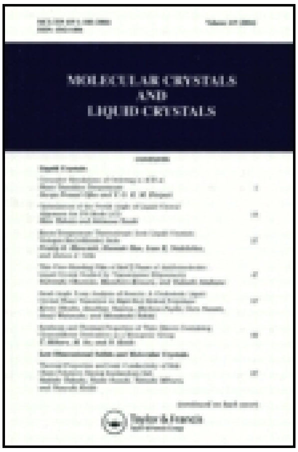

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Dielectric Properties of a Ferroelectric Liquid Crystal Mixture Under Bias Electric Field

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DIELECTRIC PROPERTIES OF A FERROELECTRIC LIQUID CRYSTAL MIXTURE UNDER BIAS ELECTRIC FIELD

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ABSTRACT The dielectric properties of a ferroelectric liquid crystal mixture of low spontaneous polarization have been studied here as a function of D.C. bias voltage, frequency and cell thickness. A small sharp peak of dielectric constant is found to become prominent at the chiral smectic C-smectic A phase transition under the condition in which Goldstone mode is suppressed. The relaxation frequency, dielectric strength and the rotational viscosity of the Goldstone mode have been determined under D.C. bias field.

INTRODUCTION

The discovery of ferroelectricity in smectic C^* phase and its subsequent applications in electro-optic device[1,2] provides the system which has gained an increasing amount of interest during last few years. An increasing number of ferroelectric liquid crystals (FLC) materials are coming up day by day and much efforts are being made to characterize the thermodynamic properties of the systems both experimentally and theoretically[3-7]. In this context the measurements of complex dielectric constant is important because in the same measurement both the static and dynamic properties of the systems can be determined. The large contribution to the dielectric constant perpendicular to the helical axis in the smectic C^* phase is due to winding and unwinding motion of the helicoidal structure, which arises due to the fluctuation of the azimuthal angle of molecular axis around the helicoidal axis. This contribution is called the Goldstone mode. A comparatively smaller contribution to the dielectric constant comes from the tilt fluctuation of the director. This relaxation mode is commonly known as soft mode.

In most of the FLC materials, below the smectic C^*-smectic A phase transition temperature (T_{C^*A}), as the temperature start decreasing the dielectric constant increases to a maximum value a few degree below the transition temperature due to the formation of ferroelectric C^* phase. Recently Ozaki et al[6,8] observed a small peak of dielectric constant at T_{C^*A} for the FLC materials possessing high spontaneous polarization (P_S). The effect of bias field on the dielectric properties of FLC materials are rare in the literature. Only a few papers have been published so far in this field[9-15]. The bias voltage is generally applied to isolate the softmode from the Goldstone mode. In the present paper the characteristic dielectric behaviour as a function of frequency and bias electric field for a low P_S ferroelectric liquid crystal mixture (ZLI-3654-000) has been reported. The dependence of the dielectric behaviour on the cell thickness has also been investigated here. The phase sequences of the sample is shown below:

$$K \; < \!— \; 30^{\circ}C \quad SmC^* \quad 62^{\circ}C \quad SmA \quad 76^{\circ}C \quad Ch \quad 86^{\circ}C \quad I$$

EXPERIMENTAL

The complex electric permittivity was measured by using sandwich type cells. Gold-plated and ITO-coated glass plates were used as electrodes. We have used three cells of cell thickness 17 μm, 40 μm and 100 μm. The measurements were carried out for planar configuration. The sample was aligned parallel to the glass plates by slowly cooling (cooling rate 0.2°C/min.) from its isotropic phase in presence of magnetic field (10 kG) for 40μm and 100μm cell but for 17μm cell thickness we simply cooled the cell (cooling rate 0.2°C/min.)

in presence of a 20 Hz ac field of amplitude $4V_{rms}$. For ITO coated cell alignment was checked under polarizing microscope and a fairly good alignment was achieved with large focal conic texture. For gold plated cell we compared the measurements with that was obtained from the ITO coated cell with a known FLC material and a fairly comparable results were obtained. Before actual reading the cells were calibrated using air and highly pure benzene as standard references. The S_C* pitch of the FLC material is -3μm at 20°C and the P_S and tilt angle are $-29nC/cm^{-2}$ and 25 degree respectively at 20°C.

A Hewlett-Packard impedance analyzer (model-HP 4192A) was used to investigate the frequency dependence of the real and imaginary part of the electric permittivity of the sample in the frequency range from 5 Hz to 13 MHz. The temperature of the sample was controlled by using Mettler Hot Stage (model - FP 5). The D. C. bias field was applied perpendicular to the helix axis i.e. parallel to the layer plane.

