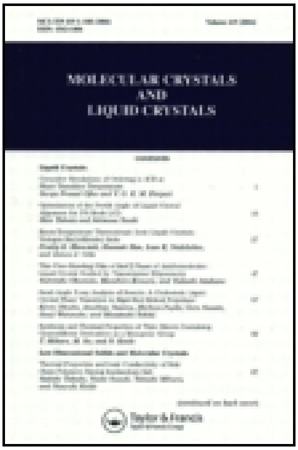

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Dynamic Light Scattering Near the Smectic A—Smectic C Phase Transition in Thin Planar Cells

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DYNAMIC LIGHT SCATTERING NEAR THE SMECTIC A - SMECTIC C* PHASE TRANSITION IN THIN PLANAR CELLS

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__Abstract__ Using the quasielastic light scattering we have studied the order parameter dynamics close to the SmA-SmC* phase transition in thin cells of ferroelectric liquid crystals, with planar boundary conditions. As a result of confinement, the bulk soft mode splits into two modes, whereas from the thickness dependence of the relaxation rate of the phase mode we measure the diffusion coefficients. The appearance of the chevron structure is manifested in the temperature dependence of the relaxation rate of the phase mode.

INTRODUCTION

Dynamic effects of the molecular reorientations in confined geometry near the SmA-SmC* phase transition have been the subject of considerable recent interest.[1-3]. The collective fluctuations of the orientational order in the SmC* phase can be derived in the frame work of the Landau theory of the second order phase transitions[4]. In the bulk SmC* phase, thermal fluctuations of the director can be decomposed into the fluctuations of the magnitude and the phase of the tilt angle. The fluctuation of the amplitude of the tilt angle is called the amplitude mode and the fluctuation of the phase of the tilt angle is called the phase or Goldstone mode. In the bulk SmA phase the collective fluctuations of the order parameter are described by a double degenerate soft mode that critically slows down at the transition point T_c.

The helical structure of the bulk SmC* phase strongly influences the properties of the dispersion relation of the amplitude and phase modes. Here the phase mode has zero relaxation rate at a finite wave vector $q_c = 2\pi/p$ in the direction of the helix axis, where p is the period of the helical structure. On the other hand, the amplitude mode, which represents a local departure of the magnitude of the SmC* order parameter from its equilibrium value, has a finite relaxation rate at all wave vectors, but again has a minimum at q_c.[4]

It is well known that the helical structure of the equilibrium bulk SmC* phase can be unwound in thin samples with planar boundary conditions[1,5]. The purpose of the present work was to observe by light scattering the effects of the finite size on the orientational fluctuations in thin samples with the unwound helix .

THEORY

In the laboratory frame, the relaxation rate of the soft mode in the bulk SmA phase for $q_x << q_z$ is given by[4]

$$\frac{1}{\tau_A}=\frac{\alpha}{\gamma}(T-T_c)+\frac{K_3}{\gamma}(q_z-q_c)^2+\frac{K_+}{\gamma}q_x^2 \tag{1}$$

whereas in the bulk SmC* the relaxation rates of the amplitude and phase modes for small q_x are[4]

$$\frac{1}{\tau_A}=\frac{2\alpha}{\gamma}(T_c-T)+\frac{K_3}{\gamma}(q_z-q_c)^2+\frac{K_+}{\gamma}q_x^2 \tag{2}$$

$$\frac{1}{\tau_P}=\frac{K_3}{\gamma}(q_z-q_c)^2+\frac{K_+}{\gamma}q_x^2 \tag{3}$$

Here α is a constant measuring the SmC* order parameter susceptibility, K_3 is the twist orientational elastic constant, $K_+=(K_1+K_2)/2$, K_1 beings splay and K_2 bend elastic constants, γ is the effective rotational viscosity and $q_y=0$. One should note that in the laboratory frame, the dispersion relation as a function of the scattering vector is shifted by q_c.

Let us consider a plane parallel sample with planar boundary conditions with thickness L small enough so that the helix is unwound. We assume a simple bookshelf structure with the tilt of the molecular director in the z-y plane. We also neglect the polar contribution of the surface anchoring energy so that no static splay is present in the sample.

