Hydrodynamic spin-orbit coupling in asynchronous optically driven micro-rotors

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Vortical flows of rotating particles describe interactions ranging from molecular machines to atmospheric dynamics. Yet to date, direct observation of the hydrodynamic coupling between artificial micro-rotors has been restricted by the details of the chosen drive, either through synchronization (using external magnetic fields) or confinement (using optical tweezers). Here we present a new active system that illuminates the interplay of rotation and translation in free rotors. We develop a non-tweezing circularly polarized beam that simultaneously rotates hundreds of silica-coated birefringent colloids. The particles rotate asynchronously in the optical torque field while freely diffusing in the plane. We observe that neighboring particles orbit each other with an angular velocity that depends on their spins. We derive an analytical model in the Stokes limit for pairs of spheres that quantitatively explains the observed dynamics. We then find that the geometrical nature of the low Reynolds fluid flow results in a universal hydrodynamic spin-orbit coupling. Our findings are of significance for the understanding and development of far-from-equilibrium materials.

Hydrodynamically coupled rotors describe the dynamics of physically diverse systems—the kinetics of proteins in biological membranes, the interactions of topological defects in superfluid Helium, and the mating rituals in dancing algae. An isotropic fluid made of rotating particles with broken parity and time reversal symmetry is expected to possess peculiar material properties, including odd viscosity and topological acoustic edge modes. Moreover, simulations show that in two-dimensional systems, hydrodynamically coupled rotors self-assemble into both random and ordered hyper-uniform arrangements.

Recently, mass-produced magnetic micro-particles rotated by an external electromagnet have been extensively used as a model system for studying rotating ensembles. The rotating external field directly spins the particles, revealing new collective dynamics, including dislocation kinetics in rotating crystals, propagation of chiral surface waves in a rotating liquid, as well as self-healing and coarsening of colloidal fluids and crystals. However, magnetic rotors are not free. Just like compass needles, the orientations of the magnetic dipoles are enslaved to the globally imposed north, as they are synchronized with the orientation of the electromagnet. Though the position of the center of particle \( i \), \( \mathbf{R}_i \), may freely diffuse in the plane, the ensemble is not isotropic, as the orientational degree of freedom, \( \theta_i \), is externally imposed such that \( \langle \theta_i(t) \rangle = \theta_M(t) \).

An alternative to magnetic rotors is particles spun by a focused beam of circularly polarized light. Photonic angular momentum can be transferred to a micro-particle through its shape anisotropy, birefringence, or simply by absorption. Unlike magnetic rotors, photonic rotors do not directly follow the rapidly rotating electromagnetic field. Instead, the optical angular momentum flux creates a torque that maintains a steady rotation, asynchronous from the external drive. However, when using a focused beam, the position of an optical rotor, \( \mathbf{R} \), is limited by the strong tweezing force at the focal point, \( \mathbf{R}_L \), constraining its translational degrees of freedom \( \mathbf{R}(t) = \mathbf{R}_L \). For truly free rotors, both the orientations and the positions are free dynamic variables, and when in a liquid, \( \mathbf{R} \) and \( \theta \) are expected to couple hydrodynamically. The motion of synthetic...
micro-rotors studied so far is incompatible with the R–θ hydrodynamic coupling that was analytically explained\(^1\), and empirically observed\(^{1,2,4}\) in pairs of biological micro-rotors. Moreover, previous studies with ensembles of synthetic micro-rotors\(^{10,11}\) spin by an externally imposed field and can not show spontaneous symmetry breaking as seen in ensembles of biological rotors\(^3\). To date, configuration space is reduced in either magnetic or photonic rotors, and the nature of the chosen drive obscures mutual hydrodynamic coupling.

In this communication, we show that optically driven rotors in a non-tweezing beam freely diffuse while spinning asynchronously. By developing a novel experimental test bed that drives hundreds of free micro-rotors (Fig. 1a, b and Supplementary Movie 1), we measure their stochastic translational and rotational dynamics independently. We find that in this system, remote particles are rotating asynchronously, and at close proximity, rotation and translation couple and rotor pairs mutually advect into an orbital motion (Supplementary Movie 2). We observe that the translation and rotation of these free optical rotors reciprocate—their spin coupling obeys a geometrical relation following the Stokes flow of spheres near a wall. To create a system of asynchronous rotors, we design an optical setup capable of producing a uniform torque field with minimal tweezing. We also develop a synthetic route for stable silica-coated birefringent particles (Fig. 2) that rotate in a circularly polarized collimated beam. We characterize the translational and rotational motion of individual particles and pairs of particles and derive an analytical hydrodynamic model that quantitatively captures their dynamics.

