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## DIELECTRIC DISPERSION IN FERROELECTRIC LIQUID CRYSTALS

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**Résumé.** — Nous avons effectué des mesures de relaxation diélectrique au voisinage de la transition ferroélectrique smectique-A smectique-C\* d'un cristal liquide chiral. Ces mesures ont permis d'obtenir pour le DOBAMBC la partie basse fréquence du spectre de fluctuation du paramètre d'ordre lors de cette transition. Dans la phase ferroélectrique (C\*), nous avons déterminé la fréquence et l'amplitude à vecteur d'onde nul du mode de Goldstone qui conserve la symétrie. Sa fréquence ne dépend que très légèrement de la température; elle décroît en approchant de la transition par températures croissantes tandis que son amplitude diminue d'un ordre de grandeur et semble tendre vers 0.

**Abstract.** — The low frequency order parameter fluctuation spectrum of chiral DOBAMBC has been studied at the ferroelectric smectic A  $\rightarrow$  C transition by dielectric relaxation spectroscopy. The frequency and the dielectric strength of the symmetry recovering Goldstone mode for  $q = 0$  in the ferroelectric phase have been determined. The frequency is approximately temperature independent, decreasing slightly when approaching  $T_c$  from below, while the intensity decreases for an order of magnitude and seems to go to zero at  $T_c$ .

In contrast to solid ferroelectrics, where a discrete symmetry group is broken at  $T_c$ , the helicoidal ferroelectric smectic A  $\rightarrow$  chiral smectic C transition involves the breaking of a continuous symmetry group. According to the Goldstone theorem [1], a symmetry recovering order fluctuation mode — the Goldstone mode — will therefore appear below  $T_c$  in addition to the soft mode.

In this work we report the first direct measurement of the low frequency order parameter fluctuation spectrum of a ferroelectric liquid crystal by dielectric relaxation spectroscopy. The frequency and temperature dependence of the dielectric constant of p-decyloxy-benzilidene p-amino-2 methyl butyl cinnamate (DOBAMBC) was measured close to the chiral smectic C  $\rightarrow$  smectic A phase transition. The sample was held between Nesa glass plates and was oriented by slowly cooling the sample from the isotropic phase in a magnetic field of 8.5 kgauss. The a.c. electric field was applied perpendicularly to the magnetic field. The temperature of the sample was stable within  $\pm 0.001$  °C and the transition temperature was  $T_c = 92.04$  °C. The phase transition of the sample was decreasing with time with the rate  $1.8 \times 10^{-3}$  °C/min. The frequency dependence of the dielectric constant was measured in the range between 20 Hz and 4 kHz. The low frequency conductivity for each temperature was determined and corresponding  $\epsilon''_{\text{con}}$  was subtracted from the measured  $\epsilon''$  values.

In figure 1 the real part of the dielectric constant

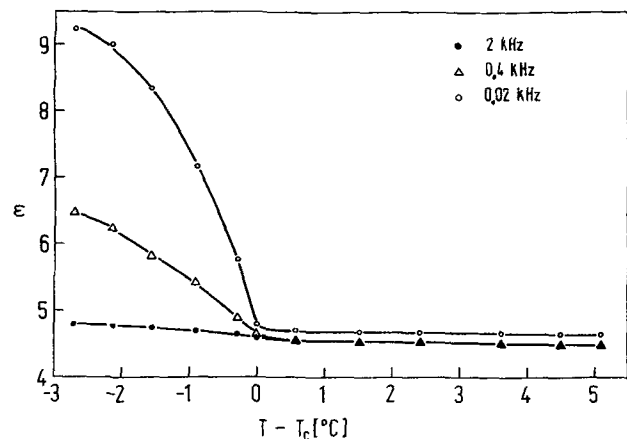


FIG. 1. — Real part of the dielectric constant as a function of the temperature at three different frequencies.

at three different frequencies is represented as a function of temperature. Above  $T_c$  the dielectric constant changes slightly with the frequency and temperature, while below  $T_c$  it is possible to observe a distinctive dispersion and temperature dependence. With the help of Cole-Cole diagrams we analyzed the experimental data (Fig. 2).

