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ELECTRO-OPTIC EFFECT IN AN ANTIFERROELECTRIC LIQUID CRYSTAL

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Abstract The antiferroelectric Goldstone mode in a free-standing film was investigated by means of the electro-optic measurement. The divergence of the relaxation time corresponding to the divergence of the helical pitch was observed. In comparison with the result obtained in a homogeneous cell the surface anchoring effect is discussed.

INTRODUCTION

The antiferroelectric Goldstone mode in the antiferroelectric phase (SmCA*) has been observed by the photon correlation spectroscopy,[1,2] the electro-optic measurement[3] and the nonlinear dielectric measurement.[4] In MHPOBC all the above measurements were carried out and the temperature dependence of the relaxation frequency was obtained. However, the experimental results are different; in the photon correlation spectroscopy the relaxation frequency at k=0 becomes zero at a certain temperature[1] while in the others a monotonic decrease of the frequency with decreasing temperature was observed.[4] In MHPOBC the wave number of the helix, q, becomes zero (the pitch diverges) to change the helical sense at α. 75°C[5] and so the corresponding divergence of the relaxation time τ is expected, since

$$1/\tau = (4\kappa q^2)/\gamma \ , \tag{1}$$

where q, κ and γ are, respectively, the wave number of the helix, the elastic constant and the viscosity. The discrepancy may come from the difference of the cells used in the

experiments; in the photon correlation spectroscopy homeotropic cells were used, while in the others homogeneous ones. The surface state may influence the helical structure.

In this paper, we study the electro-optic response in a free-standing film on the basis of a theory for the dynamics of relaxation modes in the antiferroelectric phase, and then discuss the surface anchoring effect.

ELECTRO-OPTIC EFFECT IN THE ANTIFERROELECTRIC PHASE

The dynamics of the antiferroelectric Goldstone mode under electric fields has been theoretically studied by Orihara and Ishibashi.[6] According to them, the soft and the Goldstone modes can be excited by an electric field, though they respond to the square of the field but not to the field itself. The change of the phase angle by the Goldstone mode, $\Delta\phi$, under a sinusoidal field $E=E_0\cos\omega t$ is written as

$$\Delta\phi = [c_2(\omega)\chi_{aG}(2q,0)\sin 2qz + \mathrm{Re}\{c_2(\omega)\chi_{aG}(2q,2\omega)\sin 2qz\cdot e^{i2\omega t}\}]\left(\frac{E_0}{2}\right)^2, \quad (2)$$

where the helical axis is taken along the z-axis, $c_2(\omega)$ and $c_2'(\omega)$ are coefficients depending slightly on frequency, and $\chi_{aG}(2q,\omega)$ is the linear susceptibility of the antiferroelectric Goldstone mode:

$$\chi_{aG}(2q,\omega) = \frac{\tau/\gamma}{1+i\omega\tau}. \quad (3)$$

From Equation (2), it is readily seen that the contribution of the antiferroelectric Goldstone mode to the transmitted intensity change in electro-optic measurements may be approximately expressed as

$$[a_0+a_2\mathrm{Re}\{\chi_{aG}(2q,2\omega)e^{i2\omega t}\}]E_0^2+[b_0+b_2\mathrm{Re}\{\chi_{aG}(2q,2\omega)e^{i2\omega t}\}+b_4\mathrm{Re}\{\chi_{aG}^2(2q,2\omega)e^{i4\omega t}\}]E_0^4, \quad (4)$$

where a_i and b_i are coefficients depending slightly on frequency. The experimental results will be analyzed on the basis of this equation.

EXPERIMENTAL AND RESULTS

The sample used was MHPOBC, which shows the phase sequence: SmC_A^*-SmC_γ^*-

SmC*-SmC$_\alpha$-SmA. In SmC$_A$* the helical pitch diverges so that the helical sense changes. The measurement was performed in SmC$_A$*, SmC$_\gamma$* and SmC*. A free-standing film was spread over a rectangular hole of a glass plate with electrodes for the application of an electric field parallel to the smectic layers. The sample was put between crossed polarizers and the polarizer direction was set at 45° with respect to the field direction. The sample cell was mounted in a hot stage (Mettler FP82) and illuminated by an Ar-ion laser at 514.5 nm. The transmitted light was detected with a photo-diode and the second-order and the fourth-order harmonics were measured by a lock-in amplifier (NF 5610B). The frequency of the applied field, f, was changed from 10 Hz to 60 kHz.

In Figure 1 is shown the frequency dispersion of the $2f$ component of the intensity, ΔI_{2f} at 100°C in SmC$_A$*. The imaginary part has a peak, which corresponds to the relaxation frequency of the antiferroelectric Goldstone mode. It is seen from Equation (4) that there are two different origins for the $2f$ component; one is proportional to E_0^2 and the other to E_0^4. In order to examine which contributes to the $2f$ response, we measured the applied field dependence of the amplitude of the 2f component at 1 kHz. As shown in Figure 2 (note that the abscissa is E_0^2), the $2f$ component is proportional to E_0^2 at weak voltage. In our experiment, therefore, the first term in Equation (4) is dominant. This fact is very important in a homeotropic cell. If the antiferroelectric phase can be regarded to be optically uniaxial, the first term should vanish for the following reason.

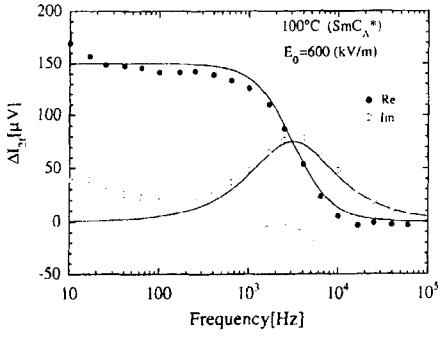

FIGURE 1 Frequency dependence of the $2f$ component in SmC$_A$*.

