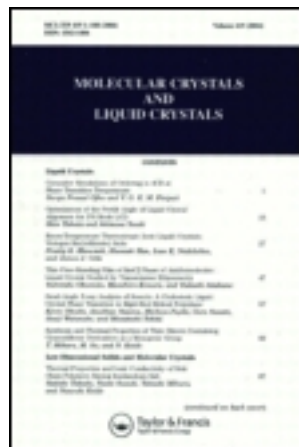


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SOFT MODE DIELECTRIC RELAXATION UNDER THE INFLUENCE OF BIAS ELECTRIC FIELD OF A FERROELECTRIC LIQUID CRYSTAL MIXTURE.

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Abstract Temperature dependent soft mode part of the complex dielectric constant has been measured as a function of bias electric field both in the smectic C* and smectic A* phases for a ferroelectric liquid crystal (FLC) mixture. From the experimental data, soft mode relaxation frequency and dielectric strength have been determined both in smectic C* and smectic A* phases. In smectic A* phase, relaxation frequency and inverse of dielectric strength of the FLC mixture does not follow the Curie-Weiss law. The critical exponent for the studied FLC mixture is 1.43. Dielectric results shows a dependence of soft mode dielectric strength and relaxation frequency with bias field near the transition temperature in smectic A* phase to an extent of 1°C above the transition temperature and above 1°C bias field does not have any effect on relaxation frequency and dielectric strength. It is observed that relaxation frequency increases with increasing bias field while the corresponding dielectric strength decreases.

INTRODUCTION :

In planar geometry of cell, two director modes of chiral smectic C liquid crystal are commonly known as Goldstone mode and soft mode^{1,2} apart from polarization modes. The polarization modes are well separated from the Goldstone mode and soft mode and its relaxation frequency is in the order of 500MHz.³ Goldstone mode arises due to a change of azimuthal angle of the molecular director by the application of measuring field. Goldstone mode relaxation frequency is of the order of 10Hz-2kHz and is almost independent of temperature except near the transition temperature. Dielectric permittivity of Goldstone mode is very high because of huge phase fluctuation. Soft mode on the other hand arises due to a change in the magnitude of tilt of the molecular director.^{4,5} It is very difficult to resolve the soft mode in the Sm-C* phase because of huge Goldstone mode contribution. This difficulty has been overcome by unwinding the helical structure with bias electric field^{6,7} parallel to layer plane and on imposition of probe field on the bias field at the vicinity of T_{SC*-SA*} transition. Another faster switching mechanism exists, which is known as electroclinic effect⁸ in Sm-A* phase. The electroclinic effect has been studied in Sm-A* phase by dielectric⁹⁻¹¹ and electro-optic method.¹² The object of the present paper is to report the soft mode behavior with and without bias field in both Sm-C* and Sm-A* phases.

In the present paper dielectric studies have been carried out for a ferroelectric liquid crystal (FLC) mixture possesses moderate range of Smectic- A* phase. The FLC mixture was available from E. Merck, Darmstadt (Code name ZLI 4237-100) having spontaneous polarization -20 nC cm^{-2} at 20°C and the phase sequences of the mixture are as follows (Ch denotes cholesteric, I isotropic, and K crystal)



EXPERIMENT

A parallel plate capacitor (ITO-coated glass plates) located at the end of a coaxial line, was used as an experimental cell. The thickness of the cell was about $20\mu\text{m}$. Before putting the sample into the cells, the air capacitance for the two cells were recorded at different temperatures and frequencies. For alignment the sample was heated to its isotropic phase by Mettler Hot Stage (model FP 5) and then cooled down very slowly ($0.2^\circ\text{C}/\text{min.}$) and simultaneously a low frequency (20Hz) electric field is applied to the cell. The temperature of the sample was stabilized to an accuracy of $\pm 0.01^\circ\text{C}$. A Hewlett-Packard impedance analyzer (HP 4192A), working in the frequency range of 5Hz to 13MHz was used for the complex dielectric permittivity measurements. 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.

