Investigation of the Polarization-dependent Optical Force in Optical Tweezers by Using Generalized Lorenz-Mie Theory

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In vectorial diffraction theory, tight focusing of a linearly-polarized laser beam produces an anisotropic field distribution around the focal plane. We present a numerical investigation of the electromagnetic field distribution of a focused beam in terms of the input beam’s polarization state and the associated effects on the trap stiffness asymmetry of optical tweezers. We also explore the symmetry change of a polarization-dependent optical force due to the electromagnetic field redistribution in the presence of dielectric spheres of selected diameters ranging from the Rayleigh scattering regime to the Mie scattering regime.

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I. INTRODUCTION

Achieving the tightest spot of highly-focused light requires a dedicated state-of-the-art optical system and a rigorous description of the light-matter interaction, from which other tuning parameters for controlling the optical field distribution might be determined. If a stable trap condition is to be realized in optical tweezers (OTs), a laser beam should be focused enough to overcome the scattering force by using a high-numerical aperture (NA) objective lens [1]. Recent studies showed that the spherical aberration (SA) exhibited by high-NA optical systems was due to a refractive index mismatch between the media interfaces [2] and could be tailored by using control parameters such as the adjusted wavefront of an input laser beam [3] or the immersion medium’s refractive index [4]. The electromagnetic (EM) field distribution of a focused laser beam is anisotropic in the focal plane even in an aberration-free ideal optical system [5–10]. The anisotropic distribution of the focused EM field manifests itself as trap stiffness (spring constant) asymmetry in OTs that utilize a linearly-polarized laser beam [8–10]. In this study, we address the anisotropy of EM field distribution of highly-focused light in terms of the input beam’s polarization state, demonstrating that the associated stiffness asymmetry in optical tweezers could be tuned via the polarization state of the trap laser beam.

For a precise description of a highly-focused EM field, the vectorial nature of light needs to be considered, as formulated in the angular spectrum method [5, 11], whereas scalar diffraction theory predicts an Airy disk pattern for a typical optical system. The tightly-focused EM field distribution of a linearly-polarized input beam has been investigated in various situations: a homogeneous medium [5], a single interface dividing immersion and specimen media [11], and a more realistic situation consisting of three interfaces including the coverslip’s thickness [2]. The focused EM field distribution of circularly-polarized light has also been investigated with respect to angular momentum transfer by using a multipole expansion [12]. Assuming aberration-free conditions which can be achieved to some extent by using a correction collar of a modern objective lens [3], we investigated the anisotropy of the focused EM field by using vectorial diffraction theory. Polarization-dependent EM field distributions in highly-focused OTs were evaluated numerically in terms of the trap stiffness asymmetry, which could be interpreted as the landscape of the optical potential. For this purpose, we revised the generalized Lorenz-Mie theory (GLMT) developed by Barton et al. [13–15] to include the polarization state of the trap beam in optical force calculations.

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II. MATHEMATICAL BACKGROUND FOR NUMERICAL CALCULATIONS

In this section, we illustrate the polarization-dependent electric field distribution of a highly-focused beam in a homogeneous medium by using the angular spectrum method. We outline the procedure for a GLMT calculation of the optical force, which was revised to encompass the general polarization state of an input beam. The coordinate systems are depicted in Fig. 1. Cartesian coordinate systems \((x,y,z)\) and \((x',y',z')\) are centered on the lens aperture \(\Sigma\) and the focal point \(O_G\), respectively, of the focused beam with the same orientation. The other, unannotated, Cartesian system centered at \(O_P\) is particle-centered for the GLMT calculation. The collimated input beam represented by its electric field \(\vec{E}^{in}\) enters the lens aperture \(\Sigma\) along the direction defined by the wave vector \(\vec{k}_0\), parallel to the optic axis \((z \text{ and } z')\), and is focused by the high-NA optical system into the medium space with a refractive index of \(n_m\). One partial plane wave component, \(\vec{E}^{in}\), in the direction \(\vec{k}_m\) is presented in Fig. 1, with a polar angle \(\theta\). The other symbols for angles in Fig. 1 are implicit. The resulting electric field distribution at \(\vec{r}_p(r_p, \theta_p, \phi_p)\) around the focal point \((O_G)\) is represented by using the angular spectrum method, which sums all relevant partial wave components, as follows [2,5,10,11]:

\[
\vec{E}(r_p, \theta_p, \phi_p) = -\frac{ikf}{2\pi} \sqrt{\frac{n_0}{n_m}} \int_0^{\alpha_{max}} d\theta \int_0^{2\pi} d\phi J(\theta) A(\theta) \times e^{-\sin^2 \theta / (f_0^2 \sin^2 \alpha_{max})} M \vec{E}^{in} e^{ikr_p\delta(\theta_p, \phi_p, \theta, \phi)}, \tag{1}
\]

where \(k\) is the wavenumber in the medium \((k = n_m k_0)\), \(J(\theta) = \sin \theta \cos \theta\) is the Jacobian for the transformation from \((\vec{k}_m, \vec{k}_p)\) to \((\theta, \phi)\), the apodization factor \(A(\theta) = 1/\sqrt{\cos \theta}\) fulfills the sine condition, \(f_0 = w_0 / (f \sin \alpha_{max})\) is the fill factor for the collimated input beam spot size \(w_0\), \(f\) is the focal length, the maximum polar angle is given as \(\alpha_{max} = \sin(\text{NA}/n_m)\), \(M\) is the polarization matrix, \(\kappa(\theta, \phi, \theta_p, \phi_p) = \sin \theta \sin \theta_p \cos(\theta - \theta_p) + \cos \theta \cos \theta_p\), and integration is performed over the solid angle defined by \(0 \leq \phi \leq 2\pi\) and \(0 \leq \theta \leq \alpha_{max}\).

The polarization matrix \(M\) given in Eq. (2) consists of the rotation matrix \(R\) about the optic axis and the matrix of the lens action \(L\):

\[
M = R^{-1} LR = \begin{pmatrix}
\cos \phi & -\sin \phi & 0 \\
\sin \phi & \cos \phi & 0 \\
0 & 0 & 1
\end{pmatrix}
\begin{pmatrix}
\cos \theta & 0 & \sin \theta \\
0 & 1 & 0 \\
-\sin \theta & 0 & \cos \theta
\end{pmatrix}
\begin{pmatrix}
\cos \phi & \sin \phi & 0 \\
-\sin \phi & \cos \phi & 0 \\
0 & 0 & 1
\end{pmatrix}.
\tag{2}
\]

For a more practical description that includes media interfaces, a matrix \(I\) consisting of the Fresnel coefficients for the s- and the p-polarization components and changes to polar angle \(\theta\) due to refraction need to be incorporated into \(M\) and \(\kappa\) [2,11]. The incident electric field is prepared for a general polarization state: \(\vec{E}^{in} = (E_x, E_y, 0)\), where \(E_x\) and \(E_y\) are complex numbers; a numerical evaluation of Eq. (1) then provides the polarization-dependent field distribution around the focal plane. The focused magnetic field distribution can be obtained by using a similar procedure.