**Results and discussion**

**Synthesis of stable birefringent micro-particles**

We couple photonic angular momentum to the particles by synthesizing a new type of birefringent colloid made of silica-coated vaterite. Vaterite has a hexagonal symmetry with a positive uni-axial optical response and birefringence of \(\Delta n = n_o - n_e = 0.1\), where \(n_o = 1.55\) and \(n_e = 1.65\) are the refractive indices along the ordinary and extraordinary axes. When illuminated with circularly polarized light, vaterite particles begin to rotate while experiencing negligible thermal absorption\(^6\), making high particle concentrations experimentally accessible without overheating (Fig. 1b). Using previous synthetic routes\(^{25,26}\), we found colloidal vaterite’s rotational dynamics to be inconsistent between tweezed and tweezing-free optical fields. In the absence of tweezing, particle rotation was intermittent and gradually diminished, suggesting that minor surface chemistry variations dominated the dynamics. We develop an alternative synthetic strategy to allow consistent rotation over prolonged durations.

Particles are synthesized via controlled precipitation of highly concentrated solutions of calcium chloride \(\text{CaCl}_2\) and sodium carbonate \(\text{Na}_2\text{CO}_3\) according to,

\[
\text{CaCl}_2\text{(aq)} + \text{Na}_2\text{CO}_3\text{(aq)} \rightarrow \text{CaCO}_3\text{(s)} + 2\text{NaCl}\text{(aq)},
\]

The vaterite phase, a product of Eq. (1), is a metastable polymorph of calcium carbonate. Deviations towards even weakly acidic conditions cause rapid dissolution and transformation of vaterite spheres to calcite cubes\(^27\). Therefore, the vaterite-to-calcite phase transition is a significant barrier encountered when synthesizing and re-suspending vaterite microspheres.

To preserve micro-particles in the vaterite phase, we control the pH of their solution and repeatedly coat particles with silica (Fig. 2). Synthesis solutions are buffered to \(\text{pH} = 9.5\) by \(\text{n-cyclohexyl-2-aminoethanesulfonic (CHES)}\) acid (see “Methods” for details). A typical yield for these conditions results in a cloudy suspension of billions of particles with a mean size of \(3.5 \pm 0.8 \mu m\) (Supplementary Fig. 3). We orientation of the polarizer (P)—is de-polarized whenever the optical axis of the rotating particles is aligned with neither the polarizer nor analyzer (A). We compute the magnitude of the Fourier transform \(\langle F^2 \rangle\) of the blinking patterns (inset) of the two particles in (b) shows that the frequencies at which the particles de-polarize the incident L.E.D. light are centered around \(0.5\) Hz, corresponding to a rotation frequency of \(0.125\) Hz. The magnitude of the sum of transforms, \(\langle F^2 \rangle\) (solid line), decays, confirming that the particles’ orientations are out of phase. Scale bar: 5 \(\mu m\).
control the particles’ size by varying the stirring speed, initial reactant concentration, and reaction time. To further promote phase stability in solution, limit flocculation, and preserve vaterite’s optomechanical behavior, particles are coated with silica by the addition of (3-aminopropyl)trimethoxysilane (APTMS) followed by tetraethyl orthosilicate (TEOS, see Methods and Fig. 2b, c). Repeated silica precipitation alters the particles to be inert with long-term stability at room temperature. Typical experiments are performed in heavy water (D2O), selected for its lower absorption of infrared radiation.

Translational dynamics of individual rotors

When the laser is turned off, vaterite micro-spheres sediment onto the capillary’s bottom surface at a constant speed. Vertically shifting the imaging focal plane from the bottom of the capillary to its top surface, we monitor the time it takes for particles to travel 100 μm, corresponding to when a focused image of a particle re-appears (see Supplementary Note 4 for details). We measure the force from radiation pressure \( F_{rad} = 4\pi\rho_d^2\alpha c \) by computing the particles’ vertical rising speeds and extracting the intrinsic reflection coefficient \( \rho = 0.22 \pm 0.01 \) (Supplementary Fig. 2).

For a power flux of 40 MW m\(^{-2}\) incident on a \( d = 3 \) μm vaterite particle, the gravitational height will increase from \( \sim 15 \) nm to \( \sim 180 \) nm resulting in \( \sim 90\% \) increase in the translational diffusion, \( D_t \), consistent with measured diffusion coefficient as extracted from the MSD (Fig. 4a, b).

