

Patterns of active dipolar particles in external magnetic fields

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Active particles with a (magnetic) dipole moment are of interest for steering self-propelled motion but also result in novel collective effects due to their dipole-dipole interaction. Here systems of active dipolar particles are studied with Brownian dynamics simulations to systematically characterize the different patterns they form, specifically in the presence of an external (magnetic) field. The combination of three types of order—clustering, orientational alignment, and chain formation—is used to classify the patterns observed in these systems. In the presence of an external field, oriented chains and bands are found to be dominant. These structures show some similarities with columnar cluster seen in (passive) ferrofluids and display columnar spacing and number of lanes per cluster that both decrease with increasing field strength.

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I. INTRODUCTION

Systems of self-propelled particles as a prime example of active matter have been studied extensively for their complex collective behavior [1–3], both from the purely theoretical point of view where these systems provide examples for systems intrinsically out of equilibrium [4] and from an applied point of view where self-propelled particles are model systems for microrobots with biomedical and environmental applications [5]. From the applications point of view, the remote control of active particles is an important feature. An important method for remote control is through magnetic fields, provided that the active particles are equipped with a magnetic dipole moment [5,6]. Active particles with a magnetic dipole moment can be found in nature, in the form of magnetotactic bacteria [6,7], but can also be made fully synthetically, e.g., active magnetic colloids [8,9] and various forms of magnetic propellers and microrobots [10,11], and as biohybrid systems by magnetically functionalizing motile microorganisms [12,13].

While a large body of work has explored the magnetic steering of active particles as well as magnetically driven propulsion mechanisms [6], the effects of the magnetic interactions, both among the active particles and with an external magnetic field, have received considerably less attention. A few studies have explored the standard two-dimensional active Brownian particle model with additional magnetic interactions based on magnetic moments in the same plane as the active motion. Liao, *et al.* [14] have determined the states of active particles with dipolar interactions in the absence

of an external field and shown various patterns, in particular aggregation into chains. Similarly, our own earlier work has addressed patterns in small systems with only a few particles in confinement and studied the emergence of chains and rings [15]. These and additional structures were also found computationally as well as experimentally using centimeter-sized toy robots in a recent study with small particle numbers and harmonic confinement [16]. Moreover, motility-induced phase separation was shown to be suppressed by the magnetic interactions [14,17]. Mixtures, in which dipolar particles are not active themselves, but interact with active particles, have also been studied and also show aggregation into chains, but also large-scale clustering of the particles [18,19]. Finally, magnetic interactions were shown to result in the clustering of magnetotactic bacteria in flow through narrow channels [20,21].

In this study, we extend the previous work [14,15] to study the patterns that emerge in systems of dipolar active particles in the presence of an external field. We use systematic computer simulations to study the ordered states in such system as a function of the density, field strength, dipole moment and activity. We distinguish up to eight different states based on three order parameters introduced by Liao, *et al.* [14] that describe the competition between chaining, clustering and orientation. As a reference, we start with the case without the external field and study how a magnetic field modulates these states. We show that under strong fields, bands of oriented particles are formed that constitute active particle equivalents of columnar clusters known from passive ferrofluids [22–24].

II. METHODS

A. Model

We consider spherical active Brownian particles in two dimensions that have a permanent dipole moment $\mu_i = \mu \hat{e}_i$ with the magnetic strength μ at the particle center. Here and in the following, the superscript "hat" denotes a unit vector.

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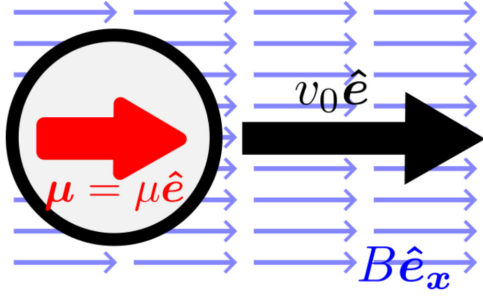


FIG. 1. Schematic illustrating single active particle with orientation $\hat{e}$ in external homogeneous magnetic field $B\hat{e}_x$ (blue). Particle has magnetic strength μ and self-propulsion speed v_0 (back).

The magnetic moment is aligned with the orientation of the particle $\hat{e}_i$, which defines the direction of self-propulsion with speed v_0 (Fig. 1). The equations of motion for a particle i with position $\mathbf{r}_i$ and orientation $\hat{e}_i$ are given by

$$\dot{\mathbf{r}}_i = v_0 \hat{e}_i + \frac{1}{\gamma_T} \mathbf{F}_i + \sqrt{2D_T} \xi_i^T, \quad (1)$$

$$\dot{\hat{e}}_i = \frac{1}{\gamma_R} \boldsymbol{\tau}_i \times \hat{e}_i + \sqrt{2D_R} \xi_i^R \times \hat{e}_i, \quad (2)$$

where γ_T and γ_R are the translational and rotational drag coefficients, ξ_i^T is the translational stochastic force, and ξ_i^R is the rotational stochastic torque. Both are described by Gaussian white noise with zero mean

$$\langle \xi_i^\alpha(t) \xi_j^\beta(t') \rangle = \delta_{i,j} \delta_{\alpha,\beta} \delta(t - t'),$$

where the upper index (α, β) runs over the x and y components of the translational noise as well as the rotational noise (z component), and the lower index (i, j) runs over the particles. D_T and D_R are the corresponding diffusion coefficients. $\mathbf{F}_i$ and $\boldsymbol{\tau}_i$ describe forces and torques acting on particle i . The forces $\mathbf{F}_i$ acting on particle i are given by the sum of the particle-particle interactions

$$\mathbf{F}_i = \sum_{j \neq i} \mathbf{F}_{ij}^{\text{dd}} + \sum_{j \neq i} \mathbf{F}_{ij}^{\text{WCA}}.$$

The pairwise forces between two particles i, j separated by the distance $r_{ij} = |\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j|$ consist of dipolar forces and excluded volume interactions. The dipolar forces are given by [25]

$$\begin{aligned} \mathbf{F}_{ij}^{\text{dd}} = & \frac{3\mu_0\mu^2}{4\pi r_{ij}^4} [\hat{\mathbf{r}}_{ij}(\hat{e}_i \cdot \hat{e}_j) + \hat{e}_i(\hat{\mathbf{r}}_{ij} \cdot \hat{e}_j) + \hat{e}_j(\hat{\mathbf{r}}_{ij} \cdot \hat{e}_i) \\ & - 5\hat{\mathbf{r}}_{ij}(\hat{\mathbf{r}}_{ij} \cdot \hat{e}_i)(\hat{\mathbf{r}}_{ij} \cdot \hat{e}_j)]. \end{aligned}$$

