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LOW FREQUENCY DIELECTRIC RESPONSE OF CHIRAL SMECTICS C*

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Abstract The low frequency dielectric response of SmC* is analysed within the framework of the electrodynamic continuum theory. The electric field induced distortion of the structure is calculated. The macroscopic contributions in the dielectric susceptibility are determined in fields parallel as well as normal to the helix. These contributions originate in the presence of the spontaneous polarization, the helical structure and the electromechanical effect.

1. INTRODUCTION

Chiral smectic C* (SmC*) liquid crystals have got into the centre of interest in the last some years.[1-3] They exhibit several electrooptic effects which open wide prospects for their application as electronic displays.[4-6] Due to the helical structure and the presence of a non-zero spontaneous polarization their dielectric response, i.e. the behaviour in infinitesimal AC electric fields, shows some specific features too. In SmC* substances a dielectric relaxation - the so-called Goldstone mode - was found at low frequencies, which was assigned to the relaxation of the helical structure.[7-10]

An interpretation of the phenomenon has been given on the basis of a Landau expansion of the free energy.[11,12] Rigorous calculations have shown that near the SmA-SmC* phase transition the situation becomes more complicated since the tilt angle may also be influenced by the electric field.[13,14] However in the great majority of experiments the electrooptical properties of SmC* are investigated far from the SmA-SmC* phase transition. In this range the tilt angle usually can be regarded as a constant depending on the temperature only. Then the hydrodynamic[15-17] or other continuum theories[5,18,19] are more convenient for the interpretation of electrooptical phenomena than the Landau expansion.[11] Unlike the high frequency dielectric relaxation modes, which provide information on molecular dynamics, the Goldstone mode is connected to a variation of a macroscopic texture. Thus an obvious claim may arise to describe it by means of the same continuum theories what are used for the interpretation of other macroscopic phenomena. Such an approach would allow a comparison of data collected by dielectric and electrooptic methods.

Inspired by the detection of an electromechanical effect[20,21] an electrodynamic continuum theory[22-24] has been derived recently which includes some novel irreversible cross-couplings as e.g. the electromechanical one. In the present paper this theory is applied to analyse the low frequency dielectric response of SmC* liquid crystals.

Section 2 is a summary of the basic assumptions of our treatment and a list of the basic equations of the continuum theory. In Section 3 we calculate the distortion of the helical texture produced by an infinitesimal AC electric field and determine the bulk polarization. In Section 4 our formulae are discussed in various geometries, i.e. when the field is either perpendicular or parallel to the helical axis, including the case when a large DC bias field is applied normal to the helix. Section 5 is a brief summary of our results.

2. BASIC ASSUMPTIONS

The orientation of the SmC* liquid crystal is characterized by two vector fields, the layer normal $\underline{n}(\underline{r})$ and the C-director $\underline{c}(\underline{r})$. The material flow is described by the velocity field $\underline{v}(\underline{r})$. We are choosing a Cartesian frame with its z-axis along the layer normal, thus[23,24]

$$\underline{n}=(0,0,1) \quad ; \quad \underline{c}=(c_x,c_y,0) \quad ; \quad \underline{v}=(v_x,v_y,0).$$

In dielectric experiments the SmC* liquid crystal is usually placed in between parallel plane electrodes held at different potential. In a homogeneous material it would yield a homogeneous electric field normal to the electrodes which we shall call applied field $\underline{E}_a=(E_{ax},E_{ay},E_{az})$. However if the sample is not homogeneous - as we shall see it later - the actual electric field $\underline{E}=(E_x,E_y,E_z)$ which enters our equations need not coincide with the applied one.

Usually the boundaries of a cell influence the orientation of an SmC* liquid crystal.[3-6,18] However it may be a reasonable assumption to neglect this influence in thick samples. Therefore throughout this paper we are considering the behaviour of an infinite, oriented SmC* sample, or by other words a sample in which no constraints are prescribed on either orientation or flow at the boundaries.

