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ELECTROMECHANICAL EFFECT IN S_C^* LIQUID CRYSTALS

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Detailed analysis is given of the electromechanical effect in planar S_C^* liquid crystals. Experimental data are presented for the amplitude of the vibration as a function of the frequency, applied voltage and temperature. The existence and the frequency dependence of the linear electromechanical effect is explained within the framework of a continuum theory.

INTRODUCTION

During the last few years chiral smectic C (S_C^*) liquid crystals have found themselves in the centre of interest because of their unique macroscopic "bulk" properties. They exhibit spontaneous polarization,[1] which is linearly coupled to the electric field thereby resulting in delicate electrooptical effects.[2–10] While investigating electrooptical properties of planar oriented S_C^* samples we recently found[11] that the reorientation of the director is accompanied by a vibrational motion of the material.

At first glance this electromechanical effect looks similar to the effects observed in piezo- and ferroelectric crystals.[12–14] However, there are important differences and their physical origin is also different. The aim of the present paper is to describe the detailed experimental results and to interpret them on the basis of the recently developed electrohydrodynamic continuum theory of S_C^*.[15–17]

EXPERIMENTAL

The behaviour of a planar S_C^* liquid crystal in an applied quasistatic electric field was investigated. The experimental set-up is shown in Figure 1.

A sandwich cell was used without any spacer. The lower glass was fixed whereas the upper one was allowed to move against a spring in the direction parallel to the lower glass and perpendicular to the helical axis.

A harmonic electric field $E = E_0e^{i\omega t}$ was applied to the sample in a direction perpendicular to the glasses.

As reported earlier,[11] in addition to the well-known switching[2–5] and helix unwinding[6–10] a new electromechanical effect was observed, namely the vibration of the upper plate against the spring.

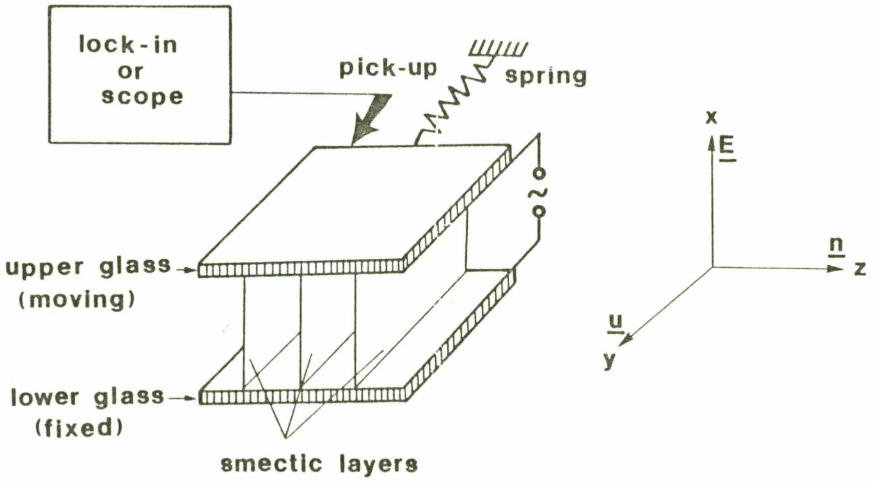

FIGURE 1 Experimental set-up. The orthogonal vectors **n**, **E** and **u** indicate the direction of the helical axis, the electric field and the displacement of the upper plate respectively.