The relaxation frequency and the distribution parameter have been calculated from Cole - Cole equation[16] using

$$v/u = (w\tau)^{1-h}$$

$$\text{Where } v = [\,(\epsilon(o) - \epsilon'(w))^2 + (\epsilon''(w))^2\,]^{1/2}$$

$$u = [\,(\epsilon'(w) - \epsilon(\alpha))^2 + (\epsilon''(w))^2\,]^{1/2}$$

$\epsilon'(w)$ = dielectric constant at a particular frequency.

$\epsilon(o)$ = low frequency dielectric constant where the Cole - Cole plot cut the abscissa axis at the lower frequency side.

$\epsilon(\alpha)$ = high frequency dielectric constant where the Cole - Cole plot cut the abscissa axis at the higher frequency side.

A plot of $\log_{10}(v/u)$ against $\log_{10} f$ should give a straight line, the intercept on the abscissa axis corresponds to f_r and the slope gives the distribution parameter h.

RESULTS AND DISCUSSION

Figure 1 shows the typical graph of dielectric constant ε' against frequency at different bias electric field. At low frequency side, a sharp change of dielectric constant is observed as the bias voltage increases, but in higher frequency bias voltage has a little effect on dielectric constant; because at higher frequency the Goldstone mode contribution to the dielectric constant in S_C* phase has already been suppressed due to frequency dispersion of dielectric constant and further suppression of dielectric constant due to D.C. bias voltage is found smaller. The temperature dependence of the dielectric constant at various frequencies is shown in Figs. 2 and 3. At lower frequency (Fig. 2), temperature dependence of dielectric constant curve shows a step wise increase of dielectric constant as the temperature decreases from T_{C*A} but no such stepwise increase of dielectric constant with temperature could be observed at higher frequencies because at higher frequency the dielectric constant is suppressed due to frequency dispersion and due to these suppression the stepwise increase of dielectric constant may not be observed at higher frequency curves in

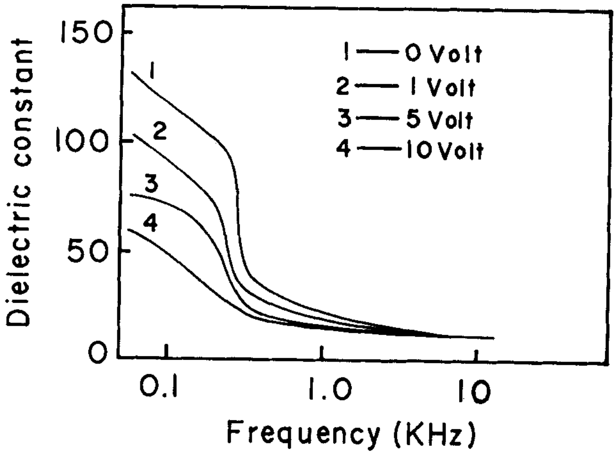

FIGURE 1 Plot of dielectric constant vs. frequency at different D.C. bias field (at 58°C).

Fig. 3. The temperature dependence of dielectric constant at

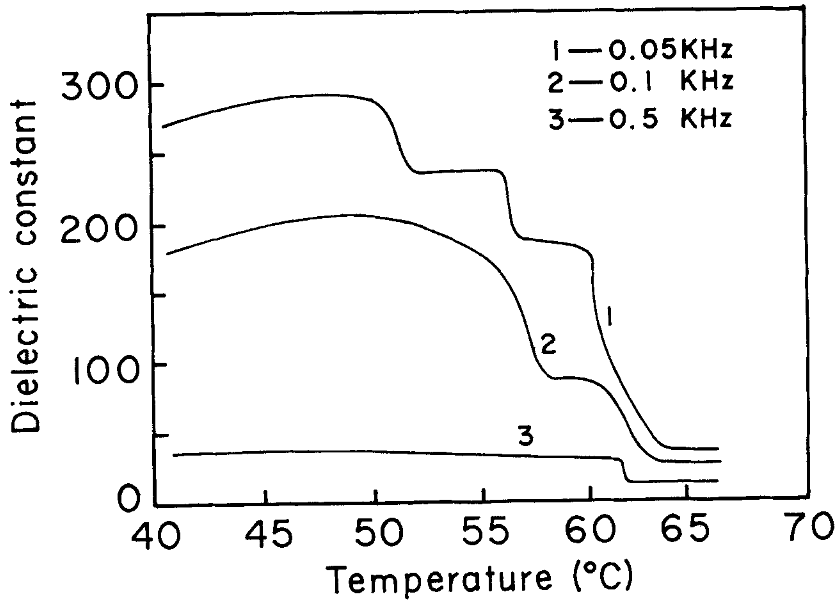

FIGURE 2 Temperature dependence of the dielectric constant in the lower frequency region (with cell thickness of 17μm).

