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Surface Effect on Goldstone Mode in Ferroelectric Liquid Crystals

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Dielectric studies of a single component ferroelectric liquid crystal material, showing chiral nematic to smectic C* phase transition have been investigated using a thin, highly conducting electrode cells. The main emphasis was given to the Goldstone mode which comes due to phase fluctuations in tilt angle of the molecules in Sm C* phase. To see the surface effect on the behaviour of Goldstone mode, one of the electrodes of the cell was highly anchored. The dielectric permittivity, particularly due to Goldstone mode has been studied for positive and negative polarity of the bias field in Sm C* phase. The Goldstone mode behaves differently during the positive and negative polarity of the bias field. The unusual dielectric behaviour has been explained by considering the charge accumulation phenomenon. The results agree with the electro-optical studies.

Keywords: Ferroelectric liquid crystals; Goldstone mode; charge accumulation

INTRODUCTION

Ferroelectric liquid crystals (FLCs) have been studied extensively due to their interesting basic properties and appear to be very promising materials for the optical displays[1]. However, they are not widely used in devices because

there are number of difficulties in their applications, including the understanding and control of alignment and switching mechanism. The use of polymer rubbed surfaces for the FLC alignment is of particular interest because of its feasibility for preparing the large area and mass production devices although, the polymer rubbed alignment creates high polar surface anchoring effect which affects the switching mechanism of the liquid crystal molecules. Similarly, the dielectric relaxation process in ferroelectric liquid crystals can be expected to be influenced by the surface anchoring effect which is created due to the charge accumulation at the interface of polymer layer and the liquid crystal material.

It is well known[2] that the dielectric relaxation in chiral smectic C* phases comes due to the collective dielectric processes and due to the molecular reorientation motion, connected to the polarisation of the molecules. The collective dielectric processes are due to the Goldstone mode, connected to the phase fluctuations in the azimuth orientations of the director and the soft mode due to the fluctuations in the amplitude of tilt angle (Θ). The dielectric permittivity in the whole Sm C* phase is dominated by the Goldstone mode. However, it can be suppressed by unwinding the helicoidal structure either by applying a bias field or by surface stabilisation. The soft mode appears near the transition temperature of Sm C* to Sm A phase as the tilt fluctuations are maximum near the transition temperature. However, soft mode can be detected few degrees in Sm C* phase by suppressing the Goldstone mode[3].

In the present investigations, the behaviour of Goldstone mode has been studied in a highly anchored surfaces of a very thin (2.5 μm) samples in a first order phase transition ferroelectric liquid crystal material having a phase sequence of smectic C* to chiral nematic phase. The dielectric studies have been performed in such samples in the frequency range of 10 Hz to 10 MHz. The influence of positive and negative polarity bias field on the Goldstone mode has been reported in the samples prepared by polymer rubbed surfaces for the alignment of ferroelectric liquid crystal molecules.

EXPERIMENTAL

Transparent conducting indium tin oxide (ITO) coating on optically flat glass plates were used as electrodes for the dielectric measurements and also to cheque the optical alignment of liquid crystal molecules. One of the electrodes of the cell was treated with adhesion promoter and polymer (Nylon 6/6) and was unidirectionally rubbed to get the planar alignment. The charge accumulation effect (surface anchoring) was obtained by thick polymer coating[4] with strong rubbing strength which was done by keeping the minimum distance between the glass plates and the rubbing block. Thickness of the sample cell was maintained around (2.5 μm) by means of photoresist film spacers. The cells were first calibrated using air and toluene as standard references. A single component ferroelectric liquid crystal material used in this investigations (IS-4362 = "FFP", E. Merck, Darmstadt, Germany) has a phase sequence as follows

$$\text{Cryst.} \underset{30^\circ C}{\overset{43.2^\circ C}{\rightleftharpoons}} \text{SmC}^* \overset{45^\circ C}{\longleftrightarrow} \text{N}^* \overset{61^\circ C}{\longleftrightarrow} \text{Iso.}$$

The material was introduced into the cell by means of the capillary action at elevated temperatures. A well aligned sample is obtained by applying an electric field, just below the transition temperature of Sm C* to N* phase as simultaneously observed under the polarising microscope. The detail procedure for planar alignment in such type of ferroelectric liquid crystal material is reported elsewhere[5].

Dielectric measurements were done in a shielded parallel plate capacitor[6] using a HP-4192A impedance analyser in the frequency range of 10 Hz to 10 MHz. The dielectric data were corrected for the static conductivity and for the high frequency deviations caused by the inductance and resistance of the cables and connectors. The frequency and bias field dependence of the real and imaginary part of the complex dielectric permittivity have been studied in Sm C* and N* phase for planar alignment in thin cells. The optical alignment was chequed by mounting the cells on a polarising microscope (Olympus BH_2).

