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Non-linear electromechanical response of S_C^* liquid crystals

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The non-linear part of the electromechanical response of four S_C^* liquid crystals were investigated. The second harmonic frequency vibrational spectra, the electric field, and the temperature dependences are presented. The experimental results are interpreted in terms of a microscopic process.

1. Introduction

Although fast bistable electro-optical switching is the most promising effect for applications of ferroelectric liquid crystals [1–3], their electromechanical behaviour [4–7] is also of interest. As a consequence of their unique symmetry properties ferroelectric liquid crystals are also piezoelectric. Due to mechanical shear electrical charges appear (both in homeotropic [4] and in planar alignment [8, 18]), and AC electric fields applied to planar aligned samples produce mechanical flows in the smectic layers perpendicular to the electric field [5, 6]. For a substance having a small spontaneous polarization the effect is mainly linear [6]: the vibrational frequency is equal to that of the applied electric field, and the amplitude is proportional to the applied electric field strength. Non-linearity has been found only at low frequencies [6].

In this paper we describe our investigations of non-linear electromechanical effects. Measurements were carried out on four room temperature ferroelectric liquid crystals. Hereafter we try to answer to the following questions.

> How do the second harmonic vibrational frequency amplitudes depend on the applied frequency?
>
> What is the connection between the vibrations at the second harmonic frequency and the fundamental vibrational frequency?
>
> How does the non-linearity depend on the spontaneous polarization of the material?
>
> What is the nature of the non-linear electromechanical effects?

2. Experiments

For these investigations four substances each with a S_C^* phase at room temperature were used. Their identification codes, phase sequences, their pitch p, and spontaneous polarization P_0, at room temperature ($T = 23°C$) are summarized in the table. The experimental set-up is almost the same as that used during the investigations of the fundamental frequency electromechanical responses (vibrations are measured at the frequencies are equal to the one of the electric excitations) [13]. A block diagram of the arrangement is presented schematically in figure 1.

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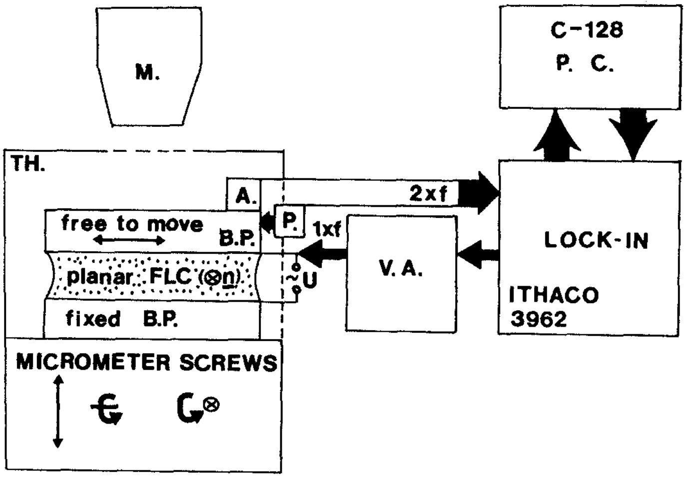

Figure 1. Schematic of the experimental set-up. The planar aligned S_C^* film is between two bounding glass plates (B.P.). The upper plate can move freely parallel to the fixed lower bounding plate and perpendicular to the smectic layer normal **n**. The vibration of the upper plate is detected either by a displacement sensitive pick-up (P.), or by a Brüel & Kjaer BK4375 accelerometer (A.). The signals of the detectors are analysed by an Ithaco 3962 lock-in amplifier. The sample is excited electrically by harmonic voltages supplied by the internal oscillator of the lock-in and then amplified by a voltage amplifier (V.A.). The sample is thermostatted (TH.) and observed by a polarizing microscope (M.). The bounding plates are micro positioned by micrometer screws.

