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Swimming modes & interactions of anisotropic active colloids

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1.

Chapter 1

Introduction

1.1 Biological Microswimmers

Studying, understanding, and mimicking self-propulsion in nature is a central goal in the field of active matter. Biological microswimmers — such as bacteria, sperm cells, or certain algae — are inherently out-of-equilibrium systems that consume energy from their environment and convert it into directed motion[1,2]. This locomotion is typically driven by the coordinated beating of flagella or cilia[3,4]. These active agents are highly efficient in exploring their environment in the search for nutrients or in moving in response to external stimuli like light or chemical gradients[2]. The collective behavior of these microorganism can also be influenced by their autonomous motion which allows them to readily organize into colonies or biofilms for more resilience.

What many of these biological motile systems have in common is their anisotropic (non-spherical) shape, which plays a critical role in determining how they swim, reorient and interact, both with one another and with their environment[5–8]. For example, experiments on E. coli with artificially modified shapes have shown that cell morphology directly impacts cell motility: crescent-shaped cells and loosely wound spirals swim along straight trajectories, while tightly coiled spirals tend to swim in counterclockwise circles[9]. These observations highlight how geometry can affect propulsion efficiency and the dynamic behavior of microswimmers. Understanding the influence of the swimmer´s shape on its propulsion and interaction mechanisms is therefore a central motivation for the work presented in this thesis.

1.2 Synthetic Microswimmers

To better understand the fundamental biological process of self-propulsion and to explore the physics underlying these inherently non-equilibrium systems, synthetic microswimmers are often used as simplified and controllable model platforms. Unlike their very complex biological counterparts, synthetic systems allow for the isolation and systematic study of key parameters such as shape, interaction forces, and propulsion mechanism. These model systems typically consist of active colloids: micrometer-sized particles suspended in a fluid medium that exhibit autonomous motion through continuous energy consumption [10,11].

For geometries beyond simple spheres and rods, the influence of the swimmer's shape on its active motion has remained relatively underexplored in experimental studies, mainly due to the challenges associated with fabricating synthetic anisotropic colloids. Spherical microswimmers, by contrast, are the most extensively studied and best understood geometry, serving as a "gold standard" for experimental and theoretical investigations in the field. This chapter therefore begins with a brief overview of the mechanisms by which self-propulsion is generally achieved, followed by a summary of the fabrication methods and behavior of spherical microswimmers, before discussing synthetic anisotropic active particles.

1.2.1 Principles of Self-propulsion

In general, self-propulsion mechanisms can be divided into two main categories: internally driven self-propulsion, which relies on local energy conversion, for example through catalytic reactions, and externally driven propulsion, where external fields such as magnetic, acoustic, or electric fields drive the system out of equilibrium [1,12]. In the case of internally driven self-propulsion, the colloidal particle autonomously generates local gradients — such as chemical [10], electrostatic [13], or thermal gradients [14,15] — which give rise to phoretic flows that propel the particle forward. Generating these gradients requires a break in symmetry of the particle's properties, which may arise from asymmetries in material composition [10], shape [16] or surface functionalization [17]. As a result, the direction of motion typically aligns with the symmetry broken axis. In this chapter, we specifically focus on catalytic self-propulsion, as it is the propulsion mechanism employed in all experimental systems presented in this work.

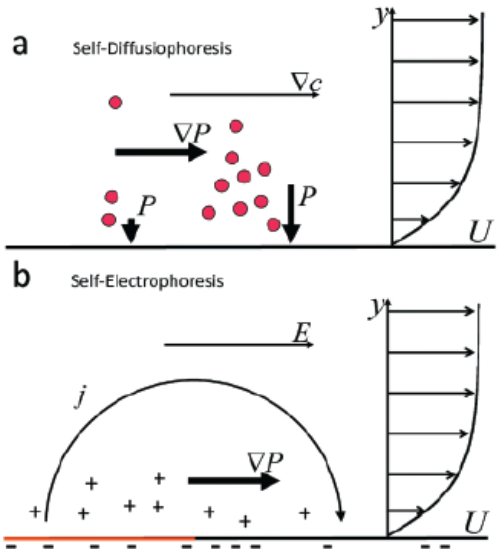

Figure 1.1: Schematic of **(a)** self-diffusiophoresis and **(b)** self-electrophoresis. Reprinted with permission from Ref. 18. © 2014 Soft Matter, Order License ID (1635936-1).