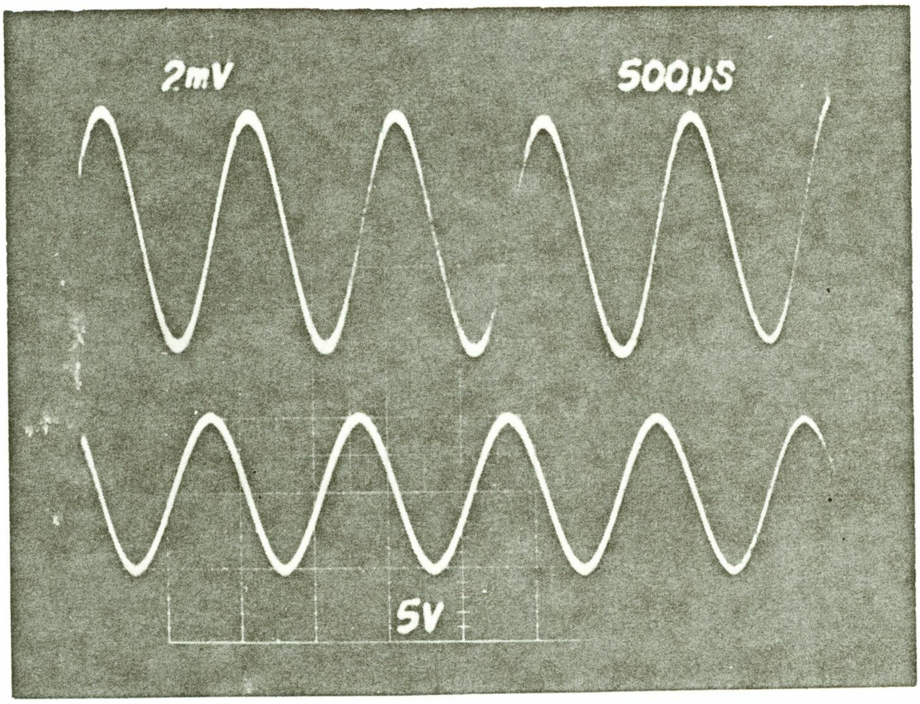

FIGURE 2 Typical oscillogram. The lower curve corresponds to the applied voltage (U = 55 V, f = 1 kHz, attenuation × 10). The upper curve shows the signal of the pick-up. Its amplitude is proportional to the displacement of the upper plate.

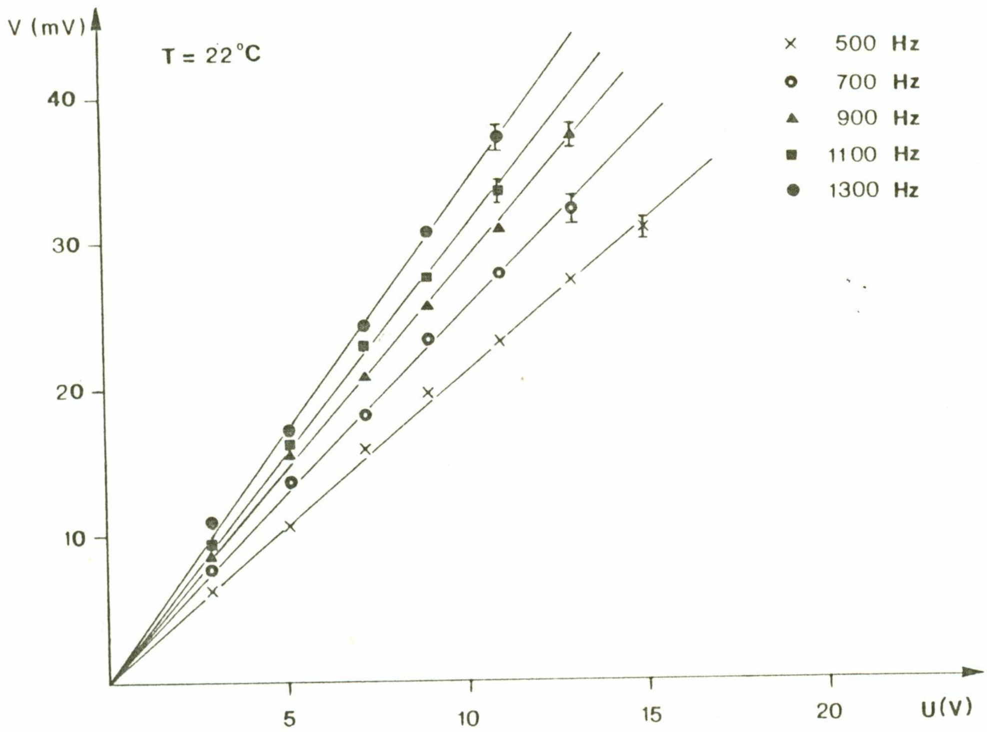

FIGURE 3 Amplitude of pick-up signal vs. applied voltage for different frequencies.

