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Mechano-Electrical Effects on Planar S_C^* Liquid Crystals

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Shear flow induced voltages (mechano-electrical responses) were detected and analyzed in two planar S_C^* liquid crystal samples. Frequency spectra, temperature and shear rate dependences were measured. Resonances observed in the frequency spectra are in accordance with the resonances were found in the electromechanical responses of these materials. The origin of the mechano-electrical effect is interpreted in the frame of shear flow alignment mechanism.

Keywords: liquid crystals (6130), ferroelectricity (7780), electrical properties (7390), electrical resonances (7785), piezoelectricity (7760)

INTRODUCTION

Due to their unique symmetry properties ferroelectric liquid crystals show piezo-electric behaviours. As it was found S_C^* liquid crystals convert alternating electric fields into mechanical vibrations,^{1–5} and vice versa, due to mechanical deformations electric polarization appears on the samples.^{6,7}

The former one is called electromechanical effect and was observed on planar S_C^* liquid crystals. Due to alternating electric fields the bounding plates vibrate relative to each other. Both responses measured at the frequency of the applied AC field and the first harmonics may have several resonances in the kilohertz frequency range. One of these resonances appears only in focal conic textures (defects are present at the cover plates), while others exist even in homogeneous planar samples⁴ (layers are nearly perpendicular to cover plates without any defect on surfaces). It is suggested, that the resonances are the consequence of chevron structure of S_C^* liquid crystals.⁸

The latter effect (hereafter we call it mechano-electrical effect) first was found by Pieranski *et al.*⁶ in homeotropic S_C^* sample (smectic layers were parallel to cover plates), and was measured at low frequencies ($f < 200$ Hz). Later similar effect was reported without any details⁷ also on planar S_C^* samples.

In this paper we describe our detailed investigations on the mechano-electrical effect. The frequency spectra of induced voltages due to unit amplitude vibrations of the upper bounding plate were determined in two planar aligned S_C^* samples.

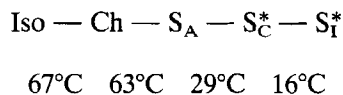
Temperature and shear amplitude dependences are mentioned too. Origin of this effect is explained in the frame of flow alignment mechanism, and the observed data are compared to the calculated formulae.

EXPERIMENTS

The investigations are carried out on sandwich cells with boundary area of $\Omega \cong 6$ cm² of two planar aligned S_C^* liquid crystal samples. The liquid crystal films were planarly aligned by means of a periodic shear¹⁰ supplied by an excited membrane of a loudspeaker, while samples were cooled down into their S_A phase. In order to study the effect of shear flow a vibration of the upper plate was maintained at S_C^* phase of the sample with the help of a vibrating loudspeaker membrane. For measuring the resulting amplitude, a Brüel & Kjaer accelerometer (BK 4375) was fixed on the top plate. The signal of accelerometer was preamplified with a BK 2645 charge amplifier, and was analyzed by an ITHACO (Model 3962) lock-in amplifier. To avoid higher harmonics the lock-in together with a sharp band pass filter was tuned on the frequency of the vibrating loudspeaker. The oscillation amplitude of the upper plate was determined by measuring the acceleration and dividing it by ω^2 ($\omega = 2\pi \cdot$ vibration frequency). Bounding plates were coated by SnO_2 transparent electrodes. The electrodes were connected to the input of the lock-in. (The input impedance of the lock-in can be regarded as a parallel RC element with $R_I = 10$ M Ω and $C_I = 40$ pF.) The loudspeaker was excited by the internal oscillator of the lock-in. Frequency was changed in steps from $f = 100$ Hz up to $f = 10$ kHz. Measurements were controlled by a personal computer.

Samples were thermostatted with the accuracy of $\pm 0.3^\circ\text{C}$, and visually observed by a polarizing microscope. The sample thickness and parallelism were set by fixing the lower plate to micrometer screws. The parallelism and sample thickness were checked with the help of a laser beam and by capacitance measurements respectively. By this method the accuracy of parallelism and sample thickness were better than 10^{-4} rad and ± 2 μm respectively. The block scheme of the experimental arrangement is presented in Figure 1.

