# Nonlinear electromechanical transformations in ferroelectric liquid crystals

A.P.Fedoryako, E.V.Popova, L.A.Kutulya, A.V.Palamar, P.P.Shtyfanyuk, V.P.Semynozhenko

Institute for Scintillation Materials, STC "Institute for Single Crystals", National Academy of Sciences of Ukraine, 60 Lenin Ave., 61001 Kharkiv, Ukraine

#### Received February 22, 2005

A parametric transformation of acoustic signal in a ferroelectric liquid crystal (FLC) cell with uniform planar orientation has been obtained for the first time. Two types of mechanical vibration types were excited in the FLC sample, namely, strong pumping and weal signal ones. The spectrum of the sample electric response comprises the lines of combination frequencies. Dependence of the combination vibration frequencies on the pumping signal parameters has been studied.

Впервые получены параметрические преобразования акустического сигнала в ячейке сегнетоэлектрического жидкого кристалла (СЭЖК) с однородной планарной ориентацией. В образце СЭЖК возбуждались два типа механических колебаний: мощные накачки и слабые информационные. В спектре электрического отклика образца обнаружены линии комбинационных частот. Исследована зависимость амплитуды комбинационных частот колебаний от параметров сигнала накачки.

The theory of nonlinear dynamics in smectic liquid crystals (SLC) has been proposed by Brand and Pleiner [1]. According to those authors, the nonlinearity is due to the relationship between the translational fluctuations along the normal to the smectic layers and the rotational fluctuations of the director in the plan thereof, that is, a strain of the layer thickness is accompanied by a wavelike mode. Later, the same authors have considered the case of strong straining in SLC and in ferroelectric liquid crystals (FLC) [2]. In FLC, there is a helical structure, and its straining is referred to as a large-scale one. The large-scale straining contributes little to the straining energy. However, at large strains, the relation between the wave vector of the layer periodicity and that of the helical structure may result in a nonlinearity of the dynamic properties. Carlsson et el. [3, 4] have used another approach. They postulated that the interlayer spacing and inclination angle in smectic layers remain constant at any strains. In this case, the wavelike straining mode is accompanied by violations of the director field in the layer plan and arising flows that define the nonlinear dynamic properties of the liquid crystal.

It is just the nonlinear electromechanical response observable in some liquid crystals [5] that is among experimental evidences of the FLC dynamic nonlinearity. Later, in the course of investigation of electromechanical transformation regularities in various phases of the liquid crystal, an anharmonism was revealed in the dynamic response of a FLC in the SmC\* phase. The anharmonism causes multiple harmonics in the electromechanical response signal when a monochromatic alternating electric field is applied to the sample [6]. It is just the nonlinearity that is the most probable reason for the anharmonism. Consideration of literature data shows that the nonlinear dynamics in SLC and FLC was not studied systematically to date. The studies of nonlinear electromechanical transformation in FLC are of a

#### Mixture I

$C_6H_{17}$ $OC_6H_{13}$	Matrix
C4HBCHCHEOOC — COOCHECHC4HB	CD - 24.27 % mass

practical interest, because the materials showing nonlinear dynamic properties make it possible to transform parametrically an acoustic signal.

Three mixtures were used in the experiments. Each mixture consisted of an achiral smectic C (matrix) and a chiral dopant (CD).

Mixture I (achiral smectic I, CD I 24.75~% mass). The phase transition sequence and temperatures are as follows:

$$I \rightarrow 80.5 \rightarrow two$$
-phase region I,  
 $SmA^* \rightarrow 77.5 \rightarrow SmA^* \rightarrow 54 \rightarrow SmC^* \rightarrow Cr$ .

The main macroscopic parameters of the composition at  $\Delta T = 20$ °C, where  $\Delta T = T_{\rm SmA}^* \rightarrow {\rm SmC}^* - T_{m_eas}$ : spontaneous polarization  $-27.6~{\rm nC/cm^2}$ , inclination angle of molecules in smectic layer 21°, rotation viscosity 0.3 Poise.

