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## Flexoelectric Effects in Cholesteric Liquid Crystals\*

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We discuss flexoelectricity in cholesteric liquid crystals. In particular we point out that experimental results obtained by Patel and Meyer can be understood as surface effects. In addition we discuss similarities and differences between static and dynamic effects associated with flexoelectricity in cholesteric and nematic liquid crystals.

**Keywords:** Flexoelectricity; cholesterics

Flexoelectricity in nematic liquid crystals is a phenomenon that has been studied in many publications for over two decades [1–11]. In 1984 it became clear that there is also a dissipative analogue to flexoelectricity [12], although this contribution is very often not taken into account when analyzing the experimental results, even when they are obtained by dynamic methods. In the present note we discuss flexoelectric effects in cholesteric liquid crystals, for which there is only very little previous work [13–15]. Basically the work presented here is an extension and generalization of our earlier, unpublished work [14]. In addition to flexoelectricity we critically analyze how dynamic experiments in cholesteric liquid crystals are not only modified by the dissipative analogue of flexoelectricity, but by the electric analogue [11, 16, 17] of the Lehmann effect [18] as well.

For uniaxial nematics the preferred direction is characterized by a direction  $\mathbf{n}$ , which does not distinguish between head and tail. Accordingly the

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\*It is a pleasure to dedicate this note of Alfred Saupe with whom we had many interesting and stimulating discussions of the physics of liquid crystals in general and of cholesteric liquid crystals in particular.

variables that emerge in addition to the conserved quantities of an isotropic liquid are the variations  $\delta \mathbf{n}$  of the director, i.e. changes of  $\mathbf{n}$  with

$$\delta \mathbf{n} \cdot \mathbf{n} = 0 \quad (1)$$

Condition (1) is equivalent to using a normalized length for the director  $\mathbf{n}(\mathbf{n}^2 = 1)$ . In the local description of cholesteric liquid crystals [11] one uses the same variables, although strictly speaking only one additional hydrodynamic variable exists [19, 20]. The presence of the helix is taken into account by implementing the pseudoscalar quantity  $\mathbf{n} \cdot \text{curl } \mathbf{n}$ , which reflects the fact that cholesteric liquid crystals with one screw sense are different from their mirror image.

The elastic energy associated with deformations of the director field takes the classical form [11, 21–23]

$$2F_n = \int d\tau [K_1(\text{div } \mathbf{n})^2 + K_2(\mathbf{n} \cdot \text{curl } \mathbf{n} + q_0)^2 + K_3(\mathbf{n} \times \text{curl } \mathbf{n})^2] \quad (2)$$

where  $q_0 = -(\mathbf{n}^0 \cdot \text{curl } \mathbf{n}^0)$  is the equilibrium wavevector of the helix. The flexoelectric term introduced for nematics in [1] (under the misleading term piezoelectric) has the form

$$F_f = \int d\tau f_{\text{flexo}} = \int d\tau e_{ijk} E_k \nabla_i n_j \quad (3)$$

with  $e_{ijk} = e_1 \delta_{ij}^\perp n_k + e_3 \delta_{jk}^\perp n_i$ . It also applies to cholesterics in the local description.

We now come to the discussion of the observation of a flexoelectric effect in a sample of cholesteric liquid crystals [13]. Applying a static electric field  $\mathbf{E}$  perpendicular to the helix axis the pure twist structure acquires some bend and splay contributions due to flexoelectricity. The structure observed in [13] can be described by the director field

$$\hat{\mathbf{n}} = (\cos \theta, \sin \theta \cos \alpha, -\sin \theta \sin \alpha), \quad (4)$$

where  $\theta = \theta(x, y, z)$ . Here  $\alpha$  is the constant angle between the optical axis (i.e. the normal to the planes, where  $\hat{\mathbf{n}}$  lies) and the helix axis in the field-free state (chosen as the  $z$ -axis). Without the electric field the helical phase  $\theta = q_0 z + \text{const.}$ , while  $\alpha = 0$ .

In the following we will show that the angle  $\alpha$  is *not* fixed by bulk elastic forces and the electric field, but given by boundary conditions. Choosing the field in x-direction without loss of generality we obtain for the flexoelectric free energy density

$$f_{\text{flexo}} = -\frac{1}{2}E_x \left\{ \left( \cos\alpha \frac{\partial}{\partial y} - \sin\alpha \frac{\partial}{\partial z} \right) \left( [e_1 + e_3]\theta + \frac{1}{2}[e_1 - e_3]\sin 2\theta \right) + \frac{1}{2}[e_1 - e_3] \frac{\partial}{\partial x} \cos 2\theta \right\} \quad (5)$$

