# Light-Driven Molecular Motor

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An all-optical control of light-angular-momentum transfer to nematic-liquid-crystal molecules is carried out by two copropagating noncoherent waves of orthogonal circular polarization. This allows the continuous variation of the total angular momentum of the light while conserving its circular symmetry. The molecular precession frequency is thus effectively and thoroughly controlled. The light-induced molecular reorientation threshold is shown to depend significantly on the azimuthal symmetry of the excitation light, while remaining the same for different values of its total angular momentum. [S0031-9007(97)02929-3]

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The unique property of liquid crystals (LC) to spatially transfer the rotation momentum gives rise to fascinating intrinsic retroaction phenomena [1]. In addition, the collective character of the interaction of LC molecules with external electromagnetic fields provides extraordinary efficient light-matter coupling [1,2]. The above-mentioned conditions may lead to the light-induced first- [3,4] and second- [5] order phase transitions in the nematic LC (NLC) [6]. These are optical analogs of electric or magnetic field induced threshold reorientation of NLC molecules (so-called Fréedericksz transition), which is, in fact, a phase transition, when taking into account the character of LC orientational order parameter change. The theory of light-induced Fréedericksz transition via dielectric torque in LC materials was developed by Zel'dovich *et al.* [2]. The transition threshold was predicted to depend on the polarization state of the excitation field, and the predicted doubling of this threshold value for circularly symmetric light with respect to the linear polarization case was experimentally proven in Ref. [5]. The delicate role of the light angular momentum in the system behavior above the phase transition threshold was established in Ref. [3]. The realization of the transfer of the light angular momentum to the quasimacroscopic collective of regularly precessing molecules [3] (this would be difficult for the case of macroscopic objects [7]) was rather surprising. The dependence of this precession frequency $\Omega$ upon the external control parameters, such as the excitation light intensity $I$ and ellipticity $\gamma$, is, however, strongly limited [8]. These limitations are determined, on the one hand, by the abrupt (first-order) phase transition induced by the circularly polarized light and, on the other hand, by the requirement of azimuthal symmetry in the light-matter system. Namely, the light intensity must be close to the light-induced phase transition value $I_{\text{th}}$, and the circularity of the electromagnetic field must be high enough as to maintain the regular precession regime [8]. The precession frequency $\Omega$ was proven to exhibit a rather small variation versus $I$ and $\gamma$ even in this regular precession regime, allowing, for example, a maximum of 7% of the $\Omega$ variation upon $I$ [8].

We propose and demonstrate, for the first time, to our best knowledge, an electromagnetic field configuration and light-matter interaction geometry, which enables the continuous modulation of the total angular momentum of the excitation light (keeping at the same time its azimuthal symmetry), and therefore allowing an effective and complete all-optical control of the regular molecular precession frequency $\Omega$ (up to 100% of its corresponding value in the ordinary geometry). This system thus might be called a light-driven molecular motor. The proposed configuration also allowed the experimental study of the light-induced phase transition dependence upon the excitation light symmetry and its total angular momentum.

It is known that the optimal condition for the angular momentum transfer from the light to the matter is the one-direction transformation of the angular momentum of the pure circularly polarized light into the opposite circular polarization [7]. Small static rotation (but not dynamic precession) of a half-wave plate had been experimentally detected [7]. In this case, each circular polarized photon traversing the half-wave plate undergoes an inversion of the circularity sign, transferring an amount of $2\hbar$ of the angular momentum to the macroscopic half-wave plate. Obviously, the light angular momentum cannot be transferred just to a part of the solid half-wave plate, say, to a microscopic domain within it. In LC, however, a quasimacroscopic portion of molecules can be rotated without rotating the whole NLC cell with glass substrates, holders, etc. The size of the affected cluster is self-determined by the so-called coherence length $\xi_c$ [1] of the field-induced torque in LC.

