Controlled transfer of transverse orbital angular momentum to optically trapped birefringent microparticles

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The interaction between structured light beams possessing optical angular momentum and small particles promises new opportunities for optical manipulation, such as the generation of light-induced torque and rotation of objects. However, so far, studies have largely centred on nanoscale particles. Here we report the observation and measurement of the transfer of transverse angular momentum to birefringent spherical vaterite particles several wavelengths in size. We outline the physics behind the beam used to control the particles, perform quantitative measurements of the transverse spin angular momentum transfer and demonstrate the generation of fluid flow around multiple rotation axes. The findings show that light can impart controllable rotational degrees of freedom to microparticles. In the future, the approach may prove useful for investigating the dynamics of complex fluids in three dimensions, studying the shear force on cell monolayers or cooling an optically trapped particle to the quantum ground state.

Conservation of momentum is a fundamental principle in physics. As a consequence of the energy carried by a photon, there is a corresponding momentum to its movement in space. It can transfer momentum to matter through absorption and scattering1–5. An object scattering light experiences an impulse from the field through the conservation of momentum. Optical tweezers trap particles in three dimensions through linear momentum transfer from strongly focused light2,6–8. Optical forces have found major applications ranging from studying a large number of complex biological systems in terms of mechanical components9 to contributing to experiments on exotic states of matter such as Bose–Einstein condensates10. Later experiments demonstrated optical angular momentum transfer to trapped particles1, which led to developments in controlling and characterizing microscopic systems11–18.

Angular momentum transfer is usually restricted to the axis defined by the beam propagation direction19–21. More recently, the transfer of angular momentum around an axis orthogonal to the beam propagation direction was demonstrated on absorbing particles using retro-reflected laser beams of non-uniform circular polarizations22,23. These investigations have led us to believe that trapping and control (in three dimensions) of larger birefringent particles should also be possible. Substantially higher angular momentum can be transferred to larger particles, and thus, they can perform more work on the surrounding environment as microscopic motors. The concept behind this is shown in Fig. 1a. A bespoke beam depicted as propagating from the bottom of the page interacts with the larger birefringent particle to induce spinning and rotation about the axis denoted with the curved arrow.

We report on the observation and measurement of the transfer of transverse angular momentum to birefringent vaterite particles (positive uni-axial crystals) several wavelengths in size and trapped with optical tweezers using a transmission optical system. Vaterite is one of the crystal structures of calcium carbonate and can be grown in the spherical form. It has been demonstrated that these spherical forms of vaterite are highly birefringent24, can be produced with sizes varying from hundreds of nanometres up to several micrometres25,26, and have been used as rotating particles in optical tweezers in both liquids and vacuum27,28. Vaterite’s size and shape can be precisely controlled29,30 and it is stable in water as well as certain ionic buffers; it can even be coated for added protection when used in more hostile environments31. The trapping of such a particle requires highly focused structured light, which can lead to the emergence of effects such as spin–orbit coupling27–31 and phenomena such as Brownian vortices32, which affect the measurement of particle dynamics. We estimate the transfer of spin angular momentum to the particles using Stokes measurements. However, we also show that these measurements cannot yield the actual angular momentum transferred in this system due to coupling between the spin and orbital momentum of light. We discuss the difficulties and prospects for the quantitative measurement of angular momentum that this system poses.

The transfer of transverse angular momentum to micrometre-sized particles can be advantageous for studies of diverse microsystems. It enables an increased level of control over vaterite spheres and potentially other birefringent particles, too. The particles used in our study are much larger than the nanoparticle systems for which transverse angular momentum was previously investigated30,23,33. The larger birefringent particle, by virtue of its large surface area, interacts more strongly with the trapping beam as well as the fluid surrounding it. Thus, it can be used for the investigation of the dynamics of complex fluids in three dimensions as well as for shear on cell monolayers.

