Dynamic consequences of optical spin–orbit interaction

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Field symmetries and conservation laws are closely associated through Noether’s theorem. Light field inhomogeneities lead to changes in linear and angular momenta and, consequently, to radiation pressure\textsuperscript{3,4}, spin or rotation of objects\textsuperscript{3,4}. Here we discuss a new type of mechanical action originating in the exchange between spin and orbital angular momenta. We demonstrate theoretically and experimentally that, when mirror and central symmetries of scattering are broken, a force appears acting perpendicularly to the direction of propagation. This new force completes the set of non-conservative forces (radiation pressure and tractor beams) that can be generated with unstructured light beams.

When an optical wave interacts with matter, there is an exchange between the spin and orbital parts of the angular momentum carried by the wave. This optical spin–orbit interaction (SOI) is responsible for a number of wave polarization effects\textsuperscript{3,6}. For instance, when the azimuthal symmetry of the field is preserved around an axis, the projection of the total angular momentum along that axis is conserved according to Noether’s theorem. As a result, the mechanical action on matter is due to the linear momentum, and points along this axis of symmetry. When the rotational symmetry is broken as a result of the interaction, the direction of the mechanical action is affected such that the conservation of canonical angular momentum is obeyed.

The field ‘mirror symmetry’ can be perturbed even when interacting with rotationally symmetric objects. A vortex field emerges that is centred at the location of a spherical scatterer when a circularly polarized plane wave is incident on it\textsuperscript{7}. This vortex structure arises because of a partial transformation of spin to orbital angular momentum. Previously, we demonstrated that, because of this swirling around a sphere, the power flow experiences a shift analogous to the spin-Hall effect of light\textsuperscript{8}. The field asymmetry during this SOI was tested by directly measuring the light to the right or to the left of a scattering particle with an optical fibre. A variation of this experiment was later proposed\textsuperscript{9,10} in which the measurement fibre is replaced with different waveguide structures and interfaces. In all these publications it was demonstrated that the rotational symmetry of light scattering can be broken by the presence of nearby interfaces.

As a consequence of angular momentum conservation, spin–orbit transformations could lead to mechanical effects. For example, the transverse force exerted by a medium on a field was predicted when spin–orbit transformations occur during light reflection and refraction (spin–Hall effect of light)\textsuperscript{11}. In another example, on being illuminated by circularly polarized light, two interacting spheres would experience spin and orbital torques because of SOI\textsuperscript{12}. If the symmetry of light scattering during SOI is affected by nearby interfaces, this could also lead to transversal forces acting on the object. To understand this quantitatively, let us consider a spherical, dipole-like particle with a radius equal to approximately one-tenth of the wavelength. The particle is located at the interface of two media with different refractive indices and is illuminated by a circularly polarized wave as shown in Fig. 1a. The distribution of intensity and the power flow (time-averaged Poynting vector) in a $y$–$z$ plane, perpendicular to the plane of incidence, is shown in Fig. 1b. As can be seen, the spiralling of energy flow breaks the mirror symmetry of the light scattering. Because of the optical spin–Hall effect in spherical geometry\textsuperscript{9}, light to the left of the $y = 0$ plane propagates mostly in the upper medium, and the opposite happens at the right side of the particle. The presence of the interface further breaks the central symmetry of the light distribution. As the magnitude of the wave’s momentum is determined by the properties of the medium through which it propagates\textsuperscript{13}, this scattering asymmetry unbalances the transversal linear momentum. Consequently, a side force perpendicular to the original wave propagation should act on the particle with a magnitude determined by its scattering phase function\textsuperscript{14}. In this Letter, we further elaborate on this idea analytically, and confirm it by exact numerical calculations and by experimental results.

If a dipolar particle is located entirely inside one of the media, the problem can be solved analytically. In the first example, we consider a point dipole located a distance $z_0$ away from the interface between two semi-infinite transparent media 1 and 2. Dipole $d$ is placed in the lower medium 2, and a circularly polarized plane wave of amplitude $E_0$ is incident from the upper medium 1. We are interested in the force $F_z$ acting on the dipole in a direction perpendicular to the plane of incidence $x$–$z$ (Fig. 1a). This force can be found by integrating the scattering phase function over a closed surface surrounding the particle. An asymmetric field distribution would ensure that these integrals are non-zero. In the case of a dipole, however, one can follow a much simpler approach derived directly from the Lorentz force formulation and find the force to be\textsuperscript{15} $F_z = (1/2)\text{Im}(\partial \phi / \partial z)\text{Im}(\partial / \partial y) T_y$, where $E$ is the local field at the location of the dipole. $\phi$ is the electric field of the incident wave with $\phi = E_0 \cos k_0 z_0$. $\partial \phi / \partial y$ represents the superposition of the transmitted field $E_T$ and the dipolar radiation $E_d$ scattered from the interface back to the dipole. Obviously, $F_z$ is zero for a normal incidence $\theta_0 = 0^\circ$ because of the azimuthal symmetry of the field. The force tends also to zero for grazing angles of incidence $\theta_0 \rightarrow 90^\circ$ when no light reaches the dipole inside medium 2. Because of spin–orbit transformations at intermediate angles of incidence, $E_d$ lacks symmetry with respect to the $y = 0$ plane and, consequently, its $y$ derivative is non-zero. In these conditions, the transversal force becomes (Supplementary Section 1)

