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Switching Characteristics of a Field-Induced Structure Near the Isotropic-Cholesteric Transition

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The static and dynamic properties of the flexoelectric effect in cholesteric liquid crystals are presented. In a simple geometry, the application of an external electric field perpendicular to the cholesteric helix results in a linear electro-optic effect. The switching time for optical modulation is almost independent of the field strength. The temperature dependence of this optical switching follows the Arrhenius type behavior.

Keywords: electrooptics, cholesterics, switching characteristics, flexoelectric effect

It has been established both theoretically[1] and experimentally[2,3] that the helical structure of a cholesteric liquid crystal (LC) can be modified or even completely unwound by the application of an external magnetic or electric field. This is well understood in terms of the dielectric (or diamagnetic) coupling with an external electric (or magnetic) field which is quadratic in the magnitude of the field. In this paper we describe the switching characteristics of a linear electro-optic effect, arising from flexoelectric coupling,[4] in a field-induced helical structure of a cholesteric LC.

This linear effect can be easily observed in a well-aligned cholesteric sample in which the helix axis lies in the plane of the surfaces. In the absence of an electric field, this cholesteric sample behaves as a uniaxial material with its optic axis perpendicular to the mean molecular axis and parallel to the helix axis. When an electric field is applied, the cholesteric helix distorts to lower the free energy of the system, which results in the rotation of the optic axis. This problem is mathematically equivalent to the rotation of the helix axis, keeping the director fixed.[5] The expression for the free energy in the helix-rotated coordinates is much simpler

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and enables us to get the main features of the problem easily. It should be noted, however, that the two phenomena are physically quite different. In the first case, the optic axis of the system is rotated, keeping the helix axis fixed, and the effect is easily observable. In the second, only the helix axis is rotated and thus the resultant electro-optic effect will be negligible.

First, we derive the expression for the free energy of a cholesteric LC in which the helix uniformly distorts in the presence of an electric field. We then calculate the magnitude of the helix distortion as a function of the field strength under the assumption of free boundary conditions at the surfaces. Consider a cholesteric LC of equilibrium twist t_o in which the director $\hat{n}$ is parallel to the $x - y$ plane, with $n_x = \cos\theta$ and $n_y = \sin\theta$. In the presence of an electric field $\vec{E}$ along the x direction, it is assumed that the helix axis (initially along the z direction) uniformly rotates by an angle ϕ about the x axis. Denoting partial derivatives by subscripts, the free energy density f is then given by[5]

$$f = \frac{1}{2} K_1\theta_y^2 + \frac{1}{2} K_2(t_o - \theta_z)^2 - \bar{e}E\theta_y + \frac{1}{8\pi} \varepsilon_a E^2\sin^2\theta, \tag{1}$$

where the elastic constant $K_1 = K_3$ and $\bar{e}$ is the mean flexoelectric coefficient. Here ε_a denotes the dielectric anisotropy.

The equilibrium structure is determined by the balance between the elastic energy and the field energy provided by the flexoelectric coupling with the applied field if the dielectric contribution is negligible. With the choice of $\varepsilon_a \approx 0$, the free energy, Equation (1), is clearly minimized by $\theta_z = t_o$ and $\theta_y = \bar{e}\,E/K_1$. If the rotated axis of the helix is represented by a wavevector $\vec{k}$ with $\theta = \theta_o + \vec{k} \cdot \vec{r}$, then $|\vec{k}|\cos\phi = t_o$ and $|\vec{k}|\sin\phi = \bar{e}\,E/K_1$ for free boundary conditions. Accordingly, the magnitude of the wavevector $k = t_o\,[1 + (\bar{e}E/K_1t_o)^2]^{1/2}$ and the rotation angle ϕ is given by the relationship $\tan\phi = \bar{e}\,E/K_1t_o$, which is linear in E for small rotations. For fixed helical pitch, i.e., $k = t_o$, the free energy density f becomes only a function of ϕ, and is expressed as

$$f = \frac{1}{2} K_1t_o^2\sin^2\phi + \frac{1}{2} K_2(t_o - t_o\cos\phi)^2 - \bar{e}Et_o\sin\phi. \tag{2}$$

In this case, $\tan\phi = [\bar{e}E - (K_1 - K_2)t_o\sin\phi]/K_2t_o$, which gives the same result for ϕ as before when $K_1 = K_2$. Also, for a general case that the dielectric contribution is non-negligible, the mean free energy averaged over one period of the helical pitch enables us to derive the equation for ϕ as a function of E by the variational principle. Surprisingly, the simple result for ϕ, obtained before, remains valid even in this more general case.[5,6]

As a consequence, we see that the system behaves as if the optic axis is rotated with respect to the electric field direction. The direction of this rotation depends on the polarity of the field, and its magnitude is directly proportional to the strength of the field. Thus, this effect results in a linear electro-optic response and phenomenologically resembles the electroclinic[7] and the ferroelectric[8] effects.

