



Active turbulence in a gas of self-assembled spinners

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Colloidal particles subject to an external periodic forcing exhibit complex collective behavior and self-assembled patterns. A dispersion of magnetic microparticles confined at the air–liquid interface and energized by a uniform uniaxial alternating magnetic field exhibits dynamic arrays of self-assembled spinners rotating in either direction. Here, we report on experimental and simulation studies of active turbulence and transport in a gas of self-assembled spinners. We show that the spinners, emerging as a result of spontaneous symmetry breaking of clock/counterclockwise rotation of self-assembled particle chains, generate vigorous vortical flows at the interface. An ensemble of spinners exhibits chaotic dynamics due to self-generated advection flows. The same-chirality spinners (clockwise or counterclockwise) show a tendency to aggregate and form dynamic clusters. Emergent self-induced interface currents promote active diffusion that could be tuned by the parameters of the external excitation field. Furthermore, the erratic motion of spinners at the interface generates chaotic fluid flow reminiscent of 2D turbulence. Our work provides insight into fundamental aspects of collective transport in active spinner materials and yields rules for particle manipulation at the microscale.

active turbulence | self-assembly | magnetic colloids | spinners

Turbulent fluid motion can be found across multiple length and time scales. It has fascinated scientists for centuries and still poses a major challenge for theoretical physics (1, 2). The well-known high-Reynolds number hydrodynamic turbulence in three dimensions is triggered by energy injection at the macroscale and cascading of energy to smaller scales. As an extension, the notion of active turbulence was recently introduced in the context of active fluids exemplified by suspensions of swimming bacteria, mixtures of microtubules and molecular motors, and other nonequilibrium systems (3–10). In contrast to hydrodynamic turbulence, the complex spatiotemporal behavior is caused by energy injection at the microscopic scale and subsequent cascading of energy toward larger scales. Active turbulence formally occurs at exceedingly small Reynolds numbers, rendering the fluid inertia negligible. Not surprisingly, statistical properties of active turbulence appear to be different from its classical counterpart. Active turbulence does not exhibit a wide inertial range (4, 11), and a nonuniversal power-law behavior at large scales was recently reported (12).

A related problem is diffusion and transport in active systems (13–17). Active bacterial baths (13, 15), chemically propelled catalysts (18), field-driven colloids (19–21), or even macroscopic entities such as fish, insects, or birds (22, 23) are examples of active systems where the units driving the motion generate local forces that overwhelm the thermal agitation (if any is observable in the first place). Such systems exhibit not only a wealth of directed collective behavior but also regimes where the collective motion is on average nondirectional, which gives rise to active (self-driven) diffusion.

A predictive description of *active* fluids is challenging due to the complexity of the individual building blocks (e.g., bacteria, molecular motors, etc.). In this respect, a simple physical model system, where interactions between particles are well-characterized, is highly desirable. Suspensions of colloidal par-

ticles energized by external fields provide a unique opportunity to model active systems in a well-controlled environment. This was first demonstrated at the macroscopic level in a system of magnetized disks suspended at a liquid–air interface and powered by a rotating magnetic field. Same-wise rotation of particles resulting in stable ordered phases similar to crystals (24, 25) has been observed. Similarly, computer simulations of spinning discs (26, 27) and dumbbells (28) in two dimensions yield various ordered and disordered states. Studies of ferromagnetic colloids confined at the interfaces and energized by an alternating magnetic field demonstrated a wealth of self-organized phenomena, from the formation of dynamic clusters and self-propelled entities (magnetic snakes, asters) (29–31) to rollers (32, 33) and self-assembled spinners (34). Interfacial spinners generated by a uniaxial alternating magnetic field emerge as a result of spontaneous symmetry breaking of clock/counterclockwise (CW/CCW) rotations. The self-assembled spinners inject the energy by a torque transfer via generation of local vortex flows. The energy injection rate and the corresponding injection scale can be tuned by the frequency and amplitude of the applied magnetic field.