Finite size of the sample in the x- direction has the important consequence that only discrete values of the q_x component are possible for the director fluctuations. In the limit of strong anchoring, when the director at the two surfaces is fixed in the y-z plane at the tilt angle θ with respect to the z-axis, the allowed eigenvalues for the q_x are given by

$$q_{xn}=\frac{n\pi}{L}$$

As far as the cell thickness remains finite q_{xn} is non-zero and therefore the soft mode splits into two modes with relaxation rates

$$\frac{1}{(\tau_n)_s}=\frac{\alpha(T-T_c)}{\gamma}+\frac{K_3}{\gamma}q_z^2+\frac{K_1}{\gamma}q_{nx}^2 \tag{4}$$

$$\frac{1}{(\tau_n)_b}=\frac{\alpha(T-T_c)}{\gamma}+\frac{K_3}{\gamma}q_z^2+\frac{K_2}{\gamma}q_{nx}^2 \tag{5}$$

The finite size effects then left the degeneracy of the soft mode. However, usually K_1 and K_2 are very close together therefore it is difficult to measure the splitting of the soft mode. The relaxation rate for the amplitude and the phase modes in the SmC* phase are given by

$$\frac{1}{(\tau_n)_A}=\frac{2\alpha(T_c-T)}{\gamma}+\frac{K_3}{\gamma}q_z^2+\frac{K_1}{\gamma}q_{nx}^2 \tag{6}$$

$$\frac{1}{(\tau_n)_P}=\frac{K_3}{\gamma}q_z^2+\frac{K_2}{\gamma}q_{nx}^2 \tag{7}$$

The measured quantity in dynamic light scattering experiments is the scattered field autocorrelation function. For a given wave vector $\vec{q}$ when the dimensions of the scattering volume are comparable or smaller than $1/|q|$, the wave vectors are no longer strictly conserved in the scattering process and fluctuations at wave vectors different from $\vec{q}$ also contribute. In particular, in our case the x-dimension of the scattering volume is small, so that the scattered field correlation function contains contributions from many transverse orientational modes;

$$g^{(1)}(\tau,\vec{q})=C\sum_n\left\langle\left|\xi_n(q_z,q_{xn})\right|^2\right\rangle\frac{\sin^2(q_{xn}-q_x)\frac{L}{2}}{(q_{xn}-q_x)^2}\; e^{\frac{-t}{\tau_n(q_z)}} \tag{8}$$

The averages $\langle|\varsigma_n(q_z)|\rangle$ are given by the equipartition theorem and are proportional to the $k_B T\tau_n(q_z)$ where k_B is the Boltzmann constant and C is a constant.

The main contribution to Eq. 8 comes from the mode with q_{xn} closest to q_x. The measured autocorrelation functions are nearly single exponential functions. Therefore for the experimentally determined relaxation rates, Eq. 7 is approximately valid

EXPERIMENTAL RESULTS

We measured the temperature and the thickness dependences of the autocorrelation functions of the intensity of the scattered light with an ALV-5000 digital multi-tau autocorrelator, in a Rayleigh scattering set-up. All measurements were performed in the heterodyne regime by mixing a part of the laser light with the scattered light. A HeNe (λ= 633nm) NEC laser was used as incident light. We used wedge-type cells with thickness of 0.4 μm to 4 μm filled with ferroelectric liquid crystals [4-(2'-methyl butyl)phenyl 4'-n-octylbiphenyl-4-carboxylate] (CE8) and BDH** mixture SCE9, in our measurements.

The incoming beam was perpendicular to the sample and the scattered light was collected at a scattering angle θ. The rubbing direction of the sample surface was in the scattering plane. The incoming polarization was chosen to be ordinary, that is perpendicular to the scattering plane, and the scattered polarization was extraordinary, i.e. in the scattering plane, as is necessary for the observation of the phase mode[6]. In this scattering geometry, the q_z component of the scattering vector goes to zero with the scattering angle going to zero, whereas the q_x component remains finite. Its value in SCE9 at zero scattering angle is $q_x = 2\times10^6$ m^{-1}, and depends on the difference of the reflective indices.

