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Optical nonlinearity and bistability in liquid crystals

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Molecular reorientation and laser heating induced by an optical field can yield significant changes in the refractive indices in a nematic liquid crystal. A c.w. laser beam is intense enough to induce a phase retardation much larger than 2π in a nematic film less than 100 μm thick. Optical bistability in such a film sandwiched between mirrors can be readily observed. Coupling between the two mechanisms for induced refractive indices can lead to interesting results in the bistable operation.

Liquid crystalline materials are known to be composed of highly anisotropic molecules that can be easily reoriented by external fields (see, for example, Sheng 1975). This is particularly true for the mesophase owing to the very strong correlation among molecules. The situation is a close analogy to spins in a ferromagnetic phase. Typically, a d.c. field of $E \approx 100$ V cm^{-1} or $H \approx 0.1$ T is sufficient to induce a significant molecular reorientation and lead to a refractive index change, Δn, as large as 0.01 to 0.1. Since, for molecular reorientation, an optical field is equivalent to a d.c. field as long as there is no strong permanent dipole on the molecules (Herman & Serinko 1979), the same Δn can be induced by a laser intensity of *ca.* 100 W cm^{-2}, readily obtainable from a c.w. laser beam. It immediately suggests that liquid crystals should be an ideal medium for studies of highly nonlinear optical phenomena resulting from the optical-field-induced Δn.

An example is shown in figure 1 (Durbin *et al.* 1981), where we have presented both the experimental data and the theoretical calculation of the induced phase shift

$$\Delta\phi = \int_{-\frac{1}{2}d}^{\frac{1}{2}d} (\omega/c)\,\Delta n\,\mathrm{d}z$$

as a function of the laser intensity, I, in a thin film of homeotropically aligned 4-cyano-4′-pentylbiphenyl (5CB) of thickness $d = 250$ μm. Because of the strong anchoring force on the molecules at the boundary surfaces, Δn is not uniform across the film, but is nearly zero at $z \approx \pm\frac{1}{2}d$ and maximum around $z = 0$. The theoretical curves are calculated from the field-induced molecular reorientation, which is derived from minimization of the free energy of the system with appropriate boundary conditions. With a normally incident laser beam, the homeotropically aligned molecules cannot be reoriented unless the field intensity is above a certain threshold value. Such critical behaviour is known as the Freedericksz transition in liquid crystals (see Sheng 1975). As seen in figure 1, a small change in the laser intensity can induce a rather appreciable change of the phase shift in some region, especially near the Freedericksz transition. The operating point for differential operation can be selected by the application of a bias field, which can be either d.c. or optical because for molecular reorientation the two are equivalent. Thus, for example, if we use a d.c. magnetic field parallel to the liquid crystal

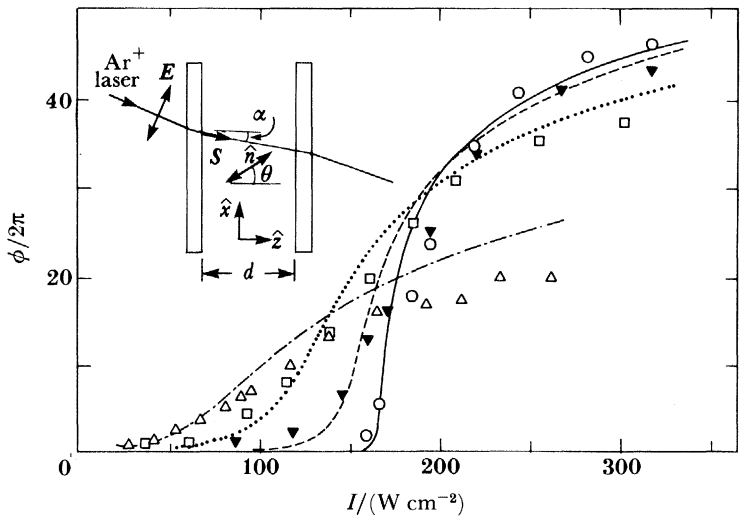

FIGURE 1. Experimental data and theoretical curves for the phase shift $\Delta\phi$ induced in a 250 μm homeotropically aligned 5CB film by an Ar^+ laser beam at different angles α: circles and solid curve, $\alpha = 0°$; solid triangles and broken curve, $\alpha = 3°$; squares and dotted curve, $\alpha = 11°$; open triangles and dot-dashed curve, $\alpha = 30°$. The inset shows the experimental geometry.