Parameters for the substances investigated

Name	Phase sequence	Pitch/μm	Polarization/Q m^{-2}
FK4 [5, 8]	$\mathrm{I}\underset{67^\circ\mathrm{C}}{\text{——}}\mathrm{N}^*\underset{63^\circ\mathrm{C}}{\text{——}}\mathrm{S_A}\underset{29^\circ\mathrm{C}}{\text{——}}\mathrm{S_C^*}\underset{16^\circ\mathrm{C}}{\text{——}}\mathrm{S_I^*}\text{——}$	5	$1{\cdot}2 \times 10^{-5}$
CS1011 [9]	$\mathrm{I}\underset{91^\circ\mathrm{C}}{\text{——}}\mathrm{N}^*\underset{78^\circ\mathrm{C}}{\text{——}}\mathrm{S_A}\underset{56^\circ\mathrm{C}}{\text{——}}\mathrm{S_C^*}\text{——}$	5	$1{\cdot}5 \times 10^{-4}$
N202 [10, 11]	$\mathrm{I}\underset{58^\circ\mathrm{C}}{\text{——}}\mathrm{S_A}\underset{57^\circ\mathrm{C}}{\text{——}}\mathrm{S_C^*}\underset{8^\circ\mathrm{C}}{\text{——}}\mathrm{C}$	0·3	7×10^{-4}
BW1 [12]	$\mathrm{I}\underset{70^\circ\mathrm{C}}{\text{——}}\mathrm{N}^*\underset{54^\circ\mathrm{C}}{\text{——}}\mathrm{S_A}\underset{38^\circ\mathrm{C}}{\text{——}}\mathrm{S_C^*}\underset{0^\circ\mathrm{C}}{\text{——}}\mathrm{C}$	> 50	$1{\cdot}7 \times 10^{-5}$

The investigations were carried out on planar aligned samples (the smectic layers were perpendicular to the substrate plate). For FK4 and N202 we obtained good quality homogeneous samples by applying a shear at the N*–S_A and I–S_A transition temperatures respectively [14]. For BW1 relatively good results were only found when an electric field ($E > 10^6\,\mathrm{V\,m^{-1}}$; $f < 5\,\mathrm{Hz}$) was applied together with shear while cooling the material. For CS1011 we could not make perfect homogeneous samples; focal conics always were present. The best results were obtained (i.e. the diameters of focal conics were the largest) when together with the shear we also used a PVA coating on the surfaces which then were rubbed perpendicular to the direction of shear.

The shear was applied by means of a vibrating loudspeaker membrane which was connected via a rod to the upper bounding plate. The upper plate was attached to the rod by creating a vacuum between them. We investigated the vibration of the upper bounding plate when it was free to move. The vibration of the bounding plate was detected both by a displacement sensitive ceramic pick-up (Tesla VK4302) which touched the edge of the upper plate, and by a calibrated Bruel & Kjaer BK 4375 accelerometer (with a BK 2645 preamplifier) which was fixed to the top of the upper plate. The signals of the pick-up and the accelerometer were analysed by an Ithaco (Model 3962) lock-in amplifier. The harmonic electric field was applied to the sample by amplifying the signal of the internal oscillator of the lock-in. To minimize harmonic components a sharp band pass filter was tuned to the double frequency of the applied voltage, thus second harmonic frequency vibrational spectra were measured. The amplitudes and the phases of the vibration were detected at the applied frequency in the range from 100 Hz to 5 kHz in steps of 10 Hz. The measurements were controlled by a Commodore 128 personal computer.

Comparing the results obtained from the pick-up signals to those obtained from the signals of the accelerometer divided by $(2\omega)^2$ ($\omega = 2\pi \times$ applied frequency) we found them to be equal in the applied frequency interval of $0{\cdot}3\,\mathrm{kHz} < f < 2\,\mathrm{kHz}$. At low frequencies ($f < 0{\cdot}3\,\mathrm{kHz}$) the accelerometer is not reliable; its signal is in the error range of the measurement. While in the region $f > 2\,\mathrm{kHz}$ the pick-up is not applicable. Calibrating the accelerometer [15] it was found that it is fully reliable until applied voltages at 5 kHz.

The parallelism of the bounding plates was ensured by micrometer screws and was checked with a laser beam. In each sample the parallelism was better than 10^{-3} rad. The sample thickness was set by either micrometer screws or by teflon spacers, and was determined by measurement of the capacitance. The glass plates were covered with evaporated SnO_2 to give transparent electrodes. The samples were heated and thermostatted. The temperature control was better than $\pm 0{\cdot}5$°C.

