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Orientation of optically trapped nonspherical birefringent particles

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While the alignment and rotation of microparticles in optical traps have received increased attention recently, one of the earliest examples has been almost totally neglected—the alignment of particles relative to the beam axis, as opposed to about the beam axis. However, since the alignment torques determine how particles align in a trap, they are directly relevant to practical applications. Lysozyme crystals are an ideal model system to study factors determining the orientation of nonspherical birefringent particles in a trap. Both their size and their aspect ratio can be controlled by the growth parameters, and their regular shape makes computational modeling feasible. We show that both external (shape) and internal (birefringence) anisotropy contribute to the alignment torque. Three-dimensionally trapped elongated objects either align with their long axis parallel or perpendicular to the beam axis depending on their size. The shape-dependent torque can exceed the torque due to birefringence, and can align negative uniaxial particles with their optic axis parallel to the electric field, allowing an application of optical torque about the beam axis.

I. INTRODUCTION

Optical tweezers have been used to manipulate and investigate microscopic particles for many years, and a wide variety of applications has been explored. The underlying principle behind optical tweezers is the transfer of momentum from the trapping beam to the particle [1]. As light can carry angular momentum as well as linear momentum, torque can also be exerted on particles in optical tweezers.

The optical torque acting about the beam axis is always a result of the alteration of orbital and/or spin angular momentum of the incident beam by the trapped particle, by absorption or by scattering if there is either external (shape) or internal (birefringence) anisotropy [2]. Consequently, the torque can either originate from a beam where the incident light itself carries angular momentum that is transferred to the particle, or it can originate from a beam where the incident light carries zero angular momentum, but where the trapped particle induces angular momentum in the beam.

A variety of methods to accomplish angular momentum transfer has been proposed and tested [3–5]. Of these, one of the best suited for actual practical applications is the transfer of incident spin angular momentum to birefringent particles [6–8]. First, spin angular momentum can easily be measured, so that the applied optical torque can be determined by purely optical means, making the system well suited for quantitative measurements [7,9]. Second, the torque can be controlled by changing the polarization state of the light, keeping the power constant. Third, the torque is quite high, typically on the order of $\hbar$ per photon per second if highly birefringent particles are used. Finally, this method can be used with Gaussian beams, ensuring high three-dimensional (3D) trapping efficiency.

However, in order to act as a wave plate, the birefringent particle cannot be oriented with the optic axis parallel to the beam axis. Only for other orientations is the polarization state of the light altered and spin angular momentum transferred from the beam to the particle, causing either constant particle rotation in circularly polarized light or particle alignment in linearly polarized light. Obviously, maintaining the required orientation is crucial for the use of birefringent particles as micromotors or for other applications requiring rotation.

Similar principles apply to flattened or elongated particles which also can alter the angular momentum of the incident light only if their asymmetry about the beam axis is conserved after being trapped. Orientation effects due to the shape of the particle have been reported previously. In particular, it has long been known that elongated particles tend to align with their long axis along the axis of the trapping beam [10]. However, this behavior is not universal, and the orientation of trapped particles depends on their size, shape, and optical properties [11].

The torques giving rise to the orientation of particles with respect to the beam axis have received little attention; this is due in part to the transient nature of the torques, which act to align the particle when it is trapped, and also to the difficulty of calculating torques on nonspherical particles. Earlier work has usually made use of the geometric optics approximation [12], or been restricted to particles of simple geometry and homogeneous and isotropic material [13]. Extension of these foundations to smaller particles for which geometric optics fails, and to more complex particles is highly desirable.

Here we will present numerical calculations—substantiated by experimental results—on how birefringent particles with different aspect ratios and radii will align after being three-dimensionally trapped. Moreover, we are completing the picture of the shape-dependent alignment of particles, showing that elongated particles can align with their long axis either parallel or perpendicular to the beam axis, depending on their aspect ratio and size compared to the beam waist.

