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Dynamic Electroacoustic Effects in Chiral Smectic Liquid Crystals

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We show that a recently observed electroacoustic effect in chiral smectic C^* liquid crystals can be explained in terms of a reversible dynamic cross-coupling between the velocity field and the polarization, which exists for chiral smectic liquid crystals, but not for other liquid crystalline phases such as nematics, cholesterics and non-chiral smectics. Further experiments to test our interpretation are suggested.

Keywords: electroacoustic effect, chiral smectics, macroscopic dynamics

INTRODUCTION

In chiral smectic C^* liquid crystals there is a polarization in each smectic layer, which rotates in a helical fashion as one moves along the layer normal[1] thus giving rise to helielectricity[1-2] and a vanishing macroscopic polarization in samples with a thickness large compared to the helical pitch. The presence of an in-plane polarization, which allows for a linear electrooptic response,[3] has led to a strong interest in chiral smectics for device applications.

However, in addition to the electrooptic aspects, chiral smectic C^* also support the existence of interesting electromechanical effects due to the lack of mirror symmetry in this phase. The first experiment in this area[4] showed that shear flow can induce a macroscopic polarization. More recently there have been studies of the mechanical response to AC electric fields[5,6] both, in low molecular weight liquid crystals and in liquid crystalline polymers.

From the theoretical point of view, we have given a macroscopic dynamic description,[7-9] which has been formulated globally[7-8] (valid on length scales larger than the helical pitch) and locally[9] (applicable on length scales smaller than the pitch).

Very recently[10] the observation of a rather exciting strong electroacoustic effect in the kHz range has been reported in this journal for thin samples ($d = 4$ μm), in which the chiral smectic C^* phase is arranged in the so-called bookshelf geometry (the smectic layers are aligned perpendicularly to the top and bottom electrodes). The application of rather moderate rectangular AC voltages (about 4 V) led to the emission of a sound wave from the sample.[10] An additionally applied DC field (of

order 0.8 V) was found to suppress the electroacoustic effect. Furthermore a pressure applied to the top glass plate led to a damped response. We note that a similar electroacoustic effect has been mentioned previously.[11,12]

In this note we will suggest an interpretation of this electroacoustic effect on the basis of a reversible dynamic cross-coupling between the velocity field and the polarization applying the macroscopic dynamic approach we have presented[7-9] before.

REVERSIBLE DYNAMIC EFFECTS IN CHIRAL SMECTIC LIQUID CRYSTALS

In a macroscopic description one keeps in addition to the classical hydrodynamic variables, namely the conserved quantities (density, density of linear momentum, energy density) and the variables associated with spontaneously broken continuous symmetries[13] (e.g. in all smectic liquid crystals the layer displacement parallel to the layer normal) the macroscopic quantities, which relax sufficiently slowly in the long wavelength limit. In chiral smectic C^* the relevant variable is the macroscopic polarization[7] $\mathbf{P}_0$, whose variations $\delta\mathbf{P}$ are rigidly coupled to the helix displacement $\mathbf{U}^c$ via

$$(\hat{\mathbf{P}}_0 \times \delta\mathbf{P})_z = q_0 U_z^c \tag{1}$$

where we have taken z to be parallel to the helical axis, $\hat{\mathbf{P}}_0 = \mathbf{P}_0/|\mathbf{P}_0|$, and where q_0 is the helical pitch.

Here we summarize briefly the parts of the macroscopic equations, which we will need in the next section for the interpretation of the experimental results.[10] In addition to a number of static and dissipative dynamic cross-coupling terms, we find,[7-9] among others, the following reversible dynamic electromechanical contributions to the dynamic equations

$$\dot{U}_i^c = \hat{p}_i \hat{p}_j v_j + \ldots \qquad \text{and} \qquad \rho_0 \dot{v}_i = h_i + \ldots \tag{2}$$

where the ellipses stand for all the other terms not given here explicitly and where $\hat{p}_i$ denotes the unit vector along the helical axis. In an infinite geometry the elastic force due to helix displacements, h_z, is given by dilation or bending of the helix, while for thin samples, where the boundaries modulate the pure helical structure (cf. below), also homogeneous displacements lead to a restoring force. In an external homogeneous electric field, E_0, transverse to the helix, the ferroelectric interaction with the polarization cancels, when averaged over a perfect helical structure, but is finite for a (slightly) distorted one. With Equation 1 this interaction is then proportional to U_z^c and adds to the force h_z, which reads

$$h_z = \gamma U_z^c - B_h \nabla_z^2 U_z^c + M P_0 E_0 q_0 \tag{3}$$

where B_h is the elastic modulus of the helix structure, γ is the effective elastic modulus due to the helix distortions in the field-free case and of the order of B_h/d^2

and M, which reflects the non-zero result of averaging over the imperfect helix, is also field dependent.