3. Experimental results

3.1. *Vibrational amplitudes*

In a given substance the amplitudes of the second harmonic vibrational frequency followed the amplitudes of fundamental vibrational frequency. For the amplitude ratios of the fundamental and second harmonic vibrational frequency λ, we generally found that for $E \approx 10^6\,\mathrm{V\,m^{-1}}$ at frequencies $f < 1\,\mathrm{kHz}$: $\lambda \approx 10$ and at frequencies $f > 2\,\mathrm{kHz}$: $\lambda \approx 10^2$. Comparing the substances, in this respect, it was found that $\lambda(\mathrm{FK4}) \geqslant \lambda(\mathrm{BW1}) \approx \lambda(\mathrm{N202}) \approx 3\lambda(\mathrm{CS1011})$.

3.2. *Frequency spectra*

Typical frequency spectra of the second harmonic vibrational frequency are seen in figures 2–5. These spectra were measured on the same samples as the fundamental frequency spectra had been investigated [13]. In the fundamental frequency spectra two types of resonances were found.

(*a*) The resonance frequencies f_1 and f_2 did not depend on the mass of the moving plate. These resonances were present both in homogeneously aligned and in focal conic planar samples.

(*b*) The resonance frequency f_0 decreased with increasing the mass of the moving plate. This resonance was present only for focal conic textures. In linear spectra at low frequencies ($f < f_0$) the vibrational amplitudes increased monotonical with increasing frequency.

In the second harmonic frequency spectra we also found resonances at the applied field frequencies $f_0/2$, $f_1/2$ and $f_2/2$. This means that the same resonances were found here as in the fundamental frequency spectra, because when the applied voltage

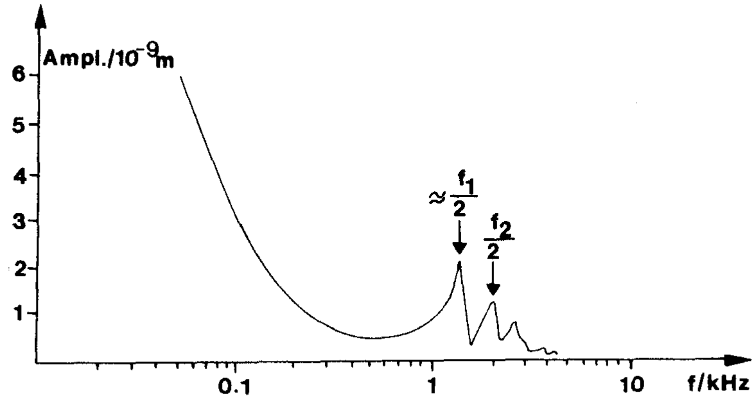

Figure 2. The second harmonic frequency spectrum of a homogeneously aligned FK4 sample. The sample thickness is $d = 19\,\mu$m and, the temperature is $T = 22°$C. The sample is excited by harmonic voltages with the amplitude of $U = 21$ V. The vibrational amplitudes at the frequencies, of the second harmonic of the applied voltages, are detected by a pick-up and by an accelerometer. The mass of the moving plate m is 10 g.

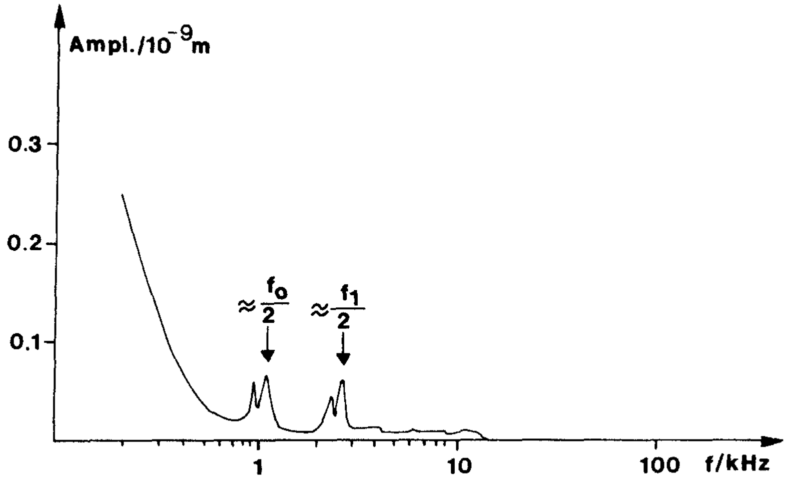

Figure 3. The second harmonic frequency spectrum of the vibrational amplitude of a CS1011 sample. The sample thickness is $d = 45\,\mu$m and temperature is $T = 22°$C. The mass of the moving plate m is 10 g. The sample is excited by harmonic voltages with the amplitude of $u = 30$ V.

frequency is e.g. $f_0/2$ the measured sample vibration is f_0. We also found that the phase function is discontinuous at the resonances. In contrast to the fundamental frequency spectra, the vibrational amplitudes decreased monotonically at low frequencies in the second harmonic frequency spectra. Disregarding the resonances, the steepest decrease was found near the Goldstone mode relaxation frequencies [16] f_G. (From switching time measurements $f_G \approx 70$ Hz; ≈ 4 kHz; ≈ 300 Hz and ≈ 150 Hz were found for FK4, N202, CS1011 and BW1, respectively.)

