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Visco-elastic modes in some ferroelectric liquid crystals

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We have studied the Goldstone-mode of three ferroelectric materials near the chiral smectic C (SmC*)–smectic A phase transition. The elastic constants corresponding to the **C** director in the SmC* phase have been evaluated from the dynamic light scattering measurements. In the material SCE6 we find a slow relaxing mode in addition to the normal Goldstone-mode. This slow relaxation is present only in the SmC* phase and vanishes in the smectic A phase.

1. Introduction

Chiral smectic C liquid crystal (SmC*) has a helical structure of layers having uniformly tilted molecules. The director makes a constant angle with the layer normal and rotates uniformly from one layer to another. The tilt θ and the azimuthal angle ϕ of the director are the amplitude and phase of the order parameters in the SmC* phase respectively. The temperature fluctuations of the director lead to fluctuations in the tilt and azimuth giving rise to strong scattering of light. Dynamic light scattering (wherein one looks at the temporal correlation of the scattered light from the sample) is a useful technique for studying the fluctuations at the SmC*–SmA transition (T_c) and in the SmC* phase [1–4]. The soft mode is associated with the fluctuations in the tilt angle and the Goldstone-mode is associated with the fluctuations in the azimuth [1, 5]. The fluctuation in tilt involves change in layer spacing and costs more energy for thermal excitation. Hence the soft mode can only be observed near the SmC*–SmA transition. Far away from the transition, the soft mode is known to be suppressed by the Goldstone-mode. In the presence of a biased electric field it is possible to observe the soft mode inside the SmC* phase [4].

In the vicinity of the direct beam for the sample aligned in the Bragg mode and in the vicinity of any diffraction order in the phase grating mode, the fluctuations in the intensity will reveal these modes. At a given temperature the measurement of relaxation time of the Goldstone-mode allows us to determine the ratio of viscosity coefficient to elastic constant. The determination of this parameter is very useful in the application of ferroelectric liquid crystals (FLCs) to optical devices such as light modulators [6, 7] and display devices.

In this paper, we describe dynamic light scattering studies carried out on three room temperature FLC materials. These were SCE6, ZL5014-100 and SCE13, and were obtained from E. Merck. The transition temperatures of these materials as obtained from a Mettler (FP82) hot stage are as follows:

SCE6

$$[\mathrm{SmC^*} \xleftrightarrow{54{\cdot}3^\circ} \mathrm{SmA} \xleftrightarrow{84^\circ} \mathrm{Chol} \xleftrightarrow{120^\circ} \mathrm{I}]$$

ZL5014-100

$$[\mathrm{SmC^*} \xleftrightarrow{65{\cdot}6^\circ} \mathrm{SmA} \xleftrightarrow{70^\circ} \mathrm{Chol} \xleftrightarrow{72^\circ} \mathrm{I}]$$

SCE13

$$[\mathrm{SmC^*} \xleftrightarrow{60^\circ} \mathrm{SmA} \xleftrightarrow{86^\circ} \mathrm{Chol} \xleftrightarrow{103^\circ} \mathrm{I}]$$

2. Experimental

We used samples aligned both in the homeotropic (smectic layers parallel to the glass substrate) and homogeneous (smectic layers perpendicular to the glass substrate) geometries. For the homeotropic alignment the cleaned glass plates were coated with ODSE (0·1% octadecyl triethoxy silane in tolune solvent) and cured at 150°C. Cells were made using 50 μm mylar spacers. The cells were filled with the sample in the isotropic phase and cooled slowly to the SmC* phase. The alignment was checked with the polarizing microscope. For the homogeneous alignment the glass plates were coated with polymide solution and cured at 300°C. They were rubbed in one direction and the cells made using 50 μm mylar spacer. The sample was filled into the cell in the isotropic phase and was cooled slowly in the presence of a magnetic field of 2·4 T applied along the rubbed direction of the glass plates.

