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Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl18

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I. Drevenšek [a], I. Muševič [a] & M. Čopič [a]
[a] J. Stefan Institute, University of Ljubljana, Ljubljana, Yugoslavia
Version of record first published: 24 Sep 2006.

To cite this article: I. Drevenšek , I. Muševič & M. Čopič (1991): Orientational Fluctuations in Ferroelectric Liquid Crystals in Static External Electric Field, Molecular Crystals and Liquid Crystals, 207:1, 199-203

To link to this article: http://dx.doi.org/10.1080/10587259108032099

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Mol. Cryst. Liq. Cryst., 1991, Vol. 207, pp. 199–203
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ORIENTATIONAL FLUCTUATIONS IN FERROELECTRIC LIQUID CRYSTALS IN STATIC EXTERNAL ELECTRIC FIELD

I.DREVENŠEK, I. MUŠEVIČ AND M.ČOPIČ
J. Stefan Institute, University of Ljubljana, Ljubljana, Yugoslavia

Abstract The effect of external static electric field on the dynamics of Goldstone orientational mode in ferroelectric liquid crystal was studied by photon correlation spectroscopy in the Sm C* phase of 4-(2'-methylbutyl) phenyl 4'-n-octylbiphenyl-4-carboxylate (CE8). In the field lower than the critical field necessary to unwind the helical structure of the crystal, the relaxation rate of the mode is almost field independent at all scattering wave vectors. At the critical field it suddenly increases and then continues to slowly increase with the field.

The spatial dispersion of the mode which is parabolic with minimum at helical wave vector q_0 in the undisturbed sample, remains unchanged below the critical field. Above the critical field the minimum in the dispersion at q_0 disappears.

INTRODUCTION

The thermal fluctuations of the director in the ferroelectric SmC* liquid crystalline phase can be decomposed into two parts: the fluctuations of the magnitude of the molecular tilt angle which exhibit the properties of a soft mode of the SmA-SmC* phase transition and the fluctuations of the azimuth of the tilt which can be considered the Goldstone mode of the transition.[1-2] Except close to the transition temperature the amplitude of the soft mode is many times lower than the amplitude of the Goldstone mode and material response due to the latter is dominant. The application of a static external electric field parallel to the smectic layers of the crystal freezes the fluctuations of the azimuth of the tilt and rapidly decreases the amplitude of the Goldstone mode.[3-6] On the other hand, static electric field deforms the helical structure of the crystal and affects also other properties of the modes.[7-9] Even some new, phonon-like modes can appear.[10] The purpose of our work was optical observation of the field effects on the spatial dispersion of the Goldstone mode.

A very simplified model, which takes into account only the dependence of the helical pitch on external electric field, can explain the main properties of the

observed effects. The Landau thermodynamic potential describing the SmA-SmC* transition gives for the inverse relaxation time of the Goldstone mode with respect to the scattering wave vector $\vec{k}$ a parabolic dispersion relation with a minimum at the helical wave vector q_o .[2]

$$\frac{1}{\tau}=\frac{K_3}{\gamma}(k_z-q_o)^2+\frac{K_\perp}{\gamma}k_y^2 \qquad (1)$$

In the external electric field the helical wave vector decreases until it vanishes at the critical field E_c. As a consequence the minimum of the dispersion shifts from $k_z=q_o$ to $k_z=0$.

$$\frac{1}{\tau}=\Delta(E)+\frac{K_3}{\gamma}k_z^2+\frac{K_\perp}{\gamma}k_y^2 \qquad (2)$$

A field dependent gap in the dispersion must also appear.[10]

EXPERIMENT

The measurements were performed in a mixture of 40% pure and 60% of racemic 4-(2'-methylbutyl) phenyl 4'-n-octylbiphenyl-4-carboxylate (CE8, BDH Ltd.) with a SmC* phase from 70.3°C to 81.0°C and a helical pitch around 5μm. The samples, prepared between clean glass plates, were 50μm thick and homeotropically aligned with a lecithin alignment layer. Homeotropic alignment was chosen because the previous measurements of the dispersion relation in the samples of purely chiral material had verified that the homogeneity of the structure in homeotropic alignment was better than in the planar cells. Four silver electrodes, wide compared to the sample thickness, were incorporated into the glass plates and provided a homogeneous electric field up to the 100kV/m in their center where the optical beam was passing through the liquid crystal (Fig.1). The temperature of the sample was stabilized to ±0.005K with a two stage temperature controller which contained glycerine as a heat exchanging as well as index matching material.

