Observation of Magnetic-Field–Induced First-Order Optical Fréedericksz Transition in a Nematic Film

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It is experimentally demonstrated that an applied dc magnetic field can transform the optical Fréedericksz transition in a nematic film from second order to first order. The results are in good agreement with theoretical predictions.

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An optical field can play the equivalent role of a dc field in inducing a phase-transition-like rearrangement of molecular orientations in a liquid-crystal film. Such an optical-field–induced Fréedericksz transition has been observed in a homeotropically aligned nematic cell with a cw laser beam.[1] The transition is usually of second order. However, it has been predicted that if the medium has sufficiently large optical anisotropy the transition can become first order.[2] The orientational rearrangement as a function of the laser intensity is then characterized by a hysteresis loop. Responding to the orientational variation, the output beam from the cell relative to the input should display an optically bistable behavior. Mirrorless, intrinsic optical bistability is presently a subject of great interest to many researchers.[3]

Unfortunately, none of the common nematic liquid crystals seems to possess enough anisotropy to exhibit a first-order optical Fréedericksz transition.[4] It was then recognized by several authors[5] that an applied dc field along a prescribed direction can enhance the effective elasticity and convert the second-order Fréedericksz transition to first order. In this paper, we report the first observation of such a first-order optical Fréedericksz transition with the help of a dc magnetic field.[6]

Let us first give a summary of the theory.[2,5] We consider here a linearly polarized laser beam normally incident onto a homeotropically aligned nematic film (director or direction of molecular alignment along the surface normal). In addition, a dc magnetic field is also applied along the surface normal. The nematic substance is assumed to have positive anisotropies, namely, $n_{\parallel} > n_{\perp}$ and $\chi_{\parallel} > \chi_{\perp}$, where n and χ denote the refractive indices and magnetic susceptibilities, and refer to directions parallel and perpendicular to the director, respectively. A strong enough optical field can distort the homeotropic molecular alignment. The reorientation angle θ of the director as a function of the distance into the film, z, is expected to be symmetric about $z = d/2$, where d is the film thickness. At $z = d/2$, $\theta = \theta_m$ is a maximum. The total free energy F of the system in the presence of the applied optical and dc magnetic fields can then be expressed in terms of θ_m. Because of the equivalence of θ and $-\theta$ in the present case, F must be an even function of θ_m.

We can treat θ_m^2 as an order parameter, using Landau's theory to describe the anticipated Fréedericksz transition. Expanding F into a power series in θ_m^2, we have[2,5]

$$F = -C\theta_m^2 + B\theta_m^4/2 + G\theta_m^6/3 + \ldots, \quad (1)$$

where the coefficients are found to be

$$C = (I_{\rm th}/I_0)[(I/I_{\rm th})^{1/2} - 1], \quad B = (1 - k - 9u/4)/4 - 9uh^2/16,$$

$$G = \tfrac{1}{96}[(\tfrac{11}{2} - k + 9u/4 + 63ku/4 - 9k^2/2 - 261u^2/32) + (9u/4 + 63ku/4 + 639u^2/16)h^2 + 1539u^2h^4/32],$$

with

$$k = 1 - k_{11}/k_{33}, \quad u = 1 - n_{\perp}^2/n_{\parallel}^2, \quad h = H/H_0, \quad H_0 = (\pi/d)[k_{33}/(\chi_{\parallel} - \chi_{\perp})]^{1/2},$$

$$I_0 = (\pi/d)^2(ck_{33}/un_{\perp}), \quad I_{\rm th} = I_0(1 + h^2).$$

The notations I and H denote the laser intensity and the dc magnetic field, respectively. Analysis of F in Eq. (1) shows that if $B > 0$, then a second-order transition occurs at $C = 0$ or $I = I_{\rm th}$, but if $B < 0$ and $G > 0$, a first-order transition can occur. The upward transition, determined by $C = 0$ and $\partial F/\partial(\theta_m^2) = 0$, happens at $I = I_{\rm th}$ with θ_m

switched from 0 to $(-B/G)^{1/2}$. The downward transition, determined from $\partial F/\partial(\theta_m^2)=\partial^2 F/\partial(\theta_m^2)^2=0$, happens at

