Irregular spin angular momentum transfer from light to small birefringent particles

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The transfer of spin angular momentum from photons to small particles is a key experiment of quantum physics. The particles rotate clockwise or counterclockwise depending on the polarization of the light beam which holds them in an optical trap. We show that even perfectly disk shaped particles will in general not rotate with a constant angular speed. The particles will periodically accelerate and decelerate their rotational motion due to a varying spin angular momentum transfer from the light. Using the Poincaré sphere we derive the equation of motion of a birefringent plate and verify the results by measuring the time dependent rotation of small crystals of Hg(I) iodide and 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA) in the trap of polarized optical tweezers. For small ellipticities of the polarized light in the tweezers the plate stops in a fixed orientation relative to the axes of the light ellipse. We discuss the origin of this halt and propose an application of small birefringent plates as self-adjusting optical retarders in micro-optics.

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I. INTRODUCTION

In 1935, Beth was the first to show that the spin angular momentum of light could be transferred to a macroscopic birefringent plate, which started to rotate in a polarized light beam [1]. Many years later, Friese *et al.* [2] repeated the experiment with microscopic calcite particles held in optical tweezers. These much smaller platelets rotate fast in the polarized laser beam [3,4]. Besides being a key experiment in physics, the spin transfer has also applications in microfluidics and biophysics [5–7]. There are other ways of rotating a small particle with the help of light [8–11]. But in contrast to most of these ways, a rotation driven by spin transfer can be simply controlled by controlling the polarization of the tweezing beam. And the amount of spin transfer is a crucial quantity in many applications of the optical tweezers [6,12]. In this work we present a general theoretical model for the transfer process of spin angular momentum from light to rotating birefringent media. The polarization of light and thus the expectation value of the photon spin can be visualized with the help of the Poincaré sphere [13,14]. It is similar to the Bloch sphere and is used in commercially available polarimeters (THOR) to display the polarization state of light. Each point on the surface of this sphere is the geometric locus of a certain polarization of the light. In order to track the polarization changes which the light undergoes through optical elements, one considers rotations [elements of the special orthogonal group SO(3)] along small or great circles on the sphere and then finds the resulting polarization with the standard formulas of spherical geometry. This method is equivalent to multiplying Jones matrices [elements of the special unitary group SU(2)], but it has significant advantages. We also present two results of our investigations: the transfer of spin angular momentum from photons to a rotating birefringent particle depends on its actual orientation and is, in general, not constant. In our experiment we analyze the time dependence of the rotational speed of the particle and compare it with a theoretical model using the Poincaré sphere. Our results show that only half wave retardation plates rotate at constant speed in an elliptically polarized light beam ($\omega_0 > 0$). In addition, we found that under certain polarization conditions a birefringent platelet starts to rotate, but it then stops with a fixed orientation of its birefringent axes. The platelet then acts as a pseudo-optically active plate and the effect can be used to self-adjust small optical parts on optical microchips.

II. EXPECTATION VALUE OF THE PHOTON SPIN ON THE POINCARÉ SPHERE

An arbitrary elliptical state of polarization P on the Poincaré sphere is given by

$$P = \sin(\delta/2)\exp(+i\varphi/2)R + \cos(\delta/2)\exp(-i\varphi/2)L, \quad (1)$$

where R and L are the vectors which represent the normalized right and left circularly polarized light waves in Cartesian coordinates,

$$L = \frac{1}{\sqrt{2}}\exp[i(\Omega t - kz)]\begin{pmatrix} 1 \\ \exp(+i\pi/2) \end{pmatrix}, \quad (2a)$$

$$R = \frac{1}{\sqrt{2}}\exp[i(\Omega t - kz)]\begin{pmatrix} 1 \\ \exp(-i\pi/2) \end{pmatrix}. \quad (2b)$$

We use the "optical convention" where the electrical vector of a right circularly polarized wave follows a right handed screw [13,15]. $\varphi = 2\lambda$ is the *azimuth or longitude* and $90° - \delta = 2\omega$ is the *altitude or latitude* of P on the Poincaré sphere. λ is the tilt angle of the light ellipse with respect to the x axis and ω is the ellipticity $\tan\omega = \pm b/a$, where a, b are the large and small half axes of the light ellipse and the P_1 sign characterizes a left handedness of the elliptical polarization P and its locus on the upper half of the sphere [13] (see Fig. 1).

