changing their polarity to one of its two neighboring orientations) at rate $r_R$. For optimal efficiency, we impose strict exclusion and parallel updating.

In the following, we use $r_P = 1$, $r_D = 2$ and $r_R = 0.032$, typical values leading to phase separation (a study of the phase diagram will be presented elsewhere). Persistent bubbles are clearly visible (Fig. 2(a-d)). The bubble area distribution in the globally phase-separated regime is similar to that observed for ABPs but with a much larger scaling region (Fig. 2(e)): $n(a) \sim a^{-\alpha}$ with $\alpha = 1.75(5)$. This region extends to a cut-off size $a_c$ that grows with the total liquid area as $a_c \propto S_\ell^\gamma$ with $\gamma = 1.40(5)$. The distributions can thus be collapsed on a master curve using these two exponents (Fig. 2(f)).

In both models presented, we find that whenever the system is globally phase separated, the dense phase contains bubbles. This phase bears the hallmarks of self-organized criticality (SOC) (for recent overviews, see [31,35]). Small bubbles are nucleated inside the liquid, diffuse and grow by merging with other bubbles. This process get slower and slower with increasing bubble size. Bubbles are eventually expelled into the reservoir of outside gas upon touching the boundary, in sudden, avalanche-like events, providing separation of timescales (see movie in [26]). As in typical SOC systems, avalanches occur at all accessible scales.

The SOC-like mechanisms leading to an algebraic distribution of bubbles do not invalidate the global picture of a phase separation between gas and liquid with fixed densities $\rho_g$ and $\rho_l$ independent of system size up to small finite-size corrections. However, the gaseous part of the system is formed here of the outside gas reservoir and of the bubbles. With this definition, the gas fraction $x_g$ fluctuates very little and is independent of system size to a good approximation (Fig. 3(a)). Moreover, $x_g$ varies linearly with the average density $\rho_0$ (Fig. 3(b)) so that the lever rule still applies: For $\rho_g < \rho_0 < \rho_l$, the average density sets the fraction of liquid and gas in the system $x_g = (\rho_l - \rho_0)/(\rho_\ell - \rho_g)$ and $x_l = 1 - x_g$. The fluctuations of $x_b$, the fraction of the system occupied by bubbles, in contrast to the gentle ones of $x_g$, are large, intermittent, and increase with system size. (Note the huge timescales over which fluctuations of $x_b$ occur even at the modest sizes shown in Fig. 3(a).) Their stronger and stronger peaks reflect the larger and larger avalanches (expulsion of bubbles) (Fig. 3(a), insets).

Microphase-separated bubbly liquid. The lever rule immediately tells us that the SOC scaling evidenced above cannot continue asymptotically when system size $S \to \infty$. Indeed, the bubble area fraction grows with system size:

$$x_b \equiv \frac{1}{S} \int_0^\infty a \, n(a) \, da \approx \frac{S_\ell}{S} \int_0^{a_c} a^{1-\alpha} \, da \propto x_l S_\ell^{(2-\alpha)},$$

(2)

where we used the scalings of $n(a)$ and $a_c(S_\ell)$. Our numerical data confirm this (Fig. 3(c)). Surely, Eq. 2...
FIG. 2. Active lattice gas. (a–d) Snapshots in the steady state at $\rho_0 = 0.8$ in systems of different sizes from $S = 384 \times 256$ to $S = 1532 \times 1024$ (colors as in Fig. 1(a,b)). For the two biggest sizes (c,d), the system is in the microphase-separated regime. (e–h) bubble area distribution in the SOC scaling regime (e,f: $\rho_0 = 0.6$) and in the microphase-separated regime (g,h). (e) $n(a)/S_t$ at various system sizes (indicated by the legends in (f)). Typical averaging time is $10^{10}$ timesteps after discarding a transient of $10^6$. (f): same as (e), but as function of $a/S_t^\gamma$ with $\gamma = 1.40$. (g): $n(a)/S_t$ at different $\rho_0$ values for $S = 768 \times 1024$ (dashed lines) and $S = 1536 \times 2048$ (solid lines). (h): same as (g), but rescaled according to Eq. (3). The grey dashed lines have slope $-\alpha = -1.75$.

FIG. 3. Active lattice gas at $\rho_0 = 0.63$. (a): timeseries of the area fraction occupied by bubbles $x_b$ (bottom) and by total gas $x_g$ (top) for system sizes $S = 384 \times 256$ and $S = 768 \times 512$; insets: two snapshots of the system taken right before and at the sharpest peak in the $x_b$ timeseries at $S = 768 \times 512$ (red curve, around $t = 1.3 \times 10^{10}$). (b): Linear variation of $x_g$ with $\rho_0$ (lever rule) computed for $S = 384 \times 256$. (c): Bubble fraction $x_b$ v system size $S$.

cesses to be possible once all the gas is contained in the bubbles, $x_b = x_g$, which happens at a typical crossover size $S^* \propto (x_g/x_t)^{1/[\gamma(2-\alpha)]}/x_t$. The cutoff on bubble size then reads

$$a_c^* \equiv a_c(S^*) \propto (x_g/x_t)^{1/[2-\alpha]}.$$  (3)

Equation (3) implies that $S^*$ and $a_c^*$ depend on the average density $\rho_0$ through $x_g$ and $x_t$ and that they diverge near the gas binodal $\rho_0 \to \rho_g$. On the other hand they get smaller when approaching the liquid binodal. Beyond $S^*$ the system settles in a micro-phase separated state, a homogeneous liquid with bubbles of all sizes up to $a_c^*$, SOC scaling then breaks down, and $n(a)$ becomes independent of system size.