Experiments were carried out on a liquid crystal binary mixture FK4[18] with the phase sequence:

$$I \underset{67^\circ C}{—} Ch \underset{63^\circ C}{—} S_A \underset{29^\circ C}{—} S_C^* \underset{16^\circ C}{—} S_I^*.$$

The pitch in the S_C^* phase was about 5 μm and an unwound sample of 10 μm thickness was used. The sample thickness was controlled with an accuracy of ± 2 μm and checked by capacitance measurement. The sample was thermostated and visually observed by polarizing microscope.

The vibration of the upper plate was detected by a ceramic pick-up sensitive to displacement below 5 kHz (with a typical sensitivity of 6–10 mV/μm).

When the signal of the pick-up was displayed on an oscilloscope as shown in Figure 2 it was found that the frequency of the vibration of the upper plate was equal to the frequency of the applied electric field. A lock-in amplifier was used to analyse the signal of the pick-up as a function of the amplitude and the frequency of the applied electric field as well as versus the temperature of the sample. Our experimental results are presented in Figures 3–5.

Figure 3 shows the vibrational amplitude as a function of the applied voltage. A linear dependence was found for each of the frequencies.

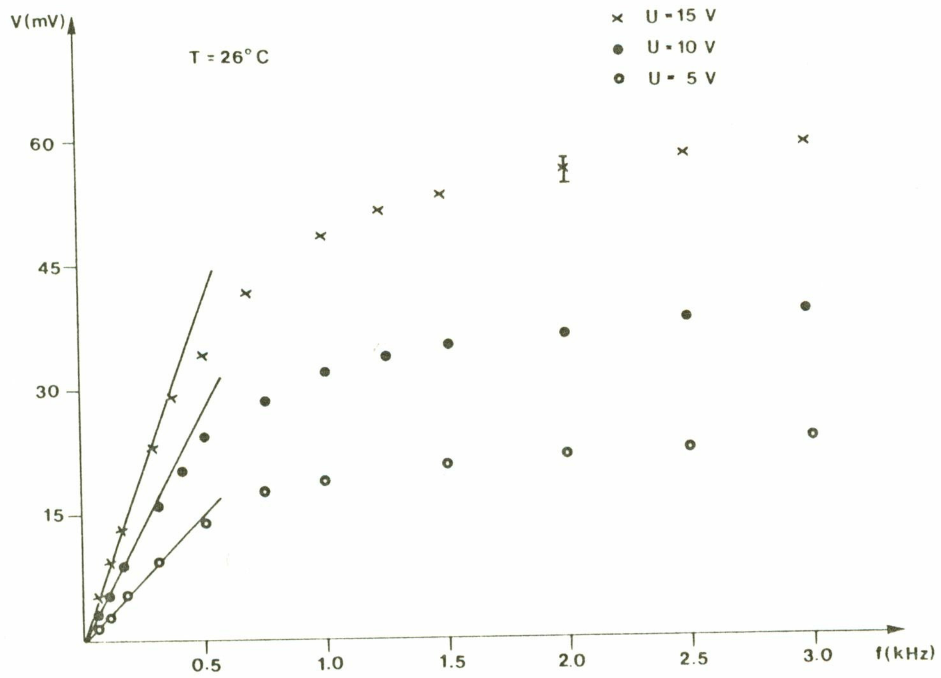

FIGURE 4 Amplitude of pick-up signal vs. frequency of the applied electric field.

In Figure 4 the vibrational amplitude is plotted versus the frequency of the applied electric field for different voltages. Below a few hundred Hz a proportionality was observed while at high frequencies a saturation was found.

In Figure 5 the temperature dependence of the electromechanical effect is shown. The vibrational amplitude is plotted versus temperature for different frequencies. These results show that the electromechanical effect in the S_A and S_I^* phases is at least one order of magnitude weaker than in S_C^* and is within the limits of the experimental error.