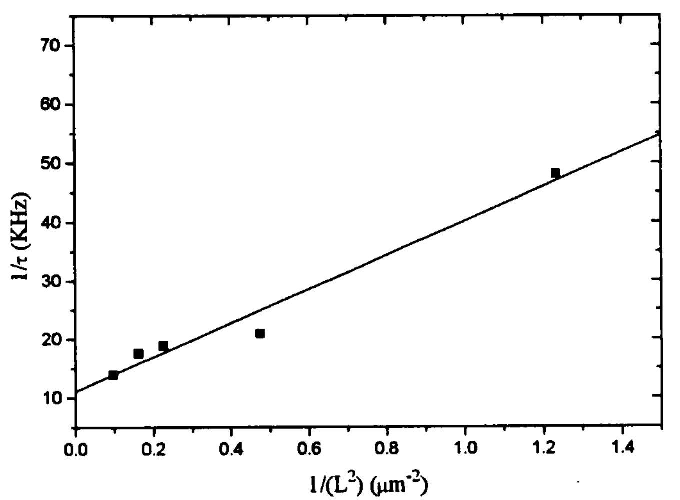

FIGURE 1 The relaxation rate of the phase mode at the transition point versus thickness in SCE9. The line is a fit to Eq.7 with $q_z = 6.77\times10^6$ m^{-1} and $q_x = 0$

In order to investigate the relaxation rates of the phase mode given by Eq.7, we plot in Fig.1 the measured relaxation rates as a function of the inverse square of the thickness

($1 / L^2$). Here the data was taken very close to the phase transition. The relaxation rate can be fitted to a line as one expects from the Eq.7. The slope of the line gives the diffusion coefficient $K_2 / \gamma = 2.9 \times 10^{-9}$ m^2s^{-1} that is in a very good agreement with our measurements in bulk sample[4] and is of the same order of magnitude as observed in other FLC materials[7].

We also measured the relaxation rate of the phase mode as a function of the cell thickness in CE8.(Fig.2). At a critical thickness of about 1μm for CE8 the helix is unwound and the relaxation rate of the phase mode increases by decreasing thickness as one expects from Eq.7. The line shows a fit of the Eq.7 to the measured data. The slope of this line gives the diffusion coefficient $K_2 / \gamma = 3.3 \times 10^{-9}$ m^2s^{-1} for CE8 that is in agreement with the previously measured values of diffusion coefficients in the bulk[4].

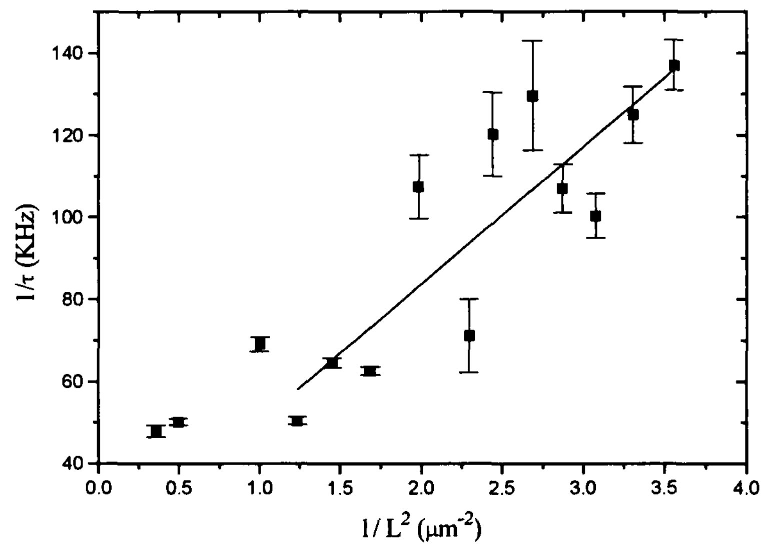

FIGURE 2 The thickness dependence of the relaxation rate of the phase mode in CE8. The critical thickness for this material is ≈ 2μm. The line shows a fit to the Eq. 7. The data were taken in a geometry $q_x = 0$, $q_z \neq 0$ and gives $K_2/\gamma = 3.3 \times 10^{-9}$.