film to bias the operating point on the $\alpha = 0$ curve near the transition, then a laser intensity of *ca.* 1 W cm^{-2} will be able to induce a π phase shift. This means that a 1 mW He–Ne laser beam will be intense enough to induce optical bistability in such a film sandwiched between two mirrors.

In addition to molecular reorientation, laser heating also contributes to the change in refractive index. Although 5CB has no absorption band in the visible region, laser heating through residual absorption, presumably due to defects or impurities, can still be noticeable. A 350 W cm^{-2} Ar^+ laser can raise the local temperature of the medium by *ca.* 2 K. Figure 2 shows how the refractive indices of 5CB change with temperature (Chu *et al.* 1980). It is seen that even well into the nematic phase, we find values of $\Delta n_{\parallel} \approx -3.2 \times 10^{-3}\ K^{-1}$ (or $-2 \times 10^{-5}\ cm^2\ W^{-1}$) and $\Delta n_{\perp} \approx 7.6 \times 10^{-4}\ K^{-1}$ (or *ca.* $4 \times 10^{-6}\ cm^2\ W^{-1}$). The corresponding

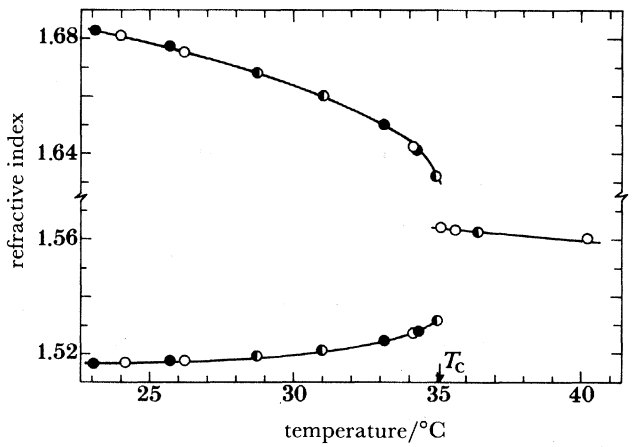

FIGURE 2. Refractive indices of 5CB against temperature, measured by the surface plasmon technique (○) and by the critical angle method (●).

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induced phase shifts in a 250 μm film are $\Delta\phi_{\parallel} \approx (10^{-2})2\pi$ rad W^{-1} cm^2 and $\Delta\phi_{\perp} \approx (2\times10^{-3})2\pi$ rad W^{-1} cm^2, respectively. This nonlinearity is not as large as that from molecular reorientation, but is certainly large enough to be observed even with a c.w. laser beam. Furthermore, the two mechanisms have very different response times, and hence their effects can be easily separated.

The dynamic response of a nematic substance to an applied laser field is actually quite complicated. The molecular reorientation angle, θ, should obey a diffusion-type equation, which, after some simplification, takes the form (Van Doorn 1975)

$$\gamma\,\partial\theta/\partial t - K\nabla^2\theta = T_H + T_{\mathrm{opt}}, \tag{1}$$

where γ is an effective viscosity coefficient, K is an effective elastic constant, and T_H and T_{opt} are the torques due to the applied magnetic and optical fields, respectively. By knowing that the spatial dependence of θ is usually dominated by the Fourier component $\cos(\pi z/d)$, and assuming that the optical-field-induced orientation is sufficiently small, an integration of the above equation with a suitable proportional constant should yield the following approximate dynamic equation for the induced phase shift $\Delta\phi_\theta$ (Durbin *et al.* 1983):

$$\left(\frac{\partial}{\partial t}+\frac{1}{\tau_\theta}\right)\Delta\phi_\theta = \frac{\alpha_\theta}{\tau_\theta} I. \tag{2}$$

