
Optothermally Driven Colloidal Matter: Activity and Guided Motion

विद्या वाचस्पति की
उपाधि की अपेक्षाओं की आंशिक पूर्ति में प्रस्तुत शोध प्रबंध

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Dedicated to Dadu and Dida....

Certificate

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The work reported in this thesis is the original work done by me under the guidance of **Prof. G V Pavan Kumar**

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List of Publications

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Abstract

Light carries energy, linear momentum, and angular momentum, which can be directly transferred to microscopic and nanoscopic objects. This transfer allows precise control and manipulation of their motion by suppressing inherent Brownian dynamics. However, optical fields often produce temperature fields as a byproduct. These thermal fields, along with fluid-mediated effects like thermo-osmosis, thermophoresis, and convection, significantly influence the dynamics of colloidal systems at micro- and nanoscales. In this thesis, we investigate the optothermal interaction-mediated dynamics of colloidal systems at the microscale, using experimental and numerical approaches to explore the effects of optical and optothermal interactions on the dynamics of colloidal particles.

We first examined the dynamics under large-area optical fields. Such beams elevate the temperature of colloidal particles based on their individual absorption properties. These temperature differences induce attractive thermo-osmotic interactions between particles, leading to the formation of self-assembled colloidal structures. Depending on the colloidal species and their arrangements, these interactions can be reciprocal or non-reciprocal. In cases of non-reciprocal interactions, imbalances in net force and torque arise, resulting in dynamic behaviors such as propulsion and curling trajectories in the assembled structures.

Next, we explored the behavior of optothermally self-assembled colloidal structures within confined environments created by focused or structured optical potential landscapes. In focused optical fields, gradient optical forces act as a confining potential, anchoring the colloidal system to the beam's center, while optothermal (thermo-osmotic) interactions drive the formation of bound structures. These structures exhibit chiral rotational states, with their

handedness adjustable by shifting the position of the focused beam. In contrast, structured optical fields, such as ring or line-shaped beams, define pathways that guide the motion of colloidal structures. At low optical intensities, the structures follow these paths in a bound manner, but at higher intensities, repulsive thermophoretic forces dominate, altering structures and their dynamics. This dynamic state can be tuned by adjusting the incident optical field intensity.

The thesis concludes by summarizing the key experimental findings on optothermal interaction-mediated colloidal dynamics. We emphasize how optical fields can both confine and drive colloidal motion, with thermal effects playing a critical role in shaping these behaviors. Finally, we outline potential avenues for extending these results in future studies, offering insights into broader applications and refinements of optothermal control techniques.

Chapter 1

Introduction

Optical manipulation, which exploits the interaction between light and matter, has become an essential technique for the non-invasive control of micro- and nanoscale objects. This is possible due to the fact that a light beam carries momentum in the form of the Poynting vector, and as a result, it can impart forces on matter [1]. In recent years, this approach has found significant applications in colloidal systems, enabling a wide range of studies related to optical trapping [2, 3], optical binding [4–6], and dynamic soft matter phenomena such as light-induced assembly [7], collective motion [8], and microswimmer navigation in fluid environments [9].

Conventional optical tweezers, which utilize a highly focused laser beam using a high numerical aperture objective lens to create a potential energy minimum through the balance of scattering and gradient forces, which can be modulated by tuning the beam’s intensity, polarization, and spatial phase. This platform has proven effective in trapping dielectric particles and biological cells across nanometre to micrometre scales [9, 10].

Beyond carrying linear momentum via the Poynting vector [1], structured light fields engineered in phase and polarization can also possess angular momentum, which includes both spin angular momentum (SAM) and orbital angular momentum (OAM) [11, 12]. These components significantly influence the dynamics of light-matter interaction [13]. Engineering these parameters enables additional degrees of control over optical manipulation, such as the simultaneous generation of optical forces that can trap and manipulate matter [14, 15]. However, these methods face challenges in manipulating absorbing particles due to their

high scattering and absorption characteristics [16, 17], as well as limitations in scalability for simultaneously handling large numbers of particles. Thus, heating of the system appears as a drawback to the manipulation of colloidal systems. In this context, dynamic optical fields have shown significant promise to manipulate the dynamics of the colloidal systems, but this requires advanced active feedback protocols, which often pose challenges [18–20].

Combining optical forces with other secondary fields, such as thermal fields, emerged as a promising alternative to address these challenges. These emerging strategies expand the landscape of optical manipulation by incorporating thermal degrees of freedom alongside electromagnetic control [21–23]. This technique harnesses spatial temperature gradients to induce forces that guide particle motion. In this context, the thermal field in the system can be efficiently generated by various means, whether it be direct optical field-mediated heating or even surface plasmon-mediated enhanced heating [23–28]. These temperature-mediated techniques significantly enhanced the capability of particle manipulation, from nanoscopic particle confinement [23, 29] to the formation of large-scale assembly [27, 28]. Besides the localized thermal fields generated by absorbing or plasmonic systems can give rise to a variety of fluidic effects, including thermo-osmosis [30–32], thermophoresis [33–36], convection [36], microbubble formation [37–39], Marangoni flows [40, 41] and temperature-dependent depletion forces [42]. Recent advances have demonstrated how these processes can be leveraged to study non-equilibrium thermodynamic behavior and to achieve energy-efficient, low-power trapping and reconfigurable assembly at the microscale and nanoscale [32, 43, 44].

This thesis explores optothermal dynamics arising from light-induced heating of active colloids under varied optical excitation conditions. These insights help bridge the understanding of optical and thermally driven forces, providing a unified framework for developing low-power, scalable platforms for particle manipulation and reconfigurable nanofabrication. The remainder of this chapter outlines key theoretical and experimental concepts, beginning with conventional optical forces and manipulation, followed by a discussion of different optothermal effects in fluidic systems.

1.1 Optical forces and manipulation

One of the most prominent manifestations of light-matter interaction at the microscale is optical trapping or tweezing, a technique first demonstrated by Arthur Ashkin [2, 3, 45]. Since its inception, optical tweezers have become a versatile tool, enabling stable confinement of micro- and nanoscale particles [46, 47], cooling and trapping of neutral atoms [48–50], and probing the dynamics of biological systems [51–53], among many other applications. The principle relies on optical forces arising from electromagnetic momentum transfer to particles, governed by momentum conservation.

In practice, optical trapping is typically achieved using a highly focused laser beam, often via a high numerical aperture objective lens. Under such conditions, considering dipole approximation ($R \ll \lambda$), the total optical force acting on a colloidal particle can be expressed as the time-averaged electromagnetic force [54]:

$$\langle F \rangle = \frac{1}{4} \epsilon_0 \epsilon_r \alpha' \nabla |E|^2 + \frac{1}{2} \epsilon_0 \epsilon_r \alpha'' \text{Im} \left(\sum_i E_i^* \nabla E_i \right). \quad (1.1)$$

Here, α' and α'' denote the real and imaginary parts of the particle's polarizability α , $i = x, y, z$, ϵ_0 is the vacuum permittivity, and ϵ_r is the relative permittivity of the surrounding medium. This expression reveals two main components: the gradient force (first term), which pulls the particle toward regions of higher light intensity, and the scattering force (second term), which pushes the particle along the direction of light propagation.

1.1.1 Confinement using gradient optical force

The behavior of these forces under various optical fields is illustrated in Figure 1.1. In a uniform field (Figure 1.1(a)), the particle experiences a net scattering force with no restoring force due to the absence of an intensity gradient. Under a Gaussian beam (Figure 1.1(b)), lateral intensity gradients give rise to a restoring gradient force that attracts the particle toward the beam center. In a tightly focused Gaussian beam (Figure 1.1(c)), the gradient force dominates, enabling three-dimensional trapping of the particle at the focal point.

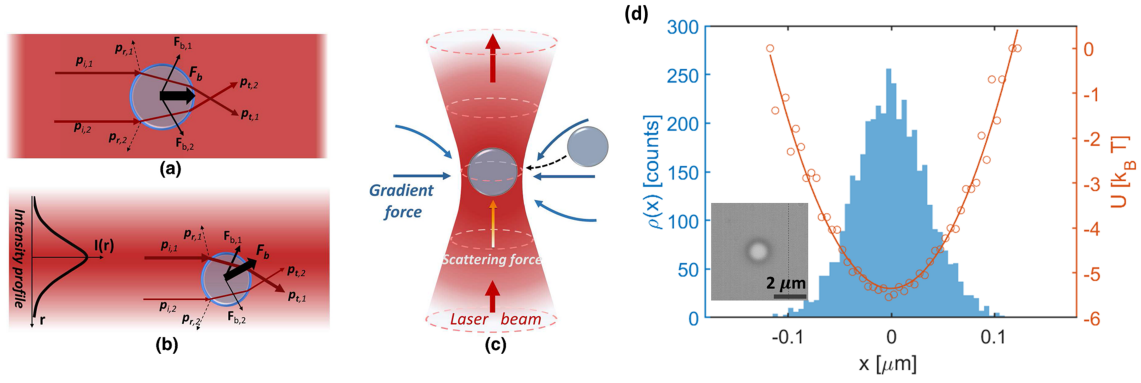


FIGURE 1.1: Illustration of optical forces on a spherical colloid. (a) In a uniform beam, the particle is pushed along the beam's propagation direction due to scattering. (b) In a Gaussian beam, lateral imbalance is created due to the gradient optical force when the colloid is at an offset position from the maximum intensity axis. (c) A focused Gaussian beam allows stable 3D trapping. (d) Experimental probability distribution $\rho(x)$ and potential $U(x)$ for a 1.3 μm polystyrene particle under focused Gaussian illumination. Figures (a)-(c) are adapted from [55] with permission.

To quantify the confinement created by the gradient force, we begin with the intensity profile of a Gaussian beam [56]:

$$I(r, z) = I_0 \left(\frac{w_0}{w(z)} \right)^2 \exp \left(-\frac{2r^2}{w(z)^2} \right), \quad (1.2)$$

where r and z are the radial and axial coordinates from the beam focus, w_0 is the beam waist, and $w(z)$ is the beam width at axial position z . At the focal plane, the intensity becomes:

$$I(r) = I_0 \exp \left(-\frac{2r^2}{w_0^2} \right). \quad (1.3)$$

The corresponding gradient force is:

$$F_{\text{grad}} \propto \nabla I = -\frac{4rC}{w_0^2} I_0 \exp \left(-\frac{2r^2}{w_0^2} \right), \quad (1.4)$$

where C is a proportionality constant. For small displacements ($r \ll w_0$), this approximates to Hooke's law:

$$F_{\text{grad}} \simeq -\kappa r, \quad (1.5)$$

where κ is the trap stiffness, characterizing the strength of the trap. The associated trapping potential is:

$$U(r) \simeq \frac{1}{2}\kappa r^2. \quad (1.6)$$

A colloidal particle undergoing Brownian motion near the trap will experience confinement near the focal point. As shown in the inset of Figure 1.1(d), a 1.3 μm polystyrene particle is optically trapped using a Gaussian beam with waist $w_0 = 1 \mu\text{m}$ and intensity $I_0 = 2.4 \text{ mW}/\mu\text{m}^2$. The particle trajectory is recorded using video microscopy, and the spatial distribution $\rho(x)$ is extracted. Under thermal equilibrium, the corresponding potential is:

$$U(x) = -k_B T \log(\rho(x)), \quad (1.7)$$

where k_B is Boltzmann's constant and T is the ambient temperature. The resulting position distribution fits well with a Gaussian profile, yielding the harmonic potential:

$$U(x) = U_0 + \frac{1}{2}\kappa(x - x_{\text{eq}})^2, \quad (1.8)$$

where x_{eq} is the equilibrium position, and κ is the measured stiffness, obtained experimentally as 3.24 pN/ μm for the particle in Figure 1.1(d). Although here we have discussed the optical confinement relying on the dipole approximation ($R \ll \lambda$), similar results also hold for the geometrical optics regime, i.e., $R \gg \lambda$ [10].

1.1.2 Manipulation using phase engineering

In addition to gradient and scattering optical forces, the in-plane energy flow through the modulation of intensity, phase, and polarization via the introduction of holographic optical tweezers has enhanced the versatility to manipulate colloidal objects [14, 15, 57–59]. Here, we would like to discuss how modulation of the phase leads to controlled manipulation of

colloidal objects. To understand this, let us first consider the vector potential of a light beam with frequency ω and polarization $\hat{\varepsilon}(r)$ given by:

$$A(r, t) = u(r)e^{i\Phi(r)}e^{-i\omega t}\hat{\varepsilon}(r), \quad (1.9)$$

where $u(r)$ and $\Phi(r)$ are the real-valued amplitude and phase. For simplicity, if we consider that light is linearly polarized, $\hat{\varepsilon}(r)$ is real. For a plane wave $\Phi(r) = kz$, where k is the wave vector of the light in the medium. Now, if we impose a transverse phase profile $\phi(r)$ on the wavefront of the light we get,

$$\Phi(r) = k_z(r)z + \phi(r), \quad (1.10)$$

where $\phi(r)$ satisfies $\hat{z} \cdot \nabla\phi = 0$. Now the modified wave vector $k(r) = k_z(r)\hat{z} + \nabla\phi$. Now, as per Abraham's formulation [60], the momentum flux associated with the beam is obtained as,

$$g(r) = \frac{1}{c^2} \text{Re}\{E^* \times H\} = \frac{k}{n_w \mu c} I(r) \nabla\Phi. \quad (1.11)$$

Now, separating the momentum flux in the propagation direction and the transverse direction, which are directly related to the forces, can be written as:

$$g_z = \frac{k k_z I(r)}{n_w \mu c} \hat{z} \quad (1.12a)$$

$$g_\perp = \frac{k}{n_w \mu c} I(r) \nabla\phi. \quad (1.12b)$$

Thus, from equations 1.12(a)-(b), we can see that by modulating the phase of the optical field, we can induce in-plane momentum flux and force on the beam, which can be transferred to the colloidal particles. If the transverse phase is $\phi(r) = qx$, then the phase gradient

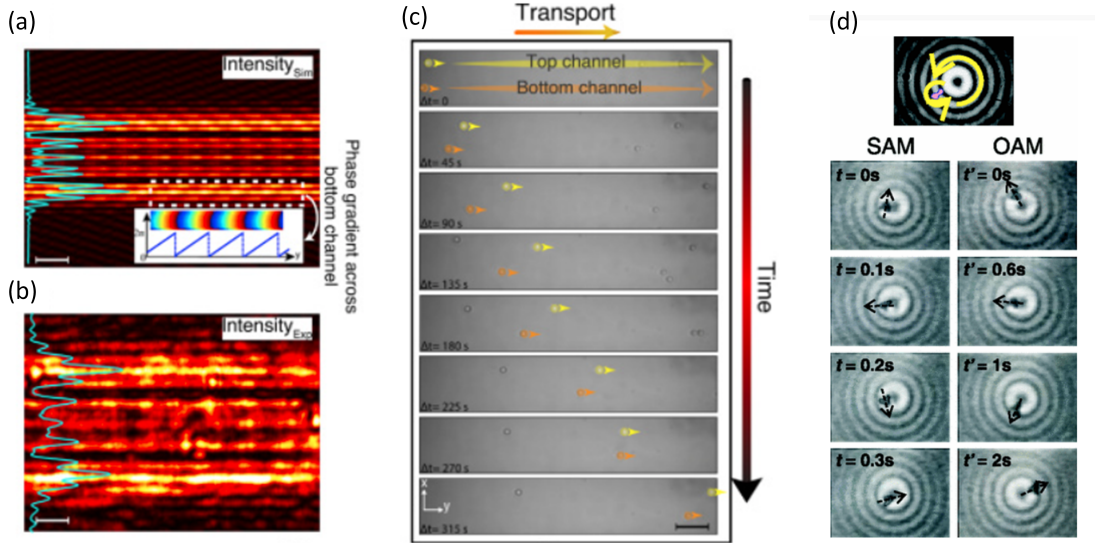


FIGURE 1.2: Optical manipulation using phase gradient. (a) The simulated intensity profile along with the phase profile is shown in the inset. (b) Measured experimental intensity profile corresponding to (a). (c) The phase gradient transverse force manipulates the colloidal particles along x direction. (d) A birefringent particle is trapped in the first ring of a higher-order Bessel beam. The particle exhibits simultaneous rotation around its axis and the beam axis due to the combined effect of the spin angular momentum (SAM) and orbital angular momentum (OAM).

Figures (a)-(c) are taken from [58], and (d) is taken from [15] with permission.

is $\nabla\phi = q\hat{x}$, thus the beam will impart force on colloidal objects so that they get manipulated in the x direction (see Figure 1.2(a)-(c)). In contrast, if the phase gradient is along the azimuthal direction, i.e., $\nabla\phi = l\hat{\theta}$, the beam possesses angular momentum, which can be used to create a rotational state of the colloidal particles (see Figure 1.2(d)).

1.1.3 Feedback controlled manipulation

Optical manipulation can be significantly enhanced using feedback-controlled mechanisms, allowing dynamic and adaptive control of particle motion beyond conventional static trapping. In such systems, the particle's position is continuously monitored, typically through video microscopy, and the optical trap is adjusted in real time based on this feedback signal [18, 19]. This enables high-precision steering, active confinement, and the creation of time-dependent potentials, which are beneficial for adaptive manipulation of absorbing particles that cannot be stably trapped using static optical fields. Feedback-based control also

facilitates the implementation of advanced protocols, such as optical cooling [61], and emulation of non-equilibrium dynamics like active Brownian motion [20]. These techniques have opened new avenues in soft matter research, biophysics, and the development of optically driven micromechanical systems. Although feedback-controlled manipulation has numerous advantages, implementing such a mechanism requires a complicated algorithm and high-end machinery.

1.2 Optothermal fields and manipulation

The forces experienced by colloidal particles in optical fields are not restricted to conventional optical forces such as gradient, scattering, or in-plane energy flow induced forces that arise purely from light-matter interactions. In systems where the colloidal particles possess absorptive characteristics, particularly in the visible or near-infrared spectral range, an additional layer of complexity is introduced. These particles absorb part of the incident laser energy and convert it into thermal energy through non-radiative relaxation processes [25, 26, 37]. This localized heating causes the colloidal particles to reach a higher temperature than the surrounding medium.

As a consequence of this temperature difference, a thermal field is established in the surrounding medium. The spatial profile and magnitude of this thermal field depend on various factors, including the absorption coefficient of the particle, the intensity and focus of the laser beam profile, the thermal conductivity of the mediums, and the geometry of the system [26]. This thermally induced environment can lead to a range of secondary phenomena such as thermophoresis [33–36] (the migration of particles along temperature gradients), thermosmosis (fluid flow near surfaces due to thermal gradients) [30–32], and alterations in the local viscosity and diffusivity of the medium. Such thermally mediated effects, when superimposed with optical forces, can result in complex, non-equilibrium dynamics and emergent behavior in colloidal systems.

To this end, in this section, we focus on the heating mechanisms of colloidal particles

and the resulting thermal field distribution in their surroundings, along with the thermally mediated phenomena that emerge from these processes. We begin by examining the heating behavior of spherical absorbing colloidal particles, as our study primarily involves such geometries. Following this, we explore the general principles governing the thermally induced motion of colloidal particles in the presence of external thermal gradients, particularly highlighting the roles of thermophoresis and thermo-osmosis. Lastly, we consider scenarios where thermal gradients are not externally imposed but instead arise dynamically due to differences in the absorption properties of the colloidal particles themselves. These self-generated thermal fields give rise to complex, interaction-driven motion and non-equilibrium behaviors within the system.

1.2.1 Heating of the colloidal particles

To understand the heating of the colloidal particles, we consider the particle a complex permittivity as $\epsilon_A = \epsilon_A' + i\epsilon_A''$, where ϵ_A' and ϵ_A'' are the real and complex parts of the permittivity. The imaginary part of the permittivity contributes to heating upon optical illumination. The corresponding heat power density (heat power per unit volume) can be defined as [26],

$$q(r) = \frac{\omega}{2} \epsilon_A'' |\vec{E}(r)|^2. \quad (1.13)$$

The differential equation associated with the heat diffusion can be written as:

$$\rho(\vec{r}) c_P(\vec{r}) \partial_t T(\vec{r}, t) - \vec{\nabla} \cdot (\kappa_c(\vec{r}) \vec{\nabla} T(\vec{r}, t)) = q(\vec{r}, t), \quad (1.14)$$

where ρ , κ_c , c_P are the density, thermal conductivity, and the specific heat at constant pressure of the material. The first term of the equation gives transient heating of the colloidal particles. For a homogeneous medium, the above equation simplifies to,

$$\rho c_P \partial_t T - \kappa_c \nabla^2 T = q. \quad (1.15)$$

Typically for microscopic particles, the heat diffusion time scale ranges from the order of $ns - \mu s$. Most of our experimental time scale is of the order of milliseconds or higher, thus, the system reaches steady state within that time scale. In such a case, the above heat diffusion equation takes the time-independent form as:

$$-\kappa_c \nabla^2 T = q. \quad (1.16)$$

This equation is three-dimensional, but if we consider spherical microscopic particles of radius a , the three-dimensional Laplacian operator reduces to only the radial component as,

$$-\kappa_c \frac{1}{r^2} \partial_r (r^2 \partial_r T) = q. \quad (1.17)$$

Outside the absorbing particles, there is no heat source, so $q = 0$, and inside the particle $q \neq 0$. Solving the above equation, we can obtain the thermal field distribution inside and outside the absorbing microparticles as,

$$T(r) = \frac{Q}{4\pi\kappa_s a} \left[1 + \frac{\kappa_s}{2\kappa_p} \left(1 - \frac{r^2}{a^2} \right) \right], \text{ for } (r < a) \quad (1.18a)$$

$$T(r) = \frac{Q}{4\pi\kappa_s r}, \text{ for } (r > a) \quad (1.18b)$$

where κ_s and κ_p are the thermal conductivity of the surrounding material and the absorbing micro-particles, respectively. Q is the total heat power dissipated by the colloidal particle and can be obtained by integrating the heat power density over the volume (V) of the absorbing particles as $Q = \int_V q dV$.

1.2.2 Thermo-osmosis

A temperature gradient across a liquid-solid interface can induce a tangential flow within the adjacent boundary layer, known as thermo-osmotic flow [30–32]. This flow is not solely

the result of heat conduction, it also involves the transport of excess free energy due to temperature-dependent interactions between the liquid and the solid surface. When a small volume of liquid near the colder region of the interface is exchanged with one from the hotter region, both heat and interaction energy are transferred. This imbalance in enthalpy gives rise to fluid motion along the interface. Since these interactions occur within a very thin region, typically only a few nanometers, the resulting hydrodynamics can be effectively modeled using the boundary layer approximation.

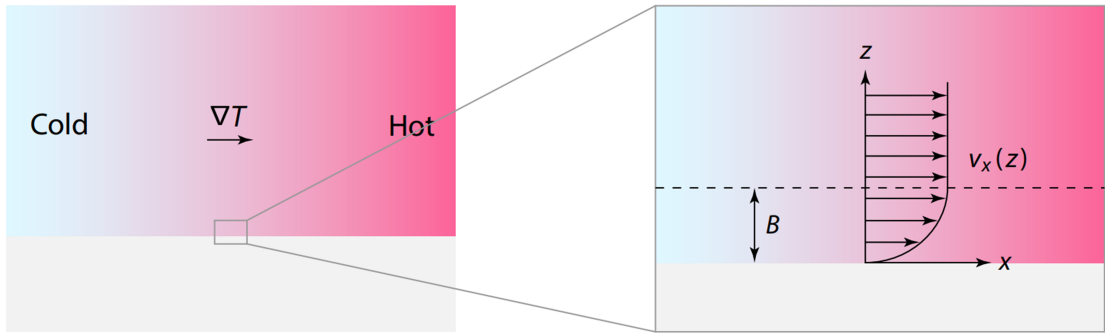


FIGURE 1.3: Illustration of the boundary layer approximation: Owing to liquid-solid interactions, the fluid velocity component parallel to the solid surface (v_x) increases with distance in the z -direction, eventually reaching a constant value beyond the interaction range B . This figure is adapted from [32] with permission.

The microscopic origin of thermo-osmotic slip flow can be understood using low Reynolds number hydrodynamics, appropriate for micro- and nanoscale systems. For an incompressible fluid ($\nabla \cdot \mathbf{v} = 0$), the velocity field is governed by the Stokes equation [32]:

$$\eta \nabla^2 \mathbf{v} - \nabla P + \mathbf{f} = 0, \quad (1.19)$$

where P is the pressure, η is the dynamic viscosity, and $\mathbf{f}$ is the force density acting on the fluid. Figure 1.3 shows the solid-liquid interface and defines the local coordinate system.

In the vicinity of the solid boundary, the vertical component of velocity vanishes ($v_z = 0$). Furthermore, variations in hydrodynamic quantities are more rapid in the vertical (z) direction than in the horizontal (x) direction. The slow variation of v_x in the horizontal

direction allows the approximation $v_x(x, z) \approx v_x(z)$. Under these assumptions, the Stokes equations simplify to:

$$0 = \frac{dP}{dz} - f_z, \quad (1.20a)$$

$$\eta \frac{d^2 v_x}{dz^2} = \frac{dP}{dx} - f_x. \quad (1.20b)$$

Solving the equations with the no-slip boundary condition $v_x(0) = 0$ yields the velocity profile parallel to the interface:

$$v_{\parallel} = -\frac{1}{\eta} \int_0^{\infty} z \left(\frac{dP}{dx} - f_x \right) dz. \quad (1.21)$$

To relate this to interfacial thermodynamics, the expression can be rewritten in terms of the local excess enthalpy $h(z)$, which characterizes the interaction energy between the liquid and solid:

$$v_{\parallel} = -\frac{1}{\eta} \int_0^{\infty} z h(z) dz \cdot \frac{\nabla_{\parallel} T}{T}, \quad (1.22)$$

where $\nabla_{\parallel} T$ is the temperature gradient along the interface and T is the absolute temperature. This can be further simplified using the thermo-osmotic slip coefficient χ :

$$v_{\parallel} = \chi \cdot \frac{\nabla_{\parallel} T}{T}. \quad (1.23)$$

The sign and magnitude of χ determine the direction and strength of the thermo-osmotic flow: if $\chi > 0$, the liquid is driven toward regions of higher temperature; if $\chi < 0$, it flows toward cooler areas. Thus, χ encapsulates the interfacial physics that governs this slip phenomenon. It considers all the interfacial interactions, such as electric double layer interaction, Van der Waals interactions, electrolyte concentration, and even the depletion interaction. For a pure milli-q water, only the electrostatic double layer interaction becomes

non-zero, resulting in positive χ , and the slip flow flows from the cold region to the hot area. This thermo-osmotic slip flow can manipulate the dynamics of colloidal particles close to the interface and drag them to the higher temperature regions.

1.2.3 Thermophoresis

Colloidal thermophoresis serves as a prototypical non-equilibrium phenomenon [33–36]. When a uniform temperature gradient ΔT is imposed across a colloidal suspension, particles ranging from the microscale to the nanoscale experience a directed drift in addition to their random Brownian motion. Depending on specific system parameters, this drift may be oriented toward either the hotter or colder region. Although this behavior mimics the response to an external force, such as gravity or an electric field, no actual macroscopic force acts on the particles. The thermophoretic velocity $\vec{u}$ is commonly described by:

$$\vec{u} = -D_T \frac{\vec{\nabla} T}{T}, \quad (1.24)$$

where D_T is the thermo-diffusion coefficient or thermophoretic mobility. A negative D_T corresponds to migration toward the high-temperature region (negative thermophoresis), while a positive D_T indicates motion toward the cold region (positive thermophoresis). Often, the thermophoretic behavior of the colloidal particles is referred to as the Soret coefficient (S_T) of the colloids rather than the thermo-diffusion coefficient (D_T). Soret coefficient is basically the ratio of the thermo-diffusion coefficient to the diffusion constant (D) of the colloid as: $S_T = D_T/D$.

Understanding the physical origin of thermophoresis and its pronounced sensitivity to interfacial characteristics is a challenge. But a widely accepted explanation connects thermophoresis to thermo-osmotic slip flow that arises within the electric double layer surrounding charged colloidal particles. According to Eq. 1.23, only the component of the thermo-osmotic flow parallel to the interface contributes to particle motion. Due to the differing

thermal properties of the particle and the surrounding fluid, the temperature gradient is distorted near the particle surface. The component of the temperature gradient parallel to the surface at any point is given by [32]:

$$\frac{dT}{dx} = \frac{3\kappa_1}{2\kappa_1 + \kappa_2} |\Delta T| \sin \theta, \quad (1.25)$$

where κ_1 and κ_2 denote the thermal conductivities of the solvent and the particle, respectively. For dielectric particles, where $\kappa_2 \approx \kappa_1$, this simplifies to $\frac{dT}{dx} = |\nabla T| \sin \theta$. Consequently, the parallel component of the velocity becomes:

$$v_{||}(\theta) = \chi \frac{|\nabla T|}{T} \sin \theta = v_{||} \sin \theta, \quad (1.26)$$

where χ is the thermo-osmotic slip coefficient. The net thermophoretic velocity $\vec{u}$ is obtained by taking the orientational average of $v_{||}(\theta)$:

$$\vec{u} = -\langle v_{||}(\theta) \hat{x} \rangle. \quad (1.27)$$

In spherical coordinates, this expression simplifies to:

$$\vec{u} = -\frac{2}{3} v_{||} \hat{r} = -\frac{2}{3} \chi \frac{\nabla T}{T}. \quad (1.28)$$

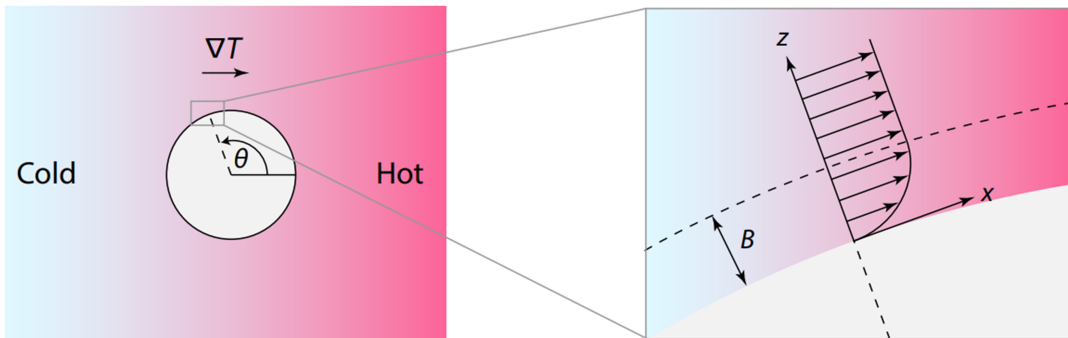


FIGURE 1.4: Schematic representation of the thermo-osmotic flow field around a spherical particle. This figure is adapted from [32] with permission.

Comparing this with Eq. 1.24, we obtain the relationship:

$$D_T = \frac{2\chi}{3T}. \quad (1.29)$$

From equation 1.25, for metallic particles, where $\kappa_2 \gg \kappa_1$, the local temperature gradient near the particle surface is significantly suppressed due to high thermal conductivity. This leads to negligible thermophoretic motion. Figure 1.5 shows the temperature distortion around polystyrene (PS) and gold particles subjected to an external gradient of $1 \text{ K} \cdot \mu\text{m}^{-1}$. As evident in panels (a) and (b), the gold particle almost completely quenches the surface temperature gradient, explaining the absence of observable thermophoretic motion.

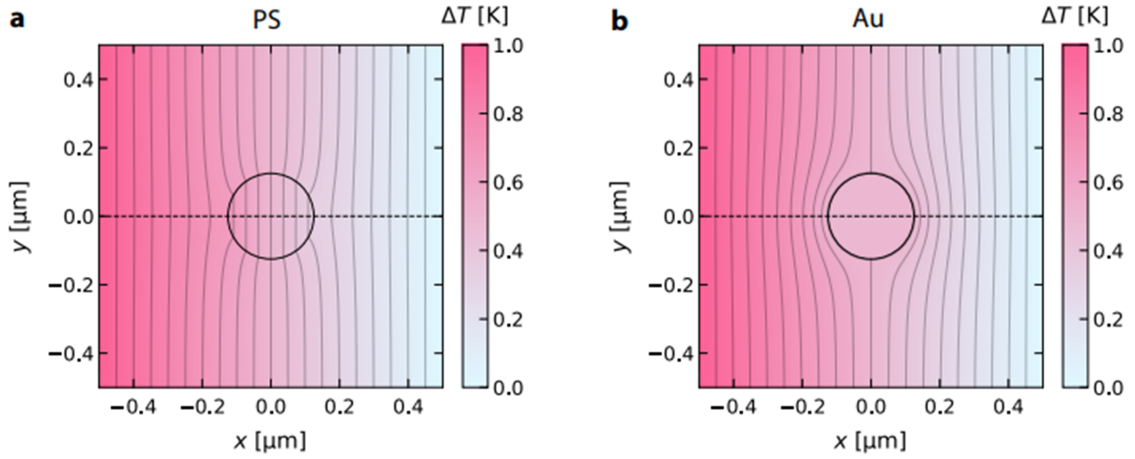


FIGURE 1.5: Temperature distortion near (a) a polystyrene and (b) a gold particle in an external thermal gradient of $1 \text{ K} \cdot \mu\text{m}^{-1}$. The vertical lines indicate the isotherms. This figure is adapted from [32] with permission.