The aligned sample was transferred to a Mettler (FP82) hot stage; the temperature of the sample was controlled to ±0·1°C. Light from a He–Ne 35 mW laser ($\lambda = 0{\cdot}6328\,\mu\mathrm{m}$) with polarization parallel to the scattering plane was incident normally on the sample;

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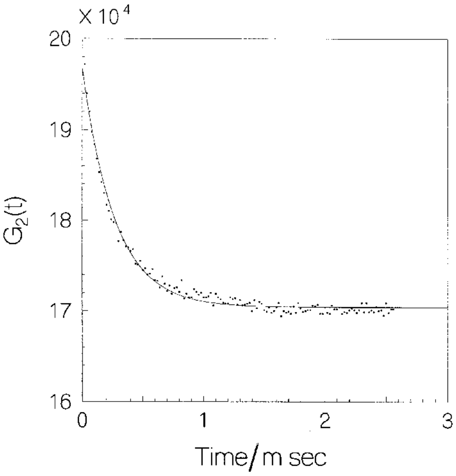

Figure 1. Intensity autocorrelation data for SCE6 at 2·0°C below the SmA–SmC* transition temperature (T_c). The data are fitted to a single exponential.

the scattered light was analysed perpendicular to the scattering plane. A photon correlator [Malvern (4700c) 8 bit] was used to acquire the intensity autocorrelation data. A typical plot of intensity autocorrelation data obtained for SCE6 is shown in figure 1. This intensity autocorrelation was fitted to an exponential function given by

$$G(t)=a+b\exp(-t/\tau) \qquad (1)$$

where τ is the relaxation time, a the background noise and b depends on the experimental conditions. Experiments were performed at various scattering angles for fixed temperatures. The experimental set-up was calibrated with previously obtained data on CE8 material [2]. The measured relaxation time for the CE8 was in good agreement with earlier reported values [1, 8–10].

The appearance of the diffraction pattern on cooling the sample from the SmA phase also indicates the phase transition to the SmC* phase. The pitch of the materials in the SmC* phase was obtained by measuring the angle of first order diffraction in the phase grating mode [11, 12].

3. Theory

Following a simple elastic theory [1, 2] one can write the relaxation time of the Goldstone-mode for the ferroelectric liquid crystal as

$$\frac{1}{\tau_G}=\frac{K_2}{\gamma}(q_z-q_0)^2+\frac{K_+}{\gamma}q_x^2 \qquad (2)$$

where q_0 is the wave vector of the helix and is given by the relation $q_0=2\pi/P$, P being the pitch of the SmC* phase. q_x and q_z are the scattering wave vector components along the x and z direction, respectively; γ is the viscosity coefficient. Here the twist axis is taken along the z axis in the laboratory frame. K_+ is given by $K_+=(K_1+K_3)/2$. K_1, K_2 and K_3 are analogous to the usual splay, twist and bend elastic constants, respectively.

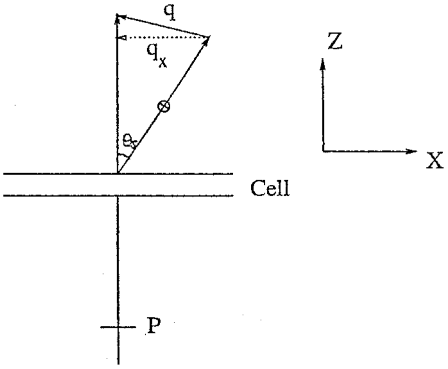

Figure 2. The scattering geometry used for a homeotropically aligned sample.

In the homeotropic geometry, for small scattering angles θ_s (see figure 2), we can write

$$q_z=|q|\sin(\theta_s/2) \quad \text{and} \quad q_x=|q|\cos(\theta_s/2) \qquad (3)$$

where $|q|=2k_i\sin(\theta_s/2)$, $k_i=2\pi/\lambda$. Here q is the scattering wave vector, k_i is the incident wave vector and λ is the wavelength of light. At a given temperature the relaxation time data at different scattering angle were fitted to equation (2) and the ratios of K_+/γ and K_2/γ were extracted.

4. Results

Temperature dependences of the helical pitch of the three materials are shown in figure 3. The pitch of SCE13 is almost constant in the SmC* phase. The temperature dependence of the relaxation time across the SmA–SmC* transition for homogeneously aligned

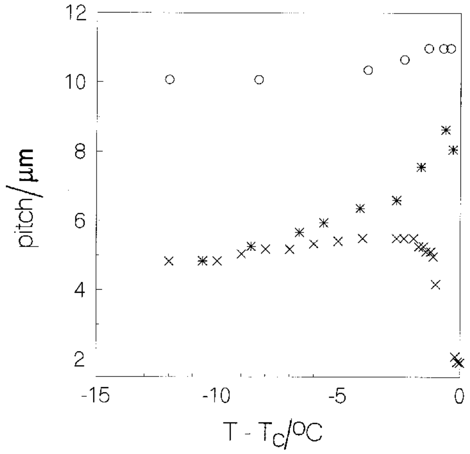

Figure 3. The measured pitch for the materials as a function of temperature. The symbols ×, * and ∘ represent the data for SCE6, ZL5014-100 and SCE13, respectively.