In the absence of an applied field the infinite SmC* sample possesses an undistorted helical structure without any flow which we shall refer to as the ground state $\underline{c}_0(\underline{r})$. An infinitesimal applied electric field may induce both a distortion of the texture and a flow, however it can be supposed in this aproximation that these distortions are homogeneous in the x-y plane, i.e. any physical quantity may depend on the z-coordinate only. Thus $\underline{\nabla}=(0,0,\nabla_z)$ and the material and partial time derivatives must coincide.

Neglecting terms quadratic in the infinitesimal fields with these assumptions the main equations of the electrodynamic continuum theory[23,24] read in our geometry:

Maxwell's Equations

$$\mathrm{curl}\,\underline{E}=0 \qquad \nabla_z E_x=0 \;;\; \nabla_x E_y=0 \tag{2.1}$$

$$\mathrm{div}\,\underline{D}=0 \qquad \nabla_z(\varepsilon_0 E_z+P_z)=0 \tag{2.2}$$

Equation of Motion of the C-director

$$\dot{\underline{c}} = (\underline{n}\times\underline{c})\left\{\frac{b_1-b_3}{2}\left[c_y(\nabla_z v_x)-c_x(\nabla_z v_y)\right]-\sigma_1 c_x E_x-\sigma_1 c_y E_y-\sigma_2 E_z\right.$$
$$\left.-\frac{1}{\eta}\left[K_2 c_y(\nabla_z\nabla_z c_x)-K_2 c_x(\nabla_z\nabla_z c_y)+P_s(c_x E_x+c_y E_y)\right]\right\} \tag{2.3}$$

Equations of Motion

$$\varrho\dot{v}_x = -\nabla_z\left\{\frac{b_1-b_3}{2}\left[K_2 c_x c_y(\nabla_z\nabla_z c_y)-K_2 c_y^2(\nabla_z\nabla_z c_x)-P_s c_y(c_x E_x+c_y E_y)\right]\right.$$
$$-\left[(\mu_8+\mu_{11})c_x^2+\mu_{12}\right](\nabla_z v_x)-(\mu_8+\mu_{11})c_x c_y(\nabla_z v_y)$$
$$\left.+(\gamma_4+\gamma_7)c_x c_y\dot{E}_x-(\gamma_4 c_x^2-\gamma_7 c_y^2)\dot{E}_y+\gamma_8 c_y\dot{E}_z\right\} \tag{2.4}$$

$$\varrho\dot{v}_y = -\nabla_z\left\{\frac{b_1-b_3}{2}\left[K_2 c_x c_y(\nabla_z\nabla_z c_x)-K_2 c_x^2(\nabla_z\nabla_z c_y)+P_s c_x(c_x E_x+c_y E_y)\right]\right.$$
$$-(\mu_8+\mu_{11})c_x c_y(\nabla_z v_x)-\left[(\mu_8+\mu_{11})c_y^2+\mu_{12}\right](\nabla_z v_y)$$
$$\left.+(\gamma_4 c_y^2-\gamma_7 c_x^2)\dot{E}_x-(\gamma_4+\gamma_7)c_x c_y\dot{E}_y-\gamma_8 c_x\dot{E}_z\right\} \tag{2.5}$$

Polarization

$$P_x = (\chi_1+\chi_2 c_x^2)E_x+\chi_2 c_x c_y E_y+\chi_4 c_x E_z-P_s c_y-e_4(\nabla_z c_x)$$
$$-(\kappa_1+\kappa_2 c_x^2)\dot{E}_x-\kappa_2 c_x c_y\dot{E}_y-\kappa_4 c_x\dot{E}_z$$
$$+(\gamma_4+\gamma_7)c_x c_y(\nabla_z v_x)+(\gamma_4 c_y^2-\gamma_7 c_x^2)(\nabla_z v_y) \tag{2.6}$$

$$P_y = \chi_2 c_x c_y E_x+(\chi_1+\chi_2 c_y^2)E_y+\chi_4 c_y E_z+P_s c_x-e_4(\nabla_z c_y)$$
$$-\kappa_2 c_x c_y\dot{E}_x-(\kappa_1+\kappa_2 c_y^2)\dot{E}_y-\kappa_4 c_y\dot{E}_z$$
$$-(\gamma_4 c_x^2-\gamma_7 c_y^2)(\nabla_z v_x)-(\gamma_4+\gamma_7)c_x c_y(\nabla_z v_y) \tag{2.7}$$

$$P_z = \chi_4 c_x E_x + \chi_4 c_y E_y + (\chi_1+\chi_3)E_z - \kappa_4 c_x \dot{E}_x - \kappa_4 c_y \dot{E}_y$$
$$- (\kappa_1+\kappa_3)\dot{E}_z + \gamma_8 c_y (\nabla_z v_x) - \gamma_8 c_x (\nabla_z v_y) \qquad (2.8)$$

