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Preliminary communication

Optical investigation of the Goldstone mode in free-standing films of ferroelectric smectic C* phases

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An optical method for investigating the dynamics in free-standing films of ferroelectric liquid crystals in the range 10 to 10^5 Hz by means of optical reflectivity is presented. The experimental results were obtained in the linear-response regime applying electric a. c. fields of strengths less than $16\,V\,cm^{-1}$. By measuring the magnitude and phase of the dynamic reflectivity, a relaxation process was found at frequencies near 1 kHz originating from the Goldstone mode.

By application of low electric a. c. fields perpendicular to the smectic layer normal of a helically structured smectic C* phase, the helix is deformed. At low frequencies, the deformation takes place instantaneously. By increasing the frequency of the applied field, the region of relaxation is reached where the phase of the process will increase from zero to 90 degrees. At higher frequencies, the relaxation no longer takes place. This relaxation process of the helix is called the Goldstone mode.

In the bulk of ferroelectric liquid crystals, the Goldstone mode has already been measured intensively by dielectric spectroscopy [1–4]. Electro-optical investigations of the Goldstone mode in surface stabilized cells are known as well [5, 6].

In free-standing films the Goldstone mode cannot be detected easily by dielectric spectroscopy because the capacitance of very thin samples such as free-standing films is extremely small. But the relaxation process causes a dynamic variation of the optical anisotropy which can be observed by reflection spectroscopy with polarized light. We therefore preferred an optical method to study the Goldstone mode in free-standing films which is described below. In this work, the first experimental results of Goldstone mode measurements on free-standing films are reported. They are different from the kink switching found by Demikhov *et al.* [7] because we only slightly deformed the helix in order to determine the Goldstone mode relaxation time, whereas they unwound the helix and switched the *c*-director by $\pm 180°$.

In our experiments, the free-standing film is drawn in a frame of two fixed teflon holders and two movable brass blades coated with polyimide and serving as electrodes. The whole set-up is held at a given temperature stabilized to $\pm 0{\cdot}01$°C. The film is produced in the smectic A phase. Homogeneous films from 60 nm up to 5 μm in thickness can be drawn. Linearly polarized light from a xenon lamp is radiated perpendicularly onto the free-standing film. The reflected elliptically polarized light passes a 1/8 m grating monochromator (Oriel 77250) and the remaining intensity is detected by a photomultiplier (Hamamatsu 1P21) and a digital multimeter (KEITHLEY DMM119). The number of layers is determined from the reflection spectra as described in [8]. The number can be controlled by the amount of substance and the drawing velocity.

After the thickness of the smectic A film has been measured, the whole set-up is cooled to the smectic C* phase. The dynamic optical reflectivity R and the corresponding phase ϕ caused by the Goldstone mode are measured as a function of the frequency of an electric a. c. field applied to the brass gliders. To obtain a sufficiently high reflectivity, the tilt angle θ of the smectic C* phase should be rather large. Now the optical reflectivity R and the phase ϕ are modulated as a function of time. Firstly, at a given temperature, a dynamic reflectivity spectrum is scanned by applying a low frequency electric a. c. field to the film and detecting the dependence of R on the wavelength with a SR830 lock-in amplifier. The monochromator is tuned to a wavelength with a large dynamic reflectivity. At this wavelength and the given temperature, the frequency dependence of R and ϕ is measured for different strengths of the electric a. c. field. This is again done by using a

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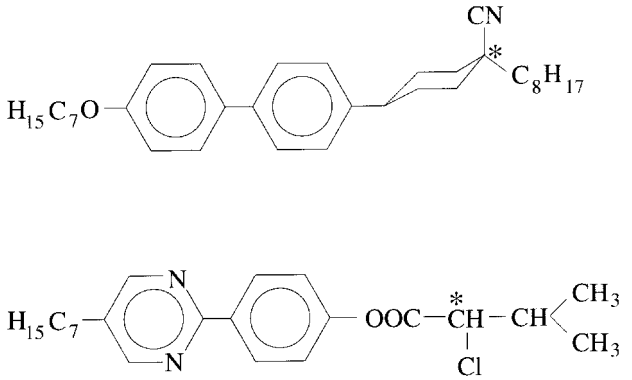

Figure 1. Structures of two chiral components of the mixture ZLI 3654 [9].

