Collective Rotation of Molecules Driven by the Angular Momentum of Light in a Nematic Film

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It is experimentally demonstrated that a circularly polarized laser beam normally incident on a homeotropically aligned nematic film can induce a collective precession of the molecules in the film if the laser intensity is above the threshold for the Fréedericksz transition. The effect is shown to result from a transfer of angular momentum from the laser beam to the medium.

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We report here an interesting new nonlinear optical phenomenon—self-induced time-dependent polarization rotation in a liquid-crystal medium. It occurs as a result of angular momentum transfer from the radiation field to the medium, causing the molecules to precess around the beam propagation direction. An elliptically polarized light beam of frequency ω, intensity I, and ellipticity $S_3=(I_+-I_-)/I$ carries an average angular momentum of $-\Phi\hbar S_3$ per unit time and unit area, where $\Phi=I/\hbar\omega$ is the photon flux and I_+ and I_- are the intensities of the right- and left-circularly polarized components of the beam, respectively.[1] The beam angular momentum can be transferred to a transparent medium if the latter is anisotropic, as some photons must emerge with their spin components reversed. The continuous transfer of angular momentum results in a torque on the medium. The torque is usually very small and is insufficient to put a macroscopic body into continuous rotation.[2] Its effect on the molecules in a liquid crystalline medium is, however, easily observable. In this Letter, we present the first observation of such an effect in a thin nematic film of 4-cyano-4'-pentyl-biphenyl (5CB). The rotation of molecules is manifested by the rotation of the polarization ellipse of the output beam. The process can also be recognized as a type of stimulated scattering. Our experimental results are in good agreement with the theoretical predictions based on angular momentum conservation.

The experimental arrangement is shown in Fig. 1. A circularly polarized argon-laser beam ($\lambda=0.515\ \mu$m) propagating along the z axis was focused at normal incidence on a homeotropically aligned nematic 5CB sample 65 μm thick by use of a lens of 10-cm focal length. The polarization state of the light beam emerging from the sample was monitored with use of a heterodyne polarimeter scheme.[3] As shown in Fig. 1, a fraction of the incoming laser beam was split off and frequency shifted by an acoustic-optic modulator driven at 10 MHz and used as the reference beam. This reference beam, linearly polarized, was then divided by a beam splitter followed by two crossed polarizers into a horizontal (H) and a vertical (V) component of equal intensities. The signal beam, after transmission through the sample, was elliptically polarized in general, and was also divided into H and V components by the same beam-splitter polarizers. The H and V components of both beams fell separately on two photodiodes and generated a heterodyne beat signal at 10 MHz from each of them. The two beat signals from the photodiodes were then sent to the x and y terminals of a sampling oscilloscope for visual display of the output polarization ellipse, or to an IBM-PC computer for data

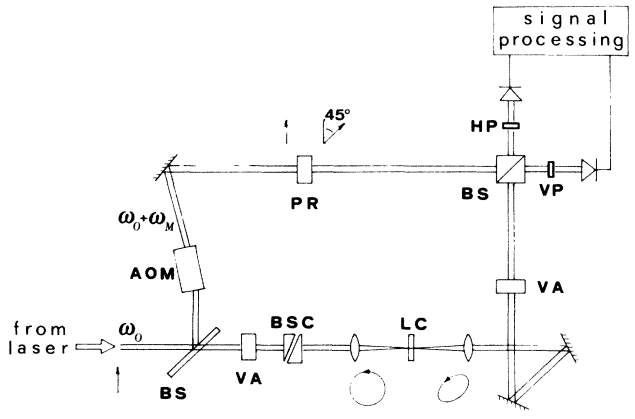

FIG. 1. The experimental arrangement: BS, beam splitter; VA, variable attenuator; BSC, Babinet-Soleil compensator; LC, liquid crystal; AOM, acousto-optic modulator; PR, polarization rotator; P, Glan prism polarizer.

processing. A Babinet-Soleil compensator and a polarization rotator were used for calibration of the apparatus.

