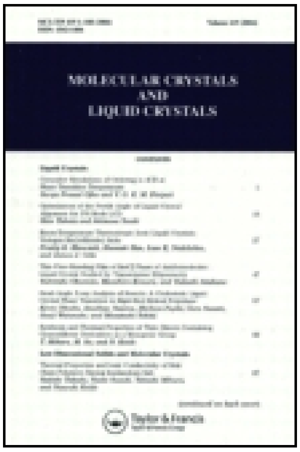

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Field-Induced Biaxiality in the Refractive Index of Chiral Smectic a Liquid Crystals

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FIELD-INDUCED BIAXIALITY IN THE REFRACTIVE INDEX OF CHIRAL SMECTIC A LIQUID CRYSTALS

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ABSTRACT The transmission of a homogeneously aligned chiral smectic A liquid crystal, placed between crossed and parallel polarizers, is measured as a function of polarization angle for a series of applied voltages. The experimental results exhibit a pronounced field-dependent birefringence, providing clear evidence for field-induced biaxiality in the refractive index in these systems. It is shown that the biaxiality is strongly correlated with the electroclinic tilt angle.

INTRODUCTION

Chiral smectic A liquid crystals are important electro-optical materials for applications which require fast analog phase modulation. The physical properties which govern device performance depend strongly on the degree of molecular ordering. In the absence of an electric field, the chiral smectic A phase is uniaxial with the long molecular axis aligned along the smectic layer normal, and the molecules are free to rotate about their long axis. When an electric field is applied parallel to the smectic layers, the transverse component of the permanent molecular dipole tends to align with the field, and a molecular tilt is induced in a plane perpendicular to the field. Known as the electroclinic effect,[1] this induced tilt should be accompanied by field-induced biaxiality,[2] as both effects arise from the action of the electric field on the dipole moment. Although there have been reports of field-induced dielectric biaxiality[3] for Sm-A* liquid crystals, the optical analog of this effect, its field dependence and relation to electroclinic tilt have yet to be investigated.

We report in this paper the observation of field-induced biaxiality in a chiral smectic-A phase. By measuring the optical transmission as a function of polarization angle and electric field, it is shown that a material exhibiting a large electroclinic effect exhibits a pronounced optical biaxiality. It is also shown that the biaxiality is strongly correlated with the electroclinic tilt.

EXPERIMENTAL

The liquid crystal studied here, designated as KN125, possesses the following phase sequence: crystal - (33°C) - chiral Sm A - (78°C) - isotropic. The sample supercools and remains in the chiral smectic-A phase at ambient temperature for prolonged periods of time. Its molecular structure has been discussed elsewhere.[4] The cells for investigation of the LC properties come from E. H. C. Co. Ltd. and are similar to those described in an earlier work.[4] Indium-Tin Oxide (ITO) coated glass substrates are overcoated with an alignment layer consisting of rubbed polyimide. Mylar spacers control the cell thickness and provide insulating barriers. The cell thickness d was measured spectroscopically using interference fringe maxima and minima. The cells were filled through capillary action, mounted in a temperature controller and mounted on a translation stage. Bipolar, square wave electric fields were applied through leads soldered to the ITO and were synchronous with the optical chopping frequency.

The homogeneously aligned chiral smectic-A liquid crystal is in the bookshelf geometry. An electric field applied normal to the cell windows causes the molecular director to rotate through an angle θ (the electroclinic tilt angle) in a plane perpendicular to the electric field. We define a tilted coordinate system (X_L, Y_L, Z_L), where the Z_L-axis lies along the molecular director, the Y_L-axis is normal to the cell windows and parallel to the electric field, and X_L is orthogonal to Y_L and Z_L.

The transmission of an optical beam propagating along Y_L is measured as a function of light polarization angle for a series of applied voltages with the sample between either crossed or parallel polarizers. For crossed polarization the transmitted signal is given by[5]

$$I_\perp = I(0)\,\sin^2(2\alpha)\,\sin^2(\psi/2)\,. \tag{1}$$

Here α is the angle between the polarization vector and the molecular director, $\psi = 2\pi\Delta n d/\lambda$ is the phase angle, and the birefringence, $\Delta n \equiv n_{Z_L} - n_{X_L}$, is the difference in refractive indices for light polarized along the Z_L and X_L axes. The variation of the electroclinic tilt angle θ (and hence *α*) with electric field causes a shift in the angular position of the maxima and minima. Any change in the value of the transmission maximum would indicate a field dependent birefringence Δn.