RESULTS AND DISCUSSION

In planar alignment the molecular director is in the plane of the substrate and smectic layer plane is perpendicular to the substrate. In the first order phase transition FLC materials, the chances of formation of chevron structure is very less because of the absence of smectic A phase. Further, planar alignment is obtained in the presence of an ac electric field in the cell where one of the electrodes is treated with polymer and rubbed[7].

As it is known[3] that in planar alignment the dielectric permittivity due to Goldstone mode is dominant in Sm C* phase which comes due to the phase fluctuations of the molecular director along the smectic tilt cone. These fluctuations can be suppressed by unwinding the helical structure either by a bias field or by the surface stabilisation. In the present FLC material, one can expect a Goldstone mode even in the very small thickness (2.5 μm) cells as the FLC material has a very small helical pitch value of 0.3 μm in smectic C* phase. Figure 1 (A&B) shows the effect of biasing field with a positive polarity to the rubbed surface, on the Goldstone mode in the form of

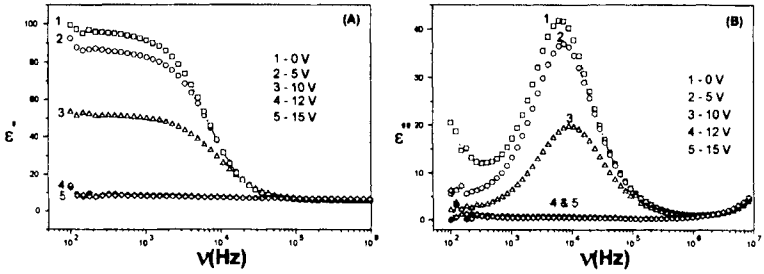

FIGURE 1 (A) Dispersion and (B) absorption curves at 42.09 °C temperature for Goldstone mode at different biasing voltages with positive polarity to the polymer rubbed surface.

dispersion and absorption curves. It is clear from the Figure that as the bias field increases the Goldstone mode also gets suppressed. At about 12 Volts of biasing voltage the dielectric permittivity (ε') becomes static. Similarly the absorption due to Goldstone mode also becomes constant (Figure 1B) at 12 Volts of positive bias field to the rubbed surface of the cell.

Figure 2 (A&B) shows the dispersion and absorption curves due to the Goldstone mode when the bias field with negative polarity to rubbed surface was applied to the cell. Here also the Goldstone mode gets suppressed with the increase in the negative bias field, but it gets suppressed with higher bias voltage (at 15 V) as seen in the dispersion and absorption curves. If one

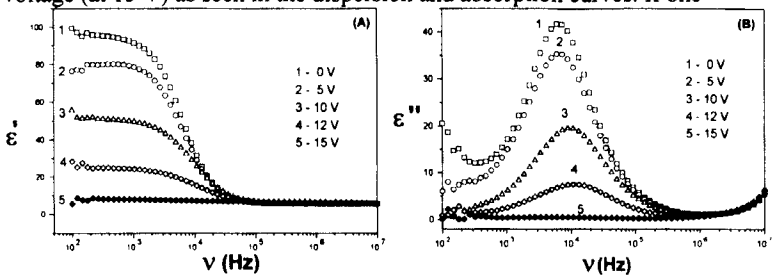

FIGURE 2 (A) Dispersion and (B) absorption curves at 42.09 °C temperature for Goldstone mode at different biasing voltages with negative polarity to the polymer rubbed surface.

analyses carefully Figures 1 & 2, the biasing voltage, required to suppress the Goldstone mode is different with the change in the polarity of the biasing field and this difference can be very clearly seen in Figure 3 at 12 Volts. This means that the Goldstone mode gets suppressed at 12 Volts when a bias field with +ve polarity to the rubbed surface and it (GM) gets suppressed at 15 Volts when a bias field with -ve polarity to the rubbed is applied.

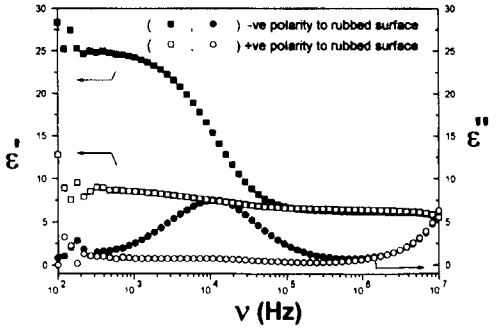

FIGURE 3 Dispersion (closed square) and absorption (closed circles) curves for 12 volts of bias field with negative polarity to the polymer rubbed surface. Dispersion (open squares) and absorption (open circles) curves for 12 Volts of bias field with positive polarity to the polymer rubbed surface at 42.09 °C temperature.