One prominent system of catalytic chemical swimmers are dielectric colloids partially coated with a thin film of catalytic material[19]. When suspended in a fuel solution such as aqueous hydrogen peroxide (H_2O_2), the local decomposition of H_2O_2 into water (H_2O) and oxygen (O_2) on the catalytic side of the particle, generates concentration gradients along the swimmer´s surface. Interactions between the reaction products and the surface generates pressure gradients, which induce phoretic flows, ultimately resulting in directed self-diffusiophoretic propulsion, see Fig. 1.1 a [10,18]. Another key example is that of bimetallic gold-platinum (Au-Pt) rods, which also self-propel in aqueous H_2O_2 solution[20,21]. Here, the decomposition of H_2O_2 occurs by a redox-reaction involving the two extremities of the rod, with protons being produced at the Pt-end and consumed at the Au-end. This establishes a proton concentration gradient and an associated local electric field, which drives electroosmotic flows along the rod's surface leading to self-electrophoretic propulsion, see Fig. 1.1 b [1,22].

The catalytic processes involved in the decomposition of hydrogen peroxide are complex and as a consequence the mechanistic details underlying self-propulsion remain a subject of ongoing debate. In particular, for dielectric Janus swimmers, observations suggest that propulsion may not be solely due to self-diffusiophoresis — as described earlier — but could also involve self-electrophoresis, similar to the mechanism proposed for bimetallic

rods[22]. For example, the strength and direction of propulsion in these systems have been found to depend sensitively on the presence of added salts and ionic surfactants[18]. Such dependencies cannot be predicted by a simple self-diffusiophoresis model based solely on neutral chemical gradients. Instead, it has been proposed that asymmetries in the catalytic film thickness on dielectric particles could give rise to ionic currents, analogous to those in bimetallic swimmers, which suggest a self-electrophoretic component in the propulsion mechanism[18,23,24].

1.2.2 Spherical Janus colloids - the current state of the art

To achieve self-propulsion, an asymmetry must be present which for isotropic spherical particles can be obtained by locally modifying their surface properties thus creating particles with two distinct hemispheres — so-called Janus particles, named after the two-faced roman god. The most widely used synthesis method for producing such particles is based on partial and temporary masking during surface treatment[1,25]. In this approach, the particles are first immobilized as a monolayer on a planar substrate, e.g. a glass cover slip. The exposed hemisphere is then coated via evaporative deposition with typically a metal catalyst (e.g., Al, Au, or Pt) or carbon. The modified colloids are subsequently released into solution, usually through sonication. This technique is also the method applied for all active colloids mentioned in this work.

Alternatively, particles can be placed at the interface of two liquid phases[26]. This configuration allows for the selective modification of the particle region embedded in one of the phases thus creating the required functional asymmetry. A related strategy was employed in the fabrication of the well-known light-activated microswimmers introduced by Palacci et al.[27], in which hematite is used as the catalytic component instead of a metal catalysts such as Pt or Pd. In this system, hematite cubes are embedded asymmetrically at the interface of TPM (3-(trimethoxysilyl)propyl methacrylate) droplets before polymerizing the droplets, resulting in solid Janus particles with hematite inclusions. The advantage of using hematite is its photocatalytic activity: it catalyzes the decomposition of hydrogen peroxide only under blue light illumination, allowing external control over the onset of propulsion.