The excluded volume interactions are modeled by the Weeks-Chandler-Anderson (WCA) [26] pair potential with the interaction energy ϵ and the particle diameter σ

$$\mathbf{F}_{ij}^{\text{WCA}} = \begin{cases} 48\epsilon \frac{r_{ij}}{r_{ij}^2} \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \frac{1}{2} \left(\frac{\sigma}{r_{ij}} \right)^6 \right] & \text{if } r_{ij} < 2^{1/6} \sigma, \\ 0 & \text{if } r_{ij} \geq 2^{1/6} \sigma. \end{cases}$$

The torques $\boldsymbol{\tau}_i$ acting on the particle i result from the pairwise dipolar interactions [27] and the interaction with the external homogeneous magnetic field $\mathbf{B} = B\hat{e}_x$ (stationary

frame of reference),

$$\begin{aligned} \boldsymbol{\tau}_i = & \sum_{j \neq i} \boldsymbol{\tau}_{ij}^{\text{dd}} + \mu \hat{e}_i \times \mathbf{B} \\ = & \sum_{j \neq i} \frac{\mu_0\mu^2}{4\pi r_{ij}^3} [3(\hat{e}_i \times \hat{\mathbf{r}}_{ij})(\hat{e}_j \cdot \hat{\mathbf{r}}_{ij}) - (\hat{e}_i \times \hat{e}_j)] + \mu \hat{e}_i \times \mathbf{B}. \end{aligned}$$

B. Simulation method

The dynamics of active dipolar particles were simulated using the simulation toolkit HOOMD-blue, version 2.6 [28,29]. The active self-propulsion of the particles and their alignment in an external magnetic field were added through modules developed in-house. The project data and its parameter space was managed within a signac framework [30,31].

All simulations were carried out using $N = 1156$ particles placed in a quadratic simulation box with side length $L_x = L_y = L$ with periodic boundary conditions. This relatively small system size was chosen to keep the simulation time manageable while systematically scanning a rather large parameter space. The same system size was used in previous work without an external field, where it was also shown that cluster sizes saturated for $N > 1000$ [14]. To avoid interactions between periodic images of the particles, a cutoff distance of the order of half the system size, $r_{\text{cut}} = L/2 - \sigma$, was introduced for dipolar interactions. To recover correct rotational diffusion dynamics for spheres, we set $\gamma_R = 3\gamma_T$ within HOOMD-blue. To study the effect of density, we varied the lateral size L of the simulation box. We chose the following three density values defined by the mean area fraction $\Phi = N\pi\sigma^2/(4L^2)$: One well below ($\Phi = 0.13$, $L = 85$), one close to ($\Phi = 0.23$, $L = 63$) and one above ($\Phi = 0.57$, $L = 40$) the critical density where MIPS is expected in systems of active Brownian particles without dipolar interactions. To investigate the effect of a constant homogeneous external field on the collective dynamics of dipolar active particles, we systematically explored different combinations of the strength B of the external field and the magnetic moment of the particles.

As in our previous work [15], we used the following dimensionless units: distances are measured in units of the particle size σ and time in units of σ^2/D_0^T , the time in which a particle diffuses over a distance of its own size. The strength of the WCA interaction ϵ is used as the unit of energy, thus temperature T has units of ϵ/k_B (here, $T = 1$). All magnetic quantities are expressed relative to a reference field strength $B_0 = 1 \times 10^{-5} \text{ T}$, which is of the order of the magnetic field of the Earth. Thus, the strength of the magnetic field has units of B_0 and magnetic moments are expressed units of ϵ/B_0 . Note that with these units, the self-propulsion velocity v_0 is identical to the Peclet number $v_0\sigma/D_0^T$. Numerical integration was performed for $N_t = 10^6$ time steps with an integration time step width $\delta_t = 2 \times 10^{-5}$. Simulations were evaluated from $t = 18$ to $t = 20$ with data taken every $\Delta t = 0.1$.

C. Order parameters

In large systems of active dipolar particles, complex structures such as branches, connected networks, bands or loops within rings, and large-scale cluster formation are expected to emerge [14,18,19,32,33]. To characterize such patterns, we

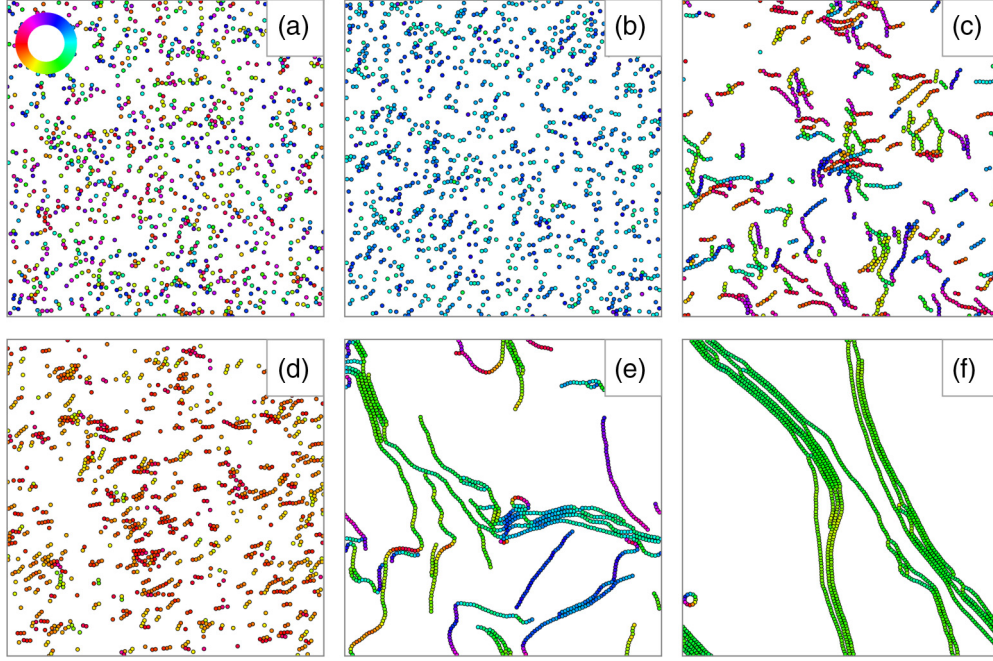


FIG. 2. Snapshots showing examples for states introduced in Table I for systems at low density ($\Phi = 0.13$), without an external field. Snapshots were taken at time $t = 20$. The following states are shown: (a) disordered gas ($\mu = 0$, $v_0 = 23$), (b) oriented gas ($\mu = 2.6$, $v_0 = 500$), (c) gas of chains ($\mu = 1.6$, $v_0 = 23$), (d) oriented chains ($\mu = 1.6$, $v_0 = 500$), (e) network of chains ($\mu = 2.6$, $v_0 = 23$), (f) bands ($\mu = 4$, $v_0 = 100$). The color wheel (top left) indicates the orientation of the particles.

used the following three global order parameters introduced by Liao, *et al.* [14]. We used combinations of these three order parameters to identify and classify different state of the system and to construct state diagrams as described below.