Here b_1, b_3 describe a reversible coupling between flow and orientation, δ_1, δ_2 an irreversible one between electric field and orientation. η is the orientational viscosity, K_2 the twist elastic constant, P_s the spontaneous polarization. μ_8, μ_{11} and μ_{12} are viscosities, γ_4, γ_7 and γ_8 electromechanical coupling coefficients, $\chi_1,\ldots,\chi_4$ dielectric susceptibilities, e_4 a flexoelectric coefficient and $\kappa_1,\ldots,\kappa_4$ are connected with dielectric loss ($\underline{\underline{\kappa}}^{55}$ in Ref.24).

In the ground state the net bulk polarization - i.e. the polarization averaged over the pitch - is zero indicating that SmC* liquid crystals are improper ferroelectrics.[1-3]

3. CALCULATION OF THE DISTORTION

Let us now apply an infinitesimal harmonic AC electric field $\underline{E}_a$ of arbitrary direction to the sample. The induced flow and the distortion of the helix should also be infinitesimal and harmonic.

Since the C-director of the perturbed ground state can be written into the form

$$\underline{c} = \underline{c}_0 + \alpha(\underline{n}\times\underline{c}_0) \quad ; \quad \underline{c}_0=(\cos qz, \sin qz, 0) \qquad (3.1)$$

where q is the helical wave vector (q_2 in Ref.24) and the derivation with respect to time is equivalent to a multiplication by $i\omega$, Eq.(2.2) can be rewritten as

$$E_z = A - B(E_x \cos qz + E_y \sin qz) + C\left[\cos qz(\nabla_z v_y) - \sin qz(\nabla_z v_x)\right] \qquad (3.2)$$

where A is an integration constant to be determined later and

$$B = \frac{\chi_4 - i\omega\kappa_4}{\varepsilon_0+\chi_1+\chi_3-i\omega(\kappa_1+\kappa_3)} \quad ; \quad C = \frac{\gamma_8}{\varepsilon_0+\chi_1+\chi_3-i\omega(\kappa_1+\kappa_3)} \qquad (3.3)$$

Then combining Eqs. (2.3)-(2.5), (3.1) and (3.3) one gets

$$
\begin{aligned}
i\omega\alpha = {} & \frac{K_2}{\eta}(\nabla_z\nabla_z\alpha)+\left(\frac{h_1-h_3}{2}+\delta_2 C\right)\left[\sin qz(\nabla_z v_x)-\cos qz(\nabla_z v_y)\right] \\
& -\left(\frac{P_s}{\eta}+\delta_1-\delta_2 B\right)(E_x\cos qz+E_y\sin qz)-\delta_2 A
\end{aligned} \tag{3.4}
$$

$$
\begin{aligned}
i\omega\rho v_x = {} & \frac{\mu^*}{2}(\nabla_z\nabla_z v_x)-\frac{h_1-h_3}{2}K_2\nabla_z\left[\sin qz(\nabla_z\alpha)\right]-i\omega\gamma_8 qA\cos qz \\
& +\frac{1}{2}(\mu_8+\mu_{11}-i\omega\gamma_8 C)\nabla_z\left[\cos 2qz(\nabla_z v_x)+\sin 2qz(\nabla_z v_y)\right] \\
& +\left(\frac{h_1-h_3}{2}-i\omega\gamma^*\right)q(E_x\cos 2qz+E_y\sin 2qz)
\end{aligned} \tag{3.5}
$$