Once active, spherical microswimmers exhibit particularly interesting collective behavior. At sufficiently high densities and even in the presence of purely repulsive interactions, they can spontaneously form clusters and undergo so-called *motility induced phase separation* (MIPS)[28–32]. The particles persistent motion leads to local crowding due to a

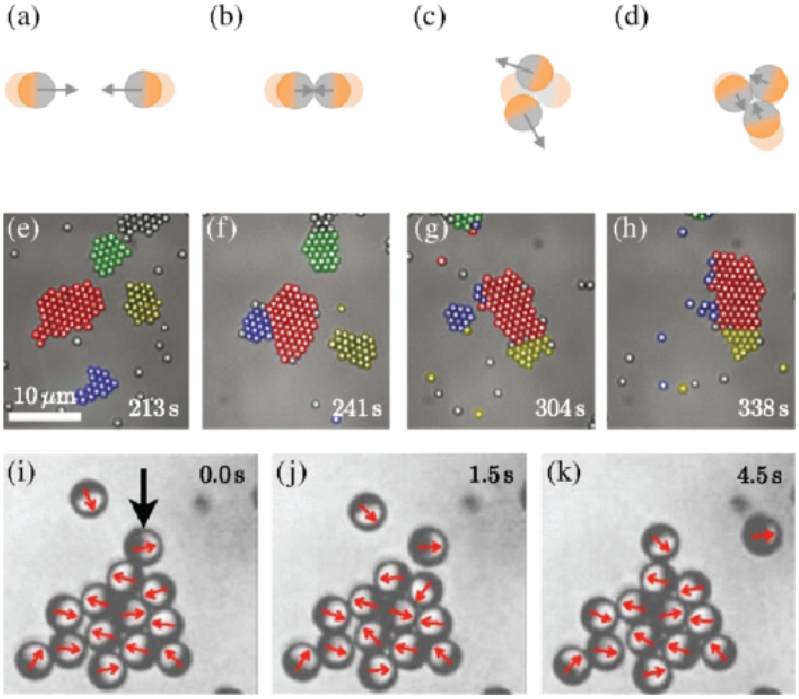

Figure 1.2: Collective behaviors of active spheres. (a) - **(d)** Qualitative explanation of the clustering process. **(e)** - **(h)** Clusters assembled from a homogeneous distribution of active particles (solid area fraction $\phi = 0.14$). The clusters are not static but rearrange, exchange particles, and merge. **(i)** - **(k)** Consecutive snapshots of a cluster of active Janus particles. Reprinted with permission from Ref. 1. © 2016 Rev. Mod. Phys., License Number (RNP/25/AUG/094032).

slowing-down of particles upon collision which results in effective aggregation. Due to their isotropic shape, spherical particles generally lack alignment interactions. In binary collisions, they therefore tend to slide past each other unless their velocity vectors are exactly anti-aligned — a configuration that is typically short-lived and disrupted on the timescale of rotational diffusion. As a result, stable cluster formation requires interactions involving at least three, but usually more, particles (cf. Fig. 1.2 a-d)[1]. In simulations of self-propelled disks, MIPS emerges above a critical packing fraction of around 40%[29], forming a single large, globally disordered macroscopic aggregate that experiences diffusive motion.

In experiments (cf. Fig. 1.2 e-k), however, additional attractive interactions, caused by the interaction of the concentration profiles around each particle, enhance aggregation and cluster formation sets in already at lower particle densities[27,33,34]. Within these multiple

disordered and dynamic aggregates, self-jamming phenomena can be observed[1,35]. Unlike passive systems, where the dynamical transition into a mechanically rigid state happens at a well-defined critical density, active matter can self-organize into locally jammed regions even below the bulk jamming density threshold of passive systems. This indicates that active systems exhibit large density fluctuations and local mechanical arrest, independent of global density.