Rotational dynamics of individual rotors

We measure the stochastic rotational diffusion with no drive by monitoring the transmitted light intensity of each particle. When confined to a two-dimensional plane, vaterite particles undergo rotational diffusion. When imaged under crossed-polarizers (PA), the particles’ birefringence modulates the intensity of the scattered light. The fluctuations of the transmitted light intensity lead to temporal decorrelation, \( g_{\Delta}(t) \), that holds information about the particle’s orientational diffusion. The decorrelation rate depends on the rotational diffusion matrix \( D_r \), which near a wall is dominated by the spinning diffusion (rotation axis is perpendicular to the wall), \( g_{\Delta} = \exp(-6D_r t) = \exp(-6D_{\perp} t) \) (see Fig. 3c, inset and the Supplementary Note 2). To leading order, the diffusive spinning approaches its bulk value

\[
D_{\perp} = \frac{k_B T}{\eta_0 d^2},
\]

and is consistent with the experimentally measured diffusion constants in the particle size range studied (Fig. 3c). The lubrication flows responsible for the reduction in translational diffusion (2) have little effect on the particle’s spinning relative to their bulk dynamics (3). This relation is significant for the spin-orbit coupling of a pair of rotors.

When illuminated with circularly polarized light, individual particles rotate at a steady angular speed of up to \( d\theta/dt = \Omega = 1.2 \) rad s\(^{-1}\). Diffusive spinning is dominated by the external drive, with the average rotational Pécelt, \( P = \Omega/D_t = 25 \). For a given particle, the spinning
Angular frequency is given by the balance of optical torque and viscous drag

$$\Omega = \frac{\mathcal{T} J}{8 \pi \eta d^2 c/4} \left[ 1 - \cos \left( \frac{2\pi \Delta n d}{\lambda} \right) \right]$$

(4)

where \( \eta = 1.25 \text{ mPa} \cdot \text{s} \) is the surrounding fluid’s viscosity\(^{37} \), \( c \) is the speed of light in vacuum, and \( \mathcal{T} \) is the transmission coefficient\(^{38,39} \) (see Supplementary Note 5 for a detailed derivation). In the chosen wavelength, vaterite has negligible absorption\(^{16} \), and in the short wavelength approximation, the transmission of the refracted rays is given by \( \mathcal{T} = 1 - R \). We measure the birefringence of the particles, \( \Delta n \), by tuning the ellipticity of the incident beam. In the presence of polarized light with ellipticity \( \phi \), the optical torque is composed of both an aligning and spinning torque. For circularly polarized light, the alignment torque vanishes. Conversely, the spinning torque vanishes for linearly polarized light. The effective birefringence of the polycrystalline vaterite colloids is then measured by considering the minimum ellipticity required to generate a net torque that overcomes the viscous torque \( \tau_v \) for a given particle size (Fig. 4c). We measure the effective birefringence as \( \Delta n = 0.075 \pm 0.015 \), consistent with \( \Delta n \) values for polycrystalline vaterite micro-particles reported in the literature between 0.06 – 0.09\(^{26,40} \). Using the measured birefringence, \( \Delta n \), and transmission, \( \mathcal{T} \), we quantitatively predict the rotation rate of individual particles as a function of flux \( J \) and size \( d \) (Fig. 4d). Note that the dependence of \( \Omega \) on \( d \) is non-monotonic due to the effectiveness of a particle as a wave plate, peaking at the thickness of an ideal half-wave plate \( d_{\text{max}} = \frac{1}{2} \lambda n = 5 \mu\text{m} \) (see Eq. (4), and Fig. 4d).

**Asynchronous rotation**

We find that particles spin asynchronously by using microscopic imaging that monitors the intensity of light transmitted through individual vaterite microspheres. Particles are imaged between crossed-polarizers using a custom-built microscope with bright field (\( \lambda = 505 \text{ nm} \)) illumination (see Fig. 1a and Supplementary Note 1 for details). When spinning, vaterite depolarizes the transmitted light from the LED source periodically. This occurs whenever the optical axis of the particle coincides with neither polarizer nor analyzer axes, corresponding to four depolarization peaks, or blinks, per period. (Fig. 1c). The relative orientation of an ensemble of vaterite particles is...
transform, for details). For individual particles, the magnitude of the Fourier transforms of different photonic fluxes is shown in the inset. b Translational diffusion increases with the gravitational height $h_g$ increases (solid line) until it approaches the bulk value (dashed line). Colors of points in (b) correspond to the legend in (a). c The effective birefringence of a particle, $\Delta n$, can be measured by monitoring the minimal ellipticity, $\phi_{m\text{inset}}$, where rotation begins for different particle sizes (inset). d Measured spin rates for different fluxes and different particle sizes (inset) as predicted by Eq. (4) (solid lines), are consistent with measured birefringence. Error bars correspond to the standard deviation.