The helicity of right (left) circularly polarized light is +1 (−1): $\hat{s}_z|R\rangle = +1\hbar|R\rangle$ and $\hat{s}_z|L\rangle = -1\hbar|L\rangle$, which can be inter-

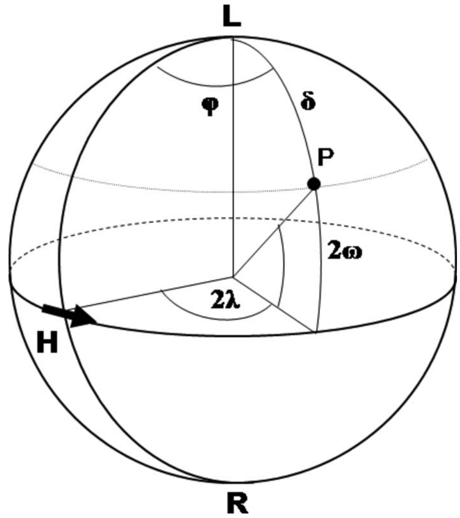

FIG. 1. Polarization of light on the Poincaré sphere. $\varphi=2\lambda$ is the azimuth and $90°-\delta=2\omega$ the altitude of the point P. λ is the tilt angle of the light ellipse with respect to the x axis; ω is the ellipticity $\tan\omega=\pm b/a$. The $(\pm)$ sign characterizes the handedness of the elliptical polarization P.

preted as the two spin states of a photon. We then find the expectation value for the spin angular momentum of a photon in a light wave with polarization P from Eq. (1),

$$\langle P|\hat{s}_z|P\rangle = -\hbar\cos\delta = -\hbar\sin 2\omega. \tag{3}$$

This expectation value does not depend on the azimuth. This is evident since the azimuth angle defines the orientation of the light ellipse in an arbitrary coordinate system which we can choose freely. Thus, all polarization states on the same altitude of the Poincaré sphere have the same expectation value for the spin angular momentum of their photons. As a consequence, an optical active plate, which transforms the polarization only along the altitude, does not change the expectation value for the spin angular momentum of the photon. On the other hand, every birefringent plate, which changes the ellipticity of the light, changes the expectation value for the photon's spin angular momentum. This is the reason why angular momentum can be transferred from light to birefringent retardation plates, an effect, which we will study in the following. Note that the expectation value for the spin angular momentum [Eq. (3)] does not depend on the energy $\hbar\Omega$ of the photon very much in contrast to linear momentum.

III. ANGULAR MOMENTUM TRANSFER FROM LIGHT TO BIREFRINGENT RETARDATION PLATES

The conservation of angular momentum is a consequence of the isotropy of space. If light passes an anisotropic birefringent plate and changes its polarization and therefore its altitudinal position on the Poincaré sphere, it changes its angular momentum and the plate starts to rotate to conserve it. The largest spin angular momentum transfer of a single photon to a birefringent plate is $2\hbar$ when the photon is right circularly polarized and passes a $\lambda/2$ retardation plate to become left circularly polarized.

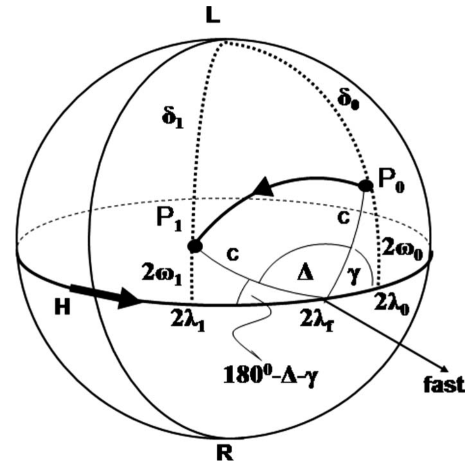

FIG. 2. Angular momentum transfer from light to a birefringent plate. The linear birefringent plate, whose fast axis is oriented at the azimuth angle 2λ with respect to the horizontal of the coordinate system, transforms the incident polarization state $P_0(2\lambda_0,2\omega_0)$ along the arc Δ into the polarization state $P_1(2\lambda_1,2\omega_1)$ behind the plate. The length of the arc is given by the retardation Δ of the plate.

