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Linear electromechanical effect in a S_C^* polymer liquid crystal

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A linear electromechanical effect, similar to that observed in low-molecular weight S_C^* liquid crystals, is reported for a S_C^* polymer liquid crystal. A 20 μm thick film of LCP1 (polymer liquid crystal, S_C^*, supplied by BDH) was prepared that could be aligned by shearing. An alternating electric field was applied perpendicular and the lateral oscillating motion of the cover glass was measured and analysed. In addition to a linear response, vibrations with the frequency of the applied field, a quadratic response was also observed. The frequency, temperature and applied voltage dependences were measured. The response depends strongly on the alignment. The qualitative analysis of the effects indicates that the oscillations are due to viscous coupling between director reorientation and flow.

1. Introduction

The study of ferroelectric liquid crystals is one of the most dynamic branches of liquid crystal research. Similarly, since their discovery in 1978 [1] the thermotropic side-chain liquid-crystalline polymers have attracted considerable attention and the study of ferroelectric liquid-crystalline polymers is of particular interest. As in low-molecular weight ferroelectrics, most efforts were devoted to the investigation of electro-optical effects [2].

Beside the promising electro-optical properties, ferroelectric liquid crystals also have other interesting physical features. For example, due to their symmetrical properties they are piezoelectric and show linear electromechanical effects. In the case of low-molecular weight S_C^* samples with a chevron texture, this effect was investigated in detail [3–5]. It was found that due to alternating electric field excitations, mechanical vibrations occur in planar S_C^* samples (smectic layers are nearly normal to the plane of the film) with a predominant linear component at the frequency of the applied field [3]. The mechanical vibrations of the cover glass were measured in the direction parallel to the bounding place and perpendicular to the helical axis, i.e. parallel to the smectic layers. It was pointed out that the vibrations are due to periodic flows or vibrations in the liquid induced by the field [4] and that in the frequency spectrum resonances may be present [5] which are related to the existence of different domains in the sample. It is anticipated that similar linear electromechanical effects should also exist in planar S_C^* polymer liquid crystals. In this paper we report the first measurements of electromechanical effects on a side-chain polymer liquid crystal material.

2. Experimental

The compound studied is denoted by LCP1 and made by BDH. It is a side-chain polymer having a S_C^* phase in a broad temperature interval, from -11°C up to 69°C

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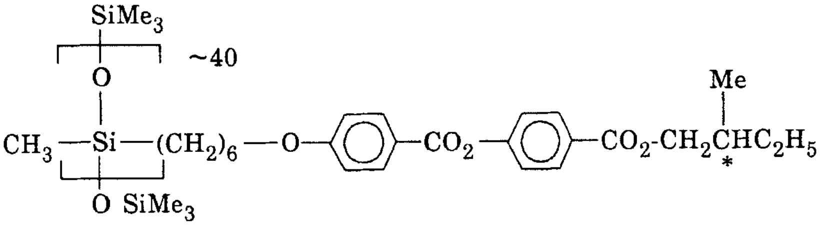
and an average magnitude spontaneous polarization $P_0 = 4{\cdot}10^{-5}\,\mathrm{C/m^2}$. Above 79°C the material is isotropic but there is a two-phase range of about 10°C (69–79°C). The molecular structure of LCP1 is the following:



The material was sandwiched between two clean glass plates, coated with transparent, electrically conductive SnO, and separated from each other by polystyrene balls. Spherical spacers of 20 μm in diameter were placed close to the edges. The bottom glass plate was attached to the oven while the upper glass plate was free to slide on the spacers. On top of the upper plate a piezoelectric accelerometer (Bruel & Kjaer, Type BK4375; sensitivity is $0{\cdot}314\,\mathrm{pC/ms^{-2}}$, $m = 2{\cdot}3$ g) was attached so that it was sensitive to vibrations parallel to the plates. The sample temperature was electrically controlled with an accuracy greater than 0·01°C. For alignment the sample was heated to the isotropic phase then slowly cooled down to its S_C^* phase while a periodic shear [6] was applied on the sample parallel to the sensitive direction of the accelerometer. No special surface treatment of the glass surfaces was used except for careful cleaning with acetone and distilled water. As a result of the shear, the smectic layers tend to align parallel to the shear and perpendicular to the glass plates, but the alignment that we actually achieved was incomplete. The texture contained focal conic defects. When the sample was cooled without shear an almost uniform alignment established itself with smectic layers parallel to the glass plates.