Experiments were carried out on two room temperature S_C^* liquid crystal mixtures. 1.) A binary mixture FK4. Its phase sequence is the following:



at $T = 23^\circ\text{C}$ its pitch is $p = 5$ μm and its spontaneous polarization is⁷ $P_o = 1.2 \cdot 10^{-5}$ C/m².

2.) A thermochromic ferroelectric liquid crystal¹¹ called N202 with a high spon-

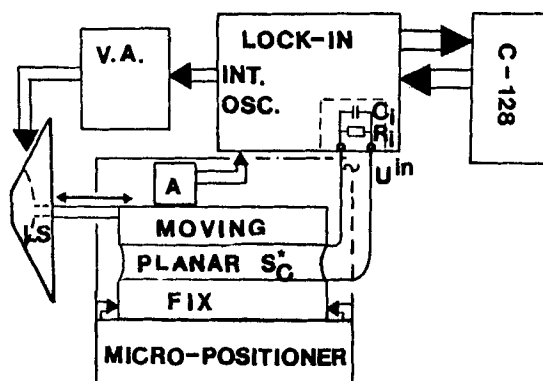


FIGURE 1 Block-diagram of the experimental arrangement. The planar S_C^* liquid crystal film is sheared by means of a vibration of the upper bounding plate. The vibration is ensured by a vibrating membrane of a loudspeaker (LS) which is excited by the appropriately amplified (V.A.) signal of the internal oscillator of the lock-in. The induced voltages U^{in} and the acceleration of the moving plate were detected on evaporated SnO_2 electrodes by a BK 4375 Brüel & Kjaer accelerometer (A), and were analyzed simultaneously by a Ithaco Lock-in Amplifier. The lock-in was controlled and its signals were processed by a personal computer.

The sample parallelism and thickness were set by fixing the lower plate to micrometer screws. The sample was thermostatted (TH).

taneous polarization ($P_o = 70 \cdot 10^{-5} \text{ C/m}^2$) and with pitch in the light wavelength regime ($p = 0.3 \mu\text{m}$)¹² at room temperature. The phase sequence of N202 is:

$$\text{Iso} - S_A - S_C^* - \text{Cr}$$

$$58^\circ\text{C} \quad 57^\circ\text{C} \quad 8^\circ\text{C}$$

EXPERIMENTAL RESULTS

In Figure 2 we plotted the frequency spectrum of the mechano-electrical response of a planar aligned FK4 sample ($T = 25^\circ\text{C}$). Induced voltages at constant vibrational amplitude were observed in the lock-in input. The sample thickness was $d = 19 \mu\text{m}$, and the sample impedances (the sample is a parallel RC element) were $R_s = 20 \text{ M}\Omega$ and $C_s = 1.13 \text{ nF}$.

The continuous line is a fit on measured data. Measuring errors are indicated in the spectrum. At low frequencies ($f < 0.5 \text{ kHz}$) the errors came from the measurement error of vibrational amplitudes,^{4,5} while at high frequencies ($f > 4\text{--}5 \text{ kHz}$) the induced voltages were within the measuring error. In the frequency interval $0.5 \text{ kHz} < f < 5 \text{ kHz}$ the measuring error everywhere was smaller than $\pm 20\%$. At this regime two main resonances were found at $f_1 = 2.9 \text{ kHz}$ and $f_2 = 4.5 \text{ kHz}$ frequencies which correspond to the resonances found in electromechanical responses of the same material.⁴

Spectrum of N202 is similar to that of FK4. At $T = 25^\circ\text{C}$ for a $d = 15 \mu\text{m}$ sample thickness sandwich cell (its electrical parameters are $R_s = 2 \text{ M}\Omega$ and $C_s =$