We studied the dynamics of the thermal director fluctuations with photon correlation spectroscopy. A He-Ne laser with a 5mW output power was passing the sample polarized as an ordinary beam (Fig.2). The scattered light with the orthogonal, extraordinary polarization, was detected by a photomultiplier connected to an autocorrelator. The relaxation time of the

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Goldstone mode was obtained from autocorrelation function by fitting to a single exponential decay. The measurements were performed at temperatures 0.1K to 1.0K below the phase transition temperature where the relaxation time of the soft mode , determined by our previous experiment,[11] is about three orders of magnitude shorter than the relaxation time of the Goldstone mode and the soft mode contribution to the autocorrelation function on a time scale of the Goldstone mode can be neglected.[11]

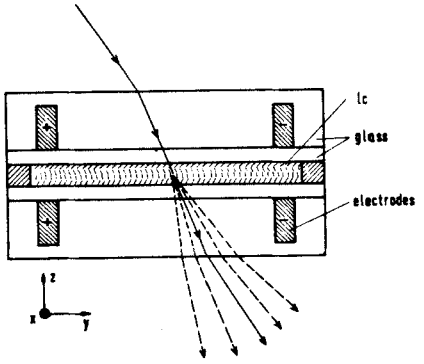

Fig.1. The configuration of the sample and the electrodes.

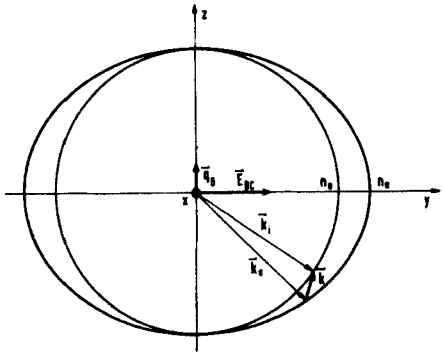

Fig.2. Scattering geometry.

RESULTS AND DISCUSSION

The dependence of the inverse relaxation time of the Goldstone mode on the scattering wave vector in two different external electric fields is represented on Fig. 3. In the field of magnitude $E=0.8E_C$ no evident effects on the dispersion were observed. In the field two times the critical field the inverse relaxation time of the mode strongly increases while the shift of a minimum of the dispersion toward $k_Z=0$ is not clear due to the large inaccuracy in the determination of the relaxation time. The main reason for it is strong elastic scattering of light from

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field induced disclinations and defects, which masks the contribution from inelastic scattering.

