Trapping of low-refractive-index nanoparticles in a hollow dark spherical spot

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Keywords: laser beam shaping, optical manipulation, polarization

Abstract
Optical trapping techniques have been of great interest and have advantages that enable the direct handling of nanoparticles. However, stable trapping of low-refractive-index nanoparticle remains challenging because the conventional two-dimensional hollow beams are only capable of trapping nanoparticle in the transverse plane. In this work, we propose a novel strategy to optically trap low-refractive-index nanoparticle in three-dimensional space with a hollow dark spherical focal spot, which is generated by 4Pi focusing of radially polarized first-order Laguerre–Gaussian beam. With the assumption that the laser power is 100 mW, the nanoparticles can be stably trapped with the maximal optical force of 0.3 pN, potential depth of $10^{3} KBT$ and stiffness of $80 \text{pN}/\mu\text{m}$. Moreover, both the number and the position of the focal spot can be controlled by modulating the focusing condition and the gradient phase of the illumination respectively, enabling the simultaneously trapping of multiple nanoparticles with complex motion trajectory. The technique demonstrated in this work may open up new avenues for optical manipulation and their applications in various scientific fields.

1. Introduction

In 1986, Ashkin and his colleagues reported the first observation of a stable three-dimensional optical trap, or optical tweezers, created using the force applied on a microparticle from a single laser beam [1]. Due to the non-contact and non-destructive features, optical tweezers has exceeded mechanical force in many applications ranging from physics to biochemical, and provides an all-optical method that allows to manipulate particle ranging in size from tens of nanometers to micrometers. The continuous development of optical tweezers has revolutionized the experimental study of small particles and become an important tool for research in biology, physical chemistry and soft matter physics [2, 3]. In order to achieve a steady optical trapping, the gradient force exerted on the particle must be large enough to overcome the scattering force, especially in the direction parallel to the optical axis. Since the gradient force is proportional to $\pm \nabla E^2$, where $\nabla E^2$ is the intensity gradient of the light. The choice of sign depends on the difference of the refractive indices between the surrounding medium and the particle. For particle with refractive index larger or lower than the medium, the sign ‘+’ or ‘−’ applies respectively. Since the conventional focused laser beam has the intensity profile of Gaussian distribution, gradient force would always point towards the focus in any directions, making it easy to realize the optical trapping of high-refractive-index particle. However, a focal field with solid intensity pattern is not always successful in trapping absorptive particles. Firstly, gradient force would drag the particle near the high-intensity focal region of the beam, therefore the particles are susceptible to optical damage through absorptive heating [4]. Secondly, the absorptive particles may be knocked out of the trap owing to the impact of photons, especially when the trapping wavelength close to the resonant wavelength of the particle [5, 6]. In recent years, optical fields with inhomogeneous spatial distribution in terms of phase, amplitude and polarization are introduced into optical tweezers as the trapping light. These complex fields with more degrees of freedom are found helpful to improve the trapping performance and realize novel optical manipulation. For example, the advantages of radial polarization in trapping metallic nanoparticle in terms of larger gradient force and zero axial scattering force

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have been demonstrated both theoretically and experimentally [7, 8]. Negative scattering force with direction against the power flow has been reported for some Bessel beams [9, 10], enabling the trapping of plasmonic nanoparticle even under the resonant condition [11–13]. Using interfering high-order Bessel beams, rotation of microscopic particles can be controlled in optical tweezers and rotators [14]. Based on the same use of an interference of counter-propagating Bessel beams, a 3D confinement of high-index micro-particles in array of optical traps has been reported [15].