A most useful property of the Poincaré sphere is the simple representation of the change in polarization in a linear birefringent retardation plate. In this plate a polarization P_0 is transformed into a polarization P_1. The retardation plate is characterized by its orientation angle 2λ of its fast axis on the equator of the Poincaré sphere and by its retardation angle Δ (Fig. 2). The polarizations P_0, P_1 now lie on the beginning and the end of a circular arc of length Δ around the position of the fast axis on the sphere, drawn from P_0 in a counterclockwise sense [13,14]. Usually this arc is part of a small circle around the point 2λ on the equator.

Linear birefringent half wave plates transfer a polarization state from an altitude above the equator of the Poincaré sphere to a polarization state with the same altitude below the equator and vice versa $(2\omega_1=-2\omega_0)$. This means that the spin angular momentum transfer to the plate is a constant regardless of the plate's orientation and does not change when the plate rotates [14]. This is not the case for birefringent plates with other retardations. In general, the drive due to spin angular momentum transfer varies during the plate's rotation and leads to an oscillatory rotational acceleration and deceleration. In the following we want to calculate the spin angular momentum transfer from a general incident polarization P_0 to a linear birefringent plate of retardation Δ using our tools on the Poincaré sphere.

Figure 2 shows the initial and final polarization states as well as the evolution between them. The linear birefringent plate with the azimuth angle 2λ of its fast axis transforms the incident polarization state $P_0(2\lambda_0,2\omega_0)$ on a small circle with radius c on the arc Δ (its retardation) into the polarization state $P_1(2\lambda_1,2\omega_1)$ behind the plate. From the basic equations of spherical geometry for rectangular triangles the spin angular momentum transfer to the plate can be calculated. One first determines $\sin 2\omega_1=\sin(180°-\Delta-\gamma)\sin c$ and separates Δ and γ by the addition theorem. Then $\sin\gamma$ is calculated from $\cos\gamma=\tan(2\lambda_0-2\lambda)\cot c$. After insertion one finds that the average spin angular momentum transfer per photon to a plate with retardation Δ is

$$\hbar \sin 2\omega_1 - \hbar \sin 2\omega_0 = \hbar\{\sin 2\omega_0(\cos \Delta - 1) + \cos 2\omega_0 \sin \Delta \sin(2\lambda_0 - 2\lambda)\}. \quad (4)$$

This angular momentum transfer consists of two terms: the first one is zero at the equator and extreme at the poles and does not depend on the orientation of the plate. It is proportional to the expectation value for the spin angular momentum of the incident photons and thus proportional to the ellipticity of the polarization. It provides a constant torque on the plate and drives its rotation.

The second term is zero for circularly polarized light (at the poles) and extreme for linearly polarized light (on the equator of the sphere). This term describes the torque of an electric field onto the induced electric dipole moment in the direction of the slow axis of the plate and inverts its sign at $\lambda = \lambda_0$ [16]. The physical reason of this second term is the following: the elliptical electric field of the light wave induces an electric dipole in the transparent optically anisotropic plate and causes a torque like the earth magnetic field on a compass needle or like an electric field on elongated particles [7]. This torque varies in magnitude and direction during the rotation of the platelet. Linearly polarized light will lead to maximum torque and zero rotational drive.

IV. DYNAMICS OF A SMALL BIREFRINGENT PLATELET IN A POLARIZED LIGHT BEAM

The angular momentum transfer per second is the torque which acts onto the birefringent platelet. We have to multiply Eq. (4) with the number N of the photons per unit time and find the equation of motion,

$$\Theta\frac{d^2\lambda}{dt^2} = N\hbar\{\sin 2\omega_0(\cos \Delta - 1) + \cos 2\omega_0 \sin \Delta \sin(2\lambda_0 - 2\lambda)\} - \eta\frac{d\lambda}{dt}, \quad (5)$$

where Θ is the moment of inertia of the platelet and $d^2\lambda/dt^2$ is its angular acceleration. To consider the realistic case, in which the birefringent platelet is suspended in a viscous liquid, we have subtracted a Stokes friction term $\eta\frac{d\lambda}{dt}$ proportional to the angular velocity of the plate.

