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Calculation of optical trapping forces on dielectric spheres at an oil-water interface with ray-optics model

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In recent years, there has been increasing interest in investigating colloidal particles trapped at an oil-water interface\(^{[1-3]}\). However, the origin of the interaction forces among particles remains unclear to date. Force measurements are important in understanding the interaction between the colloidal particles at a liquid-liquid interface. The gravity of the particle is negligible compared with the capillary force of liquid interfaces and the optical radiation force exerted on particles was found to be insufficient to induce capillary interactions among particle pairs, and the trapping forces depended on the three-phase contact angle of the particle at the interface. However, manipulating the particles at the liquid interfaces, especially at a rough interface, is difficult because the optical forces are not large enough. In this letter, the trapping forces are calculated with the ray-optics model based on spatial analytic geometry, aiming at improving the transverse trapping force on the particle at an oil-water interface.

Particles suffer from various forces when manipulated by optical tweezers at an oil-water interface, including the capillary force of liquid interfaces and the optical radiation force. The energy of attachment of a particle to an oil-water interface is related not only to the contact angle \(\alpha\) but also to the interfacial tension \(\gamma_{\text{OW}}\)\(^{[10]}\). The gravity of the particle is negligible compared with the capillary force, so the energy \(E_{\text{at}}\) required to remove a particle with radius \(R\) axially from the interface is given by

\[
E_{\text{at}} = \frac{\pi R^2 \gamma_{\text{OW}} (1 \pm \cos \alpha)}{2},
\]

where the sign inside the bracket is negative for removal into the aqueous phase, and positive for removal into the oil phase. For most oil phases, the value of \(\gamma_{\text{OW}}\) is tens pN/\(\mu\)m\(^{[11]}\), and the axial optical stiffness \(k_z\) is 1 pN/\(\mu\)m for typical optical tweezers. Therefore, the energy for removing the particle axially from the interface by optical tweezers \(E_k = \frac{1}{2} k_z R^2\) is much smaller than \(E_{\text{at}}\). It means that the radiation force is too small to overcome the capillary force to manipulate the particle in the axial direction, so axial trapping forces are neglected in the calculation.

The diameter of particles used in the experiment is always larger than the laser light wavelength and the focal spot diameter, so the optical trapping forces on particles can be calculated using a ray-optics model\(^{[12-14]}\). In this model, an incident beam can be decomposed into individual rays, and the effect of diffraction can be neglected. As shown in Fig. 1, an incident ray hits the dielectric particle located at an oil-water interface. The Cartesian coordinate system is based on the focus of an objective lens, the \(z\)-axis is parallel to the beam axis, and the objective focus is set as the origin. The surface of the particle is given by

\[
(x - x_0)^2 + (y - y_0)^2 + (z - z_0)^2 = R^2,
\]

where the point \(O(x_0, y_0, z_0)\) is the center of the particle (also the focus of the laser beam), and \(R\) is the radius of the particle. The unit vector \(\mathbf{u}\) indicating the incident ray from the objective lens is given by

\[
\mathbf{u} = (\sin \theta \cos \varphi, -\sin \theta \sin \varphi, \cos \vartheta),
\]

where \(\theta\) is the angle between the \(z\)-axis and the incident ray, and \(\varphi\) is the angle between the \(x\)-axis and the orthogonal projection of \(\mathbf{u}\) to the plane that contained the focusing lens.

At the point of incidence, \(P_1\), the incident ray is split into two rays: a transmitted ray and a reflected ray. The radiation force caused by reflection and refraction at point \(P_1\) can be expressed as

\[
f_1(\theta, \varphi) = \frac{n_{\text{att}} P(r)}{c} (\mathbf{u}_1 - R_1 \mathbf{u}_1 - T_1 \mathbf{w}_1),
\]

\[
(\mathbf{u}_1) = \left( \begin{array}{c} \sin \theta \cos \varphi \\ -\sin \theta \sin \varphi \\ \cos \varphi \end{array} \right),
\]

\[
(\mathbf{w}_1) = \left( \begin{array}{c} \sin \theta \cos \varphi \\ -\sin \theta \sin \varphi \\ -\cos \varphi \end{array} \right),
\]

\[
(\mathbf{v}_1) = \left( \begin{array}{c} \sin \theta \cos \varphi \\ -\sin \theta \sin \varphi \\ \sin \varphi \end{array} \right),
\]

\[
(\mathbf{g}_1) = \left( \begin{array}{c} \sin \theta \cos \varphi \\ -\sin \theta \sin \varphi \\ -\cos \varphi \end{array} \right),
\]

\[
(\mathbf{f}_1) = \left( \begin{array}{c} \sin \theta \cos \varphi \\ -\sin \theta \sin \varphi \\ -\sin \varphi \end{array} \right).
\]