Results

Optical momentum transfer. To create an optical trap that can also induce the transverse spinning of micrometre-sized birefringent particles, we need to consider the way light transfers momentum to matter at that scale. It is the transport of momentum by light that

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Light can also carry angular momentum and can be used to control the orientation of particles and to rotate them. An important point about the angular momentum of light is that it can either be spin or orbital angular momentum. We can write the total angular momentum density as the sum of spin and orbital angular momentum densities:

$$j = l + s,$$

where $l$ is the orbital angular momentum density and $s$ is the spin angular momentum density. The key theoretical difference between the spin and orbital angular momentum is that the spin angular momentum density is independent of the choice of coordinate system. Thus, a beam of light can carry spin angular momentum about the beam axis on the beam axis itself, since no moment arm is required. The spin and orbital angular momentum fluxes of a beam of light can be found from the densities by integrating across the cross-section of the beam. If the beam is paraxial and monochromatic (or at least quasi-monochromatic), the circular polarization of the beam simply determines the spin angular momentum:

$$\hbar \text{ per photon for left-circular polarization and } -\hbar \text{ per photon for right-circular polarization.}$$

If the beam is non-paraxial, the field structure becomes specific to particular regions of space. Although a non-paraxial beam cannot be unambiguously described by a single polarization like a paraxial beam, the far field of the beam—which is locally of the form of a plane wave—can be locally described by a polarization (this also means that the spin angular momentum can be measured by determining the Stokes parameters of light in the far field, whereas typical schemes for the measurement of orbital angular momentum require information about the phase of light).

If the light everywhere is locally right-circularly polarized, then the spin angular momentum density is $-\hbar$ per photon in the direction of the local wavevector. Only the component in the direction of the beam axis contributes to the total spin angular momentum of the beam. Although a collimated beam has a total spin of zero, it is possible to engineer a beam such that the axial components of the spin flux cancel across the beam; further, the beam has a total spin angular momentum about the beam axis of zero. It is also possible to produce a fairly abnormal condition for a beam of light: a non-zero spin flux about an axis normal to the beam axis (Fig. 1d). Such transverse spin is a potential pathway for a beam of light: a non-zero spin flux about an axis normal to the beam axis.

For a practical realization of transverse spin angular momentum transfer, we must consider something more concrete than the unspecified ‘beam’ above: a Gaussian beam is the obvious starting point, although vectorial polarization shaping of light is also possible. The superposition of two non-co-linear Gaussian beams of opposite circular polarization of equal power and both at the

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**Fig. 1 | Illustration of a particle undergoing transverse rotation in a structured beam.** a. Pictorial representation of a vaterite in a particle-centred frame with the axis of rotation (dark-grey bar) caused by the superposition of two orthogonal circularly polarized beams (red and blue arrows). Momentum from two crossed focused beams. b. Since only the axial component of the local linear momentum density contributes to the total momentum flux of the beam, a converging beam has a lower momentum flux than a collimated beam of the same power. c. In the same way that the linear momentum flux is lower in a converging beam, the spin flux must also be lower in a converging beam. However, the total angular momentum flux remains the same; therefore, an equal amount of orbital angular momentum must compensate for the reduced spin. d. If instead of combining two converging rays with equal spin, we combine two waves with opposite spin, then the combined beam has zero axial spin flux and has—unusually—non-zero transverse spin.

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allows light to be used to trap and manipulate particles, such as with optical tweezers. The momentum flux $P$ is related to the energy flux $\mathcal{P}$ by the speed of propagation in the medium:

$$P = \mathcal{P}k/(nc),$$

where $k$ is the wavevector, $n$ is the refractive index of the medium and $c$ is the speed of light in free space. The forces responsible for radial trapping result from the deflection of the trapping beam by the trapped particle. If the particle in the trap deflects the beam to the right, then conservation of momentum demands that a reaction force act on the particle to the left.