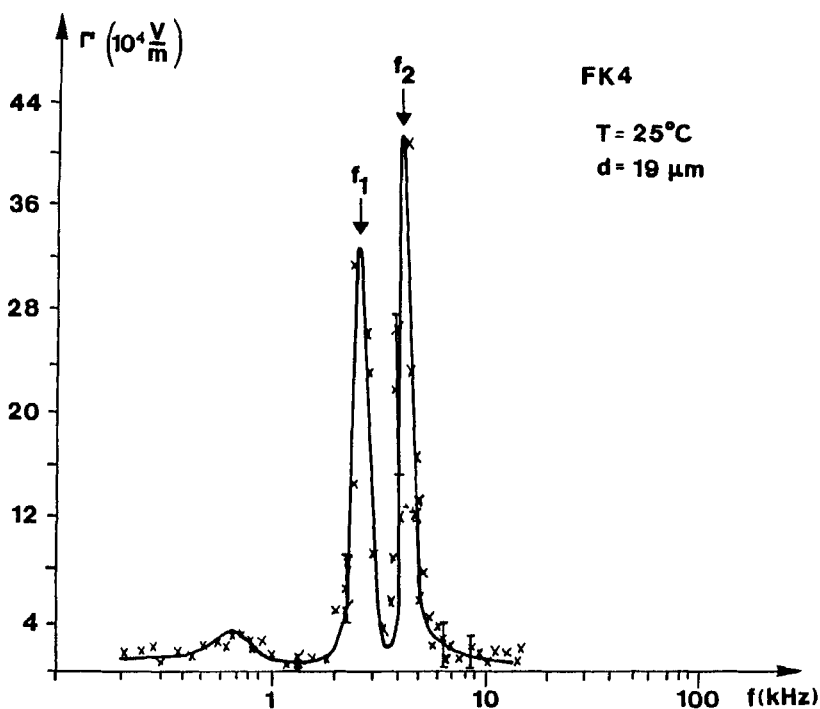


FIGURE 2 Frequency spectrum of mechano-electrical response of a periodically sheared planar aligned FK4 S_C^* liquid crystal sample. Γ means the ratio of the induced voltages and the amplitudes of upper bounding plate vibration. Induced voltages were detected in the lock-in input (which impedances are $R_i = 10 \text{ M}\Omega$ and $C_i = 40 \text{ pF}$). The sample parameters were: $T = 25^\circ\text{C}$, $d = 19 \mu\text{m}$, $R_s = 20 \text{ M}\Omega$, $C_s = 1.13 \text{ nF}$.

0.588 nF) the frequency dependence of mechano-electrical response is plotted in Figure 3.

The largest resonance was found at the same frequency as in its electromechanical response⁴ ($f_1 = 3.9 \text{ kHz}$). We also found other smaller resonances which were not present at the electromechanical effect.

Comparing the maximum amplitudes of FK4 to that of N202 we can see that in FK4 induced voltages are about two orders of magnitude larger than in N202. It seems to be in contradiction with the fact that P_o (FK4) is about two orders of magnitude smaller than P_o (N202).

For the shear amplitude dependences simple linear functions were found. Examples (N202 sample, $T = 22^\circ\text{C}$, $d = 15 \mu\text{m}$ at frequencies $f = 3.8 \text{ kHz}$ and $f = 1.0 \text{ kHz}$) are plotted in Figure 4. We could not see any deviance from the linear behaviour, and did not observe any harmonics. Temperature dependence of mechano-electrical effect was found to be similar to that measured by Pieranski *et al.*⁶ in homeotropic samples at significantly smaller shear frequencies. This temperature dependence is also similar to that measured in electromechanical effects.^{1,2,4,5} The temperature dependence of induced voltages $U^{in}(\mu\text{V})$ is plotted in Figure 5. The acceleration of the upper plate was kept constant ($a = 5 \text{ m/s}^2$) at frequency of $f = 4.02 \text{ kHz}$ while the temperature was constantly $T = 23^\circ\text{C}$.