Mixture II (achiral smectic II, CD II 14 % mass). The phase transition sequence and temperatures:

$$I \rightarrow 87 \rightarrow BP \rightarrow 86.4 \rightarrow N^* \rightarrow 74 \rightarrow SmC^* \rightarrow Cr.$$

The main macroscopic parameters of the composition at  $\Delta T=20\,^{\circ}\mathrm{C}$ : spontaneous polarization  $-50~\mathrm{nC/cm^2}$ , inclination angle of molecules in smectic layer  $28\,^{\circ}$ , rotation viscosity 0.51 Poise.

Mixture III (achiral smectic III, CD III 15.46 % mass, after mercury lamp irradiation for 1 h). The composition was in the photostationary state [7]. The phase transition sequence and temperatures:

$$I \rightarrow 106 \rightarrow N^* \rightarrow 66 \rightarrow SmA^* \rightarrow 59 \rightarrow SmC^* \rightarrow Cr.$$

The main macroscopic parameters of the composition at  $\Delta T = 20$ °C: spontaneous polarization 28 nC/cm<sup>2</sup>, inclination angle of molecules in smectic layer 29.8°, rotation viscosity 0.48 Poise.

The FLC was placed in a cell consisting of two plane-parallel glasses with transparent ITO electrodes. The plate area was at least 5 cm $^2$ . The cell thickness was set by spacers and amounted from 24 to 33  $\mu m$ .

#### Mixture II

C <sub>n</sub> H <sub>n2+1</sub> O O C <sub>m</sub> H <sub>2m+1</sub> m=6, n=8 - 50%  m=6, n=10 - 50%	Matrix
$C_aH_gO                                    $	CD -14 % mass

#### Mixture III

X X Y Y  H <sub>2 lb+1</sub> C <sub>ll</sub> ———————————————————————————————————	Matrix
OC7H17	$rac{ ext{CD} -}{15.46~\%} \  ext{mass}$

Planar boundary conditions were formed in the cell. To study the FLC nonlinear properties, a sinusoidal voltage was applied to the liquid crystal sample via an amplifier. The sound emitted was detected by a microphone, digitized, and processed using a PC. From the amplitude-frequency response of the FLC sample, the mechanical resonance frequency was determined that depends on the mobile plate mass and the viscoelastic properties of the FLC. The further studies were carried out at frequencies close to the resonance one. The setup used to obtain the parametric transformation is presented schematically in Fig. 1.

To determine the main physical parameters defining the FLC nonlinear properties, liquid crystals with various sequences of phase transitions have been studied. When examining a composition with the phase sequence I-N\*-SmC\*-Cr where the transition into the  $SmC^*$  phase is a first order one (Fig. 2, curve 1), the mechanical vibration amplitude (A) dependence on the electric field strength (E) was found to be slightly nonlinear. A low stability of mechanical vibration is typical of that composition, too. In liquid crystals with the phase sequence I-SmA-SmC\*-Cr (second order transition into the SmC\* phase), the nonlinearity is more pronounced (Fig. 2, curve 2). The highest nonlinearity, however, is observed in the mixture III (second order transition into the SmC\* phase). Within a wide tem-

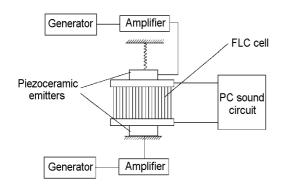


Fig. 1. Experimental setup.

perature range ( $30^{\circ}$  to  $4^{\circ}$  from the phase transition), sections where a lowering electric field strength resulted in an increasing mechanical transformation amplitude were observed in the field dependences of electromechanical response (Fig. 2, curve 3). This effect remains still unexplained.

According to the preliminary data, the highest electromechanical transformation efficiency is observed in FLC with large inclination angles of molecules in the smectic layer.

A parametrical transformation of sound has been obtained for the first time (mixture III). Along with the pumping and signal lines, combination frequencies are observed in the spectrum. Those correspond to the difference and sum of the initial frequencies (Fig. 3). Similar spectra can be also obtained using a trivial modulation of a high-frequency signal with a low-frequency one. For the parametrical transformation, the amplitudes of combination frequencies are not equal to each other; moreover, the amplitudes (A) of combination frequencies depend on the pumping vibra-

