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Surface electroclinic effect in ferroelectric liquid crystal

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Abstract. — Surface electroclinic effect in chiral nematic (N*) phase of high-tilt-angle ferroelectric liquid crystal has been studied by dielectric relaxation and electro-optical method in a homogeneously aligned, thin samples in the frequency range of 400 Hz to 100 kHz at different temperatures. The electroclinic effect in the N* phase observed in the surface layers is due to the effect of surface anchoring and smectic-like ordering of the molecules at the rubbed boundary surface of the sample which has been confirmed by dielectric and optical observations. The strong anchoring in the surface layers is also presented by applying a dc bias field.

1. Introduction.

The electroclinic effect near Smectic A-Smectic C* (Sm A-Sm C*) phase transition is well-known [1] in the ferroelectric liquid crystals (FLCs). A large number of data has been reported in Sm A and in Sm C* phases in homogeneously aligned thin cells from the optical observations [2] and dielectric measurements [3, 4]. The electroclinic effect in the chiral nematic (N*) phase of FLCs have been scarcely studied [5, 6]. The electroclinic effect occurs when an electric field applied parallel to the smectic layers induces a molecular tilt relative to the layer normal.

Alignment of ferroelectric liquid crystal molecules is generally achieved by properly treating the surfaces of the two substrates in the fabrication of FLC cells or devices. Berreman [7] first studied this problem by rubbing the glass substrates with diamond paste and creating microgrooves. He calculated the elastic distortion due to the grooves and found that the lowest energy configuration was for all the molecules to lie along the grooves, creating an uniformly aligned cell along the rubbing direction. Geary, Goodby, Kmetz and Patel [8] have proposed a different mechanism of the alignment of liquid crystal molecules on the polymer coated substrates rubbed with cloth. Rubbing of the films orients the polymer chains along a preferred direction and the molecular interaction between the liquid crystal (LC) molecules and the stretched polymer induces the alignment. It has been found [9] that alignment is either due to grooves formation or interaction between the LC molecules and the stretched polymer. The surface anchoring energy in thin cells is strongly dependent on the rubbing strength. Several surface treatments have been successfully employed, but the physical mechanisms affecting
the surface induced bulk alignment are still not well understood. For homogeneous bulk alignment the specially treated surfaces often used are rubbed polymer coated substrates. As a surface induced effect, how the first monolayer of LC molecules at the interface is oriented by the surface molecular interaction is fundamental to our understanding of homogeneous alignment.

Different techniques [9-11] have been employed to study the surface induced alignment due to the rubbing of the polymer coated substrates in ferroelectric liquid crystals. In this article, we report the electroclinic effect due to the interaction of LC molecules in cholesteric phase (N*) with the polyamide treated rubbed substrates in thin cells of high tilt angle ferroelectric liquid crystals by dielectric and electro-optical observations.

2. Experimental.

Highly conducting indium tin oxide (ITO) glass plates were used for electrodes. The distance between the plates was kept around 10 μm and this distance can be considered as thin compared with the helical pitch value (15 μm in Sm C* and 23 μm in N* phase) of the material. The high tilt angle (44°) ferroelectric liquid crystal material CS-2004 (Chisso Corporation, Japan) used in this study has the following phase sequence:

Cryst $\rightleftharpoons$ Sm-C* $\rightleftharpoons$ N* $\rightleftharpoons$ Iso

$-9^\circ C$ $62^\circ C$ $71^\circ C$

The material was introduced in the cell by means of capillary action at elevated temperatures to ensure that the filling took place in its isotropic (Iso) phase. Homogeneous alignment of the FLC sample is obtained with one surface buffered, which has been pretreated with adhesion promoter and nylon and a well defined monodomain sample was obtained by applying an ac field. A detailed procedure of the alignment and the dielectric studies in the Sm C* phase of this material has been reported elsewhere [12]. The smectic layers are assumed to be perpendicular to the plane of the electrodes for this geometry. Electric field is applied parallel to the smectic layers.

