

# Electro-optical effect of a magnetically biased ferronematic liquid crystal

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The electro-optical effect of a magnetically biased ferronematic liquid-crystal film is investigated by using birefringence measurements. When a magnetic field is applied, the threshold voltage of the Fredericksz transition no longer exists. The dependence of the birefringence on the magnetic field strength in the low field regime is presented. A theory that accounts for the results is given.

The electro-optical effect in liquid crystals (LC's) has played an important role in technological applications. It was observed by Fredericksz that, if a nematic liquid-crystal (NLC) cell is aligned so that the smallest dielectric constant is parallel to the applied field, there exists a threshold field below which the molecular orientation is unaffected.<sup>1</sup> Above the threshold the orientation of the director gradually distorts toward the minimum energy configuration and induces the birefringence. Meyerhofer<sup>2</sup> reported that a Fredericksz transition (FT) threshold for a pretilted NLC cell no longer exists.

Usually a NLC cell with a pretilt angle can be achieved by surface treatment.<sup>3</sup> In our work we used a ferronematic<sup>4</sup> LC, which has the unique property of having a high magneto-optical effect. Consequently, a pretilt angle is easily obtained and tuned by an external magnetic field. No threshold voltage of the FT has been observed. This can be verified theoretically. The birefringence measurements and theoretical results are in good agreement in the low field regime.

We consider a homeotropically aligned ferronematic LC film of thickness  $D$ . The geometry is depicted in Fig. 1. The local magnetization  $M$  is perpendicular to the local nematic director  $n$ . Both electric field  $E$  and magnetic field  $H$  are applied along the  $z$  axis, which is the direction of the unperturbed director  $n_0$ . Neglecting the effect of the disclination<sup>5</sup> defects near the magnetic grains, the system free energy per unit area of the wall is given by

$$f = \int_{-D/2}^{D/2} dZ \left[ \frac{1}{2} K_{33} (1 + K \sin^2 \theta) \left( \frac{d\theta}{dZ} \right)^2 - MH \sin \theta(z) - \frac{\Delta\epsilon}{8\pi} (E \cos \theta)^2 \right], \quad (1)$$

where  $K = (K_{11} - K_{33})/K_{33}$ ,  $K_{11}$  and  $K_{33}$  are the splay

and bend elastic constants, respectively, and  $\Delta\epsilon = \epsilon_{11} - \epsilon_{\perp}$  is the dielectric anisotropy due to the quasi-static electric field.

Considering a small deviation from the original arrangement, that is,  $\theta(Z) \ll 1$ , and using the approximation  $K_{11} \cong K_{33}$ , the free energy becomes

$$f = \int_{D/2}^{D/2} dZ \left[ \frac{1}{2} K_{33} \left( \frac{d\theta}{dZ} \right)^2 - MH\theta(Z) - \frac{\Delta\epsilon}{8\pi} E^2 (1 - \theta^2) \right]. \quad (2)$$

Let  $\theta_0(Z)$  represent the pretilt angle induced by the magnetic field  $H$ . Matching the boundary conditions  $\theta(\pm D/2) = \theta_0(\pm D/2) = 0$ , we expand  $\theta(z)$  and  $\theta_0(z)$  in Fourier series:

$$\theta(z) = \sum_{n=0}^{\infty} u_n \cos(2n+1) \frac{\pi Z}{D},$$

$$\theta_0(z) = \sum_{n=0}^{\infty} v_n \cos(2n+1) \frac{\pi Z}{D}.$$

In terms of  $u$ 's and  $v$ 's the difference of free energy is

$$\Delta f = f(\theta) - f(\theta_0) = \frac{K_{33}D}{4} \sum_{n=0}^{\infty} (u_n^2 - v_n^2) \left( \frac{(2n+1)\pi}{D} \right)^2 - \sum_{n=0}^{\infty} MH(-1)^n \frac{2D}{(2n+1)\pi} (u_n - v_n) + \frac{\Delta\epsilon E^2 D}{8\pi} \sum_{n=0}^{\infty} u_n^2 - \frac{\Delta\epsilon E^2 D}{8\pi}. \quad (3)$$

Instability occurs whenever  $\Delta f$  becomes negative.

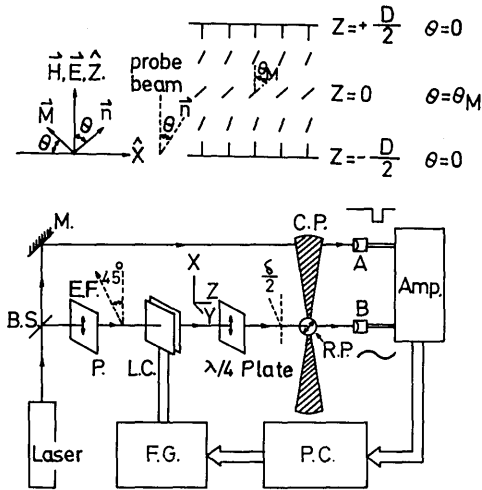


Fig. 1. Molecular geometry and block diagram of the experimental setup. B.S., beam splitter; M., mirror; P., polarizer; E.F., Earth field; L.C., liquid-crystal cell; R.P., rotating polarizer; C.P., chopper; A and B, photodiodes; Amp., lock-in amplifier; P.C., Apple II computer; F.G., function generator.