RESULTS AND DISCUSSION

Figure 1 shows the systematic variation of the measured transmission as a function of applied field and angular orientation of the sample relative to that of the polarizers. Two prominent features should be noted: (i) the variation in the angular positions of the transmission minima and maxima, from which $\theta(E)$ is determined, and (ii) the change in the amplitude of the transmission maximum. This indicates a field-dependent Δn, and is to our knowledge the first observation of field dependent birefringence in smectic-A* liquid crystals.

Equation 1 was used to fit the experimental transmission data, yielding Δn as a function of electric field. Figure 2 shows the field-dependence of $\delta\Delta n(E) \equiv \Delta n(E) - \Delta n(0)$

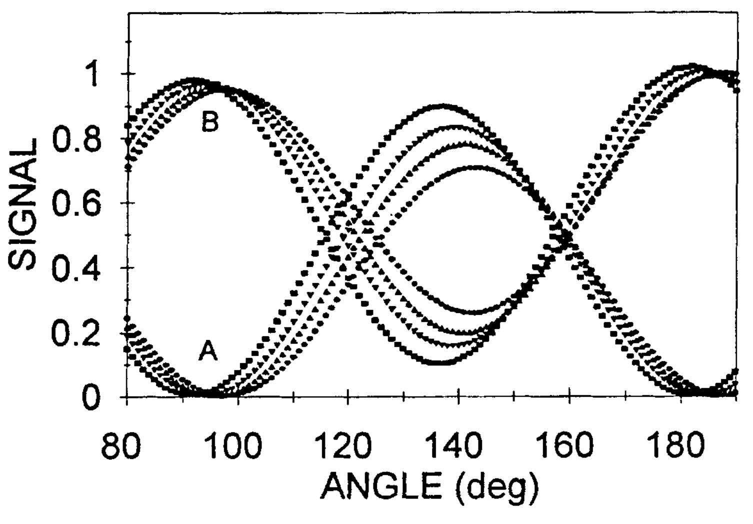

Fig. 1. Crossed- (A) and parallel-polarized (B) transmission as a function of polarization angle, for electric fields of 0.8V/μm (squares), 1.6V/μm (inverted triangles), 2.7V/μm (triangles), 4.3V/μm (circles) for KN125 at 300K.

for a 19 μm thick sample. The experimental $\delta\Delta n$ at the highest fields are roughly several percent of Δn, which varies from 0.14 to 0.16 within the visible region. The similarity in the behavior of $\delta\Delta n$ and optical tilt angle, θ,[4] strongly suggests that the field dependence of Δn arises from molecular reorientation associated with the electroclinic effect. The variation in Δn could in principle be related to either a change in n_{X_L}, arising due to field-induced biaxiality, or a change in n_{Z_L}. The latter could arise if the field were to deform the liquid crystal, causing the long molecular axis to tilt out of the X_L-Z_L plane. This would lead to a decrease in n_{Z_L} rather the observed increase. On the other hand, an increase in Δn is what would be expected for field-induced biaxiality. The larger of the two minor axes of the molecular polarizability ellipsoid should be along the transverse component of the dipole moment. The electric field tends to align the larger of the two axes in the Y_L direction, causing n_{X_L} to decrease and Δn to increase. It is clear therefore that the observed increase in Δn for this surface stabilized smectic-A* liquid crystal arises from field-induced biaxiality.

When no field is applied, the molecules rotate freely and the liquid crystal is optically uniaxial with $n_{X_L}^2 = n_{Y_L}^2 = (\epsilon_{xx}+\epsilon_{yy})/2$, where ϵ_{ii} represents the principal axes of the dielectric ellipsoid. With an applied field, the free rotation is hindered and the