$$I=I'_{\rm th}=I_{\rm th}[1-(I_0/I_{\rm th})(B^2/4G)]^2$$

with θ_m switched from $(-B/2G)^{1/2}$ to 0. Conversion from the second-order transition occurs at $B=0$, from which one obtains the tricritical field $H_{\rm TC}=[4(1-k)/9u-1]^{1/2}H_0$. Using the known values of material parameters for some common nematic liquid crystals, we find $H_{\rm TC}$ around a few hundred oersteds and $I_{\rm th}$ around a few hundred watts per square centimeter for d around a few hundred microns. Thus, the theory can be easily tested by experiment.

In our experiment, we used a homeotropically aligned nematic film of 4-cyano-4'-pentylbiphenyl (5CB) as the sample. The film was sandwiched between two glass plates coated with dimethyl-*n*-octadecyl-3-aminopropyl methoxysilyl chloride (DMOAP). The thickness of the film was 380 μm. The sample cell was regulated to 25.0 ± 0.1 °C, and was placed horizontally to minimize possible laser-induced convection. A cw Ar^+ laser beam, linearly polarized at 514.5 nm with a diameter of 0.9 mm, was normally incident on the cell. Simultaneously, a dc magnetic field was applied along the surface normal of the cell. The laser-induced molecular reorientation resulted in an optical birefringence that was measured by two techniques. In the first case, a weak He-Ne laser beam focused to the center of the pumped region in the cell was used to probe the birefringence. With the help of an iris, the effective diameter of the probe beam was ~ 10 μm. In the second case, we utilized the fact that the spatial self-phase-modulation of the pump laser beam resulting from the laser-induced birefringence leads to a multiple ring structure in the far-field pattern.[1,7] The number of rings is directly related to the maximum birefringence induced in the medium. This is quite accurate if the number of rings is large. The two techniques gave consistent results to within a 2π phase retardation for reasonable alignment of the probe beam.

The observed results are shown in Fig. 1. The experiment was conducted by our first fixing the magnetic field H at a certain value and then measuring the phase retardation $\Delta\phi$ due to the induced birefringence as a function of the laser intensity I. The latter was varied in small steps, with enough time between steps to assure static equilibrium before the measurement was taken. The random variation in the laser power is $\sim 1\%$. It is seen in Fig. 1 that at low H, the results of $\Delta\phi$ vs I behave like those of second-order optical Fréedericksz transitions reported previously,[1] but for sufficiently large H (>250 Oe) the data exhibit a hysteresis loop characteristic of a first-order transition.[2,5]

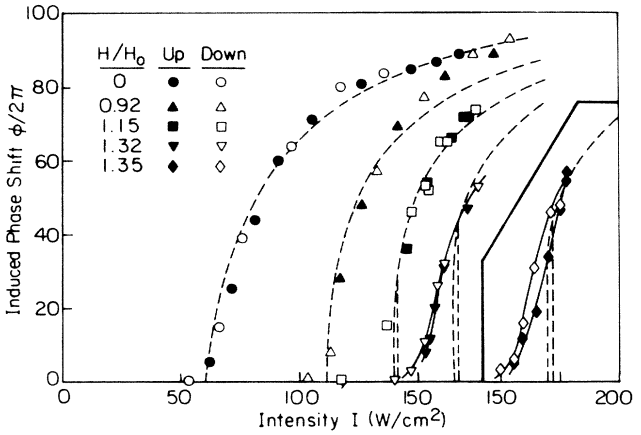

FIG. 1. Phase retardation $\phi/2\pi$ vs intensity for fixed magnetic field strengths. Open symbols were measured with increasing intensity; solid symbols, with decreasing intensity. Solid lines were drawn to aid visualization of the data. Broken lines are the theoretical fits with $k_{11}=9.00\times10^{-7}$ dyn, $k_{33}=9.51\times10^{-7}$ dyn, $n_\perp=1.54$, $n_\parallel=1.73$, $\lambda=514.5$ nm, and $d=380$ μm. The H/H_0 values are determined from the fit for each curve. The experimental H/H_0 values are 0, 0.92, 1.13, 1.20, and 1.26.