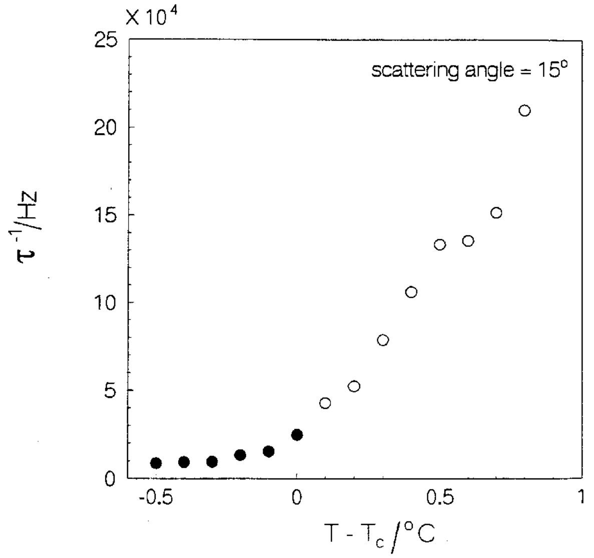

Figure 4. The inverse of the relaxation time τ^{-1} across the SmA to SmC* phase transition for SCE6. The open circles represent the inverse of the relaxation time corresponding to the degenerate mode in the SmA phase. The dark circles represent the Goldstone mode in the SmC* phase. The data were obtained for a homogeneously aligned sample at a constant scattering angle.

SCE6 is shown in figure 4. We find that the inverse of the relaxation time corresponding to the degenerate mode in the SmA phase drops sharply at the SmA–SmC* transition. One may notice that the Goldstone-mode in the SmC* phase has an almost constant relaxation time.

Scattering angle dependences of the inverse of the relaxation time of the Goldstone-mode in the homeotropically aligned SmC* phase for the three materials are shown in figures 5, 6 and 7. As expected from the simple elastic theory, the inverse of the relaxation time is dependent quadratically on the scattering angle in all three materials. Hence equation (2) can be fitted with the data to extract the values of K_+/γ and K_2/γ at different temperatures. The calculated values of K_+/γ and K_3/γ for the three materials are shown in tables 1, 2 and 3. The estimation of the errors in these visco-elastic coefficients were carried out using nonlinear fitting [13].

Table 1. Viscoelastic coefficients for the SCE6 at different temperatures.

$T-T_c$/°C	K_2/γ/cm^2 s^{-1}	K_+/γ/cm^2 s^{-1}
−0·3	$1{\cdot}08 \pm 0{\cdot}135 \times 10^{-6}$	$2{\cdot}82 \pm 0{\cdot}350 \times 10^{-7}$
−1·3	$4{\cdot}05 \pm 0{\cdot}848 \times 10^{-6}$	$5{\cdot}08 \pm 0{\cdot}722 \times 10^{-7}$
−2·3	$6{\cdot}36 \pm 0{\cdot}529 \times 10^{-6}$	$7{\cdot}33 \pm 0{\cdot}609 \times 10^{-7}$
−3·3	$7{\cdot}79 \pm 0{\cdot}569 \times 10^{-6}$	$8{\cdot}98 \pm 0{\cdot}655 \times 10^{-7}$
−4·3	$8{\cdot}43 \pm 0{\cdot}960 \times 10^{-6}$	$9{\cdot}72 \pm 0{\cdot}110 \times 10^{-7}$
−5·0	$7{\cdot}26 \pm 0{\cdot}619 \times 10^{-6}$	$8{\cdot}50 \pm 0{\cdot}724 \times 10^{-7}$
−9·3	$7{\cdot}24 \pm 0{\cdot}693 \times 10^{-6}$	$9{\cdot}16 \pm 0{\cdot}876 \times 10^{-7}$

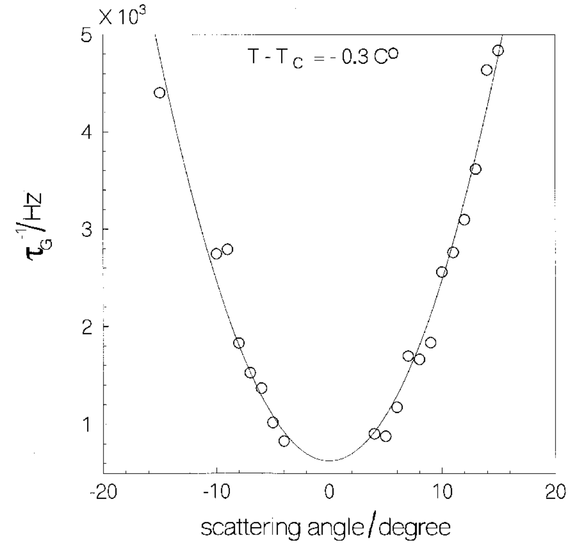

Figure 5. τ_G^{-1} corresponding to the Goldstone-mode relaxation as a function of scattering angle for SCE6 in the SmC* phase. The data are fitted to the quadratic function of scattering angle.