To measure the electromechanical response of the S_C^* phase, a sinusoidal voltage was applied on the electrodes and the acceleration of the upper plate measured. The voltage was supplied by the internal oscillator of a high precision lock-in amplifier (EG&G Princeton Applied Research, Model 5209). The current signals of the accelerometer were converted to voltages with the conversion ratio of 10^8 V/A and analysed with a lock-in amplifier. Both the acceleration and the phase shifts against the input signal were measured. In order to avoid the interference of higher harmonics, a sharp band pass filter was tuned to the frequency of the applied voltage for the linear response (first harmonic) and to twice the driving frequency (second harmonic) for the quadratic response. Three types of measurements were performed.

(1) Voltage dependence (T=const., f=const.) for linear and quadratic response.
(2) Frequency dependence for linear and quadratic response. The range of the driving voltage frequencies was $200\,\mathrm{Hz} < f < 6\,\mathrm{kHz}$.
(3) Temperature dependences of linear and quadratic response.

3. Experimental results

3.1. *Voltage dependence*

The amplitude of the first harmonic is proportional to the applied voltage. For the second harmonic the amplitude was found to be a quadratic function of the applied voltage. Typical results are seen in figure 1 where the acceleration of the glass plate is plotted versus the amplitude of the applied voltage.

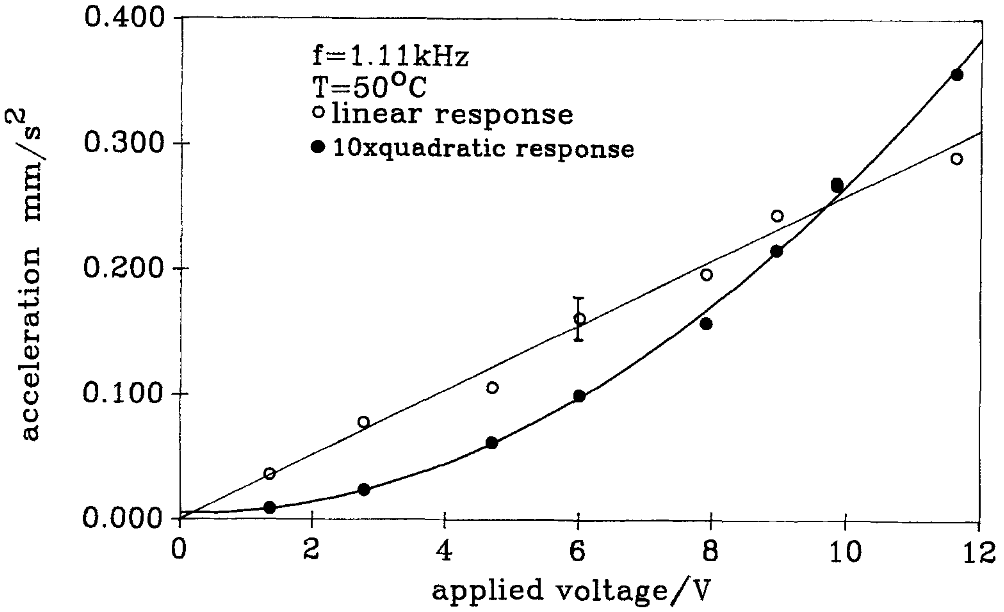


Figure 1. Voltage dependences of the acceleration. The open circles correspond to the linear response. The full circles correspond to the quadratic response amplified by ten. The sample temperature is $T=50°C$, the frequency 1·11 kHz.