This can be achieved by any  $E^2 > 0$ . Putting  $u_{2m-1} = v_{2m-1}$  for any integer  $m$ , the criterion for instability becomes

$$E^2 > \left\{ \frac{K_{33}D}{4} (u_{2m} + v_{2m}) \left[ \frac{(4m+1)\pi}{D} \right]^2 - MH \frac{2D}{(4m+1)\pi} \right\} \left( \frac{\Delta\epsilon D}{8\pi} - \frac{\Delta\epsilon D}{8\pi} \frac{1}{2} u_{2m}^2 \right)^{-1} \times (u_{2m} - v_{2m}). \quad (4)$$

It is obvious that for any  $E^2 > 0$  there exist  $u_{2m} \neq v_{2m}$  such that instability can occur. In other words, there is no threshold for the FT.

The reorientation angle of the director,  $\theta(Z)$ , can be calculated by using a free-energy minimization procedure<sup>4,6</sup> that yields

$$ZC^{1/2} = \int_0^{\theta(z)} (1 + K \sin^2 \theta)^{1/2} \times \left[ 1 - \frac{2(A \sin \theta + B \sin^2 \theta)}{C} \right]^{-1/2} d\theta, \quad (5)$$

where  $A = -MH/K_{33}$ ,  $B = -\Delta\epsilon E^2/8\pi K_{33}$ ,  $C = 2(A \sin \theta_m + B \sin^2 \theta_m)$ , and  $\theta_m$  is the maximum molecular reorientation angle in the LC sample. For a small molecular orientation distortion ( $\theta_m \ll 1$ ), it is easy to get

$$\theta(Z) = \theta_0(Z) \left( 1 - \frac{\Delta\epsilon E^2 D^2}{32\pi K_{33}} \right)$$

and

$$\theta_m = \frac{MHD^2}{8K_{33}} \left( 1 - \frac{\Delta\epsilon E^2 D^2}{32\pi K_{33}} \right), \quad (6)$$

where  $\theta_0(Z) = MH/8K_{33} (D^2 - 4Z^2)$  is the pretilt angle.

A normally incident laser beam with wavelength  $\lambda$  should have its extraordinary component experience an induced phase change

$$\delta = \frac{\pi}{60\lambda K_{33}^2} (n_e - n_o) D^5 M^2 H^2 \left( 1 - \frac{\Delta\epsilon E^2 D^2}{32\pi K_{33}} \right)^2, \quad (7)$$

where  $n_o$  and  $n_e$  are, respectively, the ordinary and the maximum extraordinary refractive indices of the sample.

In our experiment, the ferronematic LC is magnetically doped *N-p*-methoxybenzylidene-*p*-butylaniline,<sup>4</sup> with a volume filling factor  $f = 1.89 \times 10^{-6}$ . The magnetic particles are  $\gamma\text{-Fe}_2\text{O}_3$  needles 0.5  $\mu\text{m}$  long

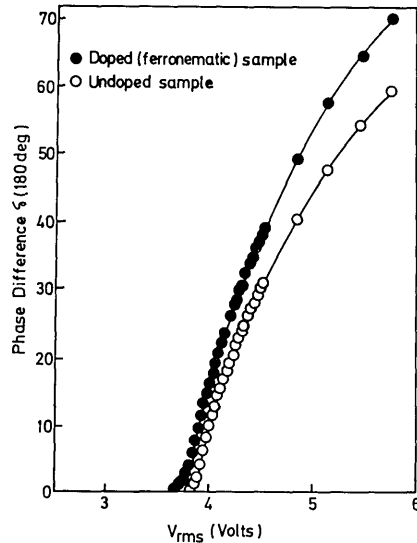


Fig. 2. Experimental values for the electric-field-induced birefringence.

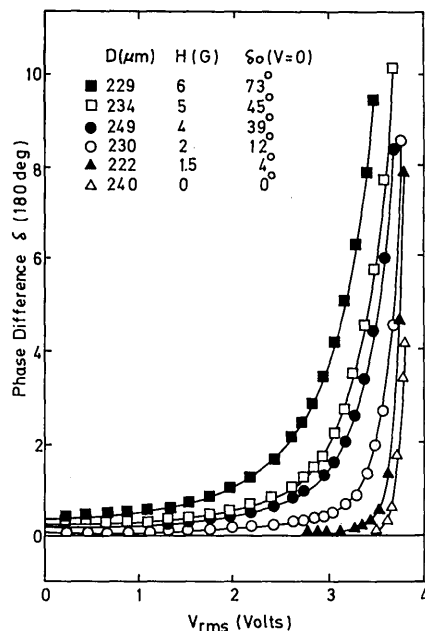


Fig. 3. Phase difference versus voltage for samples with various magnetic-field-induced pretilt angles.