Similar to other active systems, we expect active dipolar particles to form large clusters. To quantify cluster formation, we determined the fraction of all particles that are part of the largest cluster by counting the number of particles n_c^* in that cluster relative to the total particle number of the system N ,

$$\phi_c = \frac{n_c^*}{N}. \quad (3)$$

The fraction of the largest cluster is close to 0 when particles form small or no clusters and reaches 1 when the whole system forms one giant cluster. To identify clusters and, specifically, the largest cluster, we performed a density based cluster analysis (DBSCAN [34]) with the value of the cutoff distance δ_c based on the nearest neighbor distance $g_1(r)$ for each corresponding density.

Orientalional order due to dipolar interactions of alignment with the external field is characterized by the global polarization, which is given by the average over the orientations of all particles in the system

$$\phi_e = \frac{1}{N} \left| \sum_i \hat{e}_i \right|. \quad (4)$$

Finally, to assess the degree of chain formation in large systems, we applied the following chain criteria, following Liao, *et al.* [14]. Two particles are considered to be bonded if they fulfill the following three criteria: First, their distance is smaller than the cutoff distance, $r_{ij} < \delta_c$. Second, they are

aligned in parallel $\hat{e}_i \cdot \hat{e}_j > 0$, and third their orientation is in line with their connecting distance vector $(\hat{e}_i \times \hat{r}_{ij}) \cdot (\hat{e}_j \times \hat{r}_{ij}) > 0$. The fraction of particles that fulfill all three criteria gives the degree of polymerization:

$$\phi_p = \frac{n_{\text{chain}}}{N}. \quad (5)$$

III. RESULTS

In this study, we investigated how a homogeneous external magnetic field affects structure formation and collective behavior of (magnetic) dipolar active particles. To this end, we performed extensive Brownian dynamics simulations of active dipolar particles in a homogeneous external magnetic field while systematically varying the field strength B , the active velocity v_0 , and the magnetic moment μ (all in dimensionless units, see Methods). We performed our simulations for three representative densities $\Phi = 0.13, 0.23$, and 0.57 . We classified the different states of the system with respect to their clustering, orientational order, and chain formation, summarizing the results in state diagrams.

A. No external field

As a reference, we first simulated active dipolar particles without an external magnetic field ($B = 0$). Here, we briefly present our results, in particular how the observed states depend on the activity v_0 and the pairwise magnetic interactions via the magnetic moment μ . This system has previously been analyzed in detail by Liao, *et al.* [14] and the following results are in agreement with their results. In simulations for different v_0 and μ and different densities Φ , we observe different patterns. Representative snapshots are shown in Fig. 2. These

TABLE I. Order parameter combinations and their corresponding states.

$\langle\phi_c\rangle > 0.5$	$\langle\phi_e\rangle > 0.5$	$\langle\phi_p\rangle > 0.5$	State
✓	✗	✗	cluster
✗	✓	✗	oriented gas
✗	✗	✓	gas of chains
✓	✓	✗	oriented cluster
✓	✗	✓	network of chains
✗	✓	✓	oriented chains
✓	✓	✓	bands
✗	✗	✗	disordered gas

snapshots reflect combinations of three different types of ordering: alignment of the magnetic moments, clustering of the particles, and chain formation. To quantify the order in these patterns, we make use of combinations of the three previously introduced global order parameters, the fraction of particles in the largest cluster $\langle\phi_c\rangle$, the global polarization $\langle\phi_e\rangle$, and the degree of polymerization $\langle\phi_p\rangle$ (see Methods). All three parameters are dimensionless, ranging from 0 to 1. For systematic classification we define a system as ordered when its order parameter exceeds 0.5, i.e., when a majority of particles adopt the corresponding ordering. This threshold provides a rigorous criterion for order. By combining the three binary (ordered/disordered) states, eight distinct state classifications arise, as summarized in Table I. We note that this classification is more detailed than the one used in previous work [14], even though it is based on the same order parameters. It allows us to systematically distinguish different types of order, which will become crucial in the presence of an external field as studied below.

At the low and intermediate densities we simulated, we can distinguish six of these states that are shown in the snapshots in Fig. 2: the disordered gas state with no order, the oriented gas with only orientational order, the gas of chains with only chain formation, oriented chains (chain formation and orientational order), a network of chains based on the

combination of chain formation and clustering, and bands in the case where all three types of order are combined. At high system density, well above the critical density for motility induced phase separation (MIPS, $\Phi_{\text{MIPS}} \approx 0.28$ [35]), we also see cluster formation in the apolar limit ($\mu = 0$) that corresponds to MIPS, as expected [14]. The eighth possible state, oriented clusters, characterized by clustering and orientational order, but no chaining is not seen in the absence of an external field.

Based on the systematic classification, we summarized our results in the diagrams of states shown in Fig. 3. For systems with densities below the critical density of MIPS ($\Phi = 0.13$ and $\Phi = 0.23$, Fig. 3 left and center) and weak magnetic interactions ($\mu < 1$) no classification criterion is fulfilled and the system is in a disordered gas state [gray circle in Fig. 3, snapshot in Fig. 2(a)]. Order (chain formation and/or global polarization) emerges for stronger magnetic interactions ($\mu \geq 1.0$). At this point, contributions from dipolar interactions equal the energy contributed by thermal noise ($\mu^2 = T = 1$) and induce chain formation, consistent with previous observations in small systems [15]. Different ordered states can be realized by varying the activity v_0 for fixed magnetic moment. For example, in the case of intermediate magnetic interactions ($\mu = 1.4$) systems with low levels of activity tend to form a gas of chains [pink triangle in Fig. 3, snapshot in Fig. 2(c)]. Increasing the activity adds global polarization to the chaining and, thus, results in oriented chains [green pentagon in Fig. 3, snapshot in Fig. 2(d)]. For even higher levels of activity, the formation of chains is suppressed while global polarization persists, so that an oriented gas is observed [purple plus in Fig. 3, snapshot in Fig. 2(b)].