3.2. *Frequency dependence*

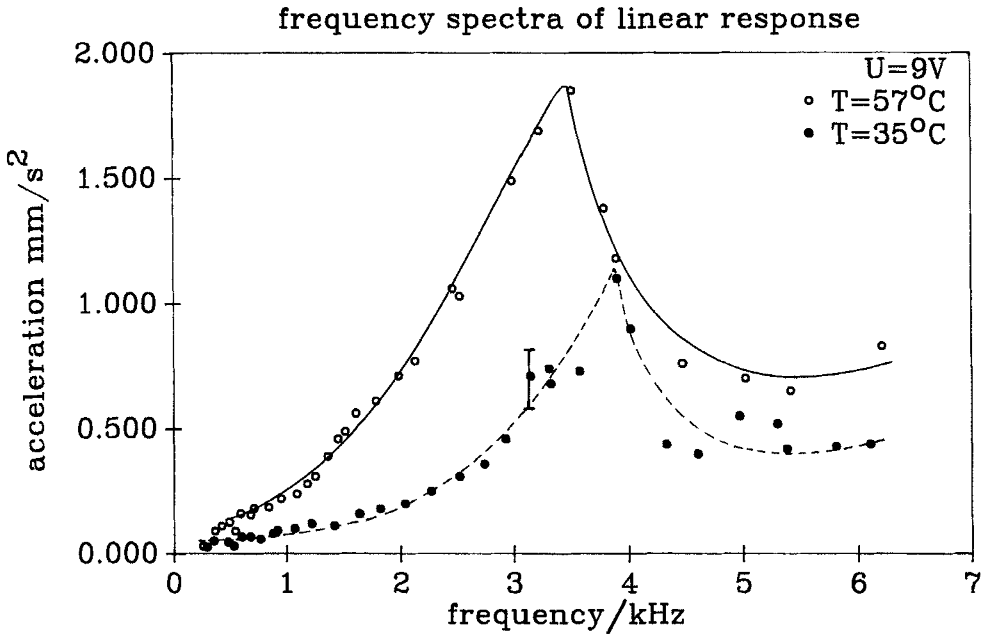
Typical spectra of the frequency dependence of the mechanical response for two selected sample temperatures, $T=35°C$ and $T=57°C$, are presented in figure 2. In the spectra of the linear response (figure 2(*a*)) large maxima can be seen at $f=3{\cdot}9$ kHz for $T=35°C$ and $f=3{\cdot}5$ kHz for $T=57°C$, respectively.

Figure 2 (*b*) gives the spectra of the quadratic responses. We see again a maximum in each curve but they are situated at lower frequencies; at half of the frequencies of the linear responses.

