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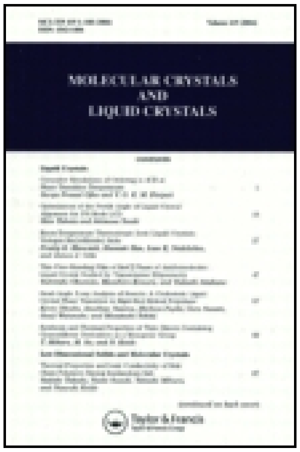

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THICKNESS DEPENDENCE OF DIELECTRIC PROPERTIES OF A FERROELECTRIC LIQUID CRYSTAL

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Abstract A room temperature ferroelectric liquid crystal has been studied by means of dielectric measurements. The results of the studies of the complex electric permittivities for aligned samples of different thicknesses and for the different electric measuring fields are presented. We discuss how dielectric properties depend on boundary conditions, thicknesses of the samples and the magnitude measuring electric fields. The obtained experimental results and the discussion make it possible to determine the spontaneous polarization from dielectric measurements.

INTRODUCTION

Since the discovery of ferroeletric liquid crystals by Meyer[1] et., a variety of experiments have been performed in order to understand the nature of this phase. In this context the dielectric constant is one of many studied properties[2-7], however in many cases the obtained results of different authors are at variance with respect to some aspects. Particularly some differences can be noticed easily in the case of the real part $\varepsilon'_\perp$ of the complex electric permittivity $\varepsilon_\perp = \varepsilon'_\perp + i\varepsilon''$ when the Goldstone-mode contribution dominates the dielectric behaviour[2,8]. In this work we will show how dielectric properties of a room temperature ferroelectric liquid crystal depend on boundary conditions, alignment and thicknesses of the samples and the applied electric field magnitude when the Goldstone-mode appears.

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EXPERIMENT

Thickness, frequency and temperature dependences of the complex dielectric constant were measured on the ferroelectric mixture of TKF described in[8,9] with the following phase sequence;

$$\mathrm{Cr}4\mathrm{SmC}^{*}54\mathrm{SmA}65\mathrm{I}$$

Dielectric measurements were made with the measuring cell and setup previously described [8,10]. Cells in the form of flat condensor were made from tin oxide (ITO) coated glass with the electrode area of approximately 16 mm^2 and thickness varying from 2 μm to 50 μm. The boundary conditions were changed by coating the electrodes with 300Å of different kinds of polyimides followed by suitable rubbings. The real $\varepsilon'_{\perp}$ and imaginary $\varepsilon''_{\perp}$ parts of $\varepsilon_{\perp}$ for FLC were measured between 10 Hz and 300 kHz employing a Precision Component Analyser WAYNE KERR 642 with a measuring voltage from 10 mV to 5V. Temperature stability was better than 0.2°C at each temperature of the sample. The sample aligmment was checked optically with a polarizing microscope.

THEORY

For chiral smectics C (SmC^{*}) the vector of dielectric polarization $\hat{P}$

$$\hat{P} = \varepsilon_o(\hat{\varepsilon} - 1)\,\hat{E} \tag{1}$$

where ε_o is the dielectric constant of free space, $\hat{\varepsilon}$ is the tensor of the complex relative permittivity of a media and $\hat{E}$ is the vector of electric measuring field, generaly consists of four parts:

$$P = P_1 + P_2 + P_3 + P_4 \tag{2}$$

In (2) one can notice that apart from P_1 (induced polarization) and P_2 (orientational one) the low frequency measuring electric field can induce in SmC^{*} phase parts which come from colective excitations such as P_3 - from soft mode and P_4 - from Goldstone mode.

The chiral ferroelectric SmC^{*} phase represents the well known spatially modulation structure[1,13]. The director n of the smectic layer coinciding with the most probable orientation of a long axes b

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of the chiral molecule in the layer processes helicoidally as one goes from one smectic layer to another. The geometry of system is presented in Fig. 1a.