While this formalism effectively relates the thermophoretic velocity of microparticles to thermo-osmotic slip, it does not fully account for all experimental observations. For instance, from equation 1.28 and Fig. 1.4, it can be seen that the motion of colloidal particles indicates toward colder regions in pure Milli-Q water, reflecting positive thermophoresis [62]. However, instances of negative thermophoresis are also reported under similar conditions, which cannot be explained using this formalism. This discrepancy highlights that thermophoretic behavior is not determined solely by bulk properties such as particle size,

density, conductivity, or surface charge. Instead, it depends strongly on the microscopic nature of the particle-solvent interface, where molecular-level interactions dominate. Although a comprehensive microscopic theory remains elusive, Eq. 1.24 serves as a reliable empirical description across various systems.

In this chapter, we have primarily focused on colloidal manipulation driven either by external optical fields or static temperature gradients. Additionally, we discussed how optically absorbing colloidal particles can locally heat the surrounding fluid, effectively acting as dynamic heat sources. These thermally induced fields can influence the behavior of nearby particles. By carefully designing the optical beam to couple optical and thermal effects, complex dynamics can emerge. For example, Fig. 1.6 shows particle propulsion within a ring-shaped beam, where motion arises without direct optical forces transfer. The inset displays the experimental intensity profile of the ring beam. The ring beam illustrated in Fig. 1.6 is generated using a spatial light modulator (SLM) where a blazed axicon pattern is projected. The detailed discussion of structured optical field generation is enlightened in chapter 2 under the subsection heading 2.1.1 Generation of structured optical field. We have used colloidal particles with different optical absorption properties, such as absorbing colloid (A) and passive colloid (P) of respective diameters $1.3\ \mu\text{m}$ and $2\ \mu\text{m}$. When colloidal particles with different absorbing properties are subject to such optical illumination, they form colloidal structures exhibiting active propulsion.

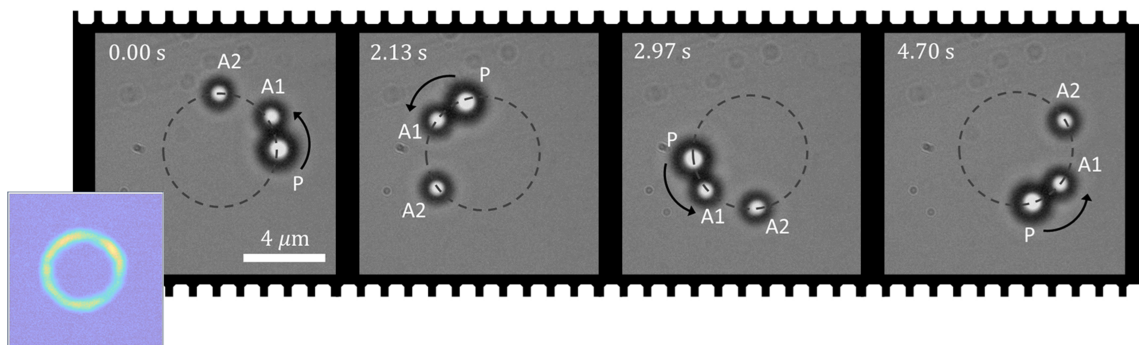


FIGURE 1.6: Optothermally induced propulsion in a ring-shaped optical illumination with the beam intensity profile in the inset.

1.3 Outline of the thesis

In this thesis, we investigate the dynamics of colloidal particles subjected to various forms of optical fields, particularly emphasizing how light-induced thermal effects govern their dynamics. When exposed to optical illumination, certain colloidal particles, particularly those containing light-absorbing materials, undergo selective heating due to their intrinsic optical absorption properties. This localized heating gives rise to thermal gradients in the surrounding fluid medium, which drive optothermal interactions such as thermophoresis [33–36] and thermo-osmosis [30–32]. These optothermal forces act as environmental cues that guide the motion and interaction of the colloidal particles. These interactions enable long-range, non-reciprocal forces between active and passive colloids, facilitating coordinated motion. As a consequence of these interactions, the system exhibits emergent behavior such as dynamic structure formation, including dimers, trimers, and other complex assemblies. These structures are stable under continuous optical excitation and exhibit controllable dynamics, such as directed propulsion or rotational motion, depending on the configuration of particles and the nature of the optical field.

Chapter 2: Methodology: experimental and numerical techniques

In this chapter, we detail the methodologies employed in setting up the experimental system, acquiring and analyzing data, and performing numerical simulations to support our experimental observations. We start by focusing on the generation of structured optical fields, specifically, ring-shaped illumination, using a phase-only spatial light modulator. This type of optical field, characterized by the absence of transverse momentum flux, enables in-plane manipulation of colloidal particles without exerting direct mechanical torque. Next, the design and components of the dual-channel optical microscope used in our experiments are discussed. We also outline the simulation framework used to model particle dynamics, which involves numerically solving the coupled Langevin equations under the influence of external forces. Additionally, we describe the finite element simulations conducted to compute the optical field distribution and associated flow profiles, which serve to validate and

complement our experimental findings.

Chapter 3: Optothermal interaction mediated dynamics of binary colloidal particles in unbound optical illumination

This chapter introduces the emergence of active motion of microscopic structures facilitated by the optothermal interactions between the colloidal particles without any direct transfer of momentum from the external field or use of the chemical environment. Under broad optical illumination, absorbing colloids generate local temperature gradients, inducing non-reciprocal thermo-osmotic forces that drive the motion of surrounding passive particles. This leads to the formation of colloidal structures composed of multiple colloidal particles. Because of the optothermal force imbalance in this colloidal structure, they exhibit resulting dynamics that include both propulsion and chiral rotation. The dynamics of the structures are corroborated by using numerical simulations based on coupled Langevin equations.

Chapter 4: Dynamics of thermally active and passive colloids under focused optical illumination

In this chapter, we experimentally demonstrate the emergence of directional rotation in colloidal assemblies composed of absorbing thermally active and non-absorbing passive particles under focused optical confinement. The rotation originates from thermal gradients generated by the thermally active colloid, resulting in unbalanced optothermal torques, without requiring spin or orbital angular momentum of light. This rotational state, such as direction and speed, can be tuned by altering the particles' spatial configuration and the laser beam's intensity, respectively.

Chapter 5: Guided motion of colloidal structures in optical ring beam

This chapter experimentally investigates controlled, guided motion of dimer and trimer colloidal structures composed of thermally active and passive particles, confined within a ring-shaped optical beam. Optothermal interactions, arising from light absorption, drive the motion, rather than direct optical forces. Attractive thermo-osmotic forces induce propulsion by generating imbalances, while repulsive positive thermophoretic interactions maintain structural parameters. The optical gradient force provides confinement, guiding motion along a particular optically laid out path. By tuning the optical intensity, the dynamic behavior of these assemblies can be precisely controlled. We have also employed the Brownian dynamics simulation by solving coupled Langevin equations to support our experimental findings.

Chapter 6: Overall summary and future directions

This thesis concludes with the present chapter, which provides a comprehensive summary of the experimental results and the central findings reported throughout this thesis. We revisit the key outcomes of our investigations on optical manipulation, optothermal interactions, and the dynamics of active and passive colloidal assemblies under structured light fields. Additionally, we critically assess the limitations of our methodologies and the constraints encountered in the experimental design and theoretical modeling.

Beyond summarizing the accomplishments, this chapter also aims to offer a broader perspective by outlining potential directions for future research. We highlight specific aspects that merit deeper investigation, such as extending the optical control mechanisms to more complex colloidal geometries, enhancing spatiotemporal control through adaptive self-feedback of the structure. These proposed directions are intended to inspire further experimental and theoretical efforts in active soft matter and non-equilibrium colloidal systems.

Chapter 2

Methodology: Experimental and Numerical Techniques

The optical forces explored in the previous chapter are not only of fundamental significance but also hold immense potential for applications across a broad spectrum of biological and technological fields [2, 9]. These forces play a critical role in the investigation of dynamic processes such as cellular interactions [63], protein dynamics [64, 65], and Brownian motion. Moreover, they are increasingly being leveraged in the development and control of artificial micromotors and microscale robotic systems [66, 67].

Beyond their standalone capabilities, optical forces can be synergistically combined with optothermal effects such as thermo-osmosis [30–32] and thermophoresis [33–36] to probe and manipulate active matter systems that are inherently out of equilibrium [68, 69]. This integration provides a powerful toolkit for studying environmental cues that influence particle behavior, enabling the exploration of complex collective dynamics and mutual interactions among colloidal particles [70]. Such interdisciplinary approaches pave the way for novel methods to control microscale systems and understand the principles underlying active soft matter.

This chapter will discuss the experimental and numerical schemes used to study colloidal dynamics owing to light-matter interaction. In the first part, we will focus on the experimental configuration employed for various studies, such as:

- Generation of structured optical field

- Microscope architecture.

The second part will discuss the different numerical schemes used to support our experimental studies and gain insight into the mechanism. This section is further subdivided into:

- Dynamics simulation by solving the Langevin equation
- Finite element method simulation for optical and flow field.

2.1 Experimental techniques

2.1.1 Generation of structured optical field

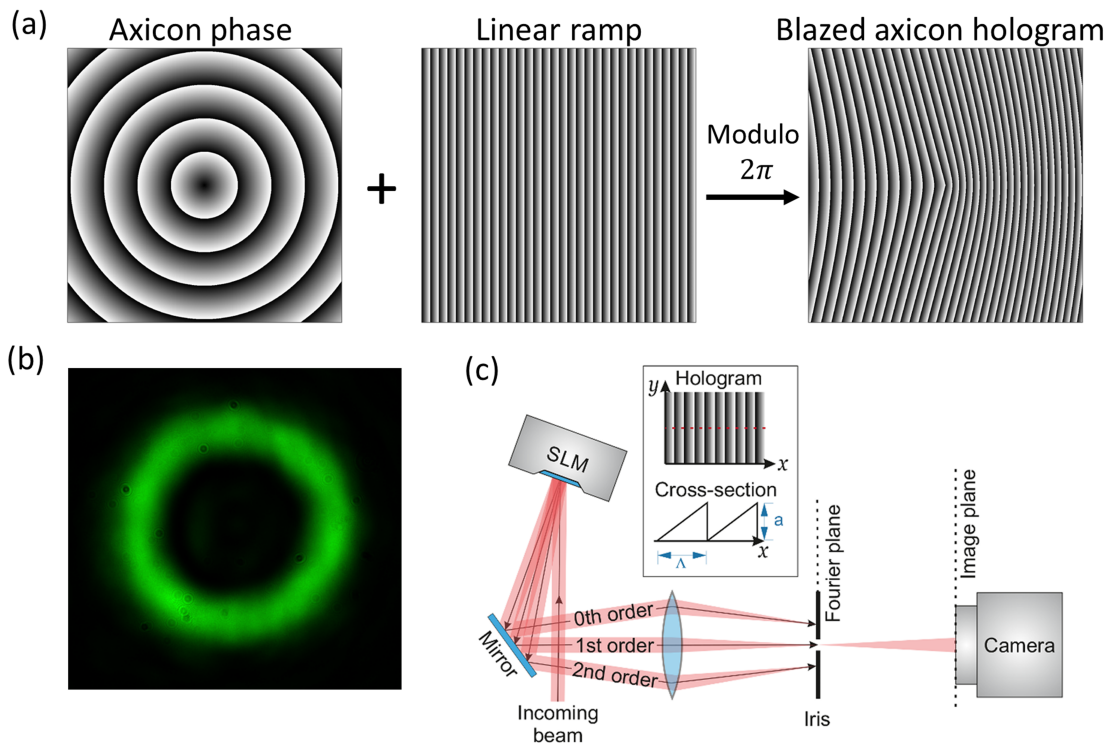


FIGURE 2.1: Schematic generation of a ring-shaped optical structured field using an SLM. (a) For that, a blaze axicon hologram is used, which can be generated by combining an axicon pattern with a Gaussian linear blazing and expressing the result in the form of modulo 2π . (b) The resulting intensity distribution represents a ring optical beam. The phase modulation protocol is implemented using SLM. Figure (c) is adapted with permission from [71].

While most laser sources naturally emit beams with a Gaussian intensity profile, many experimental applications require optical fields with specific spatial intensity and phase distributions. Shaping such fields is essential for applications in optical trapping. This section outlines the methodology for generating custom optical fields beyond the native Gaussian profile [71, 72].

Traditionally, tailored optical fields can be produced using custom-fabricated static optical elements such as axicon lenses [73]. These elements impart a desired phase modulation on the incoming light beam through their carefully engineered geometry. However, one major limitation of these devices is their wavelength specificity. They are typically optimized for a single operational wavelength and offer limited flexibility once fabricated.

To overcome these limitations, programmable devices like Spatial Light Modulators (SLMs) offer a dynamic and versatile alternative. SLMs, which are typically based on liquid crystal technology, can impose a spatially varying phase shift onto an incident beam [71, 72]. This is achieved by electronically controlling the local refractive index at each pixel, thereby enabling the generation of user-defined phase profiles. The working of the SLM spans a broad spectral range. For example, the SLM that we have used is a reflective LCOS SLM optimized for the 500-670 nm wavelength range. We can work with any wavelength within this range using the same SLM. Such broad wavelength compatibility and reconfigurability make SLMs highly suitable for a wide range of beam-shaping applications.

To generate a desired optical field using an SLM, a computer-generated phase hologram is encoded onto the SLM screen. This hologram is typically constructed by combining the phase profile of the desired beam (e.g., a Bessel or ring beam) with a linear phase ramp known as a blazed grating [72]. This blazed component diffracts the modulated beam into a well-defined order, facilitating spatial separation from undesired diffraction orders. The resulting composite hologram is then expressed in modulo 2π to ensure that the phase values remain within the physical range the SLM can display. Fig. 2.1(a) illustrates the schematic protocol used to generate the blazed phase hologram. When illuminated with a collimated Gaussian beam, the modulated output from the SLM gives rise to the desired optical field.

Figure 2.1(b) shows a representative ring-shaped intensity distribution obtained using this method. Fig. 2.1(c) indicates a typical operation protocol of SLM.

In our experimental setup, we employed a phase-only reflective SLM (Model: PLUTO-2.1-VIS-130, procured from Holoeye). The device features a resolution of 1920×1024 pixels and a pixel pitch of $8 \mu\text{m}$. Each pixel is calibrated by the manufacturer to respond linearly to the applied control voltage, which is encoded as an 8-bit grayscale value representing the desired phase level in the hologram. After reflection from the SLM, the diffracted beam contains multiple orders. A spatial filter (iris) is placed at the Fourier plane to isolate the desired first-order beam. This filtered beam is then directed toward the sample using a relay optical system comprising mirrors and lenses, ultimately focused through a microscope objective.

2.1.2 Microscope architecture

The microscope used for the experimental investigation performed in this thesis on colloidal dynamics under optical and optothermal forces is indicated in Fig. 2.2. For our studies, we have used a home-built dual-channel microscopy system. The microscope comprises two objective lenses, having respective numerical apertures NA1 and NA2. All the experiments are performed in the transmission configuration, where, using the upper objective lens, the optical field is projected to the sample plane, which is placed on a piezo stage. The lower objective lens is used for imaging purposes. For better resolution imaging in most of our experiments, the numerical aperture of the lower objective lens is higher than the upper objective lens ($\text{NA2} > \text{NA1}$). In our studies, we have used a laser diode source of wavelength $\lambda = 532 \text{ nm}$. This laser beam is directly used for all experiments with a Gaussian beam. In experiments with the structured optical fields, the laser is at first diverted to a phase-only spatial light modulator, then routed to the upper channel of the microscope. For such diversion, different mirrors (M) have been used. The SLM introduces the desired phase to the optical field to obtain a relevant structured field. To obtain continuous variation of optical intensity, a combination of a half-wave plate (HWP1) and a polarizing beam splitter (PBS)

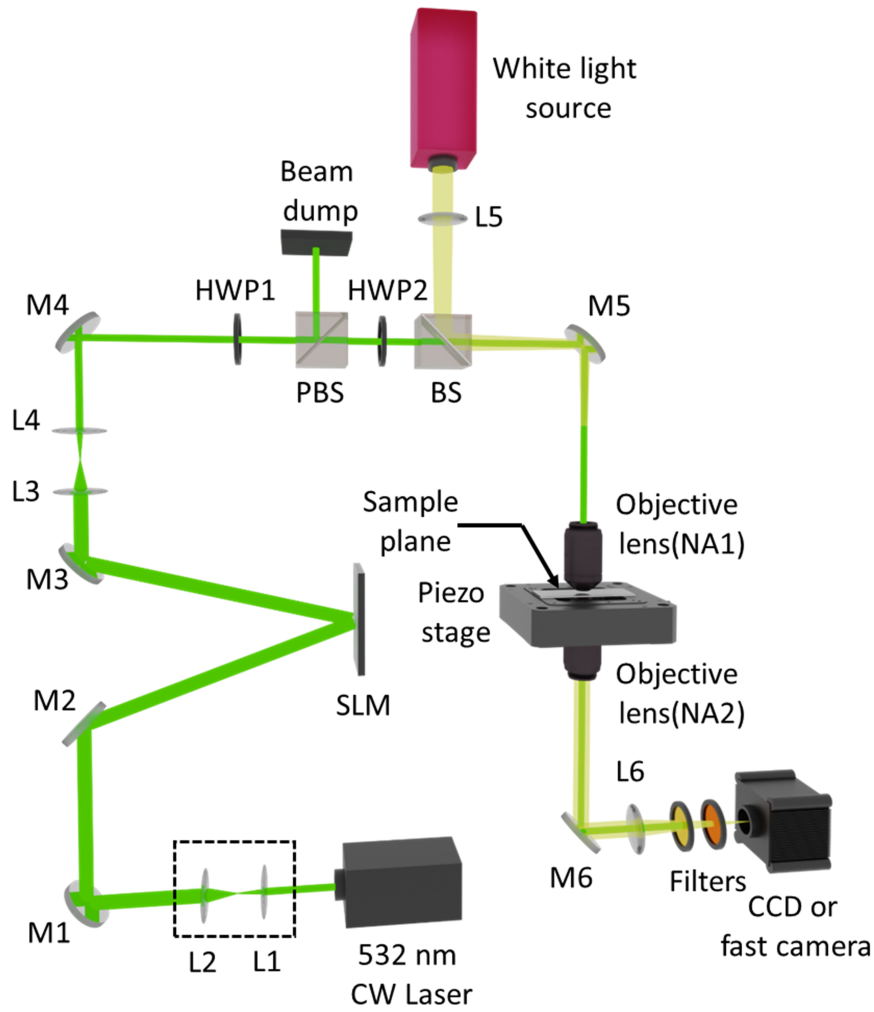


FIGURE 2.2: The schematic of the microscope design. A dual-channel optical microscopy system is used for the experiments. A laser beam from a 532 nm continuous wave (CW) laser diode is projected to the sample plane using the upper channel using an objective lens of numerical aperture NA1. A lower objective lens with a higher numerical aperture ($NA2 > NA1$) is used for high-resolution imaging. The structuring of the laser is done through a phase-only SLM. The continuous variation of the intensity of the excitation laser beam is done by using a combination of a half-wave plate (HWP1) and a polarizing beam splitter (PBS). The light collected from the sample plane is projected to the camera. Various mirrors (M1-M6) and lenses (L1-L6) have been utilized to route the optical beam path.

is used. We can get an arbitrary linear polarized state from this fixed linear polarized beam coming from PBS by placing another half-waveplate (HWP2) after PBS. To rotate the linear polarized state by θ angle, we have to rotate the HWP2 by an angle $\theta/2$ with respect to the input polarization. For the white light illumination, a 90:10 beam splitter is used to couple the white light to the microscope. The signal obtained from the lower objective lens is projected

to a charge-coupled device (CCD) or fast camera using a combination of lenses (L) and mirrors (M). Before projecting the signal to the camera, the intense laser beam is blocked using a combination of notch and edge filters. In this experimental configuration, we can change the focal plane of the top objective (trapping configuration), without disturbing the imaging configuration (focal plane of the bottom objective). Thus, our dual-channel optical setup configuration provides independent control over experimental operational parameters from the imaging conditions, which is advantageous over single-lens attached optical microscopes, where changing the trapping plane leads to a change in imaging conditions.

2.2 Numerical calculations

2.2.1 Finite element simulation of optical field distribution and flow field

Optical Field Simulation: We performed full-field simulations based on the finite element method (FEM) to explore the spatial distribution of optical and optothermal forces arising from light-matter interactions. The FEM approach divides the simulation domain into a mesh of finite elements, allowing partial differential equations (PDEs) to be solved locally within each element, subject to appropriate boundary conditions.

For this purpose, we used COMSOL Multiphysics 5.5, a commercial FEM-based simulation software. COMSOL's modular framework enabled us to model the optical field and the resulting thermally induced fluid forces within a graphical simulation environment.

To obtain the spatial distribution of the optical field, we solved Maxwell's equations under steady-state conditions using COMSOL's Wave Optics module. The simulation geometry, as illustrated in Fig. 2.3(a), includes a lower glass substrate region and an upper aqueous medium in which colloidal particles are positioned. The colloidal particles denoted by A and P indicate optically absorbing and non-absorbing particles with diameters 1.3 and 2 μm , respectively. A linearly polarized Bessel beam with a wavelength of 532 nm is introduced into the system to generate a ring-shaped illumination pattern at the sample plane.

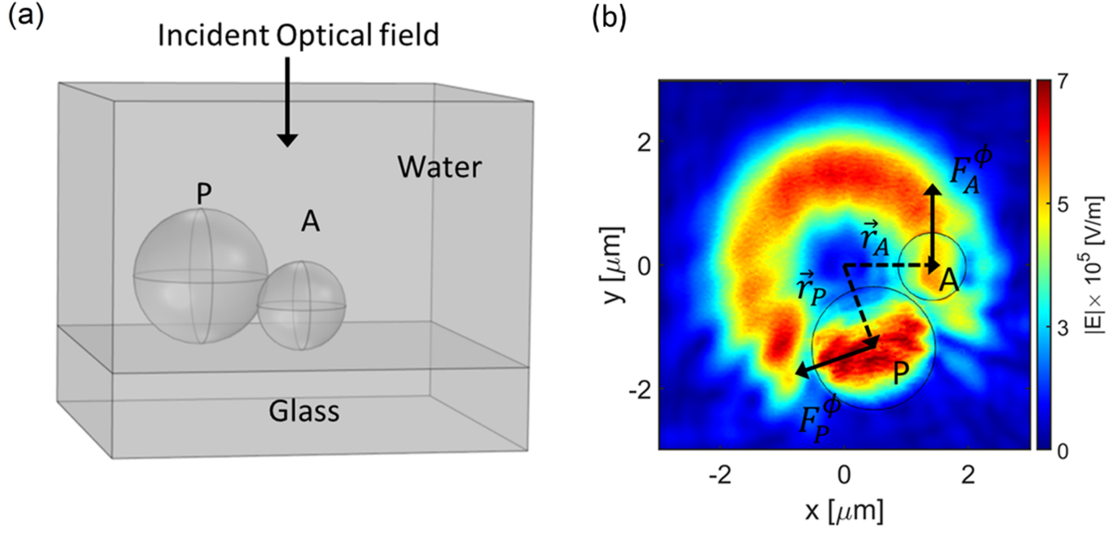


FIGURE 2.3: (a) Simulation geometry showing the layered domains and particle placement; (b) Optical intensity distribution surrounding the colloidal particles illuminated by a ring-shaped beam. A and P denote the absorbing and non-absorbing colloidal particles with diameters 1.3 and 2 μm respectively. $\vec{r}_A$ and $\vec{r}_P$ are their respective position vectors. F_A^ϕ and F_P^ϕ indicate the transverse component of optical force acting on them. The input power of the beam is 5 mW.

This figure is adapted with permission from [74].

The beam is modelled as:

$$E(\rho, z) = C J_0(k_\rho \rho) e^{-\rho^2/w^2(z)} e^{i(k_z \rho^2/2R(z) - \eta(z) + k_z z)},$$

where $J_0(k_\rho \rho)$, $w(z) = w_0 \sqrt{1 + (z/z_0)^2}$, $R(z) = z \sqrt{1 + (z_0/z)^2}$, $\eta(z) = \tan^{-1}(z/z_0)$, k_z and k_ρ are the Bessel function of the first kind, beam waist, radius of curvature, Gouy phase, wave vector components along propagation and transverse directions. Please note $|\vec{k}| = \sqrt{k_\rho^2 + k_z^2} = 2\pi/\lambda$ is the wavenumber. For the ring beam simulation, we used $k_\rho = |\vec{k}|/1.5$. Besides the refractive index of glass, water, passive particle P, and absorbing particle A are considered as 1.45, 1.33, 1.60, and $1.677 + 1.0169i$, respectively.

The resulting optical intensity distribution in the vicinity of the colloidal particles is shown in Fig. 2.3(b). The optical forces acting on the particles are computed by integrating Maxwell's stress tensor over the particle surface. Given that the incident Bessel beam carries no orbital angular momentum, the resulting optical forces are governed solely by the intensity distribution and distortion due to the presence of the particles. This interaction

creates optical forces that contribute to particle manipulation and confinement within the ring-shaped illumination. These optical field simulations provide insight into the intensity-dependent optical forces experienced by the particles. Besides optical forces, optothermal flow simulations can also be done using COMSOL, which is discussed in later sections.

Flow field simulation: Complement to the analysis of optical forces, we performed finite element simulations to account for optothermal forces arising due to heat-induced flow fields around colloidal particles. A similar simulation geometry as shown in Fig. 2.3(a) is considered. In this case, we considered only an absorbing colloid embedded in a fluid environment, which is also acting as a heat source. The surface temperature increase of the colloid is considered as $\Delta T = 2$ K. The corresponding temperature distribution in the system is computed using the Heat Transfer in Solids and Fluids module in COMSOL Multiphysics, which solves the heat diffusion equation:

$$\rho c_P \frac{\partial T}{\partial t} - \kappa_c \nabla^2 T = q, \quad (2.1)$$

where ρ is the density, c_P is the specific heat at constant pressure, κ_c is the thermal conductivity of the material, and q represents the heat power density of the heat source. In this model, the absorbing colloid acts as a localized heat source, where the surface temperature is prescribed as an input parameter. This localized heating leads to a spatial temperature gradient in the surrounding medium, as depicted in Fig. 2.4(a).

The resulting thermal gradients give rise to various flow fields, notably thermo-osmotic and convective flows. To simulate these effects, we couple the heat transfer module with the Laminar Flow module, which solves the Navier-Stokes equations based on the mass and momentum conservation:

$$\nabla(\rho \vec{v}) = 0 \quad (2.2a)$$

$$\rho \frac{\partial \vec{v}}{\partial t} + \rho(\vec{v} \cdot \nabla) \vec{v} = -\nabla P I + \eta \nabla \cdot (\nabla \vec{v} + \nabla \vec{v}^T) + \vec{F} \quad (2.2b)$$

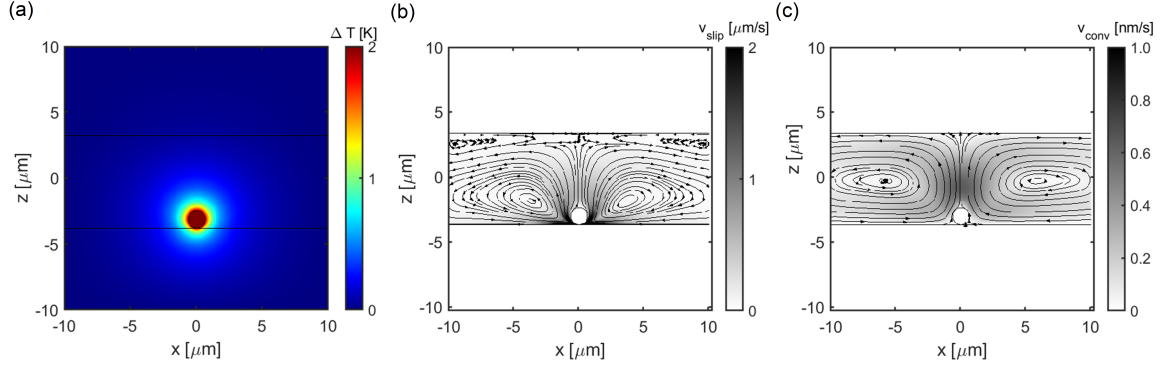


FIGURE 2.4: Temperature and flow field simulation in Comsol. An absorbing colloid is considered a heat source. The surface temperature is considered to $\Delta T = 2$ K higher than the ambient temperature. (a) The temperature distribution in the surrounding medium. The corresponding (b) slip and (c) convective flow field.

where ρ is density of fluid, $\vec{v}(\vec{r}, t)$ is the fluid velocity field profile, η is the dynamic viscosity, $\vec{v}^T$ is transpose of velocity field, I is unity tensor, ∇P is pressure gradient, and $\vec{F}$ is body force acting on fluid which accounts all the external force such as gravity, temperature induce force depending on the local temperature distribution.

To simulate thermo-osmotic flows, we apply a slip boundary condition at all solid-liquid interfaces using a dimensionless slip coefficient that characterizes the strength of thermo-osmotic coupling. The resulting flow profile is shown in Fig. 2.4(b), with characteristic velocities on the order of micrometers per second. In contrast, to isolate pure convective effects, no-slip boundary conditions are applied at all solid interfaces. Additionally, the density of the water medium is treated as temperature-dependent, i.e., $\rho_w = \rho_w(T)$. The corresponding convective flow field is shown in Fig. 2.4(c), with velocities typically in the nanometers per second range.

When a colloidal particle is immersed in these thermally induced flows, it experiences a hydrodynamic drag force. This force is calculated by integrating the viscous stress tensor over the particle's surface:

$$\vec{F}_{\text{drag}} = \int_S \boldsymbol{\sigma} \cdot \hat{n} dS, \quad (2.3)$$

where $\boldsymbol{\sigma}$ is the viscous stress tensor and $\hat{n}$ is the outward normal vector on the colloid's

surface S . This simulation framework enables quantitative estimation of optothermal flow effects, which is essential for understanding colloidal particles' observed motion and interactions under structured optical heating.

2.2.2 Dynamics simulation by solving Langevin equation:

To support and validate our experimental observations on the dynamics of colloidal particles, we performed numerical simulations based on stochastic differential equations. Specifically, we modeled the Brownian motion of colloidal particles using the Langevin equation, incorporating both deterministic forces and thermal fluctuations.

We consider a colloidal particle of radius a and mass m , whose instantaneous position at time t is denoted by $\vec{r}(t)$. The motion of this particle under an external force field $\vec{F}(t)$ is governed by the Langevin equation:

$$m\ddot{\vec{r}}(t) = -\gamma\dot{\vec{r}}(t) + \vec{F}(t) + \sqrt{2k_B T \gamma} \vec{\zeta}(t), \quad (2.4)$$

where γ is the viscous drag coefficient, T is the system temperature, k_B is the Boltzmann constant, and $\vec{\zeta}(t)$ is a delta-correlated Gaussian white noise term representing random thermal forces. The term on the left-hand side represents the inertial response, while the terms on the right-hand side correspond to viscous damping, deterministic external forces (including optical, thermophoretic, and inter-particle interactions), and thermal noise, respectively.

In typical colloidal systems, the inertial timescale $\tau_i = m/\gamma$ is very short (on the order of microseconds), whereas the timescales of interest in our experiments are in the millisecond regime. Thus, the inertia becomes negligible, and the Langevin equation simplifies to its overdamped form:

$$\dot{\vec{r}}(t) = \frac{1}{\gamma} \left(\vec{F}(t) + \sqrt{2k_B T \gamma} \vec{\zeta}(t) \right). \quad (2.5)$$

To solve this stochastic differential equation numerically, we use a finite-difference time-stepping method. The time domain is discretized into intervals of size Δt , and the relevant

quantities are updated as follows:

$$\dot{\vec{r}}(t) \rightarrow \frac{\vec{r}_{k+1} - \vec{r}_k}{\Delta t}, \quad \vec{\zeta}(t) \rightarrow \frac{\vec{\zeta}_k}{\sqrt{\Delta t}}, \quad \vec{F}(t) \rightarrow \vec{F}_k, \quad (2.6)$$

where k denotes the discrete time-step index. This yields the following discretized Langevin equation:

$$\vec{r}_{k+1} = \vec{r}_k + \frac{\Delta t}{\gamma} \left(\vec{F}_k + \sqrt{\frac{2k_B T \gamma}{\Delta t}} \vec{\zeta}_k \right). \quad (2.7)$$

Here, $\vec{\zeta}_k$ is a vector of normally distributed random variables with zero mean and unit variance. By iteratively solving this equation, we can simulate the trajectory of colloidal particles subjected to optothermal and other interaction-driven forces, allowing for direct comparison with experimental results. This framework allows us to investigate the interplay between optical confinement, thermal gradients, and colloidal interactions in a controlled and systematic manner.

Chapter 3

Optothermal interaction mediated dynamics of binary colloidal particles in unbound optical illumination

Abstract:

Engineered soft matter systems have become vital in enabling controlled motion at microscopic scales. Typically, such motion is induced either by external driving fields or through interactions among colloidal particles. When relying on inter-particle interactions, motion often results from non-reciprocal forces in a chemical medium. Here, we present an alternative strategy that bypasses the need for a chemical environment. By employing broad optical illumination, we establish thermal gradients that give rise to non-reciprocal optothermal interactions, initiating active motion in colloidal assemblies.

Our setup includes two categories of colloids: passive particles and thermally active ones. The latter incorporate light-absorbing materials that convert optical energy into localized heat, forming temperature gradients in their vicinity. These thermal fields generate attractive thermo-osmotic forces that influence the surrounding passive particles, effectively driving their motion. The system exhibits various dynamic behaviors within such a suspension, such as active propulsion and rotational (chiral) movement.