$$
\begin{aligned}
i\omega\rho v_y = {} & \frac{\mu^*}{2}(\nabla_z\nabla_z v_y)+\left(\frac{h_1-h_3}{2}K_2\nabla_z\right)\left[\cos qz(\nabla_z\alpha)\right]-i\omega\gamma_8 qA\sin qz \\
& +\frac{1}{2}(\mu_8+\mu_{11}-i\omega\gamma_8 C)\nabla_z\left[\sin 2qz(\nabla_z v_x)-\cos 2qz(\nabla_z v_y)\right] \\
& +\left(\frac{h_1-h_3}{2}-i\omega\gamma^*\right)q(E_x\sin 2qz-E_y\cos 2qz)
\end{aligned} \tag{3.6}
$$

where we introduced

$$\mu^*=\mu_8+\mu_{11}+2\mu_{12}+i\omega\gamma_8 C \quad \text{and} \quad \gamma^*=\gamma_4+\gamma_7-\gamma_8 B.$$

The solution of these coupled differential equations can be written into the form of a Fourier series. After some algebra one can get the nonvanishing Fourier coefficients

$$
\begin{aligned}
\alpha &= X_0 A - X_1(E_x\cos qz+E_y\sin qz) \\
v_x &= U_1 A\cos qz + U_2(E_x\cos 2qz+E_y\sin 2qz) \\
v_y &= U_1 A\sin qz + U_2(E_x\sin 2qz-E_y\cos 2qz)
\end{aligned} \tag{3.7}
$$

where we used the notation

$$X_0 = \frac{\frac{h_1-h_3}{2}i\omega\gamma_8 q^2-\delta_2\mu_{12}q^2-i\omega\rho\delta_2}{i\omega\rho+\mu_{12}q^2+i\omega\gamma_8 Cq^2}\;\frac{1}{i\omega} \tag{3.8}$$

$$U_1 = \frac{-\,i\omega\gamma_8 q}{i\omega\varrho + \mu_{12}q^2 + i\omega\gamma_8 C q^2} \tag{3.9}$$

$$X_1 = \frac{\left(\frac{P_s}{\eta} + \delta_1 - \delta_2 B\right)(i\omega\varrho + 2\mu^* q^2) + (b_1 - b_3 + 2\delta_2 C)\left(\frac{b_1 - b_3}{2}P_s - i\omega\gamma^*\right)q^2}{\left[i\omega\varrho + 2\mu^* q^2 + \frac{(b_1 - b_3)K_2 q^2\left(\frac{b_1 - b_3}{2} + \delta_2 C\right)}{i\omega + \frac{K_2}{\eta}q^2}\right]\left(i\omega + \frac{K_2}{\eta}q^2\right)} \tag{3.10}$$

$$U_2 = \frac{\frac{b_1 - b_3}{2}\,\frac{i\omega P_s - K_2 q^2(\delta_1 - \delta_2 B)}{i\omega + \frac{K_2}{\eta}q^2}\,q - i\omega\gamma^* q}{i\omega\varrho + 2\mu^* q^2 + \frac{(b_1 - b_3)K_2 q^2\left(\frac{b_1 - b_3}{2} + \delta_2 C\right)}{i\omega + \frac{K_2}{\eta}q^2}} \tag{3.11}$$

Combining Eqs.(3.2) and (3.7) one gets the expression for E_z

$$E_z = (1+qCU_1)A - (B-2qCU_2)(E_x\cos qz + E_y\sin qz) \tag{3.12}$$

showing that unlike E_x or E_y, E_z varies periodically along the helical axis. However this modulation integrates to zero over distances equal to the pitch thus a relation can be obtained between the electric field in the sample and the applied field.

$$E_{ax} = \bar{E}_x = E_x \quad ; \quad E_{ay} = \bar{E}_y = E_y \quad ; \quad E_{az} = \bar{E}_z = (1+qCU_1)A \tag{3.13}$$

Here and in the following a dash above a variable indicates averaging over the pitch.