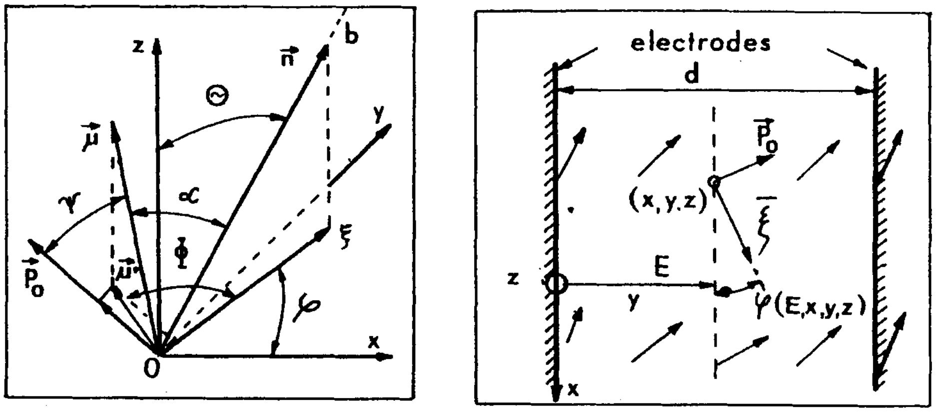

FIGURE 1 a - Geometry of the SmC*, b - The local spontaneous polarizations of a chosen smectic layer.

Let's OXYZ be the laboratory reference frame. Its OZ axis is the normal to the smectic layers. The orientation of the n with respect to the OXYZ frame is determined by a tilt angle θ and an angle φ which is connected with the wave vector of helix q: $\varphi = q \cdot z$ where $q = 2\Pi/\lambda$, λ is the helical pitch.

The projection of the molecules into the smectic planes (the xy planes) is described by $\xi = (\xi_x, \xi_y)$ the primary order parameter[13,14]. Let the permanent dipole moment of a molecule μ forms an angle α with b axis. Since the rotation of chiral molecules about long axis b of their ellipsoid of inertia in the absence of external electric field is not free[15], the most probable projection μ' of μ into the xy plane forms with ξ the angle Φ . Hence the secondary order parameter in the plane polarization, P_o appears. The vector of P_o perpendicular to ξ is given by:

$$P_o = N_o \, \mu < \cos\Psi > \qquad (3)$$

where N_o is the number of molecules in $1m^3$ of a single smectic layer.

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From geometrical considerations based on Fig. 1a one notices that[16]

$$\cos\Psi = \cos\alpha\,\cos\theta + \sin\Phi\,\sin\alpha\,\sin\theta \tag{4}$$

So, in the absence of an electrical field and special surface restriction one can see that even if the system with C_2 symmetry exibits a local net polarization P_o, the macroscopic average polarization P of the sample is zero because the P_o changes with the helix.

When one measures $\varepsilon_\perp$ in the aligned sample of SmC^* bounded by electrodes, one applies an electric field of magnitude E parallel to the smectic layers. The electric field disturbs the helix in such a way that:

a) - the phases φ of a chosen layer are not only the function of z , as they are in the case of unbounded one, but the E, x and y as well (see Fig. 1b) - Goldstone mode:

aa)- the magnitude of the tilt angle θ is the function of E,x,y,z.

The above distortion of the helicoidal ordering by E induces the vector field of P(E,x,y,z) in the interelectrodes space.

If we restricted our considerations to the Goldstone mode, the P_x and P_y components of P(E,x,y,z) may be written as:

$$\begin{aligned} P_x(E,x,y,z) &= -P_o \sin\varphi(E,x,y,z) \\ P_y(E,x,y,z) &= P_o \cos\varphi(E,x,y,z) \end{aligned} \tag{5}$$

Taking into consideration (1), (3) and (4) the Goldstone mode conribution $\delta\varepsilon'_\perp$ to the measured value of $\varepsilon'_\perp$ is the following

$$\delta\varepsilon'_\perp = \frac{1}{\varepsilon_o E} P_4 = \frac{d}{\varepsilon_o U_{AC}} P_4 \tag{6}$$

where

$$P_4 = P_o \langle\cos\varphi(E,x,y,z)\rangle_{x,y,z} \tag{7}$$

The averaging in (7) is carried out over the whole sample.

According to (1),(6) and (7) both $\varepsilon'_\perp$ and calculated from

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dielectric measurements the total average polarization P_4 should depend on d, E and boundary conditions of the sample. Egn. (7) also indicates that calculated P_4 should approach the spontaneous polarization P_0 for thick cells and rather high applied electric fields. However, from the other hand the measuring field should be weak enough to ensure a linear relation between the field and its response.

RESULTS AND DISCUSSION

To learn about the role of E and d in the process of distortion of helicoidal structure, the TKF with λ=3 μm at 22°C was investigated by means of dielectric and microscopic measurements.