When the system density exceeds the critical density of MIPS ($\Phi = 0.57$, Fig. 3 right), apolar ($\mu = 0$) and weakly magnetic particles ($\mu < 1$) also show ordered states like a network of chains [yellow square in Fig. 3, snapshot in Fig. 2(e)] or a gas of chains (pink triangle). In case of apolar particles, increasing the activity results in cluster formation (brown triangle) which has previously been identified as MIPS [14] (and was confirmed here by a bimodal distributions of

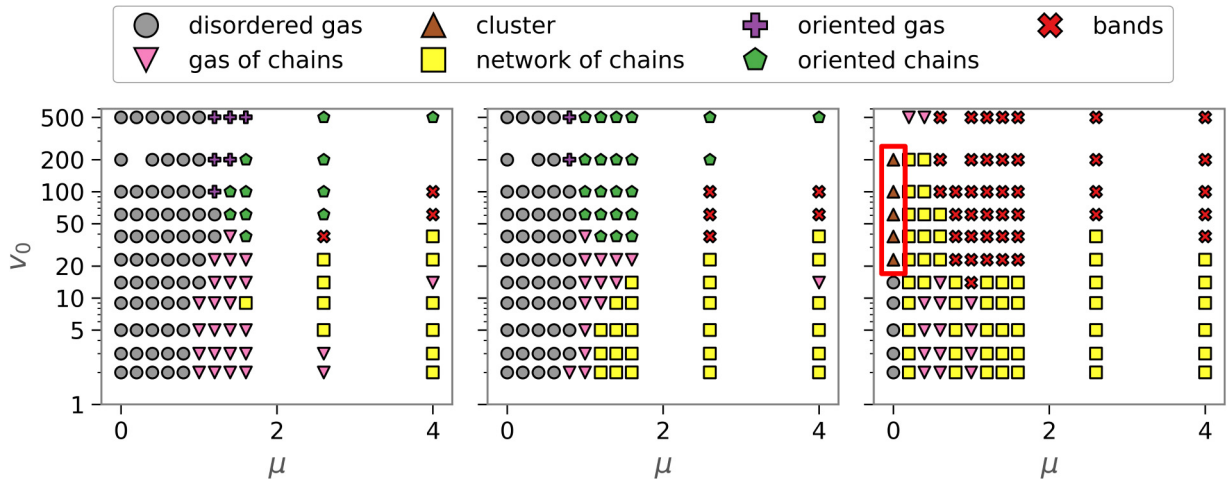


FIG. 3. Diagram of states in the absence of an external field for three densities $\Phi = 0.13, 0.23$, and 0.57 (left to right). The states result from order parameter criteria combinations according to Table I. The red box in the right diagram ($\Phi = 0.57$) highlights cluster formation that corresponds to MIPS for apolar ($\mu = 0$) active particles.

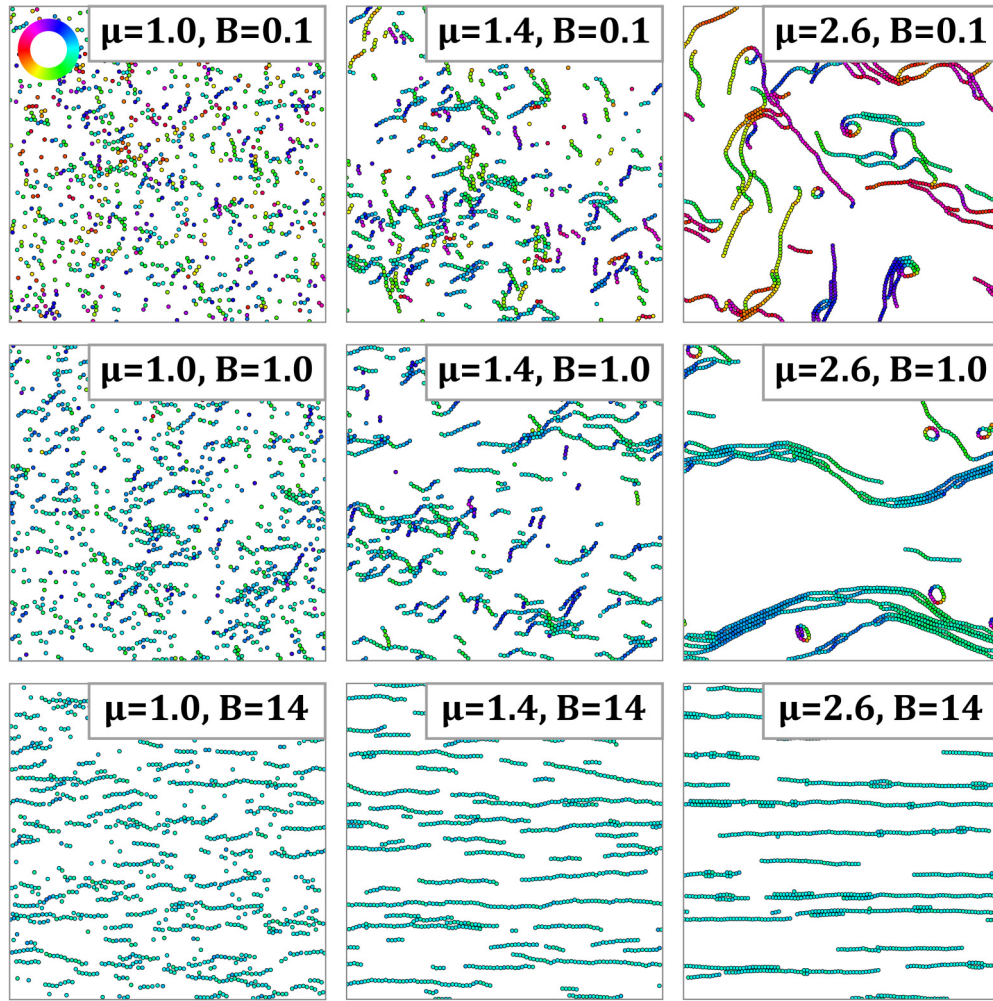


FIG. 4. Collection of snapshots of the system at low density ($\Phi = 0.13$) for intermediate self-propulsion speed $v_0 = 23$ for different magnetic field strengths B (rows) and values for magnetic interaction strengths μ (columns). External magnetic field points from left to right. All snapshots have been taken at time $t = 20$.

the local density). At high system density, network of chains (yellow square) and bands [red cross in Fig. 3, snapshot in Fig. 2(f)] dominate the diagram of states.

B. Influence of an external field

With the definitions of order parameters and the state diagram for active dipolar particles without an external magnetic field in place as a reference point, we investigated how a homogeneous external magnetic field affects those states. We first studied the lowest density simulated above ($\Phi = 0.13$), results for higher densities will be reported below. Based on the expected average orientation of a reference system ($\mu = 1$, $T = 1$), we simulated three magnetic field strength, weak ($B = 0.1$), intermediate ($B = 1$), and strong ($B = 14$), which we expect to represent different regimes.