To explore these phenomena in detail, we simulate the system by numerically solving the coupled Langevin equations that govern the particle dynamics under thermal interactive

forces. This investigation aims to understand how light-induced thermal effects and colloidal interactions can be harnessed to control motion in non-equilibrium systems. Our findings offer valuable insights for applications in directed self-assembly, microscale transport, and the broader study of active soft matter. This chapter is an adaptation of the research article "Soft Matter Accepted Manuscript (2025), (arXiv:2411.12488)" [75].

3.1 Introduction and motivation

Soft matter systems operating far from equilibrium exhibit a rich spectrum of dynamic behaviors, including autonomous propulsion [76–86], self-organized patterns [87–89], and collective emergent phenomena [90–94]. These behaviors typically emerge from the interplay between constituent particles and external stimuli [95, 96]. Artificial colloidal systems appeared as an important testbed for probing such dynamics in a controllable manner [97].

A variety of external fields are employed to manipulate colloidal motion, including optical fields [98–106] and chemical gradients [102, 103, 107], as well as electric [108–112] and magnetic fields [111–114]. Inherent structural or composition asymmetry is key in producing unbalanced forces that drive the structure’s motion in most such systems [69, 99, 115–121].

In contrast, recent advances have introduced approaches to exhibit active motion by the use of multiple colloids with different chemical activity [70, 122–126]. In such systems, active motion is enabled by non-reciprocal interactions between the colloidal particles rather than geometric asymmetry. However, implementing such mechanisms often relies on specific chemical environments such as hydrogen peroxide solutions [122] or critical binary mixtures like water-lutidine [70, 123] which can limit their applicability in broader contexts.

It has been demonstrated recently that, in aqueous environments, colloidal particles can be drawn toward heat sources through thermo-osmotic slip flows generated at the substrate [31, 32, 43]. This phenomenon offers a purely physical route without requiring chemical

reagents to generate non-reciprocal attractive forces. These temperature-induced interactions can produce active-like motion in systems comprising heated and non-heated colloids. The interplay between the flow field around the colloidal particles plays a central role in initiating and sustaining this motion. Understanding such unconfined dynamics is important for potential applications in programmable self-assembly [127], microfluidic manipulation [128–131], and probing non-equilibrium phenomena in active matter systems [132]. Optically induced thermal fields offer higher precision and real-time tunability than chemical heating methods. This optothermal approach simplifies control and opens up new opportunities for micromanipulation and engineered self-organization in soft matter systems [39, 133–136].

Taking motivation from this, in this chapter we discuss our investigation of the dynamics of composite colloidal structures formed by absorbing thermally active (A) and non-absorbing passive (P) colloids under exposure to spatially broad optical illumination (see Fig. 3.1(a)). This light exposure creates temperature gradients in the surrounding medium without relying on trapping potentials. We analyze the resulting particle dynamics and characterize various emergent motion patterns, including distinct swimming behaviors (see Fig. 3.1(b)). Complementary simulations further reinforce our experimental observations.

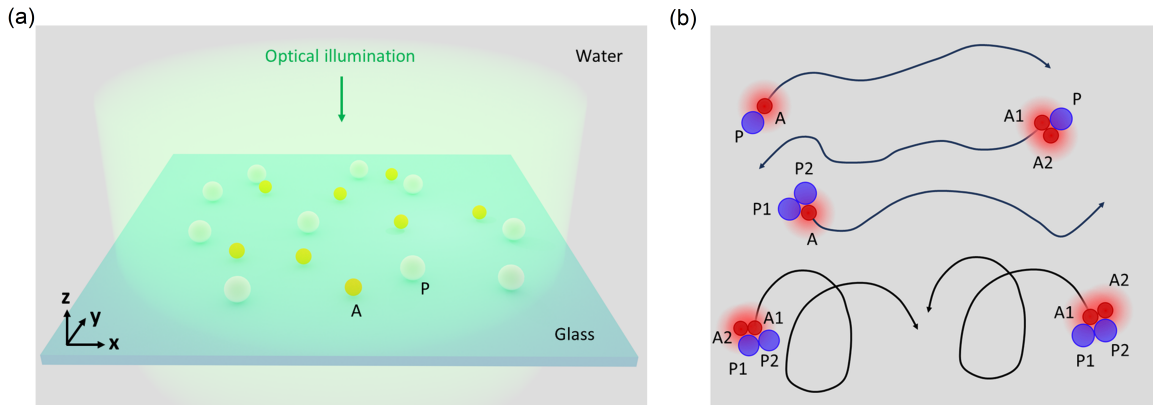


FIGURE 3.1: Illustration of colloidal dynamics under optical illumination. (a) Schematic representation of suspension of passive (P) and thermally active (A) colloids in a widefield optical illumination. Owing to their absorbing characteristics, the thermally active colloids get heated up absorbing the optical beam. (b) These gradients lead to thermo-osmotic interactions, driving the formation of self-assembled structures like dimers, trimers, and quadromers, which exhibit distinct motility behaviors. This figure is reproduced with permission from [75].

3.2 Experimental methods and characteristics of the colloidal particles

3.2.1 Absorbing properties of colloidal particles

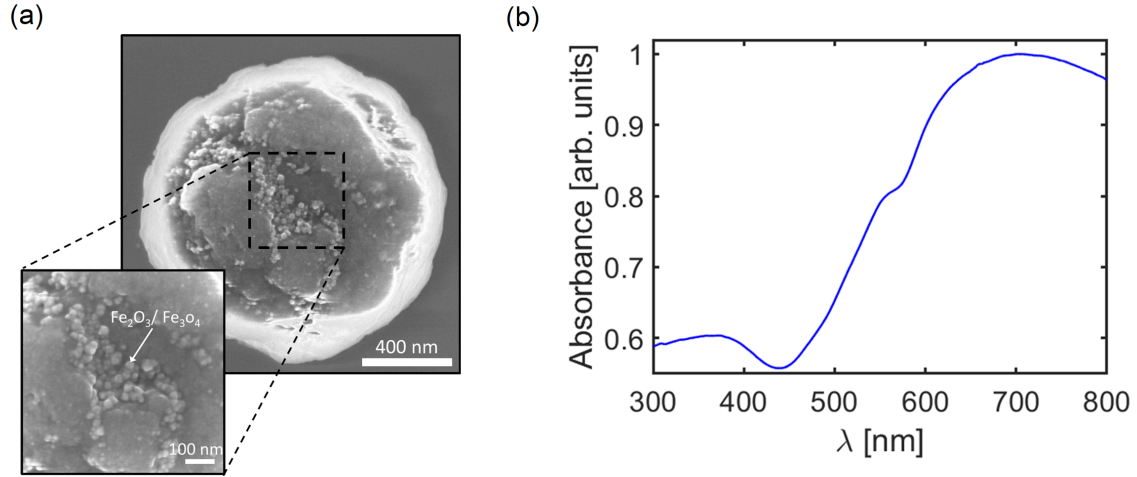


FIGURE 3.2: (a) Scanning electron microscope (SEM) image of a thermally active polystyrene (PS) colloid of diameter of roughly 1.3 μm. The enlarged view reveals the presence of iron oxide nanoparticles (Fe₂O₃/Fe₃O₄), embedded within the colloid, with sizes close to 15 nm. (b) UV-visible absorbance spectrum measured for an aqueous dispersion of these thermally active PS colloids.

In our experiments, we used two distinct types of colloidal particles: passive and thermally active. The passive colloids were melamine formaldehyde (MF) spheres with a diameter of 2 μm. In contrast, the thermally active colloids comprised 1.3 μm polystyrene (PS) particles embedded with iron-oxide nanoparticles of diameter $\approx 15 - 20$ nm. Because of these iron-oxide nanoparticles, the thermally active colloids exhibit optical absorption characteristics. The SEM image of the thermally active colloids, indicating the iron-oxide nanoparticles, is shown in Fig. 3.2(a), and the corresponding UV-visible absorbing profile is shown in Fig. 3.2(b). As a result, they can absorb the optical beam and dissipate heat, resulting in the development of thermal fields around them. These thermally active colloids exhibit interesting temperature and polarization-dependent dynamics as reported in [137]. Both the colloidal microparticles have been procured from microParticles GmbH. A dilute aqueous

suspension of these particles was prepared and introduced into a sealed sample chamber of height $\simeq 120 \mu\text{m}$ for experiments.

3.2.2 Experimental setup

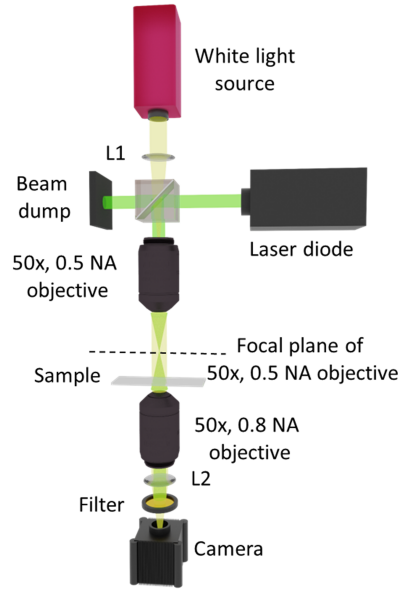


FIGURE 3.3: Schematic of the experimental setup. This figure is adapted with permission from [75].

A dual-channel optical microscopy system is utilized as indicated in Fig. 3.3. A continuous-wave Gaussian laser beam at 532 nm was directed into the sample plane through a 50 $\times$, 0.5 NA objective lens. To create a wide field of optical illumination, we need a plane wave illumination. Due to the limited output power of the laser diode ($<120 \text{ mW}$), we used a defocused beam instead of a proper plane wave. This was accomplished by shifting the sample plane away from the focal plane of the objective (indicated by the dashed line labeled in Fig. 3.3). A white light source, coupled into the optical path using a beam splitter, was incorporated to allow simultaneous bright-field imaging. For most experiments, a laser power of 45 mW was applied. This produced a defocused illumination spot with an approximate diameter of $32 \mu\text{m}$ in the sample plane, resulting in an average optical intensity of $56 \mu\text{W}/\mu\text{m}^2$. The optical intensity could be further modulated by adjusting the laser power or

changing the extent of the beam defocus. Kindly note, due to using a defocused beam, we are not able to use the full extent of optical illumination to perform the experiments because of the variation of optical field intensity. To minimize the effect of the optical field intensity variation and resulting optical force, we have considered the trajectories of the colloidal structures near the centre of the optical field. In our experiments, we consider trajectories less than $10 \mu\text{m}$ in a defocus beam of the beam waist diameter $31 \mu\text{m}$. Within such a span of the trajectories, the colloidal particles experience an intensity variation of $< 18\%$ over their trajectories. For a larger span of trajectories, the influence of the intensity variation and the role of direct optical forces will be significant, which will defy our central experimental goal.

The image of the sample plane was captured through a $50\times$, 0.8 NA objective lens and projected onto a CCD or high-speed camera sensor. Particle motion was recorded at frame rates of 30 or 500 frames per second, depending on the experimental requirements. The resulting videos were analyzed using the TrackMate [138] plugin in Fiji [139] to extract particle trajectories.

3.3 Dynamics in the absence of optical field

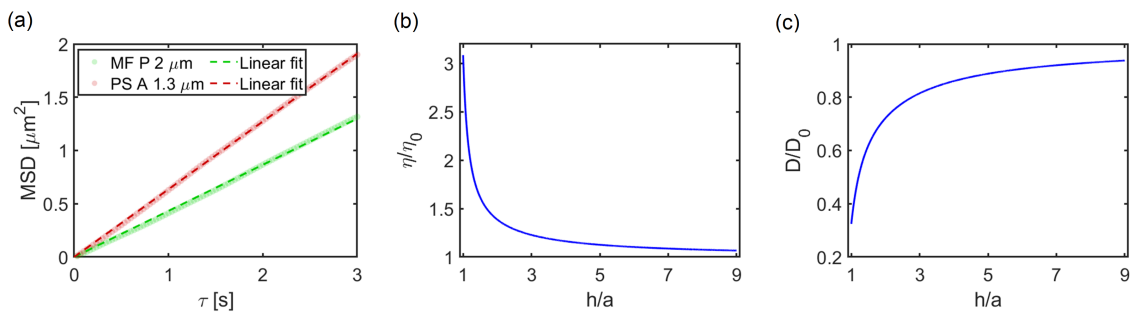


FIGURE 3.4: Dynamics of thermally active and passive colloids in the absence of optical fields. (a) Mean squared displacement (MSD) vs. lag time τ for active (A) and passive (P) colloids, fitted using $MSD(\tau) = 4D\tau$. (b) Height-dependent viscosity and (c) diffusion coefficient, both showing deviations from bulk values η_0 and D_0 . (a) Reproduced from [74] and (b)–(c) from [140] with permission.

Without any external field, both the thermally active and passive colloids exhibit unbiased Brownian diffusion. We quantified the diffusion dynamics by calculating mean squared displacement and fitting with the equation 3.1, and the diffusion constants are summarized in Table 3.1.

$$MSD(\tau) = 4D\tau, \quad (3.1)$$

where τ is lagtime and D is the experimental diffusion constant of the colloids. It can be noted that the experimental diffusion constants are significantly different from their bulk counterparts ($D_0 = k_B T / \gamma_0$, where k_B and γ_0 are the Boltzmann constant and bulk viscous drag coefficient of the colloids). Such differences arise because the colloids are diffusing close to the glass coverslip, resulting in enhanced viscosity and, hence, enhanced viscous drag forces (see Fig. 3.4(b)). Because of this, the colloids exhibit slower diffusion (Fig. 3.4(c)). The experimental and the bulk diffusion constant can be connected by the Faxen correction factor Ξ as given below [10].

$$\Xi = \frac{\gamma}{\gamma_0} = \frac{\eta}{\eta_0} = \frac{D_0}{D} \quad (3.2)$$

$$\Xi^{-1}(h) = 1 - \frac{9}{16} \left(\frac{a}{h} \right) + \frac{1}{8} \left(\frac{a}{h} \right)^3 - \frac{45}{256} \left(\frac{a}{h} \right)^4 - \frac{1}{16} \left(\frac{a}{h} \right)^5 + \dots \quad (3.3)$$

where a and h are the colloid's radius and the colloid's average distance from the glass substrate. From the above Faxen's relation, we can find the average distance of the colloids from the glass substrate.

Colloids	$2a$ [μm]	D [$\mu\text{m}^2/\text{s}$]	D_0 [$\mu\text{m}^2/\text{s}$]	D/D_0	$\gamma_0 \times 10^{-8}$ [N · s/m]	$\gamma \times 10^{-8}$ [N · s/m]	h/a
PS A	1.3	0.16	0.33	0.44	1.2	2.7	1.2
MF P	2.0	0.11	0.23	0.47	1.9	4.0	1.2

TABLE 3.1: Comparison of experimental and bulk diffusion constants of colloids.

3.4 Colloidal dynamics in localized thermally active colloids

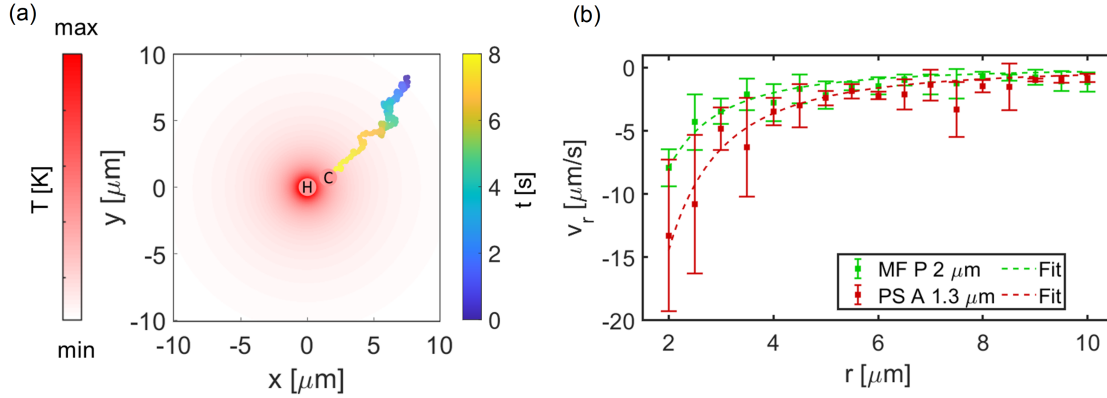


FIGURE 3.5: Colloid migration driven by thermo-osmotic flow. (a) Trajectory of a colloid (C) moving in the flow field generated by a static heat source (H). (b) Radial velocity (v_r) vs. distance (r) from the heat source, showing an inverse-square dependence ($v_r = A/r^2$, dashed fit). Error bars indicate standard deviations from five measurements. The intensity of the optical field used is $0.64 \text{ mW}/\mu\text{m}^2$, which corresponds to the temperature rise of 7.5 K . For temperature estimation, we have followed a similar protocol as discussed in section 4.5.2. This figure is reproduced from [74] with permission.

To understand the motion of the colloids in the presence of an external thermal field, we have used drop-casted thermally active colloids illuminated with focused light (with intensity $I = 0.65 \text{ mW}/\mu\text{m}^2$) serving as a localized heat source. This results in a temperature gradient in the surrounding fluid, which harnesses thermo-osmotic slip flow in the substrate and around the colloids. Because of this flow field, the colloids experience a drag force that effectively pulls them towards the heat source [31, 32]. Fig. 3.5(a) illustrates the motion of a thermally active colloid (C) moving toward the heat source H. Similar behavior is also exhibited by the passive colloids.

The radial velocity of this migration depends on the distance from the heat source. As shown in Fig. 3.5(b), particles accelerate as they approach the hot region. This velocity can be used to determine the effective thermo-osmotic slip-driven drag force acting on the colloids as expressed below.

$$v_r = \frac{\chi \nabla T}{T_s} = -\frac{\chi \Delta T_0 a}{T_s r^2} = \frac{A}{r^2} \quad (3.4)$$

$$F_d = \gamma v_r. \quad (3.5)$$

Here, χ denotes the effective thermo-osmotic slip coefficient, $T_s = 293$ K is the ambient fluid temperature, and ΔT_0 is the surface temperature increment of the heat source H due to laser heating.

To calculate the radial speed of the colloid due to thermo-osmotic flow, first, we have captured the trajectories. Then we have estimated the instantaneous velocity by using consecutive frames as a function of the instantaneous position. The data is binned in equally spaced predefined radial distance bins (bin width $0.5 \mu\text{m}$). The average velocity at each bin is then considered as the velocity at the bin center. We have performed the same calculation for $\simeq 10$ trajectories of the colloids, and the average and standard deviation of the velocity data are considered as distance-dependent experimental radial velocity and the corresponding error bar. Equation 3.4 fits the experimental velocity data well as illustrated in Fig. 3.5(b), and the fit yields values of the parameter A for the system. These values are used to compute the effective thermo-osmotic slip coefficient χ for the system and are summarized in Table 3.2.

Colloids	Diameter [μm]	A [$\mu\text{m}^3 \cdot \text{s}^{-1}$]	$\chi = -\frac{AT_s}{\Delta T_0 a}$ [$\text{m}^2 \cdot \text{s}^{-1}$]
MF P	2.0	-31.8	1.87×10^{-9}
PS A	1.3	-57.8	3.39×10^{-9}

TABLE 3.2: Thermo-osmotic slip coefficients χ of the system.

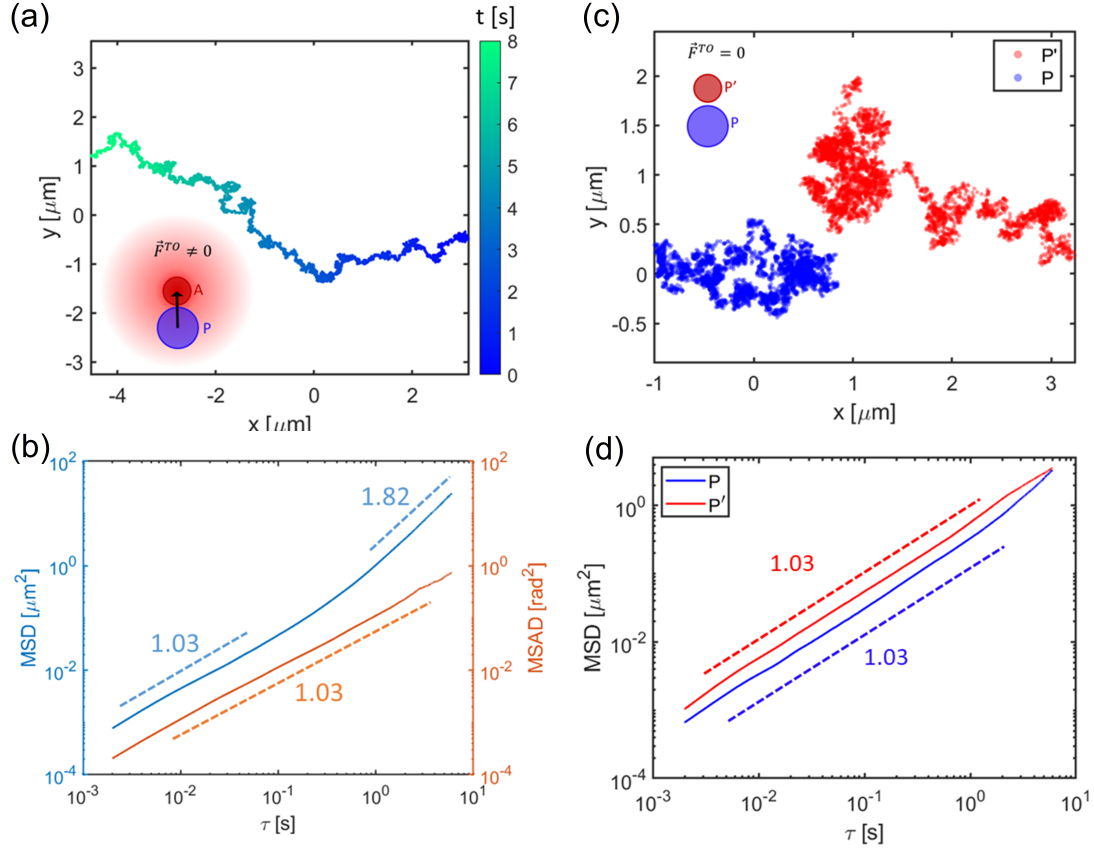


FIGURE 3.6: Motion of colloidal dimers under a defocused optical beam. (a) Trajectory of an active dimer formed by a thermally active (A) and passive (P) colloid, showing self-propulsion due to thermo-osmotic forces. Inset: The dimer orientation vector (black arrow). (b) The mean squared displacement (MSD) of the active dimer shows a ballistic to diffusive transition. The mean squared angular displacement (MSAD) shows only a diffusive trend. (c) A dimer with two passive colloids (P, P') does not show active motion. (d) Corresponding MSD scales linearly, indicating purely diffusive dynamics. 3-5 independent trajectories are recorded to perform the MSD and MSAD calculations. This figure is reproduced from [75] with permission.

3.5 Emergence of activity in colloidal dimer structures under wide-field optical illumination

Under broad optical illumination, thermally active colloids absorb laser energy and dissipate heat efficiently. This heating results in a temperature gradient around the thermally active colloids, which harness thermo-osmotic slip flows in the surrounding fluid. These flows act as environmental cues that influence the dynamics of nearby colloids. When an active colloid (A) interacts with a passive colloid (P), the resulting thermo-osmotic forces become non-reciprocal, leading to the formation of various bound configurations. The simplest of

these is a dimer composed of one active and one passive particle. Due to the asymmetric interaction, the dimer acquires net propulsion, as shown in the trajectory displayed in Fig. 3.6(a) (also see video 1 [141]). To quantify this motion, we compute the mean squared displacement (MSD), shown in Fig. 3.6(b). The MSD exhibits a crossover from an initial diffusive regime to a ballistic one, consistent with typical active matter behavior [142]. Active particles usually show three characteristic regimes in their MSD: an early-time diffusive regime, a mid-time ballistic phase, and a late-time diffusive regime. However, since the rotational diffusion time of colloids is typically on the order of 10^2 seconds, and our experiments span approximately 10 seconds, only the first two regimes are observed here.

The active dimer's orientation can be defined by a vector pointing from the passive to the active colloid, as shown by the black arrow in the inset of Fig. 3.6(a). The mean squared angular displacement (MSAD) of this orientation vector is plotted in Fig. 3.6(b), where the linear scaling (slope ≈ 1) indicates unbiased rotational diffusion. In contrast, replacing the active colloid with a passive colloid of identical size and composition (denoted P') prevents the formation of a propelling dimer. Instead, both particles diffuse independently, as shown in Fig. 3.6(c) (also see video 1 [141]). Their corresponding MSDs, depicted in Fig. 3.6(d), exhibit linear scaling, confirming purely Brownian motion. These observations highlight the critical role of non-reciprocal thermo-osmotic interactions, driven by slip flows around active colloids, in enabling self-propulsion without the need for any additional chemical stimuli.

3.6 Forces acting on the system

3.6.1 Thermo-osmotic flow induced drag force

We have simulated the thermo-osmotic flow field to understand how the thermo-osmotic flow-induced drag force harnesses non-reciprocal interactions in the active dimer structure. To do that, we have coupled the heat transfer module with the laminar flow module in the finite element method-based solver COMSOL Multiphysics. The simulation geometry is

shown in Fig. 3.7(a). The water column, where the colloidal particles are present, is sandwiched between two glass regions. We have considered the surface of a thermally active colloid as a heat source with elevated surface temperature ΔT_A compared to the surrounding bath. We have considered all the solid-liquid surfaces (indicated by 1-4 in red text) to have non-zero slip boundary conditions and a dimensionless slip coefficient $\sigma_T = \chi\rho/\eta$ $m^2 \cdot s^{-1}$. If we consider $\chi \simeq 3 \times 10^{-9} m^2 \cdot s^{-1}$, then $\sigma_T \simeq 0.003$. All other material parameters used like thermal conductivity (κ_c), heat capacity at constant pressure (C_p) and density (ρ) are as given in Table 3.3.

Material	κ_c [$W \cdot m^{-1} \cdot K^{-1}$]	C_p [$J \cdot Kg^{-1} \cdot K^{-1}$]	ρ [$Kg \cdot m^{-3}$]
MF (P)	0.50	1120	1500
PS (A)	0.18	1070	1215
Glass	1.38	703	2203
Water	0.60	4180	997

TABLE 3.3: Material parameters used in the simulation.

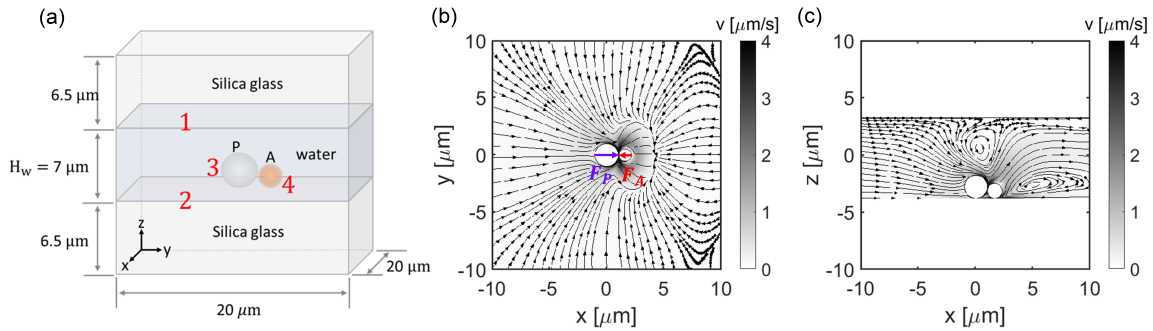


FIGURE 3.7: Thermo-osmotic flow field simulation at $\Delta T_A = 2$ K surface temperature increment of the thermally active colloids. (a) Schematic simulation geometry. The (b) in-plane and (c) out-of-plane thermo-osmotic slip flow field around the active dimer structures.

The resulting in-plane and out-of-plane thermo-osmotic flow fields around the active dimer structures are shown in Fig. 3.7(b) and (c), respectively. The flow is of the order of magnitude $1 \mu m/s$, and the flow-induced drag force acting on the passive and thermally active colloids are approximately $\vec{F}_P \simeq 0.14$ pN and $\vec{F}_A \simeq -0.06$ pN. From Fig. 3.7(b), we can see that these forces lead to a compelling attraction between the colloids. In addition, since the force acting on passive and thermally active colloids are of different magnitudes,

harnessing non-reciprocity. This leads to a net force imbalance in the system: $\Delta \vec{F} \simeq 0.08$ pN, acting from the passive colloids to the thermally active colloids. Thus, because of this force imbalance, the structure will tend to move heading towards the thermally active colloid. Now if we compare this force imbalance with the approximated thermo-osmotic force acting on passive colloids by considering the inverse-square flow field approximation as $\vec{F}_P = \gamma \frac{\chi \Delta T_A a}{T_s r^2} \simeq 0.07$ pN, which is comparable to as we observed from the thermo-osmotic flow field simulation.

3.6.2 Convective flow-induced force

Apart from the thermo-osmotic flow field, in the microscopic systems, the convective flow may also play a significant role. To understand the convective flow-induced drag forces, we have also simulated the convective flow field in COMSOL. To do that, we have followed the protocol we discussed for the thermo-osmotic flow field in the previous subsection. The only deviation is that all the solid-liquid interfaces are considered to have a no-slip boundary condition and a temperature-dependent density of the water medium is considered. At the same $\Delta T_A = 2$ K surface temperature increment of the thermally active colloid, the density profile around the colloidal dimer is shown in Fig. 3.8 (a). The resulting in-plane and out-of-plane convective flow fields around the structure are shown in Fig. 3.8(b) and (c), respectively. Convective flow is of the order of magnitude 1 nm/s, almost three orders of magnitude less than thermo-osmotic flows. The corresponding convective drag forces indicate a repulsive interaction between the colloids, which are approximately $\vec{F}_P \simeq 2.08 \times 10^{-6}$ pN and $\vec{F}_A \simeq 0.67 \times 10^{-6}$ pN. The magnitude of net convective force imbalance is $\Delta \vec{F} \simeq 1.41 \times 10^{-6}$ pN, which is almost four orders of magnitude compared to the slip-flow induced force imbalance. Thus, we can say that convective flow-induced forces play an insignificant contribution to the dynamics of the active dimers.

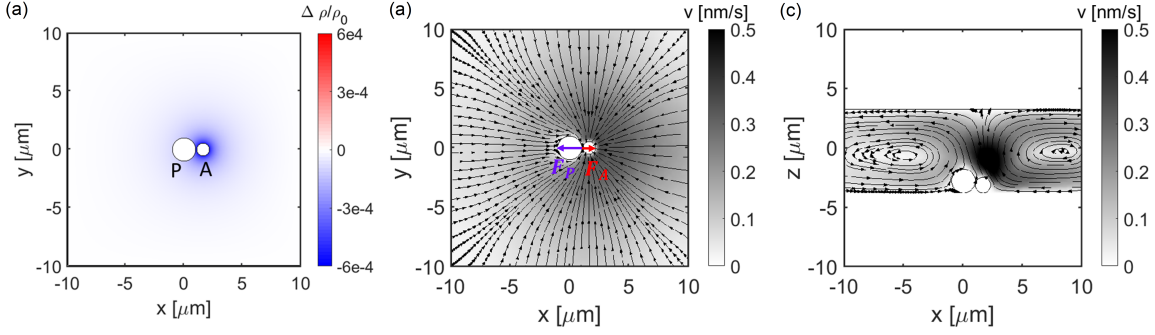


FIGURE 3.8: Convective flow field simulation at $\Delta T_A = 2$ K surface temperature increment of the thermally active colloids. (a) Density profile around the active dimer structures. The (b) in-plane and (c) out-of-plane convective flow field around the active dimer structures.

3.6.3 Optical forces

To understand the role of optical force on the active dynamics. At first, we consider the electric field of an optical beam polarized along the $\hat{y}$ axis and propagating in the $-\hat{z}$ direction is expressed as:

$$\vec{E} = \hat{y} E_0(x, y) e^{-ikz}, \quad (3.6)$$

where $E_0(x, y)$ represents the spatial variation of the electric field amplitude. This spatial dependence is negligible in the case of an ideal, wide-field uniform illumination. Under such an approximation, the electric field simplifies to:

$$\vec{E} = E_0 e^{-ikz} \hat{y}. \quad (3.7)$$

The associated optical intensity is given by:

$$I = \frac{1}{2} c \epsilon_0 E_0^2, \quad (3.8)$$

where c is the speed of light, and ϵ_0 is the vacuum permittivity.

For this configuration, the scattering force $\vec{F}_{\text{scat}}$ and gradient force $\vec{F}_{\text{grad}}$ acting on a colloid are described as [143]:

$$\vec{F}_{\text{scat}} = -\hat{z} \frac{8}{3} \pi \frac{n_w}{c} k^4 \alpha^2, \quad \vec{F}_{\text{grad}} = 2\pi \frac{n_w}{c} \alpha \nabla I,$$

where α denotes the polarizability of the colloid and n_w is the refractive index of the surrounding water medium. Ideally, for uniform optical intensity I , the intensity gradient ∇I vanishes, leading to a vanishing gradient force. The scattering force is the only significant optical force, which acts along the beam propagation direction ($-\hat{z}$), effectively pressing the colloids against the bottom glass substrate in our geometry. As a result, in combination with gravitational settling, the motion of the colloids is effectively constrained to two dimensions. The convention gradient and scattering optical field do not exert any significant in-plane force component to alter or influence the dynamics of the active dimer structures.

