

# TRAPPING, MANIPULATION AND RAPID ROTATION OF NBD-C8 FLUORESCENT SINGLE MICROCRYSTALS IN OPTICAL TWEEZERS

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Hemos construido un experimento de pinzas ópticas basado en un microscopio invertido para atrapar y manipular cristales simples de tamaño micro o sub-micrométrico de moléculas fluorescentes de 4-octylamino-7-nitro-enzoxadiazole (Nbd-c8). Estos cristales simples tienen formas de paralelepípedo y exhiben propiedades de birrefringencia evidenciadas con experimentos ópticos entre polarizadores cruzados en un microscopio polarizante. Los cristales son uniaxiales con su eje óptico orientado a lo largo de su dimensión mayor. Atrapados en la trampa óptica, se orientan los micro-cristales orgánicos de una manera tal que su eje largo esté en la dirección de la propagación del haz, y su eje corto siga la dirección de la polarización lineal. Por lo tanto, con la luz linealmente polarizada, simplemente rotando la luz polarizada se puede orientar el cristal. Al usar luz circularmente o solo elípticamente polarizada, el cristal puede rotar y alcanzar espontáneamente una velocidad de rotación de varios centenares de vueltas por segundo. Se ha observado un resultado sorprendente: cuando la potencia incidente está creciendo, la velocidad de rotación aumenta hasta alcanzar un valor máximo y después disminuye incluso cuando la potencia todavía está aumentando. Por otra parte, esta evolución es irreversible. Diversas explicaciones posibles pueden ser consideradas. El desarrollo de un control 3D de los cristales por la holografía dinámica, empleando moduladores espaciales cristalinos líquidos será presentado y discutido en base de los resultados más recientes obtenidos.

We have built an optical tweezers experiment based on an inverted microscope to trap and manipulate single crystals of micro or sub-micrometer size made from fluorescent molecules of 4-octylamino-7-nitro-benzoxadiazole (NBD-C8). These single crystals have parallelepiped shapes and exhibit birefringence properties evidenced through optical experiments between crossed polarizers in a polarizing microscope. The crystals are uniaxial with their optical axis oriented along their largest dimension. Trapped in the optical trap, the organic micro-crystals are oriented in such a way that their long axis is along the direction of the beam propagation, and their short axis follows the direction of the linear polarization. Therefore, with linearly polarized light, simply rotating the light polarization can orient the crystal. When using circularly or only elliptically polarized light, the crystal can spontaneously rotate and reach rotation speed of several hundreds of turns per second. A surprising result has been observed: when the incident power is growing up, the rotation speed increases to reach a maximum value and then decreases even when the power is still growing up. Moreover, this evolution is irreversible. Different possible explanations can be considered. The development of a 3D control of the crystals by dynamical holography using liquid crystal spatial modulators will be presented and discussed on the basis of the most recent results obtained.

**Palabras Claves.** Beam trapping, 42.65.Jx. Optical instruments and equipments, 07.60.-j. Optical elements, devices, and systems, 42.79.-e. Radiation effects on specific materials, 61.82.-d.

## INTRODUCTION

Since the seminal paper and subsequent contributions of Ashkin [1,2], a huge number of publications have been written about optical tweezers [3]. Many fields of applications have been developed in biology and medicine, as well as in chemistry, microfluidics and nanosciences. Actually, optical tweezers are presently one of the most powerful tools to maintain and control small particles without any mechanical or invasive manipulation. They can be applied to a wide variety of objects whose size is in the micrometer scale, or much below.

They are produced by a laser beam, which is focused through a high-numerical aperture microscope objective in a micro-sized spot. Scattering and gradient forces are used for trapping and moving nano- or micrometer sized particles. However, predicting how non-spherical micro-objects behave when grabbed by optical tweezers is difficult. There is a need for systematic investigations carried out on objects with well defined shape and size. Micro-crystals offer an interesting alternative for studying the orientation effects in the laser beam.

Few crystals were already studied including tetra-gonal lysozyme crystals [4], organic semiconductor perylenetetracarboxylic dianhydride platelets [5], cubic NaCl crystals [6], roughly spherical calcite crystals [7,8], and irregular crystals made of quartz [9].

In the following work, we were interested in micro-crystals of an organic dye, more exactly 4-n-octylamino-7-nitrobenzoxadiazole (1), which belongs to a well-known family of fluorescent probes. Micro-crystals were prepared by a solvent-exchange method [10] and obtained as a suspension in aqueous medium. This procedure is easy to implement, leads to reproducible samples and is particularly versatile.