Figure 4 shows typical system configurations for three different values of the magnetic moments $\mu = 1.0, 1.4$, and 2.6 (columns from left to right) at these three field strengths ($B = 0.1, 1.0, 14$ in the top, middle, bottom row, respectively) for particles with moderate self-propulsion speed ($v_0 = 23$). The orientation of the particles is indicated by their color

according to the color wheel. The magnetic field points from left to right, so alignment with the field can be seen by the cyan color of the particles. Systems with a weak external magnetic field (top row) display similar configurations to what was observed in systems without external magnetic field (compare with Fig. 3). In systems with intermediate external magnetic fields (middle row), particles align with the direction of the external field (as seen by their cyan color). In addition, particles assemble into chains for strong external magnetic fields ($B = 14$). These chains often have defects such as branches or small loops. Here, the average chain length is seen to increase with the magnetic moment of the particles, spanning the whole system size for strong magnetic interactions and strong external field strengths (for $\mu = 2.6$, $B = 14$, bottom right). These columnar ordered clusters are qualitatively similar to structures formed by nonequilibrium ferrofluids with external magnetic fields [23]. In contrast to systems without an external magnetic field, these columnar clusters mostly stay evenly separated and do not combine into connected networks of chains or into broad bands (at the low density simulated here, broad bands are seen for higher density, as discussed below). In fact, we observed in systems with strong

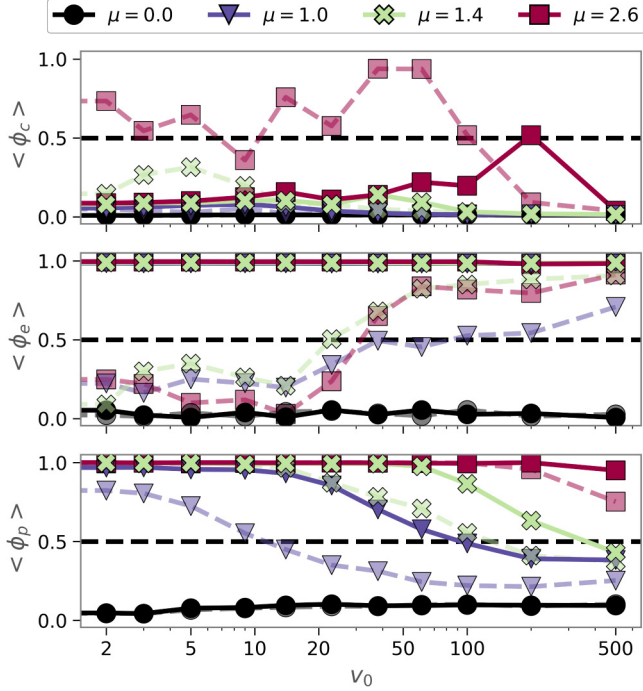


FIG. 5. Three order parameters: fraction of largest cluster (top), polarization (center), and polymerization (bottom) against self-propulsion speed v_0 for four magnetic moments μ (colors). Solid color and solid lines show data for strong external magnetic field $B = 14$, transparent colors and dashed lines show data with weak magnetic field strength $B = 0.1$. System density was set to $\Phi = 0.13$. Cutoff for cluster formation set at $\delta_c = 1.28$.

magnetic interactions ($\mu = 2.6$) that an increase of the external magnetic field strength causes broad bands to separate into thinner bands with fewer lanes or into individual chains of dipolar particles (compare the cases for $\mu = 2.6$, $B = 1.0$ and $\mu = 2.6$, $B = 14$ in Fig. 4).

We analyzed the numerical simulations quantitatively by calculating the three global order parameters introduced above. They are plotted in Fig. 5 for different μ and v_0 and for

strong and weak magnetic field. Clustering (with $\langle\phi_c\rangle > 0.5$) is largely absent at the low density used here and is only observed for the highest magnetic moment and a weak magnetic field (corresponding to the bands or networks of chains seen in the absence of a magnetic field). The strong magnetic field suppresses clustering also under these conditions (red lines).

The degree of polymerization $\langle\phi_p\rangle$ (bottom) shows a slight increase with increasing field strength. The degree of polarization $\langle\phi_e\rangle$ (middle), not surprisingly, is high for strong external fields and independent of self-propulsion speed. By contrast, it shows a transition from no polarization to strong polarization at moderate propulsion speeds for the case of the weak magnetic field. $\langle\phi_p\rangle$ decreases with increasing self-propulsion speed v_0 .

Using again the criteria based on the three order parameters, we classified the observed states of the system and summarized the results in a diagram of states, shown in Fig. 6. The figure shows the diagrams of states for the three different magnetic field strengths $B = 0.1, 1.0$, and 14 (left to right) and for the density $\Phi = 0.13$. Based on these diagrams we can make three key observations: First, disordered gas states (gray circles) are strongly suppressed with increasing magnetic field strength. In addition, the onset of the oriented gas state (purple plus) shifts to lower values of μ . Second, intermediate as well as strong magnetic fields (center and right diagram) suppress the formation of networks of chains (yellow square) and instead promote the formation of oriented states, specifically the oriented gas (purple plus), oriented chains (green pentagon) and bands (red cross, for intermediate field strength). Third, and most importantly, strong external magnetic fields suppress the formation of bands. For strong external magnetic fields (right diagram), two states dominate the diagram, the oriented gas (purple plus) and oriented chains (green pentagon). The line separating these two states resembles the corresponding line separating the gas and chain state in small confined systems of dipolar active particles investigated in previous work [15].

For strong magnetic fields, band patterns were largely absent (right panel in Fig. 6). We only found bands for two parameter combinations ($\mu = 2.6$, $v_0 = 200$ and $\mu = 4$,