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However, the internal anisotropy (birefringence) also contributes to the torque that determines orientation relative to the beam axis. This torque—like the one due to the shape—only occurs at the beginning of a trapping event and has also not yet been studied in detail. Our results reveal that the birefringence-induced torque responsible for alignment to the beam axis has the same order of magnitude as the birefringence-induced torque about the beam axis. This is in contrast to the shape-induced torque which can be an order of magnitude larger relative to the beam axis as compared to the one about the beam axis [13].

We show both experimentally and by computational modeling that, depending on the respective anisotropy, one aligning effect can dominate the other. Using growing lysozyme crystals we were able to observe the transition where the alignment torque due to birefringence overcomes the torque due to the shape when we changed the aspect ratio of a particle while trapped. These findings explain how negative uniaxial calcite crystals can be spun in circularly polarized light [6], despite the fact that such crystals tend to align with their optic axis parallel to the beam axis [8].

II. ORIENTATION OF LYSOZYME CRYSTALS

Lysozyme crystals are a widely used model to study nucleation and growth of protein crystals. In its most common (tetragonal) form, lysozyme forms a positive uniaxial birefringent crystal, with a well-characterized morphology. A schematic drawing showing the crystallographic axes and faces can be found in Ref. [14]. The optic axis, which coincides with the crystal's c axis, points from one tip to the other tip of the crystal. The difference in the indices along the [001] and [110] axes at 1064 nm is 1.66×10^{-3} [15]. The relative ordinary refractive index ($m = n_{crystal}/n_{medium}$) was found to be 1.039 from the measured trap stiffness [16]. The aspect ratio is defined as the length along the optic axis to the width of the [110] face.

The size and shape distributions of lysozyme crystals can be controlled by varying the initial salt and protein concentrations. This allows the experimental study of the shape dependence of the orientation of optically trapped particles. The size and aspect ratio can even be changed dynamically, while the lysozyme crystal is held in the optical trap.

The experiments were carried out using a setup described in detail by Singer *et al.* [15]. Briefly, linearly polarized light from a Yb-doped fiber laser operating at 1070 nm in the range of 200–500 mW was coupled into a $60\times$ oil-immersion objective of numerical aperture 1.4, with a resulting beam waist radius of 0.44 μm. A half-wave plate in the beam path could be used to adjust the direction of the plane of polarization of the trapping beam. The exact alignment of the individual particles in the trap could be determined using a software package developed by Gibson and Kou [17].

We calculated the optical force and torque acting on lysozyme crystals in an optical trap by using the T-matrix method to determine the scattering of the trapping beam by the crystal, and finding the difference between the inflow and outflow of electromagnetic momentum and angular momentum, which is equal to the force and torque exerted on the crystal [5,18–20].

Since the calculation of the T matrix is much faster for axisymmetric particles [21], the lysozyme crystals were modeled as cone-tipped cylinders. Since their relative refractive index is close to 1, and their birefringence is small, even compared with this small refractive index contrast, it was possible to assume an effective isotropic refractive index (which is a function of the orientation of the crystal) to separately calculate the torque due to the shape. The torque due to the birefringence was then calculated using the angle dependence of the orientation energy of the birefringent material in the applied field; this procedure, while approximate, is valid in the low-contrast low-birefringence case of lysozyme crystals.