From Equation 2 we read off immediately that a velocity in the direction of the helical axis leads to a time-dependent helix displacement and from Equation 3 we see that an electric field applied in the planes perpendicular to the helical axis will lead to temporal variations in the velocity component parallel to the helical axis.

When going from the global to the local description valid on length scales smaller than the pitch, one must use instead of the helix displacement U_z^c as a macroscopic variable the phase variations $\delta\phi$ related to the rotation of the in-plane director (and thus also of the in-plane polarization) as one moves along the helical axis. The structure of Equations 2 and 3 is preserved by this procedure although one then has a biaxial description reflecting the fact that chiral smectic C^* phases are globally uniaxial and locally biaxial.

INTERPRETATION OF THE EXPERIMENTAL RESULTS

The experiments described in Reference 10 have been performed for samples in the bookshelf geometry for which the layers are oriented perpendicularly to the electrodes. As has been shown by Handschy et al,[14] however, this does not imply that one has a spatially homogeneous configuration for the director and the helix. Instead one has in cells with a thickness comparable to, or smaller than, the pitch a spontaneous splay in both the director and the polarization.[14] This implies that there are spatial variations with a characteristic length-scale of the cell thickness d (in the planes of the smectic layers) and the pitch $p_0 = 2\pi/q_0$ (in the direction parallel to the helical axis). Correspondingly one has also a characteristic band of wavevectors around $k_\perp = 2\pi/d$ and $k_\parallel = q_0$, respectively. Clearly the amplitude of this splay will be largest in thin cells and will become less important in cells with a thickness large compared to the pitch. While the authors of Reference 10 do not give a value for the pitch in the smectic C^* phase of their compound, it appears—judging from most of the compounds showing a smectic C^* phase—that for the experiments described the sample thickness is of the same order as the pitch. This implies that spatial inhomogeneities induced by spontaneous splay, and perhaps also due to surface interactions,[14] can play a significant role in the strong electroacoustic effect observed.

From this consideration it emerges that all macroscopic variables assuming a nonvanishing value will show spatial inhomogeneities with comparable wavevectors $k_\parallel$ and $k_\perp$ parallel and perpendicular to the helical axis even without an external electric field.

First we discuss the case of a pure AC electric field. In this situation all deformations of the helix are arising from its ground state, where one can define the macroscopic variables U_z^c and $\delta\phi$, respectively, (compare the last section) to characterize the deformations of the helix. From Equations 2 and 3 we can read that the externally applied AC field will give rise to a temporally varying U_z^c and v_z, the velocity component in the direction of the helical axis. Due to the spontaneous splay these quantities will show spatial inhomogeneities. To satisfy all macroscopic

equations, in particular the continuity equation (which reads $\nabla_\perp v_\perp + \nabla_z v_z = 0$ for the coordinate system used in the incompressible case, which is of interest here), necessarily the velocity field has also components transverse to the helical axis. Since splay gives rise to three-dimensional deformations in a chiral smectic phase, all macroscopic variables, in particular U_z^c, v_z and $v_\perp$ are functions of $k_\parallel$ and $k_\perp$. However, $v_\perp$ gives rise to vibrations of the top glass plate with the frequency of the applied electric AC field. And this motion in turn is converted to a sound wave at the electrode air interface. This picture agrees with the experimental observation that a pressure applied to the upper glass plate leads to a damping of the electroacoustic effect.

One thus obtains a system of linear coupled inhomogeneous equations for v_z, v_x and U_z^c, where the inhomogeneity is provided by the externally applied AC field via the reversible cross-coupling. Solving these equations for the variables involved, one observes that the amplitudes of these quantities contain the dispersion relation of second sound in their denominator, a result which is rather familiar from the hydrodynamics of condensed matter systems.[13] When applying this explanation to the actual experiments, one must also take into account possible mechanical resonance effects of the set-up. This implies that a large effect can be expected close to resonances of the whole system. In the present case this resonance should be rather broad because of the spatial inhomogeneities and thus of a broad band of possible wavevectors, as is indeed observed experimentally.[10] The observation of the effect is further simplified in the experiment in question by the fact that rectangular pulses in the external electric field were applied which contain many different frequencies by construction.