We would mention that the shape of the curves is similar to that of the shear induced polarization measured by Pieranski *et al.*[19]

Moreover in the S_C^* phase the curve is reminiscent of the temperature dependence of the helical pitch[20] or that of the dielectric permittivity.[21] But in the last case the permittivity increases with decreasing frequency while the electromechanical effect becomes more intensive at high frequencies.

The electromechanical effect was detected for different sample thicknesses ($5\ \mu m < d < 70\ \mu m$), and similar dependences on the electric field and temperature were observed.

In the case of a homeotropic structure no electromechanical effect was found.

Finally, we should like to mention a very simple but powerful method for qualitatively detecting the electromechanical effect. If one uses a rigid connection between the upper plate and the membrane of a loudspeaker, the membrane

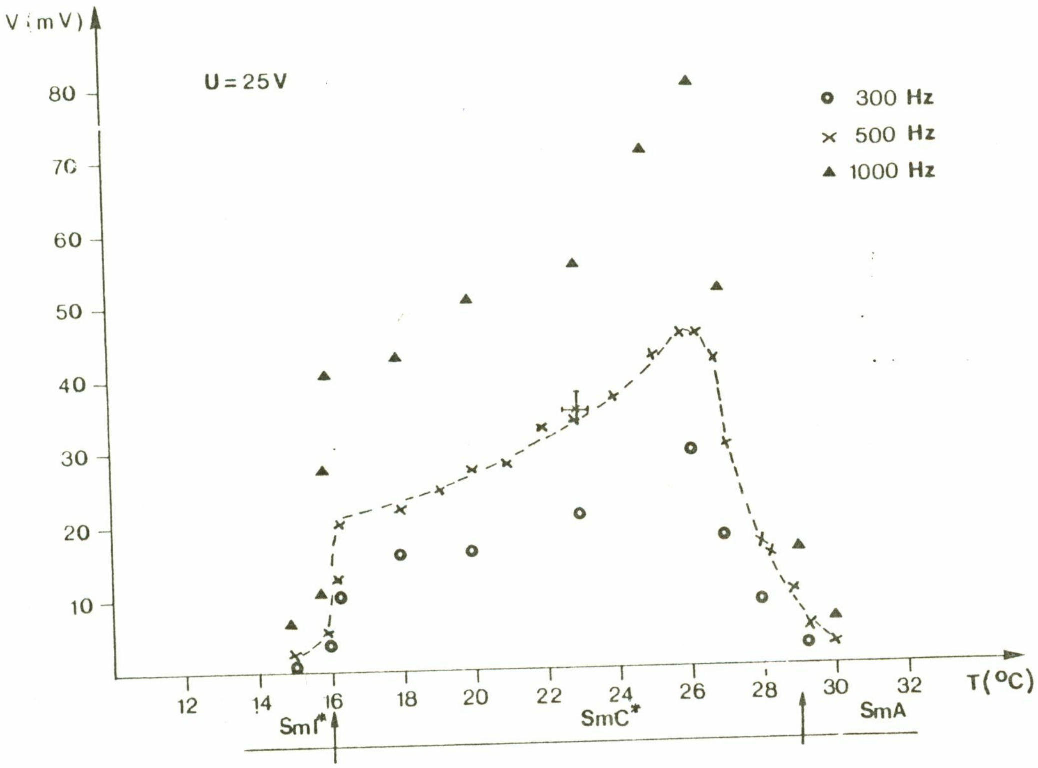

FIGURE 5 Amplitude of pick-up signal vs. temperature. (The sensitivity of the pick-up used here differs from that used in experiments shown in Figures 3 and 4.)

replaces the spring. The motion of the glass plate then makes the membrane vibrate —an effect that can simply be detected by listening to the loudspeaker. In fact in the S_C^* phase the pitch of the note produced by the electromechanical effect corresponds to the frequency of the applied field. In the S_A, Ch and isotropic phases this basic harmonic could not be heard and only a considerably weaker sound could be detected but it was an octave higher, i.e., the frequency of the vibration was twice that of the applied electric field. This second harmonic indicates the existence of a quadratic electromechanical effect in the higher temperature phases.