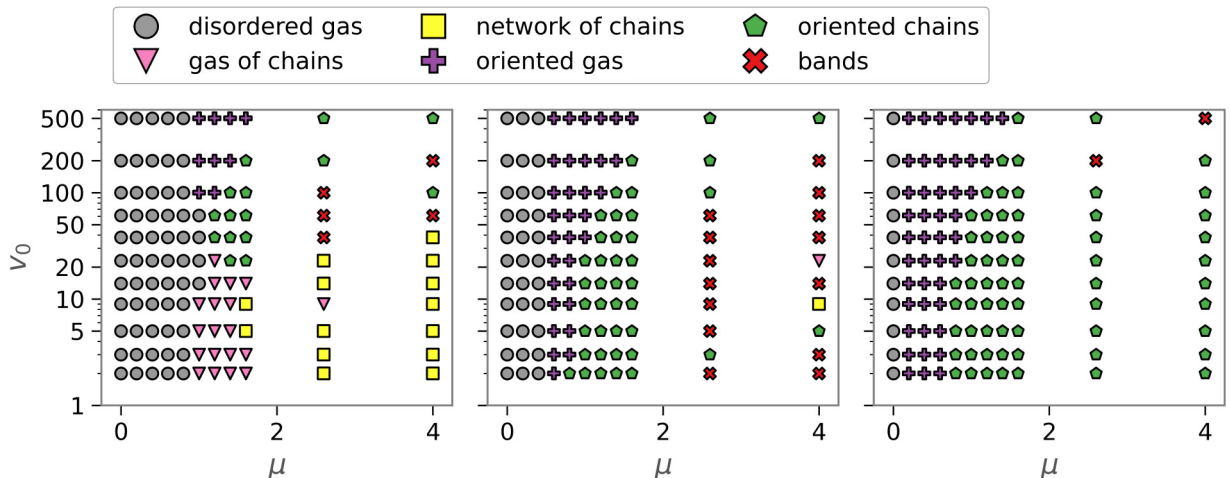


FIG. 6. Diagrams of states for three different external magnetic field strengths $B = 0.1, 1.0$, and 14 (left to right). Classification of states is based on combinations of order parameter criteria (see Table I). System at low density ($\Phi = 0.13$).

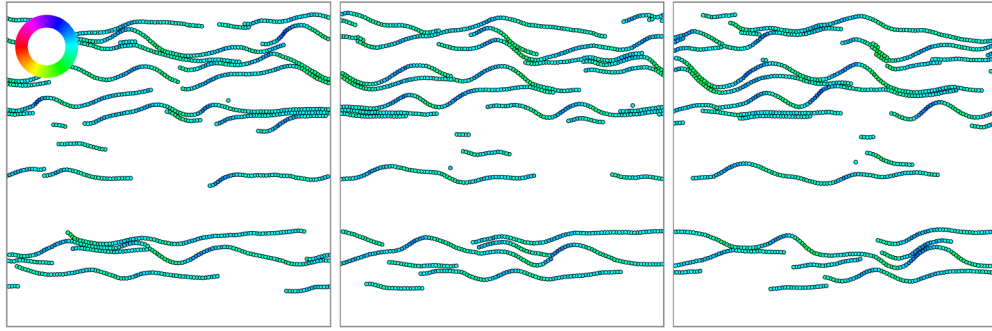


FIG. 7. Series of snapshots showing oscillating chains for dipolar active particles with strong magnetic interactions $\mu = 4$ and high self-propulsion speed $v_0 = 500$. Snapshots taken at times 26.46, 26.56, and 26.66 (left to right), color wheel indicates orientations. External magnetic field points from left to right, magnetic field strength was set to $B = 14$.

$v_0 = 500$, indicated by the red crosses in Fig. 6). In these cases we observed the following noteworthy behavior, shown by the snapshots in Fig. 7. Dipolar active particles that have self-assembled into long chains transiently show oscillations. The amplitude of these oscillations was seen to grow toward their tail of a chain. We suspect that such oscillations might be caused by a buckling instability: In the chains, particles with strong dipolar interactions are packed rather closely. The combination of high swimming speeds and translational noise can then cause a temporary compression and stress along the chain that is resolved by buckling of the chain. When the dipolar interactions were reduced, long chains were seen to break into smaller segments. These breaks typically occurred near the tail of the chain, where the oscillations reach their maximum amplitude. This observation suggests that fragmentation induced by buckling may provide a mechanism for the coexistence of chains of different lengths. We note that the breaking of chains resembles the fragmentation of passive chains observed in magnetorheological suspensions driven by a rotating field [36,37]. In our simulations these oscillating chains are seen as a transient state toward band formation. At some point, individual oscillating chains meet. Then, alignment with the neighboring particles suppresses oscillations

and a band starts to form. Again, similar lateral aggregation of individual chains into bands and columnar lanes has been observed in magnetorheological fluids [22–24,38].

C. Effect of density

To investigate the effect of the density of the system on the patterns formed by the active dipolar particles in a magnetic field, we simulated systems with two higher densities ($\Phi = 0.23$ and $\Phi = 0.57$) and repeated the calculation of the order parameters. The resulting state diagrams are shown in Fig. 8 (for $\Phi = 0.23$) and in Fig. 9 (for $\Phi = 0.57$).

In systems with the intermediate density $\Phi = 0.23$, we observed that intermediate external magnetic field strengths of $B = 1$ do not suffice to suppress formation of complex networks (yellow squares, compare the middle panel in Fig. 8 with that for low density in Fig. 6). Because of the increase in density and therefore a reduced mean particle distance, short chains may connect and form a complex network.

Furthermore, the diagram of states indicates that strong external magnetic fields diminish the effect activity has in systems at intermediate density (Fig. 8, right). Here, the line separating the oriented gas state (purple plus) and the oriented

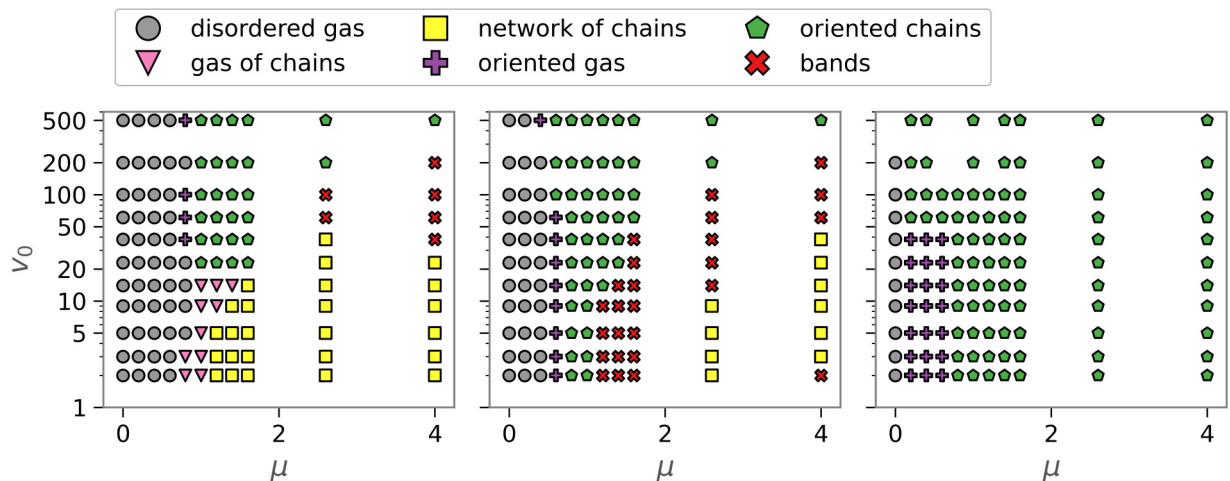


FIG. 8. Diagrams of states for three different external magnetic field strengths $B = 0.1$, 1.0 , and 14 (left to right). Classification of states is based on combinations of order parameter criteria (see Table I). System at intermediate density ($\Phi = 0.23$). Cutoff for cluster formation set at $\delta_c = 1.21$.