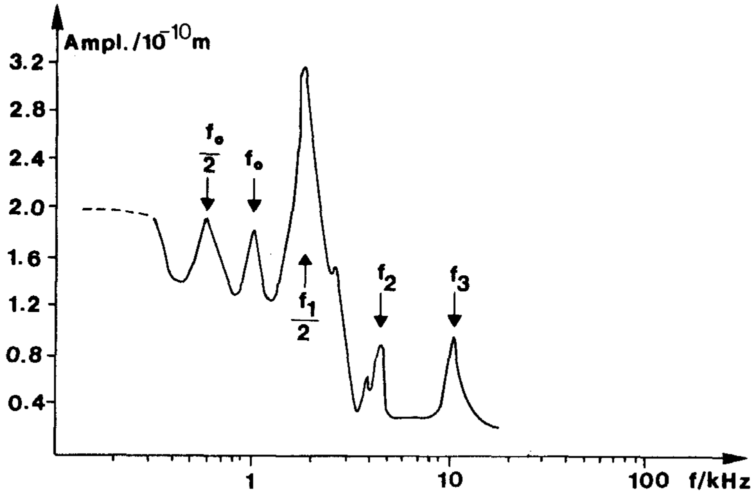

Figure 4. The second harmonic frequency spectrum of the vibrational amplitudes of a N202 sample. The sample thickness is d = 15 μm and the temperature is T = 26°C; the upper plate mass m is 9·5 g and the applied voltage amplitude is U = 5 V. $f_3/f_2 = f_2/(f_{1/2})$ = 2·4.

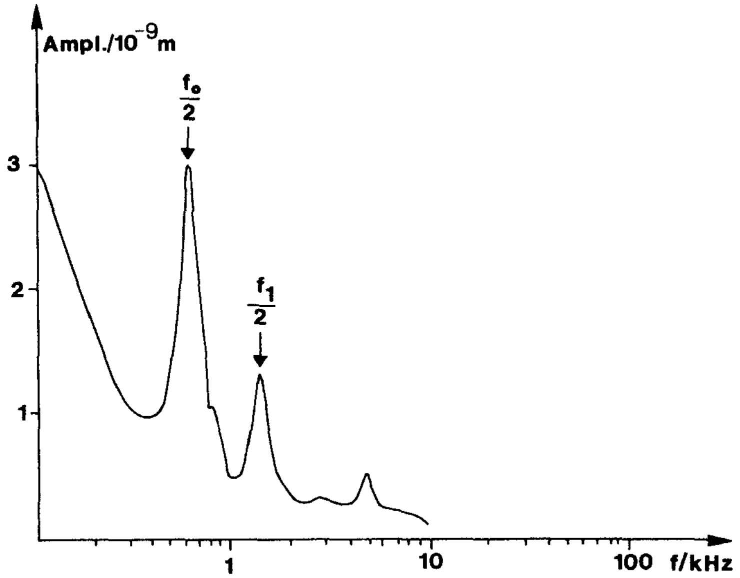

Figure 5. The second harmonic frequency spectrum of a focal conic planar BW1 sample. (d = 15 μm, T = 25°C, U = 15 V, m = 10 g).

3.3. *Non-linearities in the applied voltage dependence*

We have investigated the applied voltage dependence of the fundamental and second harmonic frequency responses. For FK4 (which has the smallest spontaneous polarization) the fundamental frequency dependence was linear, while the second harmonic frequency response was quadratic. This behaviour held until $E = 5 \times 10^6\,\mathrm{V\,m^{-1}}$. The applied voltage dependence of the fundamental and the second harmonic frequency responses can be seen in figure 6 (d = 19 μm, T = 22·5°C,