The measured temperature dependence of the relaxation rate of the elementary excitations close to the SmA-SmC* phase transition in bulk samples is shown in Fig.3. It can be seen that in the SmC* phase close to T_c the relaxation rate has a peak and departs from the Eq.3. It is well known[4] that in these materials the helical pitch is temperature dependent. It has a maximum value about one degree below T_c and then slowly decreases by reducing the temperature. Therefore by increasing the pitch the wave vector q_c

decreases and the relaxation rate of the phase mode for $q_z<q_c$ increases according to Eq.3.

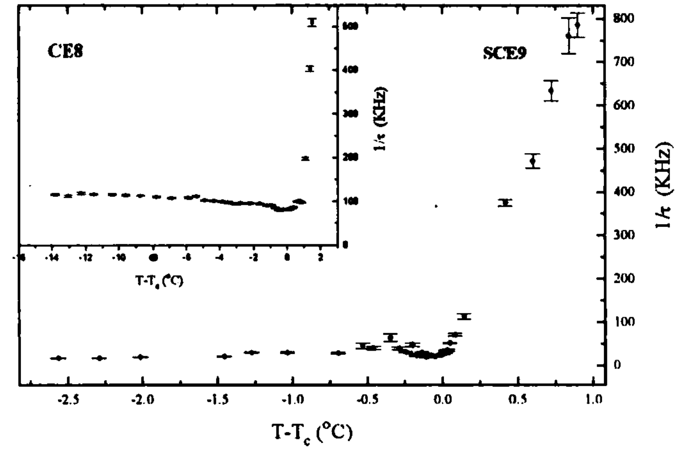

FIGURE 3 Temperature dependence of the elementary excitations close to the phase transition in thick samples of the CE8 and SCE9. (CE8: Homeotropic ,$q_x=7\times 10^6$ m^{-1} , $|q_z|=1\times10^6$ m^{-1}, SCE9: Planar, $q_x=0.2\times10^6$ m^{-1} , $q_z=6.4\times10^6$ m^{-1})

In the thin cells with homogeneous structure of the director field in the SmC* phase, the situation is different (Fig. 4). The relaxation rate of the phase mode gradually increases by reducing the temperature. This increase can not be attributed to the temperature dependence of the period of the helix, because the sample is unwound.

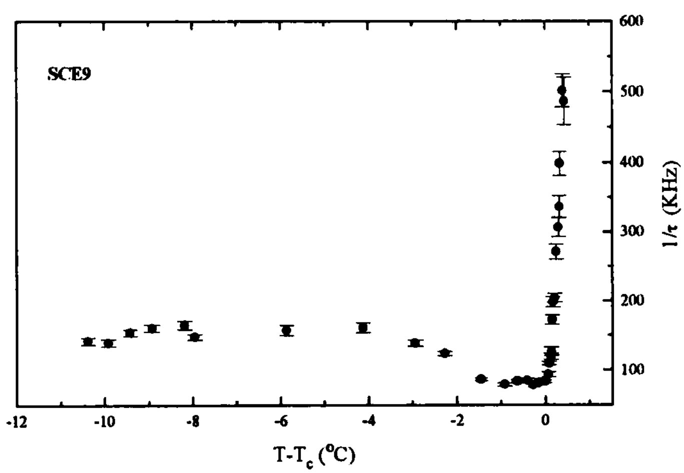

FIGURE 4 The temperature dependence of the elementary excitations close to the phase transition in SCE9 (SCE9: Planar, $q_x\approx0.$, $q_z=6.7\times10^6$ m^{-1} , L=2.6 μm)

We explain this rise of the relaxation rate of the phase mode by assuming that the sample has chevron structure. From the light scattering point of view, each branch of a symmetric chevron structure can be considered as a cell that is rotated by the layer tilt angle δ and its thickness is reduced to one half. In most materials δ increases with decreasing temperature. This changes the incident and the scattering angles and consequently q_z, which is reflected in the change of the relaxation rate of the phase mode.

