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# *Actio et reactio* in optical binding

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**Abstract:** The symmetry in action and reaction between interacting particulate matter breaks down when the interaction is mediated by an out-of-equilibrium environment. Nevertheless, even in this case, the space translational invariance still imposes the conservation of canonical momentum. Here we show that optical binding of an asymmetric material system can result in non-reciprocal interactions between constituents. We demonstrate that a non-conservative force applies to the center of mass of an optically bound dimer of dissimilar particles, which leads to an unexpected action in the transversal direction. The sign and the magnitude of this positional force depend on the abrupt phase transitions in the properties of the asymmetric dimer.

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**OCIS codes:** (290.5850) Scattering, particles; (350.4855) Optical tweezers or optical manipulation.

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## 1. Introduction

The structure of matter is determined by bonding of its parts. This bonding is commonly treated in terms of the pair-potential energy of interaction between identical particles, which leads to structure formation at different scales: atomic, molecular, etc. The interaction forces derived from this potential energy are conservative. It is also known that, when illuminated by external field, microscopic particles can form stable structures due to their mutual interaction and the corresponding optical forces. This effect, known as optical binding, ceases when the external field is shut down. Optical binding was first discussed and observed in [1] and, since then, different aspects of optical binding have been studied [2,3].

Optical binding is usually described in terms of a potential landscape with periodic minima and maxima [4]. Particles in this landscape reside in energy minima and tend to maintain certain particle-particle distance. Following this description, the potential forces acting on two bound particles are equal and oppositely directed. However, this is not always the case as we will show in the following. The forces on optically interacting but unequal particles are not necessarily reciprocal, which, at first glance appears to violate the *actio et reactio* principle.

The apparent inconsistency can be resolved if one realizes that particles interacting with the external field constitute an open system. Optical forces between particles are mediated through their interaction with the external field and incorporate not only conservative, but also nonconservative parts. One of the consequences of the asymmetric interaction with the optical environment is the occurrence of a nonzero force acting on the center of mass of the bound system. The presence of such type of force was theoretically predicted for a pair of emitting dipoles placed nearby [5] and it also suggested for longitudinal optical binding [6]. In the past, we have shown that the action of nonconservative forces determines nonzero torques leading to a rotational motion of optically bound particles which are illuminated with circularly polarized light [7]. In this Letter we demonstrate that nonconservative forces can induce translational motion of optically bound matter perpendicular to the direction of light illumination.

By itself, the oblique trajectory of an object with respect to direction of illumination is not surprising. It is a well-known behavior of anisotropic particles placed in a beam of light [8]. Recently, the possibility of transversal action of radiation pressure was demonstrated for semi-cylinders in the limit of geometric optics (“optical lift”) [9] and for chiral particles in the presence of surface [10]. We have shown that a transversal optical force can also be generated for multiple scattering objects by manipulating scattering directionality with structured light [11]. Hereinafter we will demonstrate that a transversal force can be created for a dimer consisting of isotropic spherical particles even when these particles do not form a rigid object.

## 2. System of dissimilar particles in external field

To qualitatively understand why a transversal force can appear in optical binding, let us consider a system of two dipole particles with scalar polarizabilities  $\alpha_1$  and  $\alpha_2$ . An external monochromatic plane wave illuminates this system perpendicular to the axis connecting the dipoles as shown in Fig. 1. In these conditions, the electric fields at the positions of the dipoles can be expressed as [12]:

$$\begin{cases} \mathbf{E}_1(\mathbf{r}_1) = \mathbf{E}_{0l} + \mathbf{G}(R)\alpha_2\mathbf{E}_2(\mathbf{r}_2) \\ \mathbf{E}_2(\mathbf{r}_2) = \mathbf{E}_{0l} + \mathbf{G}(R)\alpha_1\mathbf{E}_1(\mathbf{r}_1) \end{cases} \quad (1)$$