The origin of the axial force opposite to the direction of propagation that is necessary for three-dimensional trapping is less obvious. The key point is that a collimated beam (or a plane wave or ray) of a given power has a higher momentum flux than a converging or diverging beam. This is because the local wavevector is always parallel to the direction of propagation in a collimated beam, whereas it is at an angle to the direction of propagation for a converging or diverging beam (Fig. 1b). In this case, only the vector component in the propagation direction contributes to the total momentum flux of the beam. If the trapped particle changes the convergence or divergence of the beam, it changes the component of momentum flux in the axial direction. If it makes the beam more collimated (that is, less convergent or divergent), it increases the momentum of the beam in the direction of propagation, resulting in a restoring force acting on the particle in the opposite direction, as required for three-dimensional trapping against axial forces resulting from reflection and absorption.

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**Notes**

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If the light everywhere is locally right-circularly polarized, then the spin angular momentum density is $-\hbar$ per photon in the direction of the local wavevector. Only the component in the direction of the beam axis contributes to the total spin angular momentum of the beam. Although a collimated beam has a total spin of $\hbar$ per photon, a converging beam must have less spin (Fig. 1c). This is the same geometric reasoning that shows that a converging or diverging beam has a lower linear momentum flux than a collimated beam (Fig. 1b).

However, this reduction only applies to the spin angular momentum flux. If the converging beam is produced (as usual) by focussing a collimated beam using a rotationally symmetric lens, the angular momentum per photon remains unchanged. Since the spin angular momentum is reduced, an equal amount of orbital angular momentum is introduced (Fig. 1c). This spin–orbit conversion of angular momentum can have measurable consequences, as shown below.

A key difference between the behaviour of spin density and linear momentum density is that the spin density can be anti-parallel to the local wavevector, that is, $-\hbar$ per photon. Thus, the axial component of the spin flux of a complex beam need not always be in the same direction, and it is possible to engineer a beam such that the axial components of the spin flux cancel across the beam; further, the beam has a total spin angular momentum about the beam axis of zero. It is also possible to produce a fairly abnormal condition for a beam of light: a non-zero spin flux about an axis normal to the beam axis (Fig. 1d). Such transverse spin is a potential pathway to the rotation of particles in optical traps about axes normal to the beam axis.

For a practical realization of transverse spin angular momentum transfer, we must consider something more concrete than the unspecified ‘beam’ above: a Gaussian beam is the obvious starting point, although vectorial polarization shaping of light is also possible. The superposition of two non-co-linear Gaussian beams of opposite circular polarization of equal power and both at the
same angle to the common beam axis results in zero total axial spin and maximum transverse spin. If both beams are focused using a high-numerical-aperture lens, they can form an optical trap. Noting that in a microscope system, changing the angle of a beam is equivalent to changing its position at the back aperture of the main focusing optics, the two oppositely polarized beams can be initially parallel and directed onto opposite sides of the back aperture of the microscope objective.

A simple way to transfer the transverse spin angular momentum to a trapped particle is to use absorption. However, absorption forces inherently limit the maximum size of the particle that can be trapped in optical tweezers, since absorption also transfers linear momentum, acting to push the particle out of the trap. Thus, the direct transfer of transverse spin angular momentum has previously been restricted to nanoparticles.

Birefringent vaterite microspheres are commonly used for the transfer of axial spin angular momentum in optical traps. In this case, they approximately act as waveplates, and change the degree of circular polarization of the trapping beam. Such vaterites can be trapped in a transverse spin beam (as described above) and are a good prospect for the transfer of transverse spin angular momentum. However, whether the desired transfer of angular momentum can be achieved depends on the orientation of particles in the optical trap. It will be very useful to investigate this using computational modelling before proceeding to experiments.