This is in the form of a Debye relaxation equation, with τ_θ being the relaxation time and α_θ representing the reorienting strength. We can show that $\tau_\theta \approx \gamma/[\pi^2K/d^2 - GH^2]$, which is of the order of a few seconds to a few tens of seconds for a 5CB sample of *ca.* 100 μm thick, where G is a constant depending on the initial orientation induced by the d.c. magnetic field (Hsiung *et al.* 1984). The constant α_θ also depends on the initial orientation and can be of the order of 6 rad W^{-1} cm^2. The temperature rise in a medium due to laser heating of course obeys the thermal diffusion equation. A similar simplification procedure as in the reorientation case leads to the dynamic equation for the thermally induced phase shift $\Delta\phi_T$ (Durbin *et al.* 1983):

$$\left(\frac{\partial}{\partial t}+\frac{1}{\tau_T}\right)\Delta\phi_T = -\frac{\alpha_T}{\tau_T} I, \tag{3}$$

where $\tau_T \approx d^2/\pi^2D$ is of the order of 0.1 s for a 100 μm sample (D being the heat diffusion constant), and α_T is directly proportional to the absorption coefficient of the medium.

The above values of Δn (or $\Delta\phi$) and their dynamic characteristics allow us to estimate the strengths of various nonlinear optical effects in liquid crystals. We consider here only optical bistability. It is well known that if a laser beam is intense enough to induce a phase shift of $\Delta\phi = \pi$ in a nonlinear medium filling a Fabry–Perot interferometer, one should be able to observe optical bistability. With a 100 μm 5CB film in an appropriate bias magnetic field, it requires a beam intensity of only a few watts per square centimetre, assuming that molecular reorientation is the dominant mechanism for nonlinearity. Indeed, as shown in figure 3, optical bistability can be easily observed in an 83 μm 5CB film sandwiched between two mirrors (Durbin *et al.* 1983). The first bistable loop is complete at $I_{\mathrm{in}} \approx 6$ W cm^{-2}. With increasing laser intensity, $\Delta\phi$ soon becomes larger than multiples of π, and hence multiple bistable loops are readily generated.

In the above case I have assumed that the laser polarization is parallel to the biasing magnetic field. The laser heating contribution to Δn is then negligible in comparison with the molecular

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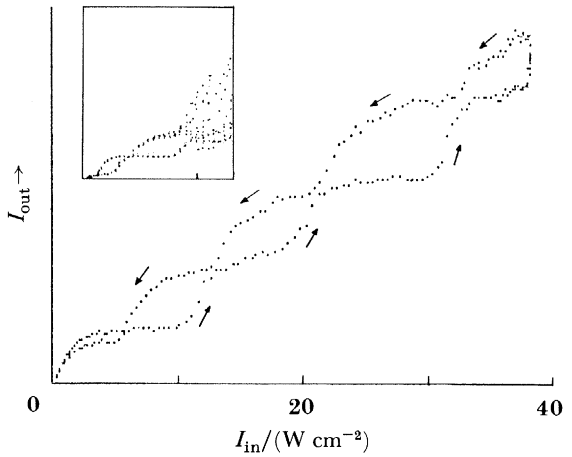

FIGURE 3. Multiple bistable loops obtained from a Fabry–Perot interferometer consisting of an 83 μm 5CB nematic film sandwiched between two mirrors of reflectivities 55 and 75% at 514.5 nm. A bias magnetic field of 103 kA m^{-1} parallel to the laser polarization was applied to the system. The inset shows bistable loops and the onset of oscillation.

reorientation effect in the steady-state operation. Laser heating also induces a $\Delta n_\perp$ that is even smaller than $\Delta n_\parallel$. It is, however, possible to increase the laser heating contribution by dissolving some absorbing molecules in the liquid crystal so that $\Delta\phi_T$ and $\Delta\phi_\theta$ become comparable. We can then have an interesting case of a nonlinear Fabry–Perot interferometer with two input beams or orthogonal polarizations, which are coupled through nonlinearity in the cavity (Kaplan & Meystre 1982), i.e.

$$\Delta\phi_\parallel = aI_\parallel T_\parallel + bI_\perp T_\perp, \quad \Delta\phi_\perp = cI_\parallel T_\parallel + dI_\perp T_\perp, \tag{4}$$

where a, b, c, d are constant coefficients and $T_\parallel$ and $T_\perp$ are the transmission coefficients of the interferometer for the two polarizations.