Here, we report on an experimental and computational study of active turbulence and transport in a system of self-assembled ferromagnetic spinners. We find that the spinners and added inert particles exhibit active diffusion (diffusive motion is promoted by the activity of the system), while the diffusion arising from thermal noise is negligible in our system. We show that the active diffusion coefficient increases nearly linear with the spinner density and is approximately independent of the frequency of the driving magnetic field. We reveal a nonmonotonic

Significance

Turbulent fluid motion is widespread in nature and is observed across diverse length and time scales, ranging from high-Reynolds number hydrodynamics to active fluids, such as bacterial suspensions and cytoskeletal extracts. It is recognized as one of the unsolved challenges in theoretical physics. Here, we explore out-of-equilibrium magnetic colloidal particles at liquid interfaces that exhibit complex collective behavior, resulting in emergence of an active spinner phase. Self-assembled spinners (active spinning without self-propulsion) induce vigorous vortical flows, demonstrating the properties of a 2D hydrodynamic turbulence. Our findings provide insight into the behavior of active spinner liquids and ways to control the collective dynamics and transport in active colloidal materials.

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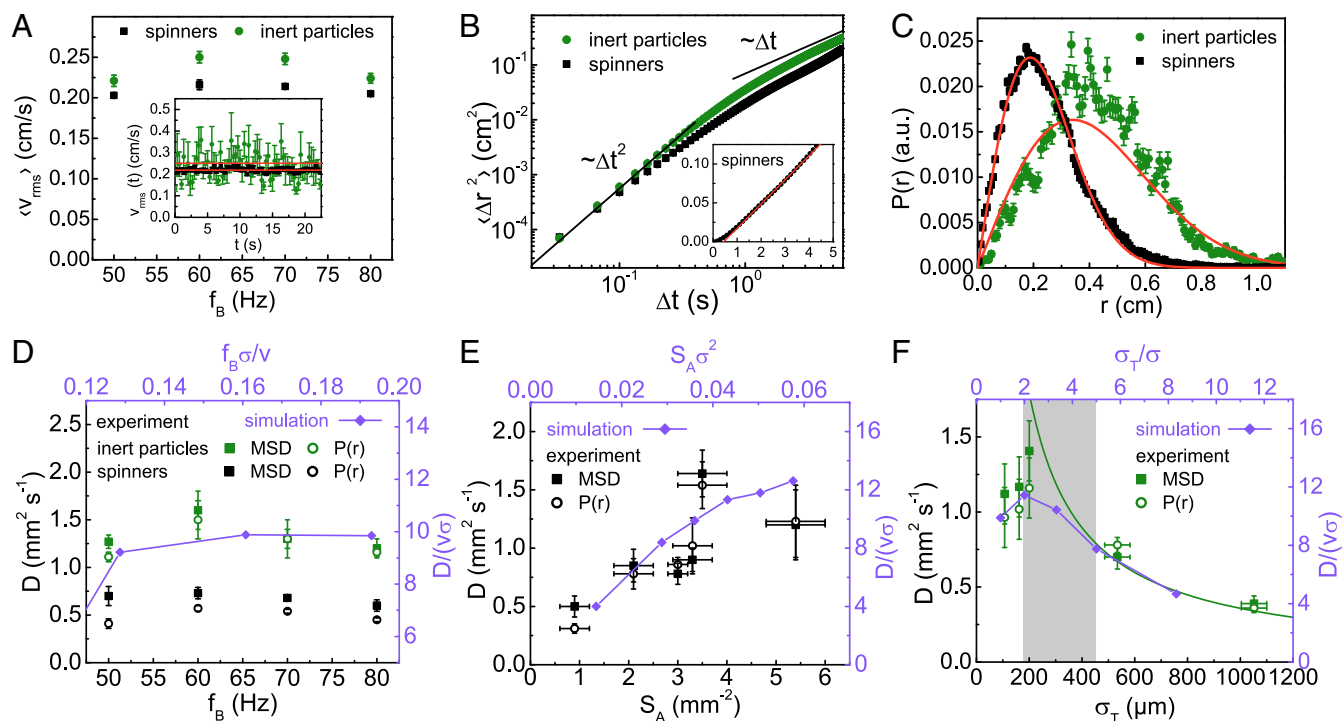