We solved Eq. (5) numerically (MATHLAB: ODE23); the results for a quarter wave plate ($\Delta = \pi/2$) are shown in Fig. 3 and depend mainly on the ellipticity ω_0 of the incident light. Two typical solutions are found: for large ellipticities the platelet rotates with periodically varying angular speeds; for small ellipticities (including linear polarization) it performs a damped oscillation and then stops at a fixed angle. In the limiting case, where the incident light has a circular polarization—with the extreme ellipticity $\omega_0 = \pi/4$—the rotation angle λ of the quarter wave plate increases quadratic in time until friction compensates the acceleration. After this transient period the rotation angle increases linearly with

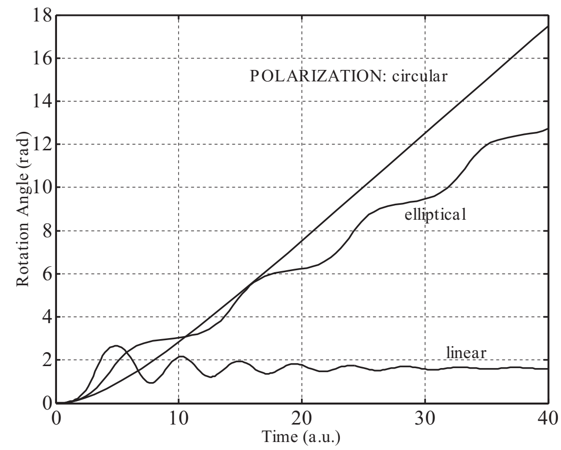

FIG. 3. Rotation angle of a birefringent platelet $\Delta = \pi/2$ versus time at three different incident states of polarization.

time and the angular velocity becomes a constant, very much like the linear fall of a mass through the atmosphere in the gravitational field of the earth. The same linear behavior is obtained if the platelet is a half wave plate ($\Delta = \pi$). In this case, the second term in the acceleration [Eq. (5)] vanishes and only the pure rotation term persists (see also below). If the incident light has an elliptical polarization with a larger ellipticity, an oscillation due to the interaction of the light's anisotropic electric field with the induced electric polarization in the plate (the compass needle effect) is superimposed on the rotation with constant velocity. From the inversion symmetry of the platelet and the very high frequency of the light in the tweezers we expect an oscillation with the period π. For incident light with a small ellipticity—that is with a slender light ellipse—this effect overbalances and the platelet stops at a fixed orientation angle after a transient damped oscillation.

This fixed orientation angle can again be illustrated on the Poincaré sphere [Fig. 4]. If the birefringent platelet stops, the momentum transfer onto the platelet must be zero. This means that the incident light P_0 and the light in the exit P_1 have the same angular momentum $\omega_0 = \omega_1$ [Eq. (4)] and thus lie on the same altitude. In principle, there are two possible positions of the platelet on the equator of the sphere, but the only stable one is the one where the slow axis lies exactly in the center of the smaller arc(P_0P_1). The polarization state inside the plate now develops clockwise along the arc(P_0P_1), which results in a rotation of the light ellipse from the orientation angle λ_0 to angle λ_1 (Fig. 4). In contrast to a compass needle, which has its potential minimum in the exact direction of the magnetic field, the slow axis will orient near the large axis of the light ellipse only if the retardation is small or nearly 2π or if the incident light is nearly linearly polarized. With spherical trigonometry we find for the orientation λ_s of the slow axis of the plate in the halt position,

$$\sin(2\lambda_s - 2\lambda_0) = \tan 2\omega_0 \tan \Delta/2, \quad (6)$$

a result we would also obtain if we set Eq. (4) equal zero (Fig. 5).

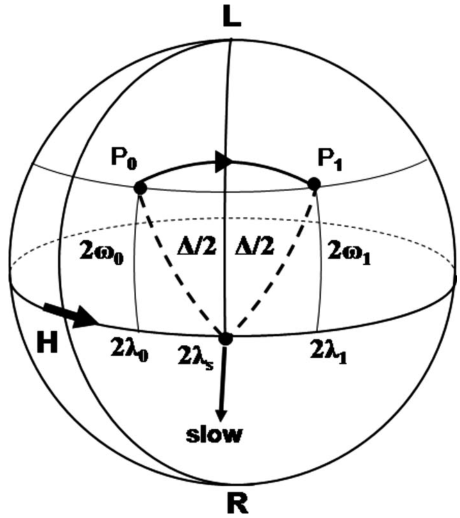

FIG. 4. Stationary solution on the Poincaré sphere. The birefringent platelet stops if the momentum transfer onto the platelet is zero. Therefore, the incident light P_0 and the exiting light P_1 must have the same expectation value for the spin angular momentum $\omega_0=\omega_1$ and thus lie on the same altitude. The slow axis of the birefringent platelet comes to rest exactly in the center of the small arc(P_0P_1).