The corresponding detailed theoretical analyses of the light-NLC dynamic interaction regime above the reorientation threshold has been carried out in Ref. [8]. We will use in this work the half-wave plate analogy only to analyze qualitatively the above discussed optimal transfer condition, as well as to describe the light field symmetry and corresponding interaction geometry (see Fig. 1 and the inset). Molecular reorientation in the homeotropic NLC cell (i.e., initial orientation of molecules being

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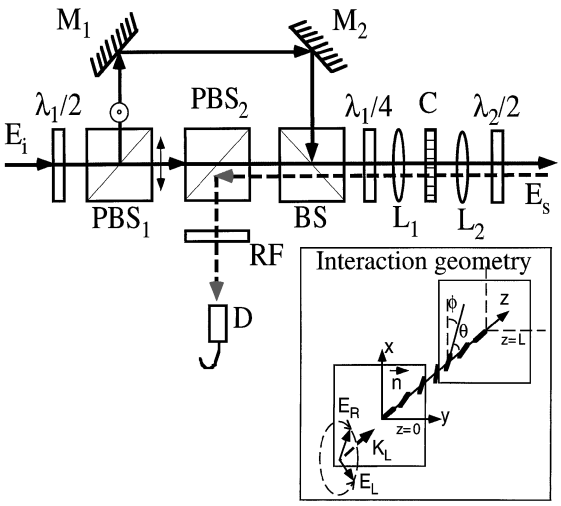


FIG. 1. Experimental setup and interaction geometry. $E_i$—initial linearly polarized beam of argon ion laser (operating at 514 nm), $\lambda_1/2$—half-wave plate (for 514 nm), $PBS_{1,2}$—polarization beam splitters, $M_{1,2}$—mirrors, BS—50/50% beam splitter, $\lambda_1/4$—quarter-wave plate (for 514 nm), C—homeotropic liquid crystal cell (with thickness $L = 90\ \mu$m), $L_1$—lens (with focal length $f_1 = 13$ cm), $E_s$—linearly polarized weak probe He-Ne laser beam, $L_2$—lens (with focal length $f_1 = 10$ cm), $\lambda_2/2$—half-wave plate (for 632.8 nm), RF—red filter, D—detector. Inset: $\vec{n}$—director of NLC (initially parallel with $\vec{z}$), $\theta$—polar angle, $\phi$—azimuthal angle, $E_{R,L}$—electric field of right and left circular polarized incident beams, $K_L$—wave vector of incident beams, L—thickness of the cell, squares are glass substrates (placed at $z = 0$ and $z = L$). $x, y, z$—coordinate system.

normal to the cell substrates) for normally incident light is achieved in the intensity range above a certain threshold $I_{\text{th}}$, when the balance between the elastic and light-induced torque takes place [2],

$$I_{\text{th}} = 2I_{\text{th}}^{L}\{1 + (\zeta_1^2 + \zeta_3^2)^{1/2}\}^{-1}, \qquad (1)$$

where $I_{\text{th}}^{L}$ is the threshold value for linear polarized light (depending on the thickness, dielectric, and elastic parameters of the NLC cell), and $\zeta_\iota$ are the components of the normalized Stokes vector of the excitation field. Thus, both completely depolarized ($\zeta_1 = \zeta_2 = \zeta_3 = 0$) and pure circularly polarized ($\zeta_1 = \zeta_3 = 0, |\zeta_2| = 1$) lights have the same transition threshold $I_{\text{th}}$ (which is twice the linear polarized light threshold). The value of $I_{\text{th}}$ is equal for the same azimuthal symmetry of the excitation field (see the inset in Fig. 1), in spite of the fact that these two fields carry different angular momentum. We have to emphasize, however, that the above-threshold behavior is not taken into account in the derivation of this formula.

