Feedback-induced first-order Freedericksz transition in a nematic film

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It is shown that optical-electrical feedback can transform the Freedericksz transition in a nematic film from second order to first order. The criteria that indicate whether the Freedericksz transition is first or second order are obtained. The hysteresis accompanying the first-order transition versus the feedback intensity is discussed. The feedback-induced first-order Freedericksz transition has been observed experimentally. The experimental results are in good agreement with the theoretical analysis.

It has been shown that the usual Freedericksz transitions in insulating nematic liquid crystals are always second order.[1] Deuling *et al.*[2] proposed that if the conductive anisotropy in a conducting nematic liquid crystal is large enough, the deformation of the molecules can exhibit a discontinuous change and hysteresis. Lately the optically induced and enhanced first-order Freedericksz transition in nematics has received particular attention. The theoretical prediction of this kind of first-order transition was provided by Ong.[3] Recently the magnetic-field-induced first-order optical Freedericksz transition in a nematic film was observed experimentally.[4] In the liquid-crystal hybrid optical device,[5] the intensity of the output light can exhibit bistability because of the electrical feedback. The purpose of this Letter is to study the Freedericksz transition in a nematic film with electrical feedback. It is found that if there is no feedback, the Freedericksz transition is second order. But when the feedback intensity is increased gradually, the second-order transition can become first order and show hysteresis. The width of the hysteresis depends on the feedback intensity. The tricritical point separating the second- and first-order transitions, and the criterion for the physical parameters that indicate whether the transition is first or second order, are obtained. The experimental results are in agreement with the theoretical analysis.

A sketch of the system is shown in Fig. 1. The light source injects a light beam I_1 into the liquid-crystal system LC. Let $T(U)$ be the transmission of the LC and U be the total voltage applied to the liquid-crystal film. The transmitted light intensity is $I = I_1T(U)$. The feedback voltage U_F is proportional to the transmitted intensity, $U_F = KI = KI_1T(U) \equiv f(U)$. Here we define a function $f(U)$, incorporating the feedback intensity K. The total voltage applied to the liquid-crystal film is composed of the feedback voltage and a bias voltage U_B:

$$U = U_B + f(U). \tag{1}$$

We consider a planar parallel-aligned nematic film of thickness d confined between the plates at $z = 0$ and $z = d$ in a Cartesian-coordinate system, where the z axis is along the normal of the plates and the x axis is along the direction of the molecules at the surfaces. In the presence of an external electric field, the orientation of the nematic director is determined by the minimization of the free energy[1]

$$F = \frac{1}{2}\int_0^d \left[(k_1 \cos^2\varphi + k_3 \sin^2\varphi)\left(\frac{d\varphi}{dz}\right)^2 - \mathscr{E}_a E^2 \sin^2\varphi\right] dz.$$

We then obtain

$$(k_1 \cos^2\varphi + k_3 \sin^2\varphi)\left(\frac{d^2\varphi}{dz^2}\right) + (k_3 - k_1)\sin\varphi \times \cos\varphi\left(\frac{d\varphi}{dz}\right)^2 + \mathscr{E}_a E^2 \sin\varphi\cos\varphi = 0, \tag{2}$$

where k_1 and k_3 are the elastic constants, $\mathscr{E}_a$ is the dielectric anisotropy, E is the electric field, and φ is the angle between the director and the x axis. Near threshold, the angle φ is very small, and

$$\varphi \simeq \varphi_m \sin(\pi/d)z, \tag{3}$$

where φ_m is the maximum of φ at $z = d/2$.

By expanding Eq. (2) up to and including terms of the order of φ^3, and inserting expression (3), we obtain

$$\frac{U - U_0}{U_0} = \left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)\varphi_m{}^2, \tag{4}$$

where $U_0 = \pi(k_1/\mathscr{E}_a)^{1/2}$ is the threshold voltage.

Therefore we can expand the function $f(U)$ near the threshold U_0, i.e., in terms of $\varphi_m{}^2$. From Eq. (1) we obtain

$$U = U_B + f(U_0) + f'(U_0)(U - U_0).$$

This can be rewritten as

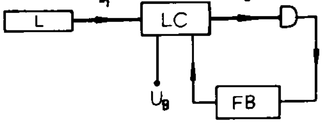

Fig. 1. Sketch of the system: L, light source; LC, liquid-crystal system; FB, feedback system.