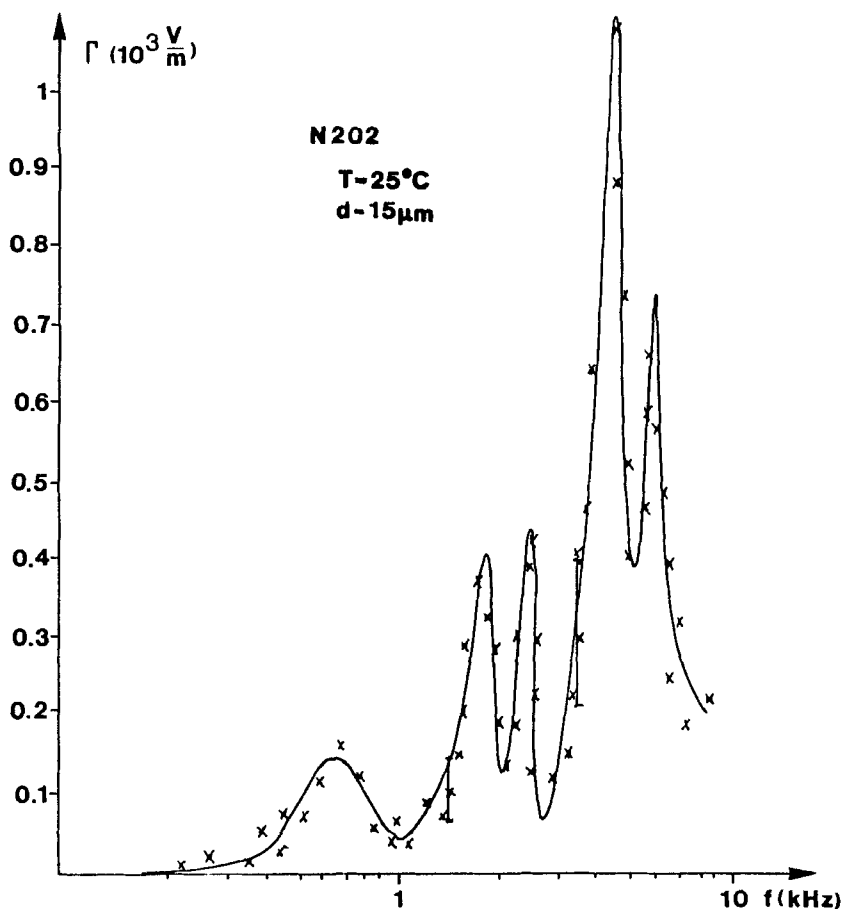


FIGURE 3 Frequency spectrum of mechano-electrical response of a periodically sheared planar N202 liquid crystal sample. In the vertical axis the ratio of the induced voltages and shear amplitude of upper bounding plate (Γ) are plotted. Induced voltages were detected in the lock-in input ($R_i = 10 \text{ M}\Omega$, $C_i = 40 \text{ pF}$). The sample parameters are the followings; $T = 25^\circ\text{C}$, $d = 15 \text{ }\mu\text{m}$, $R_s = 2 \text{ M}\Omega$ and $C_s = 0.588 \text{ nF}$.

INTERPRETATION

Presented mechano-electrical effect should be explained by the same mechanism which was considered by Pieranski *et al.*,⁶ because the relative positions of shear, induced polarization and helical axis are the same (they are perpendicular to each other) both in measurement of Pieranski *et al.* and our cases.

Accordingly we describe the mechanism resulting the observed effect as follows. Shear distorts the original director structure which, as C-director, \mathbf{c} and polarization, \mathbf{P}_0 , are rigidly coupled, results in a net polarization current flow through on electrodes.