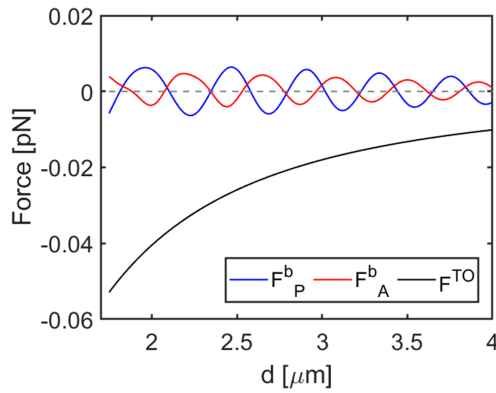


FIGURE 3.9: The computed optical binding forces on passive (blue) and thermally active (red) colloids are significantly weaker than the corresponding attractive thermo-osmotic force (black).

This figure is reproduced with permission from [75].

Apart from the conventional gradient and scattering optical forces, optical binding forces often influence the dynamics of colloidal structures. To investigate the contribution of optical binding between thermally active and passive colloids, we numerically simulated it using the MiePy toolbox [144]. For that, the refractive indices used $n_P = 1.6800$ and $n_A = 1.6777 + 0.0169i$ for passive melamine formaldehyde (MF) colloids and active polystyrene (PS) colloids [137] respectively. A linearly polarized light beam along the $\hat{y}$ -axis with an intensity of $100 \mu\text{W}/\mu\text{m}^2$ is considered, and the colloids were placed along the $\hat{x}$ -axis. The resulting optical binding forces on passive (P) and active (A) particles were calculated by integrating Maxwell's stress tensor over the particle surfaces. These forces are depicted in Fig. 3.9 with blue and red curves, respectively. The calculated force profiles exhibit an oscillatory behavior with a characteristic spacing of approximately 400 nm. This

length scale corresponds to the effective wavelength of the laser in water:

$$\lambda_w = \frac{\lambda_0}{n_w} = \frac{532}{1.33} = 400 \text{ nm},$$

This is indicative of optical binding effects arising due to interference.

Now, if we consider the attractive thermo-osmotic flow-induced drag force between colloids:

$$\vec{F}^{\text{TO}} = \gamma_P \frac{\chi}{T_s} \vec{\nabla} T = -\gamma_P \frac{\chi}{T_s} \frac{\Delta T a_A}{d^2} \hat{n}, \quad (3.9)$$

where $\gamma_P = 4 \times 10^{-8} \text{ Ns/m}$ is the drag coefficient of the passive colloid, $\chi = 1.87 \times 10^{-9} \text{ m}^2/\text{s}$ is the thermo-osmotic slip coefficient, $T_s = 300 \text{ K}$ is the ambient temperature, ΔT is the temperature difference, $a_A = 0.65 \mu\text{m}$ is the radius of the active colloid, and $\hat{n}$ is a radial unit vector pointing away from the active particle. The black curve in Fig. 3.9 represents the thermo-osmotic force for $\Delta T = 1 \text{ K}$, which is consistent with experimental estimates of the temperature rise for thermally active colloids under similar laser intensities [74].

Now, comparing the optical binding and the thermo-osmotic forces in Fig. 3.9, we can see that the optical binding forces are considerably weaker than the thermo-osmotic attraction. Therefore, optical binding also does not play a significant role in the active dynamics of the colloidal dimer structures and the only dominant contribution arises from thermo-osmotic flows.

3.7 Dynamics of higher order colloidal structures

To better understand how thermal gradients influence the collective behavior of colloidal assemblies, we experimentally investigated the motion of trimer and quadromer configurations composed of thermally active and passive particles. Two trimer variants were analyzed: (i) **AT1**, consisting of a single thermally active colloid (A1) and two passive colloids (P1 and

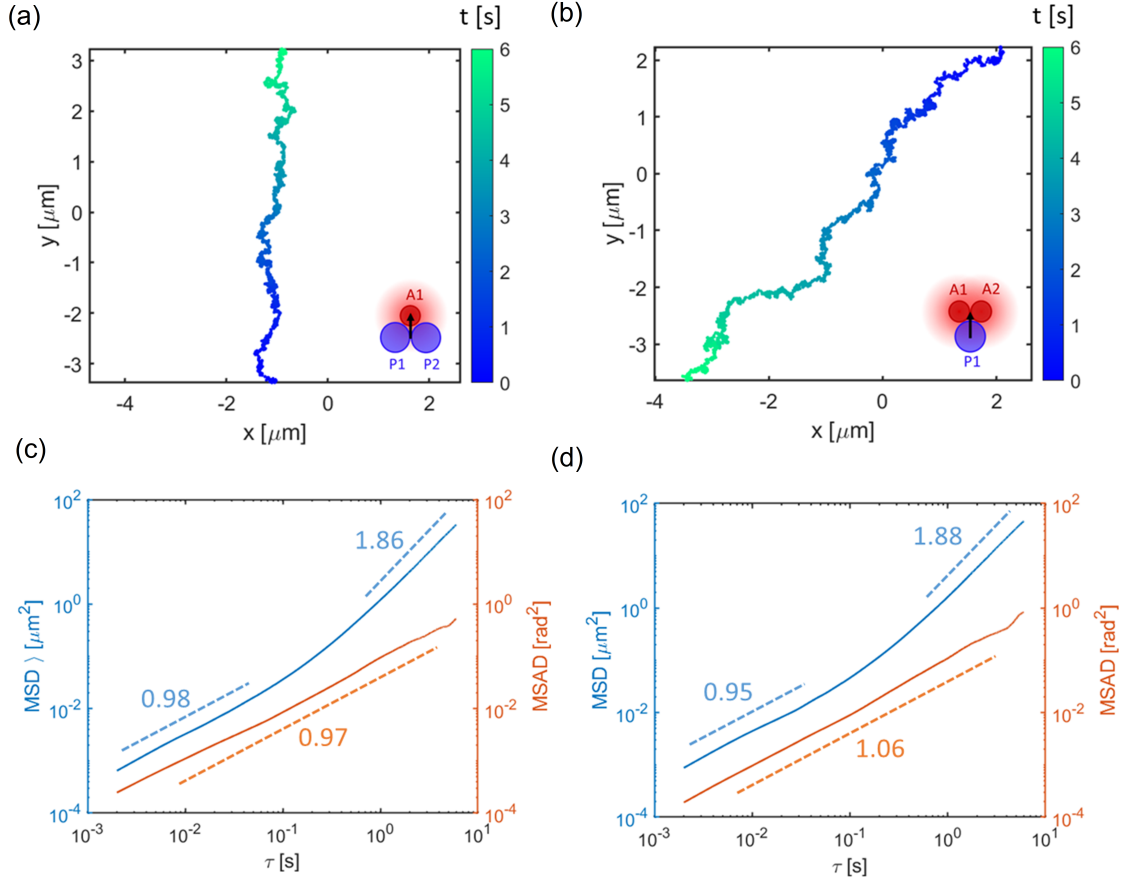


FIGURE 3.10: Dynamics of active trimer structures. (a)-(b) Trajectories of trimers with (a) one active and two passive colloids, and (b) two active and one passive colloid, both showing persistent propulsion without chirality. (c)-(d) The MSD and MSAD of the trimer structures in (a) and (b). The MSD in both cases indicates active motion, whereas MSAD shows diffusive behaviour, confirming the absence of directional bias. This figure is reproduced with permission from [75].

P2), and (ii) **AT2**, comprising two thermally active colloids (A1 and A2) and one passive colloid (P1). In both arrangements, the non-reciprocal thermo-osmotic forces are aligned along the longitudinal symmetry axis of the structure, resulting in net propulsion akin to that observed in active dimer systems. Figures 3.10(a) and 3.10(b) display respective time evolution trajectories of the AT1 and AT2 trimers (see video 2 [141]). The corresponding MSD and MSAD of the two trimer configurations are shown in Fig. 3.10(c) and 3.10(d). The trajectories and the MSD and MSAD reveal apparent translational motion without sustained angular displacement, indicating a lack of rotational preference.

In contrast, the quadromer configuration composed of two passive and two thermally

active colloids (Fig. 3.11(a)) exhibits qualitatively different behavior due to its asymmetric layout. Here, the passive particle P1 is positioned farther from the active colloid A2 than P2 is. This spatial asymmetry leads to a disparity in the magnitude of the attractive thermosmotic forces experienced by the passive colloids, with P2 subjected to a stronger pull than P1. As all four colloids migrate collectively during motion, this imbalance causes the entire structure to drift toward P1, generating a net rotational tendency (see video 2 [141]). The result is a counter-clockwise chiral motion, as visualized in Fig. 3.11(a).

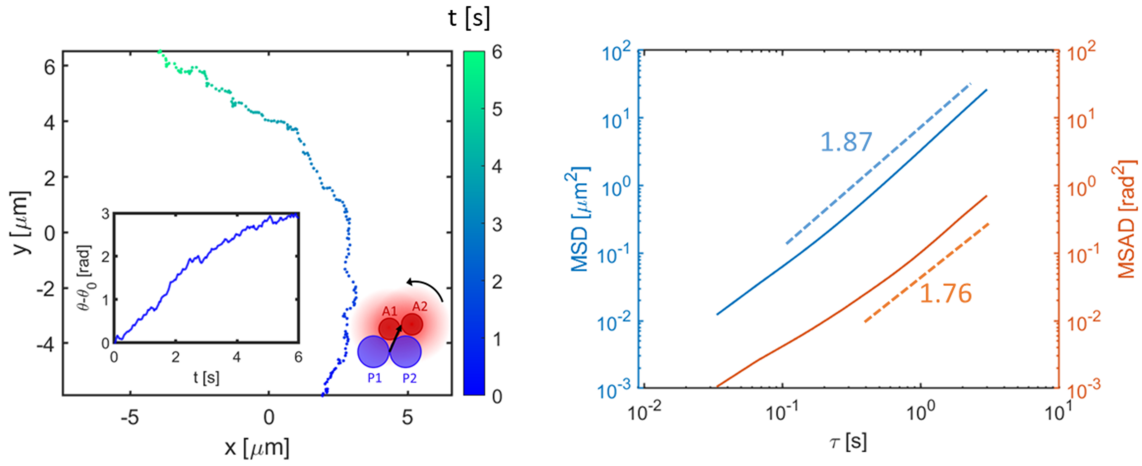


FIGURE 3.11: Dynamics of active quadromer structures and the emergence of chirality. (c) The active quadromer structures, composed of two passive (P1, P2) and two thermally active colloids (A1, A2), exhibit anticlockwise handedness based on the arrangement of the constituent colloids. The corresponding time evolution of the orientation vector for active quadromers is shown in the inset. (b) The MSD and MSAD indicate the activity signature in both linear and angular coordinates. This figure is reproduced with permission from [75].

Due to the finite size of the illumination region, a complete circular trajectory was not captured experimentally. The motion was recorded using a CCD camera at 30 frames per second. To quantify the observed rotation, we defined an orientation vector extending from the center of mass of the passive pair (P1 and P2) to that of the active pair (A1 and A2). The temporal evolution of this vector, shown in the inset of Fig. 3.11(a), confirms persistent angular displacement. Further, the calculated mean-squared displacement (MSD) and mean-squared angular displacement (MSAD) of the quadromer are shown in Fig. 3.11(b). Unlike the linear trends observed in active dimer and trimer systems, both MSD and MSAD exhibit ballistic scaling, indicating sustained propulsion with angular bias.

To further elucidate the role of thermo-osmotic forces in generating both linear and chiral motion, we performed simulations of the colloidal dynamics as discussed in the following section.

3.8 Dynamics simulation of the active colloidal structures

To investigate the motion of colloids induced by temperature gradients, we modeled passive (P) and thermally active (A) particles with diameters of 2 μm and 1.3 μm , respectively. In the simulation, active colloids are treated as localized heat sources, possessing a higher temperature than their passive counterparts. The stochastic dynamics of each particle i are described using the Langevin framework:

$$\gamma_i \vec{v}_i = \vec{F}_i^{\text{ext}} + \sqrt{2k_B T_i \gamma_i} \vec{\zeta}_i, \quad (3.10)$$

where $\vec{v}_i$ is the velocity, γ_i is the viscous drag coefficient, T_i is the temperature of colloid i , and $\vec{\zeta}_i$ is a delta-correlated random noise term. The term $\vec{F}_i^{\text{ext}}$ includes all deterministic interactions, discussed below.

We modeled the optical field as an extended plane wave in simulations (ideal condition), in contrast to the finite beam profile in our experiments. Under such conditions, no spatially varying optical gradient force exists in the lateral direction. The scattering force component directs colloids toward the substrate, effectively confining motion to two dimensions. Hence, lateral optical forces were neglected, and illumination-induced temperature gradients are the only contributions from the optical field. Potential contributions from optical binding were considered negligible compared to thermo-osmotic forces.

A thermally active colloid at temperature $T_A = T_s + \Delta T$ produces a radially decaying temperature profile:

$$T(r > a_A) = \Delta T \frac{a_A}{r} + T_s, \quad (3.11)$$

where a_A is the radius of the active colloid, and T_s is the ambient temperature. This gradient creates a thermo-osmotic slip velocity $\vec{v}_j$ at the substrate-liquid interface:

$$\vec{v}_j = \frac{\chi}{T_s} \vec{\nabla} T_j, \quad (3.12)$$

where χ is the slip coefficient. This flow exerts a drag on neighboring colloids:

$$\vec{F}_{ij}^{\text{TO}} = \gamma_i \vec{v}_j. \quad (3.13)$$

Only thermally active colloids generate such flows. Passive colloids do not produce but can experience this flow-induced drag. Thus, $\vec{F}_{ij}^{\text{TO}} = 0$ when j is passive, and $\vec{F}_{ij}^{\text{TO}} \neq 0$ when j is active.

To enforce excluded volume interactions, we included Lennard-Jones (LJ) repulsion between colloids:

$$\vec{L}J_{ij} = 4\epsilon \left(12 \frac{\sigma^{12}}{r_{ij}^{13}} - 6 \frac{\sigma^6}{r_{ij}^7} \right) \hat{r}_{ij}, \quad (3.14)$$

where $\sigma = a_i + a_j$, ϵ is the depth of the potential, and r_{ij} is the center-to-center distance.

The complete Langevin equation governing the motion of particle i is:

$$\gamma_i \vec{v}_i = \sum_{j \neq i} \vec{L}J_{ij} + \sum_{j \neq i} \vec{F}_{ij}^{\text{TO}} + \sqrt{2k_B T_i \gamma_i} \vec{\zeta}_i. \quad (3.15)$$

3.8.1 Simulated dynamics of dimer structures

For a dimer composed of one A and one P colloid, the system simplifies to:

$$\vec{v}_A = \frac{d\vec{r}_A}{dt} = \frac{1}{\gamma_A} \left(\vec{L}J_{A,P} + \sqrt{2k_B T_A \gamma_A} \vec{\zeta}_A \right), \quad (3.16a)$$

$$\vec{v}_P = \frac{d\vec{r}_P}{dt} = \frac{1}{\gamma_P} \left(\vec{L}J_{P,A} + \vec{F}_{P,A}^{\text{TO}} + \sqrt{2k_B T_P \gamma_P} \vec{\zeta}_P \right). \quad (3.16b)$$

We numerically solved these coupled equations to reproduce the behavior of dimer systems. The simulation parameters used: $\epsilon = 20 \times 10^{-21} \text{ J}$, $\chi = 1.87 \times 10^{-9} \text{ m}^2/\text{s}$, $\gamma_A = 1.7 \times 10^{-8} \text{ N s/m}$, and $\gamma_P = 4.0 \times 10^{-8} \text{ N s/m}$ [74]. The trajectory of an active dimer at $\Delta T = 2 \text{ K}$ is shown in Fig. 3.12(a) (also see video 3 [141]). The corresponding MSD (Fig. 3.12(b)) exhibits a transition from diffusive to ballistic and back to diffusive regimes in MSD, while MSAD remains diffusive throughout.

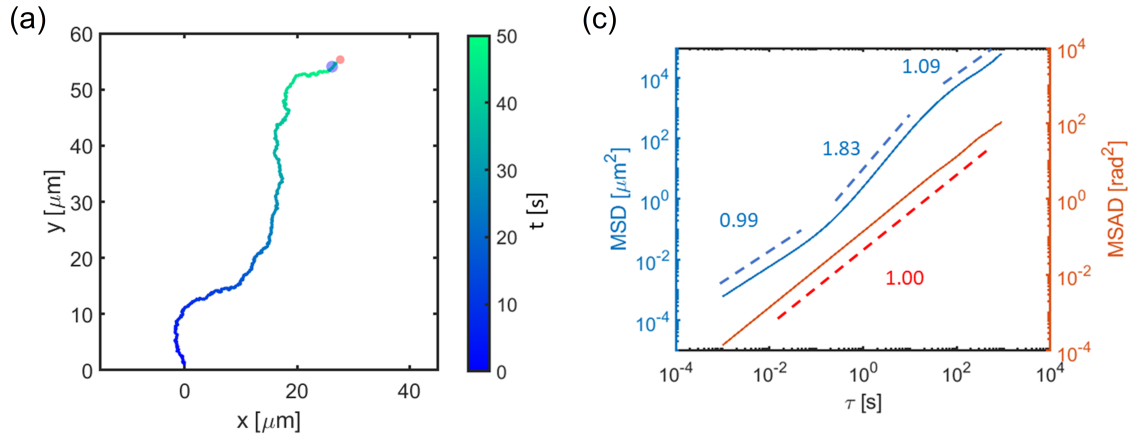


FIGURE 3.12: Simulated dynamics of active dimers. (a) Trajectory of a dimer composed of one passive and one thermally active colloid. (b) MSD shows a diffusive-to-ballistic-to-diffusive transition, while MSAD remains diffusive throughout. This figure is reproduced with permission from [75].

3.8.2 Simulated dynamics of trimer dynamics

This framework was extended to simulate trimer and quadromer configurations (see video 4 [141]). Sample trajectories for two distinct trimer designs are shown in Fig. 3.13(a,b), and their MSD/MSAD are indicated in Fig. 3.13(c,d), respectively. These mirror the dynamics of dimers.

3.8.3 Simulated dynamics of quadromer structures

Simulations of quadromers revealed chirality, as evident in Fig. 3.14(a), where the temporal evolution of orientation vectors indicates handed rotational dynamics (also see video 5 [141]). The MSD and MSAD curves in Fig. 3.14(b) show ballistic trends consistent with

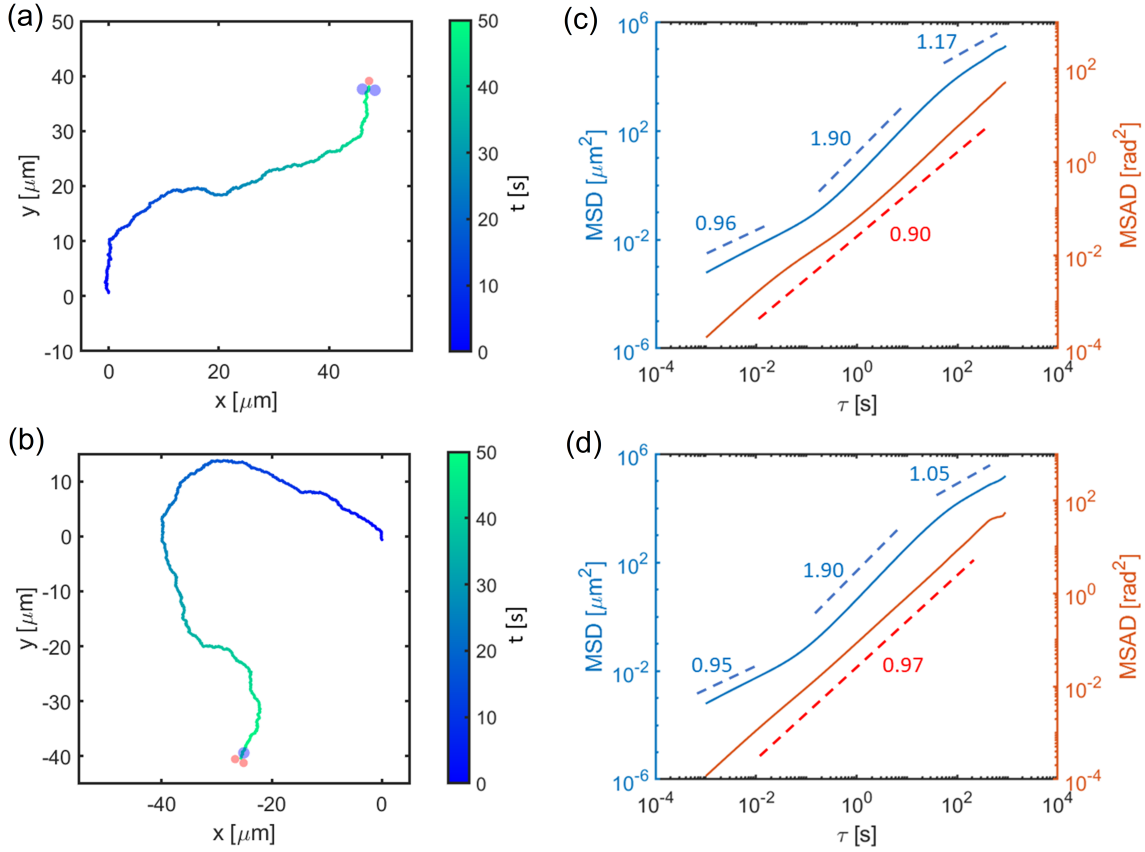


FIGURE 3.13: Simulated dynamics of active trimers. (a)-(b) Simulated trajectories of two trimer configurations. (c)-(d) MSDs and MSADs for both the trimers. MSDs show a diffusive-to-ballistic-to-diffusive trend, while MSAD remains diffusive over the entire timescale. Simulated dynamics of active trimers. The simulated trajectories of two active trimer configurations are shown in (a) and (b). This figure is reproduced with permission from [75].

persistent angular and propelling motion. In overextended time, MSAD reverts to diffusive behavior due to thermal noise, which causes transitions between opposite chiral states (see video 6 [141]). A comparison of simulation and experimental MSD timescales is summarized in the subsection given below.

3.9 Estimation of timescales

In case of active structures, there are typically two different timescales: rotational diffusive timescale (τ_r) and translational timescale (τ_t). The rotational diffusive time scale is related to the rotational diffusion constants (D_r) of the systems as $\tau_r = 1/D_r$. This rotational diffusive time scale separates the MSD into two regimes:

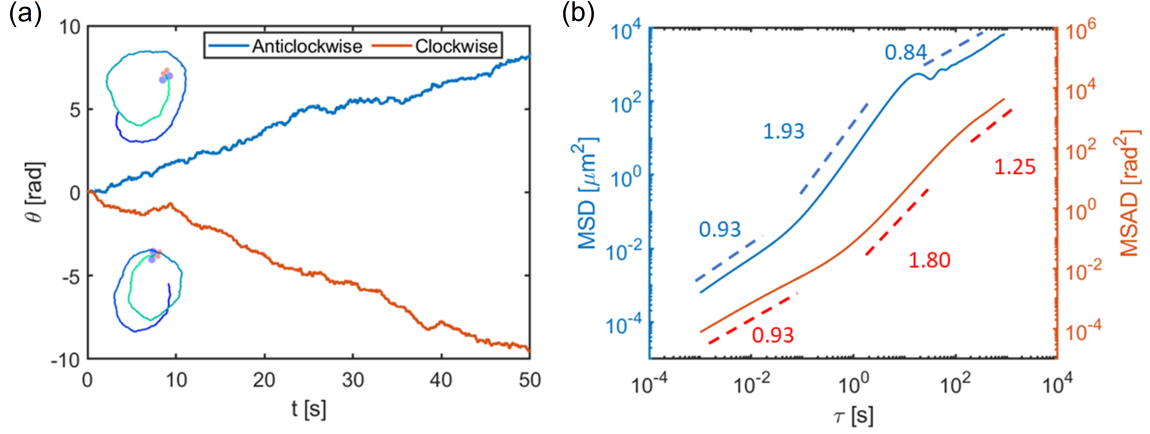


FIGURE 3.14: Simulated dynamics of active quadromers. (a) Simulated trajectory and orientation vector of a representative quadromer showing persistent motion with defined chirality. (b) MSD and MSAD confirm active translational and rotational dynamics. This figure is reproduced with permission from [75].

- large timescale linear regime: $\tau \gg \tau_r \rightarrow MSD(\tau) = (4D + v^2\tau_r)\tau$
- short timescale active regime: $\tau \ll \tau_r \rightarrow MSD(\tau) = 4D\tau + v^2\tau^2$.

Where τ , τ_r , D , and v are the lagtime, rotational diffusion time scale, translational diffusion constant, and the active velocity of the system. Now, if we closely look at $\tau \ll \tau_r \rightarrow MSD(\tau) = 4D\tau + v^2\tau^2$. In this case, we can clearly observe that the MSD has both linear and quadratic terms. Now, further depending on the time scale regime, either the linear or the quadratic term will dominate.

- The linear term is dominating when $4D\tau > v^2\tau^2 \rightarrow \tau < 4D/v^2$.
- The quadratic term is dominant when $4D\tau < v^2\tau^2 \rightarrow \tau > 4D/v^2$.

Thus, the time scale $\tau_t = 4D/v^2$ further splits the MSD into two different regimes, and this τ_t is called the translational timescale. To calculate the translational time scale, we have to consider the MSD data to be below $\tau \ll \tau_r$. Typically for microscopic systems $\tau_r \approx 10^2 \text{ s}$. We have considered the MSD for 1s, which falls under $\tau \ll \tau_r$ condition. The MSD data is then fitted with:

$$MSD(\tau) = 4D\tau + v^2\tau^2, \quad (3.17)$$

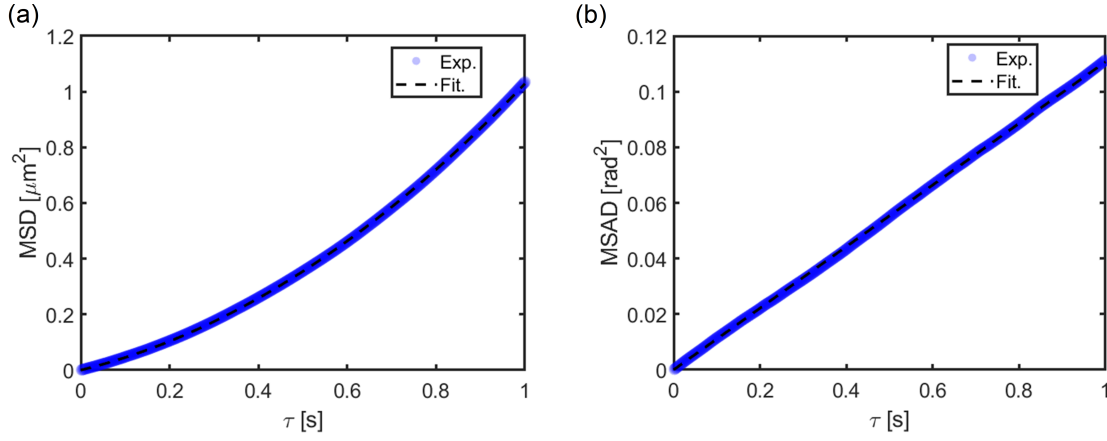


FIGURE 3.15: (a) MSD and (b) MSAD of active dimer structures over 1s. The MSD is fitted with the equation $MSD(\tau) = 4D\tau + v^2\tau^2$ and MSAD is fitted with $MSAD(\tau) = 2D_r\tau$.

where the translational diffusivity D and the active velocity v are obtained as fitting parameters. Inserting these obtained D and v into $\tau_t = 4D/v^2$ we obtained the value of translational timescale. For example, the MSD for an active dimer structure is shown in Fig. 3.15(a). The MSD is fitted with equation (3.17) and we get $D = 0.098 \mu\text{m}^2 \cdot \text{s}^{-1}$, $v = 0.799 \mu\text{m} \cdot \text{s}^{-1}$ which gives us $\tau_t = 0.61\text{s}$. To get the rotational time scale τ_r we have fitted the MSAD of the AD structure with:

$$MSAD(\tau) = 2D_r\tau, \quad (3.18)$$

The experimental MSAD and the corresponding linear fitting are shown in Fig. 3.15(b). We get the rotational diffusion constant as $D_r = 0.056 \text{ rad}^2 \cdot \text{s}^{-1}$ and the rotational diffusive time scale as $\tau_r = 1/D_r = 17.85 \text{ s}$. Please note that in case of active quadromer structures, where there is a preferential rotational bias, the MSAD $\tau \ll \tau_r$ takes the form:

$$MSAD(\tau) = 2D_r\tau + \omega^2\tau^2, \quad (3.19)$$

where D_r is the rotational diffusion constant and ω is the angular velocity of the quadromer structures.

3.10 Comparison between experimental and simulated timescales

The experimental and simulated characteristic timescales for various active colloidal structures are summarized in Table 3.4. It is evident that there is a 2.5-6.5 fold difference in the translational timescale (τ_t) between experiments and simulations, while the rotational timescale (τ_r) shows better agreement. However, the simulation aims to reproduce qualitatively similar dynamics to those observed experimentally. The observed discrepancy is attributed mainly to the assumed temperature increment of the active colloids (ΔT_A) in the simulations. All simulations were conducted using a fixed surface temperature rise of $\Delta T_A = 2$ K. This choice stems from the experimental limitation that prevents reliable temperature measurements below a 4 K increase in water. The actual temperature rise in our system is much smaller.

Structures	Experiment		Simulation	
	$\tau_t[s]$	$\tau_r[s]$	$\tau_t[s]$	$\tau_r[s]$
AD	0.61	18.06	0.16	14.10
AT1	0.29	21.46	0.12	31.17
AT2	0.26	18.60	0.04	23.32
Quadromer	0.12	55.22	0.03	44.05

TABLE 3.4: Experimental and simulated timescales (at $\Delta T_A = 2$ K) of different active structures.

We carried out additional simulations using lower temperature increments to investigate the effect of ΔT_A . Please note that the variation of temperature magnitude does not qualitatively change the dynamics. For lower temperature values, the simulations become much more computationally extensive. Fig. 3.16 presents the variation in timescales with respect to ΔT_A . The translational timescale $\tau_t = 4D/v^2$ shows a strong temperature dependence because velocity v increases with ΔT_A . On the other hand, the rotational timescale $\tau_r = 1/D_r$ remains relatively unaffected.

In our current study, the actual temperature increment of thermally active colloid is approximately $\Delta T_A \approx 1$ K [74]. We performed simulations using $\Delta T_A = 1.25$ K and summarized in Table 3.5. It shows significantly better alignment between the simulated and

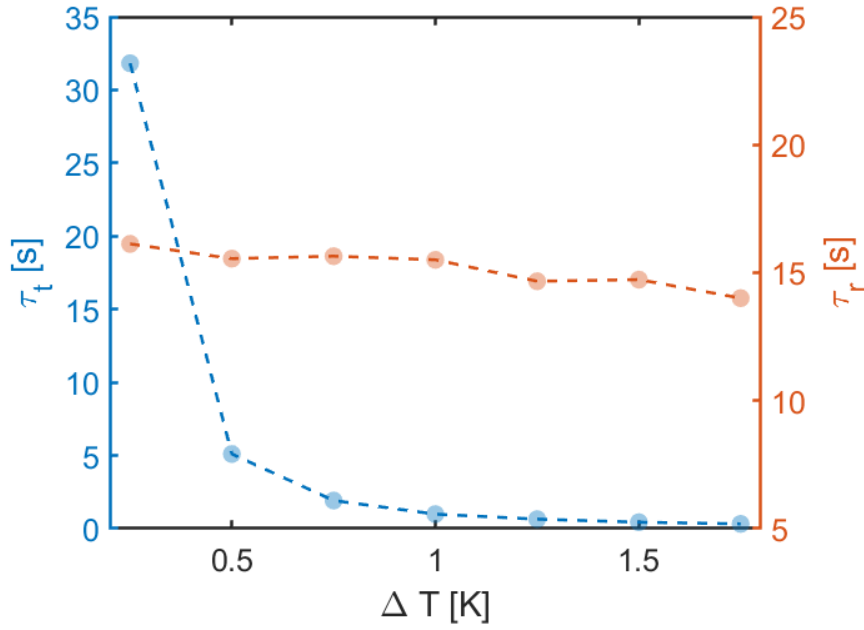


FIGURE 3.16: Simulated timescales for different surface temperature values ΔT_A of the thermally active colloid. This figure is reproduced with permission from [75].

experimental translational timescales. Since τ_r is mainly unaffected by the temperature, we focused this comparison exclusively on τ_t .

Structures	τ_t^{exp} [s]	τ_t^{sim} [s]	$\tau_t^{\text{exp}}/\tau_t^{\text{sim}}$
AD	0.61	0.65	0.94
AT1	0.29	0.39	0.73
AT2	0.26	0.21	1.24
Quadromer	0.12	0.14	0.83

TABLE 3.5: Comparison between experimental and simulated translational timescales at $\Delta T_A = 1.25$ K.

3.11 Tuning dynamics of active structures

The preceding sections demonstrated how the temperature gradient between colloids induces thermo-osmotic interactions that drive active propulsion and chiral dynamics in various colloidal assemblies. Here, we experimentally explore how the dynamics of these structures are influenced by the intensity of the incident laser light and the size of the constituent colloids.

