Dynamics analysis of nanoparticles optically driven by a Laguerre-Gaussian beam with optical spin

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\textbf{Abstract.} We theoretically discover that the spin angular momentum (SAM) enables to modulate the orbital torque through the inter-particle light-induced force (IP-LIF). Laguerre-Gaussian beam with the orbital angular momentum (OAM) can induce the orbital motion for the optically trapped objects. In addition, the SAM can also accelerate or decelerate the orbital motion due to the IP-LIF. Our discovery provides a new physical aspect, i.e. the IP-LIF plays an important role in the many-body dynamics of nanoparticles via the SAM-OAM coupling.

1. Introduction

Laguerre-Gaussian (LG) beam with a helical wave-front carries an orbital angular momentum (OAM). The OAM enables the orbital motion of trapped objects, i.e. the OAM transfer effect [1], in optical tweezers, allowing a contactless manipulation of small objects by a focused laser [2]. The spinning motion can also be induced by a SAM associated with the circularly polarized light. However, the orbital and spinning motions are not independently determined by the OAM and SAM. For example, it was reported that the OAM forces the spinning motion of a trapped object with a sufficiently large size in comparison with the laser beam radius. Thus, a spinning velocity of the trapped object should be determined by the total angular momentum defined as the sum of OAM and SAM [3]. The circularly polarized LG beam with both positive (or negative) OAM and SAM deforms the irradiated surface of photo-sensitive organic material, such as azo-polymer, to establish helical structures, whereas the LG beam with either of OAM and SAM with a negative sign does not allow such helical mass-transport [4]. The above mentioned SAM-OAM coupling effects result from the mutual contribution of the both OAM and SAM.

In recent years, we discovered that the SAM contributes to the orbital motion of the trapped nanoparticles (NPs), in which, through the inter-particle light-induced force (IP-LIF) [5–7], the orbital motion of trapped NPs is accelerated or decelerated by SAM of the left- or right-circular polarization (LCP or RCP). Our model based on the IP-LIF is entirely different from the conventional SAM-OAM coupling in the tightly focused circularly polarized LG beams previously reported in [8], and it should provide a new physical insight concerning interaction between optical fields and matter. In this paper, we reveal that the SAM certainly contributes to the orbital torque through the IP-LIF.
2. Theoretical framework
As the target of this study, the single NP and paired NPs (gold, diameter 70 nm) are irradiated with the circularly polarized LG beam. Based on the manner of discrete dipole approximation, NPs are discretized by the cubic cells (edge length: 2nm) with the induced polarization $P_i(\omega) = \chi(\omega)E_i(\omega)$, where $i$ is the index of cells. To consider the gold NPs dispersed in the water, the electric susceptibility is given by $\chi(\omega) = \varepsilon_p(\omega) - \varepsilon_{med}$ with the water dielectric constant $\varepsilon_{med} = 1.33^2$ and the gold dielectric function by the Drude-critical points model [9] with fitting the experimental result [10]. $E_i(\omega)$ is the response field expressed as,

$$E_i(\omega) = E^{inc}(r_i, \omega) + \sum_{j \neq i} G(r_i-r_j, \omega) \cdot P_j(\omega)V_j + S_i(\omega) \cdot P_i(\omega),$$