The different dynamic responses of the two mechanisms can make the bistable operation even more interesting. Since τ_T is nearly two orders of magnitude smaller than τ_θ, the bistable operation will actually be dominated by the laser heating effect if the operating time for light to go around the bistable loops is in the range $\tau_T \lesssim t_{op} \ll \tau_\theta$. As t_{op} becomes comparable with or larger than τ_θ, the bistable operation is then dominated by molecular reorientation. The two mechanisms also induce phase shifts of opposite signs ($\Delta n_\parallel > 0$ from molecular orientation and $\Delta n_\parallel < 0$ from laser heating). This, together with the different response times, can cause the output from the interferometer to self-oscillate. The phenomenon can be understood physically from a graphic construction (Durbin *et al.* 1983).

Figure 4*a* shows that as the laser intensity increases, the output (gauged by $\Delta\phi$) of the Fabry–Perot interferometer rises in steps in a stable fashion. When the operating point, O, is reached, however, one finds that the intensity is now high enough for the interferometer to go into bistable operation with the laser heating mechanism alone, as indicated by the broken line. The operating point, O, is then no longer stable against thermal fluctuations, and in a time short compared with τ_θ it shifts to either A or B. Yet neither A nor B is an operating point in the steady state. As time goes on, molecular reorientation begins to respond to the intensity change in the cavity. The operating point should then move from A (or B) along the Fabry–Perot transmission curve towards O, but as it reaches C (or C′) the system again becomes unstable

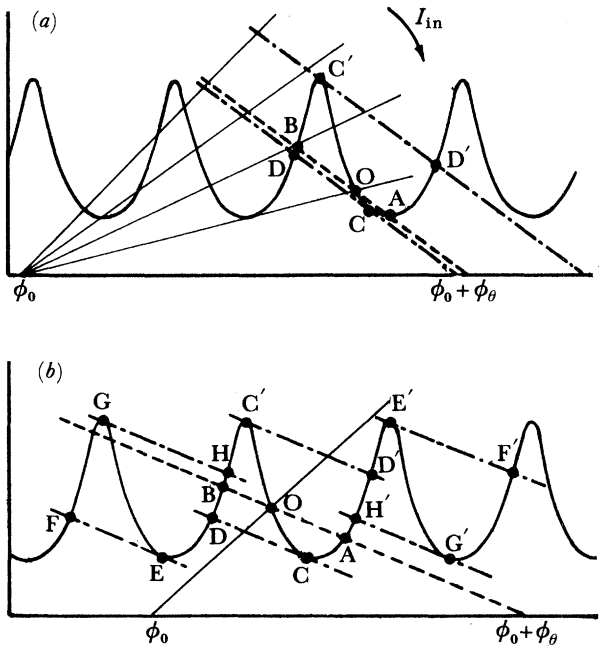

FIGURE 4. Graphic construction showing how the interplay between laser-induced molecular reorientation and thermal effect can lead to indefinite oscillation in the output of a nonlinear Fabry–Perot interferometer. Intersection of the Fabry–Perot transmission curve and solid line gives the steady-state operating point, O. The broken line describes the phase shift due to laser heating alone. In (*a*) the operating cycle is ACDBC′D′A; in (*b*) the operating cycle is ACDEFGHBC′D′E′F′G′H′A.

under thermal fluctuations, and is quickly switched to D (or D′). Afterwards, the operating point is shifted to C′ (or C) by molecular reorientation, switched to D′ (or D) by thermal action, and shifted back to A (or B). It then repeats the cycle ACDBC′D′A, and the output appears in the form of an indefinite oscillation as shown in figure 5. The period of oscillation is of the order of τ_θ.