To compare the experimental results with theory, we first obtain $k_{33}=(1\pm0.1)\times10^{-6}$ dyn from the observed $I_{\rm th}=61\pm7$ W/cm^2 at $H=0$ using the expression of $I_{\rm th}$ in Eq. (1) with $n_\perp=1.54$, $n_\parallel=1.73$ for 5CB and $d=380$ μm. The induced phase shift versus pump intensity for the $H=0$ case can then be calculated by the formalism described in Refs. 1 or 2 with $k_{11}/k_{33}=0.83$ obtained from Skarp, Lagerwall, and Stebler.[8] As seen in Fig. 1, the theory fits the data fairly well. The discrepancy presumably arises from the effect of finite beam size or a small misalignment of the pump beam from normal incidence. We then compare the observed $I_{\rm th}$ (obtained by extrapolation from the maximum slope of the data points) versus H with the theoretical expression of $I_{\rm th}$ in Eq. (1). As shown in Fig. 2 (inset), the experimental result agrees well with the predicted $1+H^2/H_0^2$ dependence. This allows us to deduce a value of $H_0=235\pm10$ Oe and hence $\chi_\parallel-\chi_\perp=(1.2\pm0.1)\times10^{-7}$ cgs units. The latter is compared with the value 1.76×10^{-7} cgs units in the literature.[9] Again, the discrepancy is presumably due to simplification in our calculation. We also compare the widths of the observed hysteresis loops for $H>H_{\rm TC}$ with the predicted values of $I_{\rm th}-I'_{\rm th}$, shown in Fig. 2. The agreement is fairly good. In particular, the tricritical field $H_{\rm TC}\cong210$ Oe calculated from the values of k, u, and H_0 is certainly consistent with the observation.

Given $I_{\rm th}(H)$, the induced phase shift versus I at various H can be likewise calculated.[5] For a better fit of the data we used $I_{\rm th}(H)$ as adjustable parameters. The results are also presented in Fig. 1. It is seen that

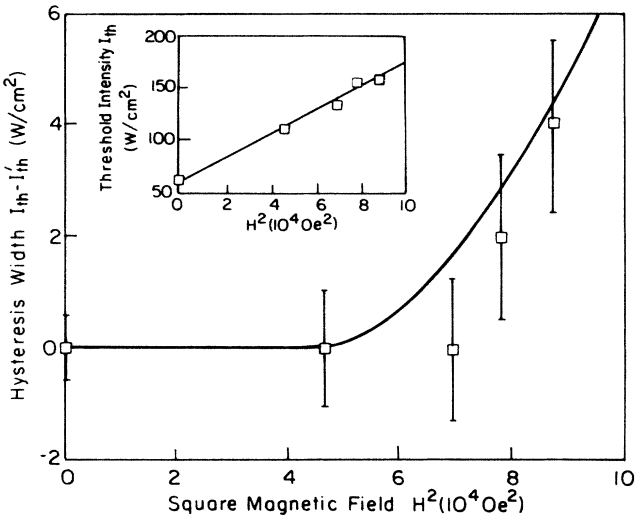

FIG. 2. Hysteresis width $I_{th}-I'_{th}$ vs squared magnetic field H^2. Thresholds are obtained by extrapolation from the maximum slope of the data in Fig. 1. The curve is the theoretical prediction using parameters given in the text. Inset: Threshold intensity I_{th} vs H^2, plotted with the theoretical line for $H_0=235$ Oe.

for $H<H_{TC}$, such that the Fréedericksz transition is second order, the theoretical calculation describes the data fairly well, including the threshold intensity $I_{th}(H)$. However, when the transition becomes first order for $H>H_{TC}$, the discrepancy between theory and experiment is rather appreciable. The theoretical $I_{th}(H)$ is significantly different from the observed $I_{th}(H)$. The most obvious difference lies in the fact that our theory for the first-order Fréedericksz transition, based on the free-energy calculation, predicts abrupt switching at I_{th} and I'_{th}, in contradiction to the experimental results. The observed smoothly varying hysteresis loops are characteristic of first-order transitions in a system with many domains. In our case, we believe that this may arise from the successive excitations of various transverse modes existing because of the finite pump beam size or the slight variation in the film thickness.[10] Such a "domain" effect also affects the average induced phase shift observed by the probe beam.