where $E^{inc}(r, \omega)$ is the electric field of incident light, $G(r, \omega)$ is the Green’s function of electromagnetic field in a homogeneous medium, $V_i$ is the volume of $i$-th cell, and $S_i(\omega) \cdot P_i(\omega) = \int_{V_i} G(r, \omega) \cdot P(r, \omega) dr$ is the analytical integration of $G(r, \omega)$ for $j = i$. For the circularly polarized LG beam propagating in $+z$-direction, $E^{inc}_r(r, \omega)$ is expressed as $E^{inc}(r, \omega) = [-\sqrt{2}i w_0 Q(z, \omega) \rho] e^{i k_p z} E^G(r, \omega)$ for the arbitrary $\ell$ and $p = 0$, where $\rho$ and $\varphi$ are variables of cylindrical coordinates. $E^G(r, \omega) = -E_0 i w_0^2 Q(z, \omega) \exp[iQ(z, \omega)\rho^2 + ikz]$ is the expression of Gaussian beam, $w_0 = 0.5 \mu m$ is the spot radius, $k$ is the wavenumber in the medium, $Q(z, \omega)$ is written by $Q(z, \omega) = k/(2z - ik w_0^2)$, and $E_0 = \varepsilon \sqrt{2\pi l_0/\varepsilon_{med}} (l_0 = 2P/|l|^2 \pi w_0^2, \ P = 1 \ W$ is power) gives the polarization and amplitude of electric field. The polarization vector is assumed as $\varepsilon = (1, \pm i, 0)/\sqrt{2}$, where + and − correspond to the LCP and RCP, respectively. From $\sigma h = \frac{\hbar}{2}(\varepsilon \times \varepsilon^*)_z = \pm h$, LCP is $\sigma = 1$ and RCP is $\sigma = -1$ for the SAM. Under these conditions, $P_i(\omega)$ and $E_i(\omega)$ can be obtained by self-consistently solving the simultaneous equations.

Based on the general expression of time-averaged LIF derived from the Lorentz force [7], the LIF acting on $i$-th cell is described as,

$$\langle F_i^{cell}(\omega) \rangle = \frac{1}{2} Re \{[\nabla E_i(\omega)]^* \cdot P_i(\omega) V_i \}.$$  \hspace{0.5cm} (2)

In addition, a sum of equation (2) yields the LIF acting on $k$-th NP,

$$\langle F_k^{NP}(\omega) \rangle = \sum_{i \in M_k} \langle F_i^{cell}(\omega) \rangle = \frac{1}{2} \sum_{i \in M_k} \sum_{j \notin M_k} \nabla E^{inc}(r_i, \omega) + \nabla G(r_i-r_j, \omega) \cdot P_j(\omega)V_j \rangle \cdot P_i(\omega) V_i \},$$  \hspace{0.5cm} (3)

where $M_k$ is a set of the cell indices composing the $k$-th NP, and equation (1) is substituted for $E_i(\omega)$, while the differential of third term in equation (1) is negligible. In [ ] of equation (3), the first term is caused by the incident light, whereas the second term is caused by the scattered light via the other cells, where the sum for $i$, $j \in M_k$ is approximately cancelled because of the action-reaction forces between dipoles. Therefore, the second term originated only from the cells composing other NPs, is namely IP-LIF, which is important for considering the orbital torque from the SAM. Then, the total torque around $z$-axis acting on NPs is simply expressed as,

$$\langle \tau_z(\omega) \rangle = \sum_{i} \left[ r_i \times \langle F_i^{cell}(\omega) \rangle \right]_z.$$  \hspace{0.5cm} (4)

However, to consider the mechanical motion of NPs, they should be regarded as a rigid body whose translational motion is driven by the force acting on its center of mass (CM): $\langle F_k^{NP}(\omega) \rangle$. Thus, this consideration yields another definition of orbital torque based on $\langle F_k^{NP}(\omega) \rangle$,

$$\langle T_z(\omega) \rangle = \sum_k \left[ r_k^{CM} \times \langle F_k^{NP}(\omega) \rangle \right]_z = \sum_k \left[ r_k^{CM} \times \sum_{i \in M_k} \langle F_i^{cell}(\omega) \rangle \right]_z,$$  \hspace{0.5cm} (5)

where $r_k^{CM} = (x_k^{CM}, y_k^{CM}, 0)$ is the CM position of $k$-th NP.
3. Orbital torque acting on the nanoparticles

Based on the configuration of figure 1(a), we estimated numerically the optical torque for the single NP by employing equation (4), and we found that the LCP-induced optical torque is relatively higher than the RCP-induced one at any wavelengths as shown in figure 1(b). In contrast, the equation (5) provided us that LCP-induced optical torque is fully identical with the RCP-induced one as shown in figure 1(c). The origin of this difference is the LIF caused by the second term of right-hand side in equation (1), which contributes to the spinning of NPs around the CM of themselves under the circular polarization. In the equation (3) for the single NP, this term is cancelled because of the sum for $i,j \in M_k$ (of course, the torque for spinning is not cancelled). Therefore, equation (4) using the LIF before the cancellation yielded the different torque for LCP and RCP, whereas equation (5) after the cancellation yielded the identical torque. This indicates the orbital motion of CM of single NP is not modulated by the SAM of circular polarization, where the IP-LIF is absent.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{(a) Intensity distribution of $E_{inc}(r,\omega)$ and the configuration of single NP (diameter $D = 70$ nm) located at the intensity maximum ($r_1^{CM} = (w_0/\sqrt{2},0,0)$). (b)(c) Torque calculated by equation (4) and (5), respectively. Red solid and blue dashed lines were for LCP and RCP.}
\end{figure}