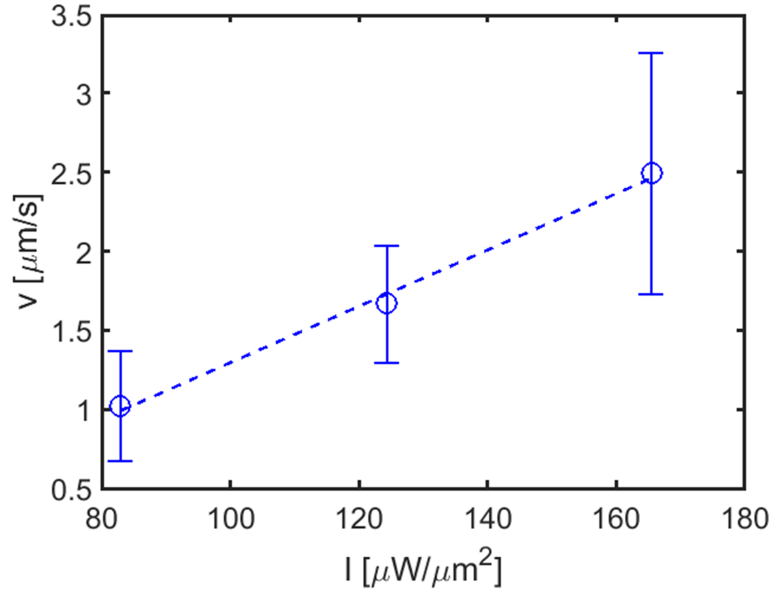


FIGURE 3.17: Linear incremental trend of the propulsion velocity (v) of active dimer structures with the intensity I of incident wide optical field. This figure is reproduced with permission from [75].

Since the thermal gradient and, thereby, the strength of the thermo-osmotic interaction scale with the optical intensity, the propulsion speed of the structures can be modulated accordingly. Fig. 3.17 illustrates the variation in the propulsion velocity of active dimer structures with respect to laser intensity. These trends are consistent with our simulation results presented in Fig. 3.18.

In addition to tuning the light intensity, we also investigated how changing the size of the colloidal components affects the dynamics. As shown in Fig. 3.19(a), the propulsion velocity of the active dimer decreases as the ratio of passive to active colloid diameters increases. In this experiment, only the size of the passive colloid was varied ($d_P = 1, 1.3, 1.6, 2 \mu\text{m}$), while the diameter of the thermally active colloid was kept constant at $d_A = 1.3 \mu\text{m}$. Since the active colloid remains unchanged, the heat generation and the associated thermo-osmotic flow remain the same. Thus, the observed variation in velocity stems solely from the change in passive colloid size. A similar decreasing trend in velocity with increasing passive colloid size was also observed in our simulations, as shown in Fig. 3.19(b). Please note that

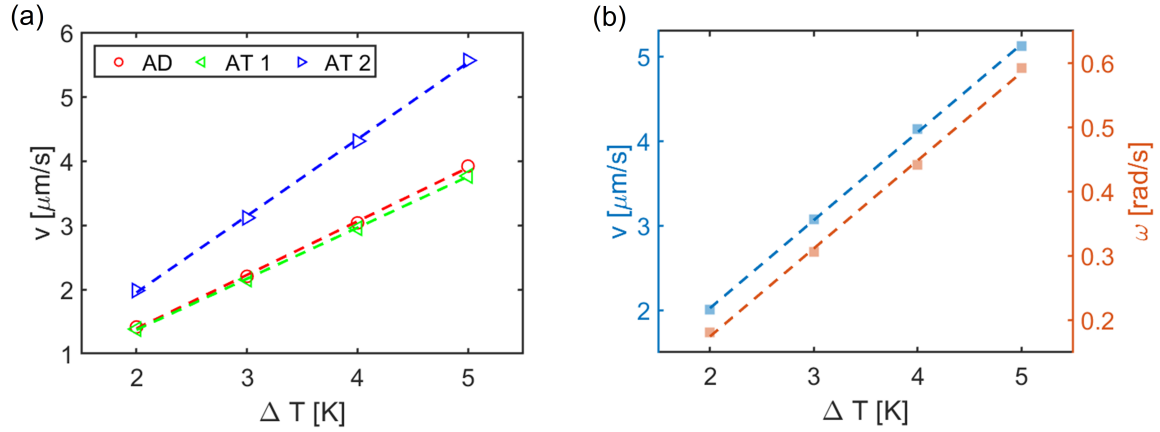


FIGURE 3.18: Simulated velocities at different temperature increment values. (a) The increasing trend of propulsion velocity (v) of the active dimer (AD) and the two trimer configurations (AT1 and AT2) as a function of surface temperature increment of thermally active colloids. (b) A similar incremental trend is also observed for both the propulsion velocity (v) and angular velocity (ω) of the active quadromer structures. The dashed lines represent linear fits to the data, highlighting the proportional relationship between the temperature gradient and the motility of the structures.

the experimental data is much more scattered compared to the simulation. Because the experiments are prone to experimental difficulties, such as: (i) Using a defocused laser beam instead of a uniform, large-area illumination. (ii) Exact control over the defocus extent of the beam. (iii) Lower experimental intensity leading to lower temperature and lower interaction. (iv) Having limited experimental realization ($\simeq 5$ sets of data) compared to more than 40 datasets in the simulation. Because of these possible reasons, the experimental data seem more scattered and have larger error bars.

3.12 Conclusion

In summary, we have experimentally investigated the dynamics of colloidal structures composed of thermally active and passive particles. Our results reveal emergent motility behaviors, such as directed propulsion and chiral motion that arise from temperature-induced non-reciprocal interactions. These behaviors vanish without such interactions, underscoring their fundamental role in driving the system's dynamics.

In contrast to previous approaches that rely on chemical fuels like hydrogen peroxide

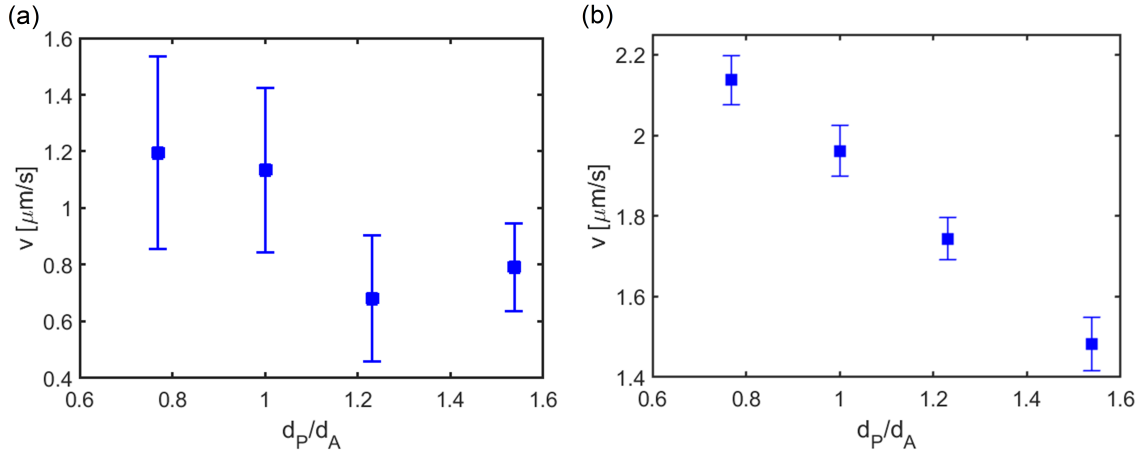


FIGURE 3.19: Effect of passive colloid's diameter (d_P) on the (a) experimental and (b) simulated propulsion velocity (v) of active dimer (AD) structures. The simulations were conducted for $\Delta T_A = 2$ K surface temperature increment of the thermally active colloids. This figure is reproduced with permission from [75].

or binary solvent mixtures such as 2,6-lutidine, our method leverages purely optothermal effects [70, 122–125]. The temperature asymmetry between colloidal components induces thermo-osmotic slip flows that mediate attractive interactions. These flows create force imbalances within the structures, leading to net propulsion and, in some cases, chirality.

We further demonstrated that the strength and nature of the dynamics can be tuned by varying the temperature difference between particles, which can be conveniently controlled through the intensity of optical illumination. To support our experimental observations, we performed numerical simulations by solving Langevin equations with the relevant physical forces, successfully capturing the key features of the observed motion. Since this propulsion mechanism is based on fluid flow generated by thermally heated colloids in water, it offers potential applications in targeted cargo delivery. More broadly, this work enhances our understanding of non-equilibrium dynamics in active colloidal systems and could inform future developments in microfluidics [128–131] and microscale engineering [145–147].

Chapter 4

Dynamics of thermally active and passive colloids under focused optical illumination

Abstract:

In this chapter, we present an experimental investigation revealing the onset of directional rotation in colloidal assemblies composed of thermally active and passive particles under optical confinement. The rotational handedness is found to be tunable by modifying the spatial arrangement of the colloidal components. Our results indicate that the angular rotation speed is influenced by both the intensity of the incident laser and the particle sizes. The origin of the observed rotational behavior lies in the asymmetric thermal distribution generated by the active colloid, which leads to an imbalance in optothermal torques within the confined system. Notably, this rotation arises without invoking spin or orbital angular momentum of light. These findings highlight the potential of optothermal manipulation for driving rotational motion in microscale systems, offering insights relevant to the development of Brownian motors and mechanisms underlying rotation in biological and ecological contexts. This chapter is adapted from the article "ACS Photonics 10(11), 4006-4013 (2023), (arXiv:2309.12740) "[[140](#)].

4.1 Introduction and motivation

Nonequilibrium systems are pervasive in nature and often exhibit a wide range of intriguing behaviors [148–151]. These behaviors include dynamic self-assembly [152–154], spontaneous pattern formation [90, 92, 93], and collective motion-manifesting in both translational and rotational modes [87, 155–158]. These collective phenomena arise from inter-element interactions and have inspired a range of experimental studies aiming to recreate similar behaviors in controlled laboratory settings. Investigating such emergent dynamics using synthetic systems lies at the heart of active matter research [76, 78, 159, 160].

In this regard, colloidal systems have become powerful model platforms for exploring active matter dynamics. These systems allow for detailed studies of inter-particle interactions and the role of environmental stimuli in driving emergent behavior. On one hand, a wide array of active colloidal particles has been designed and explored [70, 88, 97, 136, 161, 162]. On the other hand, multiple external control mechanisms, including chemical [163–166], electric [167–169], magnetic [113, 114, 170–173], and optical fields [3, 57, 174–183], have been developed to induce translation and rotation in soft systems.

In the context of the present chapter, we focus on rotational motion. Such rotational dynamics have been observed in diverse biological systems, including motile microorganisms and bacterial colonies [154–156, 184, 185]. Reproducing similar rotational behaviors using synthetic colloidal assemblies remains a compelling yet challenging goal [98, 161, 186, 187].

Among the techniques available, optical fields provide interesting avenues due to their ability to both confine and manipulate soft matter systems [188, 189]. Conventional approaches to induce rotational motion involve leveraging either the spin or orbital angular momentum of light [59, 190–192]. However, an important question remains: can one generate rotational degrees of freedom in an active colloidal system without relying on the angular momentum of light? If so, it would offer a complementary route to control soft matter systems.

One promising mechanism is through optothermal effects [193], where light-induced

localized heating gives rise to temperature gradients that break symmetry and induce rotation [98, 186, 187, 194, 195]. To utilize this strategy effectively, it is essential to identify the material properties of colloids and understand the characteristics of the optothermal fields responsible for driving such motion. Developing controlled, directional rotation in artificial active systems has thus emerged as a significant endeavor, both for fundamental insights and for applications in biological mimicry and microscale technologies [70, 166, 194].

Motivated by these ideas, here we report experimental results demonstrating the emergence of directional rotation in colloidal assemblies composed of thermally active (A) and passive (P) particles under illumination with a 532 nm laser. Fig. 4.1(a) illustrates a representative schematic of a rotating trimer configuration consisting of two passive colloids (P1 and P2) and one thermally active colloid (A). While P1 and P2 are nearly identical in composition and size, their relative spatial positions within the optical field lead to distinct dynamical behaviors. The direction of rotation (i.e., handedness) is governed by the placement of the active colloid A within the structure, as highlighted in Fig. 4.1(b) and 4.1(c). In addition to demonstrating the emergence of rotational dynamics, we explore how such motion can be tuned by varying the types of constituent colloids and the intensity of the applied optical field. In the following sections, we first describe the experimental framework used to generate these dynamics and then present a detailed investigation of the resulting directional rotation.

4.2 Experimental implementation and methods

Similar to chapter 3, in these experiments, we employed two species of colloidal particles: (i) passive colloids (denoted as P) and (ii) thermally active colloids (denoted as A). The passive species included melamine formaldehyde (MF) particles with a diameter of 2.0 μm and polystyrene (PS) particles of diameter 1.3 μm . The active colloids were iron oxide-doped PS particles of the same size (1.3 μm), procured from Microparticles GmbH. The embedded iron oxide nanoparticles enable these colloids to absorb light efficiently and generate

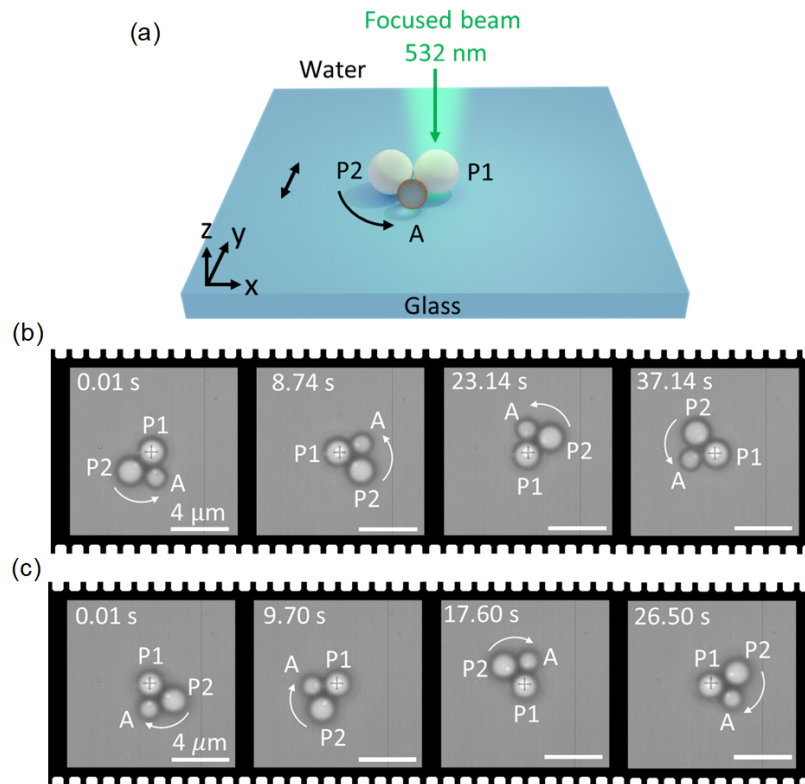


FIGURE 4.1: Rotational dynamics of a trimer structure in a focused optical field. (a) Schematic representation of the experiment. A trimer, comprising two passive melamine formaldehyde colloids (P1 and P2, diameter 2.0 μm) and one thermally active polystyrene colloid (A, diameter 1.3 μm), is within the vicinity of a focused Gaussian laser beam (wavelength 532 nm) that is linearly polarized along the y -axis, as indicated by the double-headed black arrow. The observed rotational direction is depicted with a curved black arrow. (b) and (c) show time-lapse images of two trimer structures exhibiting opposite rotational handedness under an optical intensity of 5.7 $\text{mW}/\mu\text{m}^2$. The grey '+' symbol denotes the center of the optical beam. This figure is reproduced from [140].

localized thermal fields under laser illumination.

Both active and passive colloids were suspended in deionized water. A 10 μL drop of the dispersion was placed in a sample chamber approximately 100 μm in height, constructed by spacing two glass coverslips. The prepared sample was mounted on a custom-built dual-channel optical microscope. A linearly polarized Gaussian laser beam (wavelength 532 nm) was focused at the lower interface of the chamber using a 100 $\times$, 0.95 NA objective. The resulting light signal was collected with a 100 $\times$, 1.49 NA oil immersion objective and imaged using a high-speed camera at 100 frames per second. Particle tracking was carried out using the TrackMate plugin in Fiji [138].

Fig. 4.2 presents a false-color plot of the two-dimensional experimental intensity distribution of the laser beam at the sample plane. The intensity profiles along the x and y directions can be fitted using a Gaussian function, $I(x) = I_0 \exp(-x^2/w_0^2)$, yielding an average beam waist radius of $w_0 = 0.80 \mu\text{m}$.

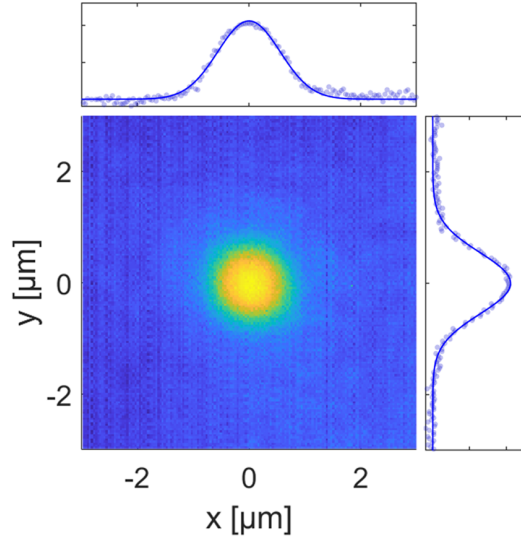


FIGURE 4.2: False-color representation of the two-dimensional experimental laser beam intensity distribution, accompanied by line profiles along the x and y axes. This figure is reproduced from [140].

4.3 Dynamics of individual colloids under focused optical field

While illuminated with a tightly focused laser beam, micron-sized colloidal particles experience a restoring optical gradient force that confines them near the beam center. As illustrated in Fig. 4.3, the stiffness of the optical trap exhibits a linear dependence on the laser intensity I_0 , and this behavior can be modeled using the following linear relation:

$$k(I_0) = A \cdot I_0 + B \quad (4.1)$$

The coefficients A and B in equation (4.1) were extracted by fitting experimental data for different colloidal types, as summarized in Table 4.1. The nonzero value of B indicates

the counterintuitive phenomenon of non-zero trap stiffness even at zero intensity. This might be due to the trapping configuration used, as we trapped the colloids near the bottom cover slip. The colloids are being pushed towards the substrate due to the forward scattering force, which might lead to additional help to trap the colloids in addition to the gradient optical force. Which might result in the nonzero B parameter. Besides, this non-zero B is significant at a lower intensity. For the higher intensity regime, the offset parameter B is negligibly small compared to the intensity-dependent part AI_0 , which leads the equation to $k(I_0) = AI_0$.

Colloid Type	Diameter [μm]	A [fs]	B [pN/ μm]
MF P	2.0	0.37	0.08
MF P	1.3	0.85	0.17
PS P	1.3	0.83	0.34
PS A	1.3	2.05	0.25

TABLE 4.1: Fitting parameters corresponding to the linear dependence of trap stiffness on laser intensity for various passive and thermally active colloids, as per equation (4.1).

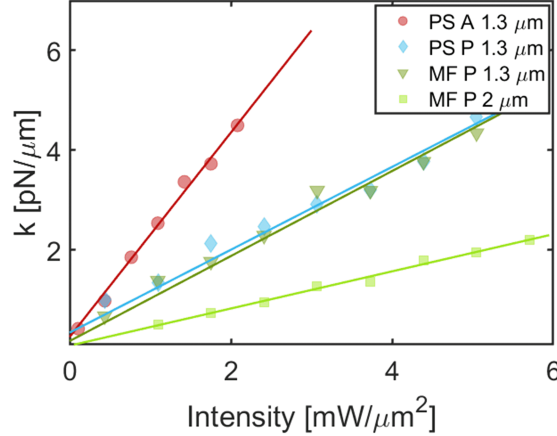


FIGURE 4.3: Linear increase in trap stiffness (k) with laser intensity of Gaussian laser beam. This figure is reproduced from [140].

From the data shown in Fig. 4.3, it is evident that thermally active PS colloids exhibit a significantly higher trap stiffness at any given laser intensity compared to the other colloidal types. This enhanced stiffness likely arises due to their increased polarizability and stronger interaction with the optical field. The trap stiffness is dependent on a lot of factors, such as surface charge, functionalization, and effective material refractive index, which might lead

to higher in-plane trap stiffness of thermally active colloids compared to the passive colloids (both MF and PS).

4.4 Dynamics of multiple colloids under focused illumination

To investigate the structural prerequisites for the onset of rotational dynamics, we systematically examined four distinct colloidal configurations under optical confinement, as shown in Fig. 4.4(a)-4.4(d) (also see video 7 [141]). These configurations include: (a) a dimer of two passive colloids (P1 and P2), (b) a mixed dimer comprising one passive (P1) and one thermally active colloid (A), (c) a trimer formed by three passive colloids (P1, P2, P3), and (d) a trimer with two passive colloids (P1 and P2) and one thermally active colloid (A). Among these, only the structure depicted in Fig. 4.4(d) displays rotational behavior.

In Fig. 4.4(a) and 4.4(b), the colloidal pairs are radially confined to the beam center by optical gradient forces and do not exhibit any rotational dynamics. When an additional passive polystyrene colloid (P3) with diameter $d_{P3} = 1.3 \mu\text{m}$ is introduced to the configuration in Fig. 4.4(a), a trimer structure emerges, as seen in Fig. 4.4(c). This structure, held together by optical gradient forces, remains non-rotating and is therefore referred to as a passive trimer (PT).

By substituting P3 with a thermally active colloid of the same size (A), we obtain the configuration shown in Fig. 4.4(d), which exhibits clear rotational dynamics designated as an active trimer (AT). The inset shows the probability distribution of P1's position, indicating that the structure rotates around a common center. The angular velocity of this motion is extracted from the differential cross-correlation function (*dccf*) computed using the (x, y) coordinates of the individual colloids:

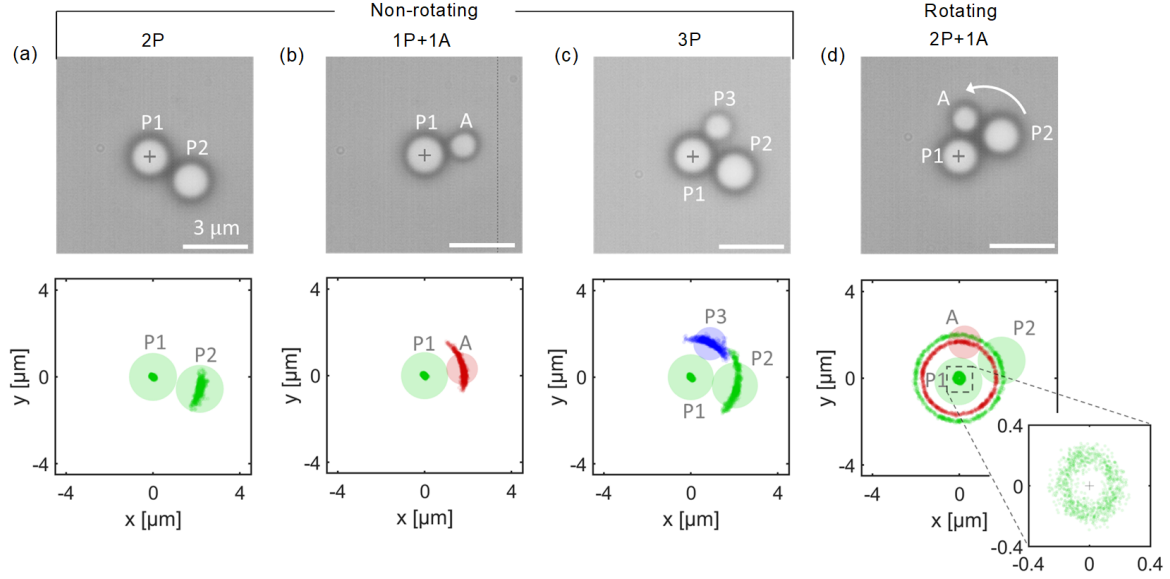


FIGURE 4.4: Dynamics of colloidal assemblies composed of passive and thermally active particles under focused laser illumination of $5.7 \text{ mW}/\mu\text{m}^2$. No rotational behavior is observed in (a) a dimer composed of two passive melamine formaldehyde (MF) colloids (P1, P2), and (b) a dimer consisting of one passive MF colloid (P1) and one thermally active polystyrene (PS) colloid (A). (c) A passive trimer formed by two MF colloids (P1, P2) and one passive PS colloid (P3) also lacks rotational motion. In contrast, (d) an active trimer comprised of two passive MF colloids (P1, P2) and a thermally active PS colloid (A) demonstrates clear counterclockwise rotation. The inset displays a zoomed-in spatial distribution of the central passive colloid (P1), which remains centered at the optical trap location. The diameters of the colloids used are $d_{P1} = d_{P2} = 2.0 \mu\text{m}$, and $d_{P3} = d_A = 1.3 \mu\text{m}$. All positional distributions are measured over a duration of 50 seconds. This figure is reproduced from [140].

$$\begin{aligned} dccf(\delta t) &= \frac{\langle x(t)y(t + \delta t) \rangle - \langle y(t)x(t + \delta t) \rangle}{\sqrt{\langle x^2(t) \rangle \langle y^2(t) \rangle}}, \\ &= Ae^{-\delta t / \delta t_{\text{ot}}} \sin(\omega \delta t), \end{aligned} \quad (4.2)$$

where δt is the lag time, ω denotes the angular velocity, and δt_{ot} is the characteristic relaxation time of the optical trap [186]. The calculated angular velocity for the AT structure in Fig. 4.4(d) is $\omega = 0.60 \text{ rad/s}$.

4.4.1 Estimation of Experimental Torque

To quantify the torque generated in a rotating active trimer (AT), we evaluate the torques' vector sum on each constituent colloid. This is given by $\vec{\tau} = \sum_i \vec{r}_i \times \vec{F}_i^{\text{d}} = \sum_i \vec{r}_i \times (\gamma_i \vec{v}_i)$,

where $\vec{r}_i$ represents the position vector of colloid i , $\vec{v}_i$ its instantaneous velocity, γ_i the friction coefficient, and $\vec{F}_i^d$ the thermo-osmotic flow induced drag force acting on it. The index i runs over all three colloids in the structure: P1, P2, and A.

Assuming rigid body rotation and using the relation between angular momentum and moment of inertia (MoI), $I_i = L_i/\omega$, we can reformulate the torque magnitude as:

$$|\tau| = \sum_i \frac{\gamma_i}{m_i} I_i \omega, \quad (4.3)$$

where m_i denotes the mass of colloid i , L_i its angular momentum, and ω the angular velocity of the rotating trimer.

To determine I_i , we invoke the parallel axis theorem, which gives:

$$I_i = \frac{2}{5} m_i a_i^2 + m_i r_i^2, \quad (4.4)$$

where a_i is the radius of colloid i , and r_i is its radial distance from the center of rotation.

Substituting this into equation (4.3), the mass terms cancel out, leading to a simplified expression:

$$\tau = \sum_i \gamma_i \omega \left(\frac{2}{5} a_i^2 + r_i^2 \right). \quad (4.5)$$

This expression reveals that the net torque acting on the active trimer is independent of the individual masses of the colloids and is governed instead by their geometric and dynamic, specifically, their sizes, radial positions, friction coefficients, and the overall angular velocity of the structure. The AT configuration's estimated torque is $|\tau| \simeq 1.7 \times 10^{-19}$ Nm.

Although the PT and AT structures (Figures 4.4(c) and 4.4(d), respectively) are nearly identical in geometry, only the AT configuration exhibits rotational motion. This highlights the critical role of the thermally active colloid and the accompanying optothermal field in breaking symmetry and driving rotation. To understand this, we have studied the associated thermal field profile, which will be discussed in the next section.

4.5 Estimation of thermal field

4.5.1 Numerically estimated temperature field

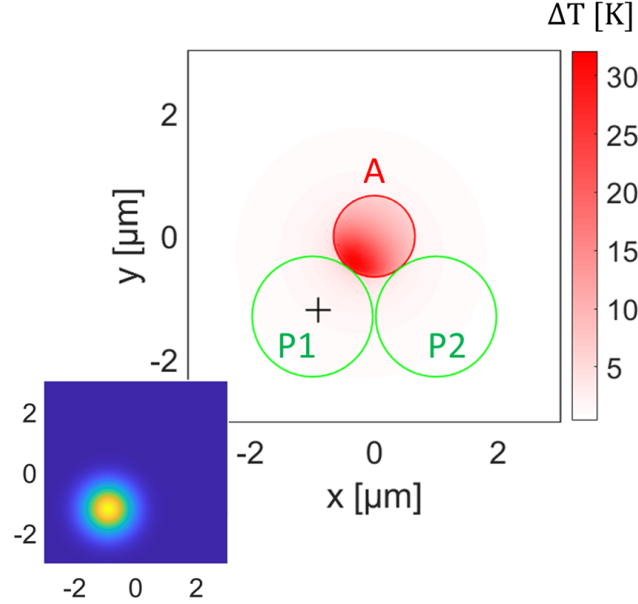


FIGURE 4.5: Numerically calculated temperature profile in an AT structure under $5.7 \text{ mW}/\mu\text{m}^2$ illumination, with beam center marked by '+'. Inset: corresponding laser intensity profile. This figure is reproduced from [140].

In an AT configuration, the thermally active PS colloid (A) is positioned off-center with respect to the laser beam, such that it lies within the Gaussian tail of the optical field. This leads to a localized heating of colloid A, as it absorbs light and dissipates the energy in heat. To numerically capture the resulting spatial temperature distribution, we model the colloid as containing approximately $N \simeq 37000$ point-like iron-oxide nanoparticles, uniformly distributed within its volume. The laser beam is focused on colloid P1, simulating the experimental configuration shown in Fig. 4.5.

Each nanoparticle acts as a localized heat source, and the temperature increment ΔT at any spatial location is computed using the relation [19]:

$$\Delta T = \sum_N \frac{I(\vec{r}) \sigma_{\text{abs}}}{4\pi\kappa r}, \quad (4.6)$$

where $I(\vec{r})$ denotes the local laser intensity at the position of the nanoparticle, $\sigma_{\text{abs}} \simeq 9.5 \cdot 10^{-18} \text{ m}^2$ is the absorption cross-section at 532 nm, κ is the thermal conductivity of the surrounding medium, and r is the distance from the nanoparticle to the point where the temperature is being evaluated.

The intensity profile of the laser beam used in the simulation was modeled based on experimentally obtained parameters, with a beam waist $w_0 = 0.80 \text{ }\mu\text{m}$. To ensure consistency with experimental data, the computed temperature distribution ΔT was scaled by a constant factor such that the average surface temperature of colloid A matched that obtained from experimental estimates. As shown in Fig. 4.5, the resulting temperature profile exhibits a strong asymmetry, with the side of colloid A closer to the beam center attaining higher temperatures. This asymmetric heating is critical to generating directional thermo-osmotic flows that drive the observed rotational dynamics in AT structures.

4.5.2 Experimental Estimation of Temperature

The experimental surface temperature of the thermally active colloid is estimated in two sequential steps:

Step 1: Surface Temperature of a Centrally Heated Colloid A

To determine the maximum surface temperature of colloid A under focused illumination, we utilize the nematic-to-isotropic phase transition of the liquid crystal 5CB, which occurs above a phase transition temperature $T_{\text{pt}} = 35^\circ\text{C}$ [32]. Upon illumination with a focused 532 nm laser, colloid A heats its surroundings, setting up a radially decaying temperature profile $\Delta T(r) \sim r^{-1}$.