The light-induced perturbation of the director $\vec{n}$ (unit vector showing the average molecular axis direction) may be considered as a weak two-dimensional (in a plane) sinusoidal reorientation, in the simplest case of linearly polarized light. The director polar reorientation angle $\theta$ may then be represented as a deformation of a "cord" (see the inset in Fig. 1), which has fixed ends on the opposite walls of the cell, $\theta(z) = \theta_0 \sin(\pi z/L)$, where $z$ is parallel to the initial NLC director orientation $\vec{n}_0$, $L$ is the thickness of the cell, and $\theta_0$ is the amplitude of the polar angular perturbation. This reorientation contains also a twist (three-dimensional) rotation component in the case of elliptical polarized light [8], but we will confine our discussion to very weak and in-plane reorientation. The input excitation light (elliptically polarized, in general case) may be decomposed into parallel and perpendicular components relative to the plane of the reoriented director. An additional nonlinear phase shift [2], experienced by the extraordinary (in-plane) light component, leads to the change in the ellipticity of the excitation beam. As noted above, the optimal angular momentum transfer condition is met when the NLC film acts as a half-wave plate for the light, that is, when the relative nonlinear phase shift is a multiple of $\pi$. In this case, each circular polarized photon should get out of the NLC cell with inverted sign of its circularity, transferring to the quasimacroscopic collective of molecules the amount of $2\hbar$ of the angular momentum. This transfer forces the "cord" of reoriented molecules to turn around the $z$ axis. However, the total amount of the transferred angular momentum $M$ and the corresponding molecular rotation cannot be effectively controlled by the photon number $N$, since the latter determines the change of $\theta_0$, and therefore it may break the optimal transfer (i.e., half-wave plate) condition. The attempts to control the input angular momentum by introducing *coherent* photons with opposite circular polarization lead to the formation of additional ellipticity $\gamma$ of the light. This breaks down the initial azimuthal symmetry condition, creating a preferred reorientation direction and resulting in suppression of the regular precession regime. Hence, this possibility is also limited.

We used in our work an electromagnetic field composed of two *copropagating* and *noncoherent* beams of orthogonal circular polarization, to induce the dielectric torque. The intensity ratio of these beams becomes an efficient control parameter for the input light angular momentum $M$ and for its transfer to the NLC. At the same time both the total photon number $N$ and the required azimuthal symmetry of the electromagnetic field may be kept constant. The proposed experimental setup is sketched in Fig. 1. The beam $E_i$ of linear polarized argon ion laser (operating at 514 nm) is divided in two separate arms by a half-wave plate $\lambda_1/2$ (for 514 nm) and the polarization beam splitter ($PBS_1$). The deviated beam is returned to the initial optical axis after reflection from the mirrors $M_1$ and $M_2$, and the simple beam splitter (BS). The optical path difference of these two arms exceeds the coherence length of the laser. Hence, the obtained optical field is a mixture of two copropagating noncoherent beams of orthogonal polarization. The intensity ratio $R = I_1/I_2$ of these two beams is easily controlled by rotating the half-wave plate $\lambda_1/2$. To obtain an optical

field composed of two beams with orthogonal noncoherent circular polarization, a quarter-wave plate $\lambda_1/4$ (for 514 nm) is placed in the optical path of the recombined beam. The latter is then focused by the lens $L_1$ (with focal length $f_1 = 13$ cm) at the homeotropic liquid crystal cell C (with thickness $L = 90$ $\mu$m) at normal incidence. The spot diameter at the NLC film is 80 $\mu$m. The NLC used is E7 from Merck Ltd. The weak linearly polarized probe beam $E_s$ of the He-Ne laser is counterpropagating with respect to the excitation beams, and is focused in the director perturbation area by the lens $L_2$ (with focal length $f_1 = 10$ cm). The probe beam is then collimated by the lens $L_1$ and is reflected from the second polarization beam splitter ($PBS_2$) away from the principal axis. The polarization plane of the probe beam is rotated by means of the second half-wave plate $\lambda_2/2$ (for the 632.8 nm). The noise or reflections of the excitation beams are cut off by the red filter (RF). The polarization state of the output probe beam is analyzed by means of the polarization beam splitter ($PBS_2$) and the detector (D). The period of the output probe beam intensity modulation (resulted from its polarization dynamical change) is detected for different fixed orientations of $\lambda_2/2$ plate. In the case of stronger excitations, the induced nonlinear phase retardation is observed at the screen by the aberration ring pattern in the transmitted excitation light. These rings are generated due to the light-induced spatially nonuniform molecular reorientation, and their number (multiplied by $2\pi$) represents roughly the nonlinear phase retardation in the center of the beam with respect to the edges [2,5].