From eq. (5) we read off immediately that the flexoelectric energy is a surface term, as long as the electric field is spatially homogeneous and  $\alpha = \text{const.}$ , i.e. the total flexoelectric energy can be transformed into a surface integral and when minimizing the complete free energy the flexoelectric part does not contribute to the Euler equation. Disregarding for the moment (as in ref. [13])  $e_1 - e_3$  and  $\varepsilon_a$ , the anisotropy of the dielectric tensor, and putting  $K_1 = K_3 = K$ , the problem is effectively two-dimensional ( $\theta = \theta(y, z)$ ). Then minimization with respect to  $\theta$  and  $\alpha$  of the total free energy, Eqs. (2) and (3), reveals that the angle  $\alpha$  is not fixed by this procedure. What is fixed is the relative angle between the orientation of the helix and the orientation of the optical axis,

$$\tan(\phi - \alpha) = \frac{(e_1 + e_3)E_x}{2q_0 K}, \quad (6)$$

where  $\tan \phi = \frac{\partial \theta}{\partial y} / \frac{\partial \theta}{\partial z}$  and  $\phi$  is the angle between the helix axes in the field-free and the field-dependent cases (in the latter case the helix is still defined by layers of constant phase  $\theta$ ).

In physical terms this means that depending on boundary conditions and geometry one can find experimentally a rotation of the optical axis (with respect to the laboratory frame set by the external field and the helix in the field-free case) or not. The former case was reported for the experiment in ref. [13]. In a different geometry (probably in a small aspect ratio cell) and under different boundary conditions one might well find, however, a smaller rotation (or none at all) of the optical axis accompanied by an appropriate rotation of the helical axis (as to keep the value of  $\phi - \alpha$ ). This possibility of rotating the helical axis exists, because in the absence of boundary effects the direction, in which translational symmetry is spontaneously broken in

cholesterics, is not fixed—a feature that apparently is not destroyed by flexoelectricity, if the external, constant electric field is orthogonal to the undisturbed helix. This physical picture is essentially unchanged, if  $\epsilon_a$ ,  $e_1 - e_3$  and  $K_1 \neq K_3$  are taken into account.

Now we turn to a discussion of some dynamic aspects. Already more than 25 years ago Helfrich [24] pointed out that the flow behavior in cholesteric liquid crystals is rather different from that of nematics leading to phenomena like plug flow due to premeation effects. As we will discuss in the following these qualitative differences between cholesteric and nematic liquid crystals also apply to the dissipative coupling terms between director deformations and the effects of electric fields. In nematics one has a dissipative analogue of the flexoelectric term [12], which in the dissipation function gives rise to the contribution

$$R_f(\mathbf{E}) = -\zeta_{ijk}^E h_i \nabla_j E_k \quad (7)$$

where the material tensor  $\zeta_{ijk}^E$  is of the form  $\zeta_{ijk}^E = \zeta^E (\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j)$  containing one transport parameter, provided the quasi-static condition  $\text{curl } \mathbf{E} = 0$  is implemented. Clearly this effect also arises in cholesteric liquid crystals when the local description is used. However, in cholesteric liquid crystals there is an additional dissipative process, namely the electric analogue of the Lehmann effect [11, 16, 17]

$$R_L(\mathbf{E}) = \psi_E q_0 \mathbf{E} \cdot (\mathbf{n} \times \mathbf{h}). \quad (8)$$

This contribution to the dissipation function contains one gradient less than that of Eq. (7) and cannot exist in nematics due to parity. As we have pointed out in ref. [17] there is no static contribution to the electric analogue of the Lehmann effect, in contrast to the case of temperature and concentration gradients, where both, static and dynamic Lehmann-type effects exist. Thus the experiments of ref. [16] were directly focusing on  $\psi_E$ .

From the fact that for cholesteric liquid crystals there are two dynamic dissipative cross-coupling terms between director variations and electric fields, it follows that it will be rather difficult to evaluate from measurements the static flexoelectric coefficients alone. In ref. [13], e.g., the dissipative electric Lehmann effect (8) drives the system into a stationary state, which is not identical to the (constraint) equilibrium state (6). The latter was obtained by  $(\delta/\delta\alpha)(F_n + F_f) = 0$ , while the stationary state is given by

$(\delta/\delta\alpha)(F_n + F_f) = \gamma_1 \psi_E q_0 E_x$ , which does not allow to obtain  $e_1 + e_3$  from measured values of  $\phi - \alpha$  without knowing  $\psi_E$ .

We note that even for nematics the evaluation of the static flexoelectric terms from dynamic experiments did not take into account the contributions  $\propto \zeta^E$  [4–10], although typically a combination of the two types of flexoelectric terms enters the dynamics of the director, a feature that will be discussed in detail elsewhere [25].

Interpreting such experiments is even more complicated in (globally) polar cholesterics, which e.g. show a longitudinal piezoelectric effect along the helix which was possibly observed in refs. [26, 27], because they must have some additional local structures (compared to ordinary cholesterics considered above) as has been discussed in [28]. We would also like to point out that the analysis given above can easily be generalized to cholesterics II [29], which can arise when biaxial nematic phases are chiralized [30]. It will also play role for defect phases [31], which can emerge when for chiralized biaxial nematic phases the three directors of the system develop a tendency to twist around different axes. Such phases have probably been observed in lyotropic systems [32]. We close by mentioning the famous cholesteric blue phase, for which similar considerations can be made when the local approach to cholesterics is used. Those are also defect lattices as pointed out first by A. Saupe in one of his classical papers [33].

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