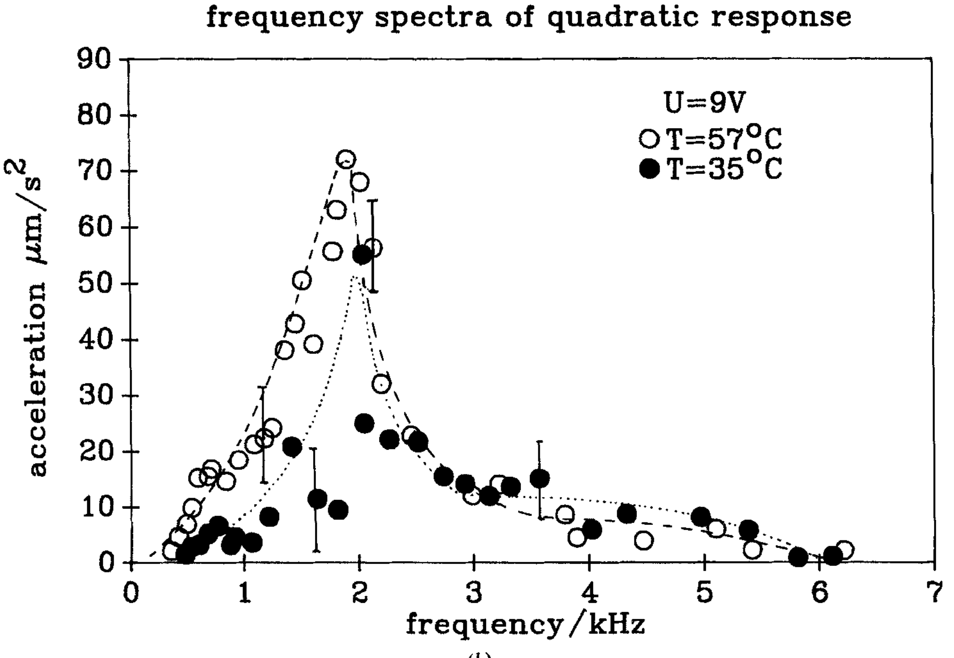
Figure 2(*c*) presents the frequency dependence of the phase shift between the velocity of the linear response and the applied voltage. Zero phase shift corresponds to a motion of the plate in phase with the field and the velocity in the positive y direction when the field is in the positive x direction. Experimentally we did not determine the sign of the velocity; however, we observed that the phase shifts for different alignments could differ by approximately 180°. Let us say the phase shift is zero at zero frequency. Then we find that it is almost constant for low frequencies and reaches $-90°$ at frequencies that correspond to the maxima of the response curves. With further increasing frequencies the phase shift difference increases further and exceeds even values of $-180°$. The actual displacements of the plate are small. From the measured linear response at $f=3{\cdot}9$ kHz, $U=10$ V and $T=57°C$, one can estimate that the largest displacements are in the range of 10^{-10} m. They are about an order of magnitude smaller for the quadratic response.

3.3. *Temperature dependences*

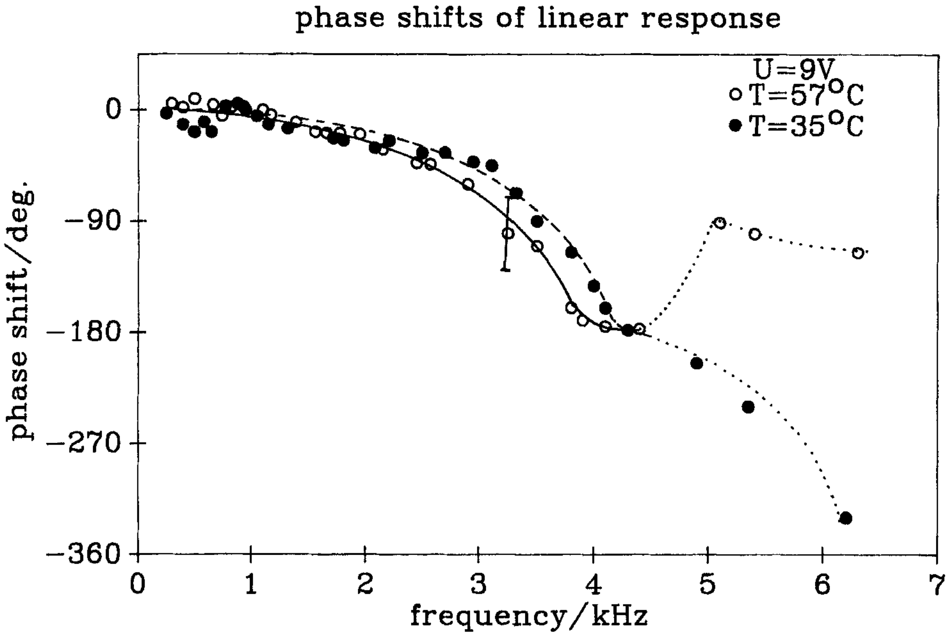
The temperature dependence for linear and quadratic responses are shown in figure 3 for shear aligned and untreated samples. The figures show typical results with general features that reproduce nicely and that were also observed with other frequencies. The


frequency spectra of linear response
U=9V
○ T=57°C
● T=35°C
acceleration mm/s^2
2.000
1.500
1.000
0.500
0.000
0
1
2
3
4
5
6
7
frequency/kHz