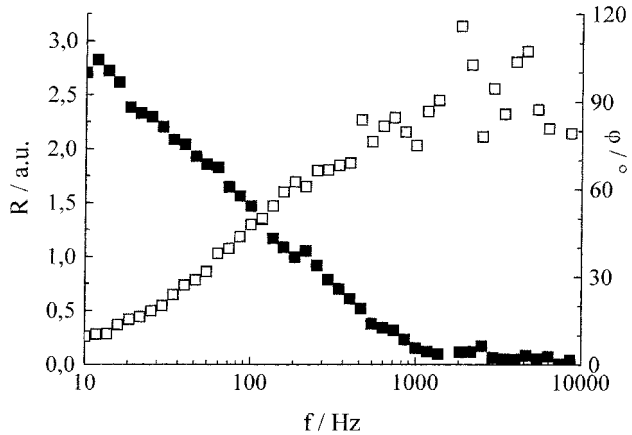

Figure 2. Frequency dependence of the dynamic reflectivity R (filled symbols) and the phase ϕ (open symbols) of a 1126 layer film at $T=54°C$ and with $\mathbf{E}=13{\cdot}6\,V\,cm^{-1}$.

lock-in amplifier locked to the first harmonic of the frequency of the a. c. electric field. The procedure is repeated at other temperatures and with different film thicknesses.

All of the measurements were made with a commercial ferroelectric liquid crystalline mixture (ZLI 3654, Merck/Hoechst) which consists of several compounds. Two of them responsible for the dielectric behaviour are shown in figure 1 [9]. The phase transition temperatures of ZLI 3654 (in °C) are Cr30 SmC* 64 SmA 80 N* 88 I.

Before a measurement of the Goldstone mode is started, the film is examined by means of polarizing microscopy in the ground state at zero field and in external electric fields of several frequencies and field strengths in order to exclude disclination lines (e.g. at zero field) and convective flow (above a critical field strength) and to get a homogeneous area larger than the diameter of the light beam. If a proper area is found, however, disclinations may move during the measurement, reducing the magnitude of R and thus mimicking a relaxation process. To avoid such artefacts, 10 to 20 identical measurements were made.

In figure 2 the frequency dependences of the reflectivity R and of the phase ϕ are shown for a 1126 layer film at 54°C and with a field strength of $13{\cdot}6\,V\,cm^{-1}$. The dynamic reflectivity $R(f)$ is large at low frequencies and decreases with increasing phase shift of the Goldstone mode relative to the external electric field. At high frequencies the Goldstone mode no longer takes place and the dynamic reflectivity falls to a value of zero. At frequencies $f>1\,kHz$ where R equals zero, the error in the phase increases because of the lock-in technique. The phase increases from 10 degrees at 10 Hz to 90 degrees at about 1 kHz. This is the frequency range of the Goldstone mode. To a first approximation, we assume a Debye type relaxation. From the inflexion point of $\phi(f)$ at 85 Hz we derive a relaxation time of 2×10^{-3} s which is of the same order of magnitude as the relaxation time of ZLI 3654 observed in cells.

In figure 3 the frequency dependences of R and of ϕ are shown for a 73 layer film at different temperatures. On decreasing the temperature the tilt angle θ increases ($\theta=17{\cdot}85°$ at 51°C, 10·7° at 59°C). Consequently, the difference between the refractive indices of the ordinary and extraordinary ray increases, which in principle would result in a higher value of the dynamic reflectivity at lower temperature. $R(f)$ is not modulated only by the frequency dependent collective dynamics. In this case R would decrease continuously with increasing frequency. As can be seen from figure 3, the curve $R(f)$ obviously also depends on other parameters [10, 11]. For instance, the difference in the effective refractive indices at zero