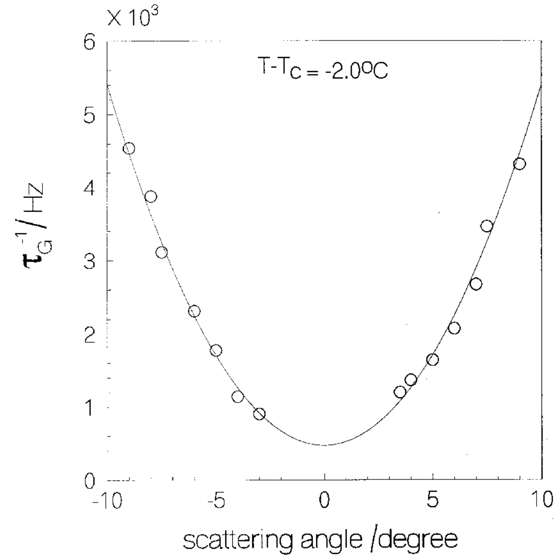

Figure 6. τ_G^{-1} corresponding to the Goldstone-mode relaxation as a function of scattering angle for ZL5014-100 in the SmC* phase.

We find that the values do not follow a regular trend with temperature. However, the values are of the same order of magnitude as those reported in the literature for FLCs [2, 8–10].

An interesting result of our study is the observation of a new relatively slow relaxation mode in SCE6 material, in addition to the Goldstone-mode, in a sample aligned in the homogeneous geometry. We have shown in figure 8, the intensity autocorrelation data taken for this slow mode and fitted to an exponential. The relaxation

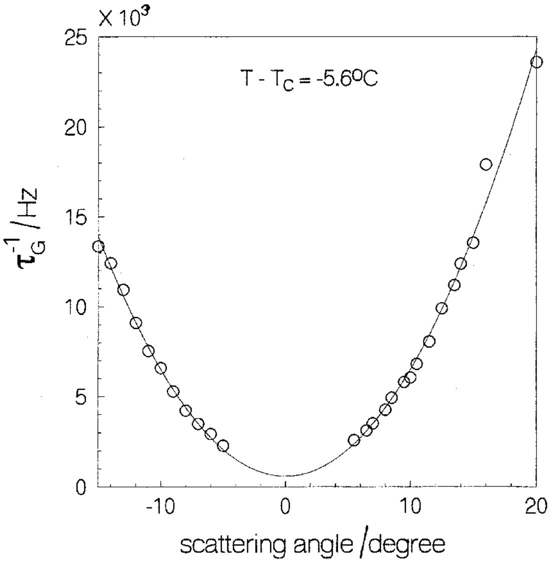

Figure 7. τ_G^{-1} corresponding to the Goldstone-mode relaxation as a function of scattering angle for SCE13 in the SmC* phase.

Table 2. Viscoelastic coefficients for the ZL5014-100 at different temperatures.

$T-T_c$/°C	K_2/γ/cm^2 s^{-1}	K_+/γ/cm^2 s^{-1}
− 1·0	$4{\cdot}55 \pm 1{\cdot}40 \times 10^{-6}$	$3{\cdot}57 \pm 1{\cdot}32 \times 10^{-7}$
− 1·6	$9{\cdot}91 \pm 1{\cdot}94 \times 10^{-6}$	$8{\cdot}35 \pm 1{\cdot}63 \times 10^{-7}$
− 2·6	$3{\cdot}59 \pm 2{\cdot}98 \times 10^{-6}$	$3{\cdot}50 \pm 2{\cdot}36 \times 10^{-7}$
− 3·6	$5{\cdot}62 \pm 1{\cdot}13 \times 10^{-6}$	$5{\cdot}63 \pm 1{\cdot}12 \times 10^{-7}$
− 4·6	$8{\cdot}42 \pm 1{\cdot}35 \times 10^{-6}$	$8{\cdot}70 \pm 1{\cdot}39 \times 10^{-7}$
− 5·6	$5{\cdot}19 \pm 1{\cdot}60 \times 10^{-6}$	$5{\cdot}59 \pm 1{\cdot}70 \times 10^{-7}$
− 10·6	$3{\cdot}49 \pm 1{\cdot}49 \times 10^{-6}$	$4{\cdot}63 \pm 1{\cdot}96 \times 10^{-7}$
− 15·6	$3{\cdot}37 \pm 1{\cdot}51 \times 10^{-6}$	$4{\cdot}76 \pm 1{\cdot}11 \times 10^{-7}$

Table 3. Viscoelastic coefficients for the SCE13 at different temperatures.

