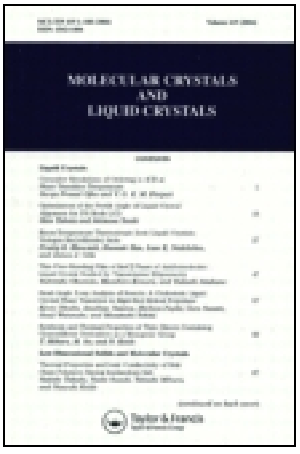

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LOW FREQUENCY RELAXATIONS IN A FERROELECTRIC LIQUID CRYSTAL WITH THE HELIX TWIST INVERSION

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Abstract Dielectric dispersion has been studied in the frequency range of 10Hz÷1MHz in a liquid crystal with 2-alkoxypropionate chiral group, which exhibits the helix twist inversion in the SmC* phase at about 109°C. The dispersion reveals two modes, dielectric strengths as well as relaxation frequencies of which exhibit a strong dependence on the sample thickness. Both modes are also detected in the electrooptic response. The mode with the relaxation in the kHz range is the Goldstone mode, which is combined with the thickness mode. In the temperature range near the inversion temperature, where the helicoidal structure is unwound, it becomes the pure thickness mode. The origin of the lower frequency mode, which is detected for the first time, remains unexplained.

INTRODUCTION

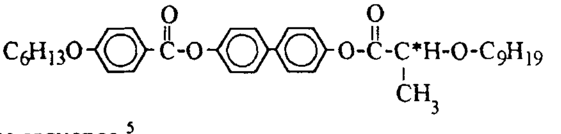
In the ferroelectric chiral smectic C (SmC*) phase two low frequency modes exist, soft and Goldstone modes, which are connected with the structural change at the SmA→SmC* phase transition. This change,[1] namely tilt of molecules from the smectic layer normal, is described by a 2-dimensional parameter $\xi_1 = \theta \sin\varphi z$, $\xi_2 = \theta \cos\varphi z$, where θ denotes the amplitude and φ the phase of the mean molecular tilt from the layer normal (z-axis). The soft mode represents fluctuations $\delta\theta$, the Goldstone mode, existing due to the breaking of the continuous symmetry, represents the phase fluctuations $\delta\varphi$. As the local optical axis is parallel to the mean molecular long axis (director), both modes can be detected in the electrooptic response. The electrooptic response of the soft mode in the SmA phase is known as the electroclinic effect.[2] Due to a linear interaction of the order parameter with the polarization, both modes can be seen in dielectric response as well.

In thin planar samples, where the helix (and therefore also the Goldstone mode) is suppressed by surface interactions, dielectric constants ε originates mainly in the contribution of a twisted structure, which is in-smectic-layer director twist-bend along the sample plane normal arising due to the polar surface anchoring.[3] A contribution of the fluctuations of the twisted structure ("thickness mode"[4]) to ε is as high as that of the Goldstone mode.[4] The director twist-bend, which is accompanied with the twist of P_s vector, exists also in usual thick samples, in which the helix is well developed. When studying either dielectric or electrooptic response in the SmC* phase the Goldstone mode and the thickness mode are not distinguishable, probably because both modes exhibit their relaxations in the same frequency region.

In this contribution we report results of the study of dielectric and electrooptic properties in a material exhibiting a spontaneous helix twist inversion in a certain temperature T_i in the SmC* phase. In this material an increase of pitch when approaching T_i and spontaneous helix unwinding in the vicinity of T_i takes place. In this temperature range the Goldstone mode is suppressed and we can detect the thickness mode separately. This way enables to distinguish between both modes.

EXPERIMENTAL

The liquid crystalline material used is



which exhibits a phase sequence [5]

Cryst. 25°C → SmC* 124°C → SmA 128°C → N* 136.5°C → BP 137°C → Iso

and the helix twist inversion temperature T_i =109°C.[5] The samples were filled into cells composed of glasses which are provided with ITO transparent electrodes and polyimid layers unidirectionally rubbed. The thickness was defined by mylar sheets as 6, 12, 25, 50, and 100μm. The samples exhibit planar (book shelf) geometry. The alignment was improved by electric field (10÷20Hz, 40kV/cm) applied for 10÷30min. During measurements the alignment was checked in polarizing microscope. No chevrons occurred in the sample texture, likely due to rather week surface anchoring.

