Optical forces and torques on asymmetric nanoscale core-shell particles

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Abstract

The optical trapping and manipulation of small particles is an important tool for probing fluid properties at the microscale. In particular, micro rheology exploits the manipulation and rotation of micron-scale particles to probe local viscosity, especially where these properties may be perturbed as a function of their local environment, for example in the vicinity of cells. To this end, birefringent particles are useful as they can be readily controlled using optically induced forces and torques, and thereby used to probe their local environment. However the magnitude of optical torques that can be induced in birefringent particles is small, and a function of the particle diameter, meaning that rotational flow cannot readily be probed on length scales much smaller than the micron level. Here we show modelling that demonstrates that asymmetric, spherical core-shell nanoparticles can be used to generate considerable optical torques. The asymmetry is a result of the displacement of the centre of the core from the shell. Our results show that, for particles ranging from 90 nm to 180 nm in diameter, we may achieve rotation rates exceeding 800 Hz. This fills a missing size gap in the rotation of microparticles with optical forces. The diameter of particle we may rotate is almost an order of magnitude smaller than the smallest birefringent particles that have been successfully rotated to date. The rotation of asymmetric core-shell nanoparticles therefore offers a pathway to nanorheology.

1 Introduction

The field of optical trapping, initiated by Arthur Ashkin [4] received the Nobel Prize in physics in 2018. The topic area describes the control and manipulation of mesoscale particles using the momentum of light. The added feature of including light fields with spin or orbital angular momentum has fuelled the topic of rotating as well as translating trapped particles. It has been widely applied across the sciences and has made particular impacts in areas spanning fundamental physics [5, 34] to biological sciences [7, 6, 40, 24, 28, 13, 30]. This opens up the prospect of new studies including those in microfluidics, namely cellular microrheology and rotational dynamics for the burgeoning area of levitated optomechanics.

From the fundamental physics standpoint, the area of levitated optomechanics has emerged as a powerful way to explore the boundary between classical and quantum physics with mesoscopic particles well isolated from their environs [10, 18, 26, 27]. In turn, this has led to the recent demonstration of cooling of a particle to the quantum ground state [14]. In other work, the motion of trapped particles has elucidated fundamental concepts for both the linear and angular momentum of light by using a trapped particle as a probe of the incident field [31, 35, 16].

In the area of biological science, the use of optical traps as calibrated force transducers has led to the measurement of exquisite, minuscule forces associated with a range of linear and rotary molecular motors [3]. However the challenge of understanding how cells respond to their environment requires probing of viscosity at the nanoscale, and this length scale is not yet accessible through the transfer of sufficient optical torque to induce rotation of nanoparticles. Currently, birefringent particles are mostly used as the optical rotation probes for local rheology measurements. However, given the small difference of the refractive indices between the optical paths of birefringent particles and optical torque in proportion to \(a^3 \sim d^6\) [22] with \(a\) being the characteristic size of the particle, the optical torque becomes negligible as the size of the birefringent particle reduces, especially down to sub-micron scale. Smaller nano-scale probes of viscosity should unlock cellular environmental responses as
they should be taken up more readily in cells, and may perform as minimally invasive probes of the intracellular environment.

From the material science point of view, there has been considerable development of nanoparticles with complex and bespoke compositions including metallic particles or high refractive index materials. Such more complex nanoparticles offer great promise for novel applications, and the emergence of new synthesis capabilities at the mesoscale are becoming more accessible. In this domain core-shell nanoparticles [11, 17, 15] can be utilised to develop novel materials to perform multiple tasks and functions.

Here we investigate the optical forces and torques acting on non-concentric asymmetrical core-shell particles. This is the first study of its type and opens up a hitherto unrecognised area of exerting optical torques and initiating rotation for particles of very small size. Although the use of spin angular momentum is already a powerful tool, standard techniques rely on a polarisation change through the trapped object, which can be prohibitively small for particles below a micron in diameter. Accordingly, rotation studies have been performed with larger particles [2]. These studies include viscosity measurements. Conversely, as we show, asymmetric core-shell objects allows rotation for objects of sizes 50-500nm in diameter. This is crucial for future studies in cellular media that may not be able to take up larger objects, for example.

To date, the majority of studies on optomechanical response of particles trapped by light has focused on particles that comprise of a single homogenous (typically dielectric) material. By tailoring the material property of the particle new modalities may be envisaged that cannot be readily achieved using shaped light alone with dielectric objects [34]. A key example is combining the advantage of core-shell particle and optical trapping, which can open up a promising direction of nanotechnology for scientific, engineering, biological and medical applications. For instance, Jannasch et al [19] enhanced optical forces to the nanonewton level by coating a titania particle with a silica shell, which is useful for biological studies [13]. Spadaro et al [33] studied how the relative thickness between the core and shell can affect the optical force on a Au–PEG core–shell particle. Ali et al [1] investigated the effects of the chirality of the core-shell particle on the optical torque under a circularly polarised beam. To date, only concentric core-shell nanoparticles have been considered. As we show, breaking centro-symmetry in such systems leads to increased optical torques.