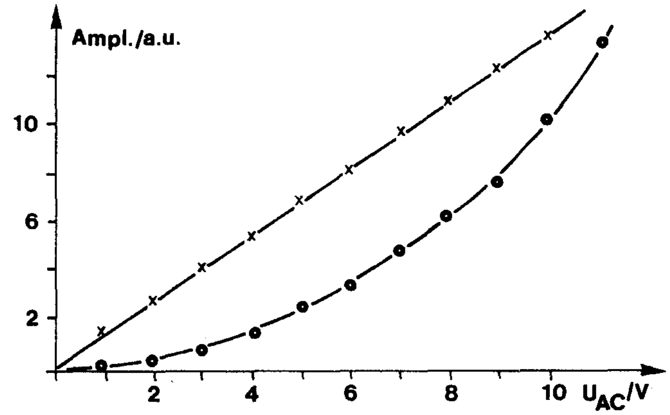

Figure 6. Electric field dependence of the fundamental and second harmonic frequency electromechanical response of a FK4 sample (d = 19 μm, T = 22·5°C, f = 3 kHz). (×): fundamental frequency response; (○): second harmonic frequency response.

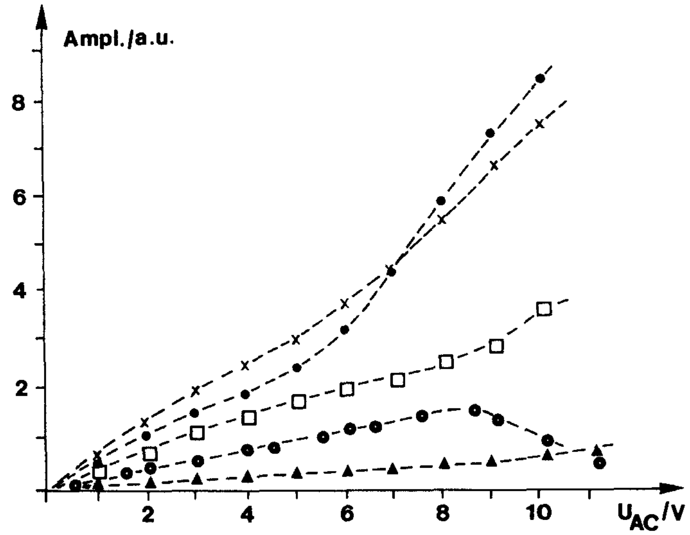

Figure 7. Electric field dependence of the fundamental frequency electromechanical response of a CS1011 sample (d = 19 μm). ●, T = 41°C, f = 4·14 kHz; ×, T = 28°C; f = 4·14 kHz; □, T = 16·5°C; f = 4·23 kHz; ○, T = 41°C; f = 414 Hz; ▲, T = 16·5°C; f = 2·34 kHz.

f = 3 kHz). This type of ideal behaviour was also found for BW1 (its spontaneous polarization is also very small). For these two materials deviations from this ideal behaviour were found only at low frequencies (f < 0·6 kHz). However CS1011 and N202 do not behave according to this ideal applied voltage dependence. For CS1011 the applied voltage dependence of the fundamental frequency response under different conditions are presented in figure 7. Deviation from the linear behaviour already occurs at small electric fields ($U \leqslant 5$ V; d = 19 μm). The non-linearity is most apparent at low frequencies. For example at f = 414 Hz at large enough voltages (U > 9 V) the vibrational amplitude decreased with increasing voltages. Similar tendencies were found also in the second harmonic frequency response of the CS1011 samples.

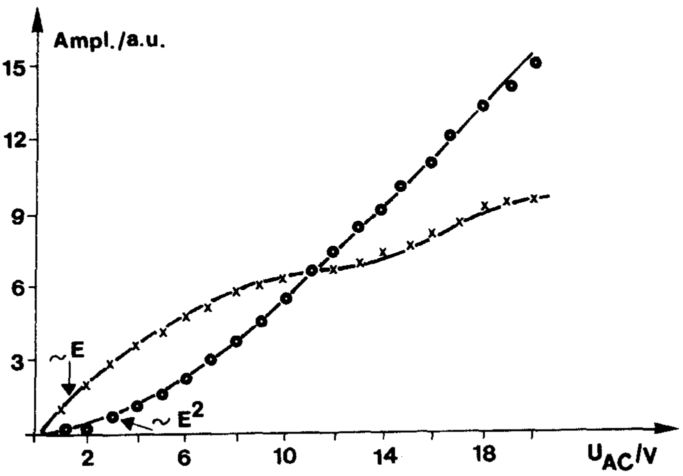

Figure 8. Electric field dependence of the fundamental and the second harmonic frequency electromechanical response of a N202 sample ($T = 23°C$, $d = 15\,\mu m$, $f = 1\,kHz$). ×, fundamental frequency response; ○, second harmonic frequency response.