$$F_z = \frac{I_0^2}{\epsilon_0} \text{Im}(d d^* y) \text{Im}(\partial / \partial y) T_y$$

$$\partial \phi / \partial y = \frac{1}{8\pi\epsilon_0} \int_0^\infty r_p(k_0) k_0^2 \exp(2ik_0 z_0) dk_p$$

$F_z$ is approximately one-tenth of the wavelength squared times the imaginary part of the object wave function $d$. This new force completes the set of non-conservative forces (radiation pressure and tractor beams) that can be generated with unstructured light beams.

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that, even though the electrostatic approximation can be used to calculate dipole moments for \( k z_o \ll 1 \), it is unsuitable for evaluating our transversal non-conservative force. The electrostatic approximation does not include retardation effects (field phases) and thus can account only for conservative forces. This demonstrates that the electrostatic approximation imposes even more severe limitations than previously believed.

For a dipole far from the interface \( (k z_o \gg 1) \), the integral in equation (1) can be evaluated asymptotically (see Supplementary Section 3 for details) to obtain

\[
F_z(z_0 \to \infty) = -\frac{k_z^4}{4\pi \varepsilon_0 \varepsilon_2} \Im(d_\perp d'_\perp) \sin(2k_z z_0) \frac{(n_{1z} - 1)}{(2k_z z_0)^2} (n_{1z} + 1)
\]

(2)

The presence of a reflection coefficient \( (n_{1z} - 1)/(n_{1z} + 1) \) factor in equation (2) suggests another interpretation of the transversal force for particles located far from the interface. The vortex spherical wave created by the dipole is reflected from the interface and causes a back-influence on the dipole itself. The force can be understood as originating from the phase of this vortex. The same phase, for example, makes particles orbit in vortex beams\(^5\). In our case the particle drags the vortex with itself, creating a self-propelling linear motion. This interpretation becomes somewhat similar to the one suggested for chiral particles at an interface\(^7\). However, our demonstration indicates that this phenomenon is more general and does not necessarily rely on exotic material properties.

The oscillating behaviour of \( F_z \) with distance \( z_0 \) resembles the behaviour occurring in optical binding when optically bound particles are illuminated with circularly polarized light\(^12\). However, the distance-dependent force in that case decays faster\(^12\) than the force in equation (2).

In a similar way one can estimate the transversal force for a particle located in the same medium as an incident wave (Supplementary Section 2). This may be more favourable in terms of the magnitude of the lateral force, which can be comparable to the radiation pressure. However, one should be careful in interpreting the nature of this transversal force: the force in equation (1) should be distinguished from the action of the transversal spin momentum that can appear during the interference of incident and reflected waves\(^14\). Because the spin momentum does not affect dipolar particles, its detection requires the presence of multipoles. This type of transversal force was detected recently in an Aharonov–Bohm optical setting\(^11\). As opposed to the force due to spin momentum, the transversal force described here would exist even if the reflected wave were completely eliminated by, for example, antireflection coatings at the interface.

Another interesting observation derived from equation (2) is that the transversal force is long-range and thus can affect dipoles located far from the surface. This suggests that such a lateral force should also appear for objects with dimensions larger than the wavelength.

To assess the magnitude of this transversal force for larger particles, we performed systematic numerical simulations (Comsol Multiphysics) for a geometry similar to our experiments. We evaluated the force acting on a 4.5 μm polystyrene (PS) particle located at an air–water interface and illuminated it with circularly polarized light. Because of the large scales of this simulation (~(10μm)), special methods were developed to perform scattering calculations (see Methods for details). The force magnitude shown in Fig. 2a was calculated using a modified Maxwell stress tensor, which was integrated over a closed surface around the particle (see Methods) to directly take into account the asymmetry of the field in the transversal direction illustrated in Fig. 2b. For irradiances of 0.15 mW μm\(^{-2}\), the force reaches values of tens of femtonewtons, which, in principle, can be detected experimentally.
Lateral force on Mie-size particles. θ = 52°, corresponding to the zero scattering angle = 52°, which leads to synchronous changes in the sign of the lateral force, as predicted by equation (1).

