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Low Frequency Dielectric Response in the Smectic C* Phase of a FLC Mixture

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LOW FREQUENCY DIELECTRIC RESPONSE IN THE SMECTIC C* PHASE OF A FLC MIXTURE.

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Abstract Experimental studies of the dielectric response of a ferroelectric liquid crystal (FLC) mixture having low spontaneous polarization at different temperature and frequency have been done in smectic C* (Sm-C*) phase. The dielectric measurement were carried out on homogeneously aligned samples. In the Sm-C* phase the temperature dependent dielectric contribution of the Goldstone mode has been measured using two cells of thicknesses of 20μm and 100 μm. Dielectric strength and corresponding relaxation frequencies are determined. From the experimental results it has been observed that the variation of dielectric strength and relaxation frequency with temperature are similar for the two cell, but the dielectric strength obtained for 20μm cell is higher than that obtained for 100μm cell at all temperatures. On the other hand, the temperature dependence of relaxation frequency for both 20μm and 100μm cell is almost the same. The mean field coupling coefficients between tilt and polarization are obtained from a fit of extended microscopic model for the interpretation of experimental results. The Goldstone mode rotational viscosity has been calculated from dielectric data. The results have been discussed.

INTRODUCTION

Since date a vast number of papers dealing with the experimental and theoretical aspect of the dielectric properties of chiral smectic-C* phase of ferroelectric liquid crystal molecules have been published[1-7]. It is shown that the dielectric response of this system consists of four modes apart from electronic contribution. Two of these are associated with the director fluctuation and have characteristic relaxation frequencies around several Hz to a few KHz range. The other two are associated with fluctuation of the polarization order parameter and their characteristic frequency is around 500MHz. One of the director mode is Goldstone mode appears in the C* phase, due to the phase fluctuation of the director. The other director mode is known as the soft mode. The soft mode appears in the vicinity of the transition temperature due to the amplitude of the tilt fluctuation of the director. Majority of the published works are related to the frequency, temperature and bias dependence of the Goldstone mode. A few dielectric studies in the Sm-C* phase are also found at different thickness of the cell[8,9]. Recently Vij et. al[10]. theoretically explained the dynamics of surface stabilized ferroelectric liquid crystal (SSFLC) cells of various thickness. The rotational viscosity is an important physical parameter that influences the switching time in Sm-C^* phase. The rotational viscosity in Sm-C^* was determined by optical method[11] in a thin cell. The rotational viscosity of the Goldstone mode was also measured by Levstik et al. from the dielectric measurement[12] in a thick sample.

In this paper we have reported the low frequency dielectric relaxation studies using 20μm and 100μm thick cells on a ferroelectric liquid crystal mixture, Merck ZLI-4237-100, obtained from E. Merck. The rotational viscosity has also been determined from dielectric measurement. The sample possesses low spontaneous polarization (=-20 nC cm^{-2} at 20°C). The phase sequences of the sample are as follows (Ch denotes cholesteric, I isotropic, and K crystal)

$$K \xleftrightarrow{-20^0C} Sm\,C^* \xleftrightarrow{61^0C} Sm\,A \xleftrightarrow{73^0C} Ch \xleftrightarrow{83^0C} I$$

EXPERIMENT

The ferroelectric liquid crystal samples were held between two conducting glass plates whose surfaces had been treated properly for obtaining planar alignment (soaked with IPA and rubbing). We have used two different pairs of cells, made of indium-tin oxide coated glass plates having a separation of 20μm and of 100μm. For alignment the sample was heated to its isotropic phase by Mettler Hot Stage (model FP 5) and then cooled down very slowly (0.2°C/min.) and simultaneously a low frequency (20Hz) electric field is applied to the cell. The temperature of the sample was stabilized to an accuracy of ±0.01°C. A Hewlett-Packard impedance analyzer HP 4192A working in the frequency range of 5Hz to 13MHz was used for the complex dielectric permittivity measurements. Before putting the sample into the cells, the air capacitance for the two cells were recorded at different temperatures and frequencies. A measuring voltage of .5 Volt was applied in a direction parallel to the smectic layers. The stray capacitance for each cell was measured by using spec-pure benzene.