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$$U - U_0 = \frac{U_B - U_{B0}}{1 - f'(U_0)}$$

or

$$\varphi_m^{\ 2} = \frac{U_B - U_{B0}}{U_0\left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)[1 - f'(U_0)]}, \tag{5}$$

where

$$U_{B0} = U_0 - f(U_0) \tag{6}$$

is the rising threshold bias voltage, below which no molecular reorientation can be induced.

Equation (5) shows that if $f'(U_0) < 1$, the Freedericksz transition is second order, in which case $\varphi_m = 0$ for $U_B < U_{B0}$ and φ_m changes continuously as U_B increases for $U_B > U_{B0}$. But if $f'(U_0) > 1$, Eq. (1) no longer holds. We must expand $f(U)$ to include the higher-order term $(U - U_0)^2$, i.e., $\varphi_m^{\ 4}$; then

$$U = U_B + f(U_0) + f'(U_0)(U - U_0) + \tfrac{1}{2}f''(U_0)(U - U_0)^2, \tag{7}$$

or

$$U_B/U_{B0} = 1 - \alpha\varphi_m^{\ 2} + \tfrac{1}{2}\beta\varphi_m^{\ 4} \equiv g(\varphi_m^{\ 2}), \tag{8}$$

where

$$\alpha = \frac{U_0}{U_{B0}}\left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)[f'(U_0) - 1],$$

$$\beta = -\frac{U_0^{\ 2}}{U_{B0}}\left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)^2 f''(U_0). \tag{9}$$

At the critical point $U_B = U_{B0}$, the stability of the state $\varphi_m = 0$ is determined by the sign of $[dg(\varphi_m^{\ 2})/d(\varphi_m^{\ 2})]_{\varphi_m=0} = -\alpha$. If $\alpha < 0$, the state $\varphi_m = 0$ is stable. This is the case of the second-order Freedericksz transition described above. If $\alpha > 0$, i.e., $f'(U_0) > 1$, the state $\varphi_m = 0$ is unstable and a first-order Freedericksz transition occurs. The conversion from the second-order to the first-order transition occurs at $\alpha = 0$, from which one obtains the tricritical point $f'(U_0) = 1$.

From Eq. (8) we obtain

$$\varphi_m^{\ 2} = \frac{\alpha + [\alpha^2 + 2\beta(U_B/U_{B0} - 1)]^{1/2}}{\beta}. \tag{10}$$

Equation (10) shows that for a first-order transition, if $\beta > 0$, as U_B increases from zero, $\varphi_m = 0$ for $U_B < U_{B0}$ and the state changes discontinuously at $U_B = U_{B0}$ from $\varphi_m = 0$ to $\varphi_m = (2\alpha/\beta)^{1/2}$ and then changes continuously as U_B increases. But if U_B decreases from a value greater than U_{B0}, the state assumes a finite amount of distortion, even at $U_B < U_{B0}$. On reaching a lower falling threshold voltage $U_{B0}' = U_{B0}[1 - (\alpha^2/2\beta)]$, the state changes discontinuously from $\varphi_m = (\alpha/\beta)^{1/2}$ back to $\varphi_m = 0$. The changes at both the rising and falling thresholds are discontinuous. The increase and decrease of the bias voltage induce the same deformations for $U_B > U_{B0}$ and $U_B < U_{B0}'$ but different deformations for $U_{B0}' < U_B < U_{B0}$. Thus we obtain bistability. We define the width of the hysteresis as

$$W = U_{B0} - U_{B0}' = U_{B0}(\alpha^2/2\beta). \tag{11}$$

From Eqs. (9) and the definition of $f(U)$, we find that if the input intensity I_1 is fixed, $W = (AK - 1)^2/(BK)$, where A and B are two constants that do not contain K. Let K_c be the feedback intensity at the tricritical point. Because $W = 0$ at $K = K_c$, $A = 1/K_c$. Letting $W = W_0/2$ at $K = 2K_c$, we then obtain $B = 1/(W_0K_c)$. Therefore

$$W/W_0 = (K/K_c - 1)^2/(K/K_c). \tag{12}$$

Equation (12) shows that if $K < K_c$ the Freedericksz transition is second order, and if $K > K_c$ it is first order. Both K_c and W_0 can be obtained from the experiment.