The picture here predicts that oscillation can occur only when the laser intensity has increased

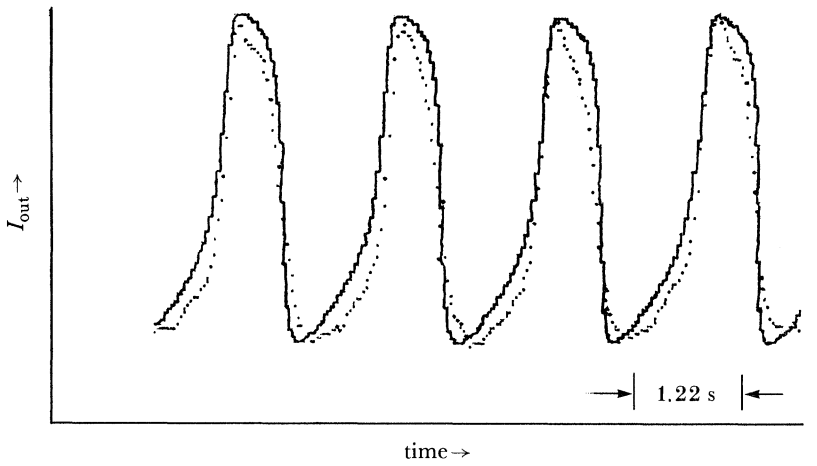

FIGURE 5. Oscillatory output (dotted line) from the Fabry–Perot interferometer described in figure 3 observed with an input laser intensity $I_{in} = 70$ W cm^{-2} and a bias magnetic field $B = 123$ kA m^{-1}. The solid curve is a theoretical fit.

to such a value that the slope of the broken line in figure 4 is less negative than the most negative tangent that one can draw from the point O to the Fabry–Perot transmission curve. The lowest possible intensity for oscillation, I_{th}, arises when O happens to be at an inflection point of the Fabry–Perot curve. It is determined from the relation

$$|C\alpha_T I_{\text{th}}|^{-1} = |\mathrm{d}\mathscr{I}/\mathrm{d}\phi|_{\max} \approx F/\pi$$

at which the oscillation condition is no longer satisfied. As the laser intensity is increased further, spatial self-phase modulation may become significant and complicate the oscillation pattern of the output. The effect of the intensity distribution of the transverse beam on oscillation has not yet been worked out.

From the graphical construction, it can be seen that if the laser heating mechanism is stronger, the output will break into oscillation at a lower laser intensity. The oscillation pattern will also change with the laser intensity in a rather interesting way. At a sufficiently high intensity with $\alpha_\theta + \alpha_T < \alpha_\theta$, the operating point O can be moved to a situation shown in figure 4*b*. Here again, the point O is not stable against thermal fluctuations. Assume that the operation is first shifted to A, moved to C, and shifted again to D. Now, because of the lower Fabry–Perot transmission coefficient at D, the laser intensity in the cavity is not sufficient to maintain the corresponding $\Delta\phi_\theta$ present. The molecules therefore have to reorient to move the operating point further down the curve towards E, at which it switches to F. The movement down the curve would continue if the laser intensity in the cavity at F is still too low. Otherwise, the operating point will move up the curve from F to G, H, C′, D′, E′, F′, G′, H′, and back to A. Clearly, in this case, the oscillation pattern involving six switching actions in a cycle is much more complicated than the one shown in figure 5. At even higher intensities, more switching actions should occur in a cycle, and the oscillation pattern could become hard to analyse and look somewhat like noise.

Whether liquid crystals can be used as the nonlinear medium to study bifurcation and chaos in an all-optical system has yet to be explored. Interesting results of such study, however, have already been obtained on hybrid systems (Song *et al.* 1983; Zhang *et al.* 1983). In general, liquid crystals as nonlinear media in bistable devices offer a lot for our imagination. The many, easily controllable parameters of the material should allow us to study the various physical phenomena connected with bistability in great detail.

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