free to vary. Tracking the transmitted light intensities of individual particles as a function of time, $I_i(t)$, allows for a direct measure of each particle’s rotation frequency and phase. Simultaneously monitoring two spinning particles shows that the periodically oscillating intensities of the light they transmit are close in frequency but differ in phase (Fig. 1c). Computing the Fourier transform of the light intensities of individual rotors, $\mathcal{F}_I[I_i(t)](\omega) = \int dt e^{-i\omega t}I_i(t)$, allows us to globally compare the phases of multiple particles (see Supplementary Note 6 for details). For individual particles, the magnitude of the Fourier transform, $\sqrt{\mathcal{F}_I[I_i(t)]^2}$ peak at $\approx 0.5$ Hz, corresponding to four times the typical particle spinning frequency (~0.125 Hz). However, the sum of the individual Fourier transforms, $\sum_{i} \mathcal{F}_I[I_i(t)]^2$, decays with the number of particles. The different phases of the light intensities do not necessarily add up constructively, indicating that particles are globally asynchronous with respect to one another (Fig. 1d). Yet when two rotating particles approach each other, they mutually advect through their flow fields. Moreover, we observe a change in their blinking rate, indicating a change in their angular speed.

**Single particle flow field**

To understand the coupling of rotor pairs, we first consider the flow field generated by a single rotor. In a uniform optical torque field, an isolated spinning vaterite micro-sphere stirs the surrounding fluid, generating an algebraically decaying flow. A single rotor can be modeled as an isolated sphere with radius $a$ centered at $r$, subjected to a constant torque $\tau = 8\mu a^2 \mathbf{\Omega}^s$, where $\mathbf{\Omega}^s$ is the angular velocity of the isolated rotor (Fig. 5). A multipole expansion well approximates the resulting flow generated by the sphere’s rotation. We introduce a singularity at position $(x_0, y_0, z_0)$, acting as a point-torque disturbance (rotlet). The corresponding Green’s function, $G_{\text{rotlet}}$, satisfying the Stoke’s equations is $G_{\text{rotlet}} = -\frac{1}{4\mu} \frac{\delta(r)}{r}$, where $\delta$ is the Levi-Civita symbol whose indices represent components of the rotlet’s position in the Cartesian coordinate system, and $r$ is a 3D vector pointing from the rotlet to a point $(x, y, z)$ in space. In an unbounded 3D Stokes fluid, the magnitude of the far-field flow of a force monopole (a Stokeslet) is $|\mathbf{u}_{\text{Stokeslet}}| \propto 1/r$, and a force dipole $|\mathbf{u}_{\text{dipole}}| \propto 1/r^2$. To leading order, the rotors are force-free but experience a torque. The resulting flow is given by an anti-symmetric derivative of the Stokeslet (a rotlet) that decays as $|\mathbf{u}_{\text{rotlet}}| \propto 1/r^2$.

In all experiments, the micro-rotors are found near a solid wall, imposing a no-slip boundary condition ($\mathbf{u}(z=0) = 0$) Fig. 5). Their flow field obtained by the method of images decays like the following derivative -1/r$^{a_2}$. Explicitly, we may approximate the far-field fluid flow in the presence of a no-slip boundary as the superposition of two rotlets with equal and opposite torques $\tau = \pm \mathbf{\tau}$ located at $z = \pm \delta$, respectively (Fig. 5). The resulting fluid flow is then $\mathbf{u}(r) = \mathbf{\Omega}^s \mathbf{a}^{\frac{3}{2}} \left\{ \frac{1}{R^3} - \frac{1}{r^3} \right\} (\mathbf{y} - \mathbf{x})$ (see Supplementary Note 7 for a detailed calculation). Here $\mathbf{x}, \mathbf{y}$ are Cartesian unit vectors, and $|R_+| = \left( x^2 + y^2 + z^2 \right)^{\frac{3}{2}}$, representing the distance to a point $(x, y, z)$ in space from the source and image charges, respectively. In the far-field limit, $|R_+|^{-3} = \frac{1}{r^3} \left( 1 \pm \frac{3\delta}{r} \right)$. When close to the wall ($\delta = a$), the 1/r$^3$ contribution vanishes, and the next term in the multipole expansion is now proportional to 1/r$^2$. This scaling arises from noting that
Recalling that rotating particles generate an algebraically decaying tangential flow field \( \mathbf{u} \propto 1/r^2 \), Eq. (7) becomes
\[
\Delta \Omega = \frac{1}{4}(1-\alpha)\mathbf{\omega},
\]
connecting the spin angular frequency change to the rotating pair’s orbital frequency. The above derivation follows closely the hydrodynamic description of biological micro-rotors near a no-slip boundary \( \frac{1}{4} \) while keeping a implicit, which only changes the slope of the linear spin-orbit coupling. The relation in Eq. (8) is general—

