Capped colloids as light-mills in optical traps

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Abstract. Custom-designed colloidal particles in optical tweezers act as light-mills in a fluid. In particular, aqueous suspensions of capped silica colloids, in which half of the surface is covered with metal layers, are investigated. Due to their asymmetry, the capped colloids can act as rotators when exposed to intense laser fields. Particles of 4.7 µm in diameter are observed rotating around the focus of a laser beam. For low intensities, particles become trapped close to the spot of highest laser intensity. Above a threshold value of about 4 mW in total beam intensity, the particles move away from the centre of the focus and start to rotate at frequencies of about 1 Hz. The balance of forces due to light pressure and hydrodynamic forces gives a constant rotation rate. The speed of the spinning particle increases linearly with laser power to above 2 Hz until the particles are ejected from the focus for intensities higher than 7 mW. Magnetic caps introduce further possibilities to tune the rotation rates.

Contents
1. Introduction 2
2. Sample preparation and experimental set-up 2
3. Results 4
4. Discussion and comparison with similar systems 7
5. Conclusions and outlook 8
Acknowledgments 8
References 9

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1. Introduction

Laser tweezers allow microscale particles to be positioned with high precision and reproducibility. The main advantage of this technique is its versatility. It can be performed in almost any transparent medium, especially in cells containing a liquid. Thus, a large number of systems (ranging from biological samples such as cells to colloidal particles) can be manipulated using this technique. A well-defined rotation of the particles can be used to further investigate particles on the micrometre and even nanometre scale. Therefore numerous experiments have been performed during the past years in order to investigate the rotational motion of such particles. Typical approaches used the absorption of polarized light that transfers spin or angular momentum [1, 2] and refraction of the beam by custom-made objects. Rotation around the beam axis [3, 4] and perpendicular to it [5] could be studied and theoretically explained [6]. Additionally, turning the angle of polarization of optical tweezers can also lead to rotation of birefringent particles [7]. Wherever light pressure propels the motion of particles in symmetric optical traps, the rotating object needs to be asymmetric. In the system investigated here, asymmetry is introduced by coating one side of the spheres with metal caps. Visualization of Brownian motion of similar particles [8] and rotation of particles with magnetic caps in a magnetic field have already been demonstrated [9]. Accordingly, charged particles with metal caps move in an electric field [10]. More detailed studies on rotation in optical traps use biochemically joined particles as dimers [11] and pieces of glass powder [12] as rotating agents. In this study, the controlled off-axis rotation of capped colloids around a Gaussian laser spot is described.

2. Sample preparation and experimental set-up

The samples, named capped colloids, were prepared in the process illustrated in figure 1. It starts with the production of a monolayer, which is similar to the method described in [13]. A drop of suspension with 4.7 µm silica particles (SS05N from Bangs Laboratories) totally wets a thoroughly cleaned borosilicate glass cover slip. It dries in a cell at water saturation pressure, which is tilted by about 5°. Through self-assembly during evaporation of the liquid, the particles arrange in hexagonal order.

The sample is then transferred into an evaporation chamber and a coating is added. After an adhesive layer of 1–2 nm Ti, various metals such as Ni, Au or Co–Pd multilayers are deposited on the particles [14]. The final coating, added by e-beam evaporation, consists of a SiO₂ layer, which should guarantee uniform surface properties such as surface charge.

To detach the particles from the substrate, a stepper motor stage dips the cover slips into a deionized water basin under 45° at about 10 µm s⁻¹. Surface tension peels the colloidal particles off the substrates so that they float on top of the water surface [15]. After pipetting the excess fluid from the reservoir, the particles are mixed with the residual liquid. Using this recipe, they disconnect from each other and give suspensions of variable concentration in capped colloids. Alternatively, ultrasound also removes particles from the surface [16].

Finally, the suspensions are confined between two horizontal glass plates separated by an O-ring. The lower glass slip is coated with a polymethyl methacrylate (PMMA) layer to provide a smooth surface, which prevents sticking of the particles. Due to gravity, the particles sediment until Coulomb repulsion originating from surface charges stabilizes them at a certain height above the substrate.
Figure 1. Preparation of capped colloids. (a) A 30 µl drop of a 10% solids stem suspension of 4.7 µm silica particles dries on a clean cover slip. The particles arrange into a monolayer and diverse metals are evaporated on top. After dipping into a water reservoir, the layer of particles detaches from the substrate and floats on the fluid surface. Finally, stirring breaks the particle bonds and the suspension is enclosed in transparent samples cells. (b) SEM image of two 4.7 µm Si particles on borosilicate glass object slides that have been coated with 50 nm Ni, 50 nm Au and 20 nm SiO₂ layers.

Such sample cells are examined in a vertically aligned optical tweezers set-up. The light from a frequency doubled Nd : YVO₄ laser (λ = 532 nm) is collimated through a 20× objective placed directly above the sample cell. This leads to an opening angle of about 25° and a spotsizes for the laser focus of about 25 µm² in the centre plane of the particles. When one of the particles is illuminated by this focused Gaussian beam, light forces can trap it in the horizontal plane, while in the vertical direction it is pushed somewhat closer to the repulsive substrate.