Though this quadratic effect exists in the S_C^* phase too, it is hidden there by the linear electromechanical effect discussed above. Consequently, it can be detected only at relatively higher fields and lower frequencies in the form of second harmonic distortions of the pick-up signal (compare Figure 6 and 2).

The main features of the experimental results are the following:

(a) The linear electromechanical effect exists in the chiral smectic C^* phase only.

(b) At low frequencies the amplitude of vibration is proportional to $\dot{E} = i\omega E$.

We should now like to offer an interpretation of the phenomenon.

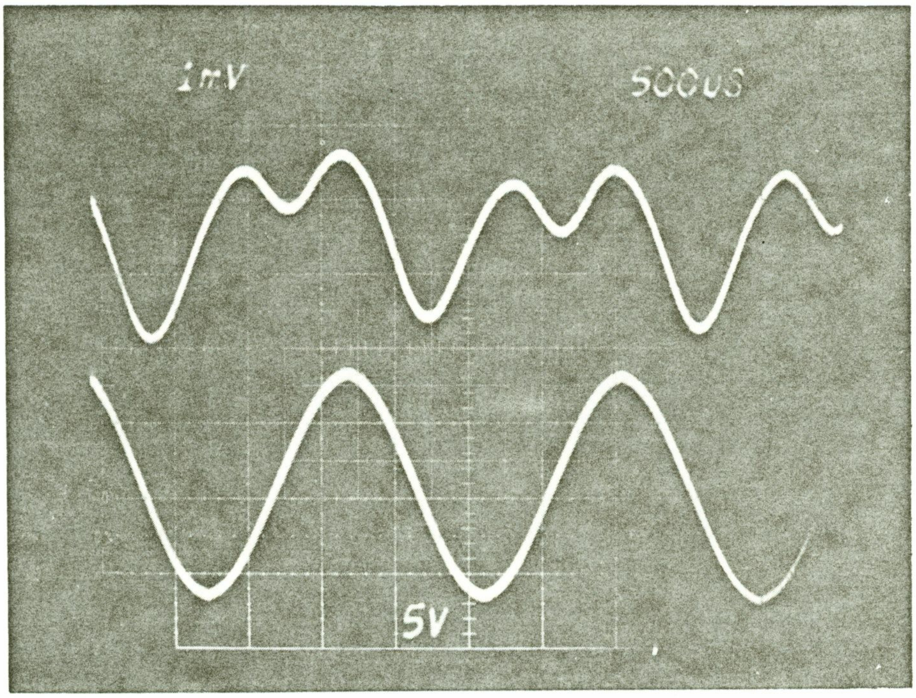

FIGURE 6 Oscillogram from the electromechanical effect. The lower curve corresponds to a tenth of the applied voltage ($U = 70$ V, $f = 540$ Hz). The upper curve shows the signal of the pick-up. The appearance of the second harmonic can be seen.

INTERPRETATION

Since the upper plate of the set-up moves parallel to the smectic layers this motion should result from molecular displacements within the layers. Usually a smectic C layer can be regarded as a two-dimensional liquid because of the lack of positional ordering; consequently, the molecular motion in the layers may be considered a shear flow rather than an elastic displacement. This means that the physical process resulting in the observed electromechanical effect of S_C^* must be different from that which is present in piezo- or ferroelectric solids.[12-14] This is also reinforced by the fact that, in contrast to solids, there is no static electromechanical effect in the S_C^* phase.

A continuum theory has been developed recently in order to explain the behaviour of uniformly layered S_C^* in the presence of an electromagnetic field.[15-17] It has been shown that due to chirality and the biaxial symmetry of S_C^* liquid crystals there exists a cross-effect between dielectric relaxation and viscous flow. This electromechanical coupling leads to the appearance of a mechanical stress proportional to the time derivative of the applied electric field thus inducing a periodic shear flow of the material. Due to this flow the liquid crystal exerts a force on the upper plate resulting in a forced oscillation of that plate.