RESULTS AND DISCUSSION :

The complex dielectric permittivities ϵ' and ϵ'' has have been measured both in Sm-C* and Sm-A* phase of a FLC mixture. Dielectric data has been analysed using cole cole ¹³fitting programme. Soft mode in Sm-C* phase has been obtained after suppression of helical structure with an application of bias electric field. Soft mode relaxation frequency and dielectric strength thus observed in Sm-C* phase is shown in Figs.1 and 2. As is seen from the Figs. 1 and 2 that on approaching to the transition temperature, soft mode relaxation frequency sharply decreases and dielectric strength increases to a maximum value at the $T_{\text{SC}^*-\text{SA}^*}$ transition temperature. Theoretical model ²⁷ predicts an approximate linear dependence of the relaxation frequency and inverse of dielectric strength with temperature. The theory qualitatively is in reasonable agreement with experimental data (Figs. 1 and 2) except very near to the transition temperature where it deviates from linearity. As on of the purpose of the present paper also to investigate the influence of bias electric field on the soft mode behavior in Sm-A* phase, we therefore measured the complex permittivity in Sm-A* phase with and without bias electric field. One of the reason of the investigation of soft mode behavior with bias electric field is to identify the exact soft mode contribution and to what extent soft mode is effected in Sm-A* phase under bias electric field. To extract information of soft mode in Sm-C* phase, we applied bias electric field for unwinding of helix. Again, bias electric field not only

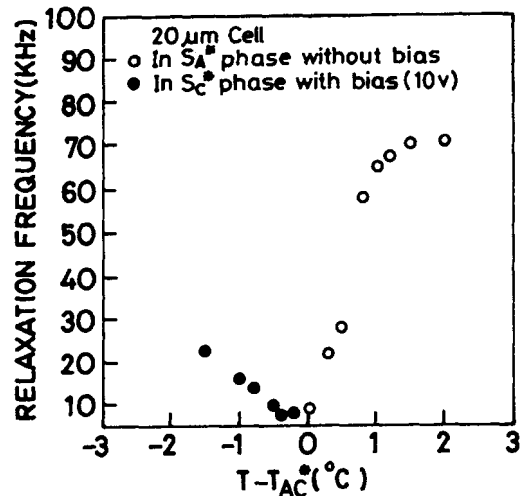


FIGURE 1. Variation of relaxation frequency of soft mode in $Sm-C^*$ and $Sm-A^*$ phases with temperature.

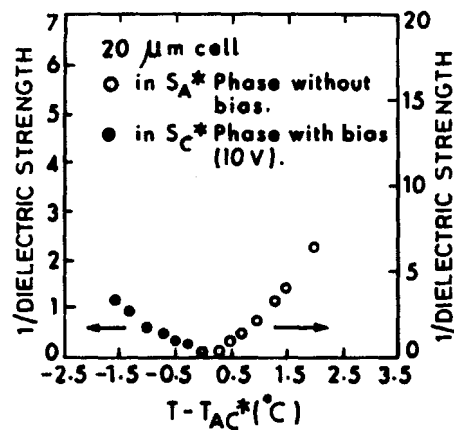


FIGURE 2. Temperature dependence of the soft mode dielectric strength in $Sm-C^*$ and $Sm-A^*$ phases.

decreases Goldstone contribution, soft mode behavior is also affected. So to extract exact soft mode contribution in $Sm-C^*$ phase, the study of bias electric field effect on the soft mode behavior in $Sm-A^*$ phase is also necessary. In $Sm-A^*$ phase, only soft mode is

present so any change of dielectric constant is only due to soft mode behavior. Cole-Cole diagram (Fig. 3) showing soft mode at different bias electric field in Sm-A* phase. As is seen from Fig. 2 that at zero bias field, the inverse of soft mode dielectric strength deviates from linearity with temperature. The critical exponent is 1.43. The nonlinear behavior of $1/\Delta\epsilon_s$ with temperature is also observed¹⁴ in a FLC mixture. The reason for this behavior in FLC mixture may be explained as follows.

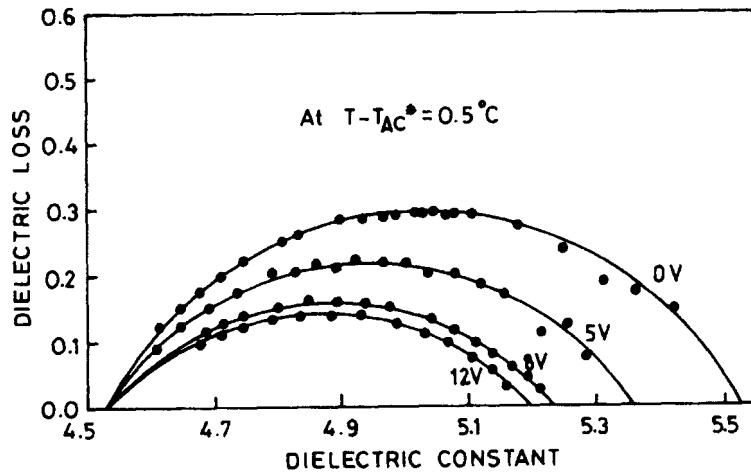


FIGURE 3. Cole-Cole representation of the soft mode at different bias field in the Sm-A* phase.