1.2.3 Anisotropic Microswimmers - the effect of shape

Deviating from the spherical geometry has a significant impact on both the single-particle dynamics and the interparticle interactions that govern the collective behavior in active systems. Anisotropic particle shapes can affect motion at the single-particle level and give rise to complex motion patterns[36–40]. For instance, chiral trajectories emerge in geometries lacking left-right symmetry: L-shaped microswimmers have been experimentally shown to follow stable circular paths due to their asymmetry[41].

Moreover, the shape strongly influences the flow fields generated around each swimmer, which in turn directly affect the particle-particle interactions[42,43]. Upon collision, anisotropic microswimmers can align, reflect, or rotate which often leads to rich collective behaviors such as segregation, polar alignment, or the formation of dynamic micro-rotors[44–46]. For example, transversely propelled rods can form long-lived doublets, a behavior not observed in active spheres[47]. The alignment interactions and torques generated during collisions among anisotropic particles, such as rods and dumbbells, have been theoretically shown to modify[48–50] or even suppress MIPS[43,51,52], see Fig. 1.3. Longitudinally propelled rods, for instance, self-organize into polar bands rather than clusters, as their elongated shape promotes lateral alignment over aggregation[52–55].

Another key difference between anisotropic and spherical swimmers is the fact that their velocity vector can be oriented in different ways with respect to their shape, which leads to a diverse range of exciting new phenomena[56]. For example, self-propelled squares exhibit markedly different collision dynamics and cluster evolution depending on whether their propulsion is directed perpendicularly to a side or towards a corner[57]. For surface-forward particles, collisions often result in immobilization, producing smaller, less mobile and more loosely packed clusters. In contrast, corner-forward propulsion allows particles to continue navigating along the surface of a cluster after impact, leading to larger, more dynamic and denser aggregates.

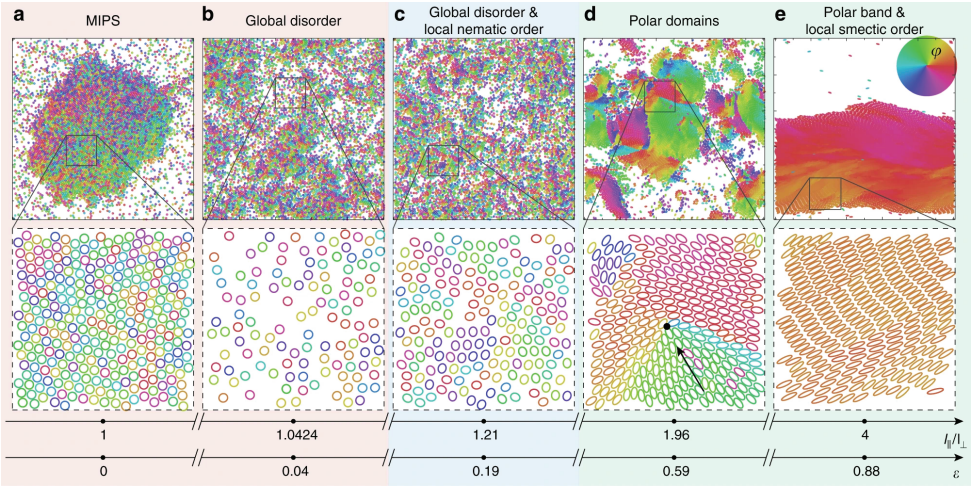

Figure 1.3: Breakdown of MIPS. Large-scale patterns for rigid self-propelled particles with increasing anisotropy ϵ and aspect ratio $l_{\parallel}/l_{\perp}$. The individual particle shape is transitioning from spheres to ellipsoids. Reprinted from Ref. 53. © 2020 Nature Communication, under the Creative Commons license.

The critical density required for cluster formation is also shape-dependent, largely because anisotropy affects how particles decelerate upon collision[46,47,58]. Recent studies demonstrated that active hexagons slow down more effectively upon contact than spheres and other polygons, leading to the formation of many small clusters at significantly lower particle densities than would be expected for isotropic particles[46].