**Computer simulation of controlled momentum transfer.** We modelled the torque transfer to vaterite particles using the optical tweezers toolbox (Methods). We calculated the torque transferred to a 3.5-μm-diameter vaterite at the stable axial trapping location with its optical axis aligned with the beam and orthogonal to the axis of transverse angular momentum. At large separations, the torque per photon continues to increase, whereas the reduction in radiance begins to dominate the contribution to absolute torque. A mask with varying thicknesses was introduced between the two beams to cut the overlapped regions of opposing spin angular momentum. We checked whether the torque per photon and difference between the two alignments could be improved with this mask. As shown in Fig. 2b, a slight improvement in both torque per photon and

**Fig. 2 | Transverse angular momentum transfer to a vaterite particle and the field that was simulated to achieve this.** a,b, Transverse angular momentum as a function of beam separation (a) and mask thickness (b) for a fixed beam separation. c,d, Transverse spin angular momentum density orthogonal in the x-y (c) and z-x (d) planes.
difference in alignments can be seen when the mask blocks more than 75% of the beams. The transverse spin angular momentum density of the composed beam is shown in the focal plane (Fig. 2c) and through the beam (Fig. 2d). An analysis of the transverse spin angular momentum density in plane-wave interference has been shown previously.36

There is a practical limit of improvement for the transverse momentum transferred as there is a trade-off between the enhancement in per photon transfer and observed rotation as the transmitted light reduces. The mask-fill level used in our experiment is estimated to be about 60% of the back aperture of the objective lens. Torque transfer per photon in our model is about 0.01ℏ per photon for both alignments, which is similar in magnitude to the value of 0.02ℏ per photon found for axial torque transferred when the vaterite sphere was trapped in a single circularly polarized beam.35 Based on the results shown in Fig. 2, the angular velocity of the particle in a viscous fluid is the greatest when the optical axis is aligned with the beam and at most half that when it is orthogonal to it. For an aid to visualization, the transverse spin angular momentum density of the beam that results from the structuring is shown in Fig. 2c. Evidently, the transverse spin component is present in a large portion of the beam near the focus.

The difficulty in measuring the momentum transfer in a system is the result of the definitions of electromagnetic torque. The electromagnetic torque of a radiation field is

\[
\langle j \rangle = \int \mathbf{r} \times \mathbf{E}_{\text{out}}^* \times \mathbf{H}_{\text{out}} d\Omega,
\]

where \( E \) and \( H \) are the electric and de-magnetizing fields, respectively, \( \mathbf{r} \) is the position and \( \Omega \) are the coordinates of a solid surface surrounding the system. For a time-harmonic field,

\[
- \frac{\partial}{\partial t} \mathbf{H} = i \omega \mathbf{E} = \frac{1}{\mu_0} \nabla \times \mathbf{E}
\]

where \( \omega \) is the angular frequency of light, \( \mathbf{E} \) is the electric field, and \( \mu_0 \) is the permeability of free space. We, thus have

\[
\langle j \rangle = \frac{i}{\sigma \mu_0} \int \mathbf{r} \times \mathbf{E}_{\text{out}}^* \times \nabla \times \mathbf{E}_{\text{out}} d\Omega.
\]

To determine \( \langle j \rangle \), we must collect all the light on the unit sphere surrounding the system and determine the propagation of radiation at each point on it. In certain circumstances where the angular scattering is constrained to the aperture of the optical system, we still have confusion as to the origin and position to convert the angle of outgoing light. This expression (equation (4)) can be further broken down into the spin and orbital torques.37,38,39 The time-averaged spin angular momentum flux can be measured via spatially resolved Stokes parameters over a spherical surface.36 The time-averaged total spin per photon is then:

\[
\langle s \rangle = \frac{\int (|E_+|^2 - |E_-|^2) d\Omega}{\int |E_+|^2 + |E_-|^2 d\Omega},
\]

where \( E_+ \) are the left-circularly polarized (+) and right-circularly polarized (−) components of the light field, and \( \hat{r} \) represents an element of the directional unit vector (Supplementary Section 4). The sampling of the back focal plane is equivalent to sampling the spherical surface where the objective lens approximately obeys the Abbe sine condition. In the experiment, the transverse measurement in the \( x \) axis is implemented using equation (6).