In the following simplified calculation we demonstrate quantitatively that in our geometry the above mentioned cross effect can actually explain the existence and the frequency dependence of the observed linear electromechanical effect. (The non-linear, quadratic effects, leading to frequency doubling will not be considered.)

The upper glass plate moves against a spring which exerts a restoring force proportional to the displacement **u** of the plate (Figure 1). The upper plate thus performs a forced oscillation governed by the equation of motion

$$m\ddot{\mathbf{u}} = -k\mathbf{u} + \mathbf{F}^{\Omega}, \tag{1}$$

where m is the mass of the plate, k is the spring constant, and $\mathbf{F}^{\Omega}$ is the force exerted on the plate by the liquid crystal. This force is a surface force

$$\mathbf{F}^{\Omega} = \int_{\Omega} \underline{\sigma}\, d\mathbf{\Omega}, \tag{2}$$

where $\underline{\sigma}$ is the mechanical stress tensor of the S_C^*, $d\mathbf{\Omega}$ is a surface element pointing outward from the sample, and the integration should be carried out over the whole surface Ω of the upper plate.

The flow velocity **v** can be determined from the equation of motion of the S_C^*

$$\rho \dot{v}_i = -\nabla_j \sigma_{ij} + F_i^{\mathrm{el}}, \tag{3}$$

where ρ is the density, and F_i^{el} is the density of the electromagnetic volume force.[15-17] This differential equation must be supplemented by the boundary conditions:

$$\begin{aligned} \mathbf{v} &= 0 \quad \text{at the lower plate,} \\ \mathbf{v} &= \dot{\mathbf{u}} \quad \text{at the upper plate.} \end{aligned} \tag{4}$$

The S_C^* material is taken to be incompressible and insulating; moreover, the uniform layered structure does not appear to be distorted as is supported by the optical observations. To proceed further, the continuum theory[15-17] states that the mechanical stress tensor must be written as

$$\sigma_{ij} = \sigma_{ij}^{r} - L_{ijk}^{65} \dot{E}_k' - L_{ijkl}^{66} \nabla_l v_k, \tag{5}$$

where σ_{ij}^{r} is its reversible part, E_k' is the electric field in the co-moving frame, L_{ijk}^{65} is the electromechanical coupling tensor, and L_{ijkl}^{66} is the viscosity tensor. (Hereafter the notations of References 15–17 are used.)

Our calculation will be carried out in the laboratory frame, (Figure 7), with axes given by the electric field **E** (x-axis), the direction of displacement **u** (y-axis), and the smectic layer normal **n** (z-axis). Thus

$$\mathbf{E} = (E_x, 0, 0), \qquad \mathbf{u} = (0, u_y, 0), \qquad \mathbf{n} = (0, 0, 1). \tag{6}$$

The position of the C-director **c** is characterized by the azimuthal angle φ (Figure 7).