Below a threshold pump power intensity I_{th}, no effect was observed and the transmitted light remained circularly polarized. Above the threshold, however, it became elliptically polarized and the polarization ellipse rotated continuously, as could be seen on the oscilloscope screen. The period of the ellipse rotation ranged from 40 to 50 s. This optical-field–induced transition appeared to be first order. As the laser intensity decreased from $I > I_{th}$ to $I < I_{th}$, the ellipticity of the output beam varied, but switched back to circular polarization only when I was significantly below I_{th}. Figure 2 describes the ellipticity change Δs_3 of the beam in traversing the sample as a function of the normalized input laser intensity. The hysteresis loop *ABCD* characterizes the first-order transition. This is a clear demonstration of an intrinsic (mirrorless) optical bistability effect in the absence of any static field.

With the input intensity changing slowly, the hysteresis loop is well reproducible. The upward transition *BC* in Fig. 2 is much faster than the downward transition *DA*: The switching times are $t_{up} \simeq 1$ s and $t_{down} \simeq 15$ s, respectively. The observed threshold power for up transition at point *B* was $P_{th} = 110$ mW. With a beam cross section of 5×10^{-5} cm^2 at the sample, the corresponding threshold intensity was $I_{th} = 2.2$ kW/cm^2. This transition should result from the field-induced molecular reorientation usually known as the Fréedericksz transition. We found that the observed threshold for a circularly polarized input beam was twice that for a linearly polarized input beam, as expected from theory.[4] However, no polarization rotation of the output was observed in the latter case.

All data points in Fig. 2 refer to an output whose polarization ellipse rotated continuously with a nearly constant angular velocity and no appreciable change in its form.[5] The branch *CD* is stable and reversible. No diffraction ring was observed in the far field except for a weak halo. This indicates that the birefringence induced in the sample is so small that the phase difference between the ordinary and extraordinary waves accumulated in transversing the medium is always less than π. When the input beam was changed from left to right circular polarization, the direction of the ellipse rotation was reversed accordingly, but the speed of the rotation remained unchanged.

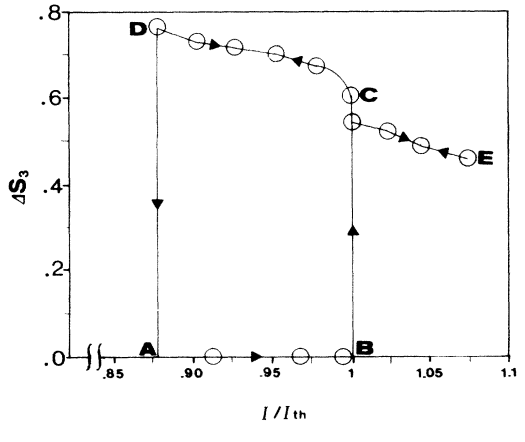

FIG. 2. Ellipticity change Δs_3 of the laser beam traversing the sample as a function of the normalized light intensity. The branch *CD* is characterized by a stable uniform rotation of the output polarization ellipse.

The branch *CE* is also reversible, but is metastable, in the sense that after a certain time (ranging from 10 to 60 min) the system switches spontaneously in 1–10 s to a different regime, characterized by a continuous variation of the form of the output polarization ellipse as well as by the appearance of a large number (up to 10) of diffraction rings in the far field. The number of rings is an indication of the magnitude of the induced birefringence in the sample.[6] During switching, the output polarization ellipse was continuously changing in form, until a steady-state regime was reached where the ellipse rotated again uniformly but with a much longer period of about 12 min. This slow-rotation regime could be reached only if the laser power is varied sufficiently slowly, $(\partial/\partial t) I/I_{th} < 2.5 \times 10^{-3}$/min. Otherwise, a dynamic regime would set in with the diffraction rings undergoing oscillating motion of expansion and contraction. If the laser intensity was beyond *E* in Fig. 2, then the system switched directly to the dynamic regime without going through any metastable or steady-state regime.

As the results for $I > I_B$ in Fig. 2 are not yet well understood, we shall, in this paper, focus our attention on the results for $I < I_B$. The fact that the ellipse rotation occurred with a circular rather than linear input polarization suggests that the effect arises from angular momentum transfer between the radiation field and the medium. While details of the theory are yet to be worked out, we present here a simple calculation that can explain the main observations.