When the principal optical axis of the quarter-wave plate $\lambda_1/4$ is chosen so that it does not change the polarization states of the two excitation beams, the stable reorientation regime with two linear cross-polarized noncoherent copropagating beams takes place. Optical-induced Fréedericksz transition is observed above a certain threshold of the total intensity, since the initial director orientation is normal to both incident linear polarized beams. The threshold value depends strongly on $R$ (Fig. 2). This is due to the continuous change of the azimuthal symmetry of the excitation field, in agreement with the formula (1) [2]. The threshold value at $R = 1$ (circular symmetry) is twice as high as the one corresponding to $R = 0$ (single linear polarized beam). The dependence of the threshold intensity on $R$ could be represented as $I_{\text{th}}(R) = 2I_{\text{th}}^{L}/[1 + \cos(\pi R/2)]$ [2]. The fit of the experimental data with this formula gives rather good agreement (Fig. 2). The NLC director orientation exhibits in this geometry a strong optical switching and a hysteresis dependence on the total input power. The hysteresis width decreases as the intensities of the two beams become unequal ($R \neq 1$) [9]. No dynamic oscillation of the probe output polarization was observed in this interaction geometry.

In the case of two orthogonal circular polarizations incident on the cell (optical axis of $\lambda_1/4$ plate being oriented at 45° with respect to the incident linear polarization), the behavior of the system changes principally due to the nonvanishing angular momentum carried by the light, and its transfer from the light to the NLC molecules.

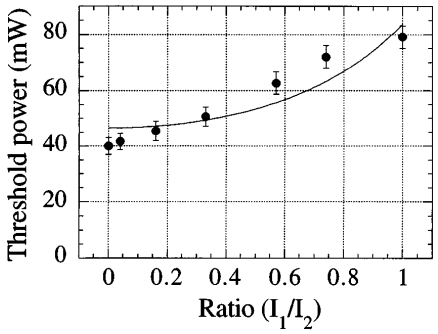


FIG. 2. Dependence of the threshold power for the light-induced Fréedericksz transition on the intensity ratio $R = I_1/I_2$ of two linearly polarized noncoherent copropagating beams (the spot diameter at the film plane is 80 $\mu$m). The line is a theoretical fit by the formula $I_{\text{th}}(R) = 2I_{\text{th}}^{L}/[1 + \cos(\pi R/2)]$.

When the two circularly polarized beams are of equal intensities ($R = 1$), an abrupt hysteresis dependence of the output ring pattern is observed. This is similar to the effect considered before for orthogonal linear polarizations with $R = 1$ [9]. Note that the symmetry of the excitation light (circular) and the carried angular momentum ($M = 0$) are the same in both cases.

As the angular momentum symmetry is broken (i.e., the intensity ratio $R \neq 1$, and hence the excitation light carries a nonvanishing angular momentum $M \neq 0$), the periodic rotation of the NLC director around the $z$ axis occurs. The effect is detected as temporal oscillations of the probe beam intensity at the output of the polarization beam splitter ($PBS_2$) (see the inset in Fig. 3). These oscillations are also observed using a similar analysis of the excitation beam (experimental setup not shown). The polarization plane of the input probe beam is rotated by means of the second half-wave plate $\lambda_2/2$ (Fig. 1) during the stabilized excitation regime (keeping the excitation beams and the $\lambda_1/4$ plate orientation unchanged). We observed periodic changes of the output probe polarization state for different fixed positions of input polarization plane, which confirms the molecular precession, but not just an in-plane perturbation. A heterodyne technique had been used in the same experimental conditions for more precise measurement of this precession amplitude and phase, but the detected precession had an almost constant rate [3], as mentioned above. Like the case of $R = 0$ (excitation by means of single circularly polarized beam [3,8]), the precession in this geometry begins at a certain threshold value (precession switch-on). The precession regime remains at lower power values until another power threshold (precession switch-off), below which the system returns to its undisturbed state.