Equation (6) can have interesting consequences for the active orientation of small birefringent plates with the help of the incident polarization of light [17]. If we change the ellipticity ω_0 of the incident light, the plate will adjust its orientation according to Eq. (6). At the same time the exiting light ellipse, which has passed the platelet, will take on the orientation $\lambda_1=2\lambda_s-\lambda_0$ and maintain the ellipticity ω_0 of the incident light. The birefringent platelet acts like an optically active plate in this case, e.g., a quartz plate cut perpendicular to the optical axis. Fluctuations of the incident ellipticity can be translated into fluctuations of the orientation of the light ellipse in the exit. For $2\omega_0>90°-\Delta/2$ the plate starts to rotate permanently (Fig. 5).

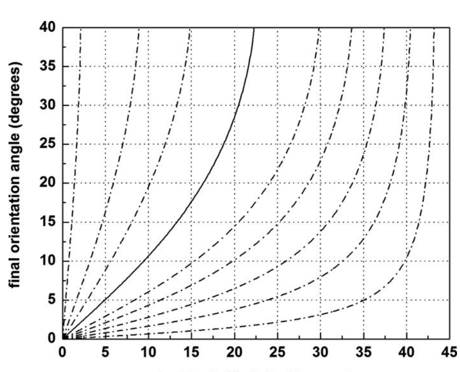

FIG. 5. The orientation $\lambda_s-\lambda_0$ of the slow axis of the plate in the halt position versus ellipticity ω_0 of the incident polarization for various phase shifts Δ. From left to right: $\Delta=\Lambda/2.1, \Lambda/2.5, \Lambda/3, \Lambda/4$(solid curve), $\Lambda/6, \Lambda/8, \Lambda/12, \Lambda/20, \Lambda/50$, where Λ is the wavelength of light. The plate starts to rotate permanently for $2\omega_0>90°-\Delta/2$.

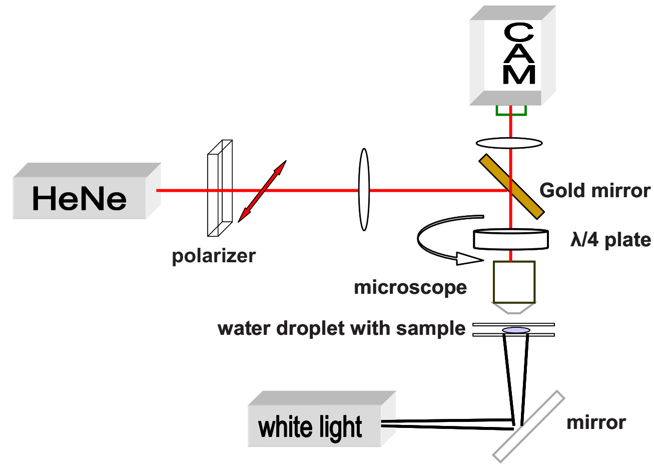

FIG. 6. (Color online) Birefringent platelets were held in optical tweezers and filmed with a video camera. The tweezing light from a HeNe laser was polarized with a polarizer and a rotatable quarter wave plate. The beam was focused via a gold mirror and the high aperture microscope objective onto the object slide with the sample suspension of the microcrystals in water.