The dielectric measurements have been carried out using a HP 4192A Impedance Analyzer in the frequency range 400 Hz to 100 kHz. The dielectric relaxation in N* phase at different temperatures has been studied by applying a dc electric field (±) 0 to 30 V/10 μm. The electroclinic effect in N* phase has been further, analyzed by observing under polarizing microscope (Olympus BH-2).

3. Results and discussion.

The electroclinic effect in N* phase has been analysed by dielectric relaxation method. This mode is most commonly observed in Sm A phase as a soft mode (which corresponds to an electroclinic effect). Figure 1 shows the tan δ (loss factor) versus log $\nu$ plot at different temperatures in N* phase. The critical frequencies of this dielectric mode have been computed from the position of the maxima on the plot which is almost constant with temperature. As described earlier one of the glass plates used in the cell is coated with polymer and rubbed whereas other plate is without any surface treatment for aligning the FLC molecules. This means that the surface which is rubbed with polymer is highly anchored [13] and other surface is weakly anchored. A high dc electric field is applied to unwind the twisted structure to see whether the dielectric mode in N* phase comes due to surface or bulk effect of the cell. Figure 2 shows tan δ as a function of frequency at different applied dc bias field in N* phase.

The dielectric relaxation in N* phase in this study is not due to flexoelectric effect which appears on weak anchoring surfaces or due to the breaking of the cell symmetry (more than one
Fig. 1. — Frequency dependence of $\tan \delta$ (loss factor) measured at different temperatures in $N^*$ phase.

Fig. 2. — Frequency dependence of $\tan \delta$ measured at different biasing voltages at $69 \, ^\circ C$ in $N^*$ phase.

domain) [14] in $N^*$ phase. Recently Lee and Patel [15] have observed the flexoelectric effect in nematic phase where the difference between anchoring strength on the two surfaces is very small. In the present investigations the possibility of flexoelectric effect is ruled out because i) the anchoring energy in polymer rubbed surface is very strong [13], ii) the alignment is uniform and a single domain is checked under polarizing microscope. The dielectric relaxation
in N* phase (Fig. 1) appears due to strong anchoring effect on the polymer rubbed surface which preserves the smectic-like ordering near the rubbed surface layers. As it is known [16, 17] that the FLC material, which has a Sm C* phase after the Sm A phase is likely to form a firm layer structure in the Sm C* phase. Contrary to this situation, the FLC material that has Sm-C* phase without passing through the Sm A phase would not form a firm layer structure and consequently the direction of the molecules near the surface is anchored rather tightly due to the nature of rubbed films (grooved formation) and this anchoring effect would be more pronounced in thin samples. The electroclinic effect arises from interaction of the liquid crystal molecules with the local electric field at the rubbed boundary surfaces in N* phase. This local field originates from the polar anchoring of the few liquid crystal monolayers on the rubbed surface [10]. The anchoring effect at the rubbed surface is so strong that even at the dc bias field of 10 V/10 μm the electroclinic effect in N* phase was observed (Fig. 2). This means that a high bias field is needed to nullify the anchoring effect on the rubbed surface so that the surface and bulk molecules behave identically. However, above this biasing field (above 10 V/10 μm in Fig. 2) the dielectric losses or conductance due to electroclinic effect was not consistent in N* phase which is shown by the error bars in the figure and hence at higher biasing field the dielectric relaxation frequency could not be computed.

The smectic-like ordering in N* phase is confirmed due to the fact that the electroclinic effect was observed in N* phase during the heating cycle (Sm C*-N*-Iso). Whereas in the cooling cycle (Iso-N*-Sm C*) this effect was not observed. This means that when the phase transition is from Iso-N*-Sm C* then the LC molecules in N* phase are not aligned along the rubbing direction resulting into the randomly lying (isotropically) the molecules on the surface and therefore, there would not be any surface anchoring of the LC molecules on rubbed surface. When it is further cooled in Sm C* phase the layer structure also disappears (multidomain cell) which has been confirmed by optical microscopic observations. Recently Lee, Patel and Goodby [18] have also reported a surface electroclinic phenomenon in the isotropic phase of an FLC on a rubbed polymer surface and interpreted as an electric field induced molecular tilt in a chiral liquid crystal which has a direct Iso-Sm A transition. They have found that the molecular tilt exists in about 1 °C above the transition to the bulk isotropic phase and predicted smectic-like ordering within the surface layer which contributes to the electroclinic effect.