Despite the many fascinating phenomena observed in suspensions of active anisotropic microswimmers, the effect of the swimmer shape on their behavior remains relatively underexplored experimentally and the number of experimental studies still lags behind the abundance of numerical simulations. This gap is largely due to the challenges in synthesizing geometries beyond simple shapes like rods and ellipsoids. In the following section, we review the main fabrication strategies for non-spherical colloids and discuss their potential for enabling more complex shape anisotropies.

1.2.4 Fabrication strategies to realize shape anisotropy - reaching beyond the spherical geometry

Several established techniques exist for making synthetic anisotropic colloids with relatively simple shapes like rods, ellipsoids, dumbbells, or patchy particles[59,60]. The most straightforward is the generation of ellipsoids through uniform mechanical deformation of spherical particles. This is typically achieved by stretching spherical latex beads embedded in a polymeric film at temperatures above the glass transition temperature of the particles[61].

Colloidal dumbbells can be synthesized via a seeded emulsion polymerization process, in which monomer swelling of the seed particle is followed by phase separation of a monomer droplet prior to its polymerization[59]. This approach has also been extended to obtain patchy particles by exploiting the coalescence of liquid protrusions on cross-linked latex particles, which self-assemble into colloidal molecules with directional bonding sites[62–65]. Another way to obtain colloidal clusters with sharper geometries is by combining capillary assembly with evaporation techniques. Here, colloidal particles positioned at emulsion interfaces spontaneously assemble into defined clusters driven by the capillary interactions that they experience as the dispersed phase evaporates[59,66,67]. All of these methods typically yield passive anisotropic particles. However, self-propulsion can be achieved if the particles are selectively functionalized in a follow up step to obtain chemically patterned particles. For example, catalytic activity can be introduced by applying a metal coating on the patches using solution-based protocols[39].

A more versatile approach with more freedom in terms of compositional and geometrical programmability is sequential capillarity-assisted particle assembly (sCAPA)[68]. Here, capillary forces are used to stepwise deposit colloidal particles of varying materials into predefined traps, allowing the fabrication of multifunctional colloidal clusters with organic and inorganic moieties. After deposition and mechanical linking, the assembled particles can be redispersed in solution. The key advantage of sCAPA lies in its precise control over particle placement, orientation, and composition at the microscale. The anisotropic colloidal molecules produced through this method can then be actuated by applying an AC electric field[69].

For more complex and precisely tailored shapes, template-assisted fabrication methods such as photolithography can be used[60,70]. In this approach, light is shone through a photomask onto a film of photoresist thus transferring the pattern from the mask onto