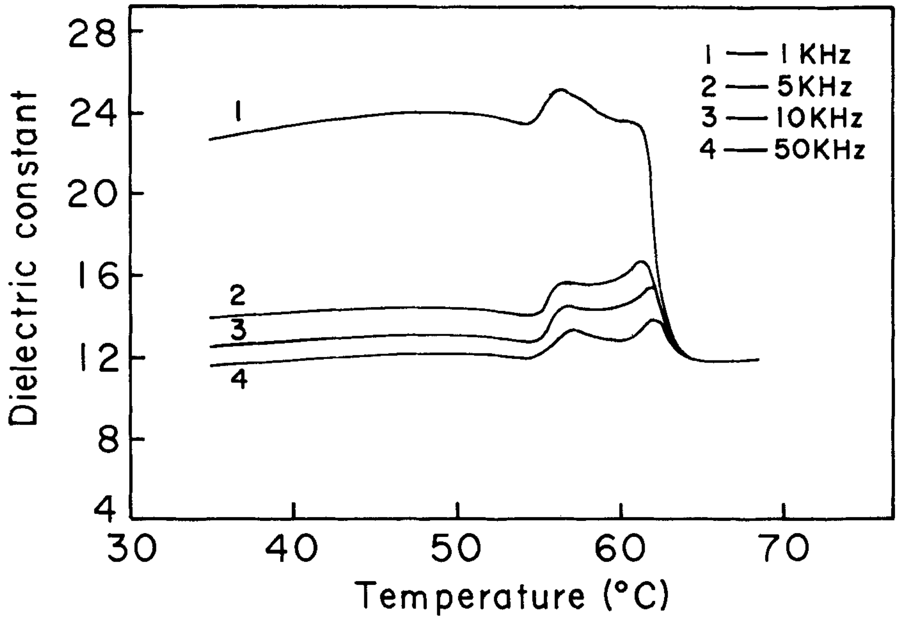

FIGURE 3 Variation of dielectric constant with temperature in high frequency region (with cell thickness of 17 μm).

different cell thickness in figure 4 have been done at 1KHz, which is higher than the relaxation frequency of the sample and the dielectric constant has been suppressed due to frequency dispersion and for the same reason no such stepwise increase of dielectric constant is observed in Fig. 4. A sharp increase of the value of the dielectric constant a few degree below T_{C^*A} with the decrease of temperature can be explained as follows - As the temperature cools down below T_{C^*A} the helicoidal structure begins to form but surface interaction with the wall of the cell prevents the formation of helicoidal structure. As the temperature further goes down, the chiral force dominates over the surface force and at temperatures around 10°C below T_{C^*A} a complete formation of the helicoidal structure is over. As a result dielectric constant attains a maximum value due to helicoidal motion.