THEORETICAL BACKGROUNDS

A. Microscopic model and Landau Coupling Coefficients

Recently meister and stegemeyer[13] presented an extended microscopic model of the spontaneous polarization in FLC as follows.

$$P=(A_1/T)\theta+(A_1A_2/T^2)\theta^3 \qquad (1)$$

Both A_1 and A_2 are time independent coupling constant. A_1 related to dipolar ordering and A_2 related to quardrupolar ordering. These parameters are also related to Landau Mean Field Coupling Coefficients (C, Ω) by equation (2) and (3)

$C=(1/\chi_0T)A_1$ (2) and $\Omega=(1/\chi_0T)A_2$ (3)

where C and Ω are the bilinear and biquadratic coupling coefficients respectively of Landau Free Energy expansion. χ_0 is the high frequency dielectric susceptibility.

B. Goldstone mode in the bulk cell and surface stabilized FLC

The dielectric response of the Goldstone mode in a bulk cell is associated with small deformation of helical structure following an application of low electric field so that there is no complete unwinding of the helix. In that case the dynamics distribution of azhimuthal angle φ of the director is along Z axis, pointing towards the normal to the layer.[3,4] This process referred to as Z mode. But in surface stabilized FLC cell, the director orientation is independent of Y and Z axes.[10] The dynamic distribution of azhimuthal angle φ with respect to X axis, pointing towards cell surface. This is referred to as X mode.

RESULTS AND DISCUSSION

In a planer aligned cell when the measuring field is parallel to the layer plane in Sm-C* phase, the main contribution to the polarization comes from the Goldstone mode process due to huge phase fluctuation. By plotting dielectric loss (ε'') against dielectric permittivity (ε') at different temperatures in a Cole Cole[14] diagram, the relaxation frequency (f_G) as well as the dielectric strength ($\Delta\varepsilon_G$) have been determined.

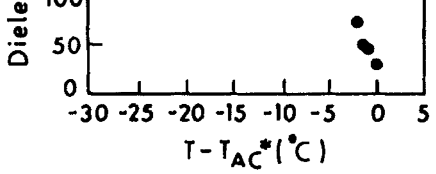

FIGURE 1 Temperature dependence of Goldstone mode dielectric strength ($\Delta\varepsilon_G$).

Fig 1. show the nature of the variation of Goldstone mode dielectric strength as a function of temperature in 20μm and 100μm cells and Fig. 2, showing the variation of relaxation frequency with temperature for 20μm and 100μm cells. As is shown in Fig. 1 that the nature of variation of $\Delta\varepsilon_G$ for the two cells with temperature is almost the same, but $\Delta\varepsilon_G$ for 20μm cell is more than approximately two times as that for 100μm cell. The theoretical relation[12] for Goldstone mode dielectric strength $\Delta\varepsilon_G$ and relaxation frequency (f_G) that have been derived from Landau equation is

$$\Delta\varepsilon_G = 1/2K_3\,[P_s/q\theta_0]^2 \quad (4) \qquad f_G = \Gamma K_3/2\pi.q^2 \quad (5)$$

where P_S and θ_0 is the spontaneous polarization and tilt angle respectively and q is the wave vector of pitch. K_3 is the twist elastic constant. Γ is the inverse of rotational viscosity. P_S/θ_0 has been determined experimentally as shown in Fig.3. The pitch of the

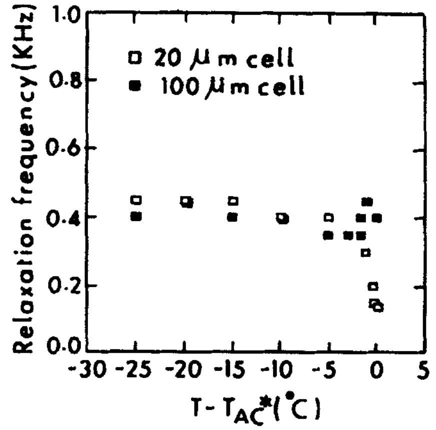

FIGURE 2 Variation of relaxation frequency (f_G) of Goldstone mode with temperature.