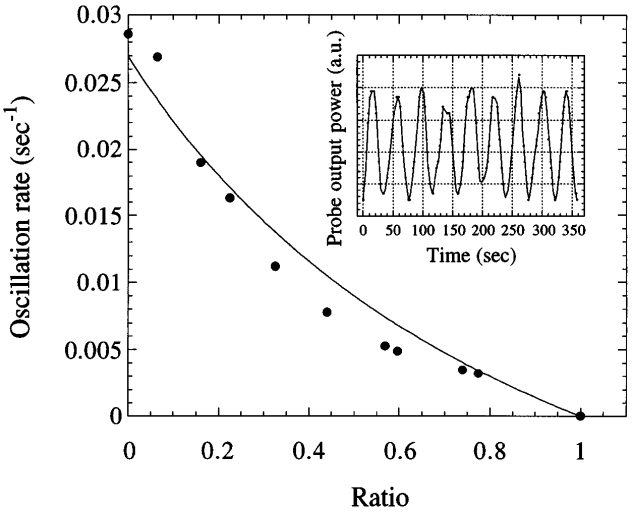


FIG. 3. Molecular precession rate (inverse period) versus $R = I_1/I_2$ for a total input intensity of $I = 1.65$ kW/cm$^2$. The line is a theoretical fit by the formula $\Omega = \text{const} \times (1 - R)/(1 + R)$. Inset: Demonstration of output probe beam intensity periodic changes due to nonlinear modulation of its polarization state.

Far above the precession switch-on threshold, where a stronger director perturbation—and therefore azimuthal symmetry break—is expected [8], the two processes of director precession and in-plane reorientation are coupled. In this intensity range the precession of the director due to angular momentum transfer may be accompanied by its in-plane oscillation due to dynamic energy exchange between polarization components [8,10]. This was observed in the present experiment as an oscillation of the number of rings in the diffraction pattern, and as a rotation of the polarization ellipse in the central part of the beam.

Above the precession switch-on threshold, a strong dependence of the precession period (at fixed total input power) on $R$ is observed. This measurement was carried out by means of the rotation of the half-wave plate $\lambda_1/2$, which allows the continuous redistribution of the number of photons carrying positive and negative angular momentum, while keeping unchanged their total number and the azimuthal symmetry of the field. Inverted oscillation period versus $R$ is plotted in Fig. 3 for total input intensity $I = 1.65$ kW/cm$^2$. The oscillations are damped as the proportion of photons of opposite circularity in the beam rises. The inverted period tends to zero while approaching $R = 1$. The precession angular speed ($\Omega$) dependence upon input light angular momentum was theoretically established in Ref. [8]. Following that work we can write $\Omega = DI(I_R - I_L)/(I_R + I_L)$, where $D$ depends on the total nonlinear phase difference between extraordinary and ordinary wave at the exit face of the sample (defined by reorientation amplitude $\theta_0$ and several fixed material parameters), $I$ is the dimensionless intensity (normalized to the threshold intensity), $I_R$ and $I_L$ are input right and left circular polarization intensities, respectively. For fixed $D$ and $I$ (as it is in our experiment) the dependence $\Omega(R)$ becomes $\Omega = DI(1 - R)/(1 + R)$, which fits very well the obtained experimental data (Fig. 3) with $DI = 0.269$ ($10^{-1}$s$^{-1}$) [9].

In conclusion, we have used the spatial separation of two polarizations of the light with their further recombination, to obtain two copropagating noncoherent beams forming an electromagnetic field with varied angular momentum, keeping at the same time its azimuthal symmetry. This allows the effective all-optical control of the light angular momentum transfer to the quasimacroscopic ensemble of the precessing molecules as well as the modulation of their rotation rate. We have confirmed the earlier predicted dependence of the light-induced phase transition threshold on the azimuthal symmetry of the light and, in particular, the doubling of this threshold value in the case of the light with circular symmetry and vanishing angular momentum (cf. [11]).

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