Finally the net bulk polarization is calculated from Eqs.(2.6)-(2.8) and (3.7) yielding

$$\overline{P}_x = \left[(\chi_1 + \frac{\chi_2 - B\chi_4}{2}) - i\omega(\kappa_1 + \frac{\kappa_2 - B\kappa_4}{2})\right]\overline{E}_x - \frac{P_s}{2}X_1\overline{E}_x$$

$$- (\gamma_4 + \gamma_7 - \chi_4 C + i\omega\kappa_4 C)qU_2\overline{E}_x = \chi_\perp^* E_{ax} \qquad (3.14)$$

$$\overline{P}_y = \left[(\chi_1 + \frac{\chi_2 - B\chi_4}{2}) - i\omega(\kappa_1 + \frac{\kappa_2 - B\kappa_4}{2})\right]\overline{E}_y - \frac{P_s}{2}X_1\overline{E}_y$$

$$- (\gamma_4 + \gamma_7 - \chi_4 C + i\omega\kappa_4 C)qU_2\overline{E}_y = \chi_\perp^* E_{ay} \qquad (3.15)$$

$$\overline{P}_z = \left[(\chi_1 + \chi_3) - i\omega(\kappa_1 + \kappa_3)\right]\overline{E}_z - \frac{\gamma_8 qU_1}{1 + qCU_1}\overline{E}_z = \chi_\parallel^* E_{az} \qquad (3.16)$$

4. INTERPRETATION

4.1. Applied Field Perpendicular to the Helical Axis

An applied AC electric field perpendicular to the helical axis ($E_{az}=0$) induces an oscillating deformation and an electric field along the helical axis which both have a spatial periodicity equal to the pitch. The induced flow is also oscillating with the same frequency ω, but has a spatial periodicity equal to the half pitch (cf. Eqs.(3.7) and (3.12)).

As can be seen in Eqs.(3.14) and (3.15) the bulk polarization is proportional to the applied field and all directions normal to the helical axis are equivalent, i.e. an infinite SmC* behaves macroscopically as a uniaxial material. The effective complex dielectric susceptibility $\chi_\perp^*$ is composed of three contributions.

i. The elements of the local biaxial dielectric susceptibility $\underline{\underline{\chi}}$ and the loss $\underline{\underline{\kappa}}$ tensors are combined to a uniaxial contribution. They describe relaxations due to fast molecular motions which usually take place at higher frequencies. Their influence on the low frequency behaviour usually can be neglected, i.e. one can regard $\underline{\underline{\kappa}}$ as zero and $\underline{\underline{\chi}}$ as constant.

ii. The distortion of the helical texture leads to a partial ordering of the spontaneous polarization of the layers. This corresponds to the Goldstone-mode.[7-9] The factor $1/(i\omega+K_2q^2/\eta)$ in X_1 (cf.Eq.(3.10)) would alone describe a relaxation of Debye-type with a critical frequency $\omega_0=\sqrt{K_2q^2/\eta}$. However this frequency dependence is altered by other factors which are mainly originating in the field induced flow.

iii. The third contribution is the flow induced polarization which is the result of an irreversible cross-coupling, the inverse electromechanical effect.[24,25]

The low frequency dielectric response of chiral smectics C* is characterized by the last two macroscopic contributions whose presence is due to the chirality and the local biaxial symmetry of this phase. Unfortunately the orders of magnitude of several material parameters are still unknown therefore it is not yet predictable theoretically which term dominates the frequency behaviour. Experimental data[7-9] suggest that this low frequency dielectric relaxation is nearly Debye-type however deviations have also been observed.[25]

4.2. Large DC Bias Field Normal to the Helical Axis

A large DC bias electric field $\underline{E}_b$ destroys the helical structure completely.[1-3,26] In this homogeneous unwound state the spontaneous polarization points toward the direction of the field. If the bias field is along the x-axis the unwound state can be characterised by

$$\underline{E}_b=(E_b,0,0) \ ; \ \underline{c}_b=(0,-\mathrm{sgn}(P_sE_b),0) \ ; \ \underline{v}=0 \ ; \ E_z=E_y=0 \qquad (4.1)$$

with a net bulk polarization of

$$\overline{P}_x=P_b=X_1E_b+P_s\mathrm{sgn}(P_sE_b) \ ; \ \overline{P}_y=0 \ ; \ \overline{P}_z=0 \qquad (4.2)$$