To verify the existence of this new type of force, we performed experiments with surface-bound microparticles. Monodispersed PS microspheres with diameter D = 4.5 μm were deposited onto the water surface (see Methods). The beam of a continuous-wave (c.w.) laser (wavelength, 532 nm; optical power, 2 W) was focused into a linear trap to prevent the longitudinal movement of the particles caused by radiation pressure. In this geometry and for the highly scattering PS particles, we did not observe the interfacial tractor beam effect13. A rotating quarter-wave plate allowed switching between left and right circular polarizations of the incident field (see Methods for a detailed description of the experimental set-up). As can be seen in Fig. 3, switching the polarizations leads to synchronous changes in the sign of the lateral force, as predicted by equation (1).

In the experiment, the weak effect of optical forces was effectively amplified by the long-range hydrodynamic interaction between particles23,24. Assuming pairwise additivity, the velocity of an arbitrary particle i is

$$v_i = v_{\text{flow}} + \sum \mu_j(r_i,r_j)F_{\perp}(r_j)$$

where $v_{\text{flow}}$ is the macroscopic flow on the water surface, $\mu_j$ is the tensor defining the hydrodynamic interaction25 and the summation is performed over all particles affected by the optical force24,26. The monodisperse PS particles experience the transversal force $F_\perp$, pointing in the same direction. The long-range $1/R$ hydrodynamic interactions27 make their velocities diverge logarithmically with an increase in the number of particles. Figure 3b shows the velocities of several particles in a linear trap as a function of time (Supplementary Movie). As the number of trapped particles increases over time (Fig. 3c), the particles’ velocity also grows. This hydrodynamic interaction results in a threefold increase in the observed velocities.

As is evident in Fig. 3b, the particles’ velocity follows the changes in polarization handedness. The regression analysis performed on the base of linear model (4) yields the velocity of an isolated particle $(F_\perp)/(3\eta D/2) = 0.62 \pm 0.06 \mu m s^{-1}$ with a confidence level of 95% (the drag coefficient here is two times smaller than that of the Stokes formula as the particles are half in air28). Knowing the dynamic viscosity of water, $\eta$, and the size of the particles, $D$, we evaluate the average lateral force to be $\langle F_\perp \rangle = 11 \pm 1 fN$ for 0.15 mW μm⁻² average power density. This force magnitude is in good agreement with the predictions of numerical calculations (Fig. 2a), $\langle F_\perp \rangle(\theta = 52°) \approx 4.7 fN$.

As a side note, we do not expect SOI to produce additional spin motion of the particles. Frictional liquid forces are on the order of micronewtons29, which is at least six orders of magnitude larger than typical optical forces.

The proposed technique completes the set of methods for arbitrary manipulation of colloidal particles on the surface of liquids without having to rely on beam structuring. Forward action is provided by the radiation pressure, and backward movement can be achieved through momentum enhancement effects31. Here, we have demonstrated that a side motion can also be induced due to the transformation of the spin angular momentum of incident photons into orbital angular momentum of scattered ones. To detect this new transversal force, we systematically applied,
for the first time, a method of hydrodynamic amplification of optomechanical effects.

Methods

Methods and any associated references are available in the online version of the paper.

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Author contributions

S.S. and A.D. conceived the idea and designed the experiments. S.S. performed theoretical analysis. V.K. and R.R.N. performed the experiments. S.S. and V.K. contributed materials/analysis tools. S.S., V.K., R.R.N. and A.D. analysed the data. S.S. and A.D. wrote the paper.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.D.

Competing financial interests

The authors declare no competing financial interests.
Methods

Numerical simulations. Simulations of light scattering and calculation of optical forces acting on a 4.5 μm particle at the interface of two media were performed with commercial software Comsol Multiphysics v.4.3a. The model dimensions (−10b)³ make excessive demands on computer memory and could not be handled directly on the available PC with 128 Gb of RAM. Also, as mentioned in the text, the calculations cannot be simplified because there are no symmetry planes for the field distribution. However, the computation efficiency can be significantly improved using a special technique where the field is decomposed into s- and p-polarized waves and two separate solutions are calculated. The final field is then built using these solutions and exploiting the symmetry properties of the two field components: \( E'(x,y,z) = -E'(x,-y,z) \), \( B'(x,y,z) = -B'(x,-y,z) \), \( E''(x,y,z) = -E''(x,-y,z) \), \( B''(x,y,z) = -B''(x,-y,z) \), \( E'(x,y,z) = E'(x,-y,z) \), \( B'(x,y,z) = B'(x,-y,z) \), \( E''(x,y,z) = E''(x,-y,z) \), \( B''(x,y,z) = B''(x,-y,z) \). Using these symmetry properties with respect to the \( y = 0 \) plane, only half the simulation volume has to be modelled in practice. Finally, the transversal force on a particle was found by integrating Maxwell stress tensor \( \mathbf{T} \) (in Minkowski formulation) over the surface of the particle \( \Sigma \):