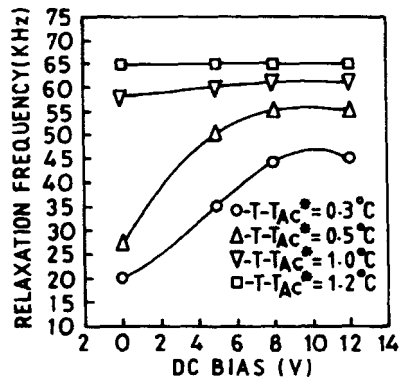


FIGURE 4. Bias dependent of the relaxation frequency of the soft mode in the Sm-A* phase.

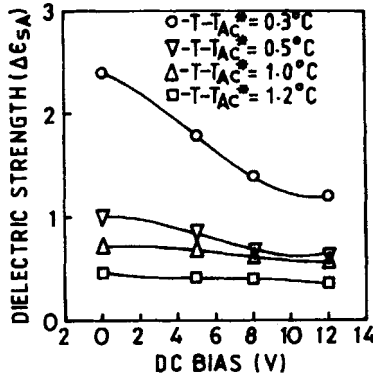


FIGURE 5. Bias dependence of the soft mode dielectric strength in the Sm-A* phase

As the temperature gradually decreases in the Sm-A* phase combined amplitude of tilt fluctuation of the individual components of the mixture due to application of probe voltage may not be uniform and this non-uniformity might have reflected in the soft mode dielectric strength in the FLC mixture. Bias dependent curves of soft mode relaxation frequency and strength are shown in Figs. 4 and 5. As is seen from Figs. 4 and 5 that at temperature 0.3°C and 0.5°C above transition temperature relaxation frequency gradually increases with the increase of bias electric field, but at about 1°C and above the transition temperature, the relaxation frequency is almost constant with bias electric field in the Sm-A* phase. Dielectric strength on the other hand behaves in opposite manner. At 0.3°C and 0.5°C above the transition temperature dielectric strength gradually decreases with bias electric field, but 1°C and above the transition temperature dielectric strength shows a constant value. In the absence of bias field soft mode in Sm-A* phase is observed due to instantaneous tilt developed as the system approaches to the Sm-A* to Sm-C* phase transition. Without bias field, tilt is developed due to a small change of temperature. As the system approaches nearer and nearer to the transition temperature, the instantaneous tilt developed is relatively high due to softening of elastic constant. The fluctuation of instantaneous tilt developed due to application of small measuring voltage is responsible for the observed soft mode in Sm-A* phase without bias field. In Sm-A* phase as the temperature approaches to SmC*-SmA* transition, the magnitude of tilt fluctuation with the applied probe voltage reflects the magnitude variation of dielectric strength with temperature. But in presence of bias field an induced tilt is developed over and above the instantaneous tilt and the induced tilt that has been developed¹⁵ has linear and some higher order terms. The closer the system to the transition in Sm-A* phase, the higher is the induced tilt and thus the linear and higher order terms of tilt as well as polarization are effective. The induced tilt thus obtained is non linear¹⁵ in character when bias field is applied. Due to non linear character of induced tilt nearer the transition temperature, relaxation frequency and dielectric strength (Figs. 4 and 5) are found increases when a measuring field is applied in conjunction with bias electric field. Away

from the transition temperature in Sm-A* phase induced tilt has a smaller value and only the linear terms comes to play. The variation of induced tilt is thus linear with bias electric field. A consequence of which the equilibrium induced tilt position is shifted without any change of relaxation frequency and dielectric strength (Fig. 4 and 5). Thus the predicted theory¹⁵ is in consistent what we have observed experimentally as shown in Figs. 4 & 5 So we have observed that inside 1 °C to SmA*-SmC* transition temperature, relaxation frequency and dielectric strength both perturbed with bias field, but above 1°C relaxation frequency and dielectric strength is independent of bias field for our FLC mixture. So if we consider the bias field will have an effect to the same extent above and below the SmC*-SmA* transition. We can explain the non-linearity of the soft mode relaxation frequency and dielectric strength curves near the transition temperature in Sm-C* as described earlier in Fig.1 and 2. By unwinding the helix with bias electric field we can study the exact soft mode behavior in Sm-C* phase avoiding the region where soft mode is perturbed by bias electric field.

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