The relaxation frequency was determined from the expression

$$\epsilon(\omega) - \epsilon_{\infty} = \frac{\epsilon_0 - \epsilon_{\infty}}{1 + (i\omega\tau)^{1/1-h}}$$

$$f = \frac{1}{2\pi\tau}$$

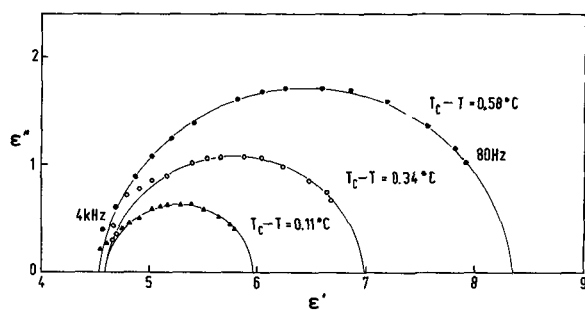


FIG. 2. — Cole-Cole diagrams in the chiral smectic C phase of DOBAMBC.

Since the measurement at one temperature takes at least 10 min and  $T_c$  of the sample is decreasing with time, the parameter  $h$  is different from zero. The temperature dependence of the relaxation frequency is represented in figure 3. The corresponding dielectric strength  $\epsilon_0 - \epsilon_\infty$  as a function of temperature is shown in figure 4.

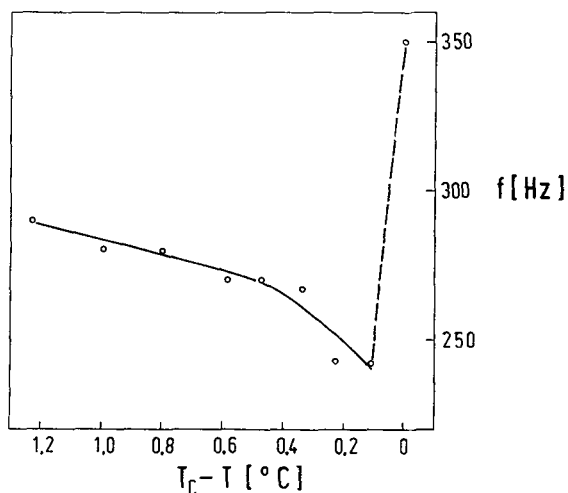


FIG. 3. — The dielectric dispersion frequency as a function of the temperature.

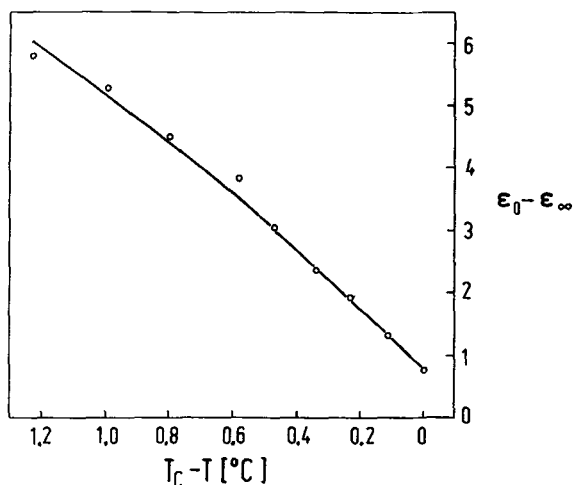


FIG. 4. — The dielectric strength  $\epsilon_0 - \epsilon_\infty$  as a function of the temperature.

The low frequency part of the polarization fluctuation spectrum is expected to have a soft mode behaviour above  $T_c$ ; while below  $T_c$ , it should be a superposition of the soft mode part and the Goldstone mode part. The soft mode frequency has been determined for  $T > T_c$  by Garoff and Meyer [2]. It is equal to  $\sim 10$  kHz at  $T - T_c = 0.1$  K and decreases critically when approaching  $T_c$ . This means that the soft mode could be observed in our frequency interval only in the very vicinity of  $T_c$ . On the other hand one expects to observe the Goldstone mode which should have a finite frequency because the dielectric response corresponds to the wave vector  $q = 0$ , while the critical wave vector  $q_0$  is determined by the pitch of the helix.