Experimental results

To observe the transfer of transverse angular momentum, we used a modified optical trapping apparatus (Methods). The key addition to the system is beam displacers that offset a single, diagonally polarized beam into horizontal and vertical components. These components are then changed to orthogonal left- and right-circular polarizations with a \( \lambda/4 \) waveplate. The setting of the waveplate after the collection objective (O2) is set to maximize the contrast between orthogonal polarizations. The noise floor was approximately 1% of the dynamic range of the camera. Typical measurement contrast values were about 15% to 45% of the dynamic range per pixel, which means that the presence and rotation of the vaterite particle caused a measurable change to the polarization state of the incident light. The two active regions on the camera used for our spin angular momentum transfer measurements are approximately \( 300 \times 300 \) pixels each, which average out the per-pixel noise. Our calculation of the estimated spin angular momentum transfer per photon is the difference in light level normalized by the sum for every pixel (equation (6)).

Figure 3a,b shows the results from our simulations and measurements of transverse spin angular momentum transferred to the vaterite particle. First, when comparing the simulated and theoretical transverse spin angular momentum (Fig. 3, solid lines), we see that simulation and theory produce a similar level of spin angular momentum transfer. The average angular momentum transfer in the simulation was about 0.12ℏ per photon. In the experiment, it was about 0.09ℏ per photon. Given the necessary assumptions regarding birefringence, structure, beam aberration and some depolarization from the apparatus, these average values are in good agreement. The oscillatory behaviour of spin angular momentum transfer predicted by the model was observed in the experiment, too. This arises due to the difference in interaction with the beam.

Fig. 3 | Comparison of spin angular momentum determined from simulation and experiment. a, Spin angular momentum and total angular momentum transferred to the simulated particle. b, Estimated spin angular momentum from the particle trapped in the experiment.
The spinning particle could generate a hydrodynamic flow field strong enough to drive another particle around it. Our modelling of the vaterite is consistent with our observations and provides good support for the changes in rotation rate as a function of orientation. We noted a small orbit of the particle around the beam as its orientation changed. These observations are consistent with the presence of spin–orbit coupling. Due to the generation of transverse angular momentum through interference, the details of the transfer of angular momentum to particles in the size regime of a few wavelengths is much more complex than for nanoparticles and needs further investigation.

We believe there are several potential future studies and experiments that can be performed using the transfer of transverse angular momentum of light. This system provides new challenges in both transfer and measurement of electromagnetic torque transfer—the development of direct and general measurements of the total angular momentum would greatly improve our understanding of the interactions observed here and contribute to the characterization of other phenomena of optical torque transfer.

Transverse angular momentum transfer to probe particles would be highly advantageous in microscopic systems such as living cells as they are often very flat, and azimuthal spin would not create much flow (shear stress) across the surface. Shear stress induced by transverse rotation can be used to study mechanotransduction within the cell. The observation of light–matter interaction within optical traps gives us the ability to probe the mechanical effects of optical torque transfer around all the principal axes.

Combined with existing methods for controlling, applying and transferring spin and orbital angular momentum about the beam axis, transverse angular momentum offers the opportunity for the controlled application of full three-dimensional torques, with three independent orthogonal axes. Since the geometry of combining left- and right-circularly polarized beams at an angle to give transverse angular momentum also applies to beams carrying opposite orbital angular momentum (such as counter-helical optical vortices), a desired combination of both transverse spin and transverse orbital angular momentum can be produced.

This opens opportunities in optical micromanipulation by adding controllable rotational degrees of freedom to microparticles. For example, the three-axis rotation of birefringent microparticles or engineered particles driven by orbital angular momentum can be used in novel optically driven microfluidic devices. Such three-dimensional control might also allow the cooling of the rotational motion of an optically trapped particle to the quantum ground state.