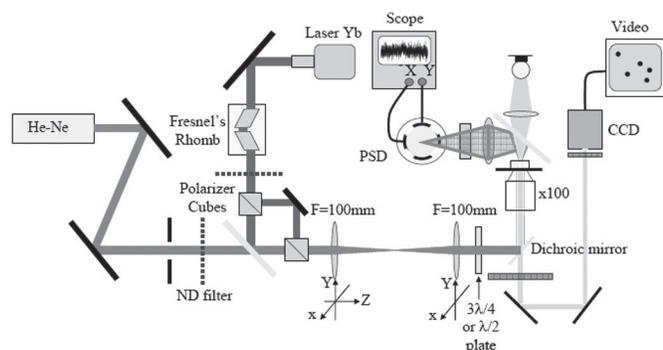


Figure 1: Scheme of the experimental set-up.

Actually, the shape and size of the micro-crystals can be controlled by placing additives, such as macro-molecules, in the crystallization medium [11-14]. A large variety of micro-crystals of this type can thus be obtained [15]. In the present case, regular parallelepiped micro-crystals were generated and their behavior was studied with optical tweezers.

Fig. 1 shows the experimental set-up used in this work, which is a conventional one, fully described elsewhere [16]. The trapping laser source is an Yb-doped fiber laser from IPG, working at  $1,064 \mu\text{m}$  and delivering 1W at maximum power. This laser generated a high quality TEM<sub>00</sub> mode, linearly polarized.

The set-up was improved for the generation of two traps by the use of two polarizing cubes as beam-splitters. For detecting and measuring fluctuations in the position of the trapped crystals, a 10 mW HeNe laser, made perfectly collinear with the IR trapping beam was used.

An optical system records the light scattered by the crystal on a Position Sensor Detector (PSD).

## RESULTS AND DISCUSSION

Although the crystal structure of (1) is still unknown, there is no doubt about the crystalline nature of the micro-objects. In the experimental conditions used for their preparation, a large majority of them are of parallelepiped shape. Their observation between crossed polarizers indicated that they were single birefringent crystals with neutral lines oriented along their long and short axis. They also were uniaxial with the optical

axis oriented along the long axis of the crystals, as proven by the observation of a black cross when observing the crystals with converging light oriented parallel to the optical axis.

*Orientation effects with linearly polarized light.* The crystals were easily captured in the optical trap, even in the case of the largest ones. On the whole, the crystals trapped in linearly polarized light were directed in the same way, i.e. with their long z-axis lying in the direction of light propagation. Thus, in practice, we observed the face of the crystal that was directed perpendicularly to the z-axis. This result agrees well with that reported for micro-cylinders, the long axis of which is oriented in the direction of the beam [17]. In the most frequent situation where the long axis is parallel to the direction of the beam, the orientation of the x- and y-axes of the crystal was directly controlled by the state of polarization of light. To do so, a  $\lambda/2$  crystalline quartz plate was placed before the objective as shown in Fig.1. By rotating the  $\lambda/2$  plate, the direction of vibration of the electric field was varied and therefore the crystal was constantly re-oriented. For any rotation of the  $\lambda/2$  plate by an angle  $\theta$ , the direction of polarization turns by an angle  $2\theta$ , as exactly does the crystal shown in Fig. 2.

The observation of the angular alignment of birefringent micro-objects in the presence of linearly polarized light has indeed been reported [18, 19]. However, this explanation is not valid in our case. In fact, being oriented with their optical axis along the beam propagation, the crystals appear isotropic. A different explanation must be looked for, based on their parallelepiped shape.

*Optical torque induced by circularly or elliptically polarized light.* In some experimental conditions, the crystals were put in rotation with quite a high number of revolutions per second. To do so, the  $\lambda/2$  retardation plate was replaced by a  $3\lambda/4$  plate, thus generating a circularly or elliptically polarized beam. The direction of rotation depends on the angle chosen for the crystalline plate. Indeed, according to the orientation of the axes of the  $3\lambda/4$  plate with respect to the direction of linear polarization of the incoming beam, a right or left circularly or elliptically polarized wave was created and a transfer of the angular momentum of the laser beam could take place. The signal associated with the rapid rotation of a crystal under high incident laser power is displayed in Fig. 3a. This signal does not correspond to a pure sinusoid. Nevertheless, the value of the rotational frequency was easily extracted from its periodicity. Fig. 3b shows the corresponding Fourier transform spectrum in which a neat peak appears at 317 Hz.

Owing to the symmetric shape of the micro-crystals, the peak displayed by the Fourier's spectrum is centered on a frequency value that is twice the number of crystal revolutions per second. Therefore, in the example of Fig. 3, the crystal is rotating at approximately 150-160 turns/s. For some crystals, these frequencies were up to 1 kHz, which means that the number of crystal revolutions per unit of time could reach 500 turns/s.