Neglecting the flexoelectric coupling between the polarization and tilt in the free energy expansion, one can express the relaxational frequencies of the soft mode ( $1/\tau_S$ ) and Goldstone mode ( $1/\tau_G$ ) in the following way [3]

$$\begin{aligned} \tau_S^{-1} &= \Gamma_1 [(T - T_c) + K_{33} q_0^2], & T > T_c \\ \tau_S^{-1} &= \Gamma_1 [2 \alpha (T_c - T) + K_{33} q_0^2], & T < T_c \\ \tau_G^{-1} &= \Gamma_1 K_{33} q_0^2, \end{aligned}$$

Using for the viscosity  $\Gamma_1$  the value [2] of  $100 \text{ m}^3/\text{J}$ , for the elastic constant  $K_{33}$  the value [4] of  $10^{-11} \text{ N}$  and  $5 \mu\text{m}$  for the pitch we get for the Goldstone mode frequency  $1/2 \pi \tau_G \cong 250 \text{ Hz}$  which is close to the measured value (Fig. 3). With decreasing temperature the measured Goldstone frequency is increasing slightly and reaches a constant value of  $\sim 500 \text{ Hz}$   $20^\circ\text{C}$  below  $T_c$  (not shown in Fig. 3). Close to the phase transition the frequency changes slowly with the temperature. In contradiction to the frequency, the dielectric strength of the mode changes for an order of magnitude in the same temperature interval (Fig. 4). With respect to frequency our experimental results are close to relaxation times determined by Pieransky [5] using a different method

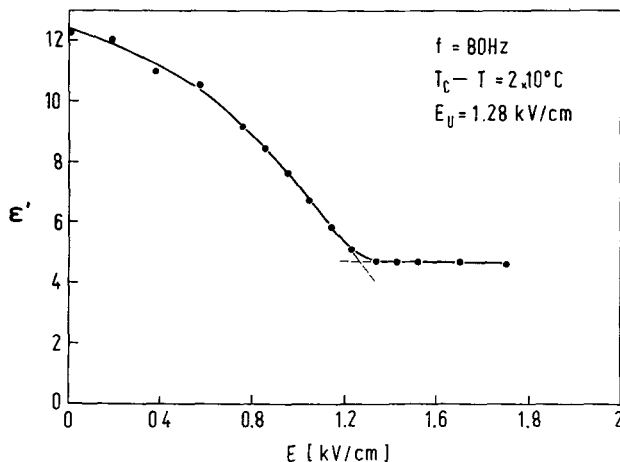


FIG. 5. — The determination of the unwinding field  $E_0$  at the frequency 80 Hz.

and to dielectrically measured values of Ostrowski (private communication). From the theoretical analysis (Žekš B., Levstik A. and Blinc R., this conference EP 50) it follows that strong temperature dependence of  $\varepsilon_0 - \varepsilon_\infty$  means that in the free energy expansion the flexoelectric interaction term is very important.

Figure 5 presents the electric field dependence of the real part of the dielectric constant in the chiral smectic C phase. Dielectric dispersion disappears when the d.c. electric field is higher than unwinding field  $E_u$ . Figure 6 represents the Goldstone mode for  $E < E_u$  and  $E > E_u$ . As the Goldstone mode represents the fluctuation of polarization perpendicular to the equilibrium polarization, it is possible to see from the right hand side of the figure 6 that when the helix is unwound the electric field is not coupled with the Goldstone mode. Hence, for  $E > E_u$  the

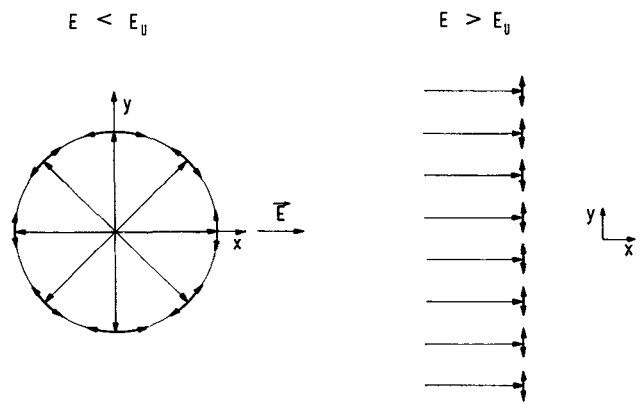


FIG. 6. — The representation of the Goldstone mode for  $E < E_u$  and  $E > E_u$ .

Goldstone mode does not contribute to the dielectric response.

### References

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