(*a*)

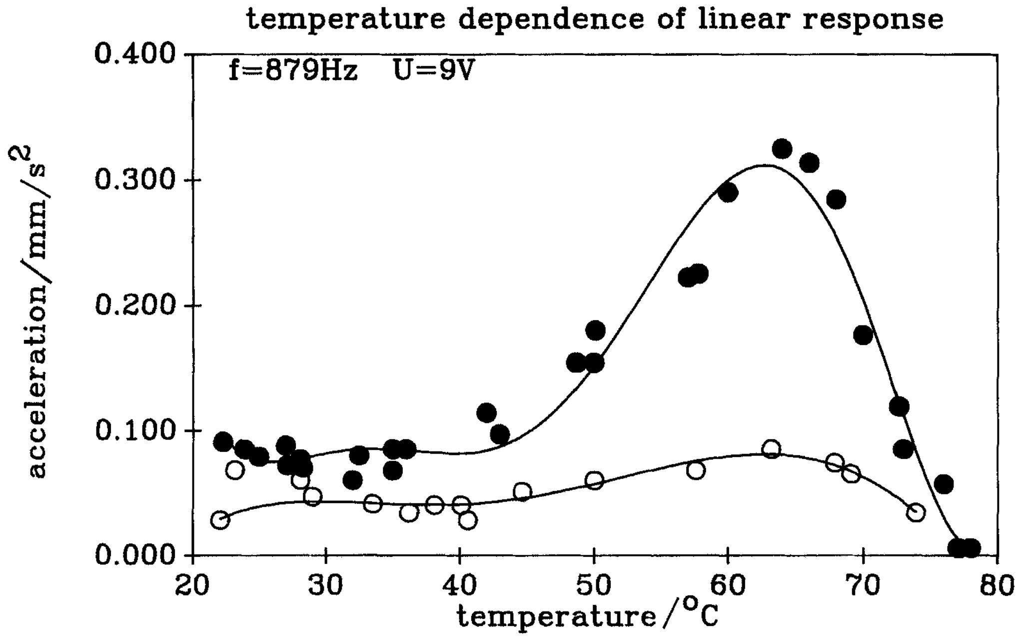

frequency spectra of quadratic response
U=9V
○ T=57°C
● T=35°C
acceleration μm/s^2
90
80
70
60
50
40
30
20
10
0
0
1
2
3
4
5
6
7
frequency/kHz

(*b*)



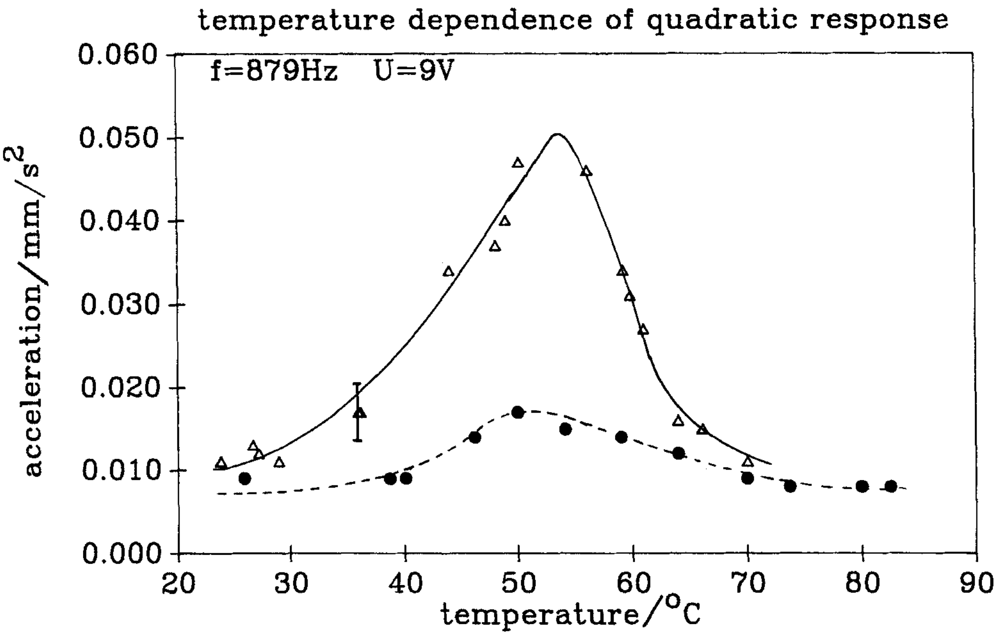
(c)