We explore linearly polarised Gaussian illumination on three types off-concentric core-shell particles at the sub-micro scale: a gold nanoparticle coated with a silica shell (Au@SiO$_2$) [21, 23], a titanium dioxide nanoparticle with a silica shell (TiO$_2@$SiO$_2$) [12] and a silicon dioxide particle with a titanium dioxide shell (SiO$_2@$TiO$_2$) [32]. These are chosen to represent prospective nanoparticles particles that may be fabricated with asymmetric cores. We show that the optomechanical response of an asymmetric core-shell particle is richer than for a symmetric particle even under a linearly polarised beam, in particular due to the exertion of optical torques, as demonstrated in Sec. 3. This optomechanical response of such an asymmetric core-shell particle can expand its remit in optical traps, namely inducing appreciable rotation rates in nanometre-sized spherical particles which opens up the prospect for nanorheology, which is the key motivation of this study.

2 Asymmetric core-shell particle with Gaussian linearly polarised illumination

We consider particles where both the core and shell are spherical, and where the centers do not align, as shown in Fig. 2 (a).

To induce optical effects, we model a trapping beam with Gaussian profile and linear polarisation. The focal position of the trapping beam is set at the origin, and the beam propagates along the $z$-axis with its electric field linearly polarising along the $x$-axis, as shown in Fig. 2 (b). The centre of the particle’s shell is fixed at the origin (the focal point of the beam), and the core-centre is varied on the $xz$ plane. The distance between the core-centre and shell-center is denoted $h$, and the angle from the line connecting core-center and shell-center to the electric field polarisation direction ($x$ axis) is $\theta$, so that when $\theta = 90^\circ$ the centre line measured from the shell-centre to the core-centre is along the beam propagation direction and when $\theta = 270^\circ$ it is opposite to the beam propagation direction.

When the beam is not highly focused, its Gaussian profile and the corresponding incoming electric and magnetic fields, $E^{inc}$ and $H^{inc}$, are well described by the approximate expressions given by Barton and Alexander [8]. Assuming a linearly polarised single colour Gaussian beam propagating along the $z$ direction with its focal point locating at the origin in a source-free homogeneous medium with relative permittivity $\epsilon$ and
Figure 1: (a) Sketch of the geometric composition of the off-concentric core-shell particle; (b) The electric field magnitude of the electric field of the Gaussian beam on the $xz$ plane. The beam propagates along $z$-axis and its electric field is linearly polarised along the $x$-axis.

The scattered fields, $E_{\text{sca}}$ and $H_{\text{sca}}$, and the transmitted fields in the particles are calculated by using the robust field only surface integral method [37, 20, 39, 38], which is ideally suited to calculating the electric and magnetic fields efficiently and accurately on particle surfaces.
Table 1: Refractive indices $n$ and extinction coefficients $k$ of Au [29], SiO$_2$ [25] and TiO$_2$ [9]

|       | $n + ik$                                      |
|-------|----------------------------------------------|
|       | $\lambda$=532 nm  | $\lambda$=775 nm  | $\lambda$=840 nm  | $\lambda$=1064 nm |
| Au    | 0.54+12.14      | 0.18+i4.51        | 0.20+i5.02        | 0.31+i6.63        |
| SiO$_2$ | 1.46            | 1.45              | 1.45              | 1.45              |
| TiO$_2$ | 2.17            | 2.10              | 2.09              | 2.07              |

By obtaining the surface electric and magnetic fields accurately, we then calculate the optical force by integrating the time-average Maxwell’s stress tensor over the surface of the shell, $S_{\text{shell}}$, as

$$F_i = \int_{S_{\text{shell}}} \frac{1}{2} \left\{ \text{Real} \left[ (D_i E_j^* + E_i D_j^* + B_i H_j^* + H_i B_j^*) n_j \right] - (D_j E_i^* + B_j H_i^*) n_i \right\} dS.$$  \hspace{1cm} (4)

In Eq. (4), subscript $i = 1, 2, 3$ is the $i$th component of the vector field, subscript $j = 1, 2, 3$ is the $j$th component of the vector field, superscript * indicates the conjugate of the field. Also, $F_i$, $E_i$, $D_i$, $H_i$, $B_i$ are the optical force, electric field, displacement, magnetic field, and magnetic displacement, respectively. $n$ is the refractive index of the surrounding medium.

The optical torque can be calculated by

$$N_i = \int_{S_{\text{shell}}} \varepsilon_{ijk} r_j^* \frac{1}{2} \left\{ \text{Real} \left[ (D_k E_l^* + E_k D_l^* + B_k H_l^* + H_k B_l^*) n_l \right] - (D_l E_k^* + B_l H_k^*) n_k \right\} dS.$$  \hspace{1cm} (5)

where subscript $l = 1, 2, 3$ is the $l$th component of the vector field, subscript $k = 1, 2, 3$ is the $k$th component of the vector field, $\varepsilon_{ijk}$ is the Levi-Civita symbol, and $r_j^*$ is the location vector of a point on the shell surface relative to the centre of mass of the core-shell particle. The centre of mass of the core-shell particle is on the line between the centre of the core and that of the shell which distance to the centre of the shell, $h_c$, can be calculated by

$$h_c = h (\rho_{\text{core}} - \rho_{\text{shell}}) / (\rho_{\text{core}} + \rho_{\text{shell}})$$

where $\rho_{\text{core}}$ is the density of the core and $\rho_{\text{shell}}$ is the density of the shell.