Fig. 3. Active transport and diffusion. (A) The dependence of the time-averaged velocity $\langle v_{rms} \rangle$ on the frequency f_B for spinners (squares) and inert particles (circles). (Inset) A typical time evolution of the rms velocity v_{rms} at $f_B = 60$ Hz; the red line is a linear fit. (B) MSDs for spinners (squares) and inert particles (circles). The black lines illustrate ballistic ($\propto \Delta t^2$) and active diffusion, with the same scaling as normal diffusion ($\propto \Delta t$). (Inset) The spinner MSD on linear scales. The red line is a least squares fit to Eq. 1 for the active diffusive part of the curve. (C) Radial probability density function $P(r)$ in the active diffusive regime for spinners (squares, $t = 3$ s) and inert particles (circles, $t = 4$ s). Red lines are least squares fits to Eq. 2. (A–C) $f_B = 60$ Hz, $S_A = 3.5 \pm 0.5$ mm⁻², and $\sigma_T = 500 \pm 20$ μm. (D) Frequency dependence of the active diffusion coefficient for spinners (solid symbols) and inert particles (open symbols) as obtained from the MSD (squares) and $P(r)$ (circles); $S_A = 3.5 \pm 0.5$ mm⁻² and $\sigma_T = 500 \pm 20$ μm. Results of simulations (violet) are shown for comparison. (E) Active diffusion coefficient as a function of the active particle number density S_A for inert particles as obtained from experiments [MSD, squares, $P(r)$; circles, $f_B = 60$ Hz and $\sigma_T = 500 \pm 20$ μm] and simulations (violet). (F) The active diffusion coefficient is a nonmonotonic function of the inert particle size for experiments (green; $f_B = 60$ Hz and $S_A = 3.0 \pm 0.2$ mm⁻²) and simulations (violet). The green line indicates the dependence $\propto 1/\sigma_T$. The gray area corresponds to the range of spinner sizes. (A–F) $B_0 = 2.7$ mT.

(Fig. 3A, Inset). Correspondingly, the time average value $\langle v_{rms} \rangle$ of inert particles is $\sim 10\%$ larger than that of spinners. In the frequency range of the spinner phase, $\langle v_{rms} \rangle$ depends only weakly on the frequency of the applied external field.

A characteristic velocity scale can be estimated from the Stokes flow around a spherical (disk-like) particle of diameter L_s (spinner length), which is given by $v(\bar{r}) = \pi f_B L_s (L_s/2\bar{r})^{\delta-1}$ in δ dimensions. Here, the typical distance $\bar{r}$ of inert particles and spinners is determined by the spinner concentration—that is, $\bar{r} \sim 1/\sqrt{S_A}$, where S_A is the colloid number density. For the experimental parameters (see SI Appendix), this implies $v \approx 0.13$ cm s⁻¹, in reasonable agreement with Fig. 3A. Note that the flow field decays less rapidly in strict two dimensions opposite to the quasi-2D experimental situation with 3D hydrodynamics, which implies larger characteristic velocities in simulations.

To characterize activity-induced transport in the system, we determined the diffusion coefficient D for spinners and inert particles via the mean square displacement (MSD)