$T-T_c$/°C	K_2/γ/cm^2 s^{-1}	K_+/γ/cm^2 s^{-1}
− 1·0	$2{\cdot}22 \pm 0{\cdot}663 \times 10^{-5}$	$1{\cdot}27 \pm 0{\cdot}363 \times 10^{-7}$
− 2·0	$1{\cdot}36 \pm 0{\cdot}226 \times 10^{-5}$	$0{\cdot}81 \pm 0{\cdot}134 \times 10^{-7}$
− 3·0	$1{\cdot}95 \pm 0{\cdot}255 \times 10^{-5}$	$1{\cdot}18 \pm 0{\cdot}154 \times 10^{-7}$
− 4·0	$1{\cdot}89 \pm 0{\cdot}352 \times 10^{-5}$	$1{\cdot}16 \pm 0{\cdot}326 \times 10^{-7}$
− 5·0	$1{\cdot}34 \pm 0{\cdot}386 \times 10^{-5}$	$0{\cdot}83 \pm 0{\cdot}239 \times 10^{-7}$
− 8·7	$1{\cdot}86 \pm 0{\cdot}589 \times 10^{-5}$	$1{\cdot}16 \pm 0{\cdot}368 \times 10^{-7}$

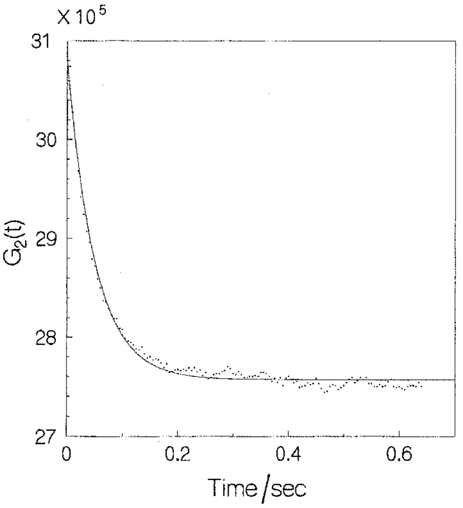

Figure 8. The intensity autocorrelation data of the slow relaxation mode for a homogeneously aligned SCE6 sample at $\theta_S = 15°$ and at 0·2°C below T_c.

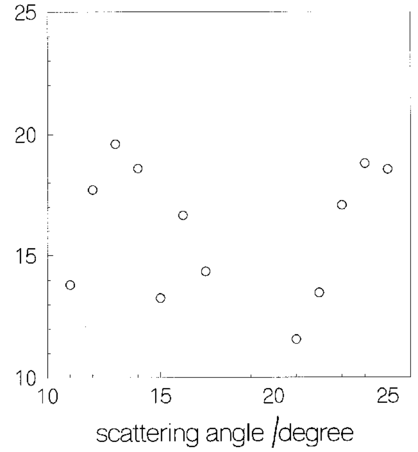

Figure 9. The inverse of the relaxation time of the slow mode as a function of scattering angle for SCE6 in the SmC* phase at 0·2°C below T_c.

time of this slow mode as a function of scattering angle is shown in figure 9. The relaxation time appears to be nearly independent of the scattering angle. It is worth noting that this slow relaxation mode is not observed in the homeotropically aligned sample and is only present in the sample aligned in the homogeneous geometry.

It is well known that a FLC with high polarization placed under an external biased electric field or confined in a restricted geometry can give rise to additional modes in an attempt to recover the broken helicoidal symmetry [4, 14, 15]. One can attribute the origin of the slow mode in SCE6 to the boundary effects which can deform the helical structure of the SmC* phase. However, this type of slow mode was not observed in the ZL5014-100 and SCE13 samples, even though the cell thicknesses used were comparable to that of SCE6. Hence the occurrence of this mode needs further study. It would be interesting to probe this mode as a function of sample thickness and in the presence of an external applied d.c. electric field.

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