Highly-focused electric-field patterns are obtained through numerical integrations of Eq. (1) for two extreme polarization states: the linear and the circular polarization states. We consider the following parameters: NA \(= 1.2\), \(f = 3\) mm, input-beam vacuum wavelength \(\lambda_0 = 780\) nm, \(f_0 = 1\), and medium refractive index \(n_m = 1.33\). The electric-field components \(|E_x|, |E_y|, |E_z|\) and the normalized intensity \(I\) in the focal plane \((x'y'-plane)\) are presented in Fig. 2(a) for a linearly-polarized incident field: \(\vec{E}^{in} = E_0 \hat{x}\), where the electric field components and the intensity are normalized such that \(|E_x| = 1\) at the focal point \(O_G\). The frame ticks in Fig. 2 are given in units of the minimum spot size \(w_G = 293\) nm, which was obtained from a Gaussian fit of the numerically-calculated field distribution. The white lines (dashed line) outline the \(5 \times 10^{-2}\) level of the magnitude of the electric-field components (the \(e^{-2}\) level of \(I\)). Additional polarization components, \(E_y\) and \(E_z\), produce an anisotropic field distribution, thus resulting in an elongation of the electric field.
polarized trap beams. Frame ticks are given in units of the normalized intensity distribution. The main contribution to the circularly-polarized incident field: \( |\vec{E}| \)\( \approx \hat{z} \times (n^2 - 1)/(n^2 + 2) \vec{E} \), where \( \varepsilon_p \) is the permittivity of a particle, \( a \) is the particle radius, the relative refractive index \( \bar{n} \) is the ratio of the refractive index of particle \( n_p \) to that of medium \( n_m \), and \( \vec{E} \) is the electric field. In the Rayleigh scattering regime, the exerted radiation force can be represented in terms of the gradient force, \( \vec{F}_{\text{grad}} \approx (\vec{p}(r', t) \cdot \vec{\nabla}) \vec{E}(r', t) \), and the scattering force, \( \vec{F}_{\text{sc}} = \frac{\pi a^2}{\mu_0 c^3} I(r') z' \), where \( c \) is the speed of light, \( \mu_0 \) is the magnetic permeability of free space, \( \mu_0 \) and \( c \) are the permittivity and the permeability of the surrounding medium, respectively, and \( \vec{r} \) is the unit vector along the radial direction (\( \vec{r}_a \)) in spherical coordinates, with the origin at \( O_p \). For a spherical particle of radius \( a \), EM fields can be represented by a series expansion of the Riccati–Bessel functions [13, 14], \( \psi_l(r) \) and \( \xi_l(r) \), and the spherical harmonics, \( Y_{lm}(\theta^a, \phi^a) \), with the following expansion coefficients: \( \{A_{lm}, B_{lm}\} \) for the incident field, \( \{a_{lm}, b_{lm}\} \) for the scattered field, and \( \{l, m\} \) for the spherical harmonic mode numbers. The boundary condition across the sphere’s surface provides the following relations between \( \{a_{lm}, b_{lm}\} \) and \( \{A_{lm}, B_{lm}\} \) [13]:

\[
\begin{align*}
A_{lm} &= \frac{\psi'_l(\bar{n}a)\psi_l(\bar{n}a) - \bar{n}\psi'_l(\bar{n}a)\psi_l(\bar{n}a)}{\bar{n}\psi_l(\bar{n}a)\xi'_l(\bar{n}a) - \bar{n}\xi_l(\bar{n}a)\psi'_l(\bar{n}a)}, \\
b_{lm} &= \frac{\bar{n}\psi'_l(\bar{n}a)\psi_l(\bar{n}a) - \psi'_l(\bar{n}a)\psi_l(\bar{n}a)}{\psi_l(\bar{n}a)\xi'_l(\bar{n}a) - \bar{n}\xi_l(\bar{n}a)\psi'_l(\bar{n}a)},
\end{align*}
\]
Numerical investigation of the polarization dependence of the optical force in OTs begins with a linearly-polarized trap beam and a homogeneous polystyrene (PS) sphere as the trap object. The following parameters were used for the virtual OTs experiments: $\lambda_0 = 780 \text{ nm}$, NA = 1.2 (water immersion), $f = 3 \text{ mm}$, $f_0 = 1$, $n_m = 1.33$, $n_p = 1.57$, and the beam power $P_0$ in the trap region is 10 mW. The radiation forces in the transverse and the longitudinal directions are presented in Figs. 3(a)–(c) for particles with radii $a = 50$, 150, and 500 nm, respectively. The transverse force components, $F_{x'}$ (red solid line) and $F_{y'}$ (blue dotted line), were estimated at equilibrium positions $z_{eq} = 20, 205$, and 226 nm for $a = 50$, 150, and 500 nm, respectively, where the longitudinal force component, $F_{z'}$ (green solid line), is zero; the scattering force pushed the equilibrium position away from the focal point. The trap potential anisotropy in the transverse plane was estimated in terms of the trap stiffness asymmetry factor $s_T = 1 - k_{x'}/k_{y'}$, where the trap stiffness was deduced by using a nonlinear fit to the numerically-calculated optical force for the corresponding degrees of freedom. For the 100-nm PS spheres (Fig. 3(a)), the trap stiffnesses $k_x$ and $k_y$ were 0.65 and 1.16 pN/μm, respectively, yielding a transverse plane stiffness asymmetry factor of $s_T = 0.44$. For the 300-nm PS sphere (Fig. 3(b)), the trap stiffnesses $k_x$ and $k_y$ were 33.3 and 29.8 pN/μm, respectively, and $s_T = 0.3$. Results for the 1-μm PS sphere (Fig. 3(c)) clearly indicate not only an averaging effect on the EM field distribution over the particle, but also an EM field distribution modification due to the particle’s presence: the trap stiffnesses $k_x$ and $k_y$ of the 1-μm PS sphere were 33.3 and 29.8 pN/μm, respectively, with a corresponding stiffness asymmetry factor $s_T = -0.12 < 0$. This counterintuitive sign change of the asymmetry factor was reported in previous studies that used a linearly-polarized trap beam [8–10].