We have measured the complex permittivity by Schlumberger 1260 impedance analyzer in the frequency range of 10Hz÷1MHz, keeping the temperature of the sample stable within ±0.1K. With the sample 25μm thick the electrooptic response has been measured in the frequency range of 10 Hz÷100kHz as a light transmitted through the sample, when the a.c. electric field has been applied.

EXPERIMENTAL RESULTS

Frequency dependences of imaginary (ε'') parts of permittivity are shown in Fig. 1 for 25μm thick sample at selected temperatures. They exhibit two relaxation processes denoted as mode 1 and 2 which are clearly seen as two maxima in $\varepsilon''(f)$ dependences (see Fig.1). Both these modes are also detectable in the frequency dispersion of the electrooptic response.

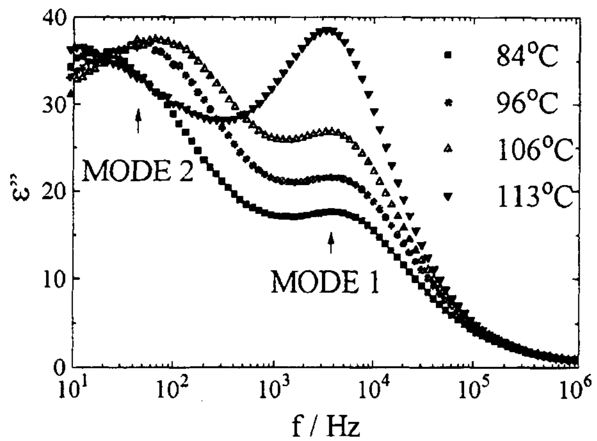


FIGURE 1 Frequency dispersions of imaginary parts of permittivity ε'' measured on 25μm sample at indicated temperatures.

Under an applied electric bias field the imaginary part of dielectric response connected with mode 1 decreases while that connected with the mode 2 increases when the bias field is increasing. The helicoidal structure was suppressed under the bias field 6÷8 kV/cm, which was observed as disappearing of dechiralization lines.

The frequency dispersion data were analysed using the Cole-Cole formula:

$$\varepsilon^* - \varepsilon_\infty = \frac{\Delta\varepsilon_1}{1+(jf/f_1)^{(1-\alpha_1)}} + \frac{\Delta\varepsilon_2}{1+(jf/f_2)^{(1-\alpha_2)}} \qquad (1)$$

where f_1, f_2, $\Delta\varepsilon_1$, $\Delta\varepsilon_2$, and α_1, α_2 are relaxation frequencies, dielectric strengths and distribution parameters for mode 1 and 2. The relaxation frequencies f_1, f_2 were obtained by fitting the imaginary part of eq.1, the dielectric strengths $\Delta\varepsilon_1$, $\Delta\varepsilon_2$ were determined from the Cole-Cole plot $\varepsilon''(\varepsilon')$ (see Fig. 2).

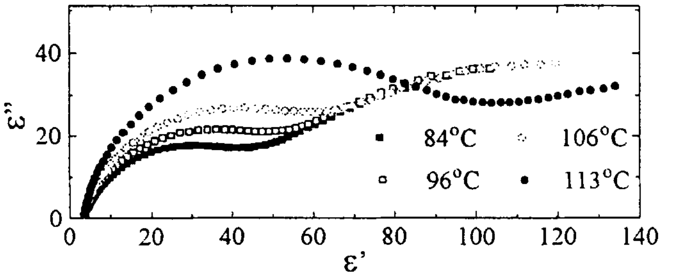


FIGURE 2 Cole-Cole plots for the sample 25μm thick at indicated temperatures.

The temperature dependences of fitted f_1 and $\Delta\varepsilon_1$ are shown in Figs. 3 and 4 for all studied sample thicknesses. Pronounced thickness dependences of both quantities are seen in the whole measured temperature region. The values of parameters α_1 are in the range 0.08÷0.12 and independent on the sample thickness. The low value of α_1 gives evidence for nearly monodispersive character of mode 1. Fig. 5 shows thickness dependence of f_1 and $\Delta\varepsilon_1(T)$ for temperature T=90°C. Relaxation frequency f_1 increases remarkably with diminishing cell thickness, dielectric strength $\Delta\varepsilon_1$, in opposite, decreases very steeply.