The difference in the biasing voltages with the change in polarity, to suppress the Goldstone mode is due to some internal field in the cell itself. This internal field is because of the charge accumulation between the alignment layer and the FLC material. As only one surface of the cell was polymer coated and rubbed for the alignment of the FLC molecules. Therefore, the field created due to surface charge accumulation would be very high on the polymer coated surface which results into the different biasing voltages with the change in polarity, to suppress the Goldstone mode. It should be mentioned here that the charge accumulation would be very high if a thick polymer coating with a high spontaneous polarisation (P_s) FLC material[8] is used. In the present study also one can expect a high charge accumulation on one surface of the cell as very high P_s (160 nC/cm^2) FLC material and a thick polymer coating (1000 Å) is used. Such behaviour of asymmetric switching[9,10] due to charge accumulation phenomenon has also been observed in such type of FLC materials by electro-optical studies when one electrode of the cell is coated with polymer and rubbed for the liquid crystal alignment.

It should be mentioned here that when the cell is optically well aligned, the behaviour of Goldstone mode with biasing field is not consistent, particularly for lower frequencies as seen in Figure 4. This means that the field formed due to charge accumulation on one of the electrodes is enhanced in the optically well aligned cells and creates hindrance in the phase fluctuations of the tilt angle, resulting into the nonconsistence behaviour of the dielectric permittivity of the Goldstone mode, particularly at lower frequencies. The fluctuations in the dielectric permittivity at lower frequencies is understandable due to the fact that the mobility of the charges is very slow. The charge accumulation phenomenon can be seen very clearly at higher temperatures in chiral nematic phase as shown in Figure 5. One may argue that why the charge accumulation dielectric process is seen in N* phase as there is no local spontaneous polarization in this phase. This is because of the fact that smectic-like ordering is retained in the surface layers, resulting into the appearance of low frequency dielectric process in chiral nematic

phase. The smectic-like ordering in the surfce layers has also been observed in N* phase by electro-optical method[11,12] in this type of FLC materials. However, for lower temperatures in smectic C* phase it could not be seen very clearly as the relaxation process appears to shift to lower frequency (curves a&b in Fig. 5).

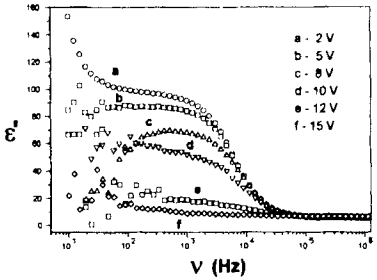

FIGURE 4 Dielectric permittivity (ε') as a function of frequency at 38.26°C temperature in optically well defined cell.

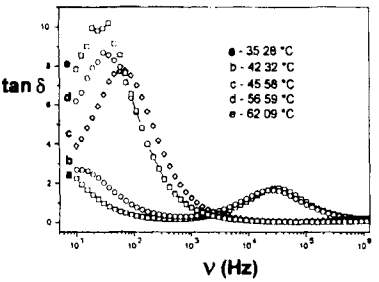

FIGURE 5 Loss factor (tan δ) versus frequency at different temperatures in SmC* and chiral nematic phase in optically well aligned cell.

This slow frequency dielectric process cannot be due to the Goldstone mode in chiral nematic phase and it is not due to the soft mode which appears at high frequency (100 kHz). This mode also cannot be due to the ionic impurity effect[13] because this process could not be seen when this material was aligned by magnetic field[14]. So, this slow frequency dielectric process, seen in the chiral nematic phase is due to the charge accumulation phenomenon between the polymer rubbed surface and the ferroelectric liquid crystal material. The dielectric measurements for this slow frequency relaxation due to charge accumulation phenomenon are being carried out at lower temperature in smectic C* phase in the frequency range of 0.01 Hz to 10 kHz by 1250 frequency response analyser (Schlumberger, Solartron) and would be published in detail.

CONCLUSIONS

1. The Goldstone mode suppression behaves differently during the positive and negative bias field when the surface charge accumulation is created on one of the electrode cell.

2. The slow frequency dielectric process is detected at higher temperatures in chiral nematic phase due to the charge accumulation phenomenon.

3. It would be interesting to study the surface charge accumulation effect by dielectric method in smectic C* phase for lower frequencies (below 10 Hz).

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