3 Results

We study the optical forces and torques acting on three types of off-concentric core-shell particles, namely Au@SiO$_2$, TiO$_2$@SiO$_2$ and SiO$_2$@TiO$_2$. The refractive indices and extinction coefficients of Au, SiO$_2$ and TiO$_2$ are listed in Table 1 and their densities are $\rho_{\text{Au}} = 19.30$ g/cm$^3$, $\rho_{\text{SiO}_2} = 2.50$ g/cm$^3$ and $\rho_{\text{TiO}_2} = 4.23$ g/cm$^3$, respectively. As the centre of the shell is located at the focal point of the beam, we can concentrate on how the orientation of the core particle will affect the optical trapping of the off-concentric core-shell particle.

Fig. 2(a, b) present the optical trapping force along the wave propagation direction. Taking Au@SiO$_2$ with shell radius $a_{\text{SiO}_2} = 90$ nm and $a_{\text{Au}} = 60$ nm as an example, when the particle is trapped in air with $n_{\text{air}} = 1.0$ by a Gaussian beam with wavelength $\lambda = 532$ nm and beam waist radius $w_0 = 1 \mu$m, we can see from Fig. 2(a) that as the distance between the Au core and the SiO$_2$ shell, $b$ increases, the optical force, $F_z$, along the wave propagation becomes larger and larger with all orientation angle $\theta$. It is noticeable that the force curves have two local minima when the orientation angle $\theta = 90^\circ$ and $\theta = 270^\circ$, respectively, where $F_z$ is larger at $\theta = 90^\circ$ compared to that at $\theta = 270^\circ$. When the surrounding medium is water with $n_{\text{water}} = 1.33$, the variations of the optical forces, $F_z$ due to the asymmetry of the Au core change accordingly because of the difference of the ratio of the relative refractive indices between the SiO$_2$ shell and the surrounding medium. One obvious difference is that $\theta = 90^\circ$ becomes the orientation of Au core corresponding to the maximum optical force along the wave propagation as shown in Fig. 2(b). Converting the magnitude of the optical trapping force, $F_z$, in Fig. 2(b), we obtain the $Q$-factor, $Q = F_z c / (n_{\text{medium}} P_0)$ larger than 0.029 which is reasonable.

As the core particle is not concentric with the centre of the shell, there will be an optical force perpendicular to the beam propagation direction, as presented in Fig. 2(c, d). It is intuitive to predict that the maximum
value of this optical force, $F_x$ appears when the core particle is at the maximum displacement from the beam axis at $\theta = 0^\circ$ and $\theta = 180^\circ$ which is demonstrated in Fig. 2(d) for the surrounding medium as water. If the surrounding medium is air, the orientation of the Au core corresponding to the largest value of $F_x$ is shifted a little away from $\theta = 0^\circ$ or $\theta = 180^\circ$ as displayed in 2(c). This is because the change of the refractive index of the surrounding medium tuned the scattered field that consequently affects the optical force, $F_x$. Nevertheless, compared to the optical force along the beam propagation axis $F_z$, $F_x$ can be deemed as a secondary effect since the magnitude of $F_x$ is 2 orders lower than that of $F_z$.

In Fig. 3, we show the optical torques acting on three types of off-concentric core-shell particles embedded in water under a linearly polarised Gaussian beam with a few different wavelengths when fixed beam waist radius $w_0 = 1 \, \mu m$. We chose readily available standard wavelengths of $\lambda = 532 \, \text{nm}$, $\lambda = 775 \, \text{nm}$, $\lambda = 840 \, \text{nm}$, and $\lambda = 1064 \, \text{nm}$. Also, these wavelengths avoid the consequence of surface plasmon resonance for Au@SiO\textsubscript{2} in water with $a_{\text{SiO}_2} = 90 \, \text{nm}$ and $a_{\text{Au}} = 60 \, \text{nm}$ that happens at wavelength around 640 nm.

We can see, from Fig. 3, that as the wavelength becomes longer, the magnitude of the optical torque decreases. Also, the optical torque on Au@SiO\textsubscript{2} is nearly one order of magnitude higher than that on TiO\textsubscript{2}@SiO\textsubscript{2} or SiO\textsubscript{2}@TiO\textsubscript{2}. This is mainly because the difference of the refractive index between Au and SiO\textsubscript{2} is higher than that between TiO\textsubscript{2} and SiO\textsubscript{2} (see Table 1). Also, the trend of optical torque along with the orientation of the core particle, $\theta$ on TiO\textsubscript{2}@SiO\textsubscript{2} with $n_{\text{core}} > n_{\text{shell}} > n_{\text{medium}}$ is opposite to that on SiO\textsubscript{2}@TiO\textsubscript{2} with $n_{\text{shell}} > n_{\text{core}} > n_{\text{medium}}$, as shown in the second and third columns of Fig. 3. This is another indicator of how the material refractive index can affect the optomechanical response of an off-concentric core-shell particle. Similar to the optical force perpendicular to the beam direction, the maximum optical torques appear at $\theta = 0^\circ$ and $\theta = 180^\circ$ for the core-shell particle with shell radius 90 nm and core radius 60 nm. For Au@SiO\textsubscript{2} and TiO\textsubscript{2}@SiO\textsubscript{2} off-concentric core-shell particles, when $\theta = 0^\circ$, the optical torque will rotate the off-concentric particle counter-clockwise if we define that the beam propagation direction is pointing at 12 o’clock, and when $\theta = 180^\circ$ the optical torque will rotate them clockwise. However, for SiO\textsubscript{2}@TiO\textsubscript{2}, the direction of the optical torque flips relative to Au@SiO\textsubscript{2} or TiO\textsubscript{2}@SiO\textsubscript{2}.