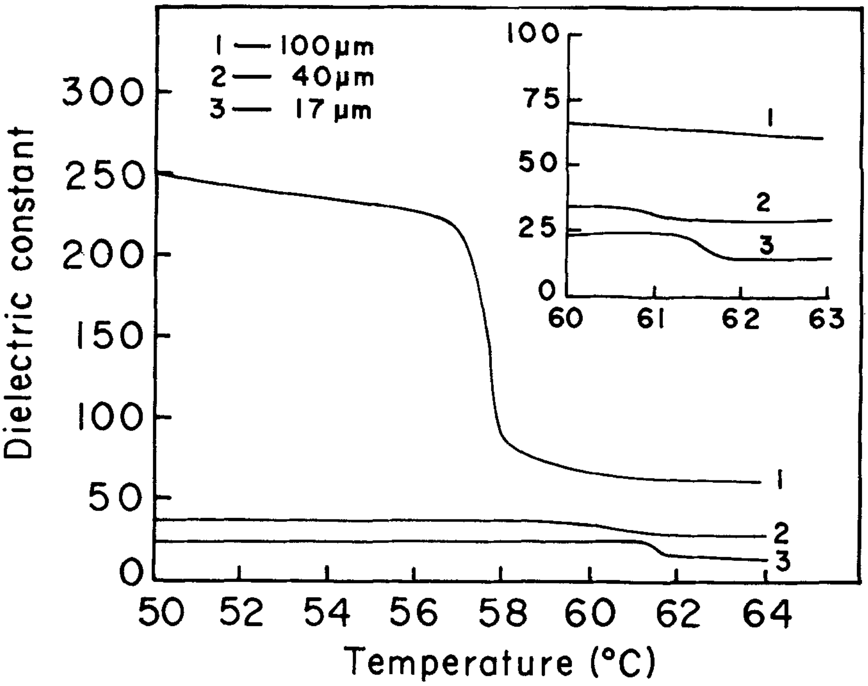

FIGURE 4 Dielectric constant vs. temperature at different cell thickness (at 1 KHz).

But as the temperature further decreases the dielectric constant begins to fall at far below the T_{C^*A} owing to the fact that the viscosity of the medium increases with the decrease of temperature and the system get stiffer against phase fluctuation and consequently dielectric permittivities decreases. This type of behaviour of dielectric constant with temperature in the S_C* phase was also observed by others[17,18], but no such decrease of dielectric constant was observed at far below T_{C^*A} temperature in higher frequency curves, due to the suppression of Goldstone mode and as a consequence of which the dielectric constant is greatly reduced at higher frequencies and any further reduction of dielectric constant due to lowering of viscosity at far below T_{C^*A} may be negligibly small. In Fig. 3 a small peak of dielectric constant is appeared near T_{C^*A}. This peak is prominent at 5 KHz and higher frequencies which are much higher than the relaxation frequency of the Goldstone mode of this compound. Therefore it is not related to Goldstone mode contribution because at higher frequency around 10 KHz, there should not be any peak in S_C* phase and at such high frequencies there is practically no contribution from the Goldstone mode due to dispersion. Therefore it can be assumed that this peak becomes observable owing to the suppression of the Goldstone mode at high frequency. Such a characteristic peak was reported earlier[8] for FLC materials with high values of spontaneous polarization. To get the better understanding regarding this peak in our case we also measured the dielectric constant as function of cell thickness and D.C. bias field.

The helicoidal structure of FLC materials in S_C* phase is influenced by the surface effect and dielectric constant in such phase is also suppressed by the decrease of the Goldstone mode contribution in thin cell. For FLC materials with high P_S value the goldstone mode is suppressed even if the thickness of the cell is greater than 200-300 μm.[8] Temperature dependence of the dielectric constant at three different cells of various spacer thickness is shown in Fig. 4. Dielectric constant is found to decrease with the decrease of cell thickness which is quite consistent by the fact that the helicoidal motion in S_C* is suppressed and its contribution to the dielectric constant is greatly reduced in thin cell. Consequently a small dielectric constant peak is become prominent in a thin cell at T_{C*A}. This peak is become prominent owing to the suppression of Goldstone mode contribution at smaller thickness of the cell. From Fig. 5 it is also observed that as the D.C. bias voltage increases in

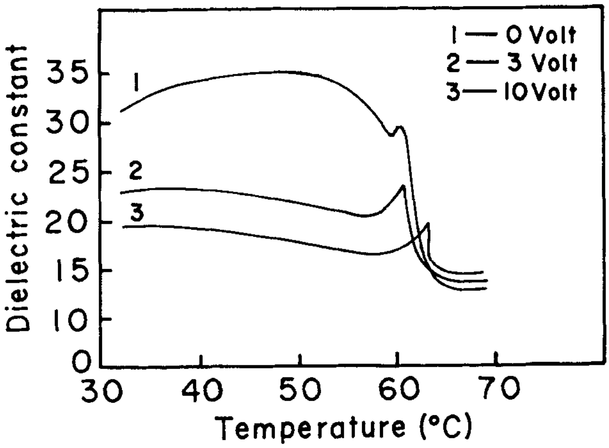

FIGURE 5 Dielectric constant vs. temperature at different D.C. bias field.