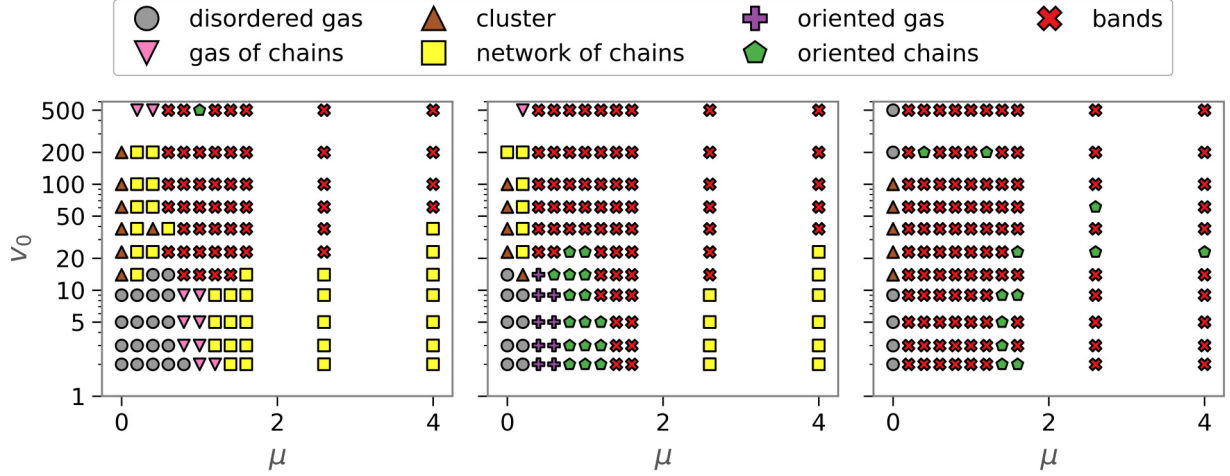


FIG. 9. Diagrams of states for three different external magnetic field strengths $B = 0.1, 1.0$, and 14 (left to right). Classification of states is based on combinations of order parameter criteria (see Table I). System at high density ($\Phi = 0.57$). Cutoff for cluster formation set at $\delta_c = 1.05$.

chains state (green pentagon) is independent of v_0 . This observation is markedly different from the previous observations for systems without external magnetic fields and to systems at low density with external magnetic fields, where activity had a strong influence on the collective behavior and structure formation.

For systems of high density (our case with $\Phi = 0.57$), three phenomena compete with one another: polar order, chain formation and MIPS, as pointed out by Liao, *et al.* [14]. The addition of an external magnetic field introduces global polar order and suppresses formation of complex networks of chains. Instead, we observed that strong external magnetic field strengths of $B = 14$ clearly organize the system with high magnetic moments into columnar structures (see Fig. 4 $\mu = 2.6, B = 14$). Because of the increase in density, these columnar structures are made up of multiple lanes in off-register alignment and not of single lanes, as seen in systems at low density.

In the diagram of states, we observed that the increased density causes the previously suppressed band states (red crosses) to reappear. In fact, bands dominate the corresponding diagram of states for strong external magnetic fields and are independent of activity (Fig. 9, right). Off-register band formation has been previously observed in passive dipolar systems with constant external magnetic fields [39–41]. In systems at high density, a clear discrimination between bands and columnar states can no longer be made. A limiting factor might be the finite system size, as observed in other systems of active particles [42].

D. Columnar clusters

At intermediate and high density and strong fields, the behavior of the system is dominated by oriented chains and bands. Snapshots of the system show additional structure in the scenarios classified as bands or oriented chains: Fig. 10 shows that as the magnetic field strength increases, a disordered network of chains first organizes into a broad band, which then, as the field strength is increased further, splits

into parallel clusters of oriented chains. These structures are aligned in the direction of the external field and, for sufficiently strong magnetic interactions, span the whole system. This effect seems to be more pronounced at the intermediate density $\Phi = 0.23$, on which we will focus in the following. Snapshots of the simulations indicate (see Fig. 10) that the number of lanes, i.e., of parallel chains in each cluster can vary and depends on the field strength. The clusters appear to be quite regularly spaced in the direction perpendicular to the external magnetic field. All these properties are reminiscent of magnetic field-induced columnar clusters in ferrofluids [22–24].

We characterized these columnar clusters with two parameters: the average number of lanes in a cluster $\bar{n}_l$ and the columnar spacing Δr_y , which is the average distance between neighboring cluster centers perpendicular to the external magnetic field. These two parameters were calculated as follows for simulations showing oriented chains or band (i.e., for simulations with $\langle \phi_e \rangle > 0.5$ and $\langle \phi_p \rangle > 0.5$): First, we identified individual clusters (color coded in Fig. 11, left), using a density based cluster algorithm (see Methods). We then calculated the center of geometry (red crosses) and the length of each cluster in direction of the external magnetic field l_x^i . Averaging the distances

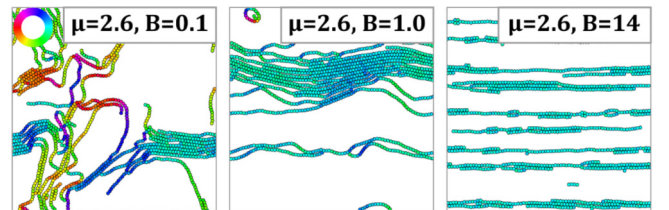


FIG. 10. Snapshots of system with intermediate density and strong magnetic interactions show columnar clusters, parallel bands spanning the whole system in the direction of the field that shrink with increasing external field. The density is $\Phi = 0.23$, the velocity $v_0 = 23$.