As expected, the optical torque increases as the core particle is positioned further away from the centre of
Figure 3: Optical torque acting on three types of off-concentric core-shell particles in water with $n_{\text{water}} = 1.33$ under the linearly polarised Gaussian beam illumination with different wavelength $\lambda$ which beam waist radius is fixed as $w_0 = 1 \mu m$.

In the first two rows of Fig 3, we demonstrated the optical torques on Au@SiO$_2$, TiO$_2$@SiO$_2$ and SiO$_2$@TiO$_2$ with $h = 25$ nm (top row) for high asymmetry and $h = 10$ nm (middle row) for moderate asymmetry when the shell radius is 90 nm and the core radius 60 nm. The optical torques with $h = 25$ nm are more than twice those with $h = 10$ nm. By using the Stokes drag of a sphere in fluid, $N_y = 8\pi\mu a^3 \Omega$ where $\mu$ is the viscosity of water and $\Omega$ is the particle rotation frequency, we can estimate the possible largest rotation frequency of the off-concentric core-shell particles under linearly polarised Gaussian beam illumination with different wavelength $\lambda$ when the beam power is $P_0 = 20$ mW. We observe that even for small particle sizes where the radius is 90 nm (diameter of 180 nm), the Au@SiO$_2$ off-concentric core-shell
Table 2: Potential maximum rotation frequency \( \Omega \) (in Hz) for three types of off-concentric core-shell particles suspended in water \( (n = 1.33) \) and illuminated with a linearly polarised Gaussian beam at different wavelengths, \( \lambda \). The beam waist radius is \( 1 \mu m \) and beam power is \( P_0 = 20 \text{ mW} \) in all cases.

| Particle Type | \( a_{\text{shell}} = 90 \text{ nm} \) | \( a_{\text{core}} = 60 \text{ nm} \) | \( a_{\text{shell}} = 45 \text{ nm} \) | \( a_{\text{core}} = 30 \text{ nm} \) |
|---------------|-----------------|-----------------|-----------------|-----------------|
| Au@SiO\(_2\)  | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) |
| TiO\(_2\)@SiO\(_2\) | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) |
| SiO\(_2\)@TiO\(_2\) | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) | \( h = 25 \text{ nm} \) | \( h = 10 \text{ nm} \) |
| \( \lambda = 532 \text{ nm} \) | 807 | 327 | 130 | 53 |
| \( \lambda = 775 \text{ nm} \) | 407 | 169 | 28 | 11 |
| \( \lambda = 840 \text{ nm} \) | 234 | 95 | 20 | 8 |
| \( \lambda = 1064 \text{ nm} \) | 61 | 22 | 7 | 3 |

A particle can be rotated at frequencies of a few hundred Hz with 532 nm. For \( \lambda = 1064 \text{ nm} \), the rotation frequency of the Au@SiO\(_2\) can reach up to 61 Hz with \( h = 25 \text{ nm} \) and 22 Hz with \( h = 10 \text{ nm} \). As to TiO\(_2\)@SiO\(_2\) and SiO\(_2\)@TiO\(_2\) off-concentric core-shell particles, the potential rotation frequencies can still range from a few Hz to 50 Hz. Our results are summarised in Table 2.

If the particle size is reduced by half, for instance \( a_{\text{shell}} = 45 \text{ nm} \) and \( a_{\text{core}} = 30 \text{ nm} \), the optical torques on the off-concentric core-shell particles are still observable, as shown in Fig. 3(g,h,i). Note that in Fig. 3(g), we did not show the optical torque with \( \lambda = 532 \text{ nm} \) since that wavelength is close to the surface plasmon resonance wavelength that is around 550 nm for Au@SiO\(_2\) core-shell particle in water when \( a_{\text{SiO\(_2\}}} \) = 45 nm and \( a_{\text{Au}} = 30 \text{ nm} \). Based on the Stokes drag, the rotation frequency of such a small particle with diameter less than 100 nm can be a few Hz when the beam power is only 20 mW, as displayed in Table 2. This result is significant for practical applications, in particular biological applications, as particle sizes below 100 nm are potentially more bio-compatible than larger particles. Furthermore, using such low powers for optical rotation should reduce potential issues associated with photo toxicity.