As illustrated in Fig. 4.6(a), the isotropic phase ring appears where the local temperature exceeds T_{pt} . This transition is visible in bright-field imaging due to the difference in refractive indices between the two phases, as seen in Figure 4.6(b). Given that the temperature profile follows $\Delta T(r) = \Delta T_0 \cdot a/r$, where a is the colloid radius, the maximum surface temperature ΔT_0^{5CB} in 5CB is given by:

$$\Delta T_0^{5CB} = (T_{pt} - 298 \text{ K}) \cdot \frac{R - a}{a} \quad (4.7)$$

To convert this to the corresponding value in water, we account for the differing thermal conductivities using:

$$\Delta T_0^{\text{water}} = \Delta T_0^{5CB} \cdot \frac{\kappa_{5CB}}{\kappa_{\text{water}}} \quad (4.8)$$

where $\kappa_{\text{water}} = 0.6 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and $\kappa_{5CB} = 0.15 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. As shown in Fig. 4.6(c), both the phase boundary radius R and the corresponding surface temperature increase linearly with laser intensity I_0 (in $\text{mW}/\mu\text{m}^2$), resulting in the relation:

$$\Delta T_0^{\text{water}} = 9.27 \cdot I_0 - 0.78 \quad (4.9)$$

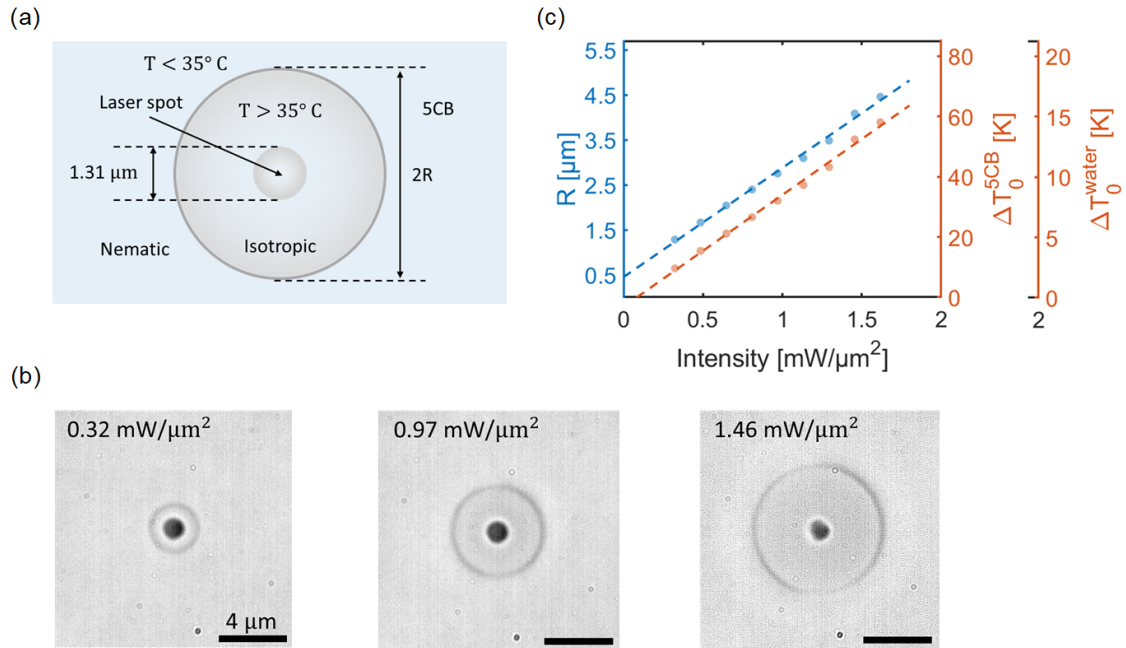


FIGURE 4.6: Measurement of maximum surface temperature of centrally heated thermally active colloid (A) using 5CB. (a) Schematic of colloid A under focused illumination showing induced isotropic phase region. (b) Bright-field images of the phase transition boundary at different laser intensities. (c) Linear increase in phase boundary radius R and surface temperature ΔT_0 with laser intensity. This figure is reproduced from [140].

Step 2: Surface Temperature of an Off-Centered Colloid A

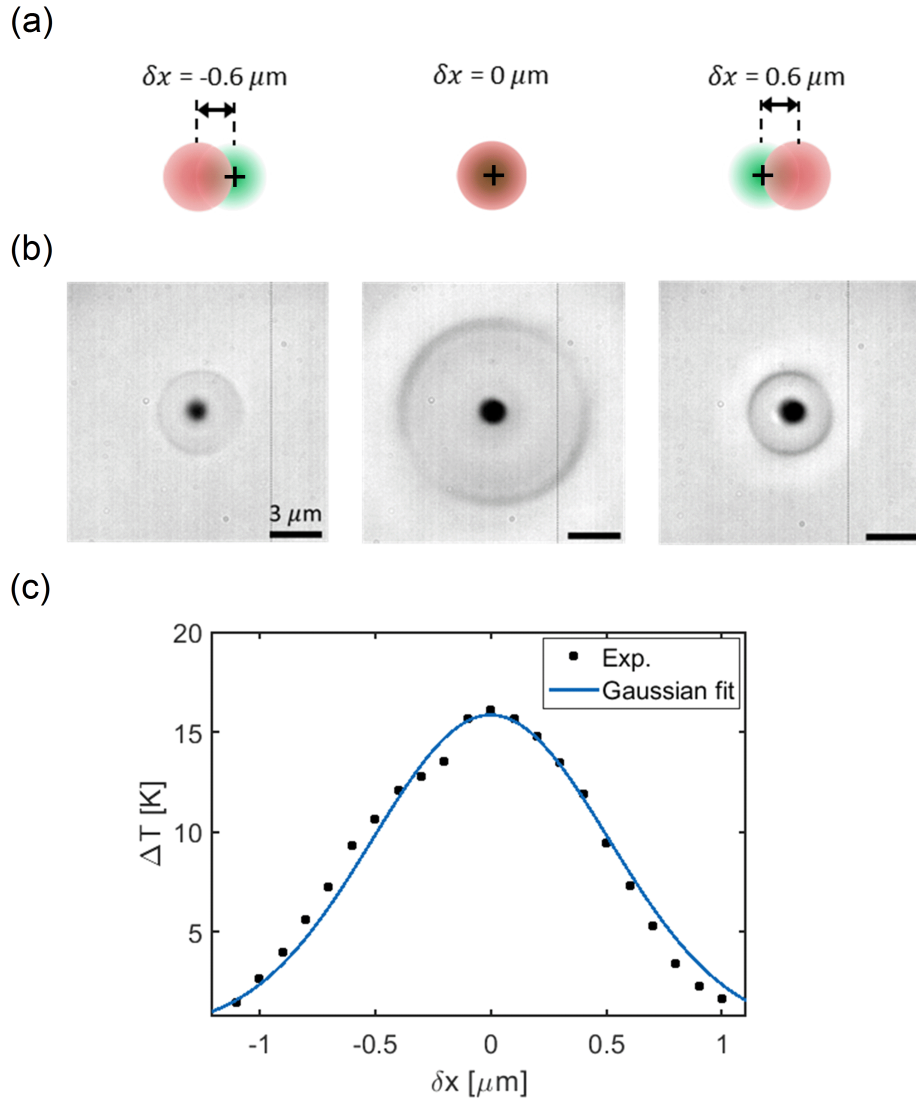


FIGURE 4.7: Estimation of average surface temperature of colloid A at off-center positions. (a) Schematic of displaced colloidal particles along the x -axis from the beam center. (b) Bright-field images showing asymmetric isotropic rings with varying displacements. (c) Gaussian fit of surface temperature versus x -axis displacement. This figure is reproduced from [140].

In the AT structure, colloid A is located at an off-center position with respect to the beam, thus experiencing heating from the Gaussian tail of the optical field. To experimentally estimate the average surface temperature under such conditions, a colloid A in 5CB medium is scanned across the beam along both x and y axes in $0.1 \mu\text{m}$ steps at a laser intensity of $2.50 \text{ mW}/\mu\text{m}^2$ (Fig. 4.7(a)). The resulting asymmetric isotropic phase rings are captured via bright-field imaging (Fig. 4.7(b)).

The average surface temperature at each position is calculated from the radius of the

isotropic phase boundary, similar to Step 1. Since the laser intensity follows a Gaussian profile, the surface temperature also exhibits a Gaussian dependence on lateral displacement δx :

$$\Delta T(\delta x) = \Delta T_0(I_0) \cdot \exp\left(-\frac{\delta x^2}{w_0^2}\right) \quad (4.10)$$

Here, $\Delta T_0(I_0)$ is the maximum surface temperature increment from Eq.4.10, δx is the lateral distance from the beam center, and w_0 is the beam waist of the laser. Fig. 4.7(c) shows the experimentally obtained temperature profile, well approximated by a Gaussian function.

4.6 Optical and thermo-osmotic forces acting on the AT structures

We qualitatively studied the relevant forces acting on constituent colloids to elucidate the mechanism behind the rotational motion observed in the active trimer (AT) structure. Without the peripheral colloids P2 and A, the optical gradient force confines P1 to the laser beam center. However, when P2 and A are present, the optical gradient forces acting on them are denoted by $\vec{F}_{O,P2}$ and $\vec{F}_{O,A}$ (green arrows) displace P1 from the beam center (Fig.4.8(a)).

The optical gradient forces acting on a colloid i ($i = P1, P2, A$), indicated by green arrows in Fig. 4.8(a), are acting radially inward, i.e., $\vec{F}_{O,i} \parallel \vec{r}_i$. As a result, the net torque contribution of these forces is zero, since $\vec{\tau} = \sum_i \vec{r}_i \times \vec{F}_{O,i} = 0$. This explains the absence of rotation in the passive trimer (PT) due to solely optical forces. The strength of the optical trap on P1 can be estimated using $F_{O,P1} = k(I_0) \cdot r_c \simeq 0.36$ pN, where $k(I_0)$ is the trap stiffness at laser intensity I_0 and r_c is the radial distance from the beam center.

In addition to optical forces, thermo-osmotic forces arise due to the thermal field generated by the thermally active colloid A. These thermo-osmotic forces $\vec{F}_{T,i}$, indicated by the violet arrows in Fig. 4.8(b)). These $\vec{F}_{T,i}$ are not necessarily aligned radially. The asymmetric

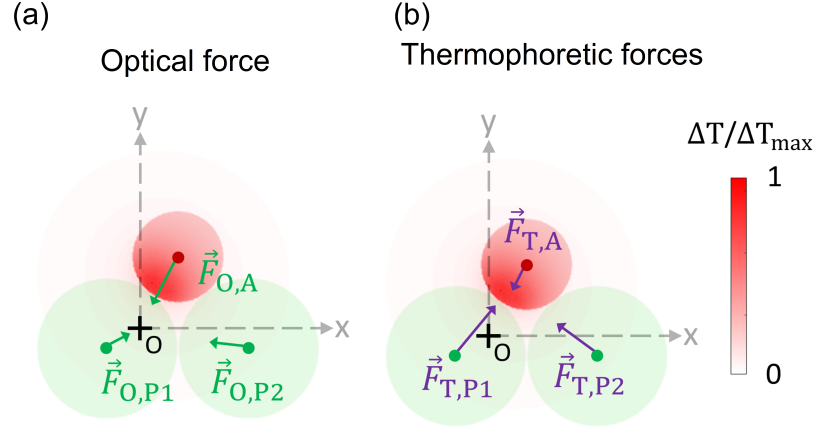


FIGURE 4.8: Optothermal interactions forces in an active trimer (AT) structure. (a) and (b) illustrate schematic representations of the optical gradient forces and thermo-osmotic forces acting within the AT configuration, with the laser beam center denoted by '+'. This figure is reproduced from [140].

positioning of A results in non-uniform temperature distribution that gives rise to unequal thermo-osmotic forces on P1 and P2 and also a non-zero thermo-osmotic force $\vec{F}_{T,A}$ on colloid A. Experimentally probing the non-uniform heating of these thermally active colloids is very difficult. We considered the thermally active colloids to have an average uniform surface temperature approximated by equation 4.10.

With such approximation, only non-zero thermo-osmotic forces are acting on P1 and P2, which can be estimated using equation (3.9) as:

$$F_{T,i} \simeq \gamma \frac{\chi}{T_s} \nabla T \simeq 0.04 \text{ pN},$$

where γ is the drag coefficient of the colloids and χ is the thermo-osmotic slip coefficient of the system. These forces are approximately directed from the passive colloids (P1 and P2) to the thermally active colloid A.

4.6.1 Thermo-osmotic forces leading to rotation of the colloidal structures

To understand the contribution of these thermo-osmotic forces to the rotational state of the AT structures, we first estimate the thermo-osmotic forces in vectorial form as:

$$\vec{F}_{T,P1} = |F_{T,P1}| \hat{D}_1 = |F_{T,P1}| \frac{\vec{D}_1}{|\vec{D}_1|}, \quad (4.11a)$$

$$\vec{F}_{T,P2} = |F_{T,P2}| \hat{D}_2 = |F_{T,P2}| \frac{\vec{D}_2}{|\vec{D}_2|}, \quad (4.11b)$$

where $\hat{D}_1$ and $\hat{D}_2$, are the direction of the thermo-osmotic forces on P1 and P2, as illustrated in Fig. 4.9(a) (represented by violet arrows). These directions can be obtained as $\vec{D}_1 = \vec{r}_A - \vec{r}_{P1}$ and $\vec{D}_2 = \vec{r}_A - \vec{r}_{P2}$, where $\vec{r}_i$ denotes the position vector of colloid i . Now we can decompose these thermo-osmotic forces into radial ($F_{T,i}^r$) and transverse ($F_{T,i}^t$) components:

$$F_{T,i}^r = \vec{F}_{T,i} \cdot \hat{r}_i, \quad F_{T,i}^t = \vec{F}_{T,i} \cdot \hat{t}_i, \quad (4.12)$$

where $\hat{r}_i$ and $\hat{t}_i$ are unit vectors in the radial and transverse directions, respectively, for colloid i (with $i = P1, P2$) that can be expressed in terms of the Cartesian coordinates (x_i, y_i) of the colloids as:

$$\hat{r}_i = \frac{x_i \hat{x} + y_i \hat{y}}{r_i}, \quad \hat{t}_i = \frac{-y_i \hat{x} + x_i \hat{y}}{r_i}, \quad (4.13)$$

where r_i is the distance of the i -th colloid from the center of the optical beam.

Only the transverse components of the thermo-osmotic forces contribute to the rotational state via a net torque:

$$\vec{\tau} = \sum_i \vec{r}_i \times \vec{F}_{T,i} = \sum_i r_i \cdot F_{T,i}^t. \quad (4.14)$$

For our case, we obtain $F_{T,P1}^t \simeq -0.02$ pN and $F_{T,P2}^t \simeq 0.03$ pN. The signs reflect the force direction with respect to the local transverse direction. Such as from the schematic, we

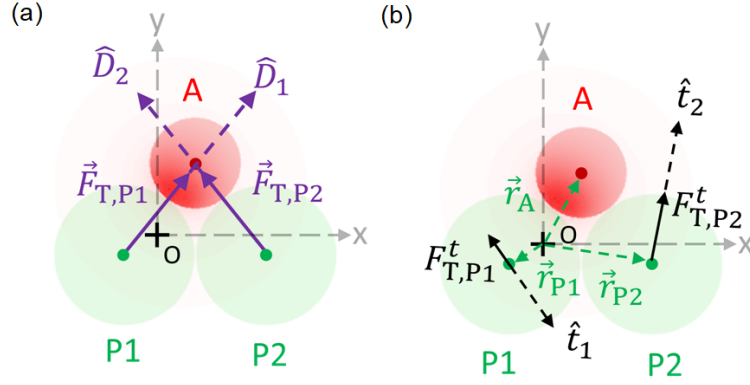


FIGURE 4.9: Schematic (a) thermo-osmotic force directions and (b) their transverse components under the approximation of uniform surface temperature of the thermally active colloid. This figure is reproduced from [140].

found that $F_{T,P1}^t$ acts in the direction opposite to $\hat{t}_{P1}$, whereas $F_{T,P2}^t$ aligns with $\hat{t}_{P2}$. From Fig. 4.9(b), we can see that $F_{T,P1}^t$ on the P1 tends to rotate the structure in a clockwise direction, whereas $F_{T,P2}^t$ on P2 tends to rotate the structures in a counterclockwise direction. Although the magnitudes of the tangential forces are similar, the larger radial distance of P2 ($r_{P2} > r_{P1}$) results in different magnitude of the corresponding torques as $\tau_{P1} \simeq -0.03 \times 10^{-19}$ Nm and $\tau_{P2} \simeq 0.62 \times 10^{-19}$ Nm, respectively. As a result, a net counterclockwise torque imbalance leads to directional rotation of the AT structures.

4.7 Different rotational states of the AT structures

From the previous subsection, we found that the observed rotation in AT originates from optothermal interactions, leading to the torque imbalance. Thus, adjusting the relative position of colloid A enables control over the rotation direction (see video 8 [141]). Specifically, if A is placed to the left of P2 (Fig. 4.10(a)), the AT rotates counterclockwise, and when placed on the right (Fig. 4.10(b)), it rotates clockwise. Directional control is also achievable by switching the laser focus from one passive colloid to another (see video 9 [141]). The angular velocities corresponding to Fig. 4.10(a) and Fig. 4.10(b) are $\omega = 0.60$ rad/s and $\omega = 0.58$ rad/s, respectively.

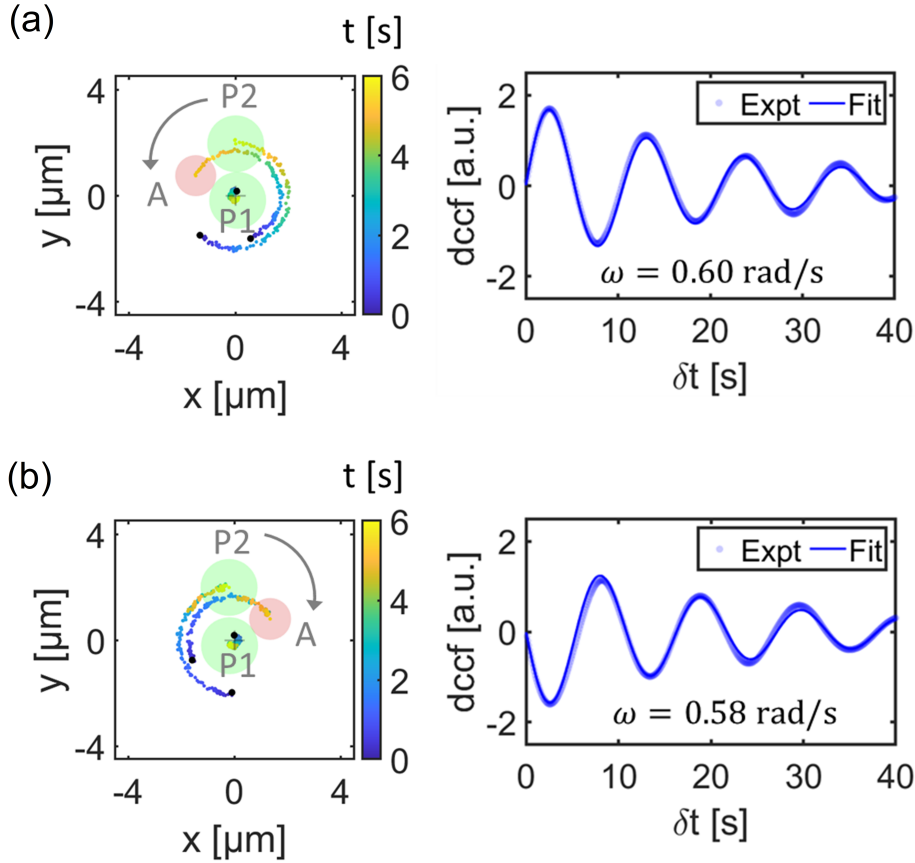


FIGURE 4.10: Different directional rotational state of active trimer structures. (a) and (b) display schematic trajectories of two AT configurations exhibiting counterclockwise and clockwise rotation, respectively, along with their differential cross correlation functions (*dccfs*'s). The angular velocities extracted at a laser intensity of $5.7 \text{ mW}/\mu\text{m}^2$ are $\omega = 0.60 \text{ rad/s}$ and $\omega = 0.58 \text{ rad/s}$ for the two oppositely rotating ATs. This figure is reproduced from [140].

4.8 Controlling the rotational dynamics

While the direction of rotation of the AT structure can be modulated by altering the spatial arrangement of its constituent colloids, the angular velocity can be independently tuned by varying the intensity of the incident laser beam. As shown in Fig. 4.11(a), both the rotational speed and the torque generated within the AT exhibit an approximately linear increase with increasing laser intensity. This trend is primarily attributed to the linear rise in the average surface temperature of the thermally active colloid A with laser intensity, which in turn enhances the magnitude of thermo-osmotic forces (Fig. 4.11(b)). The resultant enhanced slip flow and torque lead to faster rotation.

Beyond laser intensity, the rotational behavior of the AT structure can also be tuned by

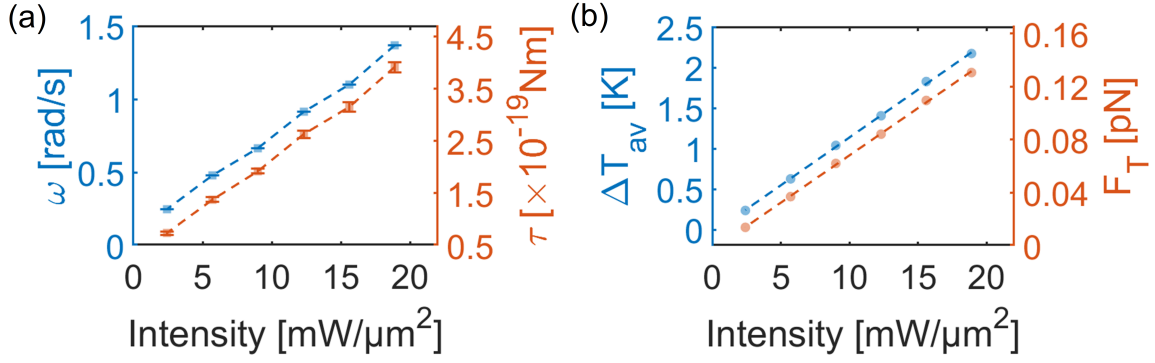


FIGURE 4.11: Tuning the rotational behavior of the AT structure via laser intensity. (a) The angular velocity of rotation (ω) and the associated torque (τ) exhibit an increasing trend with rising laser intensity. (b) Both the average surface temperature increase (ΔT_{av}) of the thermally active colloid, as computed using equation (4.10), and the thermo-osmotic force (F_T), scale linearly with laser intensity. This figure is reproduced from [140].

altering the diameters of the passive colloids (P1 and P2). Variations in colloid size do not affect the thermal field. Instead, it alters the distance of the passive colloids from the heat sources, leading to a change in thermo-osmotic drag force, thereby modulating the dynamics of rotation. To investigate the effect, we examined two AT configurations with different passive colloid diameters, as shown in Fig. 4.12(a) and Fig. 4.12(b). In these structures, P1 and P2 have diameters of $d_{P1} = d_{P2} = 2.0 \mu\text{m}$ and $d_{P1} = d_{P2} = 1.3 \mu\text{m}$ respectively, while the thermally active colloid A is maintained at $d_A = 1.3 \mu\text{m}$ in both cases.

In these configurations, the estimated thermo-osmotic forces acting on the colloids are $F_T \simeq 0.08 \text{ pN}$ and 0.13 pN for the larger and smaller trimers. This leads to the corresponding torque imbalances of $\tau = 1.2 \times 10^{-19} \text{ Nm}$ and $1.45 \times 10^{-19} \text{ Nm}$. Using these values from equation (4.5), we can obtain the corresponding angular velocities to be $\omega \simeq 0.43 \text{ rad/s}$ for the larger-diameter case and $\omega \simeq 1.2 \text{ rad/s}$ for the smaller-diameter case. These predictions qualitatively match the experimental measurements, where the larger trimer exhibited a slower rotation rate of $\omega \simeq 0.92 \text{ rad/s}$, compared to $\omega \simeq 1.30 \text{ rad/s}$ for the smaller trimer.

The observed discrepancies in the magnitude of the angular velocities, particularly in the case of the larger trimer, arise from several simplifying assumptions in our estimation. Notably, hydrodynamic interactions between colloids, which are known to play a role in such

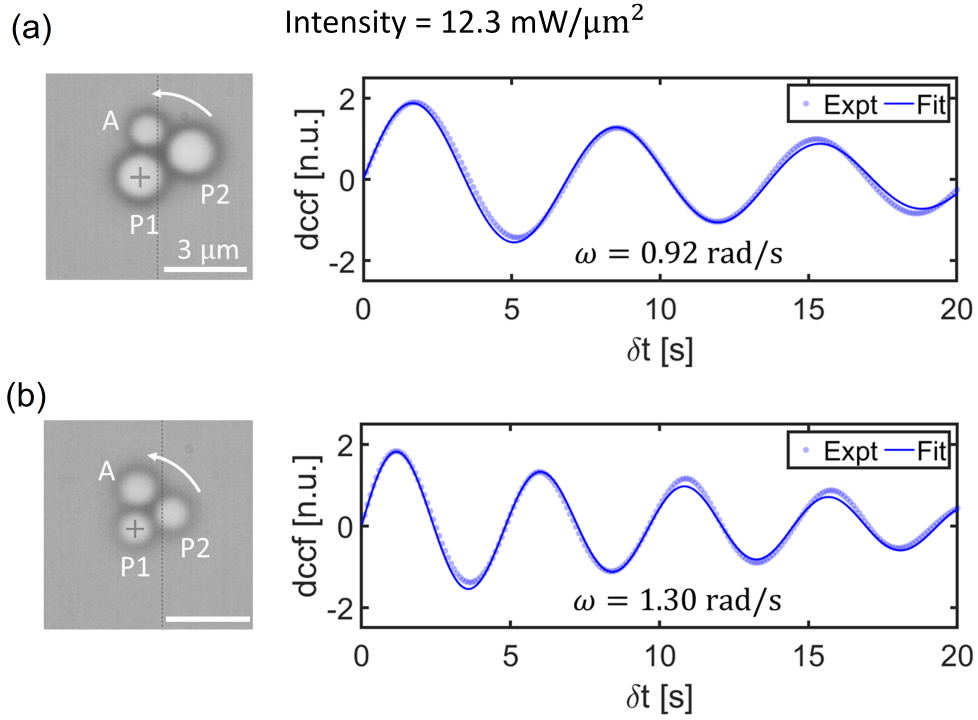


FIGURE 4.12: Tuning the rotational behavior of the AT structure via the size of passive colloids. (a) and (b) show two AT configurations with different passive colloid sizes: $d_{P1} = d_{P2} = 2.0 \mu\text{m}$ in (a), and $d_{P1} = d_{P2} = 1.3 \mu\text{m}$ in (b), while the active colloid remains fixed at $d_A = 1.3 \mu\text{m}$. The corresponding differential cross-correlation functions (dccfs) for each structure are also shown. This figure is reproduced from [140].

systems, were not considered. Additionally, the friction coefficients γ_i used in torque calculations were derived from measurements of freely diffusing colloids near a glass surface, which is significantly influenced by nearby surfaces or neighboring colloids. Incorporating these factors is expected to yield improved agreement between theoretical predictions and experimental results.

4.9 Investigation of structural dynamics of colloidal structures

To understand the mechanical stability of the colloidal assemblies, we examine the interparticle spacing in both passive and active trimer configurations. Fig. 4.13(a) and (b) present the distribution of distances among colloids in passive trimer (PT) and active trimer (AT)

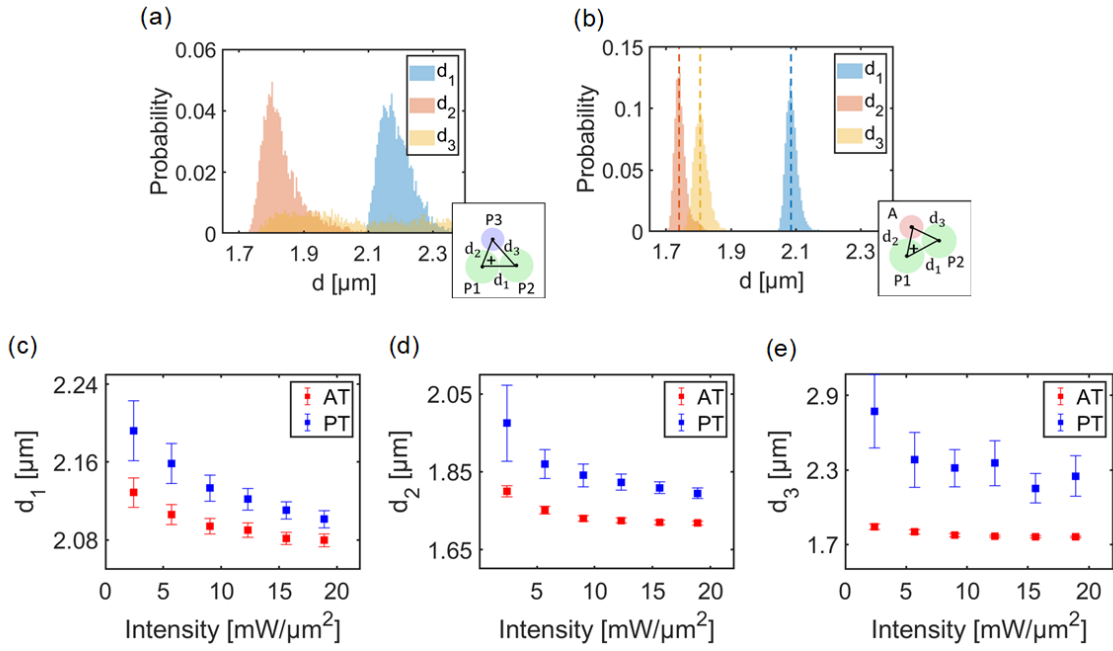


FIGURE 4.13: Interparticle distances in passive (PT) and active (AT) trimers. (a,b) Distance distributions for PT and AT, with dashed lines showing mean values at $5.7 \text{ mW}/\mu\text{m}^2$ laser intensity. (c-e) Evolution of average interparticle distances with increasing laser intensity for both AT and PT. This figure is reproduced from [140].

structures. The AT exhibits notably narrower distance distributions than the PT, suggesting more localized confinement of its constituent particles. Since the gradient optical force always pulls the colloids to the beam centre, resulting in the colloids being radially confined to the beam centre, where the colloid P1 is at the centre and P2 and P3 are located at an offset position from the beam centre in the case of a passive trimer. This results in well-defined distances d_1 and d_2 between P1, P2, and P1, P3, with a smaller distribution spread. In contrast, there is no interaction present between the peripheral colloids (P2 and P3) to bind them together. As a result, the distance between the peripheral colloids (d_3) is much more widely distributed compared to d_1 and d_2 . For the AT, the average center-to-center distances between colloids (d_1 , d_2 , and d_3) are found to be $2.09 \mu\text{m}$, $1.74 \mu\text{m}$, and $1.80 \mu\text{m}$, respectively. These distributions' full-width at half-maximum (FWHM) values are approximately $0.04 \mu\text{m}$, $0.03 \mu\text{m}$, and $0.05 \mu\text{m}$, respectively. These small fluctuations indicate strong confinement and minimal positional deviation, reflecting a stiffer, more stable structure. This

increased rigidity in the AT arises from two contributing factors. First, higher laser intensities increase the optical gradient forces that pull particles toward regions of higher intensity, thereby decreasing interparticle separation. Second, the thermo-osmotic attraction induced by the heated colloid in the AT draws neighboring colloids towards the thermally active colloids, further reducing the spacing between them. Figures 4.13(c)-4.13(e) illustrate how the average interparticle distances systematically decrease with increasing laser power, emphasizing the combined role of optical forces and thermal interactions in promoting tighter binding and structural rigidity.

4.10 Higher-order rotating structures

Given the crucial role of the induced thermal field in governing rotational dynamics, including additional thermally active colloids provides a means to modulate the dynamics further. In this section, we investigate the emergent structures and their corresponding dynamics when multiple thermally active colloids are incorporated.

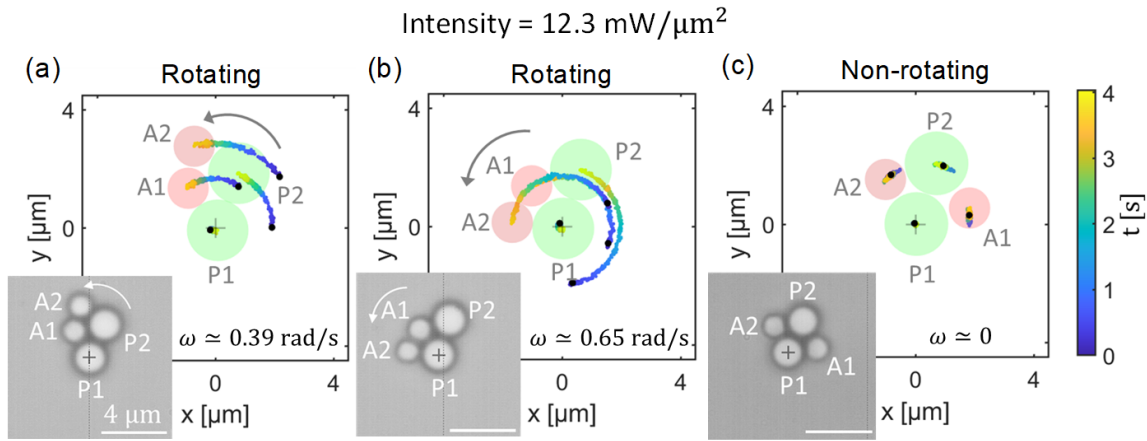


FIGURE 4.14: Rotational motion in higher order colloidal structures. (a)-(c) illustrate the trajectories of structures composed of two passive melamine formaldehyde colloids (P1, P2) and two thermally active polystyrene colloids (A1, A2), with diameters $d_{P1} = d_{P2} = 2 \mu\text{m}$ and $d_{A1} = d_{A2} = 1.3 \mu\text{m}$, respectively. The insets display corresponding bright-field microscopic images. Structures shown in (a) and (b) demonstrate rotational behavior, exhibiting angular velocities of 0.39 rad/s and 0.65 rad/s , respectively, while the configuration in (c) does not show any observable rotation. This figure is reproduced from [140].

Fig. 4.14(a) and 4.14(b) display asymmetric structure configurations containing two thermally active colloids (A1 and A2) positioned on the same side of passive colloid P2 (see video 10 [141]). Both structures exhibit rotation but with distinct angular velocities. In Fig. 4.14(a), colloid A2 is situated farther from the beam center than in Fig. 4.14(b), resulting in a lower effective surface temperature due to the Gaussian beam profile. Consequently, the magnitude of the induced thermo-osmotic slip flow is reduced in Fig. 4.14(a), leading to a slower rotation rate (0.39 rad/s) compared to the structure in Fig. 4.14(b) (0.65 rad/s).

In contrast, when the active colloids A1 and A2 are symmetrically placed on opposite sides of P2, as shown in Fig. 4.14(c), the thermo-osmotic torques generated by A1 and A2 counteract each other, resulting in no torque imbalance in the structure, leading to suppression of rotational motion (see video 10 [141]).