\[
F_x = \int_\Sigma (\mathbf{T}_x)_{ij} \, da
\]

\[
T_{ij} = \text{Re} \left\{ \mathbf{E} \mathbf{E}^* + \mathbf{H} \mathbf{B}^* - \frac{1}{2} (\mathbf{E} \mathbf{D}^* + \mathbf{H} \mathbf{B}^*) \delta_{ij} \right\}, \quad i, j = x, y, z
\]

where \( \mathbf{n} \) is the unit external normal to the surface \( \Sigma \), \( \delta_{ij} \) is Kronecker delta and the total electric and magnetic fields are \( \mathbf{E} = \mathbf{E}' + \mathbf{E}'' \) and \( \mathbf{B} = \mathbf{B}' + \mathbf{B}'' \), respectively. Taking into account the symmetry properties of \( \mathbf{E} \), \( \mathbf{B} \) and \( \mathbf{n} \), the lateral force can be found by integrating over just the \( y > 0 \) part of the surface \( \Sigma \):

\[
F_x = \int_{\Sigma_{+}} (T_{xx} n_x + T_{yy} n_y + T_{zz} n_z) \, da
\]

where the modified Maxwell stress tensor accounts directly for the field differences on the two sides of the plane \( y = 0 \) and is defined as

\[
T_{ij} = \text{Re} \left\{ \mathbf{E} \mathbf{D}^* + \mathbf{B} \mathbf{E}^* + \mathbf{H} \mathbf{H}^* + \mathbf{H} \mathbf{H}^* - \frac{1}{2} (\mathbf{E} \mathbf{D}^* + \mathbf{B} \mathbf{E}^* + \mathbf{H} \mathbf{H}^* + \mathbf{H} \mathbf{H}^*) \delta_{ij} \right\}
\]

One can clearly see that a transversal force \( F_x \) appears only if both s- and p-polarization components are present in the incident field.

Sample preparation. A suspension of 4.5-μm-diameter PS particles (Polysciences) was dissolved in methanol (99.9 mol% pure, FisherScientific). A single small drop of the solution (~6 μl) was gently released from a 10 μl syringe (World Precision Instruments) onto the surface of the water. On release the solution methanol evaporated rapidly, leading to the spontaneous formation of a well-dispersed monolayer of micrometre-sized particles on the interface.

Experimental set-up. Monochromatic light from a cw laser (wavelength, 532 nm; optical power, 2 W; Coherent Genesis CX Series model) was incident onto a water surface at an angle of 52°. The beam was focused into a linear trap using a combination of a positive lens and a cylindrical lens with focal lengths of 50 and 120 mm, respectively. The resultant linear trap (dimensions of 30 × 550 μm²) prevented longitudinal movement of the particles as a result of radiation pressure. A rotating quarter-wave plate allowed the linear and circular polarizations of the incident field to be set. Note that the long focal distance of the lens did not greatly disturb the circularity of the polarization states. To visualize the particles, the surface was illuminated with low-power incoherent red light from a light-emitting diode. The motion of the particles at the air–water interface was imaged through a ×10 microscope objective (NA 0.25, Olympus) and recorded by a charge-coupled device camera (Andor sCMOS) with a frame rate of 1 fps. Two 532 nm notch filters (OD4, Edmund) were used to remove the scattered laser light. During the recording time, the polarization was switched between different states of circular polarization. The rotation of the quarter-wave plate was driven by a stepping motor (Oriental Motor, Model PK243M-03B) controlled by NI 7340 motion controllers (National Instruments). The prepared sample was contained in a cavity inside a large aluminium plate. The container was enclosed by a square glass column to prevent excessive particle movement due to external air currents. A slight directional flow of the particles was generated against the direction of radiation pressure to allow stable trapping while providing a population of particles in the linear trap.

Data analysis. The image processing algorithm was implemented in a homemade MATLAB code to precisely locate the particle centres in each frame. The linking of particles from different frames into trajectories was performed using a third-party MATLAB code. The location of the optical trap in recorded images was determined by analysing the intensity distribution of the scattered light and was confirmed by observation of the longitudinal motion of the particles. Instantaneous velocities for each particle were calculated on the basis of tracking data. The linear model of equation (3) was used to single out the optically induced component of velocity. The hydrodynamic interaction tensor in equation (3) was taken in the Rotne–Prager form. As all the particles are the same size and are virtually indistinguishable, multivariate regression analysis was used instead of a general linear model to determine the regression coefficients.

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