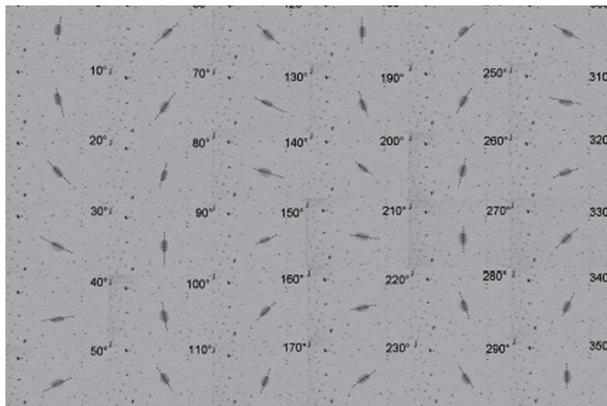


Figura 2: From top to bottom and left to right: Optical micro-scropy images showing the orientation of a micro-crystal upon rotation of the linear polarization of the trapping beam. The ro-tation angle of the  $\lambda/2$  retardation plate is indicated.

To our knowledge, it is the first time that such a high ro-tation speed only induced by light is reported for objects of the size of our micro-crystals. The curve of Fig. 4 shows the evolution of the rotational speed when the incident power is growing up. A strange behavior was observed. As the incidental power continued growing up, the rotational speed reached a maximum value, and then began to decrease. The most in-triguing fact remains the irreversible character of this evolution. Indeed, reducing the laser power did not induce the micro-crystal to turn more rapidly again.

A crude model taking into account a movement of pre-cession and damping forces due to fluid viscosity can predict a parabolic evolution. Nevertheless, more effort is still needed for a better understanding of this behavior. The question of the irreversibility is indeed a most in-triguing point.

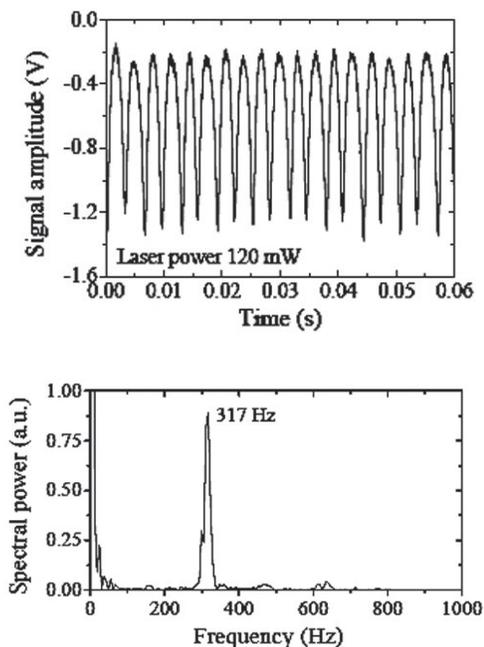


Figura 3: (a) Rotation signal for a micro-crystal under a laser power of 120 mW measured at the laser output and (b) its Fourier transform.

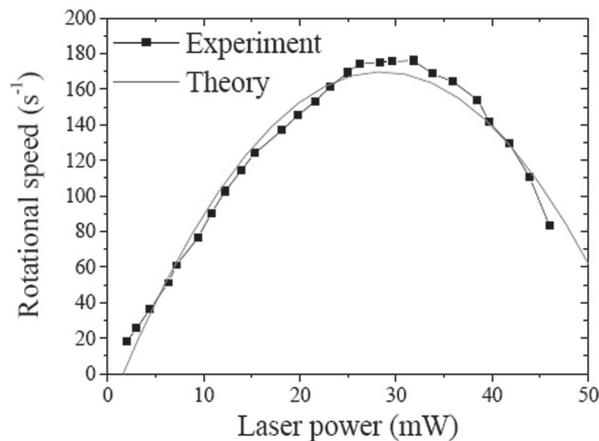


Figura 4: Variation of the rotational speed of a micro-crystal with monotonous increase of the laser power measured near the front focal plane of the objective. Points are experimental. The fitting curve in red is based on a crude model developed in [20].

## CONCLUSION AND PERSPECTIVES

Dye micro-crystals used have proved to be interesting tools to study the behavior of non-spherical particles with optical tweezers. The most remarkable fact is the high rotational speed reached by the crystals. If keeping the laser power below a critical threshold, the speed is proportional to the incidental power. Micro-crystals or similar objects could then be used as stirrers in microfluidic devices. Finally, the intriguing behavior observed at high laser power attracts attention on phenomena that are still badly known but could play a significant role in the fields of micro-fluidics and optical tweezers. An interesting perspective also emerges for the 3D manipulation and control of non spherical objects using holographic tweezers owing to the use of liquid crystal spatial light modulators. In preliminary experiments, we have shown that it was possible to manipulate and orient one crystal by generating multiple optical traps (typically 4 traps were created). A challenging perspective will be to achieve such a control with only one laser beam, spatially structured by dynamic holograms in form of Laguerre-Gauss beams or more complicated spatial shapes.