Aqueous systems usually contain both high- and low-refractive index particles in the host solution, such as air bubbles and hollow particles. The ability to manipulate these two kinds of particles is highly desirable for plenty of applications. Since the gradient force would point from the focus center to the region of low intensity, the conventional stationary Gaussian-beam trap is not suitable to trap spherical low-refractive-index particles, while a beam with focusing feature of hollow intensity distribution is required [16]. For example, high-order Bessel beams, optical vortex beam, and cylindrical vector beams are demonstrated to confine the low-refractive-index particles at the focal plane [17–20]. However, since the focal field sustains the transversal hollow dark pattern within the focal region, these optical fields with phase or polarization singularity can only trap the low-refractive-index particle in two-dimensional space due to the absence of gradient force along the optical axis. To create an axial equilibrium position as well, three-dimensional optical chain was generated by spatially modulating the phase of light with a diffractive optical element (DOE) [21]. However, the practical applications are hindered by the complicated DOE design with multiple regions and the tedious alignment requirement. Bottle beams that have a zero intensity region surrounded by light all around have been produced by many methods both theoretically and experimentally [22–24], and their applications in trapping atoms are reported [25, 26]. Besides, three-dimensional bottle beams have been studied and widely used for optical tweezers [27–30], however the axial gradient force is too weak to manipulate the low-refractive-index nanoparticle along the optical axis. In the last decade, 4Pi microscopy was developed to improve the axial resolution and synthesize focal field with various impressive characteristics through modulating the polarization and phase of the illumination [31–35]. For example, diffraction-limit focal spots with specific intensity pattern (spherical shape with dark/solid center [33, 36, 37]) and controllable polarization state [38] are reported. Moreover, a novel technique which can generate dark-hollow optical beams with a controllable shape has been realized and studied theoretically and experimentally [39]. In this work, we investigate the optical trapping effects of a hollow dark spherical focal spot and demonstrate an efficient method to optically trap low-refractive-index nanoparticle. It is demonstrated that a focal spot with an extremely sharp central dark region and almost-perfect spherical symmetry can be achieved, giving rise to the greatly enhanced gradient force in any directions. The interactions between the hollow dark spherical spot and nanoparticles enable not only the three-dimensional trapping, but also the precise control of the motion trajectory of multiple nanoparticles simultaneously.

2. Configuration of the optical tweezers

Figure 1 illustrates the diagram of the proposed optical tweezers. Two counter-propagating radially polarized light are tightly focused by two objective lenses with high numerical aperture [37]. As shown in the vector diagram, the two beams are phase shifted by $\pi$ with respective to each other. The electric field of a radially polarized first-order Laguerre–Gaussian beam can be expressed as:
\[ E = E_0 \exp \left( -\frac{2r^2}{w^2} \right) e^{i\varphi} \]

where \( E_0 \) is a constant, \( w \) is the beam waist, \( r \) and \( \varphi \) are the radius and azimuthal angle of the cylindrical coordinate system, respectively. The electric field in the vicinity of the focus of a high numerical aperture objective can be calculated with Richards-Wolf vectorial method [40]:

\[ E(r, \varphi, z) = iA \int_{\theta_{\text{min}}}^{\theta_{\text{max}}} P(\theta) e^{ikz} \cos \theta \sin \theta \left( \cos \theta [J_2(kr \sin \theta) - J_0(kr \sin \theta)] \hat{e}_r - i \cos \theta [J_2(kr \sin \theta) + J_0(kr \sin \theta)] \hat{e}_\varphi - 2i \sin \theta J_1(kr \sin \theta) \hat{e}_z \right) d\theta, \]  

where \( \theta_{\text{max}} \) and \( \theta_{\text{min}} \) are the maximum and minimum focusing angles determined by the numerical aperture of the objective, respectively. \( J_n(r) \) represents the \( n \)-th order Bessel function of the first kind, and \( k \) is the wave-vector in the focal region. The constant \( A \) is given by \( A = \pi f l_0 / \lambda \), where \( f \) is the focal length, \( \lambda \) is the wavelength of the incident light in the ambient environment, and \( l_0 \) is associated with the laser power. \( P(\theta) \) is the pupil apodization function of the objective, which can be expressed as \( P(\theta) = \cos^{1/2} \) for objective lens obey sine condition [40]. Note that the spherical aberration induced by the oil-glass interface can be ignored due to the short working distance of the objective lens. Finally, the electric field near the focus of a 4Pi focusing system can be expressed as:

\[ E_f(r, \varphi, z) = E_1(r, \varphi, z) + E_2(r, \varphi, -z), \]

where \( E_1 \) and \( E_2 \) denote the electric fields from the left and right objectives, respectively.