We now derive the equation of the balance of torque which governs the dynamics of the helix distortion, and obtain the expression for the characteristic time associated with the evolution of the helix distortion. Again, assuming that the helical pitch is fixed, i.e., $k = t_o$, the free energy becomes only a function of ϕ. Under this assumption, the dynamic equation for the helix distortion is then given by

$$\eta\left(\frac{\partial\phi}{\partial t}\right) = -K_1 t_o^2 \sin\phi\cos\phi - K_2 t_o^2(1 - \cos\phi)\sin\phi + \bar{e}Et_o\cos\phi, \qquad (3)$$

where η is the effective viscosity associated with the helix distortion. With linear expansions of $\sin\phi \approx \phi$ and $\cos\phi \approx 1$ for small ϕ's, it can be seen from Equation (3) that $\phi(t) \approx \phi(0)\exp(-t/\tau)$ where the equilibrium rotation $\phi(0) = \bar{e}\,E/K_1 t_o$ and the characteristic time $\tau = \eta/K_1 t_o^2$, independent of E to a first approximation.[9] The contributions from both the change in the helical pitch and the next higher order beyond the linear approximation are only of the order of $\phi^2(0) << 1$. Thus, the E^2 field-dependence of the above corrections is expected to be small.

We studied a cholesteric sample that is a mixture of 33% cholesteryl nonanoate and 67% ZLI 2141-100 (from EM Chemicals). The thickness of the sample is about 2.3 μm. This sample might have small dielectric anisotropy (ε_a), which minimizes the contribution of the dielectric coupling to the free energy, because the value of ε_a for ZLI 2141-100 is small. In fact, the unwinding of the helix results from this dielectric contribution in the limit of the high electric field.

The sample cell was made of conductive indium-tin-oxide coated glasses with buffed polymer layers (poly-1,4-butyleneterephthalate) on both surfaces. The material was filled in the isotropic state and cooled into the cholesteric phase in the presence of 10 kHz sine waves at 40 V. The application of the ac field produced a well-aligned structure of large area in which the helix axis of the cholesteric phase lies in the plane of the glass surfaces. There was no appreciable frequency dependence on the alignment quality. The magnitude of the aligning field plays a significant role in both the quality and the orientation of the helix with respect to the buffing axis. The angle between the aligned helix and the buffing axis is independent of the polarity, but depends on the strength of the field, indicating that the alignment mechanism arises mainly from the dielectric effect.

Recently, a simple model of a distorted boundary layer[10] has been developed to account for the process of the helix alignment. The angle of the helix with respect to the buffing axis is directly determined by the amount of twist at a distance of the order of the dielectric coherence length (transition layer) away from the buffed surface, which is consistent with the experimental results. The magnitude of the angle increases with the field up to the critical strength at which the cholesteric pitch dilates.

In our experiments, a well-aligned sample was mounted in a microfurnace for temperature control, and temperature fluctuations were approximately 0.1°C. In the presence of an ac electric field, an optical modulation can be observed by monitoring the transmission through the cell placed between crossed polarizers. Since the rotation of the optic axis is small in our sample, the magnitude of the modulation is expected to be linear in the strength of the applied field. Measure-

ments of the rotation angle of the optic axis were then made with a 400 Hz square wave of variable amplitudes. The transmitted light intensity was monitored with a photodiode and a digitizing oscilloscope. Details of the technique have been described elsewhere.[11]

We briefly mention the dependence of the optic axis rotation on the applied electric field, and concentrate on the switching behavior of the optical modulation. The linear relationship between ϕ and E was satisfied up to a threshold at which the helical structure disappears. A sample that has only one buffed surface, however, exhibited two different linear regimes, which led to a small offset in the rotation of the effective optic axis.[6] The width of the offset became narrower and continuously disappeared with increasing temperature. A more complete study of this behavior may provide useful information about how the surface interactions are chemically or physically modified by buffing and how the dynamic properties are influenced. It is possible to obtain the flexoelectric coefficient $\bar{e}$ from the slope $\bar{e}/K_1 t_o$ of a linear fit of the data ϕ as a function of E. With typical values[12] of $K_1 \approx 1 \times 10^{-6}$ dynes and the helical pitch $2\pi/t_o \approx 0.5$ μm, the magnitude of $\bar{e}$ is typically of the order of 5×10^{-5} cgs units, which is consistent with other measurements.[13,14]