Figure 2. Frequency spectra of the acceleration. The applied voltage is $U=9$ V. Open circles: $T=57$°C, full circles: $T=35$°C. (a) Linear electromechanical responses. (b) Quadratic electromechanical responses. (c) Phase shifts between velocity and field of the linear electromechanical responses.



(a)

Figure 3.



(*b*)

Figure 3. Acceleration versus the sample temperature. Applied voltage $U=9$ V, frequency $f=879$ Hz. (*a*) Linear effect. Full circles, aligned sample; open circles, unsheared samples. (*b*) Quadratic effect. Open triangles, after shear alignment; full circles, without shear alignment.

general trend of the aligned samples is that the linear effect increases with temperature and reaches a maximum near the S_C^*–I phase transition. During the measurement the sample was visually observed by a polarizing microscope. Smectic batonnets were seen until $T=79$°C. As the whole sample turns isotropic the accelerometer signal drops to zero. The quadratic response shows a similar trend, but it does not completely reduce to zero in the isotropic phase. In the isotropic phase a quadratic response is still possible but the induced motion of the glass plate is vertical and should not be detected. We think that the observed small signal is an artifact. The figures also show that the enhancement by shear alignment is quite significant for both the linear and the quadratic response, in spite of the relatively poor alignment that could be achieved. (We obtained much better alignment with low-molecular weight liquid crystals which form a smectic A above C* [6].)

The electromechanical response depends strongly on the alignment, but at a given alignment and temperature the measured data are quite reproducible. The error bars in the figures give estimates of the scattering errors.

4. Discussion

We found that a side-chain polymeric liquid crystal in the S_C^* phase has electromechanical effects similar to the effects observed in low-molecular weight S_C^* liquid crystals.

The similarities are summarized as follows:

(1) In both cases the samples vibrate primarily with the frequency of the applied voltage but second harmonic responses also can be measured. The amplitude of

the first type of vibrations are proportional to the applied voltage, while they are proportional to the square of the voltage for the second type.

(2) Similar to previously reported studies on low-molecular weight S_C^* liquid crystal multi-domain [5, 7], the frequency dependence shows at least one maximum in the observed frequency interval.

(3) The temperature dependences of linear and quadratic responses are also similar. The remarkable features are that the linear electromechanical responses exist only in the S_C^* range and disappear in the isotropic phase.

The comparison shows that the polymeric and low-molecular weight S_C^* materials behave qualitatively in the same way. The major difference is that the response of the polymeric material is smaller. The exact mechanism that induces electromechanical effects is not yet established. The main features of the experimental results which should be explained by theory are the following:

(*a*) The magnitude of the effect depends on the alignment.

(*b*) The phase relation of field and velocity of the cover slide at low frequencies can be close to 0° or 180°. For increasing fields, the motions may go in either a positive or negative direction.

(*c*) In the low frequency range from 0·5 to 1·5 kHz the acceleration increases linearly with the frequency.

(*d*) At about 4 kHz there is a broad resonance peak.