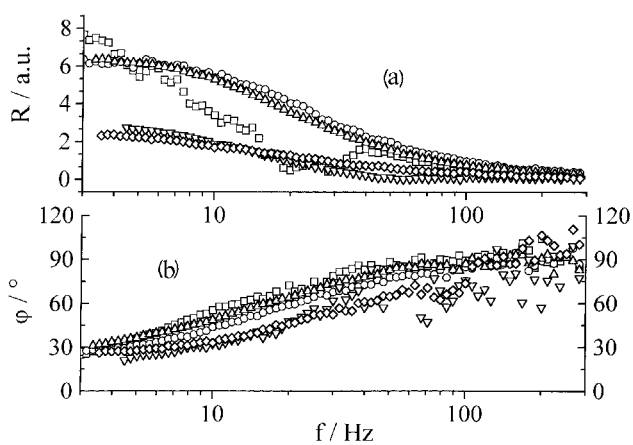

Figure 3. Frequency dependence of the dynamic reflectivity R (*a*) and the phase ϕ (*b*) of a 73 layer film at different temperatures with $\mathbf{E}=15{\cdot}4\,V\,cm^{-1}$; squares: $T-T_C=-25\,K$, circles: $T-T_C=-13\,K$, triangles: $T-T_C=-10\,K$, inverted triangles: $T-T_C=-3\,K$, rhombs: $T-T_C=-2\,K$.

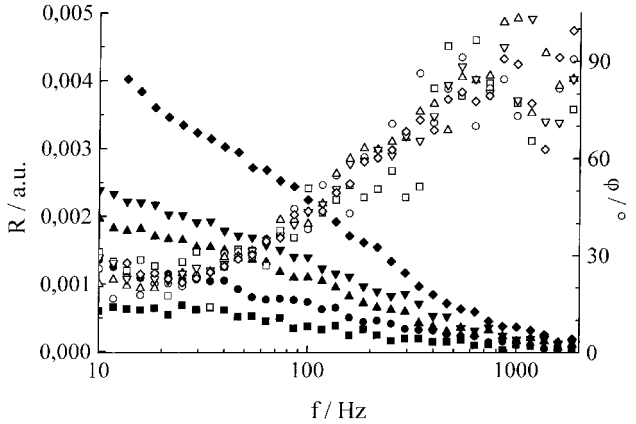

Figure 4. Frequency dependence of the dynamic reflectivity R (filled symbols) and the phase ϕ (open symbols) of an 891 layer film at $T=57°C$ with the electric field strength as a parameter; squares: $3\,V\,cm^{-1}$, circles: $4{\cdot}5\,V\,cm^{-1}$, triangles: $7{\cdot}5\,V\,cm^{-1}$, inverted triangles: $10\,V\,cm^{-1}$, rhombs: $16\,V\,cm^{-1}$.

field and at maximum or minimum field, respectively, contribute to the shape of $R(f)$. However, a possible influence of these effects is not relevant to the conclusions drawn from the phase measurement. $\phi(f)$ is not influenced by these other effects mentioned above which can be seen for $T-T_C=25\,K$ in figure 3. This is a necessary condition for the determination of τ from the inflexion point of $\phi(f)$. The figure also shows a slight shift of $\phi(f)$ to higher frequencies with increasing temperature, which indicates an acceleration of the Goldstone mode with increasing temperature.

In figure 4 the frequency dependences of the signals R and ϕ at different applied field strengths are shown for an 891 layer film at 57°C. The higher the electric field strength is, the stronger is R. In the range 3 to $16\,V\,cm^{-1}$ the measured relaxation frequency obviously does not depend on the field strength, as can be seen from $\phi(f)$. Consequently, the homogeneous films in this range of a. c. field strength are proper for a measurement of the pure Goldstone mode.

The results presented here show that the Goldstone mode which has been found in SSFLC cells is also present in free-standing films where no complete helix can be formed due to the small thickness of a film. This is an interesting observation because it allows a determination of the Goldstone mode relaxation frequency without the disturbing influence of surface anchoring which takes place in SSFLC cells.

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