the S_C* phase the helicoidal structure gets unwound and the helicoidal motion ceases. As a result a small peak appears at T_{C*A} at higher D.C. field. Therefore it may be concluded that the small peak of the dielectric constant which is become prominent at T_{C*A} is due to the soft mode contribution which arises due to the suppression of the Goldstone mode contribution to the dielectric constant. Recently Levstik et al.[19] also reported the appearance of a small peak of dielectric constant at T_{C*A} from theoretical calculations. In curve no. 4 of Fig. 3 a small anomalous dip of the dielectric constant is also observed at just below T_{C^*A}. This can be explained as follows-In the present studied sample the relaxation frequencies of the soft mode just at the transition temperature is around 6 - 10 KHz. So at frequency higher than 10 KHz, dielectric constant of the soft mode decreases with the increase of frequency due to anomalous dispersion and the small dip is observed in the temperature dependence of the dielectric constant curve. This anomalous dip of dielectric constant in higher frequency was also observed earlier by Ozaki et al.[8] From Fig. 5 it is also found that the abrupt change of the dielectric constant occurs at 61°C which is a little below the actual phase transition temperature of the material observed in texture studies under polarizing microscope. This is also observed by Biradar et al[14] for this sample. They observed the smectic C^*-smectic A phase transition 2°C below the actual T_{C^*A}. Cole-Cole plots as a function of various D.C. bias fields are shown in Fig. 6.

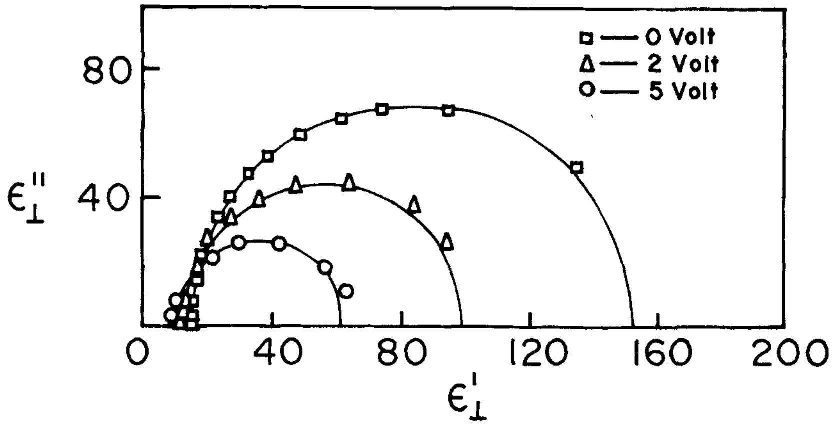

FIGURE 6 Cole-Cole plot at different D.C. bias field in the smectic C^* phase (at 58°C).

Figures 7 and 8 are given to show the variation of dielectric strength ($\Delta\epsilon_G$) and relaxation frequency (f_G) of the Goldstone mode with D.C. bias voltage. From these two graphs it is seen that the value of dielectric strength is gradually decreasing and the relaxation frequency is increasing with the increase of D.C. bias voltage which quite resembles to the earlier reported values.[20,21] The dielectric strength gradually decreases with applied D.C. bias voltage - predicts a continuous deformation of the helix. It can also be observed from the Fig. 7 that as the transition approaches the critical field for perturbing the helicoidal motion becomes less. Since the sample has low P_S value we could not unwound the helix completely at about 10 volt D.C. bias voltage. The relaxation frequency at different temperature as

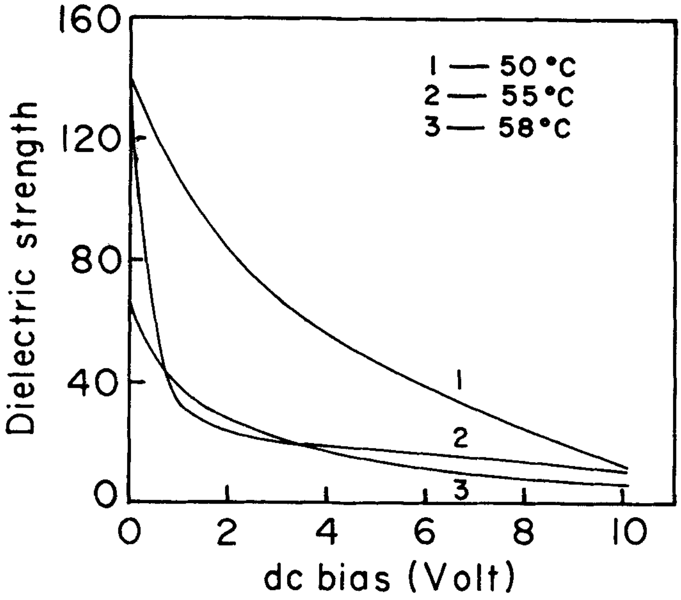

FIGURE 7 Plot of dielectric strength ($\Delta\epsilon_G$) of the Goldstone mode with D.C. bias field.