The GLMT-based optical force calculations in the transverse directions are presented in Fig. 4 for a circularly-polarized trap beam and PS spheres with radii of (a) 100 nm, (b) 300 nm, and (c) 1 μm. Compared to the linear polarization results, the circularly-polarized trap-beam results show an orthogonal pair of lateral forces that overlap within the precision of the numerical calculation for the given particle sizes, implying that the isotropic field distribution provided by the circularly-polarized light is preserved even in the presence of a trapped spherical particle. The trap stiffnesses and asymmetry factors for circular polarization are summarized in Table 1, together with the linear polarization results. We also present the longitudinal stiffness asymmetry factor, $s_L = (k_{x'} + k_{y'})/2k_{z'}$, which is the ratio of the mean lateral stiffness to the axial stiffness. The numerical values of $s_L$ only represent the numerical error; the symmetry of the EM field distribution and the
Fig. 4. (Color online) GLMT calculation of the radiation forces for circularly-polarized light for PS sphere radii of (a) 50 nm, (b) 150 nm, and (c) 500 nm. For each PS sphere radius (a-c), the transverse force components, $F_{x'}$ and $F_{y'}$, are denoted by red and blue dashed lines, respectively.

Table 1. Trap stiffness and corresponding asymmetry factor in the transverse plane for various PS sphere radii and polarization states of the input beam. The superscript distinguishes polarization states: LP and CP for linear and circular polarization state, respectively. Parameters for this calculations are as follows: $\lambda_0 = 780$ nm, NA = 1.2, $f_0 = 3$ mm, $f_0' = 1$, $n_m = 1.33$, $n_p = 1.57$, $\lambda_n = \lambda_0 / n_m$, $\Delta \phi = (\bar{n} - 1)k2a$, and $P_0 = 10$ mW. The unit of stiffness is pN/μm.

| Radius (nm) | $\Delta \phi$ | $k_{x'}$ | $(k_{x'}, k_{y'})^{LP}$ | $(s_T, s_L)^{LP}$ | $(k_{x'}, k_{y'})^{CP}$ | $(s_T, s_L)^{CP}$ |
|------------|----------------|----------|------------------------|-------------------|------------------------|-------------------|
| 50         | 0.19           | 0.27     | (0.65, 1.16)           | (0.44, 3.4)       | (0.90, 0.90)           | (2.0 $\times 10^{-3}$, 3.3) |
| 150        | 0.58           | 2.8      | (13.5, 19.3)           | (0.30, 5.9)       | (16.5, 16.6)           | (6.1 $\times 10^{-3}$, 5.9) |
| 250        | 0.97           | 5.5      | (39.2, 42.5)           | (0.08, 7.4)       | (40.7, 40.8)           | (1.4 $\times 10^{-3}$, 7.4) |
| 350        | 1.35           | 8.7      | (53.3, 46.8)           | (-0.14, 5.8)      | (50.1, 50.0)           | (-1.9 $\times 10^{-3}$, 5.8) |
| 500        | 1.93           | 7.3      | (33.3, 29.8)           | (-0.12, 4.3)      | (31.4, 31.5)           | (-3.2 $\times 10^{-3}$, 4.3) |

Fig. 5. (Color online) Stiffness asymmetry versus particle size for polarization states (LP and CP). Filled and empty circles denote Rohrbach’s experimental data and theoretical values [8], respectively. Red empty squares and blue diamonds denote our theoretical estimates for linear polarization (LP) and circular polarization (CP) states, respectively.