where  $\mathbf{G}(R)$  is the Green’s dyad and  $\mathbf{E}_{1,2}$  are the electric fields at the locations of dipoles  $\mathbf{r}_1$ ,  $\mathbf{r}_2$ ;  $R = |\mathbf{r}_2 - \mathbf{r}_1|$ . When the incident wave  $\mathbf{E}_{0l}$  is polarized perpendicularly or parallel to the dimer’s axis, a scalar version of the vector equations in Eq. (1) suffices and the system’s solution becomes

$$\begin{cases} E_1 = E_{0l} \frac{1 + \alpha_2 G}{1 - G^2 \alpha_1 \alpha_2} \\ E_2 = E_{0l} \frac{1 + \alpha_1 G}{1 - G^2 \alpha_1 \alpha_2} \end{cases} \quad (2)$$

with  $G$  being the transversal or longitudinal component of the tensor of the Green's function [12].

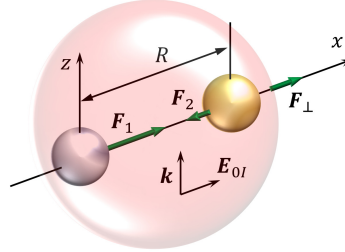


Fig. 1. Geometry of the optical bound system. Two dissimilar particles experience different forces  $\mathbf{F}_1 \neq \mathbf{F}_2$  when illuminated by an external field  $\mathbf{E}_{0l}$ . As a result, a transversal optical force  $\mathbf{F}_\perp$  acts on the whole system (represented by semitransparent sphere).

When there is no interaction, the only force acting on the isotropic particles is the radiation pressure component along the direction of wave's incidence. However, a quite different situation develops when the interaction is taken into account as described by Eqs. (1) and (2). In particular, a transversal force acting on a system of particles can appear as indicated in Fig. 1. This transversal optical force can be found by summing over all the acting forces [13,14]:

$$F_\perp = \frac{1}{2} \sum_{j=1,2} \text{Re} \left( \alpha_j \mathbf{E}_j \frac{\partial \mathbf{E}_j^*}{\partial r} \right) \quad (3)$$

Here  $r$  represents the coordinate along the dimer's axis. Using the fields in Eq. (2), one obtains the following expression for the force on the center of mass:

$$F_\perp = -E_{0l}^2 \text{Im} \left[ \frac{\partial G}{\partial r} \right] \frac{\text{Im}(\alpha_1 \alpha_2^*) + |\alpha_2|^2 \text{Im}(\alpha_1 G) - |\alpha_1|^2 \text{Im}(\alpha_2 G)}{|1 - G^2 \alpha_1 \alpha_2|^2} \quad (4)$$

From Eq. (4), one can conclude that, in general, dissimilar particles experience forces that do not compensate each other, which results in an overall movement of the system. It can be easily checked that the force in Eq. (4) is zero for identical particles ( $\alpha_1 = \alpha_2$ ).

The presence of transversal force can be also explained by considering scattering pattern of a dimer of dissimilar particles. Equivalently, the transversal force on a system of particles could be calculated through the scattering function as [8]

$$F_\perp = -\frac{\varepsilon}{2k^2} \int_{\Omega} |\mathbf{f}(\hat{k}_s = \hat{r}, \hat{k}_l)|^2 \hat{r}_\perp d\Omega \quad (5)$$

where  $k$  is the wavenumber,  $\varepsilon$  is the dielectric permittivity,  $\mathbf{f}(\hat{k}_s, \hat{k}_l)$  denotes the scattered field in the far zone:  $\mathbf{E}_s = [\exp(ikR)/(kR)] \mathbf{f}(\hat{k}_s, \hat{k}_l)$ . The integration in Eq. (5) is performed over a solid angle of  $4\pi$  steradians;  $\hat{r}$  is a unit vector corresponding to angle of integration and  $\hat{r}_\perp$  is its projection onto the dimer's axis. Formula (5) clearly shows that the transversal force on a dimer is determined by the radiation pattern of the scattering system. The presence

of unequal particles makes this pattern asymmetric, as shown in the insert in Fig. 2, creating a nonzero transversal force. Thus, the seemingly violated *actio et reactio* principle for the interacting particles is restored for the entire system “particles plus field” as part of momentum is carried away by asymmetrically scattered electromagnetic field. Another important conclusion can be derived from Eq. (5). Because in most cases one can simultaneously perform measurements on the field and on the particles, optical experiments offer unique possibilities to examine the role of conservation laws and symmetries in complex interacting systems.