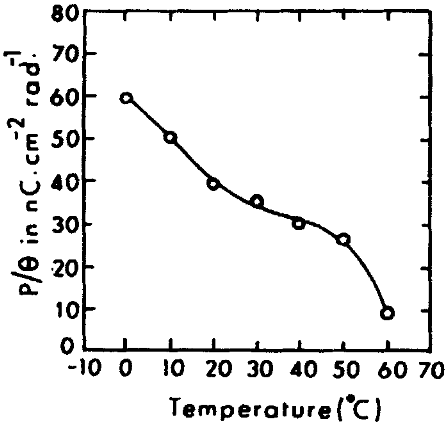

FIGURE 3 Variation of P_S/θ in the smectic C^* phase with temperature.

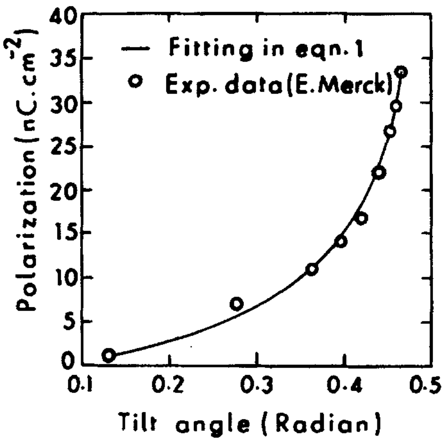

FIGURE 4 Spontaneous polarization (P_S) as a function of tilt angle (θ).

studied FLC mixture is 10μm and assuming a slow variation of pitch with temperature, except near the transition temperature, the measured dielectric strength $\Delta\varepsilon_G$ as shown in Fig.1 agrees qualitatively with what is expected from the above equation, a sharp increase of the $\Delta\varepsilon_G$ near the transition temperature is due to the sharp increase of pitch near transition temperature. The exact nature of the curve of $\Delta\varepsilon_G$ also depends on the relative importance of the biquadratic coupling between tilt and polarization of the molecule.[15] The bilinear and biquadratic coupling have been determined from equations 2 & 3. Two coupling constants A_1 and A_2, related to dipolar and quadrapolar ordering respectively have been determined by fittings (Fig. 4) the data in equation 1. χ_0 has taken from our data.[16] The values of A_1, A_2, C and Ω along with other reported data for FLC 6430[17] and DOMBAMBC[18] included in a table 1.

TABLE I Physical data for some FLC mixtures.

Compound	A_1 (CKm^{-2})	A_2 (K)	C (Vm^{-1})	Ω (VmC^{-1})	χ_0
ZLI4237-100	.0178	10518	1.07×10^6 at 300K	637×10^9 at 300K	5.5
FLC6430	.322	542	12.9×10^6 at 298K	22×10^9 at 298K	8.4
DOBAMBC			2.8×10^6	570×10^9	2.6