shown in Fig. 8 increases with the increase of bias field. Near to transition temperature there is a sharp rise of relaxation frequency with low bias voltage. Therefore it may be concluded that as the transition temperature approaches the critical field necessary to distort the helicoidal

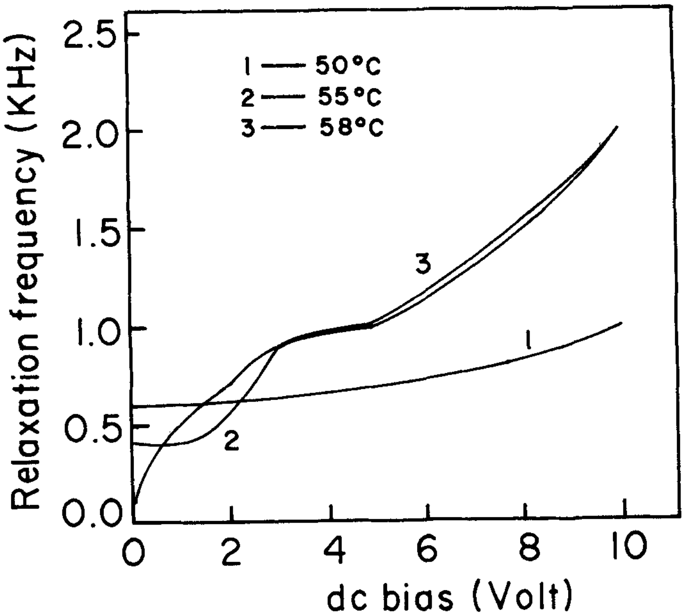

FIGURE 8 Plot of relaxation frequency of the Goldstone mode with D.C. bias field.

structure becomes less. Rotational viscosity has been calculated using the visco-elastic equation[22]

$$\gamma_{\phi} = \frac{P^2}{4\pi\epsilon_0 \, \theta^2 \, (\Delta\epsilon_G \, f_G)}$$

Where P = Spontaneous polarization
θ = tilt angle

From our $\Delta\epsilon_G$ and f_G values we have calculated the rotational viscosity co-efficient (γ_ϕ) of this sample in the S_C* phase as a function of temperature under D.C. bias voltage. We have taken the literature[23] values of P_S and tilt angle (θ) of this sample. In this case we have assumed that these two parameters vary little with D.C. bias voltage. The variation of rotational viscosity in the S_C* phase with D.C. bias voltage and temperature is shown in Fig. 9. It has been

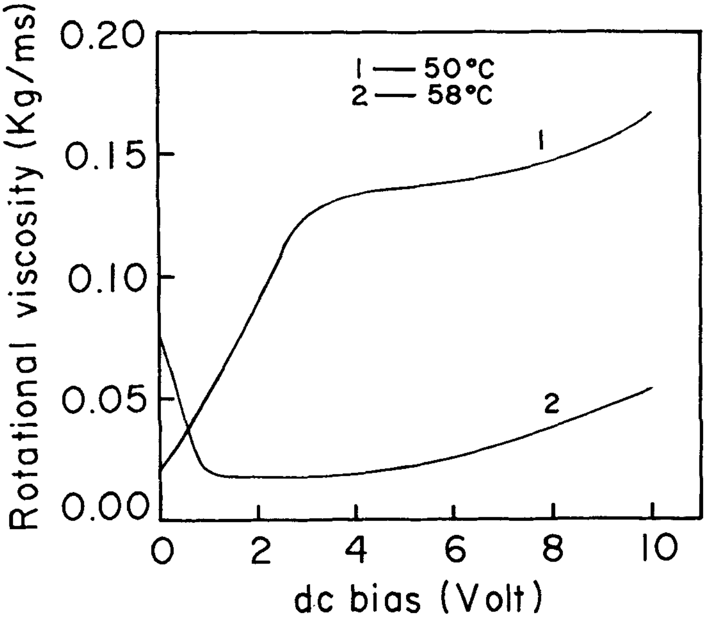

FIGURE 9 Variation of rotational viscosity in the smectic C^* phase with D.C. bias voltage at different temperatures.

shown in Fig. 9 that γ_ϕ depends much on temperature but slowly increase with bias voltage from 2Volt to 10Volts. The viscosity γ_ϕ is found to be invariant by Srivastava et al.[24] The visco-elastic equations contains P_S and tilt angle θ which are not truly independent of bias voltage but in calculation we have taken these as constant due to which our viscosity results are found to vary a little with voltage.

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