Field-induced thickness change of ferroelectric liquid crystal films

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We observe an increase of the thickness of a chiral ferroelectric liquid crystal film when subjected to ac fields. At 1 kHz at voltages slightly above the threshold for helix unwinding, the sample thickness increases by 0.3 μm in a few minutes. Under this field the cover plate vibrates vertically with a dominating component at twice the field frequency of an amplitude of almost 1 nm. This vibration is expected by the continuum theory of Leslie, Stuart, and Nakagawa [Mol. Cryst. Liq. Cryst. **198**, 443 (1991)], but a theoretically unexplained small asymmetry in the vibration is needed to explain the pumping. The reason for the asymmetry is not yet understood. [S1063-651X(96)50806-8]

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INTRODUCTION

Due to the presence of permanent electric dipole in chiral smectic-C^* (Sm-C^*) liquid crystals the director is linearly coupled to an external electric field. Director rotation induces flow (backflow) [1] and there is a corresponding mechanical vibration of the film (electromechanical effect [2]) mainly along the film surface. Ferroelectric Sm-C^* liquid crystals have a tilted layered structure. The director tilt angle is coupled to the spontaneous polarization. Electric fields have therefore also a linear coupling to the tilt angle (electroclinic effect). Applied fields induce accordingly a variation of the layer spacing that causes an extension of the layers. In bookshelf textures under ac fields this results in a periodic variation of the film thickness [3]. For weak fields the dominating electromechanical effects are linear with the field (the frequency of the induced mechanical vibration is equal to that of the applied field). For large fields quadratic and higher order terms become important (the vibration has Fourier components at twice or higher multiples of the frequency of the applied field).

In addition to the periodic vibrations nonperiodic mechanical effects are also possible. It was demonstrated recently by Zou and Clark [4] that in films with a fixed thickness alternating fields can yield steady flow in the direction parallel to the layers and the film surface. The direction of the unidirectional flow can be reversed by reversing the equilibrium direction of the polarization, so it is connected to the chirality of the system.

In this paper we describe a different pumping effect which is due to nonlinear contributions and does not depend on chirality. We studied a short pitch Sm-C^* film in bookshelf structure under various field amplitudes and frequencies and observed, in a limited frequency range, a field-induced steady increase of the film thickness.

EXPERIMENT

We studied 8 μm thick cells (some of them were wedge shaped, $\sim 10^{-3}$ rad) of uniform bookshelf structures of a liquid crystal, FLC 6430 (from Hoffmann La Roche). The material has Sm-C^* phase from -11 °C to 58 °C [5]. Between 58 °C and 65 °C it has a smectic-*A* phase, at higher temperatures it is isotropic. At room temperature it has a helical structure with a pitch of $p = 0.4$ μm, a director tilt angle of $\theta = 27°$, and a polarization of $P_s = 90$ nC/cm^2. The glass surfaces of the cells were coated with octadecyltriethoxysilane. On cooling homeotropically aligned smectic-*A* films form and the layers stay parallel to the substrate on cooling further to the smectic-*C* phases. By applying an electric field of 4 V/μm and under periodic shearing of the plates at room temperature the horizontal layer texture was transformed to a uniform bookshelf texture [6]. The bookshelf structure remained uniform after removing the field in spite of the reformation of the helix. The cell thicknesses were set by spacers placed near to the edges and were sealed by tapes which allowed small movement of the plates.

The textures were observed at room temperature by a polarizing microscope while different voltage waves of various amplitudes and frequencies were applied. During the application of the field the long term variation of the sample thickness was detected by interference microscope using a Leica microscope equipped with a Mirau interferometer. We also measured the vertical periodic vibration by placing a tiny piezoelectric accelerometer (BK 4375 from Bruel & Kjaer) on the cover plate. The first and second harmonic components of the vibration were measured using a lock-in amplifier (EG&G 5209).

At sufficiently high fields the helical structure unwinds and P_s switches fully (from ''up'' to ''down'') with the frequency of the field. Under this condition and frequencies above $\sim$130 Hz air channels appear at the edges and grow inward parallel to the smectic layers (see Fig. 1). The channels run parallel to the layers and starts at the edges; in the wedge shape cell at the thinner end. The air channels shrink again when the voltage is below the critical field for unwinding so that a helical structure forms, or when a dc bias field is applied that suppresses the switching (see Fig. 2).