The values C and Ω are of the same order of magnitude as that obtained at for FLC6430 and DOBAMBC molecules reported by Zugeumaier et. al. By comparing the A_1 and A_2, C and Ω values of the presently studied compound with those of DOBAMBC and FLC6430. We can say that biquadratic coupling is relatively high in our FLC mixture. A relatively high biquadratic coupling can also be suggested by comparing the experimentally determined $\Delta\varepsilon_G$ (Fig.1) with those of numerically calculated $\Delta\varepsilon_G$ curve.[12] $\Delta\varepsilon_G$ in Fig. 1 agrees fairly well with those of theoretically calculated curve[12] of $\Delta\varepsilon_G$ for lower β value, suggesting a high biquadratic coupling in the FLC mixture. As we did not put much voltage during alignment, so a chevron structure rather than a book-shelf structure may be expected in our cell. A bookshelf geometry is obtained when very high voltage is applied during alignment and the process is irreversible.[10] The dielectric strength $\Delta\varepsilon_G$ (Fig. 1) is fairly comparable to the $\Delta\varepsilon_G$ for the materials having almost same value of spontaneous polarization as reported by Biradar et al.[19] and Viz et al[10] in chevron structure geometry. This also suggest a chevron geometry in our cell. When the cell thickness is very low due to surface stabilization there is a unwinding of helix. There is practically no contribution from helicoidal motion and $\Delta\varepsilon_G$ value thus obtained low as reported by Viz et al[10]. We have used 20μm cell which is two times the pitch length of the material for our measurements. A large dielectric strength $\Delta\varepsilon_G$, what has been observed in the 20 μm cell is a part due to dynamic distribution of azimuthal angle φ with respect to X axis and the main part is due to the dynamic distribution of azimuthal angle φ with respect to the Z axis. On still thicker cell, X mode contribution gradually diminishes. In 100 μm cell only the Z mode contribution remains, which is arises due to helicoidal motion. Dielectric strength thus obtained in 100μm is relatively low. As the pitch of the FLC mixture is 10 μm, there is a less probability of suppression a helical structure particularly in 20 μm cell. So we can expect helical structure both in 20 μm and 100 μm cell. Due to a surface anchoring there may be a little distorted helicoidal structure in 20μm cell, but that does not affect much on relaxation frequency. Because the relaxation frequency as seen from equation (5) is proportional to K_3q^2/γ_φ. Where K_3q is the restoring torque and is counteracted by rotational torque which is proportional to rotational viscosity γ_φ. Any small change of rotational viscosity (γ_φ) and twist elastic constant (K_3) in a distorted helical structure involved in the relaxation frequency in opposite sense. Therefore in both cells the relaxation frequency is almost same as shown in fig 2.

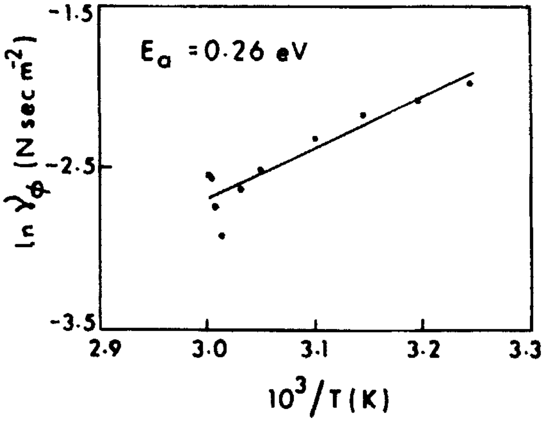

FIGURE 5 Arrhenius plot of the Goldstone mode rotational viscosity (γ_ϕ).

Rotational viscosity (γ_φ) has been determined from dielectric data on thick cells in which an unperturbed helicoidal structure is developed. So for the determination of γ_φ we have used 100μm cell. An Arrhenius plot of rotational viscosity (γ_φ) is shown in Fig. 5. As is seen from the figure5. that the rotational viscosity as well as the activation energy (E_a) is a little lower than other earlier reported result.[12,15] As is seen from the model equation[12] that γ_φ is strongly depends on $\Delta\varepsilon_G$, which in turn depends on the geometry of the cell. $\Delta\varepsilon_G$ is comparatively high in chevron structure rather than that of bookshelf structure. as reported by Vij et al. In the present case the probable geometry of our cell is chevron structure and dielectric constant thus observed is also high. The lower γ_φ value may be due to larger dielectric strength in present measurement geometry.

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