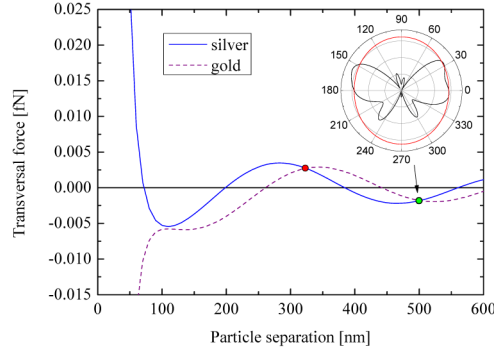


Fig. 2. Transverse component of optical forces acting on each of interacting particles within a dimer illuminated perpendicular to its axis by a plane wave ( $\lambda = 342\text{nm}$ ) with electric field strength  $E = 10^6 \text{ V/m}$ . Polarization of the wave is aligned perpendicular to the dimer's axis. The dimer consists of dipolar, spherical particles of Au and Ag having 20nm in diameter. The green and red dots indicate the particle-particle separations of stable and unstable equilibriums, respectively. The insert shows the scattering diagram for particles in stable equilibrium. The scattering angle is measured with respect to the dimer's axis (x-axis in Fig. 1).

An example of transversal force calculation for an asymmetric dimer is illustrated in Fig. 2. One can see that indeed two bound particles experience, in general, nonreciprocal forces. The equilibrium relative positions of particles in dimer can be found from the condition of equality of forces acting onto each of particles:

$$\Delta F_{\perp} = F_{2\perp} - F_{1\perp} = 0 \quad (6)$$

The stable positions of optical binding are determined by the condition  $\partial\Delta F_{\perp} / \partial r < 0$ . Including additional effects such viscous drag or hydrodynamic interactions into condition (6) is straightforward but it is beyond the scope of this paper.

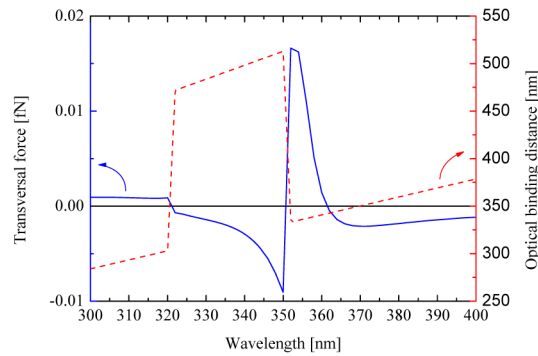


Fig. 3. Force acting on a center of mass of optically bound dissimilar particles and stable particle-particle separation of optical binding as functions of illumination field wavelength. Gold and silver particles are of equal diameter 20 nm and are placed in vacuum. External wave with electric field strength  $10^6 \text{ V/m}$  is polarized perpendicular to the dimer axis. Optical constants for gold and silver are taken from [15].

One can get more insights regarding the properties of transversal force by examining the case of small polarizabilities, i.e.  $|\alpha_i G| \ll 1$ . In this case one can disregard the last two terms in Eq. (4) and, rewriting  $\text{Im}(\alpha_1 \alpha_2^*)$  as  $-\alpha_1' \alpha_2'' + \alpha_2' \alpha_1''$ , it becomes obvious again that  $F_{\perp} \neq 0$  when one of the particles acts as radiation damper, i.e.  $\alpha'' \neq 0$ . Interestingly, in terms of maximizing the transversal force, an even better condition occurs when the real parts of polarizabilities  $\alpha_i'$  are of different signs. This dispersive effect is illustrated in Fig. 3 where we show transversal force for optically bound plasmonic (silver and gold) particles as a function of wavelength  $\lambda$ . Two features are noticeable in this graph. First, the magnitude of the force increases near the plasmonic resonance of silver particle (around  $\lambda = 360$  nm) where the imaginary part of the polarizability  $\alpha''$  increases significantly. Second, around this plasmonic resonance one can observe a sharp change in the force direction. This change is another unique feature of a system of optically bound but dissimilar particles; it corresponds to the structural phase transition for the case when the real part of polarizability of the one of the particles (silver in our case) changes its sign. This leads to changing the sign of  $\partial \Delta F_{\perp} / \partial r$ , the stable equilibria becomes unstable and vice versa, and, consequently, the distance corresponding to stable binding changes abruptly as can be seen in Fig. 3.