Equilibrium orientations were found from the dependence of torque on the angle between the axis of the crystal model and the beam axis. The torque versus the angle for different aspect ratios is shown in Fig. 1. Three distinct regimes of behavior—alignment with the crystal optic axis perpendicular to the beam axis, parallel to the beam axis, and at an intermediate angle—can be identified. This behavior can be understood in terms of the usual behavior of nonspherical particles in optical traps—such particles tend to align with their longest dimension along the beam axis. Therefore, a high-aspect ratio lysozyme crystal (elongated along the optic axis) will align with the optic axis parallel to the beam axis. For crystals with smaller aspect ratios the body diagonal is the longest axis and alignment with the body diagonal parallel to the beam axis is to be expected, leading to the optic axis being skewed to the beam axis by an angle depending on the aspect ratio [14]. This type of alignment is widely observed with flattened particles [13]. Meanwhile, since the crystals are positive uniaxial, the torque due to birefringence acts to align the optic axis perpendicular to the beam axis. For low-aspect ratio (flattened) particles, both the torque due to shape and the torque due to birefringence act in the same direction, and the crystal aligns with the optic axis perpendicular to the beam axis, as seen in Fig. 1(a). For elongated crystals, these torques oppose each other, and the transition to alignment along the beam axis requires a larger aspect ratio than would be the case without birefringence.

However, this simple picture fails to explain the existence of the third regime—alignment with the optic axis perpendicular to the beam axis. Notably, the shape torque alone would result in alignment at an intermediate angle, at an angle of 17°. Since the birefringence torque acts to align the optic axis perpendicular to the beam axis, but is insufficient to completely overcome the torque due to shape, this angle is increased to 61°.

We observed all three types of alignment with lysozyme crystals of varying size and aspect ratios. Since it is possible to grow crystals while they are trapped, we were able to observe the transitions between these regimes of alignment in individual crystals. Figure 2 shows the growth of a trapped crystal while the protein concentration in the solution was changed, with a change in orientation between frames 2 and 3. It can be seen that for the given growing conditions the crystal is growing primarily by an addition of material on the (110) faces, as would be expected at a high protein concentration [22]. The measured angle between the optic axis of the crystal and the beam axis is shown in Fig. 3. The transi-
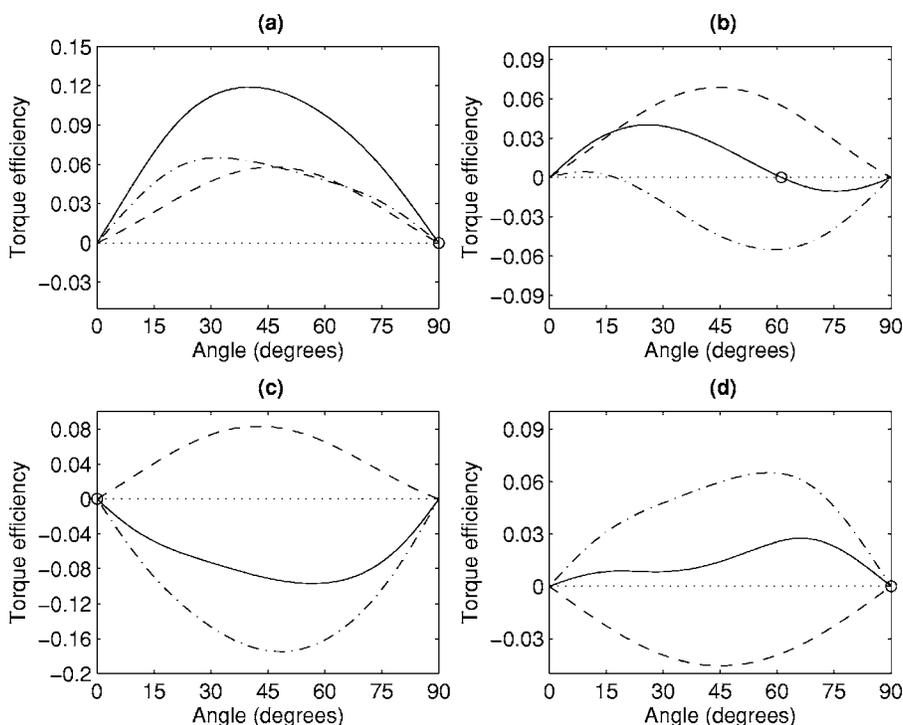
FIG. 1. Torque (shown as torque efficiency, in $\hbar$/photon) acting on trapped lysozyme crystals. Solid lines show the total torque, dotted-dashed lines the torque due to the shape alone, and dashed lines the torque due to birefringence alone. The dotted line indicates zero torque; equilibrium points occur when the total torque curve (solid line) crossed this dotted line, with the stable equilibria indicated by the small circles. In all cases, since the crystals are positive uniaxial, the torque acts to align the optic axis (the symmetry axis of the model crystals) with the electric field. (a)–(c) show the dependence of the torque on the angle between the optic axis of the crystal and the beam axis. A positive torque acts to align the optic axis with the beam axis. The torque is shown for crystals of radius 3 μm and aspect ratios of (a) 0.9, (b) 1.2, and (c) 1.6. Three different equilibrium orientations can be seen: optic axis perpendicular to the beam axis (a), with both the torque due to birefringence and the torque due to shape acting in the same direction; parallel to the beam axis (c), with the two contributions to the torque opposing each other, but with the shape contribution dominant; and at an intermediate angle (b). In (b), if the shape torque was the only torque acting, the crystal would still align at an intermediate angle. The birefringence torque still acts to align the optic axis perpendicular to the beam axis and shifts the equilibrium angle closer to being perpendicular to the beam axis, but is insufficient to overcome the shape torque. The torque about the beam axis acting on the crystal in (a) is shown in (d). A positive torque acts to align the optic axis with the plane of polarization; the angle is the angle between the optic axis and the plane of polarization. The shape torque and birefringence torques oppose each other. In this case, the shape torque is dominant; for larger crystals, the birefringence torque will increase approximately proportional to the radius, while the shape torque will decrease [5].