Considering our geometry (see Figure 6) the schematic of the mechanism is the

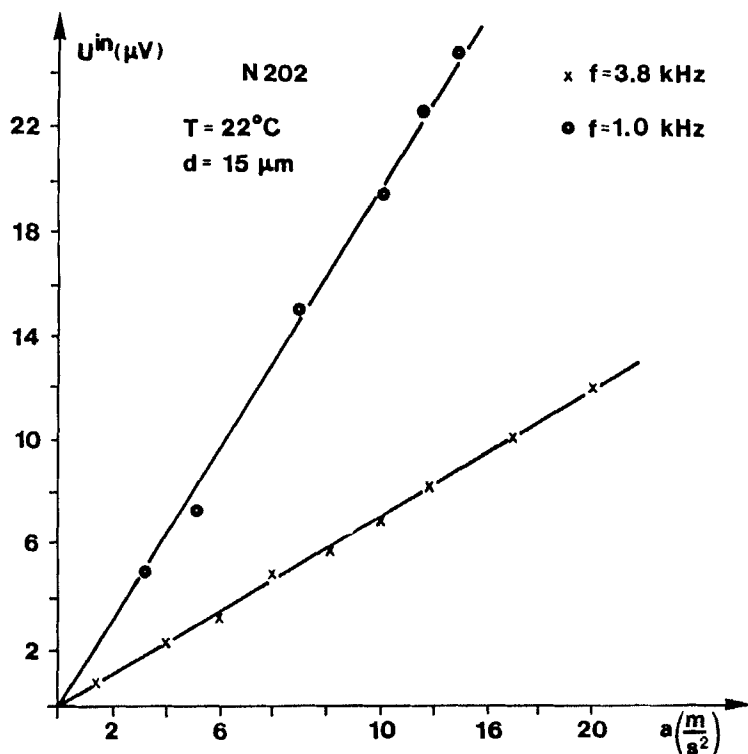


FIGURE 4 Shear amplitude dependence of mechano-electrical response of a planar aligned N202 liquid crystal sample. $T = 22^\circ\text{C}$. Other sample parameters are the same as in Figure 3. Crosses: $f = 3.8$ kHz; open circles: $f = 1.0$ kHz.

following:

$$\nabla_x v_y \Rightarrow \frac{\partial c_y}{\partial t} \Leftrightarrow \frac{\partial P_x}{\partial t} \rightarrow I(t) \Rightarrow U^{\text{in}} (= I(t) \cdot Z_{\text{eff}})$$

Here $v_y(x)$ is the shear flow velocity in the sample c_y is the y component of the C -director, \mathbf{c} (\mathbf{c} is the projection of the director into the smectic layer). \mathbf{P}_o is the spontaneous polarization ($\mathbf{P}_o/P_o = \mathbf{n} \times \mathbf{c}$; \mathbf{n} is the smectic layer normal).

As flow takes place inside smectic layers, where mechanical behaviour of the material is nematic-like, describing dynamics of flow alignment we can use a nematic like description. Till this point our description is identical to that of Pieranski *et al.*,⁶ however the form of the relevant torque balance equation should be different, because the structure of a planar film is different from that of a homeotropic.

In planar film the director configuration is affected by the cover plates. In this respect two main director structures are to be considered. a. When the sample thickness, d is smaller than the helical pitch, p (surface stabilized structure), then the sample is uniform in z direction, that is the azimuthal angle φ_o is constant. b. In case of thick samples ($d \gg p$) the director structure is helical, and the z dependence of the azimuthal angle reads: $\varphi_o = q \cdot z$ (here $q = 2\pi/p$).

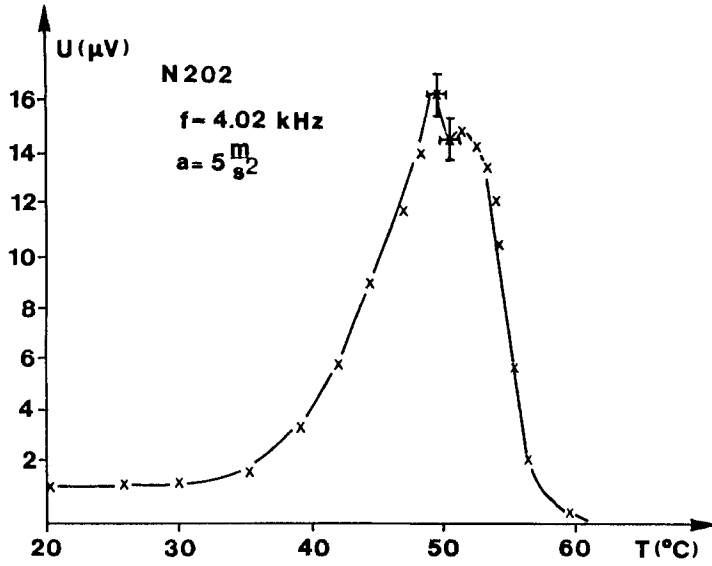


FIGURE 5 Temperature dependence of induced voltages due to constant amplitude periodic shears. The shear frequency is $f = 4.02$ kHz, the acceleration is $a = 5$ m/s. Other sample parameters are identical to that used in Figure 3.