To confirm that the rotation of an off-concentric core-shell particle is an observable phenomenon, we studied the light power scattered by the particle, \( P_{\text{sca}} \) from the side of the light beam. The scattered light is collected by a circular objective with radius of 3.75 mm with centre located at (0.13 mm, 0, 0), as shown in Fig. 2(b) (indicated by the eye). The power obtained by the objective can be calculated by integrating the time-average Poynting vector over the objective as \( P_{\text{sca}} = \frac{1}{2} \int_S \text{Re} [E_{\text{sca}} \times (H_{\text{sca}})^*] dS \). Fig. 4 presents the light power (photon counts) at the far field perpendicular to the beam propagation when an off-concentric core-shell particle with \( a_{\text{shell}} = 90 \text{ nm} \) and \( a_{\text{core}} = 60 \text{ nm} \) trapped by a Gaussian beam with wavelength of 532 nm and beam waist radius of 1 \( \mu \text{m} \). From this figure, we can see that the scattered light power, \( P_{\text{sca}} \) varies along with the orientation of the core, which can be used to monitor the rotation of the off-concentric core-shell particles in practical experiments.

Figure 4: The scattered power along with the orientation of the core when \( \lambda = 532 \text{ nm} \) that is collected by an objective facing the beam polarisation direction. The objective is a circular plane with radius of 3.75 mm which centre is located at (0.13 mm, 0, 0).
4 Conclusion

We numerically studied the optomechanical response of an off-concentric core-shell particle with sub-micro size under a linearly polarised Gaussian illumination with fixed beam waist radius $w_0 = 1 \, \mu m$ (more detailed results can found in the supplementary material [36]). We considered three types of core-shell particles: Au@SiO$_2$, TiO$_2$@SiO$_2$ and SiO$_2$@TiO$_2$ together with different optical trapping light beam wavelengths: $\lambda = 532 \, nm$, $\lambda = 775 \, nm$, $\lambda = 840 \, nm$ and $\lambda = 1064 \, nm$. One exciting observation is that such an off-concentric spherical core-shell particle with diameter of 180 nm (Au@SiO$_2$) can be rotated at over 800 Hz under a moderate illumination power of 20 mW. For the same power, a 90 nm diameter particle (SiO$_2$@TiO$_2$) can be rotated at over 10 Hz. Optical rotation of such a small particle with such moderate light powers is not achievable with typically used birefringent particles. This indicates that the use of asymmetric sub-micro scale core-shell particles will find use in fundamental and applied studies in optical trapping where researchers seek rotation of nanometric sized objects. This could include the areas of levitated optomechanics and biophotonics. In this latter area, such particles will enable the local measurement of nano-viscosity in complex fluids which is of great importance in understanding how cells respond to stimuli from their surrounding environment.

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The detailed optomechanical response of an asymmetric spherical core-shell particle under the Gaussian illumination with linear polarisation that is described in Sec. 2 of the main text is given in this document. The asymmetry of the particle is introduced by the displacement between the core-centre and shell centre. We studied the optical forces and torques acting on an off-concentric core-shell particle with the shell radius $a_{\text{shell}} = 90 \, \text{nm}$ and core radius $a_{\text{core}} = 60 \, \text{nm}$ which shell centre is at the focal point of the Gaussian beam. The beam waist radius is set as $w_0 = 1 \, \mu\text{m}$ when the beam propagates along $z$-axis and polarises along $x$-axis. The distance between the core-centre and shell-center is $h$, and the angle from the line connecting core-center and shell-center to the $x$ axis is $\theta$, so that when $\theta = 90^\circ$ the centre line measured from the shell-centre to the core-centre is along the beam propagation direction.

It is intuitive that an off-concentric core-shell particle can tune the electric and magnetic fields when the orientation of its core changes. Take an asymmetric Au@SiO$_2$ particle as an example, when it is under the illumination of a linearly polarised Gaussian beam in air, by using the computational model detailed in Sec. 2 in the main text, Fig. 1 shows the total electric and magnetic fields in the surrounding medium and the transmitted electric and magnetic fields in the particles as the centre of Au core locates at different position relative to the centre of the SiO$_2$ shell. The wavelength of the beam is 532 nm, and the displacement between the centre of the Au core and the centre of the SiO$_2$ shell, $h = 25 \, \text{nm}$. The refractive indices of the composition materials of the core-shell particle are listed in Table 1 in the main text. The contour plots of the $x$ component of the electric field in the top row of Fig. 1 indicate that we will expect a force perpendicular to the wave propagation. Also, under a propagating wave, if the particle size is not much perpendicular when compared to the light wavelength, the effects of phase across the particle can be observed.
Figure 1: Electric and magnetic fields of when a linearly polarised Gaussian beam is incident on an off-concentric Au@SiO$_2$ core-shell particle in air. The wavelength of the beam is 532 nm and its waist radius is 1 $\mu$m. The radius of the Au core is 60 nm, that of the SiO$_2$ shell is 90 nm, and the displacement between the centre of the Au core and the centre of the SiO$_2$ shell, $h = 25$ nm. The refractive indices of air, SiO$_2$ and Au are $n_{\text{air}} = 1.0$, $n_{\text{SiO}_2} = 1.46$ and $n_{\text{Au}} = 0.54 + i2.14$, respectively. (a) - (c) The $x$ component of the electric field; (d) - (f) The $z$ component of the electric field; (g) - (i) The $y$ component of the magnetic field at different orientation of the Au core.
Together with the asymmetry given by the off-concentric core-shell allocation, a torque acting on this particle will appear as shown by the contour plots of the $z$ component of the electric field and the $y$ component of the magnetic field in the middle and bottom rows of Fig. 1.