We have also measured the turnon and turnoff times of molecular reorientation induced by $I>I_{th}$. For a given value of H and I, the initial exponential rise of the induced birefringence in the sample was recorded, yielding the turnon time $\tau_{on}(H,I)$. Later the beam was blocked and the final exponential decay of the birefringence was recorded, yielding the turnoff time $t_{off}(H)$. In either case the measured birefringences corresponded to rather small molecular reorientation angles. Thus, small-angle approximations can be used in the theoretical analysis. Following the derivation of Ref. 2 with extension to include the magnetic field effect, we can find

$$\tau_{on}^{-1}=\left(\frac{k_{33}}{\eta}\right)\left(\frac{\pi}{d}\right)^2\left(1+\frac{H^2}{H_0^2}\right)\left(\frac{I}{I_{th}(H)}-1\right),$$

$$\tau_{off}^{-1}=(k_{33}/\eta)(\pi/d)^2(1+H^2/H_0^2).$$

Our experimental results indeed show the predicted dependence of τ_{on} and τ_{off} on H and I, as seen in Fig. 3.

We notice that if I_{th} vs H deduced from Fig. 3(a) is plotted, the relation $I_{th}\propto 1+H^2/H_0^2$ is still obeyed, but the value of H_0 is found to be 290 ± 10 Oe, which is different from the one deduced from Fig. 2(a). This is because different samples with somewhat different beam focusing were used; the theoretical derivation

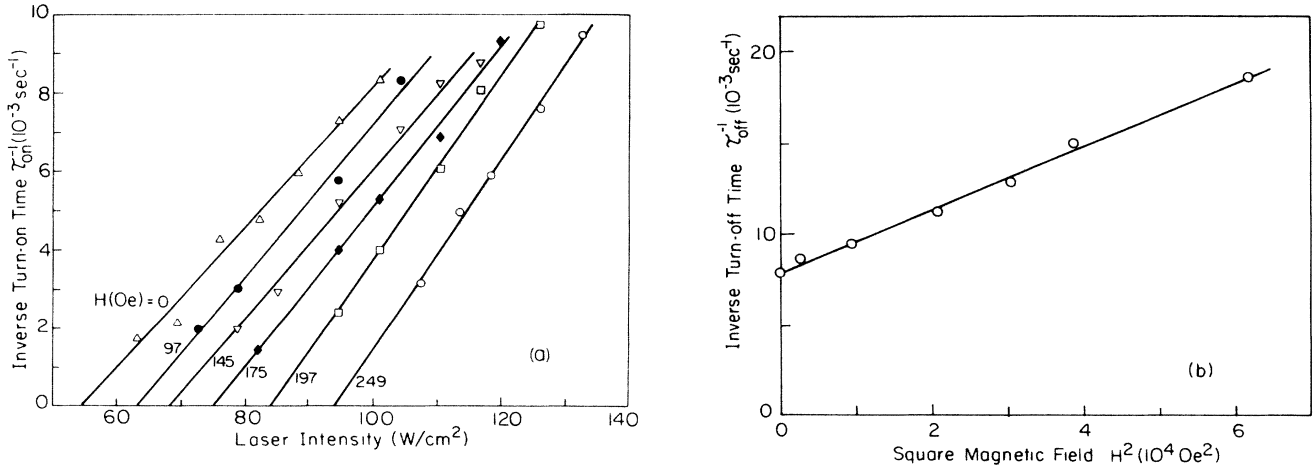

FIG. 3. (a) τ_{on}^{-1} vs laser intensity for various magnetic fields, showing the predicted divergence as the threshold intensity is approached. (b) τ_{off}^{-1} vs H^2 with the theoretical fit (solid line) using $H_0=215$ Oe.