$$\langle \Delta r^2 \rangle = 4D\Delta t, \quad [1]$$

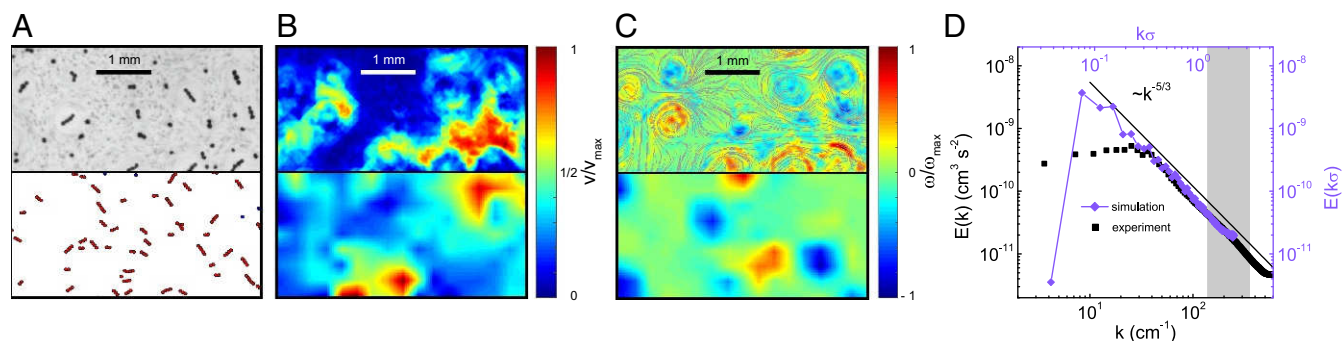
from both experimental and simulation results, as well as the probability distribution function

$$P(r, t) \propto r \exp\left(-\frac{r^2}{4Dt}\right) \quad [2]$$

from experiments, where r is the displacement at time t . Fig. 3B illustrates the initial ballistic motion followed by a crossover to

free diffusion. No anomalous diffusion was observed. The time scale for the cross-over between ballistic and diffusive regime is set by the spinner mean free time (time between a collision with another spinner or a free particle). As discussed before, the $\langle v_{rms} \rangle$ is larger for inert particles than spinners. Consequently, the diffusion coefficient for inert particles is larger than that for spinners (Fig. 3D). We attribute this to hindered spinner motion due to their strong magnetic and hydrodynamic interactions with their neighbors. Simulations yield a similar behavior (see SI Appendix, Fig. S6). However, the lifetime of the spinners in simulations just exceeds the crossover time between ballistic and diffusive motion such that no pronounced diffusive regime is obtained and no spinner diffusion coefficient can be extracted. In contrast, a clear diffusive regime is obtained for inert particles.

Diffusion coefficients for the spinners and inert particles are displayed in Fig. 3D as a function of the frequency f_B . D values were extracted independently from the MSD and the displacement distribution functions $P(r)$ (Eq. 2) (see also Fig. 3C). The latter figure shows that long-time displacements are well described by a Gaussian stochastic process. There is a good agreement between the values extracted by the two methods and also qualitative agreement with simulation results. We attribute the frequency independence of the diffusion coefficient to a competition between a faster rotation leading to faster fluid motion and the decreasing spinner length with increasing frequency f_B of the field (experiments and simulation show a similar trend; see SI Appendix, Fig. S4).



To investigate the dependence of diffusion on activity in the system, we analyzed the inert particle diffusion coefficient at different active particle number densities S_A . Obtained results are shown in Fig. 3E. The inert particle diffusion coefficient exhibits a monotonic increase with the number density until the system becomes too dense to sustain the spinner phase (immobile agglomerates of magnetic particles are formed for high number densities) (34). The observed nearly linear dependence qualitatively resembles previously observed enhanced tracer diffusion in suspensions of swimming microorganisms (13, 15, 43, 44). However, we have to keep in mind that Reynolds numbers in suspensions of swimming microorganisms are typically much smaller than unity, whereas here $Re \approx 30$. Furthermore, there are significant differences in the origin of the emerging flow fields: Swimmers usually exhibit a characteristic dipolar flow field, while spinners create a rotational flow field.