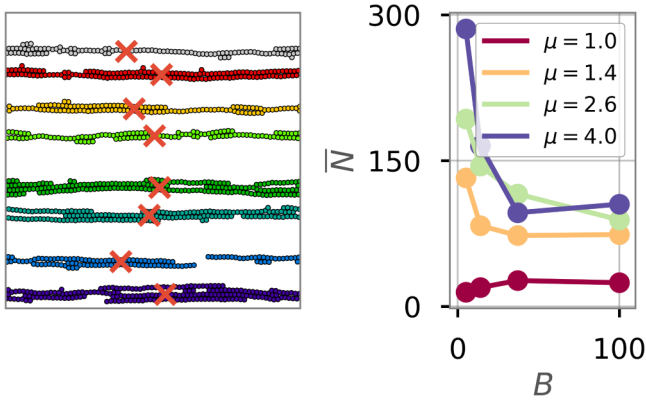


FIG. 11. Left: snapshot of system at $B = 14$, $\mu = 2.6$, $v_0 = 14$ showing columnar clusters. Individual clusters are color coded, center of geometry is marked by a red cross. Right: average cluster size $\bar{N}$ against external magnetic field strength B for particles with moderate self-propulsion speed $v_0 = 14$. System is at intermediate density $\Phi = 0.23$. Colors indicate different magnetic moments of the dipolar particles.

between neighboring centers of geometry perpendicular to the direction of the external magnetic field while accounting for periodic boundary conditions, gives us the columnar spacing Δr_y . To approximate the number of lanes in each cluster, we divided the number of particles per cluster n_i by the cluster extension in direction of the external magnetic field $n_i^l = \frac{n_i}{l_i}$.

Indeed, the average number of particles per cluster $\bar{N}$ (Fig. 11, right) decreases rapidly with increasing magnetic field strength, confirming that clusters separate into smaller sized clusters as the magnetic field strength increases in systems with $\mu > 1$. The average cluster size appears to converge to a fixed value for very strong external magnetic fields. Figure 12 shows the columnar spacing Δr_y and the average number of lanes per column $\bar{n}_l$ for systems with strong magnetic interactions $\mu = 4$. Both the columnar spacing and the number of lanes per cluster decay rapidly and approach a fixed value as the external magnetic field strength increases. Activity seems to have only a minor effect on this behavior. Columnar spacing

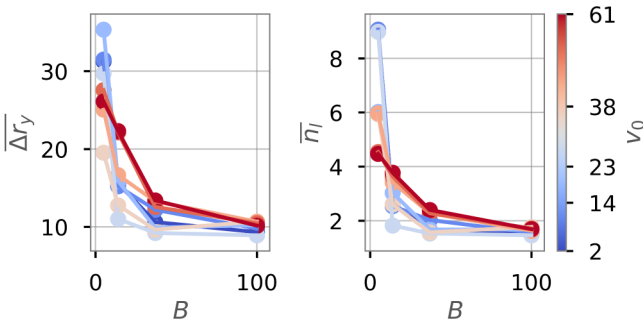


FIG. 12. Columnar spacing Δr_y (left) and average number of lanes per cluster $\bar{n}_l$ (right) against external magnetic field strength B . The magnetic moment was set to $\mu = 4$ and density to $\Phi = 0.23$. Colors indicate increasing self-propulsion speed of the particles. Data sampled over trajectories at times $t = 18-20$ every $\Delta t = 0.1$. Data points show systems that fulfill criteria $\langle \phi_e \rangle > 0.5$ and $\langle \phi_p \rangle > 0.5$.

has been reported to be independent of the external magnetic field in experiments with (passive) ferrofluids [24]. Our result for active particles at for strong magnetic fields mirror this behavior. However, we observed a strong dependence of the columnar spacing in the regime of weak external magnetic fields. We note that in some cases, a weak external magnetic field seems to promote separation between columnar structures.

As reported in Ref. [24], columnar spacing is reduced with increasing density in ferrofluids. While we did qualitatively observe in movies of our simulations that the number of lanes in each column increases with density, more systematic investigation of density effects would be needed to make quantitative statements about the influence of density on columnar structures.

IV. CONCLUSIONS

In this study, we used extensive simulations to investigate the effect of a homogeneous external magnetic field on systems of active dipolar particles. Based on the combination of three order parameters, we determined diagrams of state for different magnetic field strengths to provide a comprehensive overview of the collective behaviors observed in such system. Consistent with previous work [14,15], we see that dipolar interactions without an external field result in chains, which at higher densities form networks and bands. In our numerical experiments, motility-induced phase separation is only observed in the absence of dipolar interactions. Adding dipolar interactions, even with the weakest interaction strength we simulated, resulted in a network of chains instead. Therefore, we conclude that dipolar interactions likely prevent the directionally disordered jamming of particles and thereby inhibit the slow-down required for motility-induced phase separation.

Under the influence of an external magnetic field oriented chains are dominant and bands are seen less frequently. For weak interactions, an oriented gas of individual dipoles is observed. Both states are only weakly dependent on the level of activity and practically independent of it when a strong external magnetic field is present. This observation indicates that for strong field, the magnetic interactions dominate over activity, such that systems of active dipolar particles resemble passive ferrofluids more than classical (nondipolar) active Brownian particles. This observation for strong field stands in stark contrast to the chain formation observed in systems without external magnetic fields, where the activity plays a crucial role in collective phenomena [14,15,18].

Two observations may merit further research in future work: (i) We observed oscillating chains as a transient state toward band formation in our system. We suspect that these oscillations are caused by a buckling instability and that fragmentation of long chains due to buckling allows chains of different lengths to coexist in dilute systems, despite high magnetic moments. The self-assembled active dipolar chains we study here have some similarities to active polymers, in which the monomers are linked by a spring potential [43]. In that case, buckling has been studied both theoretically and experimentally [44–47] and was shown to be controlled by the flexure number, which characterizes the relation between active propulsion and bending rigidity [44,45].

(ii) We observed that active dipolar particles form columnar clusters that resemble structures induced by external magnetic fields in (passive) ferrofluids [22–24]. A quantitative analysis showed that the number of lanes per column and the columnar spacing decreases rapidly with increasing magnetic field strength, but become constant for strong fields. The strong-field behavior is similar to observations in ferrofluids, where columnar spacing has been reported to be independent of the external magnetic field strength [22,24]. Further features of the system and experimental protocols (such as the application of the external magnetic field, presence of hydrodynamic interactions, polydispersity and boundary conditions) may have an effect on this result. In a first step, we have shown here that activity does not strongly affect separation between columns. In future studies, the effect of density could be investigated in more detail and compared to observations for ferrofluids [22].

Our results together with those from previous studies [14,17] show that a preferred orientation (introduced by the external field) coupled to activity can significantly change the observed collective behaviors. In our model, activity is introduced as an active force acting in the direction of the magnetic moment of the particle. Therefore, orientation competes with the formation of structures based on random or conflicting

directions of self-propulsion such as MIPS. These observations indicate that the details of how activity is introduced into the system and whether or how the direction of self-propulsion is coupled to the direction of the dipoles should be important for the type of patterns and collective phenomena that are observed. They suggest that even richer collective phenomena are possible if there is loose coupling between the dipolar and the self-propulsion direction of the active particles.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [48,49].

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