The colloidal spheres are imaged with an inverted video microscope on to a CCD camera, and particle positions are determined with a rate of 1 Hz and a lateral accuracy of 100 nm. From these data we obtain particle positions and the relative orientations of the caps. Rotation rates were determined with a photodiode mounted at one side of the sample cell by recording the sequence of light pulses.
Figure 2. Counterclockwise and clockwise rotation of capped colloids around a laser focus. (a) 4.7 µm silica colloids with a 50 nm Au and 20 nm SiO₂ cap rotate counterclockwise around the focus of a laser beam of 5.44 mW in an aqueous suspension. The inner cap surface reflects the light of the optical trap similar to a mirror reflecting a signal fire in a light-house. A movie is available. (b) Filtering out most of the laser light at 4.25 mW for a clockwise rotating particle reveals that it is held eccentrically. The transparent half without cap points into the direction of motion. In both cases, the time difference between successive pictures is approx. 0.3 s. (c) The orientation of a capped colloid in the optical tweezers is visualized at two different laser powers.

3. Results

The orientation and rotational motion of capped colloids are clearly visible. This allows us to characterize the system in a controlled and precise way.

Figure 2 depicts capped colloids rotating in the optical trap. The laser beam driving the motion of the particles is reflected by the inner and outer cap surfaces. The light-house effect results from this reflection at the cap. It becomes visible in the images because of the scattering of the reflected light from the substrate surface. Figure 2(a) and a movie in the supporting material visualize this effect. In order to determine the orientation of the cap in greater detail, the intensity of the laser light in front of the camera is reduced. This reveals that the motion of the particle is eccentric. It moves on a circle with a diameter of about 1.5 µm around the focal point. Although the exact orientation of the cap is somewhat difficult to determine, it is inferred from the images that its edge aligns vertically. So the plane separating the coated and uncoated parts lies perpendicular to the substrate. Tilting a cap away from this position (as it can
Figure 3. Onset and frequency of rotation. Below a threshold of 4 mW of total laser power in the optical tweezers 50 nm Au and 20 nm SiO$_2$ capped colloids can be trapped. Above this rotation sets in, until for intensities higher than 7.5 mW the light pressure expels the particles from the laser focus.

be achieved for particles with a magnetic cap in an external magnetic field, see below) lowers the rotation rate until the particle stops. There is no preference either for rotation in clockwise or counterclockwise directions and jumps between them occur spontaneously.

To induce the rotation of the particles, a threshold value in laser power needs to be overcome. Below this intensity, the capped particles can be trapped and moved, but do not rotate. As shown in figure 3, above this value the rotation rate increases almost linearly with the laser power. Each individual particle shows a monotonous increase in rotation rate with laser power. The errorbars indicate the distribution of the frequencies for an ensemble of particles. The standard deviation in rotation rate for one specific capped colloid is approximately the symbol size. At a certain point (here $\approx 7.5$ mW), the light pressure is high enough to expel particles from the laser focus.

In further investigations, we have studied the influence of the polarization of the incident light, since one possible origin of the particle rotation might be the transfer of spin angular momentum of the laser photons [1, 7]. The phenomenon, however, turned out to be independent of the linear direction of the polarization. The particle still rotated although all circularly polarized light has been filtered out. Hence, this explanation can be ruled out.

In another sequence of measurements, we have determined the positions of the particle centres relative to the centre of the laser tweezers. After the position of the laser focus had been fixed using a picture without a particle, colloids and caps were located in images resulting after a threshold in brightness level had been applied. Dark areas corresponding to particles in the pictures were identified and located by edge detection. The position of the cap was found as the barycentre of the dark region. With this information, the normal vector of the caps could also be determined. It was given by the relative position of the particle centre and the barycentre of the cap. For each particle, the distance to the beam centre was determined. Finally, the relative
coordinates were converted into the coordinate system of the capped colloid. The resulting distribution of focal positions is depicted in figures 4(c) and (f).

In order to illustrate the bistability of the system in more detail, we have derived effective potentials for the particles in the various regimes. We assumed that the distribution of the particle positions in the potential landscape follows Boltzmann’s equation (which, however, could be a rather crude approximation considering that the role of dissipation in our system still needs to be clarified). For this purpose, first the particle positions were converted into polar coordinates.
The azimuthal angle was dropped and only the radial dependence of the effective potential was retained. In the next step, the occurrences were sorted into 19 bins up to the maximum distance between the laser focus and the particle centre. With our assumption, the probability to find a particle at a certain distance from the centre should depend on the potential energy as $p(r) = e^{-\Phi(r)/kT}$. This gives an average potential of $\Phi(r) = -\ln(p(r))$ in units of $kT$. For uncapped particles, a similarly derived potential has a minimum at the origin and increases with radius as shown in [17]. For capped particles, the pivotal minimum in the radially averaged potential shifts to values around 0.5 $\mu$m as seen in figure 4(b). Here, particles are simply held in the tweezers. The potential further develops a second minimum for higher laser powers as in figure 4(e). For increasing intensity, the dip at a distance of 1.5 $\mu$m from the centre of the laser focus becomes more and more pronounced. When the particles are trapped in this state, they rotate around the focus.