tion to the perpendicular alignment occurs when the torque due to the birefringence of the crystal becomes dominant over the shape-dependent torque. However, even if the crystals were optically isotropic, this transition would still occur (though at a smaller aspect ratio) when the shape-dependent torque changes direction.

The observed equilibrium orientations of a number of lysozyme crystals of varying size and aspect ratios are shown in Fig. 4. The calculated extents of the different regimes of
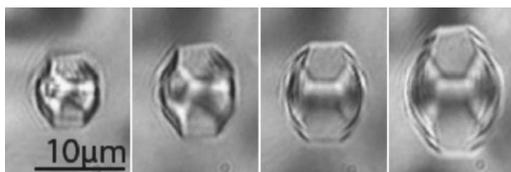
orientation are also shown and agree well with the observed orientations. The crystals were all in the same sample. Note that the crystals drawn in Fig. 4 to show the equilibrium

FIG. 2. Growth of a trapped lysozyme crystal. The aspect ratio can be changed during growth, altering the orientation of the trapped crystal.
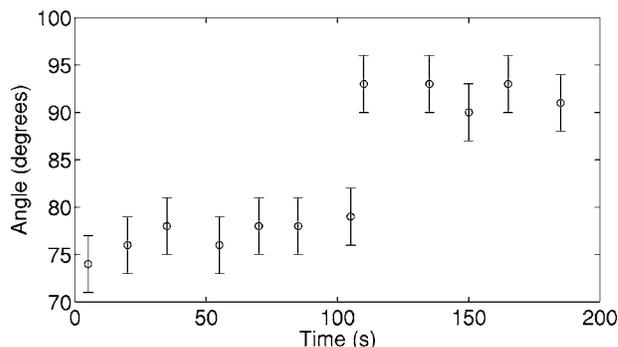