We will discuss the observations using the following model for the S_C^* phase. The layers are not deformed by the applied field. The orientational order is in good uniaxial approximation so that the director **d** for the orientational order can be defined. The tilt angle, θ, between **d** and the layer normal, **n**, and the spontaneous polarization, $\mathbf{P}_0$, remains constant. We consider the linear effects only and neglect, therefore, the anisotropy of the dielectric polarizability. According to this model, the director can move only on a cone around **n** and flow can take place only within the layers. We treat the hydrodynamics analogous to that of a uniaxial nematic which is subject to the mentioned constraints. This is justified, to some extent, since the biaxiality and the ferroelectric order are relatively weak. We considered a uniformly aligned film sandwiched between glass plates and selected a cartesian coordinate system with the x axis normal to the plane of the film and the y axis in plane to the smectic layers. The layer normal forms an angle α with the z axis and ϕ denotes the angle between the spontaneous polarization $\mathbf{P}_0$ and the y axis.

We assume that the x dependence of ϕ is negligible except for a relatively thin film at the surface. Accordingly, the flow gradient will differ from zero significantly only in this film. In order to estimate the y component of the force on the glass plates, we derive the value of the viscous stress tensor element σ_{xy} in the material below the surface range. It is

$$\sigma_{xy}=\frac{1}{2}\frac{d\phi}{dt}[\gamma_2(A\cos\phi+B\cos 2\phi)+\gamma_1(-A\cos\phi+B)] \tag{1}$$

where $A=(1/2)\sin^2\theta\sin\alpha$ and $B=\sin^2\theta\cos\alpha$.

Here $\gamma_2=\eta_b-\eta_c$, where η_c and η_b are the shear viscosities with the director in the direction of the flow gradient and parallel to the flow respectively, and γ_1 is the rotational viscosity. The torque density due to the applied field is $\mathbf{P}_0\times\mathbf{E}$. We neglect the coupling due to the dielectric anisotropy that gives

$$\frac{d\phi}{dt}=-EP_0\frac{\cos\alpha\cos\phi}{\gamma_1\sin^2\theta}. \tag{2}$$

The y component of the force per unit area is under the given assumptions approximately equal to σ_{xy}. The measurements were made on the sample in which the smectic C* phase was twisted. The resulting force for a twisted film is obtained by integration over the surface. This gives with the assumption that E is constant

$$\langle\sigma_{xy}\rangle = (1/8)\cos\theta\sin 2\alpha(\gamma_2/\gamma_1 - 1)EP_0. \tag{3}$$

The result shows that the force disappears for $\alpha = 0$, therefore, the effect will disappear for a bookshelf structure. In chevron structures α can be positive or negative and varies typically between 10 and 20° [8]. Accordingly, the force may be in a positive or negative y direction for a given field, depending on the type of chevrons.

The magnitude of the force per unit area can now be estimated. With $E = 5\times10^5$ V/m, $P_0 = 4\times10^{-5}$ C/m^2, $\theta = 25°$, $\alpha = 15°$ and $\gamma_2/\gamma_1 = -1$ we find, as an approximately value at 50°C,

$$\langle\sigma_{xy}\rangle = 3\ \mathrm{N/m^2}. \tag{4}$$

Under these conditions (see figure 1), the observed rms of the plate velocity at $f = 1{\cdot}11$ kHz is $3{\cdot}5\times10^{-8}$ m/s. It corresponds to an acceleration of $2{\cdot}5\times10^{-4}$ m/s^2 or a force of 0·006 N per unit area. (The mass of the plate plus accelerometer was 5 g and the area covered by the film 2 cm^2.)

The force on the plate is less than 1 per cent of the estimated stress (equation (4)), indicating that due to the random distribution of positive and negative domains the effect cancels to ~99 per cent. In summary, the model explains the main features at low frequences but it cannot explain any resonances as observed in the 4 kHz range. Comparing the present results with earlier data on low-molecular weight materials, we noted as a main difference that the effect in low-molecular weight materials is nearly two orders of magnitude larger. This can be explained, at least in part, by the higher viscosity of polymers. Equation (3) shows that the force depends only on the ratio γ_2/γ_1 and not directly on the viscosities, but the velocity is inversely proportional to the effective shear viscosity which is much larger for the polymer. It may be that the viscoelastic properties are also important, but this has not yet been explored. Further experimental research is in progress. We are presently studying the vertical vibration.

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