From Eq. (6), we obtain

$$U_{B0}/U_0 = 1 - KI_1T(U_0)/U_0. \tag{13}$$

This shows that the threshold bias voltage U_{B0} is linearly related to the feedback intensity K. If K is increased, U_{B0} will be decreased. But there is an upper limit of K at which $U_{B0} = 0$, i.e., the system itself can induce the Freedericksz transition in a liquid crystal, even without the bias voltage. From Eq. (13) we obtain the upper limit, $K_m = U_0/[I_1T(U_0)]$.

The above analysis uses the function $f(U)$, which includes the feedback intensity K and the light transmission of the liquid crystal. In order to determine an expression for it in our system, we should analyze the transmission first. The optical phase difference between the light polarized along the y axis and that along the x axis is given by

$$\delta = \frac{4\pi}{\lambda}\int_0^{d/2}[n_o - n(z)]dz, \tag{14}$$

$$n(z) = n_on_e/(n_e^{\ 2}\sin^2\varphi + n_o^{\ 2}\cos^2\varphi)^{1/2}, \tag{15}$$

where λ is the wavelength of the light and n_o and n_e are the ordinary and extraordinary indices of refraction, respectively.

Expanding the integrand in Eq. (14) up to the terms of the order of φ^2 and inserting Eq. (3) into the integral, we obtain

$$\delta = \frac{2\pi d}{\lambda}(n_o - n_e) + \frac{\pi d}{2\lambda}n_e\frac{n_e^{\ 2} - n_o^{\ 2}}{n_o^{\ 2}}\varphi_m^{\ 2}. \tag{16}$$

In our system the liquid-crystal cell is placed between two polarizers oriented at +45° and −45°, respectively, relative to the surface molecular orientation. The intensity of the transmitted light is given by[6]

$$I = I_1\sin^2(\delta/2). \tag{17}$$

Inserting Eq. (16) into Eq. (17), we get

$$I = I_1\sin^2\left[\frac{\pi d}{\lambda}(n_o - n_e) + \frac{\pi d}{4\lambda}n_e\frac{n_e^{\ 2} - n_o^{\ 2}}{n_o^{\ 2}}\varphi_m^{\ 2}\right]. \tag{18}$$

Therefore the expression for $f(U)$ is obtained by inserting Eq. (4) into Eq. (18):

$$f(U) = KI_1 \sin^2\left[\frac{\pi d}{\lambda}(n_o - n_e) + \frac{\pi d}{4\lambda} n_e \frac{n_e^2 - n_o^2}{n_o^2} \times \left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)^{-1} \frac{U - U_0}{U_0}\right], \quad (19)$$

and then

$$f'(U_0) = KI_1 \frac{\pi d n_e (n_e^2 - n_o^2)}{4\lambda n_o^2 U_0}\left(\frac{1}{3} + \frac{k_3 - k_1}{2k_1}\right)^{-1} \times \sin\frac{2\pi d}{\lambda}(n_o - n_e). \quad (20)$$

The above analysis can be tested by an experiment. In the experiment, we use a planar parallel-aligned nematic film of E7 as a sample. At a temperature of 22°C, we have[7] $\mathcal{E}_\parallel = 19.20$, $\mathcal{E}_a = 13.80$, $n_o = 1.5222$, $\Delta n = 0.224$, $k_1 = 10.7 \times 10^{-12}$ N, and $k_3 = 20.7 \times 10^{-12}$ N for E7. The liquid-crystal film was sandwiched between two glass plates coated with transparent electrodes in order to apply the electric field. The surfaces were rubbed to ensure planar parallel orientation of the surface molecules. The temperature of the sample was regulated to 22.0 ± 0.1°C. A 25-mW He–Ne laser was used as the light source. The liquid crystal was placed between two polarizers oriented at +45° and −45°, respectively, relative to the surface molecular orientation. The light beam was normally incident upon the cell. The output intensity was detected and converted to an electrical voltage as a feedback signal by an optical-electrical cell. In order to avoid the electrode effect of the liquid crystal, we used this feedback voltage and the bias voltage together to modulate an alternating voltage at a frequency of 0.6 kHz. Then this ac voltage was applied to the liquid-crystal cell. The input intensity I_1 was fixed. We changed $f'(U_0)$ by changing the feedback intensity K. For several typical values of K, the output intensity versus the bias voltage is shown in Fig. 2. The horizontal and longitudinal axes are the bias voltage U_B and the output intensity I, respectively. The dashed lines correspond to the theoretical results obtained from Eqs. (10) and (18). Because for large K or U_B the upper state is far from threshold, the discrepancy in this case is presumably due to the expansion of $f(U)$ at the threshold. We obtained the value of K_c from the fit of Eq. (12). It is shown that when $K < K_c$, φ_m changes from $\varphi_m = 0$ to $\varphi_m > 0$ continuously with increasing U_B, i.e., a second-order Freedericksz transition. But when $K > K_c$, φ_m shows discontinuous