To gain additional insights on activity-induced transport in active spinner material, we explored the inert particle size dependence of the diffusion. For inert particles larger than spinners (particle diameter $\sigma_T >$ spinner length), the inert particle diffusion coefficient follows the Stokes–Einstein relation $D \propto 1/\sigma_T$ (Fig. 3F). Hence, the stirred fluid appears as a random, white-noise environment. Remarkably, for smaller particles, the trend is inverted, and the diffusion coefficient decreases with size. The nonmonotonic dependence indicates a change in the statistical properties of the ambient fluid. A diffusion coefficient independent of particle size has been obtained, for example, for particles embedded in an active fluid with temporal exponentially correlated noise (45). In contrast, for larger particles, the fluid acts as a thermal bath. Similarly, a nonmonotonic size dependence of particle diffusion was recently observed in bacterial suspensions (44). The monotonic dependence breaks down once a particle size becomes comparable with a characteristic fluid flows scale. In the case of our system, this scale approximately corresponds to the size of a spinner, while in a bacterial suspension (44), it is determined by a typical size of self-organized bacterial flows. Moreover, our results imply that there is an optimal passive particle size for fastest mixing for a given active system, indicated by the maximum diffusion coefficient. These findings clearly demonstrate that active transport can be tuned. Simulations are in good qualitative agreement with the observed experimental trends (Fig. 3D–F). The results of simulations are presented as dimensionless quantities, with σ and σ/v as relevant length and time scales. It is important to note, however, that the flow fields in three (experiment) and two dimensions (simulations) imply different velocities.

ties scales (as discussed above); therefore, a quantitative match is not expected. Despite that, both experiments and simulations yield the same dependencies of diffusion coefficients on the frequency, active particle density, and inert particle size (Fig. 3 *D–F*).

Energy Spectra. The magnitude of the hydrodynamic velocity field, induced by the rotating spinners (Fig. 4A), illustrates that the flows are concentrated around the spinners (Fig. 4B). To further investigate the self-induced interface flows in the spinner phase, we calculate the energy spectrum of turbulent fluctuations in our system. A typical energy spectrum $E(k)$ of the flows as extracted from experiments is shown in Fig. 4D. It resembles that of an inverse energy cascade in 2D turbulence (1). The broad energy-injection scale (gray area in Fig. 4D) arises from a spinner-size heterogeneity. Although the size constraints of our experimental and simulation systems limit the values of the accessible wave numbers k , a characteristic power-law behavior can be clearly observed over more than an order of magnitude in length scale. The power-law decay $k^{-5/3}$ of the energy spectrum showed no dependence on spinner density within the boundaries of the spinner phase (see also [SI Appendix, Fig. S7](#)) and corresponds to that typically found in a high-Reynolds number turbulence.

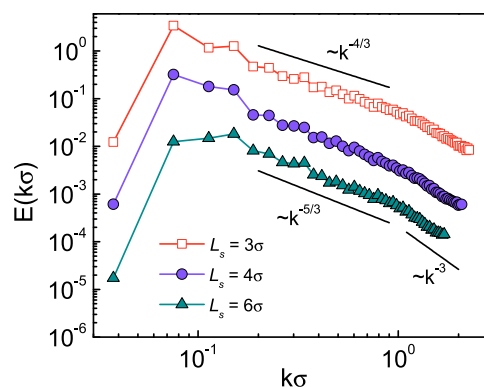


Fig. 5. Energy spectrum for monodisperse spinners from simulations. Shown are spectra for spinners of lengths $L_s/\sigma = 3, 4$, and 6 at the spinner packing fraction $\phi_s = 0.113$. In all cases, the Reynolds number is $Re \approx 38$. The top and bottom curves are shifted vertically by a constant factor with respect to the middle curve, for better distinction.