Interferometric studies revealed that while the air was flowing in the sample thickness increased by about 0.3 μm in a few minutes, then remained constant. During the outward flow of the air the sample thickness went back to its original value.

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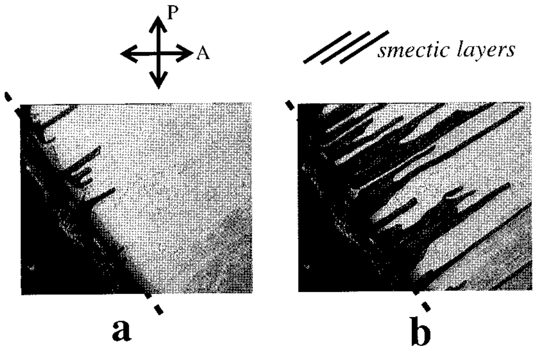

FIG. 1. 8 μm thick Sm-C^* film between crossed polarizers (P and A). Dark areas are filled with air. Dashed line indicate the edge of the cell. (a) 10 sec and (b) 60 sec after $U = 22$ V sinusoidal voltage of 1 kHz is applied. Pictures taken when the field was on. $T = 25$ °C, Magnification: 100.

The first and second Fourier components of the vertical vibrations of the cover plate (linear and quadratic responses, respectively) for a 1 kHz field is plotted in Fig. 3. The first harmonic component is almost a linear function of the voltage over the whole range. It does not show significant change at the helix unwinding. The second harmonic component is negligible below the unwinding field, but then it increases sharply to 0.7 nm. At higher fields, the quadratic response decreases again. The increase of the sample thickness starts with the onset of the quadratic effect. At higher voltages the rate of increase saturates and becomes independent of the applied voltage. At frequencies lower than 130 Hz an increase of the quadratic effect in the voltage range near the unwinding transition is observed too, but it is not accompanied by an increase of the sample thickness. In this frequency range the second Fourier component of the vibration decreases only slowly at larger fields. For example, at 75 Hz after the unwinding (6 V) the quadratic signal increases from 0.03 nm to 0.12 nm and stays at that value up to 30 V. At low frequencies the amplitude of the linear signal is larger, but again behaves smoothly during the helix unwinding process.

For fields above the unwinding threshold the rate of the growth of the area covered by the air channels increase with the frequency. It was determined by analyzing the video recording. The average growth rate is zero below 130 Hz then increases with the frequency almost linearly. For example, at a 5 V/μm field the average growth rate is about 6 μm/s at 1 kHz. At a given frequency the speed is almost independent of the voltage above the threshold.

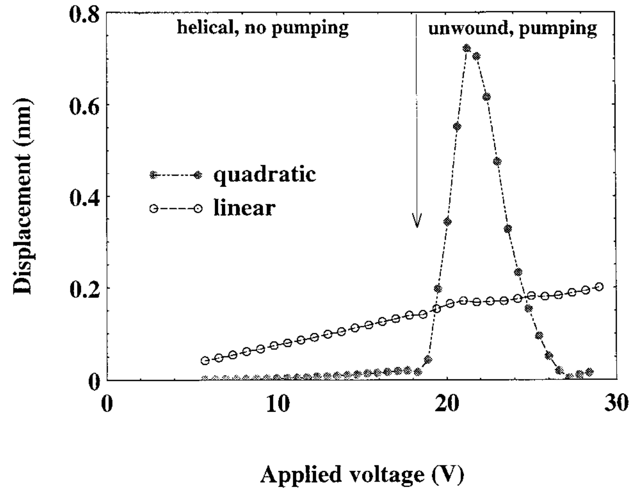

FIG. 3. Voltage dependence at 1 kHz of the amplitudes of the first and second Fourier components (linear and quadratic responses, respectively) of the vertical vibrations of the cover plate.