In figure 8 we show an interesting voltage dependence for a N202 sample ($d = 15\,\mu m$; $T = 23°C$; $f = 1\,kHz$). The fundamental frequency response (denoted by crosses) and the second harmonic frequency response (denoted by open circles) behave ideally only until U of 5 V. At larger voltages however the second harmonic frequency dependence is more linear than quadratic. Also interesting is the fundamental frequency behaviour between U of 10 V and 17 V; in this interval an inflection is seen. Our visual observation showed [11], that at this regime a special instability (an irregular stripe system) exists in the sample. Thus we observed that a static director field deformation can also be seen in the electromechanical response. Similar conclusions were arrived at on the basis of investigations with DC bias fields [19].

3.4. *Temperature dependence*

The temperature dependence of the second harmonic frequency response was found to be similar to that of the fundamental frequency response [13]. As the temperature decreases from the S_C^*–S_A transition temperature the vibrational amplitude increases and reaches a maximum 2–3°C below the transition temperature, then decreases and saturates at a significantly smaller vibrational amplitude. In the case of N202 this behaviour is presented in figure 9 ($d = 15\,\mu m$, $U = 5\,V$, $f = 4{\cdot}5\,kHz$).

4. Discussion and conclusions

We have found two, seemingly different types of non-linearity in the electromechanical response of S_C^* phases.

(i) Applying an alternating voltage on planar aligned S_C^* samples the freely moving plate vibrates not only with the frequency of the applied voltage, but, with significantly smaller amplitudes, with second harmonic frequencies too. The frequency spectra of this latter behaviour were investigated for four different S_C^* materials. The resonances which were found in the second harmonic frequency responses correspond to the resonances of the fundamental frequency response. From

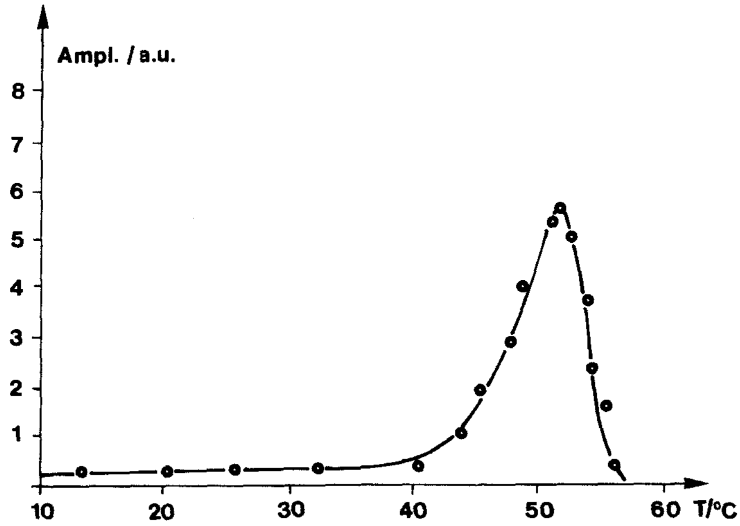

Figure 9. Temperature dependence of the second harmonic vibrational amplitudes of a N202 sample. ($d = 15\,\mu$m, $U = 5$ V, $f = 4{\cdot}5$ kHz).

this fact we conclude that the resonance must be associated with the structure of the samples. As proved in [17] the resonances are the consequences of the chevron structure [20, 21] of the S_C^* phases. The global frequency dependence of the second harmonic vibrational frequency is significantly different from that of the fundamental vibrational frequency. Disregarding the resonances the second harmonic frequency spectra decrease monotonically. The frequencies where the steepest decreases were found correspond to the Goldstone mode relaxation frequencies f_G. (f_G describes the relaxation of helical distortions [16].)

(ii) The electric voltage dependence was found to be sensitive to the spontaneous polarization of the sample. At high frequencies ($f > f_G$) in materials with small spontaneous polarizations we obtained linear and quadratic functions for the fundamental and second harmonic frequency responses, respectively. If the materials have larger spontaneous polarizations, the former ideal behaviour holds only for small voltages. The frequency spectra of the vibrational amplitudes (disregarding the resonances which are due to the domains), and the non-linearity found in the electric field dependence can be interpreted with the following model.