Differently from single-beam gradient force optical trap, 4Pi microscopy utilizes two counter-propagating beams to create various interference field in the focal region, providing more degrees of freedom to tailor the focal field and the optical force as well. In order to increase the magnitude of the gradient force, two aplanatic oil-immersion lenses with numerical aperture of 1.4 are adopted to tightly focus the light. Assuming the space between the objective lenses is filled with water (\( \varepsilon = 1.77 \)), the interference pattern of the focal fields of radially polarized first-order Laguerre–Gaussian beam is numerically calculated using equations (2) and (3) and shown in figures 2(a) and (b). Note that the parameter \( w \) in the input beam intensity distribution is chosen such that the beam width is
equal to the radius of the objective lens. It can be seen that the intensity distribution has complete circular symmetry in the \(x-y\) plane, with zero intensity at the center. Besides, this is accompanied by a nearly circularly symmetric intensity distribution along the optical axis, leading to a tightly focused hollow spherical spot. The corresponding line-scans of the axial and transversal intensity distributions are shown in figures 2(c) and (d), demonstrating that axial and transversal focal spots are nearly equal with size match about 0.02 \(\lambda\). The FWHM of the spot size is measured to be 0.35 \(\lambda\), corresponding to a focal volume of 0.02 \(\lambda^3\). Note that the hollow dark spherical spot can only be synthesized by radially polarized light with first order. It is because the total focal field is a superposition of three different modes proportional to \(m^{-1}, m\) and \(m+1\) order Bessel functions \([37]\), where \(m\) denotes the topological charge of the incident light. For illumination with \(|m|\) larger than 1, there would be zero intensity along the optical axis within the focal region, leading to the symmetry breaking of the focal pattern.

Considering a hollow spherical low-refractive-index nanoparticle with relative permittivity of \(\varepsilon_m = 1\) and radius \(a = 50\) nm is immersed in the focal region of the 4Pi microscopy, its movement is subject to the time-averaged optical force induced by the focal field. Since the size of the particle is assumed to be far less than the wavelength of light, the electric field \(E\) can be viewed as constant over the whole particle. Although the field varies over a spatial distance of 200 nm and the particle is 100 nm in diameter, as studied in [41], the dipole response still describes reasonably well the dielectric response. Then, the particle may be considered simply as an induced electric dipole, the moment of which is related linearly to the external field through the polarizability. For a spherical particle, its polarizability is a scalar that can be described as [5]:

\[
\alpha = \frac{\alpha_0}{1 - i\alpha_0 k^2 / (6\pi)},
\]

where \(\alpha_0 = 4\pi a^3 [\varepsilon_m(\omega) - \varepsilon] / [\varepsilon_m(\omega) + 2\varepsilon]\), \(k\) is the wave-vector in the medium, and \(\omega\) is the frequency. It is known that the transfer of momentum from the laser to an object would produce optical forces, which can be employed to trap, manipulate and characterize nanoparticle. The optical force exerted on this hollow spherical nanoparticle can be expressed as [5]:

Figure 3. Distributions of the optical forces induced by the focal field in figure 2 and the corresponding potential depth exerted on 50 nm (radius) low-refractive index nanoparticle along (a), (c) radial and (b), (d) longitudinal directions.
where $F_{\text{grad}}$ and $F_{\text{scat}}$ are the gradient force and scattering force, respectively. \( S \) is the Poynting vector, \( c \) is the light speed, \( k_0 \) is the wave-vector in free space, and \( \sigma = k \Im(\alpha) \) is the total cross section of the particle. In cylindrical coordinate system \((r, \phi, z)\), the optical force can be rewritten as:

\[
\begin{align*}
F_{\text{grad}} &= \frac{1}{4} \varepsilon_0 \text{Re}(\alpha) \nabla |\vec{E}|^2, \\
F_{\text{scat}} &= \frac{\pi \sigma}{c} \langle \vec{S} \rangle - \frac{\varepsilon_0 \sigma}{2k_0} \text{Im}[\vec{E} \cdot \nabla \vec{E}^*],
\end{align*}
\]  

where $F_{\text{grad}}$ and $F_{\text{scat}}$ are the gradient force and scattering force, respectively. \( S \) is the Poynting vector, \( c \) is the light speed, \( k_0 \) is the wave-vector in free space, and \( \sigma = k \Im(\alpha) \) is the total cross section of the particle. In cylindrical coordinate system \((r, \phi, z)\), the optical force can be rewritten as:

\[
\begin{align*}
F_x &= \frac{1}{4} \varepsilon_0 \text{Re}(\alpha) \nabla_x |\vec{E}|^2 + \frac{\pi \sigma}{2c} \text{Re}(E_0 E_t^* - E_2 H_z^*) - \frac{\varepsilon_0 \sigma}{2k_0} \text{Im} \left( E_x \frac{\partial E_x^*}{\partial r} + E_\phi \frac{\partial E_\phi^*}{\partial \phi} + E_z \frac{\partial E_z^*}{\partial z} \right), \\
F_\phi &= \frac{1}{4} \varepsilon_0 \text{Re}(\alpha) \nabla_\phi |\vec{E}|^2 + \frac{\pi \sigma}{2c} \text{Re}(E_2 H_t^* - E_0 H_z^*) - \frac{\varepsilon_0 \sigma}{2k_0} \text{Im} \left( E_\phi \frac{\partial E_\phi^*}{\partial r} + E_x \frac{\partial E_x^*}{\partial \phi} + E_z \frac{\partial E_z^*}{\partial z} \right), \\
F_z &= \frac{1}{4} \varepsilon_0 \text{Re}(\alpha) \nabla_z |\vec{E}|^2 + \frac{\pi \sigma}{2c} \text{Re}(E_0 H_t^* - E_2 H_z^*) - \frac{\varepsilon_0 \sigma}{2k_0} \text{Im} \left( E_z \frac{\partial E_z^*}{\partial r} + E_\phi \frac{\partial E_\phi^*}{\partial \phi} + E_x \frac{\partial E_x^*}{\partial z} \right).
\end{align*}
\]  

In this case, the wavelength of the illumination is chosen to be 808 nm, at which the specimens would have a low absorption coefficient so as to minimize heating damage to the biological material. With the assumption that the total input power is 200 mW, the optical forces along the \( z \) - and \( x \) -axis are calculated using equations (7)–(9) and shown in figures 3(a) and (b). It can be clearly seen that the axial scattering force is canceled by the counter-propagating focal fields. Besides, the transversal scattering force is also negligible for lossless particle. Consequently, the low-refractive-index nanoparticle would be confined at the dark center of the hollow spherical focal spot by the gradient force. In order to evaluate the stability of the optical trapping, the distributions of the corresponding potential depths are calculated by $U = -\int F \cdot ds$ and presented in figures 3(c) and (d). Generally, an optical trapping with potential depth larger than one \( k_B T \) can be considered as...
The simulation results clearly illustrate that potential depths of nearly 10 \( K_B T \) can be achieved in both axial and transversal directions, demonstrating a stable trap in three-dimensional space. In addition, the stiffness \( k_s \) of the optical trap at the equilibrium position is calculated to be 80 pN/\( \mu \)m. It is estimated by the Hook’s law that \( k_s = -F/x \), where \( F \) is the optical force and \( x \) is the distance from the equilibrium position. Besides, the optical trap would not be destabilized by heating effects since the particle is trapped near the low-intensity focal region. Contrarily, as for the nanoparticle with refractive index larger than medium (\( \varepsilon_m = 2.53 \)), we can see that

\[ Figure 5. \] Intensity distribution in the vicinity of the focal point of a tightly focused azimuthally polarized light in the (a) \( x-y \) plane and (b) \( z-x \) plane. Distributions of the (c) optical forces induced by the focal field in (a), (b) and the corresponding (d) potential depth exerted on 50 nm (radius) low-refractive index nanoparticle.

\[ Figure 6. \] The relationship between the size of the hollow spherical focal spot and the effective numerical aperture of the objective lens.
there are two equilibrium points along both radial and longitudinal directions (shown in figure 4), indicating that the high-refractive-index nanoparticle would be trapped along the radial direction in a spherical region at the focus and the particle can move freely along the azimuthal direction. Consequently, it is not only feasible to simultaneously trap more nanoparticles due to the wide area of the spherical surface, but also be capable of trapping both high- and low-refractive-index nanoparticles at the same time.

For comparison, focal field distributions and optical forces for single-beam optical trap with azimuthally polarized illumination are shown in figure 5. It is known that the focal field of tightly focused azimuthally polarized light only has azimuthal component \( \ell = 0 \) \cite{40}. As a fair comparison, the input power is set to be 200 mW. Similar to the case presented in figure 2, the focal field for azimuthal polarization has a null at the center in the transverse plane. The optical force distribution also shows that there is one equilibrium point at the center for particles with index lower than the ambient (shown in figure 5(c)), which demonstrates the suitability of the focused azimuthally polarized light for trapping low-refractive-index nanoparticle. Note that the magnitude of the gradient forces are smaller than the case shown in figure 3. It is because the incident energy is highly concentrated into a hollow spherical spot with diffraction-limit size by the 4Pi focusing system. Since the focal field of azimuthal polarization along the \( z \)-axis is essentially zero, the \( z \) component of the gradient forces are negligible. This implies that the path of the low-refractive-index nanoparticle would be in Brownian motion along the optical axis.