Let us first give a qualitative description. The molecular reorientation of the homeotropic alignment is initiated only if the incoming laser intensity is above the threshold for the Fréedericksz transition. With circularly polarized input, the director $\hat{\mathbf{n}}$ would tend to be reoriented in such a way that it is tilted away from the $\hat{\mathbf{z}}$ axis but random in the azimuthal plane. However, in order to keep the elastic energy low, $\hat{\mathbf{n}}$ is likely to be tilted along a single direction. Some residual anisotropy would define that direction. The medium then appears birefringent to the incoming beam and, consequently, the beam polarization becomes elliptical. Transfer of angular momentum from the beam to the medium causes $\hat{\mathbf{n}}$ to precess around $\hat{\mathbf{z}}$, and hence the rotation of the polarization ellipse following $\hat{\mathbf{n}}$. With the beam becoming elliptically polarized, the threshold for the Fréedericksz transition is reduced. Therefore, even if the incoming I is now decreased to a value below I_{th} for circularly polarized light, the director $\hat{\mathbf{n}}$ may still point away from $\hat{\mathbf{z}}$ and precess around $\hat{\mathbf{z}}$ until

the elliptically polarized light in the medium can no longer sustain the molecular reorientation, leading to the downward transition *DA* in Fig. 2. If $I > I_{th}$, then tranformation of the beam polarization from circular to elliptical tends to tilt $\hat{\mathbf{n}}(z)$ farther away from $\hat{\mathbf{z}}$ and enhances the birefringence.[7] As the birefringence increases, the beam polarization changes accordingly, and in turn, modifies $\hat{\mathbf{n}}$ again. Because the reorientation of $\hat{\mathbf{n}}$ responds rather slowly to the change of beam polarization, this could lead to a libration of $\hat{\mathbf{n}}$ in its polar angle, and hence an oscillation in the observed birefringence.

We now consider some quantitative features of the problem. The threshold I_{th} for the Fréedericksz transition with circularly polarized light is given by

$$I_{th}=2(\pi/L)^2(ck_{33}/n_o)[n_e^2/(n_e^2-n_o^2)], \quad (1)$$

where L is the thickness of the nematic film, k_{33} is the bend elastic constant, and n_0 and n_e are the ordinary and extraordinary refractive indices of the medium, respectively. With $L=65$ μm and taking $k_{33}=4.4\times10^{-7}$ dyne, $n_o=1.53$, and $n_e=1.70$ for 5CB,[8] we find $I_{th}=2.1$ kW/cm^2, which is in good agreement with the observed value.

The angular momentum per unit area lost by the beam to the medium in traversing the nematic film is $(I/\omega)\Delta s_3$, where $\Delta s_3=s_3(L)-s_3(0)$ is the ellipticity change of the beam polarization. This angular momentum transfer appears as a dynamic torque on the medium, causing the director $\hat{\mathbf{n}}$ to precess, and is balanced by the torque arising from the viscous force:

$$I\Delta s_3/\omega=\gamma_1\int_0^L \sin^2\theta\frac{\partial\phi}{\partial t}dz, \quad (2)$$

where γ_1 is a viscosity coefficient[9] and θ and ϕ are the polar and azimuthal angles of $\hat{\mathbf{n}}(z,t)$, respectively. We have neglected in Eq. (2) the inertia term and the backflow effect.

We now assume, for the *CD* section in Fig. 2, that $\partial\phi/\partial z=0$, $\partial\theta/\partial t=0$, and $\partial\phi/\partial t=\Omega$ which describes the constant rotation of the director and the output polarization ellipse. Equation (2) then yields

$$\Omega=I\Delta s_3/(\omega\gamma_1\int_0^L \sin^2\theta dz). \quad (3)$$

We recognize, however, that the phase difference between the ordinary and extraordinary waves at $z=L$ is

$$\chi=\pm\pi/2+(\omega/c)\int_0^L[n_o-n_on_e/(n_e^2\cos^2\theta+n_o^2\sin^2\theta)^{1/2}]dz\simeq\pm\pi/2-(\omega n_o/2c)(1-n_o^2/n_e^2)\int_0^L\sin^2\theta(z)dz, \quad (4)$$