FIG. 3. Change in orientation of a growing lysozyme crystal. The angle between the optic axis of the crystal and the beam axis is shown.
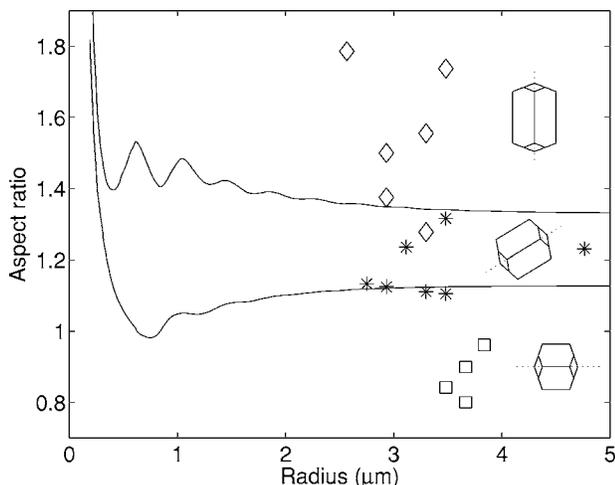
FIG. 4. Equilibrium orientation of trapped lysozyme crystals with different aspect ratios and sizes. The solid lines represent the calculated borders between the different regimes; the orientation within each regime is shown by the inset crystal profiles, showing a side view of the crystal, relative to a vertical trapping beam with plane polarization in the plane of the page. Observed orientations are indicated by ◇—optic axis parallel to the beam axis, □—perpendicular, and *—intermediate. The top-view photographs shown in Fig. 2 correspond to the intermediate (first two frames) and the perpendicular (last two frames) cases.

orientation in each regime are shown in a side view while Fig. 2 shows their appearance when viewed in the microscope (i.e., a top view).

III. OTHER PARTICLES

While the previous results apply specifically to lysozyme crystals, which have a low refractive index contrast with the medium and a small birefringence, the same general principles apply to other particles as well. While the shape-dependent torque varies with the refractive index of the particle (proportional to the refractive index contrast $m-1$ in the low-contrast limit), the orientations for which the torque is zero only weakly depends on the refractive index—a refractive index contrast ten times larger yields boundaries between the regimes of orientation very similar to those in Fig. 4.

This is especially relevant when we consider the trapping and rotation about the beam axis of birefringent particles. If the particle is positive uniaxial, the birefringence-dependent torque acts to align the optic axis perpendicular to the beam axis. The torque on a negative uniaxial particle, on the other hand, acts to align the optic axis with the beam axis. If this is

the equilibrium orientation of the particle, the particle appears to be isotropic as far as the incident beam is concerned, and no transfer of angular momentum occurs [2,8]. This raises the question of why optically trapped negative uniaxial crystals have been observed to spin [6]. It appears reasonable to suppose that, especially since the crystals in question were irregular, shape-dependent torques produced an equilibrium orientation such that the optic axis was not parallel to the beam axis.

It can also be seen in Fig. 4 that, if the particle is small compared to the beam waist, elongated particles can align with their long axis perpendicular to the beam axis, in agreement with previous results for very small particles [11]. These findings can be explained by the fact that particles that are small compared to the beam waist are trapped in the center of the focal spot, where the intensity gradient is small. The elongated objects therefore align with the axis of the highest particle polarizability—their longest axis—in the direction of the electric field vector, and therefore perpendicular to the beam axis, rather than parallel to the beam axis.

IV. CONCLUSIONS

Of the several methods to orient and rotate microscopic particles in optical tweezers, by far the most important to date, as far as quantitative measurements are concerned, is the transfer of spin angular momentum to birefringent particles [7–9]. However, the ability to use birefringent particles is restricted to those which orient with their optic axis not parallel to the beam axis after being trapped, which at first appears to rule out the use of negative uniaxial materials. We have shown that the torque due to the nonspherical shape can overcome the torque due to birefringence, and can be used to maintain negative uniaxial particles in the desired orientation. Furthermore, we showed that elongated particles small compared to the beam waist will align perpendicular to the beam axis.

The results presented are relevant to the design of particles that can be used as motors in optically driven micromachines, and have the potential to increase the range of particles that can serve to probe properties of microscopic or biological systems. The predictability and computability of these torques enable their practical use in optical micromanipulation.

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