The total electric and magnetic fields in the surrounding medium are the superposition of the incident field and the scattered field as $E = E^{\text{inc}} + E^{\text{sca}}$ and $H = H^{\text{inc}} + H^{\text{sca}}$. Introducing the above relationships into Eqs. (4) and (5) in the main text, we notice that the optical force and torque have three parts: one from the incident field, one from the scattered field, and one from the interactions between the scattered and incident fields as

$$F^{\text{inc}}_i = \int_{S_{\text{shell}}} \frac{1}{2} \left\{ \text{Real} \left[ (D_{ij}^{\text{inc}} E_j^{\text{inc},*} + E_j^{\text{inc}} D_j^{\text{inc},*} + B_j^{\text{inc}} H_j^{\text{inc},*} + H_j^{\text{inc}} B_j^{\text{inc},*}) n_j - (D_j^{\text{inc}} E_j^{\text{inc},*} + B_j^{\text{inc}} H_j^{\text{inc},*}) n_i \right] \right\} dS$$

$$F^{\text{sca}}_i = \int_{S_{\text{shell}}} \frac{1}{2} \left\{ \text{Real} \left[ (D_{ij}^{\text{sca}} E_j^{\text{sca},*} + E_j^{\text{sca}} D_j^{\text{sca},*} + B_j^{\text{sca}} H_j^{\text{sca},*} + H_j^{\text{sca}} B_j^{\text{sca},*}) n_j - (D_j^{\text{sca}} E_j^{\text{sca},*} + B_j^{\text{sca}} H_j^{\text{sca},*}) n_i \right] \right\} dS$$

$$F^{\text{ext}}_i = \int_{S_{\text{shell}}} \frac{1}{2} \left\{ \text{Real} \left[ (D_{ij}^{\text{ext}} E_j^{\text{ext},*} + E_j^{\text{ext}} D_j^{\text{ext},*} + B_j^{\text{ext}} H_j^{\text{ext},*} + H_j^{\text{ext}} B_j^{\text{ext},*}) n_j + (D_j^{\text{ext}} E_j^{\text{ext},*} + E_j^{\text{ext}} D_j^{\text{ext},*} + B_j^{\text{ext}} H_j^{\text{ext},*} + H_j^{\text{ext}} B_j^{\text{ext},*}) n_j - (D_j^{\text{ext}} E_j^{\text{ext},*} + B_j^{\text{ext}} H_j^{\text{ext},*}) n_i - (D_j^{\text{ext}} E_j^{\text{ext},*} + B_j^{\text{ext}} H_j^{\text{ext},*}) n_i \right] \right\} dS$$

$$N^{\text{inc}}_i = \int_{S_{\text{shell}}} \varepsilon_{ijk} \frac{1}{2} \left\{ \text{Real} \left[ (D_{jk}^{\text{inc}} E_k^{\text{inc},*} + E_k^{\text{inc}} D_k^{\text{inc},*} + B_k^{\text{inc}} H_k^{\text{inc},*} + H_k^{\text{inc}} B_k^{\text{inc},*}) n_l - (D_k^{\text{inc}} E_k^{\text{inc},*} + B_k^{\text{inc}} H_k^{\text{inc},*}) n_j \right] \right\} dS$$

$$N^{\text{sca}}_i = \int_{S_{\text{shell}}} \varepsilon_{ijk} \frac{1}{2} \left\{ \text{Real} \left[ (D_{jk}^{\text{sca}} E_k^{\text{sca},*} + E_k^{\text{sca}} D_k^{\text{sca},*} + B_k^{\text{sca}} H_k^{\text{sca},*} + H_k^{\text{sca}} B_k^{\text{sca},*}) n_l - (D_k^{\text{sca}} E_k^{\text{sca},*} + B_k^{\text{sca}} H_k^{\text{sca},*}) n_j \right] \right\} dS$$

$$N^{\text{ext}}_i = \int_{S_{\text{shell}}} \varepsilon_{ijk} \frac{1}{2} \left\{ \text{Real} \left[ (D_{jk}^{\text{ext}} E_k^{\text{ext},*} + E_k^{\text{ext}} D_k^{\text{ext},*} + B_k^{\text{ext}} H_k^{\text{ext},*} + H_k^{\text{ext}} B_k^{\text{ext},*}) n_l + (D_k^{\text{ext}} E_k^{\text{ext},*} + E_k^{\text{ext}} D_k^{\text{ext},*} + B_k^{\text{ext}} H_k^{\text{ext},*} + H_k^{\text{ext}} B_k^{\text{ext},*}) n_l - (D_k^{\text{ext}} E_k^{\text{ext},*} + B_k^{\text{ext}} H_k^{\text{ext},*}) n_i - (D_k^{\text{ext}} E_k^{\text{ext},*} + B_k^{\text{ext}} H_k^{\text{ext},*}) n_i \right] \right\} dS$$