Using our lattice gas model at high enough $\rho_0$, we are able to reach system sizes where all the gas is contained in bubbles, and the system settles in the micro-phase separated state (Fig. 2). Our data are in agreement with our scaling arguments: $n(a)$ is then independent of system size and is cut off at some scale that depends only on the average density (Fig. 2(g)). Plotting $n(a)/a_c^*$ collapses the distributions for different $\rho_0$, confirming the validity of Eq. (3) (Fig. 2(h)), at least close to the liquid binodal.

Reduced bubble model. The AMB+ field theory of Ref. 3 suggests that bubbles exist because of reverse Ostwald ripening, which causes large bubbles to shrink at the advantage of small ones, thus competing with coalescence. To test whether these ingredients are sufficient to reproduce the phenomenology described above, we implemented them in a reduced model whose degrees of freedom are the positions and radii of bubbles that we assume to be perfectly circular 30G.

The bubble-particles evolve in continuous time in a continuous domain. New bubbles with radius $r_0 = 1$ are nucleated in the liquid at a small rate $k_n$ per unit area. In line with the reverse Ostwald scenario, the new bubbles are nucleated at the expense of the larger ones: all other bubbles shrink by an amount $k_\rho(1-r_0/r)$ (where $r$ is their current radius), with $k$ chosen such that the total area of gas is conserved. (Note that this neglects spatial effects: In principle, bubbles would equilibrate in priority with neighboring ones.) Bubbles chosen randomly among the current $n(t)$ existing ones diffuse with a coefficient $D$, that for simplicity we assume constant. If the move brings the bubble into contact with another,
they merge into a single one located at their “center of mass”, conserving total area. To have the same geometry as in globally phase-separated microscopic models, we also add the possibility to have a gas reservoir outside two parallel interfaces that move along the dynamics to insure that $x_g$ remains constant.

While a complete presentation of the behavior of this reduced model and some variants will be reported elsewhere, here we show that it typically yields a phenomenology remarkably similar to that described above. Fixing $D = 1$ and $k_n = 10^{-4}$, we vary $x_g$ the total gas fraction and the system size $S$. At high $x_g$, or small system size $S$, we observe the SOC coexistence between a bubbly liquid and a gas reservoir (Fig. 4(a,b)).

The bubble size distribution scales as in Fig. 2(t) with exponent values close to those of the lattice gas ($\alpha = 1.77(2)$ and $\gamma = 1.48(5)$), but preliminary results (not shown) suggest that they are not universal. Decreasing $x_g$ or increasing $S$, the gas reservoir becomes smaller and smaller until it disappears, at which point we have a microphase separated regime with $n(a)$ independent of system size (Fig. 4(c,d)). Despite its simplicity, our bubble model thus captures the essential phenomenology described here.

**Coarsening process.** We finally study the growth of order following random initial conditions, considering the characteristic length extracted from the structure factor $20$. We only present results for our active lattice gas (Fig. 3), but similar ones, albeit of lesser quality, were obtained for ABPs. When the liquid is the majority phase, after an initial transient, the coarsening is dominated by vapor bubbles and follows an anomalous $t^{0.22}$ law. We currently lack an analytical explanation of such law. Instead, when the liquid is the minority phase, it is dominated by liquid droplets and coarsening is normal $t^{1/3}$ as expected both from Ostwald ripening and coalescence in models with conserved order parameter $37$. In fact, within the liquid droplets we expect the bubbles to coarsen as well with the anomalous law above. However, being slower than the liquid coarsening, it is not surprising that this is not visible in our data.

**Conclusion.** We have shown, using two very different models of active particles interacting strictly by pairwise repulsion, that the dense phase resulting from MIPS is critical, containing bubbles of gas distributed algebraically up to some cutoff scale. We observe at high density that, as long as an outer gas phase is present, this cutoff increases as a power of system size. At large enough system size and/or global density, the gas reservoir may disappear and the cutoff scale becomes independent of system size. This asymptotic regime is thus microphase separated. A “reduced bubble model” captures this essential phenomenology within a minimal framework that implements the basic idea of reverse Ostwald ripening put forward in Ref. $2$.

In the models presented here, the asymptotic cutoff scale $a_c^\rho$ grows very fast with $\rho_0$. Numerically, we are only able to access the asymptotic regime at rather high density (Fig. 2). What happens asymptotically at low densities thus remains unknown, but extrapolating the scaling laws uncovered here lead us to speculate that our scenario remains valid in the whole phase coexistence region $\rho_g < \rho_0 < \rho_c$.

This Letter leaves several important open questions. In particular, whether the scenario described here is observed whenever pairwise repulsion is present, and whether the critical exponents are universal could be addressed numerically by considering other models showing MIPS, the AMB+ field theory, and reduced bubble models with different parameters. In this context, the very recent work of Caporusso et al $25$, where a very hard potential between ABPs leads to crystalline clus-
ters that aggregate to form a dense phase with interstitial gas, might be understood within our scenario. Finally, in regards to the current controversy about the nature of the critical point of MIPS, our results make it unlikely that it belongs to the Ising universality class, but this hard problem remains thus unsettled.

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