The orientation vectors for the caps always have a large component against the direction of the motion. Brownian motion can lead to spontaneous changes in direction from clockwise to counterclockwise or vice versa. In the reference frame of the particles in figure 4(f), this corresponds to a position of the focus, which is more to the left or more to the right of the centre, respectively. The points close to the middle correspond to usual tweezing as in the case of lower intensity in figure 4(c). The ring-like distribution of the dots is due to the limited resolution of the camera, which leads to an artificial quantization of the relative positions of the particles.

Another approach to find out more about the behaviour of capped colloids in optical traps is to use magnetic caps, e.g. Ni or CoPd multilayers [14]. In this case, an external magnetic field can be used to manipulate the particles in addition to the laser tweezers. Without a magnetic field being applied, the magnetic colloids align and rotate in the optical trap like the nonmagnetic particles described above.

When a magnetic cap is, however, tilted in the beam direction by an external field from an electromagnetic coil, the rotation slows down with increasing magnetic field strength. (The coil has an inner diameter of 4.5 cm and is located 2–7 cm below the sample. It induces a magnetic field up to 20 mT perpendicular to the substrate inside the cells.)

Small magnetically stabilized clusters or short chains of particles can be rotated as well. Often CoPd-covered particles form ensembles of three particles so that their caps are pointing towards their centre. Such clusters with a larger hydrodynamic radius rotate more slowly than individual particles. The reflectivity of the caps can be tuned by layer depth and choice of material. A minimum layer depth of about 20 nm of metal coating is needed to obtain a significantly reduced transmissivity of the capped side. Particles with thinner caps do not rotate.

4. Discussion and comparison with similar systems

From a comparison with experiments on specially designed particle dimers in [11], it is assumed that light pressure drives the system. A strict theoretical approach, however, also needs to address the role of dissipation and local heating in the system. Their contributions are hard to estimate, but might be necessary to understand its nonlinearity.

In any case, employing capped colloids as rotators simplifies the geometry of the scattering object. For capped colloids, several forces contribute to the proposed rotation mechanism. Due to the scattering force, their transparent part is drawn into the focus of the laser tweezers. On the other hand, the momentum transfer from internal and external reflections at the coated surface
pushes the cap. Both components balance at an equilibrium distance from the beam centre. This leads to the second minimum in the effective potentials.

Furthermore, a torque on the particle results because of the asymmetric orientation of the cap towards the beam axis. Therefore, the angular velocity increases until it is balanced by hydrodynamic forces. Due to this asymmetric position, higher torques can be exerted compared to particles rotating about an axis through their centre. To estimate the net torque from the viscous drag on the particle, simplifications in analogy to [11] yield a value of about $2.4 \times 10^{-18}$ Nm.

The rotation of capped colloids is robust for various choices of inner cap material (Al, Au and thin layers of C), given that Au and SiO always cover the outer surface. This supports the intuitive conclusion in [11] that the light pressure due to reflection at the outside shell drives the motion.

During rotation, irregularities in the particle motion occur. Since Brownian motion leads to a change in the direction of rotation of the particles, it might also generate fluctuations in the rotation rates. For constant laser intensity, it is expected that smaller particles will rotate faster at fixed laser intensity, as is reported for chunks of glass powder [12]. Contrary to capped colloids, however, the size and shape of such irregular particles is hard to characterize.

The manipulation of rotating capped colloids with a magnetic field shows that indeed the scattering geometry determines the rotation rate. It can be used to adjust the orientation of the particles during rotation. The more the cap is tilted, the slower the particle rotates.

5. Conclusions and outlook

In summary, capped colloids rotate around the focus of optical tweezers with tunable angular velocity. Above a threshold, the particles rotate eccentrically with a rotation rate proportional to the laser intensity. Rotators can be fabricated by floating the capped particles off the substrates after evaporation of the metal layers. This prevents damage due to ultrasound and may facilitate treatments such as biofunctionalization. The rotators can be moved around and actuated by a simple optical trap.

Light pressure pushes the particle in an equilibrium distance from the beam centre and propels the motion. The contribution of heat transfer, however, is not yet clear. Effective potentials are derived from the relative positions of the trap and the particle. They visualize the transition from normal tweezing to rotation.

This design should allow one to exert higher torques for pumps, motors or drills on the micrometre scale in a fluid. Making use of the magnetic properties of the particles may become a further step towards micro-machines. A feedback loop for the laser power could be used to further control the rotation speed as in [12]. Additionally, a magnetic field can set the orientation of magnetic caps.

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