From the above simulation results, one can see that the key of the proposed trapping strategy is to create a hollow dark spherical focal spot that can provide a stable trapping of low-refractive-index nanoparticle in three-dimensional space. However, only hollow particle with small enough size can be confined in the dark center of the focused beam. Otherwise, it would be knocked out from the focus by the high-intensity focal region. As shown in figure 6, the largest size of the trappable particle is determined by the size of the dark region of the focal field, which is inversely proportional to the numerical aperture of the objective lens. It is worthy of noting that the FWHMs of the focal field in the \( x-z \) plane and \( x-y \) plane are nearly the same. Consequently, it would be a convenient way to trap low-refractive-index nanoparticle with different size by adjusting the focusing system.

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**Figure 7.** Intensity distribution in the (a) \( x-y \) plane and (b) \( x-z \) plane of a hollow dark spherical spot centered at \( P = (5\lambda, 2\lambda, 10\lambda) \).

**Figure 8.** Intensity distribution in the vicinity of the focal point for radially polarized first-order Laguerre–Gaussian illumination focused by 4Pi focusing system with \( \theta_{\text{min}} = 37^\circ \), \( \theta_{\text{max}} = 62^\circ \).
Note that the beam width may need to be optimized for objective with different numerical apertures so as to maintain the central symmetry of the hollow focal spot.

In plenty of applications that involves optical trapping, it is often desirable to move the particle along scheduled trajectory. It is known that the position of the focal spot can be changed by encoding additional phase to the trapping laser, enabling the shift of particle without the introduction of mechanical disturbance. In order to move the particle from origin to \( P(x_0, y_0, z_0) \), required phase modulation can be expressed as:

\[
\eta(\theta, \varphi) = e^{-i \vec{p} \cdot \vec{r}} = e^{-i k (x_0 \cos \varphi + y_0 \sin \varphi + z_0 \cos \theta)}.
\]  

Figure 9. (a) Optical forces induced by the focal field in figure 8 exerted on 50 nm (radius) low-refractive-index nanoparticle along the optical axis. (b)–(j) Distribution of the optical forces along radial direction and the potential depths along radial and longitudinal directions at the three axial equilibrium positions in (a).

Note that the phase modulations applied on the right- and left-propagating light beams are \( \eta_1(\theta, \varphi) = e^{-i \vec{p} \cdot \vec{r}} \) and \( \eta_2(\theta, \varphi) = e^{i \vec{p} \cdot \vec{r}} \) respectively. Figure 7 shows the intensity pattern when the focal field is shift to \( P = (5\lambda, 2\lambda, 10\lambda) \). Compared with the pattern shown in figure 2, one can see that the movement of the focal field does not change its shape and intensity, giving rise to the creation of new equilibrium position. Since the nanoparticle would follow the hollow spherical focal spot as it moves, complex motion trajectory of the nanoparticle can be achieved by a series of segment movement of the focal field, with the single maximum displacement determined by the width of the trapping potential (about 0.6\( \lambda \) shown in figure 3(c)).
In addition, dark-hollow beam array can be obtained with a filter, which is an annular aperture where only the illumination with limited range of incident angle can transmit. Figure 8 shows the focal field intensity when the focusing condition is chosen to be \( \theta_{\text{min}} = 37^\circ, \theta_{\text{max}} = 62^\circ \). Note that the restriction on the minimum incident angle is necessary to alleviate the deformation of the beam array. In this case, most energy is concentrated into three dark cores arranged along the optical axis. Besides, it is found that the dark cores are almost spherically and periodic. As shown in figure 9, there are three equilibrium positions with deep enough potential depth along the optical axis, enabling the simultaneous trapping of multiple low-refractive-index nanoparticles at the dark centers of the focal field.

3. Conclusion

In conclusion, we have numerically demonstrated the trapping of a low-refractive-index sphere \((50 \text{ nm in radius})\) in water by using a hollow dark sphere focal spot, which is generated by 4Pi focusing of radially polarized first-order Laguerre–Gaussian beam. The circularly symmetric intensity distribution of the focal field along the optical axis gives rise to a axial equilibrium position at the focus, enabling the stable trapping of both high- and low-refractive index nanoparticle in three-dimensional space. By encoding the illumination with gradient phase modulation, the movement of the nanoparticle can be designed to follow a complex motion trajectory. Besides, simultaneously trapping of multiple nanoparticle is feasible by adjusting the focusing conditions, in which case specific number of the hollow sphere focal spots arranged along the optical axis can be generated. This versatile trapping method may open up new avenues for optical trapping and their applications in various scientific fields.

Acknowledgments

National Natural Science Foundation of China (11504049, 11474052, 11774055); National Science Foundation of Jiangsu Province (BK20150593); National Key Basic Research Program of China (2015CB352002).

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