**Methods**

**Vaterite synthesis and sample preparation**

Vaterite micro-spheres are synthesized by controlled precipitation from a super-saturated solution of 0.33M calcium chloride (CaCl₂, Sigma-Aldrich) and 0.33M sodium carbonate (Na₂CO₃, Sigma-Aldrich). We buffer CaCl₂ and Na₂CO₃ to a pH of 9.5 (CHES, Sigma-Aldrich),
before mixing at 1000 RPM in a glass vial with a 1 cm magnetic stir bar. The total stirring time is ~40 s. For these conditions, the typical poly-dispersion is 3.6 ± 0.8 μm, although, by varying the synthesis conditions, a particle diameter range of $d = 2$–12 μm is readily accessible (Supplementary Fig. 3). Particles are then coated via a sequential coating process using (3-Aminopropyl)trimethoxysilane (APTMS, Sigma-Aldrich) and tetraethyl orthosilicate (TEOS, Sigma-Aldrich) Fig. 2. In a typical APTMS coating, 1.5 mL of the synthesis bath is washed in DI H2O 3 times, to which 70 μL of APTMS (Sigma-Aldrich), 25 μL of Ammonia (25% V/V in H2O, Merck) and 940 μL of ethanol (200 proof) are added. The sample is then placed in a shaker for 2.5 h. For TEOS coatings, the procedure is identical (APTMS is replaced with TEOS), except that the sample is allowed to shake for 5.5 h. Electrostatic interactions are minimized by the presence of 14 mM NaCl in the solution, reducing the Debye screening length to 2.5 nm. Microscope samples are made by dispersing the particles in heavy water (D2O, Sigma-Aldrich) and loading into a 100 μm tall glass channel (Vitrotubes W5010050) passivated through vapor deposition of hexamethyldisilazane (Sigma-Aldrich). Loaded capillaries are placed onto a clean microscope glass slide and sealed on their ends with UV-curable resin (Loon Outdoors UV Clear Fly Finish).

**Experimental setup**

Imaging is performed on a custom-built, bright-field microscope coupled to a laser source. A commercial light emitting diode (λ = 505 nm Thorlabs) with a diffuser (ground glass N-BK7 600 grit, Thorlabs), condenser, and an iris are used to achieve Köhler illumination. The scattered light is picked up by the microscope objective (HCX PL APO 40x NA = 0.85, Leica) and a tube lens (B&H), detected by a digital camera (DCC1545M, Imaging Source), and acquired using commercial video recording software (IC Capture, Imaging Source). A laser beam was introduced on a separate optical path (see Supplementary Fig. 1a). A λ = 1064 nm laser beam (YLR-10-1064-LP, I.P.G. Photonics) passes through a zero-order half-wave plate (WPH05M-1064 Thorlabs) and is contracted using a customized Galilean telescope to achieve a wide beam (Supplementary Fig. 1b). The laser beam is introduced into the sample using a polarizing beam splitter (PBS CM1-PBS253 Thorlabs). Its intensity at the sample is controlled by a combination of the electronic laser head controller and adjustment of the half-plate. The intensity is measured using an optical power meter (PM100D power meter, with S175C sensor, Thorlabs). In order to eliminate laser intensity before the camera, stained glasses (FGS900S, Thorlabs) are stacked after the objective.

**Data availability**

The data generated in this study are available at an online repository and can be accessed at the following URL: https://doi.org/10.6084/m9.figshare.22294690.

**Code availability**

The custom codes used in this study are available from the corresponding author upon request.
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Author contributions
A.M., M.Y.B.Z., and P.M.C. conceived the project. A.M. and M.Y.B.Z. designed and conducted the experiments, data analysis, and developed the theoretical model. All authors contributed to the writing of the manuscript.

Competing interests
The authors declare no competing interests.

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