DISCUSSION

The coincidence between the pumping and the onset of the quadratic effect, i.e., the second harmonic component of the vibration, suggests that they are connected. The nonlinear effect is caused by the director rotation (see Fig. 4). The following simplified description illustrates the mechanism. During the switching process the director rotates from position ***u*** (upward polarization) to position ***d*** (downward polarization) through position ***m*** (polarization is horizontal). When the director is in the position ***m*** the layers extend in the x direction. Accordingly the sample thickness would change. According to our observations the vibration is unbalanced and in each period the sample thickness increases (or decreases) by a small fraction of the vibrational amplitude (for example, in the first minute of application of a 1 kHz field it increases by about 0.1%).

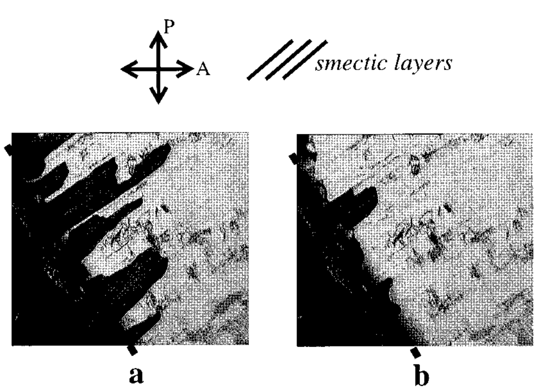

FIG. 2. As in Fig. 1, but after reduction of the applied field. (a) 10 sec and (b) 120 sec after reduction to a 16 V sinusoidal voltage of 1 kHz.

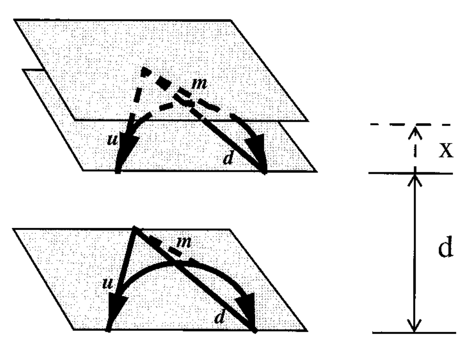

FIG. 4. Simple model for the vertical quadratic electromechanical effect in the range where the helix is unwound and the director rotates collectively around the cone angle.

The periodic pressure can be calculated using the continuum theory of Leslie *et al.* [7]. We consider constant layer thickness and director tilt angles. The experimental geometry corresponds to a uniform bookshelf structure where the layers are upright and normal to the electrodes. We choose a coordinate system so that the layers are in the x-y plane, the layer normal showing in the z direction and the external electric field is applied in the x direction. The direction of the polarization makes an angle Φ with the x axis. In the experimental situation the helix is unwound and $\Phi=\Phi(x,t)$, but we will in the following neglect the x dependence of Φ. Following the calculation of Zou *et al.* [8] the equations to solve are the following.

$$N\nu_{2,1}+G\dot{\Phi}=\sigma_{11}, \tag{1}$$

$$F\nu_{2,1}+L\dot{\Phi}=\sigma_{21}, \tag{2}$$

$$L\nu_{2,1}+\lambda_5\dot{\Phi}=\frac{1}{2}\Gamma_3. \tag{3}$$

In these expressions

$$F=1/2(\mu_0+\mu_4+\lambda_5)+(\mu_3/4)\sin^2(2\Phi)+\lambda_2\cos(2\Phi), \tag{4}$$

$$G=\lambda_2\sin(2\Phi), \tag{5}$$

$$L=\lambda_2\cos(2\Phi)+\lambda_5, \tag{6}$$

$$N=(\mu_4/2+\mu_3\sin^2\Phi+\lambda_2)\sin(2\Phi). \tag{7}$$

μ_0, μ_3, μ_4, λ_2, and λ_5 are relevant Leslie viscosity coefficients (for their nematic correspondence see Zou *et al.* [8]). A comma preceding an index means a partial derivative of the spatial coordinate. σ_{11} and σ_{21} are the xx and yx components of the viscous stress tensors. Γ_3 is the external torque in the direction of the layer normal.

$$\Gamma_3=-PE\sin\Phi+\frac{1}{2}\Delta\varepsilon\varepsilon_0E^2\sin^2\theta\sin(2\Phi). \tag{8}$$

The first term is due to the coupling with the spontaneous polarization, the second is the dielectric torque. In our samples $\Delta\varepsilon\sim1$, $P\sim10^{-3}$ C/m^2 and $E=5\times10^6$ V/m, so $PE/\Delta\varepsilon\varepsilon_0E^2\sim200$, therefore the dielectric contribution can be neglected.