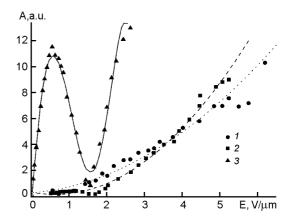
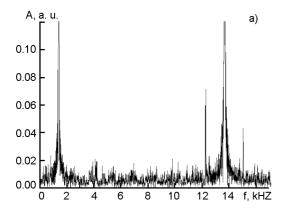


Fig. 2. Dependence of mechanical vibration amplitude (A) on the electric field strength (E) for various compositions: Mixture I,  $\Delta T = 68.6^{\circ}\text{C}$  (1); Mixture II,  $\Delta T = 34^{\circ}\text{C}$  (2); Mixture III,  $\Delta T = 31^{\circ}\text{C}$  (3).

tion parameters (amplitude and frequency) [8]. These dependences were studied. By varying the pumping vibration parameters, an output signal can be obtained that comprises in its spectrum combination frequencies corresponding to the difference of the initial ones (Fig. 3b), to the sum thereof or both the combination frequencies simultaneously (Fig. 3a). The differential and sum frequency lines are different in amplitudes, that is typical of the parametrical transformation but not of the modulation. A dependence of the amplitude (A) of combination frequencies on the pumping vibration intensity is also observed (Fig. 4), that confirms also the presence of the parametrical transformation.

Thus, the work has been shown that FLC where the  $SmA \rightarrow SmC$  phase transition is of the second order exhibit a more pro-



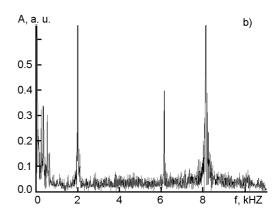


Fig. 3. Output signal spectra comprising the combination frequencies corresponding to the difference and sum of the initial ones (a) and only that corresponding to the difference (b). Mixture III,  $\Delta T = 39^{\circ}$ C.

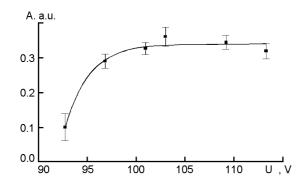


Fig. 4. Dependence of the differential frequency amplitude (A) on the pumping signal exciter voltage  $(U_d)$ . Mixture III,  $\Delta T = 39^{\circ}$ C.

nounced nonlinear electromechanical response as compared to the substances with first order SmC phase transition. Using a FLC with nonlinear electromechanical response as the active substance in an acoustic mixer, the parametrical transformation of an acoustic signal has been obtained for the first time.

Authors are thankful to Dr.V.V.Vashchenko who has synthesized the materials used in this work.

#### References

- 1. H.Brand, H.Pleiner, J. Phys., 41, 553 (1980).
- 2. H.Pleiner, H.Brand, Physica, 265, 62 (1999).
- T.Carlsson, N.Clark, Z.Zou, *Liquid Crystals*, 15, 461 (1994).
- T.Carlsson, F.M.Leslie, N.A.Clark, *Phys. Rev.*, 51, 4509 (1995).
- A.Jakli, L.Bata, Liquid Crystals, 7, 105 (1990).
- E.V.Popova, A.P.Fedoryako, L.A.Kutulya, V.P.Semynozhenko, Kristallografia, 50, 180 (2005).
- L.Kutulya, A.Krivoshey, E.Popova et al., Mol. Cryst. Liq. Cryst., 426, 99 (2005).
- 8. W.H.De Jeu, Physical Properties of Liquid Crystalline Materials, Gordon and Breach Science Publishers, New York-London-Paris (1980).
- 9. Yu.G.Kuleshov, Nonlinear and Parametrical Radio Circuits, Vyscha Shkola, Kiev (1970) [in Russian].

### Нелінійні електромеханічні перетворення у сегнетоелектричних рідких кристалах

## О.П.Федоряко, К.В.Попова, Л.А.Кутуля, О.В.Паламар, П.П.Штифанюк, В.П.Семиноженко

Вперше отримано параметричні перетворення акустичного сигналу у комірці сегнетоелектричного рідкого кристала (СЕРК), що має однорідну планарну орієнтацію. У зразку СЕРК збуджувалися два типи механічних коливань: потужні накачки, та слабкі інформаційні. У спектрі електрооптичного відгуку знайдено лінії комбінаційних частот. Досліджено залежність амплітуди комбінаційних частот коливань від параметрів сигналу накачки.