V. EXPERIMENTAL RESULTS

A. Setup

To demonstrate the periodically varying angular speed of birefringent platelets in polarized light, we held them in optical tweezers and filmed their rotation. Our apparatus (Fig. 6) was strongly influenced by the work of Bishop *et al.* [7], Smith *et al.* [18], and Bechhoefer and Wilson [19]. A HeNe laser with 40 mW at 632.8 nm was linearly polarized with a polarizer and the optics in the beam was arranged so that this linear polarization was maintained throughout the reflections at mirrors. A gold mirror reflected the red laser light into the microscope. It also allowed viewing and filming of the sample with a digital video camera from above because it transmitted the shorter visible wavelengths of the halogen light which illuminated the sample from below. A quarter wave plate was inserted in front of the microscope objective. This quarter wave plate was rotatable about the beam axis [7]. At an angle of 45° with the incident linear polarization, the quarter wave plate transforms the exiting light into circularly polarized light. By decreasing this angle, polarizations with smaller ellipticities can be produced. The beam was then focused by a high aperture objective (63×, numerical aperture=0.85) onto the object slide with the sample. The power at the focus was only 26 mW mainly due to reflection losses at the gold mirror. The beam waist radius was about 0.9 μm. This beam held the platelet in the focus of the objective and transferred its spin angular momentum to it.

As birefringent materials we used Hg_2I_2 [Hg(I) iodide] [20] and 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA) crystals levitated in water. Both materials are known to have a high birefringence. At 632.8 nm Hg_2I_2 and PTCDA have birefringences of $\Delta n=1.48$ and $\Delta n=0.87$, respectively, which are much higher than that of calcite ($\Delta n=0.17$) or quartz ($\Delta n=0.09$) [21–23]. Since we can only ro-

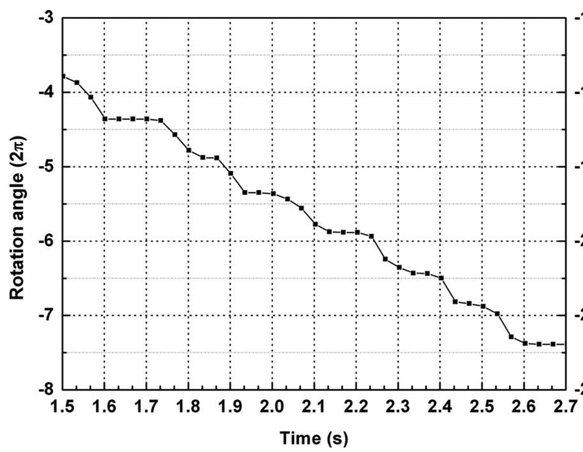

FIG. 7. Periodically varying rotational speed of a small platelet of PTCDA in elliptically polarized ($\omega_0=-30° \pm 2°$) optical tweezers. The orientation of the PTCDA particle was traced frame by frame with a software program (LOGGER PRO). It rotates clockwise with a frequency of about 3 Hz. The steplike structure should be compared with the curve of the elliptical polarization in Fig. 3.

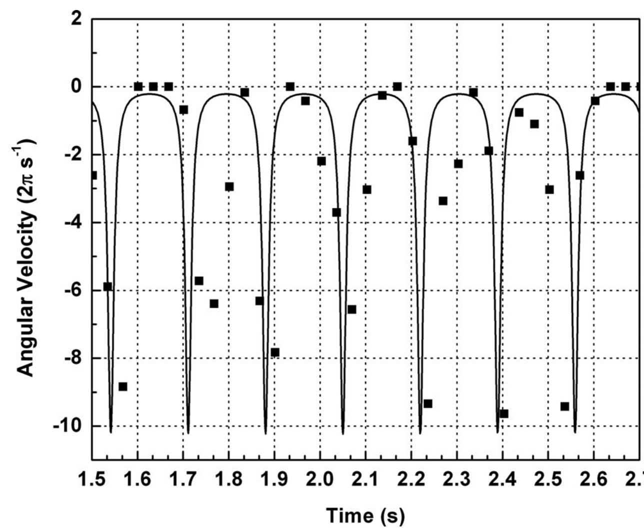

FIG. 8. Angular velocity of the PTCDA platelet. The data points are retrieved from the curve in Fig. 7; the line is the derivative of the theoretical solution $\lambda(t)$ from Eq. (5) for $\omega_0=30.51°$ and $\Delta=\Lambda/3$. The platelet accelerates and decelerates periodically; it almost seems to stop in intervals of one half revolution (180° or $T/2\sim 0.16$ s) which is due to the π symmetry of the light ellipse.

tate small crystals, a high birefringence is essential. The shape of the microcrystals must also be optimal for rotation, so crystal cleavage is important. We obtained microcrystals (diameter between 1 and 3 μm) by crushing larger ones in a mortar and then suspended them in water. Hence, only a small portion of the crystals actually displayed rotation; these were later found by trial and error during the experiment. We placed a droplet of the suspension between a microscope slide and a cover glass. The digital video camera observed this sample and also some of the backscattered laser light which passed through the gold mirror.