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1. Boffetta G, Ecke RE (2012) Two-dimensional turbulence. *Annu Rev Fluid Mech* 44: 427–451.
2. Xu H, et al. (2014) Flight–crash events in turbulence. *Proc Natl Acad Sci USA* 111: 7558–7563.
3. Dombrowski C, Cisneros L, Chatkaew S, Goldstein RE, Kessler JO (2004) Self-concentration and large-scale coherence in bacterial dynamics. *Phys Rev Lett* 93:098103.
4. Wensink HH, et al. (2012) Meso-scale turbulence in living fluids. *Proc Natl Acad Sci USA* 109:14308–14313.
5. Sokolov A, Aranson IS (2012) Physical properties of collective motion in suspensions of bacteria. *Phys Rev Lett* 109:248109.
6. Dunkel J, et al. (2013) Fluid dynamics of bacterial turbulence. *Phys Rev Lett* 110:228102.
7. Sanchez T, Chen DT, DeCamp SJ, Heymann M, Dogic Z (2012) Spontaneous motion in hierarchically assembled active matter. *Nature* 491:431–434.
8. Snezhko A (2011) Non-equilibrium magnetic colloidal dispersions at liquid–air interfaces: Dynamic patterns, magnetic order and self-assembled swimmers. *J Phys Condens Matter* 23:153101.
9. Angelini TE, et al. (2011) Glass-like dynamics of collective cell migration. *Proc Natl Acad Sci USA* 108:4714–4719.
10. Solis KJ, Martin JE (2014) Complex magnetic fields breathe life into fluids. *Soft Matter* 10:9136–9142.
11. Belkin M, Snezhko A, Aranson I, Kwok WK (2009) Magnetically driven surface mixing. *Phys Rev E* 80:011310.
12. Bratanov V, Jenko F, Frey E (2015) New class of turbulence in active fluids. *Proc Natl Acad Sci USA* 112:15048–15053.
13. Wu XL, Libchaber A (2000) Particle diffusion in a quasi-two-dimensional bacterial bath. *Phys Rev Lett* 84:3017–3020.
14. Tierno P (2012) Depinning and collective dynamics of magnetically driven colloidal monolayers. *Phys Rev Lett* 109: 198304.
15. Sokolov A, Goldstein RE, Feldchtein FI, Aranson IS (2009) Enhanced mixing and spatial instability in concentrated bacterial suspensions. *Phys Rev E Stat Nonlin Soft Matter Phys* 80:031903.
16. Deutsch A, Theraulaz G, Vicsek T (2012) Collective motion in biological systems. *Interface Focus* 2:689–692.
17. Maier FJ, Fischer TM (2016) Transport on active paramagnetic colloidal networks. *J Phys Chem B* 120:10162–10165.
18. Wang W, Duan W, Ahmed S, Sen A, Mallouk TE (2015) From one to many: Dynamic assembly and collective behavior of self-propelled colloidal motors. *Acc Chem Res* 48:1938–1946.
19. Martin JE, Snezhko A (2013) Driving self-assembly and emergent dynamics in colloidal suspensions by time-dependent magnetic fields. *Rep Prog Phys* 76: 126601.
20. Yan J, Bloom M, Bae SC, Luijten E, Granick S (2012) Linking synchronization to self-assembly using magnetic janus colloids. *Nature* 491:578–581.
21. Visser T, van Blaaderen A, Imhof A (2011) Band formation in mixtures of oppositely charged colloids driven by an ac electric field. *Phys Rev Lett* 106:228303.
22. Buhl J, et al. (2006) From disorder to order in marching locusts. *Science* 312:1402–1406.
23. Cavagna A, et al. (2010) Scale-free correlations in starling flocks. *Proc Natl Acad Sci USA* 107:11865–11870.
24. Grzybowski BA, Stone HA, Whitesides GM (2000) Dynamic self-assembly of magnetized, millimetre-sized objects rotating at a liquid–air interface. *Nature* 405:1033–1036.
25. Climent E, Yeo K, Maxey MR, Karniadakis GE (2007) Dynamic self-assembly of spinning particles. *J Fluids Eng* 129:379.
26. Goto Y, Tanaka H (2015) Purely hydrodynamic ordering of rotating disks at a finite Reynolds number. *Nat Commun* 6:5994.
27. Götze IO, Gompper G (2010) Flow generation by rotating colloids in planar microchannels. *Europhys Lett* 92:64003.