Figure 1 shows a normalized optical modulation and the switching time τ of the

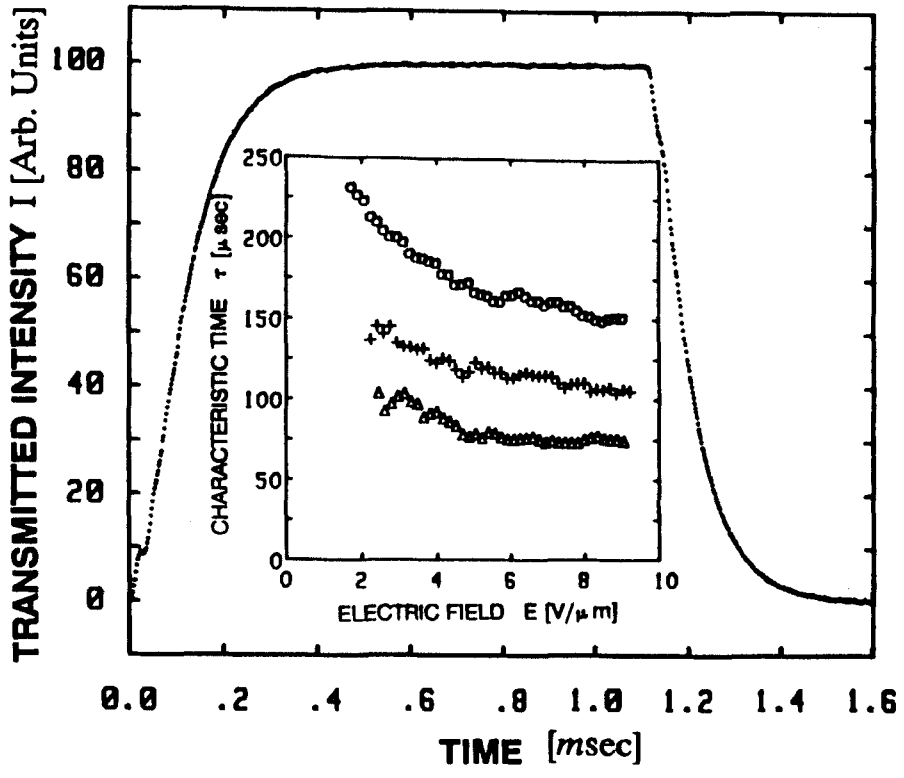

FIGURE 1 A normalized optical modulation and the characteristic time τ as a function of the applied electric field E. The circle, cross, and triangle symbols present T = 24°C, 33°C, and 48°C, respectively.

dynamic response to an applied square wave of frequency 400 Hz at three different temperatures. As shown in Figure 1, the dynamic optical response decays (or grows) exponentially in time. The rise (or fall) time was obtained by fitting the modulation of the transmitted intensity to a form of exponential growth (or decay) at various electric fields. τ is strongly dependent on the applied field at relatively low fields. For a sample that has only one buffed surface, it was found that τ remains nearly constant in the whole range of the field we studied. This suggests that both the surface anchorage and the resultant molecular orientations at surfaces predominantly govern the dynamic properties of the flexoelectric effect. Furthermore, the flexoelectric phenomena can be enhanced when the electric field is highly inhomogeneous near the surfaces as is the case of the boundary layer in a thin cell made with both buffed surfaces. The measured value of τ is on the order of 100 μsec which agrees reasonably well with the predicted value of about 10 to 100 μsec, based on the literature value for $\gamma_1 \approx \eta \approx 0.1$ to 1 poise,[12] together with the values of K_1 and t_o used before.

It is important to determine the temperature dependence of the flexoelectric response to an applied field from the practical point of view. To parameterize the temperature dependence of τ which may result mainly from the viscosity η, we write τ as $\tau_o \exp(B/k_B T)$, where k_B denotes the Boltzmann constant and B represents an effective activation energy associated with τ. Here τ_o is assumed a temperature-independent constant. Therefore, the temperature dependence essentially follows the Arrhenius type behavior. The typical value of τ_o for the material we studied is about 40 μsec, and the corresponding activation energy $B \approx 0.04$ eV, which is rather small compared to that for the rotational viscosity γ_1 of conventional liquid crystals such as MBBA.[15] However, this is not surprising because the temperature dependences of both K_1 and t_o also should be taken into account. The actual value of the activation energy associated with the viscosity η might be of the order of 0.1 eV, comparable to that for γ_1.

In summary, we have demonstrated that the flexoelectric effect in cholesteric liquid crystals leads to a fast linear electro-optic modulation of light. The rotation of the optic axis is linear up to the critical field at which the helical structure disappears. The speed of the electro-optic response is on the order of 100 μsec, and is fairly independent of the field except for the low field regime. Particularly, the sample with only one surface buffed exhibited almost no field-dependence even at low fields. Thus, for practical purposes, the switching time may be considered to be field-independent. The temperature dependence of the associated characteristic time essentially follows the Arrhenius type behavior. From the fundamental point of view, it would be very important to distinguish the relative role of bulk to surface on the observed phenomena due to the flexoelectric effect. The effect described here should be useful for electro-optic devices, particularly for signal processing and optical computing.

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