Thermal and frequency characterictics of $\varepsilon'_{\perp}$ and $\varepsilon''_{\perp}$ in the SmC^*, SmA and I states were determined for different polyimids coated glass plates, different distance between the glass plates and different measuring voltage U_{AC} .

Typical thermal characteristics for d=23μm are shown in Fig. 2.

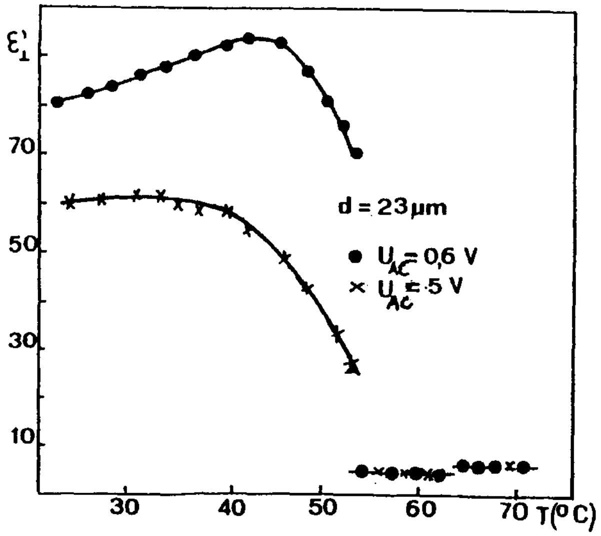

FIGURE 2 Temperature characteristics of $\varepsilon'_{\perp}$ for TKF at 20Hz.

When the applied field is small one can notice under the polarizing microscope that the helix still exists and the temperature variation of $\varepsilon'_{\perp}(T)$ is a consequence of the shape of $\lambda(T)^{8,17}$. If E increases the helix becomes unwound and the thermal characterics of $\varepsilon'_{\perp}(T)$ becomes to reproduce the order parameter S(T) for SmC* state[18].

Fig.3 shows the thickness dependence of $\varepsilon'_{\perp}$ measured in frequency range from 10 Hz to 300 kHz. The thickness dependence of $\delta\varepsilon'_{\perp}$ becomes important at frequencies below 5 kHz, where the Goldstone mode dominates.

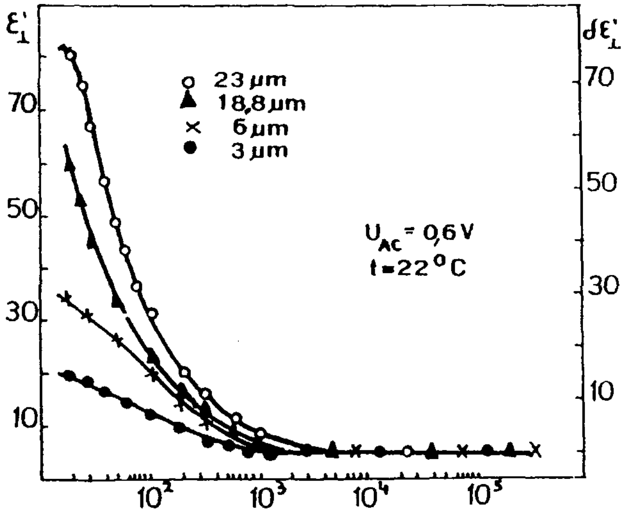

FIGURE 3 Thickness dependence of the Goldstone mode contributions for TKF. The glass plates were coated with the same polyimide of P-1[19].

When the boundary conditions of the samples are the same (the same polyimid, the same procedure of rubbing) the samples exibit a fairly good homogeneous orientation and the U_{AC} is fixed one can observe increasing of $\delta\varepsilon'_{\perp}$ with increasing of d. The above is in good agreement with eq.(5). If the boundary conditions are not the same, as it was in the samples with d=10,3 μm and d=44μm (where the

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polyimides P-2 and P-3) were used respectively) the above sequence may be even reversed as is shown in Fig.4 and also observed by Biradar et al.[2]