4.11 Evolutionary dynamics of rotating structures

Till the previous sections, we mainly discussed the rotational dynamics of asymmetric colloidal structures under focused optical illumination. If we study the evolution of such structures over a long duration, then due to the thermo-osmotic flow field, new colloids will join the system and it grows bigger (see Fig. 4.15). The system maintains the characteristic rotational motion until the dynamic asymmetry, and hence, the torque imbalance is maintained.

4.12 Conclusion

To summarize, we have experimentally demonstrated the emergence of directed rotation in multi-component colloidal structures without direct angular momentum transfer from optical fields. The observed rotation arises from optothermally induced asymmetries in the system, and we have shown that the handedness of this rotation can be dynamically controlled by tuning the relative spatial configuration of the thermally active and passive components.

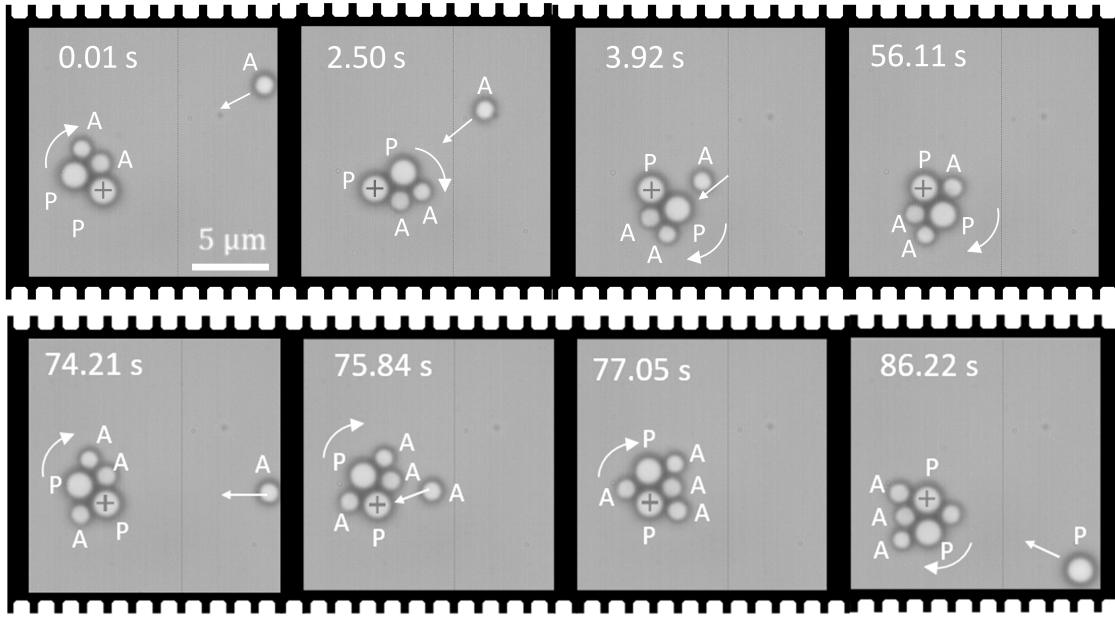


FIGURE 4.15: Rotational dynamics of optothermally driven asymmetric evolving colloidal structure, composed of passive and thermally active colloids under focused optical beam illumination of intensity $I_0 = 12.3 \text{ mW}/\mu\text{m}^2$. '+' indicates the beam center. This figure is reproduced from [140].

This dynamic control mechanism offers a significant advantage over pre-fabricated asymmetric structures, which lack flexibility in modulating rotation direction [98, 146, 186, 196–198]. Furthermore, we have demonstrated that the angular velocity of the structure can be finely tuned by varying the intensity of the incident laser beam, effectively using it as an environmental parameter to modulate rotational behavior.

Our strategy of leveraging thermo-osmotic interactions between thermally active and passive colloidal entities is broadly applicable across diverse colloidal systems due to the generality of thermo-osmotic flow fields. Interestingly, this kind of optothermally driven rotational behavior bears resemblance to phenomena observed in natural non-equilibrium systems such as bacterial vortices [155, 199, 200] and sperm-cell [201–203] swarming dynamics. Therefore, the presented system serves as a versatile platform for controlled micro-rotation and offers a testbed for exploring the physics of active matter, collective dynamics, and non-equilibrium thermodynamics at the microscale. This work paves the way for future investigations into microscale engines, responsive micromachines, and model systems for

studying emergent behaviors in active and living matter.

Chapter 5

Guided motion of colloidal structures in optical ring beam

Abstract:

In this chapter, we explore the emergence of controlled, directional revolutionary motion in dimer and trimer structures composed of thermally active and passive colloidal particles within a ring-shaped optical beam. This motion arises from optothermal interactions, without requiring direct mechanical forces from the external optical field. The optical absorption properties of the colloidal particles give rise to optothermal interactions that exhibit both attractive and repulsive characteristics. The attractive interactions create an imbalance that drives the propulsion, while the repulsive interactions regulate the structural arrangement of the system. Together, the specific configuration of colloidal particles within these dimer and trimer structures enables controlled, directional motion. The optical gradient force further acts as a confining mechanism, guiding the particles along a predefined path. By varying the intensity of the optical field, we can modulate the dynamics of these systems, offering a tunable approach to their behavior. This chapter is an adaptation of the research article "ACS Photonics 12(2), 855-863 (2025), (arXiv:2409.16792)" [[74](#)].

5.1 Introduction and motivation

Since the pioneering discovery of optical tweezers by Arthur Ashkin, optical forces have become a cornerstone for confining and manipulating micro- and nanoscale colloidal objects [2, 3, 45]. These optical fields, however, are often accompanied by heating processes that trigger various out-of-equilibrium optothermal interactions, such as thermo-osmosis [30–32], thermophoresis [33–36], diffusiophoresis [186], and depletion interactions [42]. These interactions significantly influence the dynamics of colloidal objects and can even be harnessed to design bio-mimetic artificial active matter systems in complex environments [136]. The dynamics of artificial colloidal systems have been widely studied under both hard and soft confinement conditions. Hard confinement is typically achieved using lithographic boundaries or channels [204], while soft confinement relies on optical potential landscapes [57, 174, 176, 189]. In most existing studies, the motion of colloidal particles is driven by externally applied fields, either through active steering of the field [35, 68, 98] or by establishing a static, asymmetric force distribution [14, 69, 205–207], often requiring sophisticated feedback mechanisms. However, comparable dynamic states can also emerge under static external fields through mutual interactions between colloidal objects [137, 140, 208]. For example, Yuval Yifat et al. demonstrated directional motion of a heterodimer within a ring-shaped optical illumination, where the motion arose from an imbalance in scattering efficiencies between the colloidal particles [208]. Despite this insight, their study highlighted limitations in controlling the resulting dynamics. Beyond these findings, optothermal interactions offer a promising avenue for guiding colloidal particles along optically defined paths [31, 32, 35, 133, 209]. By leveraging extended optical traps [14, 101] and holographic optical tweezers [68, 210–212], researchers can overcome localization effects and enhance motion control. This potential underscores a growing need to investigate how novel dynamic states of colloidal systems can emerge from optothermal interactions within structured optical fields.

Building on this foundation, here we present a study demonstrating the controlled directional propulsion of colloidal particles facilitated by optothermal interactions along an

optically defined path. We employ ring-shaped optical illumination to simplify the dynamics into a pseudo-one-dimensional motion, creating a continuous field that sustains uninterrupted colloidal motion. The experimental schematic is illustrated in Fig. 5.1(a), with an inset showing the intensity profile of the ring-shaped optical illumination. We observe directional motion in two colloidal configurations: a dimer, consisting of a passive (P) and a thermally active (A) colloid, and a trimer, comprising a passive (P) and two thermally active colloids (A1, A2), as depicted in Fig. 5.1(b) and Fig. 5.1(c), respectively. The dynamics of these systems can be precisely tuned by adjusting the intensity of the incident optical illumination, which modulates the interplay between attractive thermo-osmotic and repulsive thermophoretic interactions.

5.2 Experimental methods and characterization

Our experiments involve two types of colloidal particles - passive (P) and thermally active (A) as previously discussed in Chapter 3. The passive colloids are melamine formaldehyde (MF) spheres with a diameter of 2 μm , while the thermally active colloids are polystyrene (PS) spheres of 1.3 μm diameter embedded with iron oxide nanoparticles. When exposed to laser illumination, the iron oxide nanoparticles in the active colloids absorb light energy and release it as heat, generating a localized temperature field in the surrounding fluid.

To prepare the sample, a dilute aqueous suspension is created by mixing the passive and active colloids in Milli-Q water. A 10 μL droplet of this mixture is placed in a chamber formed by two glass coverslips separated by a 120 μm spacer. The sample is then mounted on a dual-channel optical microscopy setup, illustrated in Fig. 5.1(d). A blazed axicon hologram (Fig. 5.1(e)) displayed on a phase-only spatial light modulator (SLM) generates a ring-shaped optical beam from a 532 nm continuous-wave Gaussian laser source. The first-order diffracted ring beam is isolated using an iris and directed into a 50X (0.8 NA) microscope objective via a relay lens system (L3 and L4).

This linearly polarized ring beam simultaneously traps both the passive and thermally

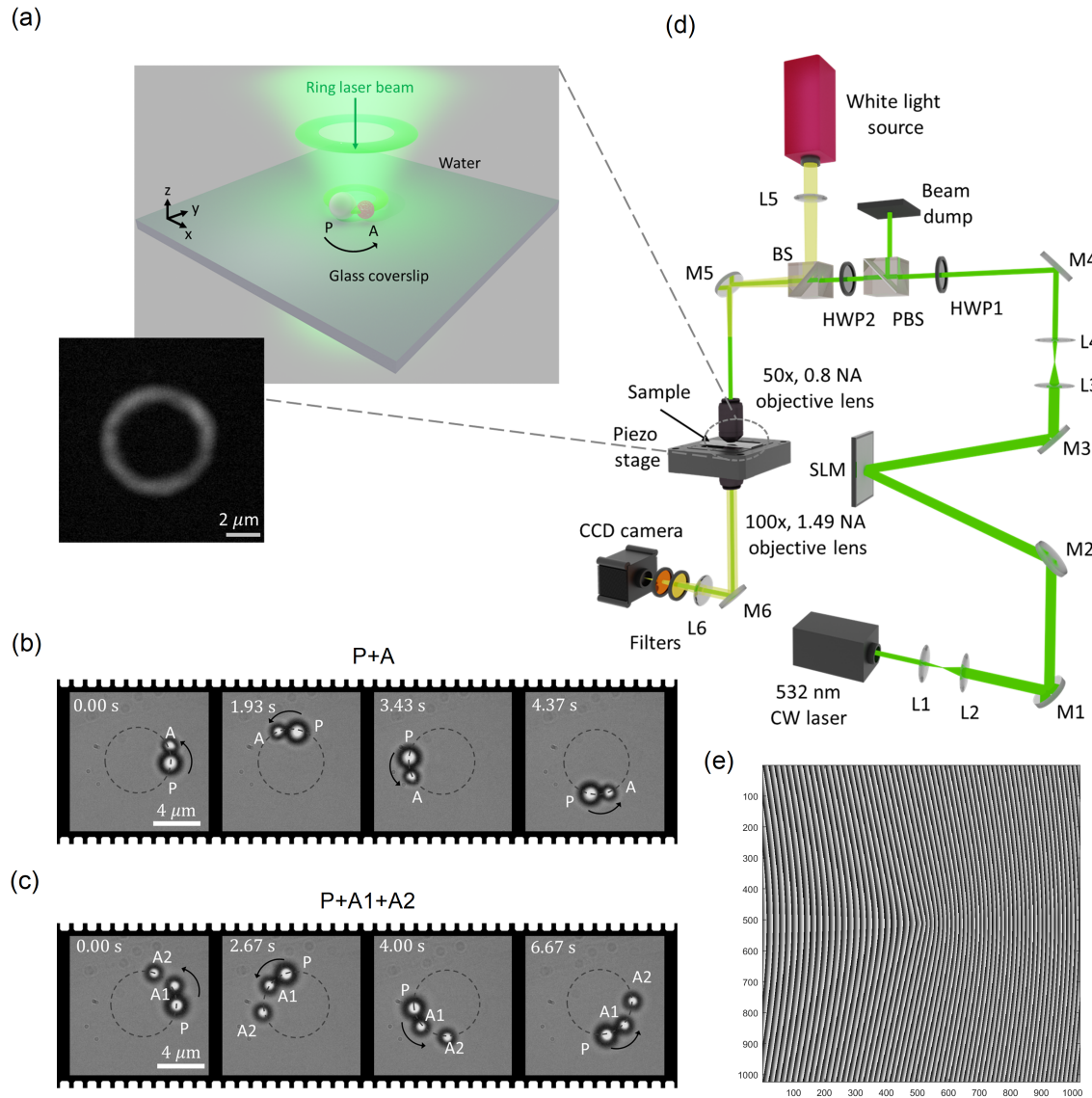


FIGURE 5.1: Guided motion of colloidal structures in ring-shaped optical illumination. (a) Schematic showing a dimer (P+A) undergoing directional propulsion in a linearly polarized 532 nm ring beam. (b) and (c) Experimental trajectories of dimer and trimer (P+A1+A2) structures, respectively. (d) Dual-channel microscopy setup. (e) A ring beam is generated using a blazed axicon pattern on an SLM and projected via a 50x, 0.8 NA objective. Dynamics are recorded using a 100x, 1.49 NA oil objective and CCD camera. This figure is reproduced with permission from the article [74].

active colloids, whereas it heats only the thermally active colloids. To capture the resulting dynamics, light from the sample plane is collected using a 100X oil immersion objective (1.49 NA) and recorded with a CCD camera at 30 frames per second. The trajectories of individual colloids are extracted from the video data using the TrackMate plugin in FIJI/ImageJ [138, 139].

5.3 Dynamics of individual colloidal particles in ring beam

5.3.1 Confinement of colloids

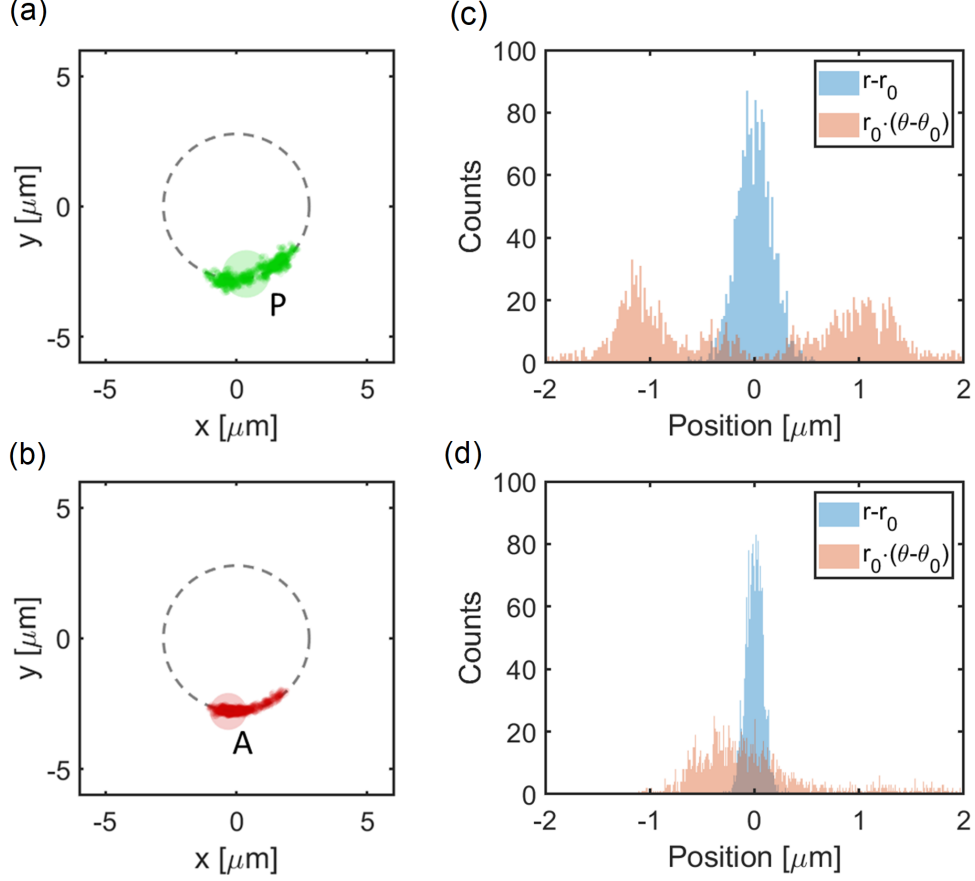


FIGURE 5.2: Dynamics of individual passive and thermally active colloids under ring optical beam illumination (radius $r_0 = 2.79 \mu\text{m}$ and intensity $I_0 = 0.1 \text{ mW}/\mu\text{m}^2$). The 53s long trajectory of a passive (P) and a thermally active colloid (A) are shown in (a) and (b). (c) and (d) show respective radial ($r - r_0$) and arc length ($r_0 \cdot (\theta - \theta_0)$) distributions, where r_0 and θ_0 are the average radial and angular position. Both colloids P and A are confined radially but azimuthally diffuse. This figure is reproduced with permission from the article [74].

We illuminated the ring beam close to the individual colloids to understand the dynamics of the colloids. The trajectories of the thermally passive and active colloids are shown in Fig. 5.2(a) and 5.2(b), respectively. The trajectories show that the colloids are confined radially (see video 11 [141]). But azimuthally, they are not strictly bound. The corresponding radial and azimuthal position distributions of colloids are shown in Fig. 5.2(c) and 5.2(d). We have plotted the arc length distribution $r_0(\theta - \theta_0)$ instead of the exact azimuthal position distribution $(\theta - \theta_0)$. The colloids get confined near the maximum intensity region of the

optical ring beam, i.e., at $r = r_0$ of the ring beam, due to the dominant in-plane gradient optical force acting on them.

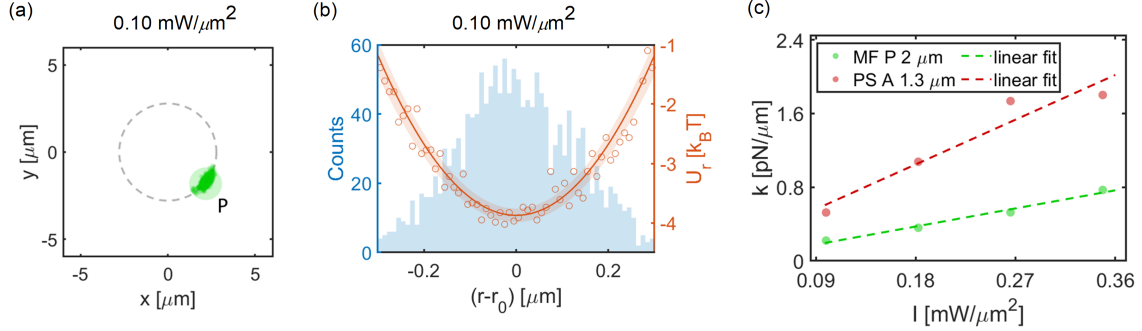


FIGURE 5.3: Radial trapping of colloids. (a) The position distribution of a passive colloid with a diameter of $2\mu\text{m}$ under the incident optical ring beam. (b) show the associated radial position and potential distribution. (c) The trap stiffness of the colloids at different beam intensities, where the linear fitting is indicated by dashed lines. This figure is reproduced with permission from the article [74].

We have also quantified the colloids' radial trapping state to quantify the radial potential depth and efficiency of confinement. From the typical trajectory of the colloids as shown in Fig. 5.3(a), we obtain the radial potential profile by taking the negative logarithm of the radial position distribution as,

$$U_r = -k_B T \log \rho(r), \quad (5.1)$$

where k_B and T are the Boltzmann constant and the ambient temperature. The corresponding radial distribution $\rho(r)$ and the potential profile U_r are depicted in Fig. 5.3(b). By fitting with a harmonic potential as $U_r = U_0 + \frac{1}{2}k_r(r - r_0)^2$ where the fitting parameter k_r is called the trap stiffness indicating the efficiency of the radial confinement. Figure 5.3(c) shows that the trap stiffness scales linearly with increasing laser intensities I and can be fitted as

$$k(I) = A \cdot I + B, \quad (5.2)$$

where A and B are the linear fitting parameters. The corresponding fitting parameters for passive and thermally active colloids are summarized in table 5.1.

Colloids	Diameter [μm]	A [$f\text{s}$]	B [$\text{pN}/\mu\text{m}$]
MF P	2.0	2.18	-0.02
PS A	1.3	5.39	0.08

TABLE 5.1: Linear fitting parameters obtained from equation (5.2) for different thermally active and passive colloids used in our experiments.

5.3.2 Temperature field generated by thermally active colloids

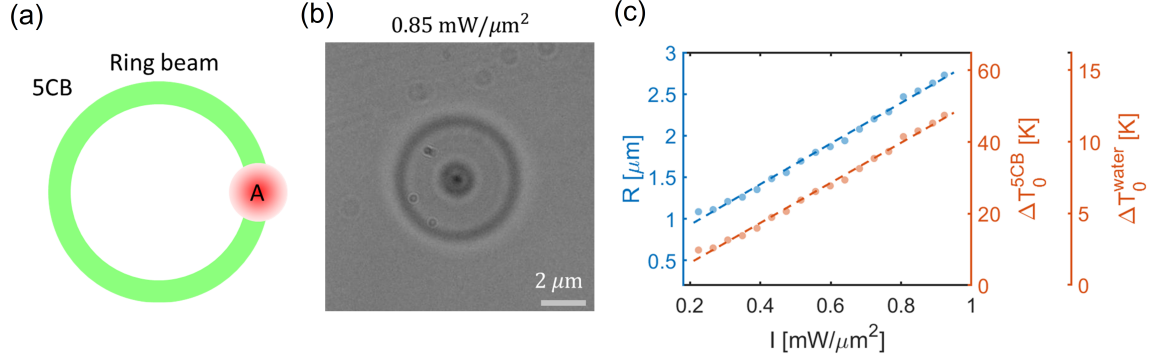


FIGURE 5.4: Temperature measurement of thermally active colloids under ring-shaped optical illumination. (a) A schematic illustrates a drop-casted thermally active colloid in a 5CB liquid crystal medium and under-ring optical beam. (b) Snapshot of the isotropic phase bubble generated around the thermally active colloid. (c) The linear incremental trend of the phase transition radius and the maximum surface temperature of the heated colloids. This figure is reproduced with permission from the article [74].

Although both the thermally active and passive colloids get trapped individually under ring optical illumination, owing to the absorbing nature of the thermally active colloids, a thermal field is generated around the thermally active colloids. One can estimate the approximate value of the surface temperature of the colloids by using the 5CB liquid crystal-based temperature measurement techniques. We drop-cast thermally active colloids on a glass coverslip and filled the sample cell with 5CB liquid crystal. The drop-casted thermally active colloid is exposed to the ring beam, as shown in the schematic Fig. 5.4(a). By quantifying the radius of isotropic phase bubble R from the bright field optical image (Fig. 5.4(b)), we can estimate the surface temperature of the heated colloid ΔT_0^{5CB} in the 5CB medium as

$$\Delta T_0^{5CB} = (T_{pt} - 293K)(R - a)/a, \quad (5.3)$$

where a and $T_{pt} = 35^\circ\text{C}$ are the radius of the thermally active colloid and the phase transition temperature. By mapping the temperature rise to the water, we obtain the linear incremental trend of the thermally active colloids to water medium as $\Delta T = A \cdot I + B$ (see Fig. 5.4(c)).

5.4 Propulsion of thermally active colloids in optically laid out ring path

The localized heating produced by thermally active colloids generates thermo-osmotic flows that draw nearby colloids toward the heat source near the glass substrate. As a result, when both active and passive colloids are exposed to a ring-shaped optical field, these induced flows give rise to asymmetric attractive interactions. This interaction, denoted as the potential $U_{A,P}$, can be quantitatively evaluated following the methodology described in [213, 214]. The procedure involves measuring the pairwise distance distribution of colloids along the circular arc, $P_{A,P}(r_0\Delta\theta_{A,P})$, and computing the interaction potential via the relation:

$$U_{A,P}(r_0\Delta\theta_{A,P}) \approx -k_B T \ln P(r_0\Delta\theta_{A,P}). \quad (5.4)$$

As depicted in Figure 5.5(a), the potential profile shows a minimum at a separation distance close to $a_P + a_A = 1.65 \mu\text{m}$, the smallest allowed center-to-center distance between the particles. This observation reinforces the attractive nature of the interaction. The resulting unidirectional attraction effectively binds the two colloids into a dimer, which exhibits a directional propulsion in an optically laid out path as demonstrated in Fig. 5.5(b). Notably, such propulsion of structures only emerges when both active and passive colloids coexist under illumination (see video 11 [141]).

To characterize this propulsion, the mean squared angular displacement (MSAD) is computed and shown in Figure 5.5(c). The experimental MSAD data fits well to the functional form:

$$MSAD(\tau) = \langle [\theta(t + \tau) - \theta(t)]^2 \rangle = 2D_r\tau + \omega^2\tau^2, \quad (5.5)$$

where D_r denotes the overall diffusion constant in azimuthal direction, ω represents the angular speed of the dimer, and τ is the lag time. From this, the translational speed of the rotating dimer can be approximated using $v = \omega \cdot r_0$.

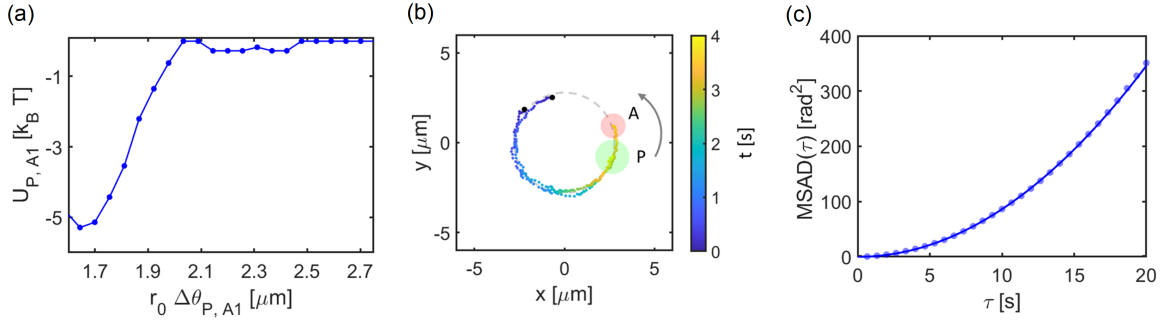


FIGURE 5.5: Colloidal dimer dynamics under ring-shaped optical illumination ($r_0 = 2.79 \mu m$, $I_0 = 0.5 mW/\mu m^2$). (a) The interaction potential between passive and thermally active colloids reveals a unidirectional thermo-osmotic attraction arising from localized heating under the ring beam. This non-reciprocal interaction drives the formation and propulsion of the active dimer. (b) The experimentally observed trajectory of the dimer highlights its propulsion dynamics in the optically guided path. (c) The corresponding mean squared angular displacement (MSAD).

This figure is reproduced with permission from the article [74].

5.4.1 Role of gradient optical and the thermo-osmotic forces

To investigate the role of slip-flow-induced drag and optical gradient forces in the dynamics of active dimers, we carried out numerical simulations by solving the Langevin equations under the influence of external forces [142, 215]. These external forces include both the optical gradient force and the thermo-osmotic flow-induced drag.

The intensity distribution of the ring-shaped optical trap can be described by the equation:

$$I(r) = I_0 \exp \left(-\frac{(r - r_0)^2}{w_0^2} \right), \quad (5.6)$$

where I_0 is the peak intensity, r_0 is the radial position of the maximum intensity, and w_0 denotes the beam waist. Figure 5.6(a) shows the simulated intensity distribution, which qualitatively reproduces the experimental ring-beam profile. Within this region, the optical

gradient force acting on colloid i ($i = P, A$) can be approximated as a harmonic restoring force:

$$\vec{F}_{o,i} \approx -k_i(\vec{r}_i - \vec{r}_0), \quad (5.7)$$

assuming $|\vec{r}_i - \vec{r}_0| \ll w_0$.

The passive colloid P experiences a drag due to the thermo-osmotic slip flow generated by the nearby active colloid A. This force can be approximated as:

$$\vec{F}_{T\{P,A\}} \approx \gamma_P \vec{v}_s \approx \frac{\gamma_P \chi \vec{\nabla} T_A}{T_s}, \quad (5.8)$$

where γ_P is the viscous drag coefficient for the passive colloid, χ is the thermo-osmotic slip coefficient, ∇T_A is the temperature gradient induced by the active colloid, and T_s is the ambient temperature.

To prevent particle overlap and account for short-range repulsion, we also included a Lennard-Jones (LJ) interaction between particles i and j , defined as:

$$\vec{L}J_{\{ij\}} = 4\epsilon \left[12 \frac{\sigma_{ij}^{12}}{r_{ij}^{13}} - 6 \frac{\sigma_{ij}^6}{r_{ij}^7} \right] \hat{r}_{ij}, \quad (5.9)$$

where $\sigma_{ij} = \sigma_0(a_i + a_j)$ sets the interaction length scale based on the radii a_i and a_j of the colloids, and ϵ is the interaction strength. We used $\sigma_0 = 1$ and $\epsilon = 20 \times 10^{-21}$ J in the simulations.

The Langevin equations governing the dynamics of the active (A) and passive (P) colloids are then:

$$\gamma_A \dot{\vec{r}}_A(t) = \vec{F}_{o,A}(t) + \vec{L}J_{\{A,P\}}(t) + \sqrt{2k_B T_A \gamma_A} \vec{W}_A^t, \quad (5.10)$$

$$\gamma_P \dot{\vec{r}}_P(t) = \vec{F}_{o,P}(t) + \vec{L}J_{\{P,A\}}(t) + \vec{F}_{T\{P,A\}}(t) + \sqrt{2k_B T_P \gamma_P} \vec{W}_P^t, \quad (5.11)$$

where the last term signifies the Gaussian white stochastic noise.

These equations were numerically solved after discretization as given below:

$$\vec{r}_A^{(k+1)} = \vec{r}_A^{(k)} + \frac{\Delta t}{\gamma_A} \left[\vec{F}_{o,A}^{(k)} + L\vec{J}_{\{A,P\}}^{(k)} + \sqrt{\frac{2k_B T_A \gamma_A}{\Delta t}} \vec{w}_A^{(k)} \right], \quad (5.12)$$

$$\vec{r}_P^{(k+1)} = \vec{r}_P^{(k)} + \frac{\Delta t}{\gamma_P} \left[\vec{F}_{o,P}^{(k)} + L\vec{J}_{\{P,A\}}^{(k)} + \vec{F}_{T\{P,A\}}^{(k)} + \sqrt{\frac{2k_B T_P \gamma_P}{\Delta t}} \vec{w}_P^{(k)} \right], \quad (5.13)$$

where Δt is the simulation time step, and $\vec{w}_i^{(k)}$ is the stochastic force sampled from a Gaussian distribution with zero mean and unit variance.

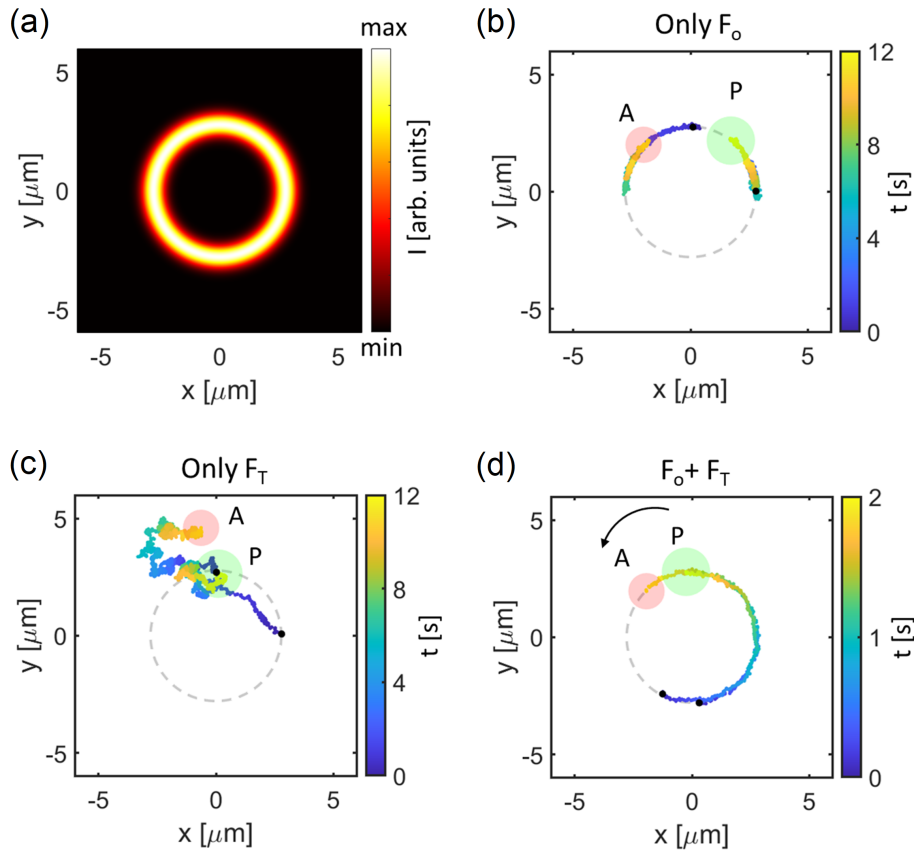


FIGURE 5.6: Simulated dynamics of active dimers and the role of various forces. (a) The simulated intensity profile of the ring optical beam profile, governed by the equation $I(r) = I_0 \exp(-(r - r_0)^2/w_0^2)$. The corresponding dynamics considering (b) only gradient optical force ($\vec{F}_o$), (c) only thermo-osmotic force F_T , and (d) both the gradient optical force and thermo-osmotic force. Only in (c) colloids form tightly bound dimer structures that undergo directional propulsion. This figure is reproduced with permission from the article [74].