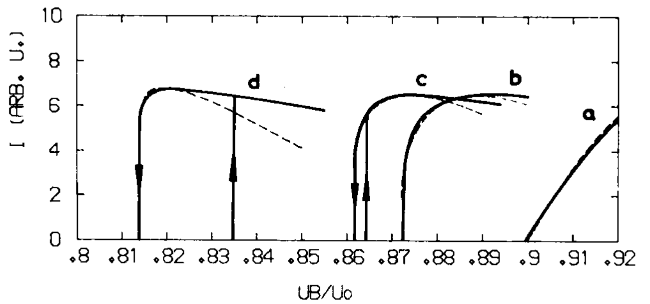

Fig. 2. Output intensity versus the bias voltage: a, $K/K_c = 0.492$; b, $K/K_c = 1.149$; c, $K/K_c = 1.313$; d, $K/K_c = 1.969$.

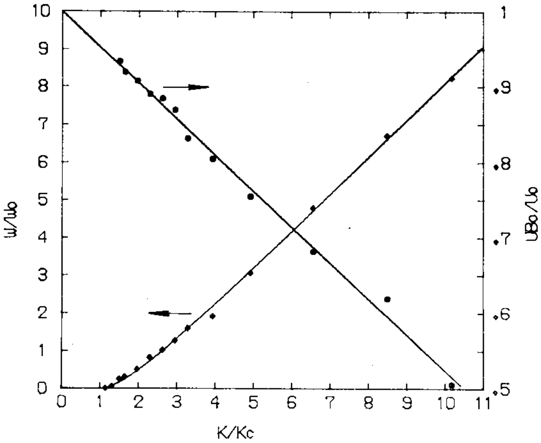

Fig. 3. Width of the hysteresis and threshold bias voltage versus the feedback intensity.

change and hysteresis, i.e., a first-order Freedericksz transition occurs. For each K, the bias voltage was changed slowly enough to ensure static equilibrium. In order to permit the hysteresis to be observed, U_B was changed in both the increasing and decreasing directions.

In the first-order Freedericksz transition, we observed the width of the hysteresis versus the feedback intensity K. The results are shown in Fig. 3. The solid line is the theoretical fit from Eq. (12). The experimental results agree quite closely with the theory. We obtained $K_c = 1.8205$ V/mW and $W_0 = 0.04226$ V from the fit of the observed data. In the experiment, $I_1 = 0.044$ mW and $d \simeq 14$ μm. In order to avoid the error in d, we obtained the value of $\sin(2\pi d/\lambda)$ $(n_o - n_e)$ directly in the expression for $f'(U_0)$ by observing the transmission of the liquid crystal. We obtained $T(U_0) = 0.529$; then $\sin(2\pi d/\lambda)(n_o - n_e) = 0.998$. From Eq. (20), we find $f'(U_0)|_{K=K_c} = 1.030$, which agrees well with the theoretical prediction $f'(U_0)|_{K=K_c} = 1$.

We also tested the linear relation between the threshold bias voltage U_B and the feedback intensity K. The experimental results are shown in Fig. 3. The solid line is the result of a linear least-squares fit from which we obtained the upper limit $K_m/K_c = 21.0$. From the expression $K_m/K_c = U_0/[K_c I_1 T(U_0)]$ we get $K_m/K_c = 21.9$. These results are in good agreement.

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