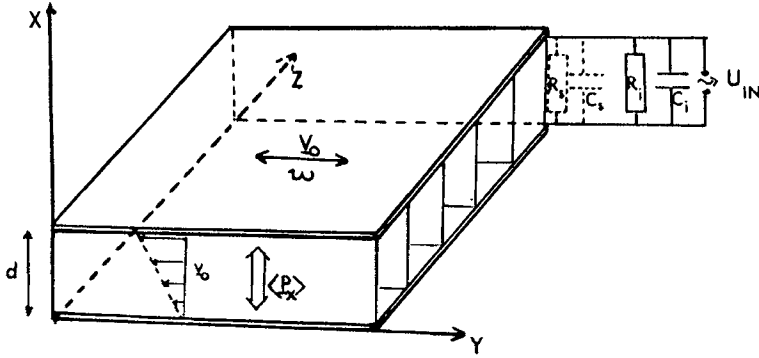


FIGURE 6 Sample geometry and the coordinate system during studying mechano-electrical responses. Top cover plate is vibrating in y direction causing a shear flow with velocity v_y inside smectic layers. This flow is coupled to director rotations, which results in an induced net polarization in x direction.

Dynamics of flow alignment should be described by solving the torque balance equation. a. For surface stabilized samples the director field is uniform, so elastic torques can be neglected. Therefore, the torque balance reads:

$$\mu(\varphi_o) \cdot \frac{\partial v_y}{\partial x} + \dot{\varphi} \cdot \gamma_1 = 0 \quad (1)$$

where $\mu(\varphi_o) = \alpha_3 \cdot \cos^2 \varphi_o - \alpha_2 \cdot \sin^2 \varphi_o$ is the relevant shear viscosity and $\gamma_1 = \alpha_3 - \alpha_2$ is the rotational viscosity⁹ (α_2, α_3 are Leslie coefficients).

The shear is a harmonic function of time and maintained by the oscillation of the cover plate, so:

$$v_y = v_o \cdot e^{i\omega t}, \text{ and } \frac{\partial v_y}{\partial x} \cong \frac{v_o}{d} \cdot e^{i\omega t} \quad (2)$$

Supposing that due to shear the variation of azimuthal angle is small, Equation (2) and Equation (3) provides that in uniform samples the shear induced azimuthal angle variation φ_1^u reads:

$$\varphi_1^u = \frac{\mu \cdot v_o}{\omega \cdot d \cdot \gamma_1} \quad (3)$$

b. In case of thick samples we should consider the spatial derivatives in the z direction too. As v_o does not depend on z , viscous torques are identical to the left hand side of Equation (1), except we have to consider the z dependence of φ_o . Now the elastic torque, M^{el} arising during the process is not zero and from Leslie continuum theory⁹ it has the form:

$$M^{el} \cong K_2 \cdot q^2 \cdot \varphi_1 \cdot \xi(z) \cdot e^{i\omega t}, \quad (5)$$

where

$$\xi(z) = 9 \cdot (\cos^3 qz - \sin^3 qz) - 6 \cdot (\cos qz - \sin qz) \quad (6)$$

is in the order of unity.

Disregarding the x dependences, furthermore using Equation (2) and Equation (3) the torque balance can be written as

$$\mu(\varphi_o) \cdot \frac{v_o e^{i\omega t}}{d} + i\omega \varphi_1^h \cdot e^{i\omega t} + K_2 \cdot q^2 \cdot \varphi_1^h \cdot e^{i\omega t} \cdot \xi(z), \quad (7)$$

from which for the variation of the azimuthal angle φ_1^h we get that

$$\varphi_1^h = \frac{\mu}{d \cdot \gamma_1} \cdot -\frac{v_o}{\sqrt{\omega^2 - (\xi/\tau)^2}}, \quad (8)$$

where $\tau = \gamma_1/(K_2 q^2)$.