A superposition of an infinitesimal applied AC electric field normal to the helical axis $\underline{E}_a=(E_{ax},E_{ay},0)$ can not influence the structure of this unwound state but contributes to the bulk polarization.

$$\overline{P}_x=P_b+(\chi_1-i\omega\kappa_1)E_{ax}\ ;\ \overline{P}_y=\left[(\chi_1+\chi_2)-i\omega(\kappa_1+\kappa_2)\right]E_{ay}\ ;\ \overline{P}_z=0 \quad (4.3)$$

As it can be seen there is no macroscopic contribution to the dielectric susceptibility due to the bias field which is in accordance with the experiments.[10] This geometry would allow measurement of two elements of the local biaxial susceptibility tensor $\underline{\underline{\chi}}$ however for technical reasons usually the applied field is parallel to the bias one. It means that only one eigenvalue can be obtained, the one belonging to the eigenvector pointing in the direction of the spontaneous polarization, i.e. in the direction of the bias field.

It is worth noticing that no field is induced along the helical axis ($E_z=0$) since the texture is homogeneous. E_z can appear only in biaxial systems which possess inhomogeneous textures.

4.3. Applied Field Along the Helical Axis

An applied AC field along the helical axis ($E_x=E_y=0$) results in a homogeneous oscillating rotation of the whole undistorted helix and in an oscillating flow with a spatial periodicity equal to the pitch (cf. Eqs. (3.7) and (3.13)). As can be seen in Eq. (3.16) the effective complex dielectric susceptibility $\chi^*_{\parallel}$ is composed of two terms. The first one corresponds to fast molecular motions and contains elements of the $\underline{\underline{\chi}}$ and $\underline{\underline{\kappa}}$ tensors. The second one is a macroscopic contribution which is owing to the presence of the irreversible electromechanical coupling thus it mainly contributes to the dielectric loss.

It should be emphasized however that according to Eq. (3.8) the amplitude of the oscillating rotation of the helix diverges at $\omega=0$. This indicates that our approach used in Section 3 was not really adequte for this geometry at low frequencies. Therefore we present here an another approach which is valid for a small DC applied field.

If a DC field is applied along the helical axis the C-director can be written as

$$\underline{c}=(\cos(qz+\Omega t),\sin(qz+\Omega t),0) \tag{4.4}$$

Then Eqs.(2.3)-(2.5) yield immediately

$$\Omega = -\delta_2 E_{az} \quad ; \quad \underline{v}=0 \tag{4.5}$$

which predicts a continuous rotation of the whole undistorted helix around the helical axis with an angular velocity Ω proportional to the applied field. It is a result of a direct irreversible coupling between electric field and the C-director.[23,24] It is analogous to the Lehmann-rotation caused by thermal gradients.[27]

In case of a low frequency applied field the probable distortion of the structure is an oscillating rotation of the helix with a finite amplitude accompanied by a flow, however we could not provide an analytical solution for this intermediate regime.

5. SUMMARY

Applying the electrodynamic continuum theory of chiral smectics C* we were able to calculate the electric field induced deformation and flow in various geometries. These distortions result in macroscopic contributions to the dielectric susceptibility which determine the low frequency dielectric response of the substance.

The recently described irreversible cross-couplings,[24] the electromechanical effect and its inverse play a role independently of the direction of the applied field.

In electric fields normal to the helical axis the partial orientation of the spontaneous polarization contributes to the susceptibility. This relaxation, the Goldstone-mode, is not exactly Debye-type. This relaxation is absent if the helix is unwound by e.g. a DC bias field.

For a DC field along the helical axis a new phenomenon, a continuous rotation of the helix is predicted.

It has been proved that owing to biaxiality an electric field normal to the helix induces a field along the helical axis supposing that the texture is inhomogeneous, i.e. the C-director varies in space.

We should like to emphasize that our calculations hold for infinite samples. The behaviour of thin sandwich cells may be considerably different due to the influence of the boundaries, but this more complicated problem unfortunately can not be treated exactly within the framework of our theory.

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