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where \( n_m \) is the refractive index of the surrounding medium determined by the location of point \( P_1 \), \( R_1 \) and \( T_1 \) are the reflectivity and transmissivity\(^{[15]} \) at point \( P_1 \), respectively, \( v_1 \) and \( w_1 \) are the unit vectors of the reflected and refracted rays, respectively, \( c \) is the velocity of light in free space, and \( P(r) \) is the laser power at a distance \( r \) from the beam axis. The beam is assumed to have a Gaussian intensity distribution at the position of the exit pupil, \( I(r) \), given as

\[
I(r) = I_0 \exp(-2r^2/\omega_0^2),
\]

where \( \omega_0 \) is the waist of the laser beam, and \( I_0 \) is the intensity at the center of the laser beam. The refracted part of the ray \( T_1w_1 \) as a new incident ray reaches the next point \( P_2 \) on the surface, and is reflected and refracted again. The radiation force caused by reflection and refraction at point \( P_2 \) can be obtained using a similar way as in Eq. \((3)\). The reflected ray repeatedly reflects and refracts at the particle-surrounding medium interface with losing intensity. The intensity of rays can be obtained using Fresnel formulae. By repeating this procedure, the optical force \( F \) of a single ray can be determined by the unit vectors of incident rays \( u_1 \), reflected rays \( v_1 \), refracted rays \( w_1 \), and their intensities for all interaction points \( P_i \), so the optical force of a single ray can be written as \( F = \sum_{i=1}^{\infty} f_i \). The optical force \( F \) can be obtained by integrating over \( f \) for all incident rays emitted from the objective lens as

\[
F = \int_0^{2\pi} \int_0^{\omega_0} f(\theta, \varphi)rdrd\varphi.
\]

In our calculation, the refractive indices of the trapped particles and water were 1.55 and 1.33, respectively, and the power of laser with wavelength of 633 nm was 1 W. The radius of the bead was 3 \( \mu \)m, and the initial size of the diameter of the laser beam at the exit pupil of the objective \( d \) was 6 mm. A laser beam is usually polarized in an optical tweezers system. Laser beams with different polarized states in the calculation may lead to different calculation results\(^{[16,17]} \). The intensity of the s-polarized component was assumed to be equal to the intensity of the p polarized component in this calculation.

The preferred emulsion type depends on the particle wetting properties, and this can be described by a three-phase contact angle \( \alpha \) as shown in Fig. 1. Particle wetting properties are of paramount importance: hydrophilic colloids tend to stabilize oil-in-water (O/W) emulsions, while water-in-oil (W/O) emulsions are better stabilized by oil-wettable particles. Figure 2 shows the transverse trapping force exerted on the particle at the interfaces as the three-phase contact angle is varied. This calculation assumed that \( n_2 = 1.40 \) and numerical aperture \( NA = 0.7 \). A negative trapping force indicates a restoring force directed back to the trap center. \( F_x \) can be seen to decrease with the increase in \( \alpha \). The increase in \( \alpha \) means that the number of interaction points \( P_i \) at oil phase increases and the total momentum transferred from the laser to the particle decreases. This decrease leads to the reduction in trapping force.

In Fig. 3, the transverse trapping force \( F_x \) is plotted as a function of transverse position for several refractive indices of the oil phase. This calculation assumed that \( NA = 0.7 \) and \( \alpha = 90^\circ \). The refractive indices of the conventional oil phase are larger than those of water, so in all of the calculations, \( n_2 > 1.33 \). The value of \( n_2 = 1.33 \) means that the particle is immersed in water totally. \( F_x \) decreases with the increase in \( n_2 \), as shown in Fig. 3. The increase in \( n_2 \) means that the total momentum transferred from the laser beam to the particle diminishes, which reduces the trapping forces.

To fully utilize the well-accepted properties of optical tweezers which are used to trap the particles at the oil-water interface, larger transverse trapping forces are always better. In practice, the three-phase angle and the refractive index of the oil phase can be altered, so \( F_x \) can be enhanced by changing the oil. Furthermore, the following calculation shows that \( F_x \) can also be improved by adjusting the parameters of the optical tweezers system. It can be obtained by decreasing the NA of the
the particles located at an oil-water interface, medium when the trapped particle was immersed in a single was decreased, the transverse trapping forces increased laser beam. microscope objective or shrinking the diameter of the laser beam.

Previous investigations have indicated that when NA was decreased, the transverse trapping forces increased when the trapped particle was immersed in a single medium. When optical tweezers are used to trap the particles located at an oil-water interface, \( F_x \) varies with the transverse displacements for a different NA of the microscope objective, as shown in Fig. 4. This calculation assumed that \( n_2 = 1.40, \alpha = 90^\circ \), and the result indicated that \( F_x \) can be improved by decreasing the NA of the objective. According to Refs. [9,12], an increase in incidence angle \( \beta \) will increase the transverse trapping forces. \( \beta \) of the ray at the edge of the beam is increased with decreasing the NA, so we can improve \( F_x \) by decreasing NA when NA > 0.5. It should be noted that when the NA is lower than 0.5, the diameter of the laser beam is very small, and the transverse trapping forces cannot be calculated with the ray optics model again. According to the above analysis, the transverse trapping force can be improved by increasing the incidence angle of the rays at the edge, so the improvement in the transverse trapping force can be obtained by shrinking the diameter of the laser beam. Figure 5 shows the transverse trapping force \( F_x \) as a function of transverse position for several beam diameters. The calculation assumed that \( \alpha = 90^\circ, n_2 = 1.40 \), and \( \alpha = 90^\circ \). Furthermore, the ray optics model is not suitable for calculating the trapping force when the diameter is too small.

We have calculated only the transverse trapping forces on a particle at a smooth interface. The deformation of liquid surfaces caused by gravity of the particles and laser-induced time-varying temperature distribution was neglected. However, the simulation can approximate the true state if interfacial properties are considered. Our results can be used to analyze the trapping ability despite its limitation.

In conclusion, we have calculated the transverse trapping forces on a dielectric particle at an oil-water interface based on the ray-optics model. The calculation results show that the forces can be increased by altering the oil phase to decrease the parameters of either \( \alpha \) or \( n_2 \). \( F_x \) can also be improved by either decreasing the NA of the objective or shrinking the diameter of the laser beam. We believe that the results will be helpful in manipulating the particles and investigating the interaction forces of colloidal particles at liquid interfaces.

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