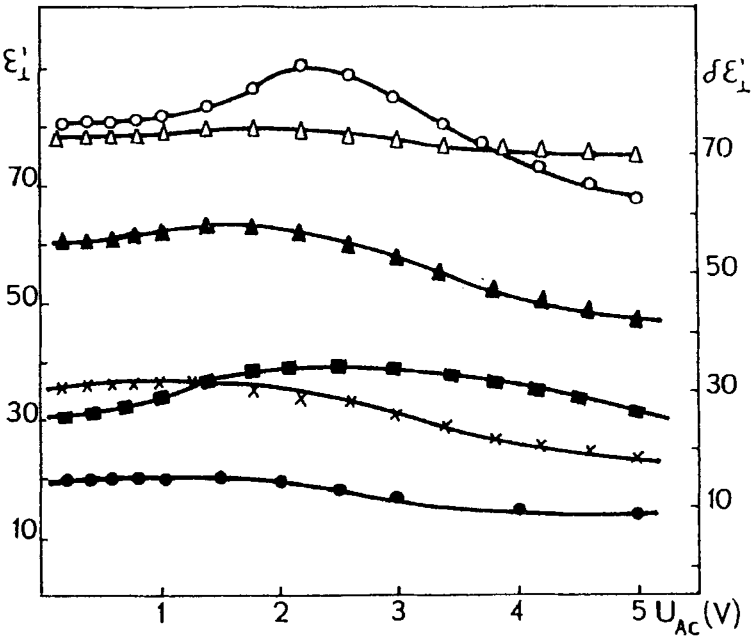

FIGURE 4 Voltage dependence of the Goldstone mode contribution for TKF at 22°C and 20 Hz : (●) d=3μm with P-1, (X) d=6μm with P-1, (□) d=10,3μm with P-2, (▲) d=18,8μm with P-1, (o) d=23μm with P-1, (△) d=44μm with P-3.

The same results may be obtained when the bookshelf aligment is not the best (see Fig.5).

Fig.6 shows calculated values of P_4 obtained from eq.(5) on the basis of results shown in Fig.4. As we can see, in case of TKF the measured values of P_4 for d smaller than 23 μm depend sharply on boundary conditions and values of E. When d is higher than 23 μm, the the influence of E (for d=23μm with P-1 and for d=44μm with P-3 one obtained the same functions P_4=f(E).

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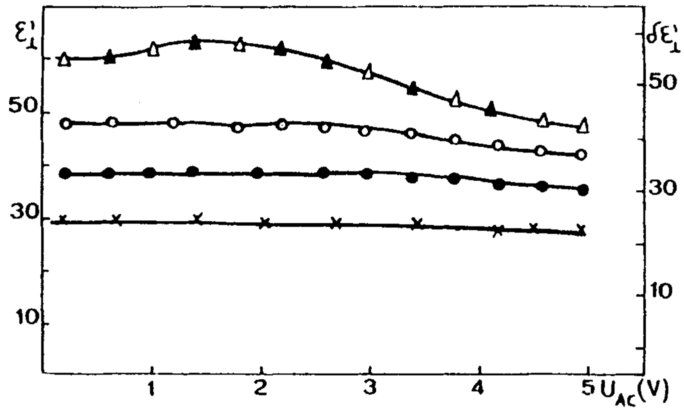

FIGURE 5 Voltage dependence of the Goldstone mode contribution for TKF at 20°C, and 20Hz, d=18,8μm, P-1 obtained: (×) - just after filling, (●) - after one cycle of heating up to $T_{SmA,I}$ and then slowly cooling to SmC* in the presence of proper electric field $E^{8,20}$, (o)- after two such cycles, (Δ) - after one cycle slowly cooling in presence of proper magnetic field B, (▲) - after two such cycles.

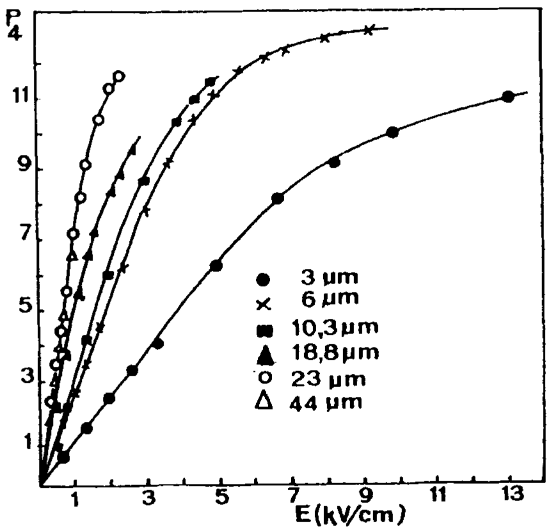

FIGURE 6 The calculated values of P_4 in function of E for TKF at 22°C and 20 Hz.

It is worth pointing out that the value of the spontaneous polarization (P_o = 13,6 nC/cm^2) to which approach all characteristics P_4 =f(E) is in good agreement with previous results[8,9].

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