- [1] A. Ashkin, "Acceleration and trapping of particles by radiation pressure," *Phys. Rev. Lett.* 24, 156-159 (1970).
- [2] A. Ashkin, "History of optical trapping and manipulation of small neutral particles, atoms and molecules in Single Molecule Spectroscopy, R. Rigler, M. Orrit, and T. Basché, eds. (Springer Ser. Chem. Phys. 2001), pp. 1-31.
- [3] M. J. Lang, and S. M. Block, "Resource letter: LBOT-1: Laser based optical tweezers," *Am. J. Phys.* 71, 201-215 (2003).
- [4] W. Singer, T. A. Nieminen, U. J. Gibson, N. R. Heckenberg, and H. Rubinsztein-Dunlop, "Orientation of optically trapped nonspherical birefringent particles," *Phys. Rev. E* 73, 021911 (2006).
- [5] C. Starr, W. Dultz, H. P. Wagner, K. Dholakia, and H. Schmitzer, "Optically controlled rotation of PTCDA crystals in optical tweezers," *AIP Conference proceedings* 772, 1099-1100 (2005).
- [6] U. Gibson, W. Singer, T. Nieminen, N. Heckenberg, and H. Rubinsztein-Dunlop, "Growth of crystals in optical tweezers," in *Optical Trapping and Optical Micromanipulation II*, K. Dholakia, and G. C. Spalding, eds., Proc. SPIE 5930, 294-302, (2005).
- [7] M. E. J. Friese, T. A. Nieminen, N. R. Heckenberg, and H. Rubinsztein-Dunlop, "Optical alignment and spinning of laser-trapped microscopic particles," *Nature* 394, 348-350 (1998); Erratum: *Nature* 395, 621 (1998).
- [8] A. T. O'Neil, and M. J. Padgett, "Rotational control within optical tweezers by use of a rotating aperture," *Opt. Lett.* 27, 743-745 (2002).

- [9] A. La Porta, and M. Wang, "Optical torque wrench: angular trapping, rotation, and torque detection of quartz microparticles," *Phys. Rev. Lett.* 92, 190801 (2004).
- [10] H. Nakanishi, and H. Oikawa, "Reprecipitation method for organic nanocrystals, in *Single Organic Nanoparticles*, H. Masuhara, H. Nakanishi, and K. Sasaki, eds. (Springer Verlag, Berlin, 2003), pp. 17-31.
- [11] F. Bertorelle, D. Lavabre, and S. Fery-Forgues, "Dendrimer-tuned formation of luminescent organic micro-crystals," *J. Am. Chem. Soc.* 125, 6244, (2003).
- [12] F. Bertorelle, F. Rodrigues, and S. Fery-Forgues, "Dendrimer-tuned formation of fluorescent organic micro-crystals. Influence of dye hydrophobicity and dendrimer charge," *Langmuir* 22, 8523-8531 (2006).
- [13] M. Abyan, F. Bertorelle, and S. Fery-Forgues, "Use of linear polymers to control the preparation of luminescent organic microcrystals," *Langmuir* 21, 6030-6037 (2005).
- [14] L. Birl, F. Bertorelle, F. Rodrigues, S. Badré, R. Pansu, and S. Fery-Forgues, "Effect of DNA on the growth and optical properties of luminescent organic microcrystals," *Langmuir* 22, 6256-6265 (2006).
- [15] M. Abyan, D. de Caro, and S. Fery-Forgues, "Suspensions of organic microcrystals produced in the presence of polymers: Diversity of UV/vis absorption and fluorescence properties according to the preparation conditions," *Langmuir* 25, 1651-1658 (2009).
- [16] M. Rodriguez-Otazo, "Réalisation de pinces optiques pour la manipulation de nano et micro objets individuels d'intérêt chimique ou biologique," PhD thesis, Université de Paris 11, 2008 (in French).
- [17] R. C. Gauthier, M. Ashman, and C. P. Grower, "Experimental confirmation of the optical-trapping properties of cylindrical objects," *Appl. Opt.* 38, 4861-4869 (1999).
- [18] E. Higurashi, R. Sawada, and T. Ito, "Optically induced angular alignment of trapped birefringent micro-objects by linearly polarized light," *Phys. Rev. E* 59, 3676-3681 (1999).
- [19] E. Higurashi, R. Sawada, and T. Ito, "Optically induced angular alignment of birefringent micro-objects by linear polarization," *Appl. Phys. Lett.* 73, 3034-3036 (1998).
- [20] M. Rodriguez-Otazo, A. Augier-Calderín, J.-P. Galaup, J.-F. Lamère, and S. Féry-Forgues, "High rotation speed of single molecular crystals in an optical trap with elliptically polarized light," *Appl. Opt.* 48, 2720-2730 (2009).