As you can see from Fig. 4, the relaxation rate of the phase mode increases from 80 KHz to ≈140 KHz in a temperature range of 10 degrees. In a typical ferroelectric liquid crystal, the layer tilt angle for this range of temperature, changes about 18 degrees (DOBAMBC)[8]. As we mentioned before, the change of the layer tilt angle ultimately leads to the change of the q_z and consequently changes the relaxation rate of the phase mode.

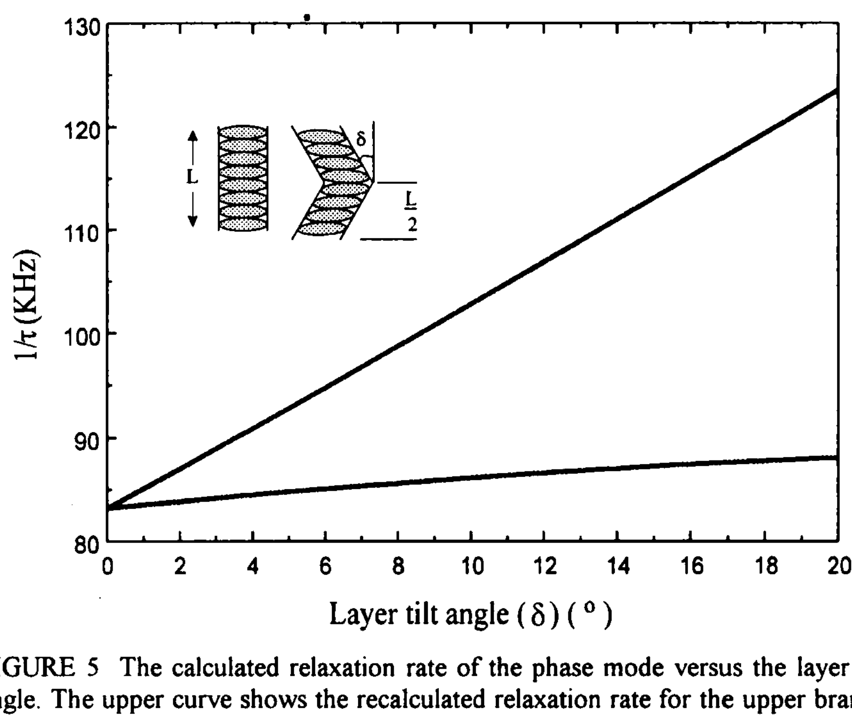

FIGURE 5 The calculated relaxation rate of the phase mode versus the layer tilt angle. The upper curve shows the recalculated relaxation rate for the upper branch of the chevron whereas the lower curve presents the calculated contribution of the lower branch The chevron layer is assumed symmetric. (The cell thickness L= 2.6μm, $K_2/\gamma = 2.9\times10^{-9}\, m^2S^{-1}$, $K_3/\gamma = 6.7\times10^{-10}\, m^2S^{-1}$)

In Fig.5 we plot the calculated relaxation rate of the phase mode in a chevron unwound SmC* phase versus the layer tilt angle. It can be seen that for the upper branch of the chevron, the relaxation rate of the phase mode changes from ≈ 80 KHz to ≈ 120 KHz when the layer tilt angle decreases to 20 degrees, whereas the lower chevron branch almost remains constant. In experiment we measure an averaged relaxation rate from both branches of the chevron structure(Fig.4). We see that in the experiment we probably observe the contribution of the upper chevron branch..

The temperature dependence of the layer tilt angle is thus reflected in the temperature dependence of the phase mode, which allows us to calculate the chevron tilt angle.

CONCLUSIONS

We have shown that in thin cells with planar boundary conditions, the transverse fluctuation modes are discrete. In the thin unwound SmC* cells the dispersion of the phase mode is not gapless and remains finite at the phase transition.

From the thickness dependence of the relaxation rate of the phase mode we have calculated the diffusion coefficients which are in very good agreement with bulk values. It seems that the appearance of the chevron structure in the SmC* phase manifests in the temperature dependence of the relaxation rate of the phase mode. It is thus possible to estimate the temperature dependence of the layer tilt angle by use of the measured temperature dependence of the relaxation rate of the phase mode.

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