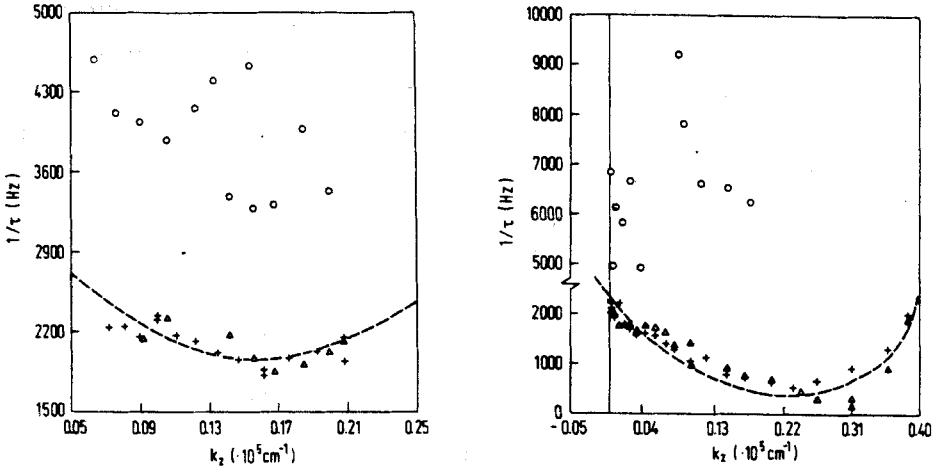

Fig.3. a)The dependence of inverse relaxation time of the Goldstone mode on scattering wave vector $k=(0, k_y=38q_o, k_z)$ in external electric field +_E=0V/m, Δ_E=34kV/m=0.8Ec, o_E=84kV/m=2Ec at $T=T_c-0.54K$. b) The dependence of inverse relaxation time of the Goldstone mode on oblique scattering wave vector $k=(0, k_y, k_z)$ in external electric field +_E=0V/m, Δ_E=34kV/m=0.8Ec, o_E=84kV/m=2Ec at $T=T_c-0.54K$.

To determine the dependence of the Goldstone mode relaxation time on the external field we choose some scattering wave vectors where the elastic scattering was minimal. The critical field was determined by the disappearance of the Bragg peaks. Fig. 4 shows the results at two scattering wave vectors. In the field below the critical field the inverse relaxation time of the Goldstone mode increases very slowly with the field. At the critical field the step rise appears. Above the critical field the inverse relaxation time continues to increase linearly with the field. A hysteresis was observed comparing the results in increasing and in decreasing field. It is probably due to slowly increasing ion concentration in static electric field, which through screening decreases the internal field at fixed external voltage.

Our experiment showed that static external electric field significantly changes the relaxation time of the Goldstone mode in ferroelectric liquid crystal only when the field is stronger than the critical field and the helical structure is unwound. Near the critical field the inverse relaxation time rapidly increases and becomes linearly field dependent. Further investigations, taking care of the field

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induced defects, are necessary to obtain more clear dispersion relation close and above the critical field.

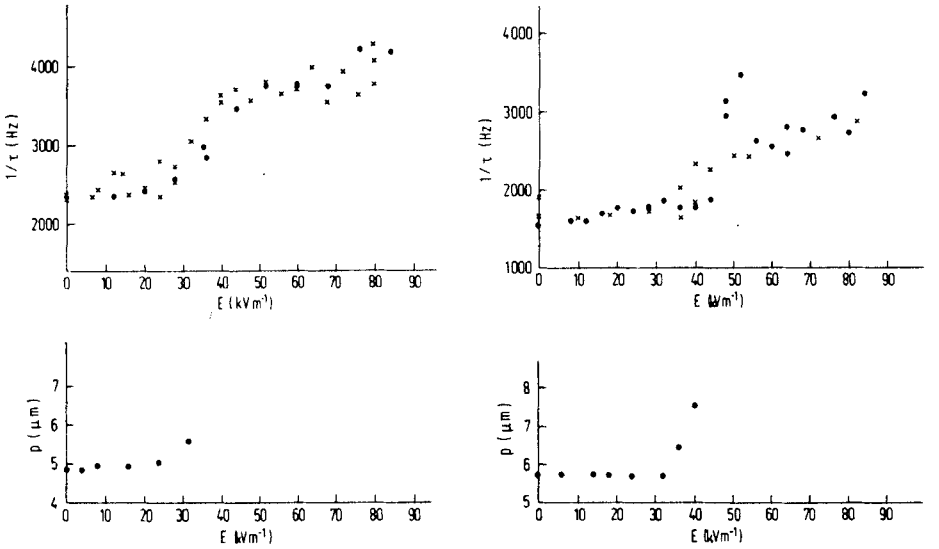

Fig.4. a) The inverse relaxation time of the Goldstone mode as a function of external electric field at k=(0, 0.67, 0)q_o and the dependence of helical pitch p=2π/q_o on external field at T=T_c−0.37K. o_Increasing, x_decreasing of the field.
b) The inverse relaxation time of the Goldstone mode as a function of external electric field at k=(0, 0.67, 1)q_o and the dependence of helical pitch p on external field at T=T_c−0.12K. o_Increasing, x_ decreasing of the field.

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