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Methods
Modelling and simulation. The calculation of optical properties and angular momentum transfer were carried out with the optical tweezers toolbox\(^4\). The current version of the toolbox (available at https://github.com/ilent2/ott) has the capacity to generate T-matrix models of vaterite particles based on a ‘sheaf-of-wheat’ model of dipole anisotropy\(^4\). However, the T-matrix model of the vaterite used in these calculations was determined using the scattered radiation pattern from plane-wave projections in a discrete dipole approximation code (ADDA)\(^4\). This was motivated by memory limitations with how MATLAB 2018a solves the coupled dipole problem. ADDA uses far less memory and thus was used instead. The plane-polarized wave projections from ADDA were incorporated into the optical tweezers toolbox by means of a basis transformation from vector plane waves to vector spherical wavefunctions. For a 3.5 μm particle, a vector spherical wavefunction expansion to degree 25 is required in the optical tweezers toolbox; 26 (N + 1) unique polar and 52 (2N + 2) unique azimuthal projections over a uniformly spaced angular grid on an Ewald sphere (far field) were required to characterize the scattering by the T-matrix model. This was generated by the superposition of the plane-polarized components of the amplitude matrix\(^4\) as the left- and right-chiral components into a matrix, \(M_{\text{out}}\)\(^5\). The set of transformations performed to generate the T matrix could then be represented as a matrix equation with

\[
T_{\text{out}} = C_{\text{PV} \to \text{VSWF}} M_{\text{out}}^{\text{ADDA}} C_{\text{VSWF} \to \text{PN}},
\]

where \(C\) denotes the basis transformations that map either plane waves to vector spherical wavefunctions or vice versa.

To obtain the absolute torque and torque per photon transferred to a vaterite particle (Fig. 3), we computed the stable trapping positions for Gaussian beams of opposite circular polarizations as a function of separation at the back aperture of a high-numerical-aperture lens. The power of the absolute torque transfer to the vaterite particle is in unit beam power for the case in which the two Gaussian beams have co-linear transmission.

Using the parameters from our experimental apparatus, we simulated the behaviour of the vaterite particles using a stochastic dynamics simulation in the creeping flow regime solved with Euler–Maruyama step integration\(^4\) over the three translational and rotational degrees of freedom. Optical forces and torques were computed using the optical tweezers toolbox with a laser power chosen to obtain comparable rotation rates to those observed in the experiment. These results are shown in Fig. 3. Supplementary Section 1 provides the visualization of a simulated vaterite spinning in the optical trap.

Apparatus. A linearly polarized 1,064 nm laser source was used in the experiments (YLR-10-1064-LP, IPG Photonics). The polarized beam was incident on a beam displacer such that its output created two equal-intensity orthogonally polarized beams. The beam block appearing in the conjugate imaging plane was measured to block approximately 3.8 mm of the back focal plane of the focusing objective lens (numerical aperture, 1.2; UPLSAPO60XW, Olympus). Imaging of the beam block at the objective was achieved with an inverting telescope. Microscopy samples consisted of two coverslips (471112250, Trajan) sandwiched with Parafilm (P 7543, Sigma–Aldrich), with holes in the middle to retain the samples. Light exiting the sample region was collected with another objective lens stretched Parafilm (P 7543, Sigma–Aldrich), with holes in the middle to retain the samples consisted of two coverslips (471112250, Trajan) sandwiched with Parafilm (P 7543, Sigma–Aldrich), with holes in the middle to retain the samples. Microscopy samples consisted of two coverslips (471112250, Trajan) sandwiched with Parafilm (P 7543, Sigma–Aldrich), with holes in the middle to retain the samples. A selection of frames from one of our experiments is included in Supplementary Fig. 6. Video microscopy of the vaterite particles was captured by a complementary metal–oxide–semiconductor camera (EoSens CL, Mikrotron) and streamed to a solid-state disc for post-processing.

Vaterite samples were prepared using the method outlined elsewhere\(^5\). Most vaterite particles were found to be stuck to the coverslips. A combination of exhaustive search and jiggling of the stage resulted in several successfully freed and subsequently optically trapped particles.

Data availability
All practically distributable raw and processed data (such as images, signal traces and tracked positions) are available from the corresponding authors upon reasonable request.

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Author contributions
A.B.S. and H.R.-D. conceived the investigation. A.B.S. designed the experiment and simulations, as well as collected and analysed the data. A.B.S., H.R.-D. and T.A.N. composed and edited the manuscript. A.B.S. composed the supplementary information.

Competing interests
The authors declare no competing interests.

Additional information
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