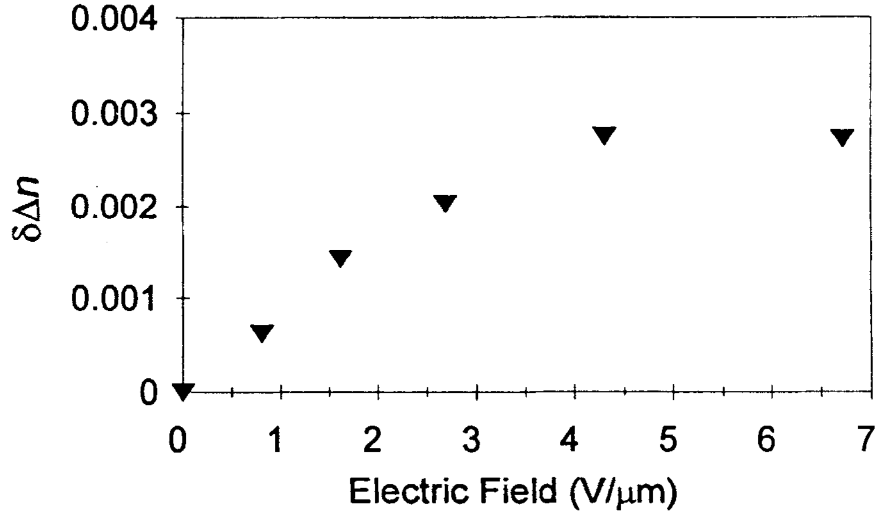

Fig. 2. δΔn as a function of electric field in the smectic-A* phase of a 19μm thick KN125 sample.

system becomes biaxial. In the high-field limit, $n_{X_L}^2 \approx \epsilon_{xx}$.

In figure 3, $\delta\Delta n$ is plotted as a function of the tilt angle θ for KN125. The solid curve is a guide to the eye, illustrating a quadratic dependence of δn on tilt angle. Galerne[6] has observed a similar correlation between refractive index biaxiality and the tilt angle in the smectic-C phase. The biaxiality measured in that work for the smectic-C phase is of the same order of magnitude as that reported here for KN125 in the chiral smectic A* phase. A quadratic dependence of δn on θ may be predicted from symmetry arguments since $\Delta n(\theta)=\Delta n(-\theta)$, yielding $\delta\Delta n \propto \theta^2$ in the low field limit. However, the relationship between δn and θ is more general and should hold for a wide range of fields, since the electroclinic tilt as well as the field-induced biaxiality result from the force exerted by the field on the dipole moment.

Finally, it should be emphasized that the primary effect caused by the electric field acting on Sm-A* liquid crystals is the reduction of the azimuthal angle degeneracy. This induces tilt as well as biaxiality, removing two important characteristics that differentiate the Sm-A* and Sm-C* phases. It is shown here that the effect of this induced biaxiality on the optical properties of the Smectic-A* phase can be quite signifi-

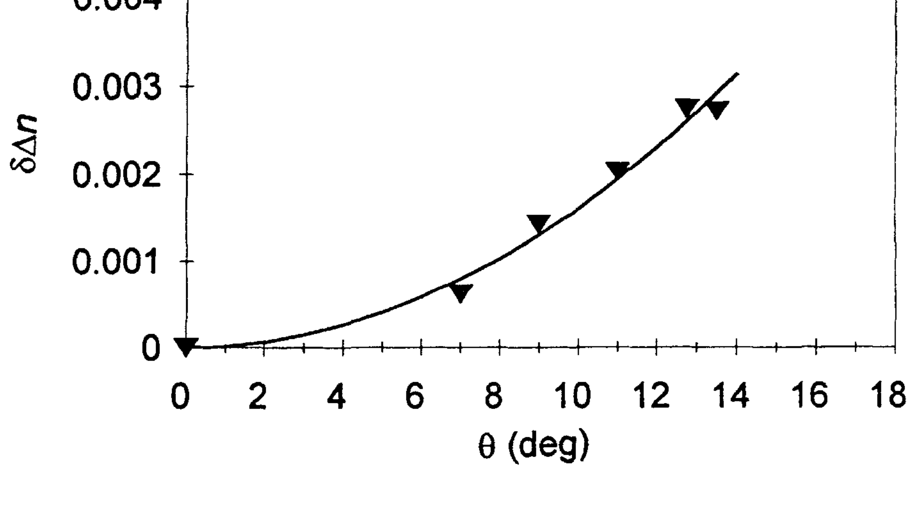

Fig. 3. δΔn as a function of θ in the smectic-A* phase of a 19μm thick KN125 sample.

cant, and that a full characterization of the physical state of this system requires both angular coordinates to be specified. In light of this, a more general model describing the electro-optic properties of the Sm-A* phase, that takes into account all three principal refractive indices, is needed. Our results also show a strong correlation between the electroclinic tilt angle and biaxiality.

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