where $\pm\pi/2$ refers to the phase difference at $z=0$ for the right- and left-circularly polarized waves, respectively, and the approximation is valid for small θ. Equation (4) indicates that the output at $z=L$ is elliptically polarized. Since $s_3(L)=\sin\chi$ and $s_3(0)=\pm1$, we find, by eliminating $\int_0^L\sin^2\theta dz$ in Eq. (3) using Eq. (4),

$$\Omega=\pm\frac{I}{I_{th}}\left(\frac{\pi}{L}\right)^2\frac{k_{33}}{\gamma_1}\frac{|\Delta s_3|}{\sin^{-1}[(|\Delta s_3|/2)^{1/2}]}. \quad (5)$$

It is seen that Ω changes sign when the circular polarization of the input beam is reversed, as observed in the experiment.

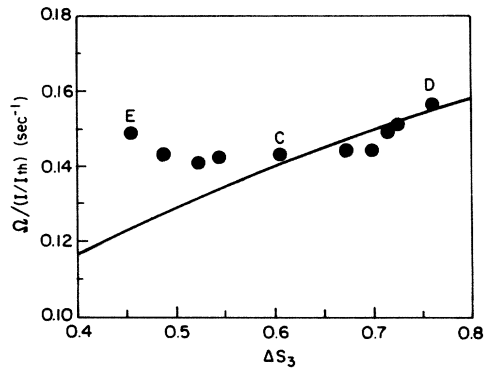

FIG. 3. Angular velocity Ω (normalized against I/I_{th}) of the output polarization ellipse as a function of the ellipticity change Δs_3 across the sample. The continuous line is obtained from Eq. (5).

From the measured I/I_{th} and Δs_3 given in Fig. 2 and $\gamma_1=0.38$ P taken from the literature,[8] we can find Ω from Eq. (5). The results are shown in Fig. 3 in comparison with the measured Ω. Note that the agreement is fairly good for the *CD* branch, where the approximations we used in the derivation are reasonable. For the metastable *CE* branch, the approximations are no longer valid, and hence the agreement is poor. The transient dynamic behavior of the problem is generally more complex. We are still in the process of finding a satisfying solution to explain all our observations quantitatively.

We also realize that the induced molecular precession requires a deposition of the beam energy in the medium. Since the medium is transparent, this can only happen if part of the beam is downward shifted in frequency. Indeed, the rotation of the polarization ellipse means that the two circularly polarized components of the elliptical polarization have different frequencies ω and ω', respectively, with $\omega-\omega'=2\Omega$. Using a heterodyne technique, we were able to measure directly the ω' component in the output. In this respect, we can also regard the present nonlinear optical effect as a stimulated light-scattering process in which a new frequency component at ω' is generated. Details of our experiment and theoretical description will be reported elsewhere.

In summary, we have demonstrated that via the optical Fréedericksz transition, a circularly polarized input beam can activate a precession of the director in a homeotropic nematic film by transferring part of its angular momentum to the medium. This yields an output beam with a

continuously rotating polarization ellipse. The phenomenon is intrinsically bistable. At input intensities above the transition threshold, the induced birefringence in the medium may break into oscillation. The observations can be understood physically, but the details are yet to be worked out.

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[1]See, for example, J. M. Jauch and F. Rohrlich, *The Theory of Photons and Electrons* (Addison-Wesley, Cambridge, 1955).

[2]Nevertheless, this torque has been measured in static conditions with a sophisticated torsional balance by R. A. Beth, Phys. Rev. **50**, 115 (1936).

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[5]A small ($\sim 3\%$) oscillation of the output polarization ellipticity was always present. This effect is due to an imperfect, rigid rotation of the liquid-crystal molecules that has been neglected in our simplified mode. The values of Δs_3 reported in Figs. 2 and 3 are obtained by averaging over this oscillation.

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[7]The same mechanism is responsible for the sharp switchon of *BC* in Fig. 2.

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[9]See, for example, P. G. de Gennes, *The Physics of Liquid Crystals* (Oxford Univ. Press, Oxford, 1974), pp. 156–158.