Further, the electroclinic effect in the present investigation has been confirmed by electrical switching method by inducing a tilt angle in molecules by applying a high electric field in N* phase. Figure 3 shows the change in the texture of the sample at different applied static dc voltages. At zero voltage the surface molecules and bulk molecules are in the same phase. Therefore, uniform texture is seen under the microscope (Fig. 3A). The increase in the dc voltage which gives rise to an induced tilt of the surface molecules with respect to the bulk molecules, results into the change in the texture of the sample (Figs. 3B, C, D and E). However, if a square pulse is applied at low frequency a complete switching in the sample is observed which is due to the fact that surface molecules and bulk molecules are in unison. This means that the switching starts at the rubbed surface molecules and percolate in the bulk resulting into the complete switching in the N* phase. Figure 4 shows optical micrograph by applying a positive or negative bias voltage to the rubbed surface. When 0 to +10 V bias is applied to rubbed surface a smectic-like ordering is clearly seen (Fig. 4A) which is due to the fact that the polynamide surface acts like positively charged surface, as suggested by Patel and Goodby [19]. When negative bias is applied cholesteric type texture is seen (Fig. 4B). The switching response and polarization reversal current due to electroclinic effect in N* phase is also shown in figure 5 by applying a square and triangular pulses at 45 Hz frequency. It should be stressed here that in the whole temperature range the surface electroclinic effect was
Fig. 3. — Microphotographs of 10 μm cell in N* phase at 69 °C at different applied dc voltages (A) $v = 0$ V, (B) $v = 8$ V, (C) $v = 12$ V, (D) $v = 17$ V and (E) $v = 24$ V.

Fig. 4. — Microphotograph in N* phase at 69 °C by applying either positive or negative voltage to the rubbed plate. (A) 0 to +10 V and (B) 0 to −10 V.
observed in N* phase for the first time, even in the blue phase which is observed just below the isotropic phase, in the FLC material possessing Sm C*-N*.-Iso phase transition.

The electroclinic effect appears due to the strong polar anchoring [10] and smectic-like ordering [18] of the molecules within the surface layers of polymer rubbed samples in N* phase. One should expect a spontaneous two-dimensional electric polarization which resides very close to the surface and is oriented perpendicular to the director. In order to probe the electroclinic effect in terms of polarization, an ac electric field ($E$) of frequency $\nu$ is applied perpendicular to the director, thereby inducing a small rotation of the surface molecules. This surface-driven motion then propagates elastically into the samples interior by a combination of bend and twist elasticity resulting into an overall motion of the bulk samples which is observed in terms of polarization and response current in the electroclinic geometry.

As is seen in the figures 1 and 2 the electroclinic effect observed by dielectric relaxation in N* phase of high tilt angle FLC material is weak due to the fact that the pitch value in N* phase is quite large and also the tilt angle and polarization value in the Sm C* phase of this material is almost independent of temperature. These temperature independent values of tilt angle and polarization are retained within the surface layers of the rubbed surface even in N* phase. This has been confirmed by optical observations (Figs. 3 and 4). It is worth mentioning here that in a thick sample (25 $\mu$m) a very weak electroclinic effect was observed very close to the transition temperature in N* phase which indicates that the electroclinic effect due to surface layers is dominant in this samples. Similar results have been observed in [5, 16, 17] by optical method using thin planar unwound samples. Large electroclinic effect has recently been observed [6] in the FLC material where the helical pitch value of N* phase is quite small (0.5 $\mu$m).

In summary, surface electroclinic effect induced in a polyamide coated rubbed surfaces was observed by dielectric relaxation and electrical switching method in N* phase of high tilt angle ferroelectric liquid crystal which has a phase sequence of Sm-C*-N*-Iso in a thin sample. It is concluded that the surface electroclinic effect in N* phase is due to the strong anchoring and smectic-like ordering of the molecules within the surface layers of rubbed samples.

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