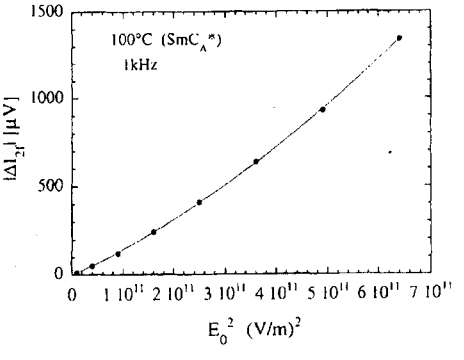

FIGURE 2 Dependence of the amplitude of the $2f$ component on the square of the field strength at 1 kHz in SmC$_A$*.

The transmitted intensity in this case is described as

$$I \propto \sin^2(\pi d n_a/\lambda) , \tag{5}$$

where d is the film thickness, n_a is the index difference in the film plane, and λ is the light wave length. For symmetry reason, n_a should be proportional to E_0^2 and therefore the intensity to E_0^4. The appearance of the first term in Equation (4) may be ascribed to defects or the helical structure with a large pitch comparable to the wave length of the incident light. We measured also the frequency dispersion of the $4f$ component. The applied voltage dependence of it was confirmed to be proportional to E_0^4, as it should be. It is seen from Figure 3 that the frequency dependence of the $4f$ component is not Debye-type, but the square of it, as is expected from Equation (4).

The temperature dependence of the relaxation frequency obtained from the measurement of the $2f$ component is shown in Figure 4. It is needless to say that the modes observed in different phases are different; they are called the antiferroelectric Goldstone mode in SmC_A^*, the ferrielectric Goldstone mode in SmC_γ^*, and the ferroelectric Goldstone mode in SmC*. The relaxation frequency of the ferrielectric Goldstone mode is remarkably low compared with those of the ferroelectric and the antiferroelectric ones. In SmC_A^* the relaxation frequency of the antiferroelectric Goldstone mode shows a parabolic temperature dependence. With decreasing temperature, it gradually decreases to a minimum at 78°C and then increases.

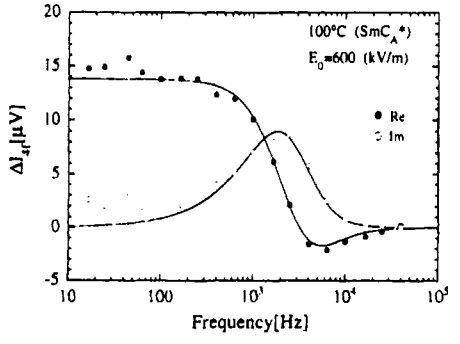

FIGURE 3 Frequency dependence of the $4f$ component in SmC_A^*.

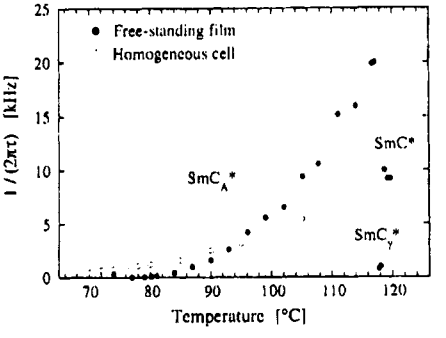

FIGURE 4 Temperature dependence of the relaxation frequency $1/(2\pi\tau)$.

The temperature dependence of the relaxation frequency obtained by the third-order nonlinear dielectric measurement in a homogeneous cell is also shown in Figure 4. The relaxation frequency in the homogeneous cell is slightly dependent on temperature and there is no minimum. Taking account of Equation (1), this discrepancy may come from the surface anchoring effect in the homogeneous cell, by which it is not easy for the helical pitch to take one realized in a homeotropic cell, where the helical pitch is determined only by the bulk nature because the molecules on the surfaces can freely rotate. According to a theory, the wave number of the helix has such a temperature dependence as

$$q = (T - T_D)/C, \tag{6}$$

where T_D is the temperature at which the pitch diverges and C is the constant. Substitution of this equation into Equation (1) yields $1/\tau \propto (T - T_D)^2$. In order to confirm this we plotted $(1/\tau)^{1/2}$ vs. T in Figure 5, where we take the negative sign for $(1/\tau)^{1/2}$ when $T<T_D$; the negative sign means that helical sense is reversed. All the data fall on a straight line as is expected from the theory.

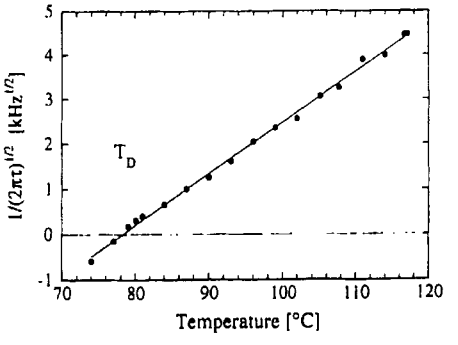

FIGURE 5 Temperature dependence of the square root of the relaxation frequency.

CONCLUSIONS

We have measured the relaxation frequency of the antiferroelectric Goldstone mode in a free-standing film. The divergence of the relaxation frequency was observed, which corresponds to that of the helical pitch. The linear temperature dependence of the helical

wave number was found. By comparing these results with the ones obtained in a homogeneous cell, it is seen that the surface anchoring in a homogeneous cell is strong enough to change the helical pitch from the natural one in the absence of the surface effect, while in a homeotropic cell (a free-standing film) the surfaces have almost no effect on the helical structure.

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