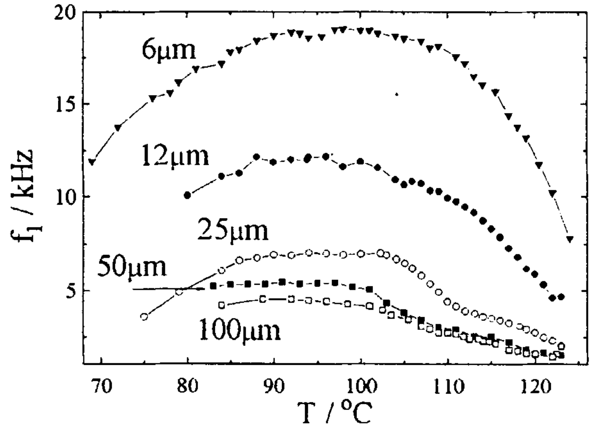


FIGURE 3 Temperature dependences of relaxation frequencies f_1 of mode 1 for all studied sample thicknesses.

For mode 2, both f_2 *(T)* and $\Delta\varepsilon_2(T)$ exhibit remarked anomaly around the inversion temperature T_i. Similarly as for mode 1, f_2 increases and $\Delta\varepsilon_2$ decreases with decreasing sample thickness. The value of α_2 =0.4÷0.5 shows a polydispersive character of mode 2.

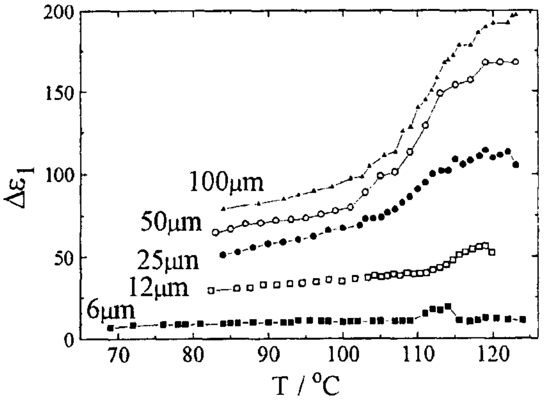


FIGURE 4 Temperature dependences of dielectric strengths $\Delta\varepsilon_1$ of mode 1 for all studied sample thicknesses.

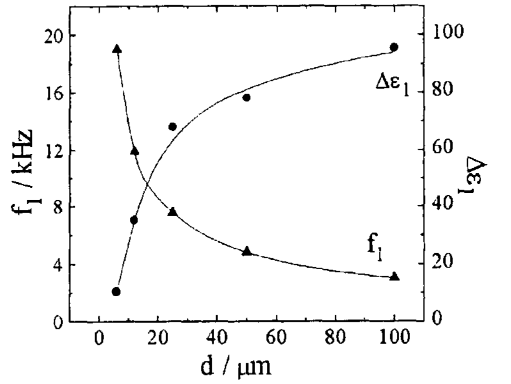


FIGURE 5 Relaxation frequency and dielectric strength of mode 1 in dependence of the sample thickness *d*, determined for temperature *T* =90°C.

DISCUSSION AND CONCLUSIONS

In the dielectric response two modes have been found in the SmC* phase, which can be also detected in the frequency dispersion of electrooptic effect. This fact establishes that both modes are director vibrations.

The higher frequency mode (**mode 1**) is easily suppressed by a bias field, which is a typical behaviour of the Goldstone mode connected with the existence of the helicoidal structure. On the other hand, mode 1 exists even in the temperature range, where the helix is spontaneously unwound due to helix twist inversion phenomenon. Moreover, the relaxation frequency f_l as well as the dielectric strength $\Delta\varepsilon_l$ of this mode is strongly thickness dependent. A similar thickness dependence has been also found in Ref. 6. This is the reason why we suppose that generally, in mode 1 contributions of two collective processes are present, namely of the Goldstone and the thickness modes. When the helix is unwound the thickness mode gives the only contribution to mode 1. The extinction of the Goldstone mode is responsible for decrease of $\Delta\varepsilon_l$ below T=105°C (see Fig. 4). The thickness dependence of mode 1 is due to the thickness mode contribution, for which it has been found[7] $\Delta\varepsilon_l \sim d^{1/2}$. A monodispersive character of mode 1 shows that the relaxation frequencies of both modes are close to each other and have the same thickness dependences.

The dielectric strength of the low frequency **mode 2** is higher than that of mode 1 and has a strong thickness dependence of the same character as mode 1. Moreover, it slightly increases under bias fields. Such a mode has been found for the first time and its origin is not understood at present.

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