$$\mathbf{c} = (\sin\varphi, \cos\varphi, 0). \tag{7}$$

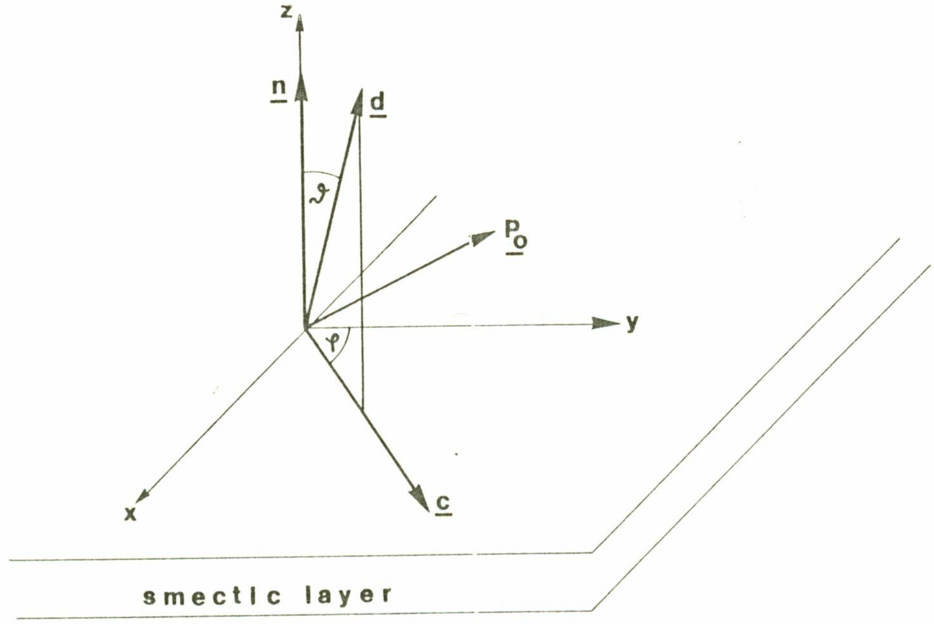

FIGURE 7 Smectic C^* coordinate system. The smectic C^* molecules form a layered structure; the z-axis is parallel to the layer normal. The average direction of the molecular long axes defines the unit director field **d**. The molecules tilt away from the layer normal **n** by angle ϑ. The projection of **d** onto the layer plane defines the C-director **c**, which makes an angle φ with the y-axis. The ferroelectric polarization $\mathbf{P}_0$ is perpendicular to **c** and lies in the layer plane.

The fixed lower plate corresponds to the plane $x = 0$; the upper plate is positioned at $x = d$.

Let us now introduce some simplifying assumptions:

(a) The dimensions of the sample in the y and z directions are much greater than the sample thickness and the pitch; this means that there are no side effects so the y dependence can be neglected.

The electromechanical effect has been observed for thick samples having undistorted helical structure as well as for thin ones where the helix is unwound by the surfaces. This suggests that the z dependence of the C-director in the sample does not influence essentially the effect, consequently the z dependence can also be neglected, i.e., in our simplified calculation all physical quantities depend on x only.

(b) A material flow normal to the layers is incompatible with the smectic structure thus $v_z = 0$. As long as $v_x = 0$ at the glass plates (see Equation (4)) and the incompressibility requires $\nabla_x v_x = 0$, it follows that $v_x = 0$, i.e.,

$$\mathbf{v} = (0, v_y, 0). \tag{8}$$

(c) It is supposed that the velocity of the material flow is small, i.e., it is sufficient to keep terms linear in **v** and **E**. This makes it unnecessary to differentiate between the laboratory frame introduced above and the co-moving material frame which was used when developing the continuum theory.[15-17] It means that the electric field in the two frames is identical $\mathbf{E} = \mathbf{E}'$; the magnetic field is zero.

(d) If a harmonic voltage $U = U_0 e^{i\omega t}$ is applied, the electric field in the medium is

$$E_x(x, t) = E(x) e^{i\omega t}. \tag{9}$$

We are interested only in the linear electromechanical effect; thus, neglecting higher harmonics, we can take

$$u_y(t) = u e^{i\omega t}, \qquad v_y(x, t) = v(x) e^{i\omega t}. \tag{10}$$

(e) σ_{ij}^r in Equation (5) is connected with the reversible deformations of the S_C^* material, i.e., with the reorientation of the C-director, while the other terms describe irreversible processes. Visual investigations have shown that the electrooptical effects, like switching[2-5] or helix unwinding,[6-10] are not influenced by the vibration of the upper plate. It suggests that the reversible changes of the C-director can be handled separately from the irreversible shear flow, thus we can take $\sigma_{ij}^r \approx 0$.

In our experimental geometry with the above assumptions one gets for the equation of motion (3) of the S_C^* material

$$i\omega\rho v(x) = -\frac{\partial}{\partial x}\left[-\mu(x)\frac{\partial v}{\partial x} - \gamma(x) i\omega E(x)\right] \tag{11}$$

with

$$\mu(x) = \mu_{11} + \mu_5 \sin^2\varphi \cos^2\varphi$$

and

$$\gamma(x) = \left[\gamma_5 - (\gamma_2 + 2\gamma_5)\cos^2\varphi\right]\sin\varphi,$$

where μ_5, μ_{11} are viscosities, and γ_2, γ_5 are electromechanical coupling constants. $\varphi(x)$ would describe the C-director in the sample, but this function is unknown. It should be calculated from the equation of motion of the C-director[16,17] but unfortunately it cannot be solved exactly. Since the sample has a planar orientation, the C-director has to be parallel to the plates (strong anchoring at the boundaries), i.e., $\varphi = 0$ or $\varphi = \pm\pi$ must be fulfilled at $x = 0$ and $x = d$.