Test spheres ensures that $s_{CP}$ is 0 (zero).

Noteworthy is that a small deviation in the polarization state from circular polarization breaks the isotropy of the focused EM field distribution and the optical forces in the transverse plane. Experimental realization of a tailored optical potential via polarization control could be limited by the retardance accuracy of the wave plate. For instance, the nominal retardance accuracy of a zero-order quarter-wave plate is about $\lambda/100$, which corresponds to the phase uncertainty $\delta \varphi = 6$ mrad, yielding the degree of circular polarization $\cos(\delta \varphi) = 0.998$. The optical field of a slightly-deviated polarization state, elliptical polarization (EP), produces a modified EM field distribution in the focal plane: $d_{EP}^{x,y'} / d_{LP}^{x,y'} = 0.985$. The associated trap stiffness asymmetry factor when using the most sensitive 100-nm PS spheres among the particles under consideration in Fig. 4 is estimated to be $s_{EP}^{T} = 3.4 \times 10^{-2}$, which implies that the strong asymmetry ($s_T$) of the optical potential landscape of OTs can be adjusted to be nearly isotropic via polarization control using a conventional wave plate.

We now apply our numerical method to Rohrbach’s experimental results [8], where the trap stiffness asymmetry of highly-focused OTs formed by focusing linearly-polarized light was investigated experimentally and theoretically by using a two-component model [8,17]. The experimental parameters were as follows: $\lambda_0 = 1064$ nm, $P_0 = 10$ mW, $n_m = 1.33$, $n_p = 1.57$, NA =1.2, $f_0 = 2$, and particle radii of 110, 265, 345, 425, 515, and 830.
nm. With these parameters, the minimum beam spot size $w' G$ was estimated to be 399 nm; we assumed an aberration-free ideal optical system. In Fig. 5, filled and empty black circles denote experimental and theoretical values from Rohrbach’s study [8], respectively. Experimental data indicated that the trap stiffness asymmetry factors for PS particles larger than the beam wavelength $\lambda_0/n_m$ were negative: $k_x < k_y$ for particle radii $a < \lambda_0/n_m$ and $k_x > k_y$ for particle radii $a > \lambda_0/n_m$. Rohrbach’s theoretical estimate of trap anisotropy by using the two-component model developed in the Rayleigh-Gans regime did not accurately describe the experimental data for particle sizes comparable to the beam’s wavelength. Significant gaps between the experimental and the theoretical results existed except for the two smallest particles ($a = 110$ and 265 nm); their theoretical method could not support the experimentally-observed sign change of the trap asymmetry factor. Our calculation results based on the revised GLMT show good agreement with Rohrbach’s experimental data, including the sign change of the trap anisotropy factor. More importantly, a demonstration of trap anisotropy control by using the polarization state of the trap beam is presented (blue diamonds in Fig. 5). With a circularly-polarized trap beam, the trap stiffness asymmetry factors were numerically estimated to be two orders of magnitude less than those for the linear-polarization case for all particle sizes. We note that the trap asymmetry factor should be zero in an ideal case because of the isotropic field distribution of circularly-polarized light in the transverse plane and the homogeneous particles with spherical shapes.

**IV. CONCLUSION**

We investigated polarization-dependent optical forces by using a revised GLMT method that incorporated the general polarization state and the minimum beam spot size. The dependence of the trap anisotropy on the particle size was investigated for a linearly polarized trap beam, and the result was compared with available experimental data; the comparison showed a better agreement than models employed in a previous study [8], especially for large spheres. Our results indicate that the trap stiffness asymmetry factor strongly depends on the particle size and the input beam’s polarization state. The trap anisotropy modified by the presence of a particle can be readjusted via trap beam polarization control to be isotropic. We believe that our investigation of polarization-dependent optical forces may be useful for precise control of particle dynamics in colloidal systems and for realizing more ideal systems for parametric resonance studies in the context of OTs.

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