An external DC field leads to a partial unwinding of the helix, or to be more precise, to the formation of domains of favorable orientation of the polarization with domain walls in between. In this case one can no longer define the macroscopic variables U_z^c and $\delta\phi$ and thus Equations 2 and 3 are no longer applicable. This picture is corroborated by the experimental observation[10] that a sufficiently large DC bias field applied in addition to the AC electric field quenches the strong electroacoustic effect. From an estimate of typical 'unwinding' fields for chiral smectics one sees that the DC field applied in the experiments is large enough to lead to strong deformations of the helical structure.

We would like to stress that no density changes are necessary for the mechanism suggested here. As for the key difference to the classical Goldstone mode,[13] we find the crucial importance of strong spatial inhomgeneities as the explanation of the electroacoustic effect observed.

Discarding possible resonance effects of the set-up, there is also an additional qualitative consistency check for the picture given concerning the frequencies for which the effect is observed. To see this we exploit our knowledge about second sound in smectics. But we apply it here to the helix and not to the layers. The large intrinsic length, the pitch, leads to a very soft behavior. To estimate the compressional constant B_h of the helix, we assume that a typical layer compressional coefficient scales with the inverse square of the intrinsic length (the layer spacing versus the pitch). Taking a layer spacing of 3 nm and a typical value for the pitch of 3 μm, we see that B_h will be about six orders of magnitude smaller than the

layer compressional constant of a typical smectic phase. A typical speed for second sound in smectic is $c = 3 \cdot 10^3$ cm/s. Keeping in mind that the sound speed scales with the square root of the compressional constant, we expect $c_h = 3$ cm/s. This compares quite well with the speed one estimates from the experimental data[10]: $\omega = 2\pi \cdot 10^4\ s^{-1}$, $d = 4\ \mu m$ and thus $c_h = 4$ cm/s.

FUTURE EXPERIMENTS

From the picture suggested in the last section, there emerge immediately several experiments which could be performed to further test the interpretation given here.

First of all it would be highly desirable to do the same type of experiment for a freely suspended thick film[2] of a smectic C^* liquid crystal. In this case no spatial inhomogeneities are generated by spontaneous splay in the case without an external electric field, since one has a free surface and no glass plates enforcing a certain director orientation. Thus one does not expect to observe the electroacoustic effect found in samples with a thickness comparable to the pitch.[10]

Based on our interpretation one should not observe an analogous electroacoustic effect in nematic, cholesteric and nonchiral smectic liquid crystals including smectic A, B, E, and smectic C. Also for the chiralized version of the recently discovered[15,16] and analyzed[17,18] smectic C_M phase this electroacoustic effect should be absent, although a smaller effect based on a different mechanism could be present there (and in the cholesteric phase as well).

In contrast we expect a similar electroacoustic effect in chiral smectic I^* and F^*, which have a tilted helical structure as smectic C^*, which coexists with the bond orientational order and for which one has therefore also the helix displacement as a macroscopic variable.[19]

CONCLUSIONS AND PERSPECTIVE

In this note we have proposed a possible explanation for an interesting electroacoustic effect observed recently in thin samples of a chiral smectic liquid crystal phase.[10] Our interpretation is based on the reversible dynamic cross-coupling[7-9] between the stress tensor and the polarization. In the absence of spatial variations of the velocity field, as is expected to be the case for a freely suspended thick film of chiral smectics, the observed electroacoustic effect should vanish.

In thicker samples we expect the electroacoustic effect to be less pronounced, since in thick samples the distortions generated by spontaneous splay are smaller in amplitude.

It would also be worthwhile to measure the intensity of the sound wave as a function of the applied DC bias field to see the crossover from the case of no helix unwinding over partial helix unwinding, where one has domain walls, to the helix free case for which the electroacoustic effect is absent.[10] This cross-over could be abrupt or gradual. The threshold field above which the sound wave vanishes, will give one a hint up to which degree of helix unwinding the use of a displacement

field[7,8] or of phase changes[9] as macroscopic variables can be justified, a question that has not been clarified as yet and for which only the answer for the limiting cases is clear.

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