outlined above is for a pump beam of infinite size, while a finite beam size should lead to effectively larger values of k_{33} and H_0.[11] Furthermore, τ_{on} is only measured at small tilt angles ($N < 2$), whereas steady-state values of I_{th} are extrapolated from large angles. The value of H_0 deduced from Fig. 3(a) is also different from the value $H_0 = 215 \pm 10$ Oe deduced from Fig. 3(b), even though the same sample and beam geometry were used in the measurements. This, we believe, is because the effect of finite beam size is generally weaker on τ_{off} than on τ_{on}.[11]

In summary, we have reported here the first observation of transformation of the optical Fréedericksz transition in a nematic film from first to second order. The results of both the static and dynamic measurements are in fair agreement with the theoretical predictions, except that the observed hysteresis loops are smoothly varying, indicating possible transverse mode structure.

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[1]S. D. Durbin, S. M. Arakelian, and Y. R. Shen, Phys. Rev. Lett. **47**, 1411 (1981); A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, N. N. Sobolev, and L. Chillag, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 170 (1980) [JETP Lett. **32**, 158 (1980)].

[2]H. L. Ong, Phys. Rev. A **28**, 2393 (1983); B. Ya. Zeldovich, N. V. Tabiryan, and Yu. S. Chilingaryan, Zh. Eksp. Teor. Fiz. **81**, 72 (1980) [Sov. Phys. JETP **54**, 32 (1981)]. In variance with the case of the dc Fréedericksz transition, the torque exerted on the molecules by the optical field is proportional to $(1 - u \sin^2\theta)^{-3/2} \sin\theta \cos\theta$ which increases as θ increases as long as θ is not too large. The $(1 - u \sin^2\theta)^{-3/2}$ term acts as a positive feedback in reorienting the molecules. For a sufficiently large optical anisotropy (u sufficiently small) in the presence of a sufficiently large splay elasticity, this positive feedback is so strong that it can give rise to a first-order transition. The optical torque can be effectively enhanced by the application of a dc bias field that increases the threshold intensity for the transition.

[3]J. A. Goldstone and E. Garmire, Phys. Rev. Lett. **53**, 910 (1984).

[4]H. L. Ong, M. Schadt, and I. F. Chang, Mol. Cryst. Liq. Cryst. **132**, 45 (1986).

[5]S. R. Nersisyan and N. V. Tabiryan, Opt. Spektrosk. **55**, 782 (1983) [Opt. Spectrosc. (USSR) **55**, 469 (1983)], and Mol. Cryst. Liq. Cryst. **116**, 111 (1984); H. L. Ong, Phys. Rev. A **31**, 3450 (1985), and **32**, 3148(E) (1985), and Appl. Phys. Lett. **46**, 822 (1985). An optical field can also convert the dc-induced Fréedericksz transition from second order to first order; see H. L. Ong, Phys. Rev. A **33**, 3550 (1986).

[6]Preliminary results were reported as a postdeadline paper by S. M. Arakelian, A. Karn, Y. R. Shen, and H. L. Ong, in *Proceedings of the 1985 Conference on Lasers and Electro-Optics (CLEO-85), Baltimore, Maryland, 21–24 May 1985* (Optical Society of America, Washington, D.C., 1985).

[7]S. D. Durbin, S. M. Arakelian, and Y. R. Shen, Opt. Lett. **6**, 411 (1981).

[8]K. Sharp, S. T. Lagerwall, and B. Stebler, Mol. Cryst. Liq. Cryst. **60**, 215 (1980).

[9]N. V. Madhusvdana and R. Pratibha, Mol. Cryst. Liq. Cryst. **89**, 249 (1980).

[10]Smooth transitions of dc-field–induced bistability were observed in a cholesteric twist cell with a thickness variation of 2%, according to D. W. Berreman, private communication.

[11]L. C. Csillag, I. Janossy, V. F. Kitaeva, N. Kroo, and N. N. Sobolev, Mol. Cryst. Liq. Cryst. **84**, 125 (1982); H. Hsiung, P. Shi, and Y. R. Shen, Phys. Rev. A **30**, 1453 (1984).