Based on the numerical experiments set up in our work, since the shell is spherical and its centre is fixed at the focus of the incident Gaussian beam, the force and torque due to the incident fields $E^{\text{inc}}$ and $H^{\text{inc}}$ are zeros: $F^{\text{inc}} = \mathbf{0}$ and $N^{\text{inc}} = \mathbf{0}$. It is worth mentioning that if the particle is not located at the focus of beam, there will be optical force generated from the incident field (gradient force). As such, in this work, we investigated the total optical force and torque, $\mathbf{F}$ and $\mathbf{N}$ defined in Eqs. (4) and (5) in the main text, respectively, and those from the scattered field, $F^{\text{sca}}$ and $N^{\text{sca}}$ defined in Eqs. (1b) and (2b), respectively, and those from the interaction between...
Figure 2: Optical force along the direction of incident beam propagation on the off-concentric Au@SiO$_2$ core-shell particle under the linearly polarised Gaussian beam illumination. (a) - (c) in air with $n_{\text{air}} = 1$; (d) - (f) in water with $n_{\text{water}} = 1.33$. The shell radius is $a_{\text{shell}} = 90$ nm, the core radius is $a_{\text{core}} = 60$ nm, the beam wavelength is $\lambda = 532$ nm, and the beam waist radius is $w_0 = 1 \mu$m.

1 Optical force along the beam propagation direction

Let us first consider the optical force along the beam propagation or the trapping force. Fig. 2 shows how the asymmetry, $h$, can affect the trapping force $F_z$ and its components $F_{z\text{sca}}$ and $F_{z\text{ext}}$ when an off-concentric Au@SiO$_2$ core-shell particle illuminated by a beam with wavelength $\lambda = 532$ nm. The top row of Fig. 2 shows the variations of $F_z$, $F_{z\text{sca}}$ and $F_{z\text{ext}}$ along with the Au core orientation, $\theta$ when the surrounding medium is air with $n_{\text{air}} = 1$. In Fig. 2(a), we can see that when the distance, $h$, between the Au core-centre and the SiO$_2$ shell-centre increases, the total optical force along the wave propagation, $F_z$ becomes larger and larger at all orientation angle $\theta$. The trends of the optical force along the wave propagation due to the interaction between the scattered field and the incident field $F_{z\text{ext}}$ and $N_{z\text{ext}}$ defined in Eqs. (1c) and (2c), respectively. Nevertheless, the optical force from the scattered field, $F_{z\text{sca}}$ is much smaller relative to that from the interaction between the incident and
Figure 3: Optical force along the direction of incident beam propagation on three types of asymmetric core-shell particles in water under a linearly polarised Gaussian beam illumination with beam waist radius as $w_0 = 1 \ \mu m$. The geometrical features of the core-shell particle are $a_{\text{shell}} = 90 \ \text{nm}$, $a_{\text{core}} = 60 \ \text{nm}$ and $h = 25 \ \text{nm}$.

When the surrounding medium is water ($n_{\text{water}} = 1.33$), the variations of the optical forces due to the asymmetry of the Au core change accordingly because of the change of the ratio of the relative refractive index between the SiO$_2$ shell and the surrounding medium. As shown in Fig. 2(d), when the asymmetry $h$ of the off-concentric particle becomes larger, the net optical force along the beam propagation, $F_z$ increases at most orientations of the Au core except for a small range around $\theta = 270^\circ$ which is the effect from the force component due to the scattered field, $F_{\text{sca}}^z$ as displayed in Fig. 2(e). When comparing Figs. 2(e-f), we can see that the optical force from the interaction between the incident and the scattered field, $F_{\text{ext}}^z$ dominates that from the scattered field, $F_{\text{sca}}^z$. 


Figure 4: Optical force perpendicular to the direction of incident beam propagation on the off-concentric Au@SiO\(_2\) core-shell particle under the linearly polarised Gaussian beam illumination. (a) - (c) in air with \(n_{\text{air}} = 1\); (d) - (f) in water with \(n_{\text{water}} = 1.33\). The shell radius is \(a_{\text{shell}} = 90\) nm, the core radius is \(a_{\text{core}} = 60\) nm, the beam wavelength is \(\lambda = 532\) nm, and the beam waist radius is \(w_0 = 1\) \(\mu\text{m}\).

The scattered field, \(F_{\text{sca}}\).

Fig. 3 presents the effect of wavelength on the optical force along the Gaussian beam propagation for three types of asymmetric core-shell particles in water: Au@SiO\(_2\), TiO\(_2\)@SiO\(_2\) and SiO\(_2\)@TiO\(_2\). Four wavelengths, \(\lambda = 532\) nm, \(\lambda = 775\) nm, \(\lambda = 840\) nm and \(\lambda = 1064\) nm are under consideration when the linearly polarised Gaussian beam waist radius is fixed as \(w_0 = 1\) \(\mu\text{m}\). As the wavelength becomes larger and larger, the magnitudes of the net optical force, \(F_z\) decreases. From the first and second rows of Fig. 3, we can clearly see that the optical force from the interaction between the scattered and incident fields, \(F_{\text{ext}}\), dominates the force from the scattered field, \(F_{\text{sca}}\), for Au@SiO\(_2\) and TiO\(_2\)@SiO\(_2\). However, for SiO\(_2\)@TiO\(_2\), the magnitude of these two parts are in the same order and they are competing with each other. The net force \(F_z\) shown in Fig. 3(g) indicates that the optical force from the interaction between the scattered and incident fields, \(F_{\text{ext}}\), overcomes the force from the scattered field, \(F_{\text{sca}}\).