In contrast to Zou and Clark [4], who considered σ_{21}, we are interested in σ_{11}. Neglecting the backflow, i.e., $\nu_{2,1}=0$, $\Phi(t)$ and $\sigma_{11}(t)$ can be calculated. With periodic electric fields $[E=E_0\sin(\omega t)]$ we get

$$\Phi=2\arctan\left(\exp\left\{\frac{PE_0}{\lambda_5}[1-\cos(\omega t)]+\ln\left[\tan\left(\frac{\Phi_0}{2}\right)\right]\right\}\right), \tag{9}$$

$$\sigma_{11}(t)=PE\frac{\lambda_2}{2\lambda_5}\sin(\Phi)\sin(2\Phi). \tag{10}$$

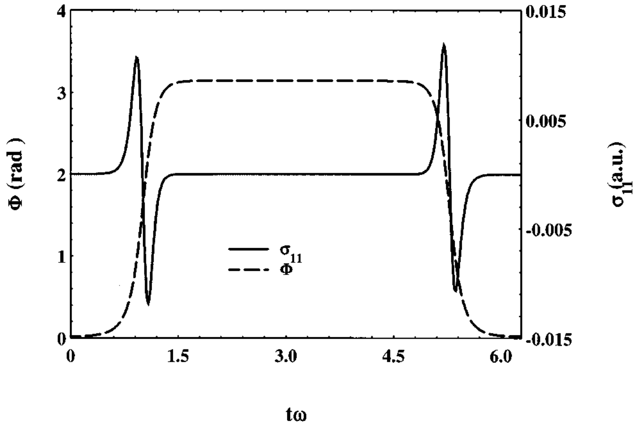

FIG. 5. $\Phi(t)$ and $\sigma_{11}(t)$ as calculated from Eqs. (19) and (20) when the different viscosity coefficients are taken to be equal, $PE/(2\lambda_5\omega)=10$ and $\Phi_0=0.02$ rad.

$\Phi_0=\Phi(t=0)$ is the ''pretilt'' angle. An example of the solutions for $\Phi(t)$ and $\sigma_{11}(t)$ are plotted in Fig. 5 (λ_5 and λ_2 were taken to be equal; $PE/(2\lambda_5\omega)=10$ and $\Phi_0=0.02$ rad). $\Phi(t)$ is asymmetric with respect to the 0 and π positions but the actual ''up'' and ''down'' starting positions are determined by thermal fluctuations and are equal in average. The time average of the pressure $\langle\sigma_{11}\rangle$ is zero, so the above model cannot explain the observed steady increase of the sample thickness. The model explains the second and higher Fourier components of the vibration.

Experimentally we find that the second Fourier component of the periodic vibration has a maximum near the threshold for unwinding, then it decreases at higher fields (see Fig. 3). This can be explained with the decrease of the switching time relative to the period which increases the amplitude of the higher harmonics. In spite of the decreasing second Fourier component the pumping effect does not decrease at higher fields. It means that $\langle\sigma_{11}\rangle$ is independent of the amplitude of the field above the switching threshold. At voltages above the switching threshold the pumping efficiency is zero below a critical frequency, than is a linear function of the frequency. The observed critical frequency for the pumping indicates that there is a threshold for the lifting (the plate rests on spacers). Accordingly at low frequencies $\langle\sigma_{11}\rangle$ is smaller than the threshold pressure. The saturation of the lifting motion in time is probably due to the increased surface tension, by forming air channels and the reduced area where pumping occurs.

Summing up we observed an increase of the sample thickness of a ferroelectric liquid crystal under ac fields strong enough for ferroelectric switching. The effect is due to an asymmetry in the switching process which results in a nonzero time average of the pressure.

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