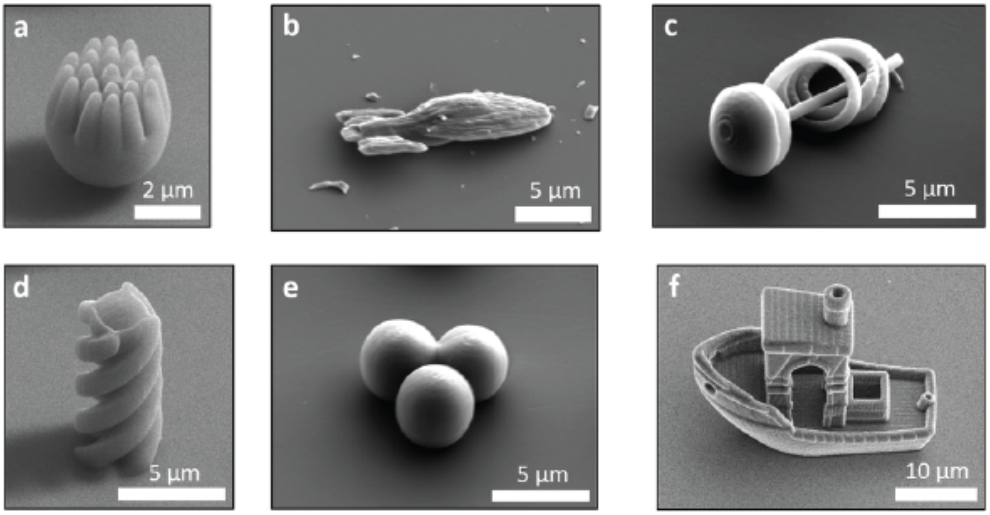

Figure 1.4: 3D microprinted anisotropic colloids. SEM images of various 3D printed particle shapes as obtained by two-photon polymerisation. **(a)** A spiky sphere, **(b)** a starship, **(c)** a spiral, **(d)** a helix, **(e)** a trimer and **(f)** 3D benchy boat. Reprinted from Ref. 36. © 2020 Soft Matter.

the film. The unexposed regions are then removed in a post-curing development step, resulting in individual colloidal particles with well-defined shapes. To improve throughput, lithography-based synthesis can be integrated in a microfluidic systems[59,71].

The fabrication method with the highest flexibility and control over the particle shape, which is also the technique applied in this work, is 3D microprinting using two-photon polymerization[36,72–74]. It enables the fabrication of complex three-dimensional structures with sub-micron resolution by focusing a femtosecond laser into a droplet of photoresist spread on a substrate, polymerizing material only at the laser's narrow focal point. For example structures see Fig. 1.4. This method also allows enhanced freedom over the active patch location since the orientation of the particle relative to the substrate can be precisely chosen during particle design[36]. After printing, the desired exposed surfaces can be selectively functionalized via sputter coating. Ultimately, this spatial control enables a fine-tuning of the propulsion direction as the particle's shape and the velocity vector can be aligned in a custom way.

1.3 In this work

In this thesis, we explore how the shape of anisotropic microswimmers influences their swimming modes, clustering behavior, and interactions with other active particles. Inspired by the *Plasmodium falciparum* malaria parasite, which enhances its transmission efficiency through its distinctive curved, crescent-shaped morphology[75,76], we focused on the fabrication and analysis of catalytically active bent rods.

In **Chapter 2**, we demonstrate how the bent rod geometry dramatically enhances out-of-equilibrium self-organization. We experimentally and numerically investigate a model class of microswimmers with a shape that can be continuously tuned from spherical to bent and straight rod configurations. We find that bent rods promote interlocking assembly even at extremely low particle densities, with semicircular shapes (crescents) showing the highest clustering efficiency. Our analysis reveals that clustering dynamics are governed by a balance between the probability of interlocking and the stability of the resulting clusters. We also report for the first time that crescent-shaped particles reverse their swimming direction at higher fuel concentrations — a phenomenon that significantly reduces their clustering efficiency.

Building on this observation, we investigate the direction reversal of crescent-shaped particles in greater detail in **Chapter 3**. We experimentally show that the swimming direction can change with increasing fuel concentration, not only for crescent-shaped particles but also for other anisotropic swimmers such as disks and tori. At low hydrogen peroxide concentrations, these swimmers propel with their inert side forward, whereas at high concentrations, they move catalytic side forward. We attribute this reversal to combined effects of pH-dependent surface properties, particle geometry, and substrate-induced solute confinement. For anisotropic prolate-like geometries and relative zeta-potential ratios of the two Janus sides close to unity, small variations in surface mobility can reverse the propulsion direction, whereas symmetric or oblate shapes require mobilities of opposite sign. The nearby substrate further confines reaction products and reshapes the concentration gradients in a shape-dependent manner, thereby amplifying this effect.

After having studied in **Chapter 2** the particle interactions in a pure crescent sample, **Chapter 4** focuses on lock-and-key type interactions of crescent-shaped swimmers with different "key" particles. We study mixtures of different types of active shape-complementary colloids and analyze the resulting pair formation. We find that the self-assembly process can

be described with a chemical equilibrium model with equilibrium constant K. We further demonstrate that K can be tuned by the shape of the key-partner and that, surprisingly, the most efficient clustering remains the crescent–crescent interaction.

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