B. Experiment

We trapped a microcrystal with a circularly polarized tweezing beam. When we detected a platelet that rotated smoothly and relatively fast (>6 Hz), we switched the helicity of the tweezing light by rotating the quarter wave plate by 90° around the beam axis and observed whether the particle switched its rotation direction accordingly. We also verified whether it still showed smooth rotation at approximately the same angular speed as before. We did this to ensure that the rotation was caused by spin angular momentum transfer and not by some angular momentum due to linear momentum transfer from the light beam. After we confirmed that we were observing spin angular momentum transfer we reduced the ellipticity of the light by slowly reducing the angle of the quarter wave plate from 45° to 0° and then to −45° and filmed the motion: the rotation of the particle began to stutter. This behavior was often more dominant at small ellipticities where the particle almost seemed to flip between two orientations. The rotational speed of the observed particles was most of the time well below the frame rate (~ 30 Hz) of our recording system. We traced the orientation of a PTCDA particle frame by frame with a software program (LOGGER PRO) by observing the orientation of the backscattered asymmetrical diffraction pattern of the laser light. Figure 7 shows the time dependence of the rotation angle in a typical time sequence where the ellipticity of the tweezing light was set to $\omega_0=-30° \pm 2°$. In this sequence the particle seemed to stop at an angle of approximately 45° and at the opposite side at approximately 225°.

The steplike function of the rotation angle versus time comes from the periodic acceleration of the platelet due to Eq. (5) and should be compared with the "elliptical" curve of Fig. 3. Two steps per one full 360° rotation of the particle are expected from Fig. 3 and are due to the π symmetry of the light ellipse. Counting the number of steps in a fixed time interval yields the rotation frequency of the particle, which is about 3 Hz (Fig. 7). The same behavior was observed at $\omega_0=+30° \pm 2°$ in the counterclockwise rotation. When differentiating the curve, we obtain the angular velocity of the platelet (Fig. 8). The graph shows the periodic acceleration and deceleration and the nearly stopping of the platelet after one half revolution, which takes about $T/2\sim 0.16$ s.

VI. DISCUSSION

Small birefringent particles in optical tweezers can be used to drive micropropellers, microcentrifuges, and microhydraulic pumps [5]. The great advantage of spin angular momentum transfer over momentum transfer from light is that it is possible to switch from clockwise to counterclockwise rotation by simply changing the helicity of the light beam. A further step in micromanipulation is the fabrication of heterogenic microstructures by self-adjustment of the construction elements. This can be achieved, for example, by using the halt of a retardation plate in an elliptically polarized light beam to orient it in front of a polarization maintaining optical fiber. Other small optical components can also be oriented by this method if they are attached to an appropriate birefringent particle made, e.g., from a high birefringent plastic. Such a self-adjustment of optical parts can be useful in the production of optical microchips.

An interesting application of rotating halting platelets lies in optical measurement techniques: for a limiting ellipticity ω_0 of the light in the tweezers, a birefringent platelet will stop rotating in its trap at the orientation angle λ_s [Eq. (6)]. The large aperture objective of the optical tweezers usually allows viewing the birefringent platelet and it is then possible to determine the difference $\lambda_s - \lambda_0$ from the conoscopical image of the platelet and the known orientation angle λ_0 of the incident light ellipse. It is advantageous for this measurement to use a weaker light source with another frequency to avoid reorientations. The retardation Δ of the platelet can then be calculated from the parameters $\lambda_s - \lambda_0$ and ω_0 without knowledge of its birefringence and its thickness. If the birefringence $n_s - n_f$ of the material and its approximate thickness are known, its exact thickness is found with $d = m\Lambda\Delta/2\pi(n_s - n_f)$, where Λ is the wavelength of light and m is an integer that reflects the approximate thickness of the platelet. This method of measuring retardations of small crystals needs no knowledge of material and particle data in contrast to a measurement using interference colors. The determination of the thickness of a small particle with size of the order of the wavelength of light can be achieved in this way and would be much easier than with a reticle under the light microscope.

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