The average polarization $\langle P_x \rangle$ induced on electrodes reads

$$\langle P_x \rangle = P_o \cdot \langle \varphi_1 \rangle \quad (9)$$

Electric charge Q appears on electrodes is:

$$Q = \langle P_x^{in} \rangle \cdot \Omega_{eff} \quad (10)$$

Here Ω_{eff} is an effective surface area. By definition it is equal the area of a monodomain sample where the director flipping in whole takes place collectively with the same phase. So, if the sample is a monodomain Ω_{eff} is equal the whole sample area; however if it consists of more domains, then $\Omega_{eff} < \Omega$, and gives the resulting area of different domains (let's say there are two domains with area Ω^+ and Ω^- , then $\Omega_{eff} = \Omega^+ - \Omega^-$).

Because $Q(t) = Q_0 e^{i\omega t}$, the induced electric current, I (the sample can be regarded as an electric current generator) reads: $I = \omega \cdot Q_0$.

Induced voltage U_{in} measured on parallel RC circuit built from sample impedances R_s , C_s and input impedances of lock-in R_I , C_I reads:

$$U_{in} = I \cdot |Z_{eff}| = I \cdot \frac{R}{\sqrt{1 + R^2 C^2 \omega^2}} \quad (11)$$

As the lock-in and the sample are electrically parallel, in this equation

$$R = \frac{R_I \cdot R_s}{R_I + R_s}; \text{ and } C = C_I + C_s \quad (12)$$

Above equations together with our experimental data and sample parameters allow us to calculate effective surface areas of measured samples.

Hence pitch of N202 is $p \cong 0.3 \mu\text{m} \ll d$, the structure of this sample is well described by ideal helical model. Therefore, for calculation of the effective surface we have to use Equation (8) and Equations (9–12). Doing so, furthermore supposing that $\mu \sim \gamma_1$ we get $\Omega_{eff}(\text{N202})$ is typically in the order of $.1 \text{ mm}^2$ which is much less than $\Omega \cong 6 \text{ cm}^2$, indicating that the material consists of small domains.

In case of FK4 $p = 5 \mu\text{m} \leq d = 19 \mu\text{m}$. Therefore the middle of the material is helical too, but close to surfaces the sample is unwound.¹³ Because total thickness of uniform regions is approximately equal to the size of the pitch, its volume is not negligible relative to that of the region of helical structure. So, for calculating the average azimuthal angle variation, φ_1 we can write that $\varphi_1 \cong (3 \cdot \varphi_1^h + \varphi_1^u)/4$. Executing the computation this way we get for Ω_{eff} that it is typically in the order of $0.1\text{--}1 \text{ cm}^2$, and at resonance it is comparable to the whole sample area. This indicates that FK4 consists of less and larger domains, which surface areas are in the range of cm^2 .

Visually observing our samples we really found so called zig-zag defects^{14,15} inside the samples separating monodomains and saw that the typical areas of monodomains are comparable to the areas concluded from the above considerations. It also agrees with earlier observations⁴ on the same materials.

SUMMARY

Frequency spectra, temperature and shear rate dependences of mechano-electrical responses of two S_C^* planar liquid crystal samples were investigated experimentally. The resonances found in frequency spectra well correlate with that of observed in

electromechanical measurements.^{4,5} The origin of the observed mechano-electrical effect is explained in the frame of shear flow alignment mechanism and calculation is completed for surface stabilized and ideal helical structures. Calculated results are compared to experimental data and found that in different domains contained in the samples the induced polarizations are not in phase, making thus the resulting effect suppressed. The resonances observed in the frequency dependences are probably also in connection with the visually observed domain structures and caused by the zig-zag defects which separate different monodomains. Effect of the zig-zag lines on frequency dependence of electromechanical responses is discussed elsewhere.⁸

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