This means that $\mu(x)$, $\gamma(x)$ and $E(x)$ in Equation (11) can be expanded into a Fourier series as follows

$$\mu(x) = \mu_{\text{eff}} + \mu^{(1)}\sin\left(\frac{\pi}{d}x\right) + \text{higher harmonics},$$

$$\gamma(x) = \gamma^{(1)}\sin\left(\frac{\pi}{d}x\right) + \text{higher harmonics},$$

$$E(x) = \delta\left[1 + \alpha\sin\left(\frac{\pi}{d}x\right) + \beta\cos\left(\frac{\pi}{d}x\right)\right] + \text{higher harmonics}.$$

Supposing that it is sufficient to retain the first nonvanishing harmonics, Equation (11) finally becomes

$$i\omega\rho v(x) = -\frac{\partial}{\partial x}\left\{-\mu_{\text{eff}}\frac{\partial v}{\partial x} + i\omega\frac{U_0}{d}\gamma_{\text{eff}}\sin\left(\frac{\pi}{d}x\right)\right\}, \tag{12}$$

where

$$\mu_{\text{eff}} = \mu_{11} + \frac{1}{8}\mu_5, \qquad \gamma_{\text{eff}} = \frac{\frac{1}{2}\gamma_2}{1 + 2\alpha/\pi}$$

and

$$U_0 = \int_0^d E(x)\,dx = \delta\left(1 + \frac{2\alpha}{\pi}\right)d$$

is the amplitude of the applied voltage. This equation is supplemented with the boundary conditions for the velocity (see Equation (4)):

$$v(0) = 0, \qquad v(d) = i\omega u.$$

With the approximations listed above the equation of motion of the upper plate becomes

$$-m\omega^2 u = -ku - \Omega\mu_{\text{eff}}\left.\frac{\partial v}{\partial x}\right|_{x=d}. \tag{13}$$

These equations can now be solved exactly resulting in the following expression for the amplitude of vibration:

$$u = i\omega\gamma_{\text{eff}}\frac{U_0}{d}\Omega\frac{2\pi}{\pi^2 + \lambda^2 d^2}\frac{\lambda d/2\,\text{cth}\,\lambda d/2}{k - m\omega^2 + i\omega\mu_{\text{eff}}\Omega\lambda\,\text{cth}\,\lambda d}, \tag{14}$$

where $\lambda = \sqrt{i\omega\rho/\mu_{\text{eff}}}$.

In the low frequency limit Equation (14) yields the asymptotic formula

$$u(\omega \to 0) = i\omega\gamma_{\text{eff}}\frac{U_0}{d}\Omega\frac{2}{\pi}\frac{1}{k}. \tag{15}$$

This formula shows that for low frequencies the displacement of the upper plate is proportional to ω in accordance with our experimental results (see Figure 4). Equation (15) gives a good approximation of Equation (14) below the natural frequency $f_0 = (1/2\pi)\sqrt{k/m}$ of the system.

f_0 has been determined experimentally applying a voltage to the loudspeaker and measuring the amplitude of vibration of the upper plate versus frequency. Depending on the quality of the alignment and the sample thickness we obtained $f_0 =$ 300–600 Hz, a value which is in good agreement with the data in Figure 4.

SUMMARY

A periodic electric field induced mechanical vibration of the same frequency was detected in a S_C^* mixture. It was proved experimentally that this linear electromechanical effect exists in the ferroelectric phase only. In contrast to piezoelectrics, a static field does not induce deformation.

The phenomenon is interpreted as a result of a new cross-effect termed electromechanical coupling. Our model is in good qualitative agreement with the experimental results.

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