The simulated dynamics of the colloidal dimer were obtained by numerically integrating the governing Langevin equations. When only the optical gradient force was present, the

particles remained confined along the radial direction of the trap. However, no stable dimer formation or net directional propulsion was observed under these conditions, as illustrated in Fig. 5.6(b).

In contrast, when only the thermo-osmotic drag force was active, the stochastic thermal noise caused the particles to drift away from the illuminated region due to the absence of a confining force. Once outside the beam, the particles were no longer heated and their motion became purely diffusive, as shown in Fig. 5.6(c).

Notably, when both the optical gradient force and the slip-flow-induced drag were simultaneously active, a stable dimer configuration emerged that exhibited persistent directional propulsion, as demonstrated in Fig. 5.6(d). These results suggest that the optical gradient force serves to confine the colloids along the designed intensity path, while the slip-flow-induced drag effectively acts as the driving mechanism for propulsion.

5.4.2 Investigation of optical binding forces on the dynamics

To ensure that the observed dynamics are not due to optical force via the optical field distortion by the colloidal systems, we performed finite element method (FEM) based field simulation in COMSOL 5.5. We considered a cuboidal geometry of dimension $6 \mu m \times 6 \mu m \times 5 \mu m$ as simulation domain as shown in Fig. 5.7(a). The simulation domain consists of glass in the bottom part and water in the upper part, where colloidal particles (P and A) are submerged. The ewfd module is used to simulate the electric field distribution. From the top of the geometry, a Bessel beam of wavelength 532 nm is used for modeling the ring beam as given below

$$E(\rho, z) = C J_0(k_\rho \rho) e^{-\frac{\rho^2}{w^2(z)}} e^{i \left(\frac{k_z \rho^2}{2R(z)} - \eta(z) + k_z z \right)}, \quad (5.14)$$

where $J_0(k_\rho \rho)$, $w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_0} \right)^2}$, $R(z) = z \left(1 + \left(\frac{z_0}{z} \right)^2 \right)$, $\eta(z) = \tan^{-1} \left(\frac{z}{z_0} \right)$, k_z and k_ρ are the Bessel function of the first kind, beam waist, radius of curvature, Gouy phase, propagation vector components along propagation and transverse directions. Please note

$|k| = \sqrt{k_\rho^2 + k_z^2}$, where $|\vec{k}| = \frac{2\pi}{\lambda}$ is the total wave number. To simulate the ring beam we used $k_\rho = \frac{|\vec{k}|}{1.5}$, and the following material parameters have been used:

Material	Refractive Index
Glass	1.45
Water	1.33
P	1.60
A	$1.677 + 0.0169 i$

TABLE 5.2: Material parameters used in the simulation

To avoid spurious reflection from the boundaries of the simulation geometry, we considered using no-scattering boundary conditions. In the absence of particles, the electric field distribution in the sample plane is shown in Fig. 5.7(b). It can be observed that the simulated field profile is qualitatively comparable to the experimental intensity distribution. Due to the field distortion created by the presence of colloids, Fig. 5.7(c), optical forces are developed on them. For a uniform ring beam simulation, the azimuthal optical force acting on the colloids should be zero. But when multiple colloids are present in the ring beam simultaneously, then the light scattered by one colloid can lead to optical force insertion of the other. Because of the simultaneous presence in the beam, there are non-zero counteracting azimuthal optical forces of the colloids. Besides, it can be clearly seen that in the simulation, the beam is not exactly circularly symmetric due to polarization, so that might lead to a non-zero azimuthal optical force on the colloids at certain positions. Thus, the counteracting forces on the colloids are not solely due to non-circularly symmetric intensity in the beam; rather, the counteracting nature arises from beam distortion due to the simultaneous presence of multiple colloids in the beam.

In the experimental scenario, radially, the colloids are confined because of gradient optical forces and exhibit directional propulsion along the azimuthal direction. Thus, we analyzed the azimuthal component of optical forces acting on the particles as denoted by F_P^ϕ and F_A^ϕ in this case. It can be seen from Fig. 5.7(c) that these azimuthal forces acting on both the colloids counteract each other, leading to repulsion between them. Thus, only optical forces cannot lead to tightly bound dimer formation. Even hypothetically, if we consider the

colloidal dimer is formed, the non-zero net azimuthal optical forces lead to net torque as:

$$\vec{\tau} = \sum_i F_i^\phi r_i \hat{z} = -0.56 \times 10^{-19} \text{ Nm}.$$

The negative sign dictates that the colloidal dimer arranged as indicated in Fig. 5.7(c) will lead to clockwise motion, whereas experimentally, we found that the colloidal dimer exhibits counterclockwise motion. In our dynamics, thus, the role of optical torque is not significant.

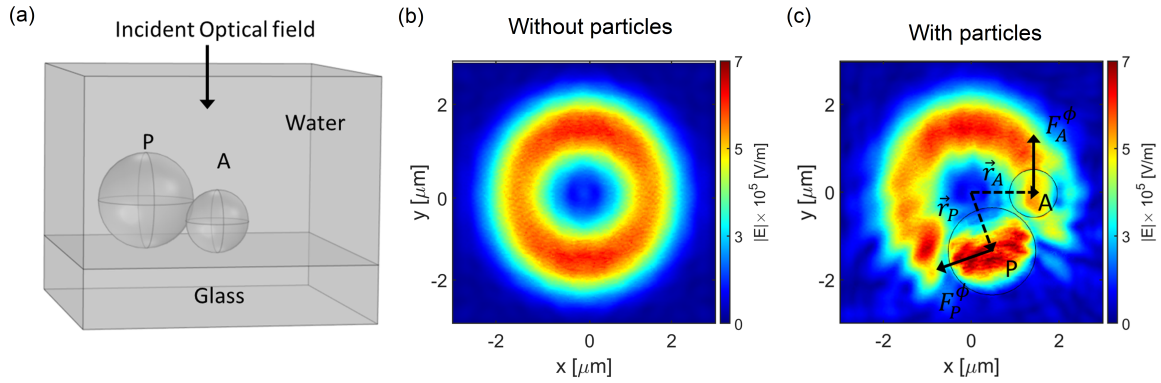


FIGURE 5.7: Finite element method (FEM) simulation showing the impact of optical forces on the dynamics of active dimer (AD) structures in a ring-shaped beam. (a) Schematic representation of the simulation geometry. Simulated optical intensity profile in the xy -plane at a height of $1 \mu\text{m}$ above the glass-water interface: (b) without colloidal particles and (c) with colloids present. The insertion of particles perturbs the optical field, giving rise to optical forces acting on them. The power of the beam is 5 mW. This figure is reproduced with permission from the article [74].

5.5 Controlling the dynamics of active dimer structures

The propulsion of active dimer structure is emerging from the non-reciprocal thermo-osmotic interaction, acting from passive to thermally active colloids. By changing the arrangement, we can control the direction of propulsion. The trajectories of two ADs propelling in different directions based on their arrangements are shown in Fig. 5.8(a) and 5.8(b) (also see video 12 [141]). Not only the direction of motion but also the velocity can be tuned by modulating the thermal field by tuning the intensity of the optical field (see video 13 [141]). The linear incremental trend of the surface temperature of thermally active colloids along

with both the angular speed (ω) and the linear speed (v) with the intensity of ring beam are depicted in Fig. 5.8(c) and Figure 5.8(d) respectively.

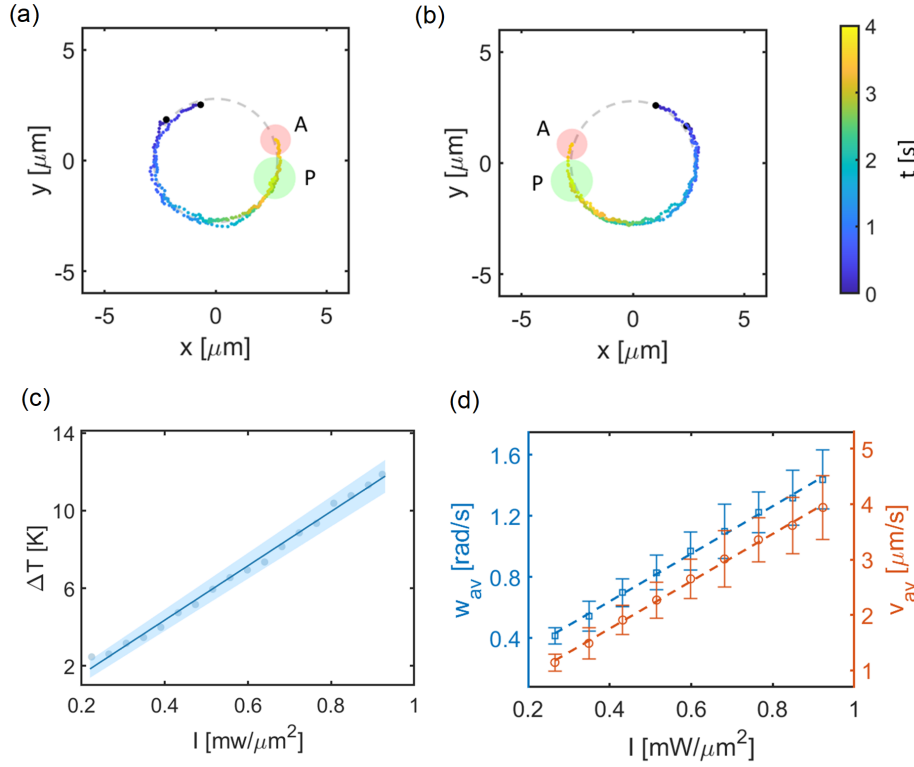


FIGURE 5.8: Tunability of the dynamics. (a) and (b) show different propulsion directions based on the initial state of organization of the colloids and the active dimer propels heading towards the thermally active colloid. (c) The surface temperature and both the (d) linear v and angular velocities ω scale linearly with increasing intensity of the incident optical field. This figure is reproduced with permission from the article [74].

5.6 Active trimer structures under ring optical beam

5.6.1 Counteracting attractive and repulsive interaction in active trimer structures

In the previous sections, we saw that the attractive thermo-osmotic interaction leverages the directional propulsion behavior of the active dimer structures. In this section, we will be demonstrating the simultaneous emergence of attractive and repulsive interactions within an active trimer (AT) structure composed of one passive (P) and two thermally active (A1,

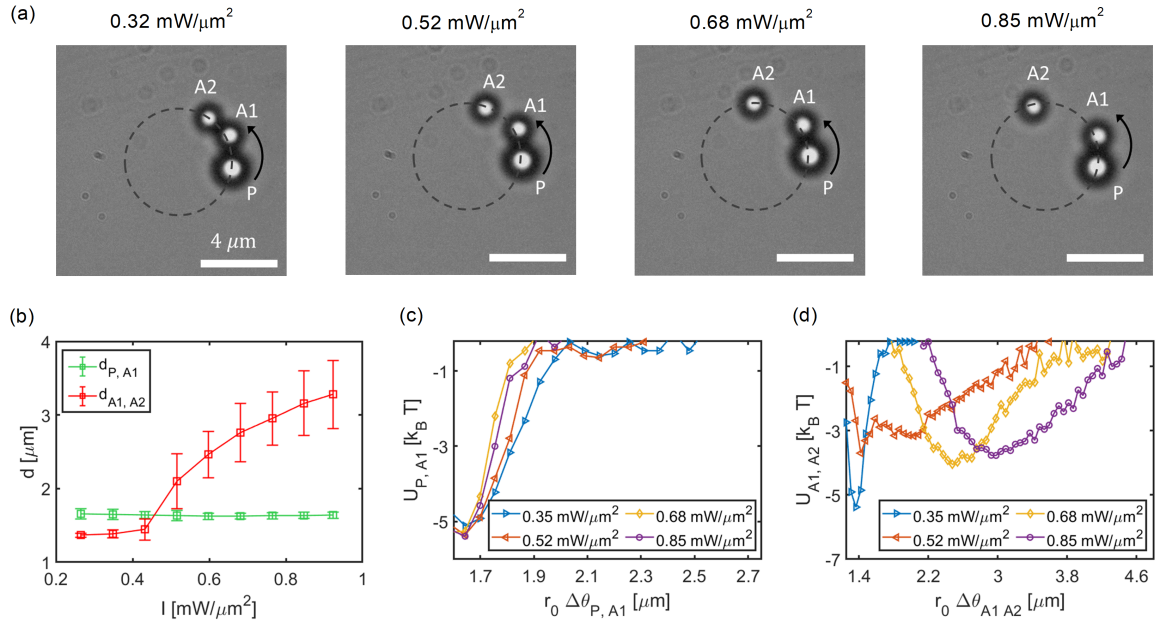


FIGURE 5.9: Controllable interactions within colloidal active trimer (AT) assemblies. (a) The motion and configuration of the AT structure are modulated by the optical beam intensity, governed by the balance between attractive thermo-osmotic and repulsive thermophoretic forces. (b) Variation of inter-particle distances with changing laser intensity: the separation between particles P and A1 remains relatively unchanged, while the distance between A1 and A2 increases with intensity. (c)-(d) Computed interaction potentials between P-A1 and A1-A2 pairs at various laser intensities. This figure is reproduced with permission from Ref. [74].

A2) colloids. These structures exhibit directional propulsion, and their structural properties can be altered using the intensity of the incident optical field as shown in Fig. 5.9(a) (also see video 14 [141]). The distances between the colloidal pair (P, A1) and (A1, A2) are shown in Fig. 5.9(b). It can be clearly seen that although the distance between the colloidal pair (P, A1) does not change significantly, the distance between the pair (A1, A2) exhibits non-linear variation with the intensity of the optical field. We have even calculated the corresponding interaction potentials ($U_{P,A1}$, $U_{A1,A2}$) as shown in Fig. 5.9(c) and 5.9(d) respectively. A clear observed shift in the potential minima position of $U_{A1,A2}$ is observed, which is absent in the case of $U_{P,A1}$. This indicates the presence of a temperature-dependent repulsive interaction between the colloidal pair (A1, A2) in addition to the attractive slip flow-induced drag force. Such repulsive interaction can only originate from the thermophoretic behavior of the colloids, which is generally characterized by the thermo-diffusion constant (D_T) of colloids. The temperature-dependent thermo-diffusion constant can be modelled as

$D_T(T) \approx C(T - T^*)$, where C is a constant, T^* is the transition temperature [62, 216]. For the colloids $T < T^*$, $D_T(T)$ is negative, leading to an attractive interaction towards the heat source. In contrast, for $T > T^*$, $D_T(T)$ is positive, leading to the development of repulsive interaction from the heat source. Due to this temperature-dependent thermophoretic behavior, a repulsive interaction between the thermally active colloids above a certain laser intensity is developed. Similar repulsive behavior is also previously observed between thermally active colloids as reported in [137]. A similar repulsive interaction is absent between the colloidal pair (P, A1). This is most probably due to: (1) the passive colloids do not get directly heated up and act as a heat source, resulting in less significant change in their thermophoretic nature. (2) The thermophoretic transition temperature T^* of the passive colloid is way beyond the experimentally induced thermal field.

5.6.2 Dynamics simulation of active trimer structures

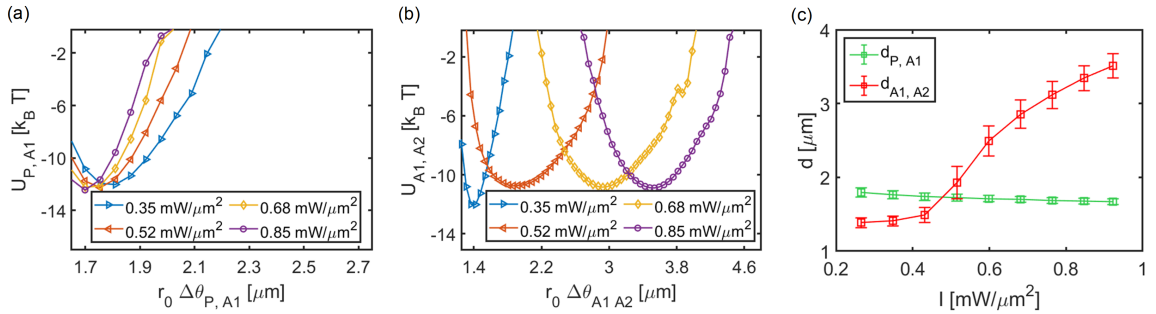


FIGURE 5.10: Simulated interaction in an AT structure with one passive and two thermally active colloids (P+A1+A2). (a)-(b) Interaction potentials $U_{P,A1}$ and $U_{A1,A2}$, showing stable minima for $U_{P,A1}$ and intensity-dependent shift in $U_{A1,A2}$. (c) Simulated distance trends between colloid pairs (P, A1) and (A1, A2). Simulated interaction between colloids in AT structure with one passive and two thermally active (P+A1+A2) colloids. This figure is reproduced with permission from Ref. [74].

We have also simulated the dynamics of active trimer systems under ring beam illumination, considering different forces acting on the system. In addition to the mentioned forces in the case of active dimer structures, temperature-dependent thermophoretic forces are considered between the thermally active colloidal pairs. The thermophoretic interaction force on i -th thermally active colloid due to the thermal field produced by j -th thermally active colloid

can be written as $\vec{F}_{Th\{ij\}} = -\gamma_i D_{T,i} \vec{\nabla} T_j$. With all these forces, the Langevin equation can be written as,

$$\dot{\vec{r}}_i(t) = \frac{1}{\gamma_i} \left(\vec{F}_{o,i}(t) + \sum_{j \neq i} \vec{L} J_{ij}(t) + \sum_{j \neq i} \vec{F}_{T\{i,j\}}(t) + \sum_{j \neq i; j=A1, A2} \vec{F}_{Th\{i,j\}}(t) + \sqrt{2k_B T_i \gamma_i} \vec{W}_i \right) \quad (5.15)$$

The above equation can be written in discretized form as,

$$\vec{r}_i^{(k+1)} = \vec{r}_i^{(k)} + \frac{\Delta t}{\gamma_i} \left(\vec{F}_{o,i}^{(k)} + \sum_{j \neq i} \vec{L} J_{ij}^{(k)} + \sum_{j \neq i} \vec{F}_{T\{i,j\}}^{(k)} + \sum_{j \neq i; j=A1, A2} \vec{F}_{Th\{i,j\}}^{(k)} + \sqrt{2k_B T_i \gamma_i} \frac{\vec{w}_i^{(k)}}{\sqrt{\Delta t}} \right). \quad (5.16)$$

Solving the above equations, we obtain the dynamics of the active trimer system. The optimized parameters used are simulation $\chi_P = 1.87 \times 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$, $\chi_{A1} = \chi_{A2} = 1.13 \times 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ and $C = 2.33 \times 10^{-12} \text{ m}^2 \cdot \text{K}^{-2} \cdot \text{s}^{-1}$ and $T^* = 295 \text{ K}$. The corresponding interaction potentials are shown in Figure 5.10(a) and Figure 5.10(b), respectively. We clearly observe a qualitatively similar trend in the simulated interaction potential $U_{A1, A2}$ minima shift with incident optical illumination as the experimental trend. The simulated interparticle distances are indicated in Figure 5.10(c), which also exhibits a similar trend as Figure 5.9(b).

5.7 Exploration of active dynamics in other structured optical fields

To generalize the guided motion of colloidal structures beyond ring-shaped optical fields, we conducted experiments using a line-shaped optical illumination. Similar propulsion behavior was observed, as illustrated in Fig. 5.11.

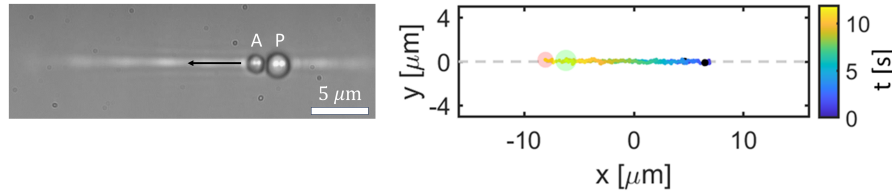


FIGURE 5.11: Guided motion of an active dimer structure in a line-shaped beam profile.

5.8 Conclusion

In this study, we explored the dynamics of colloidal, specifically dimers and trimers-driven by optothermal interactions under ring-shaped optical illumination. The observed behavior results from a complex interplay between thermo-osmotic and thermophoretic forces, which together give rise to both attractive and repulsive interactions.

The attractive interaction is mediated by drag forces originating from thermo-osmotic slip flows, leading to unidirectional motion along the predefined optical path. The direction of the propulsion depends on the spatial arrangement of passive and thermally active colloids within the structure. Additionally, the speed of motion can be tuned by adjusting the laser intensity.

At higher intensities, we observe repulsive interactions between thermally active colloids, where positive thermophoresis begins to outweigh the attractive slip-flow effects. This balance of forces enables precise control over interparticle spacing without affecting the direction of motion.

Interestingly, similar dynamics were also observed under linear optical beam profiles, suggesting that the underlying interaction mechanisms are general and adaptable to various optical field geometries. This tunable, directional transport mechanism offers promising potential for biomedical and technological applications, such as targeted drug delivery, microsurgery, and cargo transport at the microscale [109, 111, 170, 217–219]. These insights may also contribute to a better understanding of collective dynamics in natural systems, especially in constrained geometries like walls, channels, or in the presence of obstacles [154, 158, 220].

Moreover, the interplay of thermophoretic and thermo-osmotic effects is rooted in thermodynamic principles; this system is a valuable model for probing nonequilibrium thermodynamics in microscale environments.

Chapter 6

Overall summary and future directions

To summarize, the studies presented in this thesis explore our recent efforts in manipulating colloidal particles using a single static optical beam. Conventionally, optical manipulation of colloids depends either on the direct application of optical forces or on dynamically steering the optical field to overcome the random Brownian motion of the colloids. In contrast, this thesis demonstrates that controlled motion in colloidal systems can be achieved by exploiting non-reciprocal interactions between particles, without the need for any chemical reactions or functionalization.

In our approach, the optical beam serves a dual purpose: it confines the colloidal particles and simultaneously induces localized heating due to the absorption of light by thermally active colloids. This heating gives rise to temperature gradients in the surrounding fluid, which in turn activate thermodynamic processes such as thermo-osmosis and thermophoresis. These processes act as environmental mediators that facilitate interparticle interactions, thereby enabling the self-assembly and propulsion of colloidal structures. The resulting interactions are inherently non-reciprocal, as they depend on the asymmetry in thermal response between different particles.

To support these experimental observations, we have employed minimalistic numerical simulations that capture the essential physics of the system. All experimental studies were carried out using a dual-channel optical microscopy setup. This configuration allows independent control over the focusing conditions through the excitation (upper) channel while maintaining stable imaging through the observation (lower) channel.

In the following paragraphs, we summarize the key findings of each chapter. We also highlight ongoing work and outline future research directions inspired by the results presented in this thesis.

Chapter 3 focused on the dynamics of colloidal particles under defocused wide-field illumination. In this regime, gradient optical forces are minimal, and the primary effect of illumination is the creation of a temperature mismatch between colloids due to differences in optical absorption. This thermal imbalance leads to thermo-osmotic flow fields that drive non-reciprocal interactions between thermally active and passive colloids. These interactions result in the formation of bound structures such as dimers, trimers, and quadromers. The symmetry of these structures strongly influences their dynamics: while dimers and trimers exhibit directed motion, asymmetric quadromers show chiral trajectories. On a long time scale, rotational diffusivity or the rearrangement of colloids leads to effective diffusive motion of the structures.

Chapter 4 investigated the behavior of colloidal assemblies in a tightly focused Gaussian optical field. Here, confinement of colloids near the beam focus breaks the spatial symmetry of thermally induced interactions. In this configuration, active trimers exhibit rotational motion around the beam center, a behavior that contrasts with the unbound propulsion observed under wide-field illumination. This difference highlights the role of confinement and symmetry breaking in shaping collective colloidal dynamics.

In Chapter 5, we demonstrated controlled transport of colloidal structures using a structured optical field, in particular, a ring-shaped beam. The ring geometry provides an annular confinement potential, while thermal gradients induce propulsion along the ring. Systems with multiple thermally active colloids experience complex dynamics due to the coexistence of attractive thermo-osmotic and repulsive thermophoretic interactions. These competing effects enable fine control over the motion and configuration of the structures.

Taken together, the findings presented in Chapters 3 through 5 establish that optothermal forces can be harnessed for programmable colloidal manipulation. The observed dynamics are tunable not only through the intensity profiles of the optical field but also by changing

the colloid sizes, their thermal properties, and their spatial organization within a structure.

Thus, collectively, the work presented in this thesis led us to an important, intrinsic understanding of the light-matter interaction. The insights drawn can potentially serve as a bridge that combines optical and temperature-mediated interactions in the realm of colloidal manipulation and provides additional control parameters. Besides thermal field-mediated forces, combined with structured light can be employed to create novel assemblies of colloids, and provides a complementary approach to the direct optical beam-induced manipulation.

Limitations

While the results presented in this thesis offer valuable insights into colloidal dynamics under optical and thermal fields, both the experimental and numerical approaches employed have certain limitations.

In Chapter 3, we aimed to investigate the behavior of colloidal structures under wide-field optical illumination. Ideally, such an illumination should resemble a uniform plane wave extending over a large area. However, due to the limited output power of the laser diode used in our setup, we resorted to a defocused optical field as an approximation of wide-field illumination. This introduced non-negligible in-plane scattering forces, particularly at the edges of the beam profile, which in turn imposed constraints on the accessible spatial and temporal scales for tracking colloidal dynamics.

Similarly, in Chapter 5, a perfect ring-shaped illumination would require an intensity distribution with minimal variation along the azimuthal direction. In practice, our generated ring beam exhibited noticeable intensity fluctuations around the ring, resulting in non-zero azimuthal gradient forces. These forces introduced variations in the local velocity of the colloidal structures. Our current analysis does not account for these intensity inhomogeneities, which may influence the precision of the measured dynamics.

In our experimental temperature estimations, based on phase transition methods (discussed in Chapters 4 and 5), we did not incorporate the potential influence of nearby glass substrates and colloids on the local thermal field. The proximity of such interfaces can alter heat dissipation and affect the actual temperature distribution near the colloids.

On the numerical side, our simulations assume neutral colloidal particles and do not consider electrostatic interactions, which are significant in real systems where colloids typically carry surface charges. This omission could lead to discrepancies between simulated and experimental results, especially in regimes where electrostatic forces play a dominant role.

These limitations, while not detracting from the central findings, highlight avenues for future refinement in both experimental design and modeling to achieve a more comprehensive understanding of the system.

Future directions:

In the following discussions, we outline the possibility of extrapolation based on our current studies.

Exploring Out-of-Plane Colloidal Motion

All the investigations presented in this thesis are fundamentally directed towards the controlled manipulation of colloidal structures and the exploration of emergent dynamics such as linear propulsion, rotational motion, and guided navigation along defined optical paths. A careful analysis of the experimental configurations reveals a common characteristic: the in-plane motion of the colloidal systems is primarily governed by in-plane optical gradient forces and thermo-osmotic flow-induced forces, while the vertical (out-of-plane) confinement arises due to the optical scattering force, which pushes the colloids towards the glass substrate. Consequently, the observed dynamics are predominantly restricted to two-dimensional motion near the substrate surface.

This inherent planar confinement naturally raises an important question: *can we extend the control over colloidal motion into the out-of-plane (vertical) direction?* To address this challenge, one must generate a significant vertical force component that is capable of counteracting the downward optical scattering force.

Among the potential mechanisms, the thermo-osmotic backflow field, previously illustrated in Fig. 2.4(b), emerges as a promising candidate. If this fluid flow is sufficiently strong, it can potentially drive particles away from the substrate, enabling three-dimensional motion. Additionally, the temperature-dependent variation in the thermo-diffusion constant (as discussed in Chapter 5), particularly the emergence of repulsive positive thermophoretic forces between thermally active colloids, suggests another avenue for generating out-of-plane forces.

However, both of these effects, thermo-osmotic backflow and repulsive thermophoresis, are not independently sufficient to overcome the downward scattering force in a pure Milli-Q water environment. This limitation arises primarily from two factors: (i) the colloidal structures formed in such a medium are not rigid but dynamically assembled, and (ii) the magnitude of the thermally induced forces in Milli-Q water is too weak to generate measurable out-of-plane displacement.

To enhance these forces, we turned to a well-known strategy: modifying the solvent environment by adding ionic constituents. It is well established that both thermo-osmotic and thermophoretic effects can be significantly amplified in the presence of salt ions. Motivated by this, we conducted preliminary experiments using a 5 mM NaCl aqueous solution. Remarkably, in this medium, thermally active and passive colloids spontaneously form rigid and permanent structures with varied geometries, even in the absence of any optical confinement.

When these rigid colloidal structures, comprising one passive and two thermally active colloids, are subsequently trapped using a ring-shaped optical beam, we observe distinct out-of-plane motion (see Fig. 6.1). Interestingly, the directionality of this motion depends sensitively on the relative positioning of the thermally active colloid that lies outside the

ring illumination. The interplay between the optical confinement and the spatially asymmetric heating gives rise to a net vertical force, resulting in three-dimensional motion. Moreover, the colloidal structure exhibits spontaneous transitions between two different rotational handedness, indicating the existence of bistable chiral states.

At present, this out-of-plane motion remains stochastic, with the system randomly switching between two chiral states. Ongoing efforts are focused on developing strategies to actively control this handedness, potentially by tailoring the optical field geometry or by modulating the environment properties. These results point to a rich and largely unexplored regime of colloidal manipulation in three dimensions, and open up new opportunities for designing optically driven microscale machines with complex, programmable behaviors.

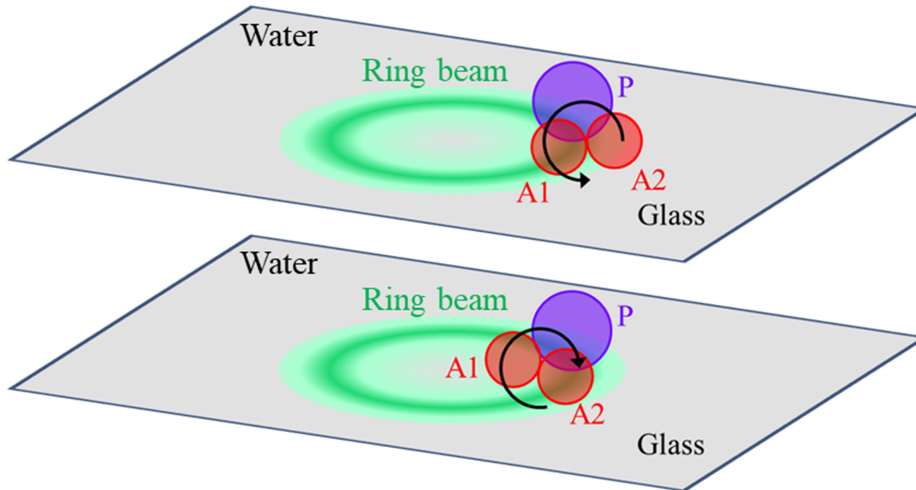


FIGURE 6.1: Schematic two out-of-plane motion mode of the colloidal structures in 5mM aqueous NaCl solution under ring optical illumination.

Study of Light Emerging from the Dynamics of Colloidal Structures

In addition to the diverse dynamic behaviors induced in colloidal systems via light-matter interaction through optothermal effects, a particularly intriguing aspect is the nature of the light emerging from these dynamically evolving structures. The scattered or transmitted light carries rich information not only about the structural configurations and motions of the colloids but also about the underlying interactions that govern their behavior.

By systematically analyzing the characteristics of the emerging light, such as its polarization, intensity distribution, it is possible to infer critical properties of the colloidal assembly. These include effective refractive indices, spatial symmetries, and anisotropies, as well as other material parameters. Thus, in this way, the light not only drives the colloidal motion via optical or opto-thermal forces but also acts as a probe of insight into the evolving dynamics of the system. This two-way interaction opens new avenues for designing self-regulating or adaptive photonic cavities. This ongoing work aims to deepen our understanding of how dynamic colloidal structures modulate the properties of light and to explore whether the emerging light can, in turn, be harnessed to further steer colloidal behavior.

To the end, we would like to say that the work presented in this thesis mainly focuses on the study of colloidal manipulation in the light of various nature of optical fields, may it be a defocused beam, a focused Gaussian beam, or even a structured ring beam, and the consequent generated optical fields. We have also extensively analyzed the dynamic feature of the motion of colloidal structures and numerically corroborated our experimental observations to find insight into the underlying driving principles. It is our ambitious hope that the findings of our studies will acquire significant interest in the community and can enrich knowledge of various light-matter interactions and colloidal manipulation.

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Optothermally Induced Active and Chiral Motion of Colloidal Structures

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Emergence of Directional Rotation in an Optothermally Activated Colloidal System

**Author:** Rahul Chand, Chaudhary Eksha Rani, Diptabrata Paul, et al**Publication:** ACS Photonics**Publisher:** American Chemical Society**Date:** Nov 1, 2023*Copyright © 2023, American Chemical Society*

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Optothermal Revolution: Colloids in an Optical Ring Trap

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