2 Optical force perpendicular to the beam propagation

Fig. 4 displays the optical force perpendicular to a linearly polarised Gaussian beam with its light wavelength as \(\lambda = 532\) nm illuminating on an off-concentric Au@SiO\(_2\) core-shell particle.
Figure 5: Optical force perpendicular to the direction of incident beam propagation on three types of off-concentric core-shell particles in water under a linearly polarised Gaussian beam illumination with beam waist radius as $w_0 = 1 \mu m$. The geometrical features of the core-shell particle are $a_{\text{shell}} = 90 \text{ nm}$, $a_{\text{core}} = 60 \text{ nm}$ and $h = 25 \text{ nm}$.

The variation trends of such a force, $F_x$ and its components, $F_{x}^{\text{sca}}$ and $F_{x}^{\text{ext}}$ with respect to the orientation of the Au core, $\theta$ are quite similar for air or water as the surrounding medium. This force is more significant when the centre of the Au core is further away from the centre of the SiO$_2$ shell. Unlike the force along the beam propagation, $F_z$ in which the contribution of the interaction between the scattered and incident fields is dominating, for the optical force perpendicular to the beam direction, $F_x$, both the force from the scattered field, $F_{x}^{\text{sca}}$ and that from the interaction of the scattered and incident fields, $F_{x}^{\text{ext}}$ have similar contributions to the net $F_x$ for the Au@SiO$_2$ core-shell particle when $\lambda = 532 \text{ nm}$.

The optical force perpendicular to beam propagation under different wavelengths on three types...
Figure 6: Optical torque perpendicular to the direction of incident beam propagation on the off-concentric Au@SiO$_2$ core-shell particle under the linearly polarised Gaussian beam illumination. (a) - (c) in air with $n_{\text{air}} = 1$; (d) - (f) in water with $n_{\text{water}} = 1.33$. The shell radius is $a_{\text{shell}} = 90$ nm, the core radius is $a_{\text{core}} = 60$ nm, the beam wavelength is $\lambda = 532$ nm, and the beam waist radius is $w_0 = 1 \, \mu\text{m}$.

The direction of the optical torque is perpendicular to the plane constructed by the light propagation direction and its electric field polarisation direction. Fig. 6 illustrates how the asymmetry, $h$, and orientation, $\theta$, of the Au core affect the optical torque on an off-concentric Au@SiO$_2$ core-shell particle under the illumination of a linearly polarised Gaussian beam with wavelength as $\lambda = 532$ nm. As shown in Figs. 6(b-c) and (e-f), when the displacement $h$ between the centre of the Au core and that of the SiO$_2$ shell increases, the optical torques due to the scattered field and the interaction between the incident and scattered fields become more and more significant. However, these two effects are out of phase (180 degree difference) along the orientation angle of the Au core.

3 Optical torque

The direction of the optical torque is perpendicular to the plane constructed by the light propagation direction and its electric field polarisation direction. Fig. 6 illustrates how the asymmetry, $h$, and orientation, $\theta$, of the Au core affect the optical torque on an off-concentric Au@SiO$_2$ core-shell particle under the illumination of a linearly polarised Gaussian beam with wavelength as $\lambda = 532$ nm. As shown in Figs. 6(b-c) and (e-f), when the displacement $h$ between the centre of the Au core and that of the SiO$_2$ shell increases, the optical torques due to the scattered field and the interaction between the incident and scattered fields become more and more significant. However, these two effects are out of phase (180 degree difference) along the orientation angle of the Au core.
core, $\theta$, when the surrounding medium is air, as shown in Figs. 6(b) and (c). This leads to a small net optical torque as presented in Fig. 6(a). When the surrounding medium is water, the optical torques due to the scattered field and the interaction between the incident and scattered fields are in phase along with the orientation angle of the Au core, $\theta$. As such, the total optical torque on the asymmetric Au@SiO$_2$ core-shell particle in water is more significant relative to that in air.

Fig. 7 shows the effect of wavelength on the optical torque acting on three types of asymmetric core-shell particles with $h = 25$ nm under the illumination of a linearly polarised Gaussian beam in water. As displayed in the first row of fig. 7, for the Au@SiO$_2$ asymmetric core-shell particle, the optical torque from the scattered field, $N^\text{sca}_y$ and that from the interaction between the scattered
and incident fields, $N_{y}^{\text{ext}}$ are in phase with respect to the orientation of the Au core, $\theta$, which leads to a significant net optical torque, $N_{y}$. As to the TiO$_2@$SiO$_2$ off-concentric core-shell particle, net optical torque, $N_{y}$ is dominated by the interaction between the scattered and incident fields, $N_{y}^{\text{ext}}$; while for the SiO$_2@$TiO$_2$ asymmetric core-shell particle, the contribution from the scattered field dominates.