28. van Zuiden BC, Paulose J, Irvine WTM, Bartolo D, Vitelli V (2016) Spatiotemporal order and emergent edge currents in active spinner materials. *Proc Natl Acad Sci USA* 113:12919–12924.
29. Snezhko A, Belkin M, Aranson I, Kwok WK (2009) Self-assembled magnetic surface swimmers. *Phys Rev Lett* 102:118103.
30. Belkin M, Glatz A, Snezhko A, Aranson I (2010) Model for dynamic self-assembled magnetic surface structures. *Phys Rev E* 82:015301.
31. Snezhko A, Aranson IS (2011) Magnetic manipulation of self-assembled colloidal asters. *Nat Mater* 10:698–703.
32. Driscoll M, et al. (2016) Unstable fronts and motile structures formed by microrollers. *Nat Phys* 13:375–379.
33. Kaiser A, Snezhko A, Aranson IS (2017) Flocking ferromagnetic colloids. *Sci Adv* 3:e1601469.
34. Kokot G, Piet D, Whitesides GM, Aranson IS, Snezhko A (2015) Emergence of reconfigurable wires and spinners via dynamic self-assembly. *Sci Rep* 5:9528.
35. Esmaeeli A, Tryggvason G (1996) An inverse energy cascade in two-dimensional low Reynolds number bubbly flows. *J Fluid Mech* 314:315–330.
36. Lance M, Bataille J (1991) Turbulence in the liquid phase of a uniform bubbly air–water flow. *J Fluid Mech* 222:95–118.
37. Mudde R, Groen J, Van Den Akker H (1997) Liquid velocity field in a bubble column: LDA experiments. *Chem Eng Sci* 52:4217–4224.
38. Kellay H, Goldburg VI (2002) Two-dimensional turbulence: A review of some recent experiments. *Rep Prog Phys* 65:845–894.
39. Vázquez-Quesada A, Franke T, Ellero M (2017) Theory and simulation of the dynamics, deformation, and breakup of a chain of superparamagnetic beads under a rotating magnetic field. *Phys Fluids* 29:032006.
40. Snezhko A, Aranson IS (2015) Velocity statistics of dynamic spinners in out-of-equilibrium magnetic suspensions. *Soft Matter* 11:6055–6061.
41. Nguyen NH, Klotz D, Engel M, Glotzer SC (2014) Emergent collective phenomena in a mixture of hard shapes through active rotation. *Phys Rev Lett* 112:075701.
42. Yeo K, Lushi E, Vlahovska PM (2015) Collective dynamics in a binary mixture of hydrodynamically coupled microrotors. *Phys Rev Lett* 114:188301.
43. Leptos KC, Guasto JS, Gollub JP, Pesci AI, Goldstein RE (2009) Dynamics of enhanced tracer diffusion in suspensions of swimming eukaryotic microorganisms. *Phys Rev Lett* 103:198103.
44. Patteson AE, Gopinath A, Purohit PK, Arratia PE (2016) Particle diffusion in active fluids is non-monotonic in size. *Soft Matter* 12:2365–2372.
45. Eisenstecken T, Gompper G, Winkler RG (2017) Internal dynamics of semiflexible polymers with active noise. *J Chem Phys* 146:154903.
46. Groisman A, Steinberg V (2001) Elastic turbulence in a polymer solution flow. *Nature* 405:53–55.
47. Wensink H, Löwen H (2012) Emergent states in dense systems of active rods: From swarming to turbulence. *J Phys Condens Matter* 24:464130.
48. Kapral R (2008) Multiparticle collision dynamics: Simulation of complex systems on mesoscales. *Adv Chem Phys* 140:89–146.
49. Gompper G, Ihle T, Kroll DM, Winkler RG (2009) Multi-particle collision dynamics: A particle-based mesoscale simulation approach to the hydrodynamics of complex fluids. *Adv Polym Sci* 221:1.
50. Huang CC, Gompper G, Winkler RG (2012) Hydrodynamic correlations in multiparticle collision dynamics fluids. *Phys Rev E Stat Nonlin Soft Matter Phys* 86:056711.
51. Noguchi H, Gompper G (2008) Transport coefficients of off-lattice mesoscale-hydrodynamics simulation techniques. *Phys Rev E Stat Nonlin Soft Matter Phys* 78:016706.
52. Theers M, Westphal E, Gompper G, Winkler RG (2016) From local to hydrodynamic friction in Brownian motion: A multiparticle collision dynamics simulation study. *Phys Rev E* 93:032604.