Invited Paper

Optical Manipulation by Nonlinear Response of Nanoparticles

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Abstract

We investigate the optical manipulation of nanoparticles with the resonant nonlinear optical response. Efficient trapping of nanoparticles observed in experiments under the resonance condition is elucidated by considering optical nonlinearity. Also, we propose the flexible optical manipulations of nanoparticles that have gain by optical pumping. The pulling force and the rotational switching are demonstrated, where the stimulated emission from nanoparticles with inverted population is considered. These results show that utilizing nonlinear optical effect will greatly enhance the degrees of freedom to manipulate nanoparticles.

Keywords: optical force, laser tweezers, nanoparticle, nonlinear optical response, resonant trapping, optical vortex

I Introduction

The targets of optical trapping have been recently expanded to nano-sized objects. It is generally difficult to trap nanoparticles because the optical force is inversely proportional to the object volume in the nanoscale regime, which cannot exceed the force received from the environment. To overcome this problem, the optical manipulation using an electronically resonant light has been theoretically proposed, where the optical force is resonantly enhanced, and further, it makes possible to select particles with particular quantum mechanical properties due to the electronic confinement effect [1]. Actually, successful optical transport of nanoparticles was experimentally reported, where the pulsed laser light with a frequency resonant with excitonic states of nanoparticles was used [2]. On the other hand, as for trapping nanoparticles, it is known that the use of the resonant condition is not favorable if only the linear optical response is considered [3]. This is because the dissipative force to push the particle from the focal spot becomes more prominent than the gradient force under the resonant condition. However, some experimental studies reported positive results of the resonant optical trapping [4-6]. The discrepancy between the experimental and theoretical results has been solved by considering the nonlinear optical response [7]. The theory including nonlinear effect successfully explains existing experimental results, and recently, it has been experimentally demonstrated that the laser blue shifted from the resonance energy of nanoparticles more efficiently trap them than the red shifted one due to nonlinearity [8]. Now, the above results show that the role of optical nonlinearity has reality, and hence, we can greatly enhance the number of degrees of freedom of the optical manipulation by introducing the nonlinear effect. Here, we propose unconventional manipulation schemes of nanoparticles by using nonlinear optical response, where the pulling force and the rotational switching are demonstrated by considering the gain of nanoparticles by optical pumping.

II Efficient trapping

Although the theory within the linear response shows the difficulty in trapping molecules under the resonant condition [3], experimental results indicate positive effects for trapping molecules due to resonant optical response [4-6]. Considering optical nonlinearity beyond the perturbative regime, it has been clarified that dissipative force is saturated with the increase of the incident light intensity, and further, the sign of the gradient force becomes inverted due to the population inversion of the excited state, which leads to a favorite condition for the efficient trapping [7]. Here, we examine the kinetic motion of nanoparticles under resonant light

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with using Brownian dynamics simulations. The exerted force is calculated according to the following expression of the time-averaged force as $\langle \mathbf{F}(\omega) \rangle = (1/2)\mathrm{Re}\{\int d\mathbf{r}\,[\nabla \mathbf{E}(\mathbf{r},\omega)^* \cdot \mathbf{P}(\mathbf{r},\omega)]\}$ [12], where **E** and **P** are the time-harmonic electric field and the induced polarization, respectively. **E** and **P** are obtained by solving the density matrix equation of the nanoparticle system considering nonlinearity beyond perturbative regime. Assuming the parameters of typical dye molecules, we obtain the following results: (1) The molecules far from the focal spot are much more efficiently drifted toward the spot due to the resonant effect than the case of the molecules without resonance. (2) The motions of the drifted molecules are braked at the focal spot and they are trapped due to the attractive force arising from the optical nonlinearity, which cannot be seen in the calculation with only the linear response. These results elucidate the mechanism of the efficient trapping of molecules under the irradiation of resonant light.

III Manipulation of nanoparticles with gain

By considering the optical nonlinearity, it will be possible to enhance the degrees of freedom to manipulate nanoparticles. In our previous study, we have proposed a pulling force exerted on the molecules with gain [7]. If the molecules are pumped and the population is inverted, stimulated emission occurs when irradiated by the manipulation laser, which results in the pulling force. We demonstrate this situation by using the Brownian dynamics simulations assuming the same parameters as the previous section. The results show that the trapped molecules are pulled toward the light source after the pumping, which can be understood from the momentum conservation law.

Then, we demonstrate the similar mechanism for the rotational motion. If we irradiate the nanoparticles with the Laguerre-Gaussian (LG) beam (Optical vortex beam), the particle simply rotates counterclockwise as shown in Fig. 1(c). Then we consider the situation where the particle is strongly pumped as depicted in Fig. 1(b). As shown in Fig. 1(d), when the particle is pumped, it receives the force to rotate clockwise, namely, the direction of rotation becomes inverted. This is because the population inversion is induced by the pumping, and the

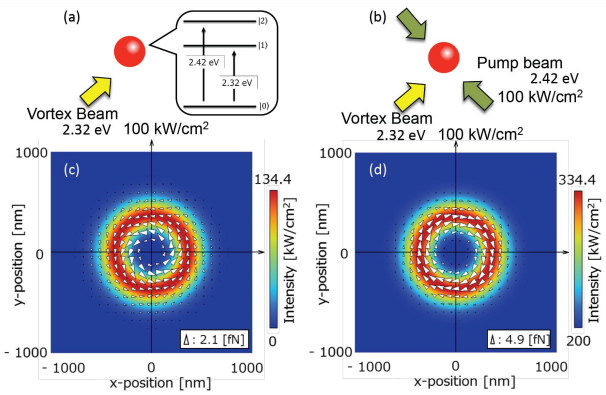

Fig.1: (a) Schematic view of the configuration of lasers and the target. (b) The configuration when the target is pumped. (c) Map of the optical force in the x-y plane in the absence of the pump beam. (d) Map of the optical force in the x-y plane in the presence of the pump beam.

stimulated emission occurs. In this case, the rotation direction of rotation is inverted because of the conservation low of the angular momentum.

IV Summary

We theoretically investigate optical manipulation of nanoparticles by resonant nonlinear optical response. We demonstrate an efficient optical trapping by using nonlinear effect, which is in striking contrast with the prediction by the theory with only the linear response. We also propose the flexible manipulation of nanoparticles with gain, where the pulling force and the rotational switching are demonstrated. These demonstrations show that utilizing nonlinear optical effect will greatly enhance the degrees of freedom to manipulate nanoparticles.

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