Next, as shown in figure 2, based on the configuration (a), the orbital torque acting on paired NPs was calculated by the equation (5). Although the torque was calculated for the LIF acting on the CM of NPs, there was a clear difference between LCP and RCP. This indicates the faster orbital motion can be expected for LCP, if the NPs keep the same relative position in the cylindrical coordinates during the motion.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{(a) Configuration of NPs, where they were located at the same $x,z$ position with figure 1(a), but different $y$ position: $y_k^{CM} = \mp (D + g)/2$ (− for NP1 and + for NP2, gap length $g = 10$ nm). (b) Torque calculated by equation (5) (averaged over the NPs by $N_p = 2$).}
\end{figure}

The reason why the torque for LCP and RCP differed is due to the presence of IP-LIF given by the second term in [] of equation (3). Figure 3 shows the LIF acting on the paired NPs, including IP-LIF. As shown in figure 3(c) and (d), the $y$-component of LIF ($\langle F_{NP}^{y;k}(\omega) \rangle$) was identical for LCP and RCP. In both cases, the NPs were attracted each other due to the IP-LIF. In addition, the IP-LIF was cancelled in the sum because of the action and reaction. The remained positive value of sum was caused by the OAM and would transport NPs for $+y$-direction in this configuration.
On the other hand, the $x$-component of LIF $\langle F^{NP}_{x,k}(\omega) \rangle$ was inverted for LCP and RCP, where the zero of summed LIF indicated that $\langle F^{NP}_{x,k}(\omega) \rangle$ was originated only from the IP-LIF. Under the circularly polarized light with SAM, the NPs are rotated around not only each CM, but also the CM of paired NPs as the two-body problem. Thus, the IP-LIF rotates NPs anti-clockwisely and clockwisely for the LCP and RCP. In particular, since the NPs were not located on the $x$-axis, $\langle F^{NP}_{x,k}(\omega) \rangle$ also remained in equation (5) as $r^CM_k \times \langle F^{NP}_{y,k}(\omega) \rangle = x^CM_k \langle F^{NP}_{y,k}(\omega) \rangle - y^CM_k \langle F^{NP}_{x,k}(\omega) \rangle$, and modulated the orbital torque. Given $y^CM_k = \mp (D + g)/2 = \mp 0.04 \mu m$ and $\langle F^{NP}_{x,k}(\omega) \rangle = \pm 70 \mu N$ for the peak wavelength 600 nm in figure 2(b), where the factor of 2 came from the difference between LCP and RCP at 600 nm in figure 2(b), where the factor of 2 came from the difference of SAM: $+\pi - (-\pi) = 2$, while the factor of 2 from $N_p = 2$ was not considered because figure 2(b) showed $\langle T_z(\omega) \rangle/N_p$. As above, since $\langle F^{NP}_{x,k}(\omega) \rangle$ originated only from IP-LIF, therefore, we can conclude that the difference in the orbital torque in figure 2(b) is originated from the SAM through the IP-LIF.

![Figure 3](image-url) LIFs acting on each NP and their sum. (a)(b) $x$-component and (c)(d) $y$-component of LIF for (a)(c) LCP ($\sigma = 1$) and (b)(d) RCP ($\sigma = -1$) are shown.

4. Conclusion

We have discovered that the SAM enables the modulation of the orbital torque through the IP-LIF. In particular, the IP-LIF contributes significantly to the CM motion of NPs. These results, manifesting the mutual induction of mechanical motion by the SAM and OAM, contribute to the further development of laser manipulation and laser materials processing by using LG beams.

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