Because the linear electromechanical effect exists even in unwound samples [19], it can be interpreted as the piezoelectric response of the constant director field. The linear electromechanical effect does not require time dependent, but only space dependent director fields. However from the electrooptical investigations we know, that due to sufficiently large electric fields the spontaneous polarization (consequently the director) follows the applied voltage variations. As the frequency decreases and the spontaneous polarization increases the depth of the director field oscillation also increases. For harmonic excitations the director field varies with the frequency of the applied voltage thus causing second harmonic frequency variations. For the vibrational amplitude $u(t)$ it was found [6, 17] that

$$u(t) = c\gamma(t)E(t). \tag{1}$$

This expression holds both when zig-zag defects are not supposed [6, 7], and when the effect of chevrons is incorporated [17]. In these different cases only the expressions for c (which is time independent) differ. $E(t)$ is the applied harmonic electric field, $E_0 \exp(i\omega t)$, where $\omega = 2\pi f$ and f is the applied frequency. $\gamma(t)$ is the electromechanical coupling constant. It depends explicitly on the director configuration, thus for moving director fields it depends explicitly on time. Consequently we can write that

$$\gamma(t) = \langle\gamma\rangle + \Delta\gamma_0 \exp(i\omega t), \tag{2}$$

where $\langle\gamma\rangle$ is the averaged electromechanical coupling constant, and Δ describes the amplitude of the director field oscillation due to the alternating electric excitations. Δ depends on the spontaneous polarization P_0, the electric field E_0, the excitation frequency f and other material parameters as the viscosity η, the elastic constants K and the helical pitch p.

From equations (1) and (2) we can see that

$$u(t) = c\langle\gamma\rangle E_0 \exp(i\omega t) + c\Delta\gamma_0 E_0 \exp(2i\omega t). \tag{3}$$

Thus the electromechanical effect has a fundamental frequency excitation contribution with a vibrational amplitude $c\langle\gamma\rangle E_0$, and a second harmonic frequency contribution with amplitude $c\Delta\gamma_0 E_0$. For high frequencies and small dipole coupling the averaged electromechanical coupling constant is $\langle\gamma\rangle \approx \gamma_0$ independent of the electric field, thus the fundamental frequency oscillation is linear with the electric field. At small fields Δ is presumably proportional to the electric field, consequently the second harmonic frequency vibrational amplitude is proportional to E^2. However at higher electric fields and lower frequencies the director field oscillation is larger, thus $\langle\gamma\rangle \neq \gamma_0$. For example, at frequencies smaller than the Goldstone mode relaxation frequency, and the electric field is larger than the unwinding critical field E_c the director field oscillates between the two unwound states, therefore $\langle\gamma\rangle \approx 0$. This is why, we found for CS1011 that at $f = 414$ Hz and $E > 0{\cdot}5 \times 10^6\,\mathrm{V\,m^{-1}}$ the fundamental vibrational frequency amplitude decreased with increasing electric field. Furthermore, physically it can be expected, that $\Delta(E)$ is saturated at large fields, so it is understandable why the electric field dependence of the second harmonic vibrational frequency was rather linear at large applied fields for N202 (see figure 8).

On the basis of this model we can also explain the inflection found in the fundamental frequency response in the 10–17 V range for the N202 sample (see figure 8). Here, due to the appearance of irregular stripes [11], $\langle\gamma\rangle$ changed causing a modification of the voltage dependence. The global second harmonic frequency spectra can be explained on the basis of equation (3). The amplitude of the director field oscillation Δ, which determines the second harmonic vibrational frequency amplitude decreases with increasing frequency, because as f increases P_0 cannot follow the applied field. The characteristic frequency, until P_0 can follow E, must be of the order of the Goldstone mode relaxation frequency.

We should note that second harmonic vibrational frequencies can be observed even in other liquid crystal phases. This might be due to the electrostriction which exists in all condensed matter. Consequently, electrostriction must contribute also to the observed second harmonic vibrational frequency in S_C^*. Nevertheless, on the basis of the observed temperature dependences we think that the contribution of electrostriction is small. The observed temperature behaviour would be hardly expected for pure electrostriction. It would be interesting to measure quantitatively the second harmonic frequency response in other liquid crystal phases, however unfortunately it is of the order of the experimental error.

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