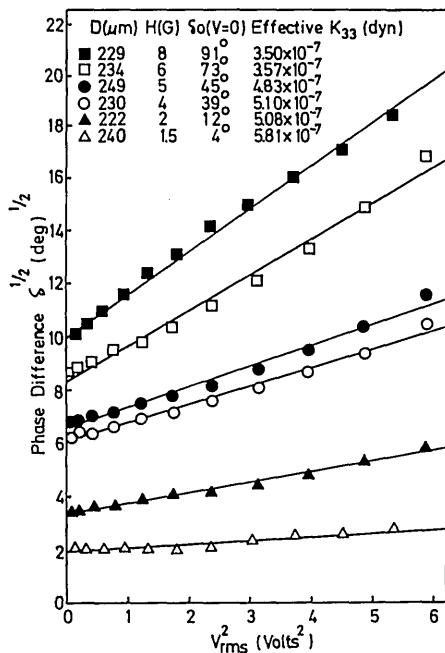


Fig. 4. Phase-difference dependence on voltage,  $\sqrt{\delta} \propto V^2$ , as predicted by Eq. (7).

and have an aspect ratio of  $\sim 7:1$ . Their magnetic dipole moment points along the long axis of the particle and is 70 emu/g. To prevent clumping, these needles are coated with dimethyl octadecyl aminopropyl trimethoxysilyl chloride (DMOAP). The sample films are made by sandwiching the ferronematic liquid crystal between two glass plates. The glass windows are first coated with indium tin oxide, which serves as transparent electrodes, and then with DMOAP to achieve homeotropic alignment.<sup>7</sup>

The field-induced birefringence is measured with a He-Ne probe laser by using a modulation technique originally devised by Lim and Ho.<sup>8</sup> During the measurement the sample film is placed horizontally. The magnetic dipole moment of the ferronematic matrix is first aligned by the horizontal component of the Earth's field. The biasing magnetic field is applied with a pair of Helmholtz coils such that  $H \parallel n$ . A quasi-static electric field at 1 kHz is applied normal to the glass window. A block diagram of the experimental apparatus is shown in Fig. 1. Briefly, the probe beam is split, and the main measurement beam passes successively through a polarizer, the sample, a  $\lambda/4$  plate, and a rotating polarizer before being detected by photodiode A. The polarization direction of the probe beam makes an angle of  $45^\circ$  with the horizontal component of the Earth's field. The axis of the  $\lambda/4$  plate is aligned with the probe-laser polarization direction. A polarizer is mounted at the end of the open shaft of a synchronous motor. The shaft of the motor

also carries a chopper, which intercepts the reference beam. The phase difference between the signals from the photodiodes is measured with a phase-sensitive detector and recorded.

The measured electrocontrolled birefringence of the undoped and doped samples is given in Fig. 2. Both samples show a typical critical behavior near the threshold voltages, which are 3.88 and 3.77 V, respectively. Using  $V_{th} = \pi\sqrt{4\pi K_{33}/\Delta\epsilon}$  and  $\Delta\epsilon = -0.5$ , the corresponding values of the elastic constant,  $K_{33}$ , are  $6.74 \times 10^{-7}$  and  $6.37 \times 10^{-7}$  dyn. It is obvious that the effective elastic constant of the doped sample is smaller than that of the undoped sample.

The phase difference versus voltage for samples with different magnetic-field-induced pretilt angles is shown in Fig. 3. One can see that no threshold voltage exists for these samples and that the magnetic field enhances the electrocontrolled birefringence significantly.

For comparison with the prediction of Eq. (7), we plot the square root of the phase difference versus the square of the voltage in Fig. 4. After least-squares fitting, all six samples with different pretilt angles show a linear relation  $\delta^{1/2} = \delta_0^{1/2}(1 - \Delta\epsilon V^2/32\pi K_{33})$ , where  $\delta_0$  is the phase difference for zero voltage. One can get the effective elastic constant  $K_{33}$  for each sample from the slope of these straight lines. It is obvious that the larger the pretilt angle the smaller the effective elastic constant.

In conclusion, we have shown that the variable pretilt angle of a ferronematic film can be achieved by a weak magnetic field. No threshold voltage exists for such a pretilted sample. Our quantitative measurement shows that in the low field regime the square root of the phase retardation varies linearly with the square of the applied voltage. This agrees with our theoretical prediction, although the effect of the disclination defects near the magnetic grains is neglected.

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