Let us note that the requirement for a nonzero imaginary part of polarizability does not necessarily imply that particles should be absorbing. The reactive dissipation required by energy conservation imposes an imaginary component of the polarizability even in the absence of absorption [16,17]. Accounting for radiation reaction the Rayleigh polarizability is

$$\alpha = \alpha_0 \left( 1 - \frac{2i}{3} \frac{1}{4\pi\epsilon} k^3 \alpha_0 \right)^{-1}$$

with  $\alpha_0$  being the static polarizability, which may or not be a complex quantity. It is also important to note that the nonreciprocal interaction for dielectric nonabsorbing particles extends even beyond the dipole approximation. To demonstrate that a transversal force exists for large nonabsorbing particles, we evaluated the interaction force between two particles having the same size but different refractive indices. Using a multiple-sphere T-matrix code [18], we calculated the field corresponding to a pair of silica (refractive index 1.49) and  $\text{TiO}_2$  (refractive index 2.76) particles placed in water. The forces acting on particles were then evaluated by integrating the Maxwell stress tensor over surrounding surfaces [19]. As can be seen from Fig. 4, the transversal forces for optically bound dimers of dissimilar particles can be much larger than that for dipolar particle.

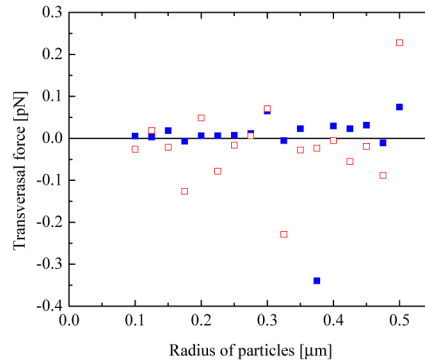


Fig. 4. Transversal force on an optically bound dimer consisting of two nonabsorbing dielectric particles with refractive indices of 1.49 and 2.76 and varying radii. The force is evaluated at the first bounding positions corresponding to each particle size. The particles are placed in water (refractive index 1.33), the external field has a strength of  $10^6$  V/m and vacuum wavelength 532nm, and the polarization is perpendicular (hollow red squares) or parallel (filled blue squares) to the dimer's axis.

The transversal force can acquire different values, both positive and negative, depending on stable equilibrium particles currently occupy. Another remarkable feature is the fact that in certain cases the direction of transversal force can be switched by the change of electric field polarization. This characteristic can provide a convenient mechanism for controlling the direction of the force.

### 3. Conclusions

In conclusion, we demonstrated that two dissimilar and optically interacting particles experience in general nonreciprocal interaction forces. This apparent violation of “action and reaction” principle is due to the presence of non-conservative optical forces which appear due to the interaction with the external field. One of the consequences of interaction forces being nonreciprocal is the appearance of side force in transversal optical binding. The transversal force exists even when the dimer is composed of spherically symmetric particles. In the case of dipole particles, the sign and magnitude of the side force is determined by the sign of real part of particles’ polarizabilities and by the magnitude of their imaginary parts. If the sign of the real part of polarizability changes (for example, because of dispersion), the optically bound dimer can demonstrate structural phase transition with particle-particle separation and magnitude and direction of the side force abruptly modifying.

We also demonstrated that this transversal force is present even when the bound particles are nonabsorbing. Because optical forces are generally determined by the volume of interaction with the electromagnetic field, the transversal force increases for larger particles. This may be of interest for certain applications of micro-manipulation with a single illumination channel where the rotational and translational movement can be easily adjusted by controlling radiations’ polarization or wavelength.

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