New theoretical results for the Lehmann effect in cholesteric liquid crystals

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We point out for the first time that the classical Lehmann effect, which arises when a temperature gradient is applied parallel to the helical axis of a cholesteric droplet, is not only due to a dynamic cross-coupling between director orientation and temperature, but has also a static contribution. In addition it is demonstrated that the analogous effect in an external homogeneous electric field is purely dynamic, in an external concentration gradient it is a mixture of a static and a dynamic contribution, whereas in a density gradient there are only static cross-couplings involved. The latter possibility has apparently never been discussed before.

One of the first experiments ever reported in liquid crystals was the Lehmann effect:[1] a rotation of constant angular velocity is observed, when a temperature difference is applied parallel to the helical axis of a droplet of a cholesteric liquid crystal.

In a cholesteric liquid crystal the typically rodlike molecules are arranged in a helical fashion, but are otherwise free to move as in a liquid. Since 1900, no one has supplied an experimental confirmation of Lehmann's early observation. Only very recently, Madhusudana and Pratibha[2] described an analogous effect with the temperature gradient replaced by a dc electric field.

A theoretical interpretation of the Lehmann effect as a phenomenon of a purely dynamic coupling between director orientation and external force has been given by several authors (compare Refs. 3 and 4, and references cited therein). Here we point out for the first time that only in the case of an external electric field is the Lehmann effect purely dynamic, whereas for an external temperature gradient as well as for an external concentration gradient (in mixtures) the Lehmann effect has a dynamic as well as a static coupling contribution. Finally, for a density gradient there is only a static cross-coupling. The static contributions discussed here have never been investigated in connection with Lehmann-type effects.

As the authors in Refs. 3–5 we adopt the local approach to cholesteric liquid crystals; that is, we take as macroscopic variables characterizing the additional long-lived degrees of freedom the deviations δn of the director from its equilibrium value $\hat{n}^0$, which can be taken to be $\hat{n}^0 = (\cos\phi, \sin\phi, 0)$ with $\phi = q_0 z + \text{const}$ for a helix with an axis parallel to z and where q_0 is the wave vector of the helix. For a strictly hydrodynamic description valid for length scales large compared to the pitch of the helix and using the displacement along the helical axis as a hydrodynamic variable we refer to Refs. 6–8. Since a helix is different from its mirror image, it is possible to associate with it a pseudoscalar quantity, which is a scalar except for its sign change under parity. It is introduced via $q_0 = -(\hat{n}^0 \cdot \text{curl}\,\hat{n}^0)$ and its presence allows for a number of cross-couplings absent in nematic liquid crystals.[3,4]

Here we focus on the statics and the dissipative dynamics, especially on the cross-couplings between the director variations and the other variables. Reversible dynamic effects will be discussed in detail in a forthcoming publication.

Using as macroscopic variables variations of density $\delta\rho$, of entropy density $\delta\sigma$, of the direction δn, and the density of linear momentum $\mathbf{g}$, we find for the generalized free energy

$$F = F_{\text{nema}} + K_2 q_0(\hat{n}\cdot\text{curl}\,\hat{n} + q_0) - \tau_\sigma q_0(\hat{n}\cdot\text{curl}\,\hat{n} + q_0)\delta\sigma$$
$$- \tau_\rho q_0(\hat{n}\cdot\text{curl}\,\hat{n} + q_0)\delta\rho , \qquad (1)$$

where F_{nema} contains all terms already present in nematic liquid crystals, including flexo- and dielectric contributions.[3] If necessary (e.g., in mixtures), the concentration can be used as an additional variable. It enters Eq. (1) analogously to the entropy density. As we well see below, τ_σ and τ_ρ are the static cross-couplings between δn and $\delta\sigma, \delta\rho$, which enter as static contributions into the Lehmann-type effects. We have given these terms very recently for compensated cholesterics[9] and we will present a detailed comparison with the other local approaches to cholesterics elsewhere.[10]

The Gibbs relation and the balance equations are set up in the usual way (compare, for example, Refs. 6 and 11). In the same way as the thermodynamic forces can be derived by taking variational derivatives of the generalized free energy with respect to the variables, the same can be done for the dissipative contributions by taking variational derivatives of the dissipation function with respect to the thermodynamic forces. We have for the dissipation function

$$R = R_{\text{nema}} + \psi_T q_0 \nabla T \cdot (\hat{n}^0 \times \mathbf{h}) + \psi_E q_0 \mathbf{E} \cdot (\hat{n}^0 \times \mathbf{h}) , \qquad (2)$$

where ψ_T and ψ_E are the dissipative dynamic couplings associated with Lehmann-type effects (called ν/γ_1 in Ref. 3) and $\mathbf{h} \equiv -(\delta/\delta\hat{n})\int F\,dV$. For a mixture the chemical potential difference enters Eq. (2) analogously to the temperature.

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To see how the static and the dissipative dynamic contributions enter the Lehmann and/or the Lehmann-type effects we proceed along the lines discussed in detail in Ref. 3. Taking an external temperature gradient along the helical axis, it is easily verified that the complete hydrodynamic equations allow for the solution $\mathbf{v} = 0$, i.e., there is no flow. The dynamics of the orientation of the director in the plane perpendicular to the helical axis is then given by

$$\delta\dot{\phi} = \left[\hat{n}^0 \times \frac{\partial R}{\partial \mathbf{h}}\right]_z = \zeta_1(K_2 - q_0^2 \tau_\sigma^2 C_V T_0^{-1})\nabla_z^2 \delta\phi$$

$$+ (\zeta_1 \tau_\sigma C_V T_0^{-1} + \psi_T)q_0 \nabla_z T , \qquad (3)$$

where $\delta\phi$ denotes the angle between the director and a fixed direction in the plane perpendicular to the helical axis, and where ζ_1 is the director friction coefficient ($\equiv 1/\gamma_1$) and C_V is the specific heat at constant density. Whether or not a uniform rotation arises as a consequence of Eq. (3) depends on the boundary conditions at the limiting surfaces. As shown, for example, in Ref. 3, free surfaces (freely suspended films) lead to a uniform rotation and we find for the rotation velocity of the director orientation, keeping both the dynamic and the static contribution

$$\omega \equiv \delta\dot{\phi} = q_0(\zeta_1 \tau_\sigma C_V T_0^{-1} + \psi_T) . \qquad (4)$$

As we read off immediately from Eq. (4), both the static contribution (τ_σ) and the dissipative contribution (ψ_T) enter and cannot be separated experimentally by the Lehmann experiment.

For an external homogeneous electric field parallel to the helical axis, the situation changes since there is only a dissipative dynamic and no static contribution, because neither the flexoelectric effect nor the dielectric effect influence the orientation of $\hat{n}$ in the plane normal to the field. The latter effect could, however, force $\hat{n}$ out of the normal plane,[4] if $\epsilon_a > 0$ (tensor components defined as in nematics), a possibility we will not investigate further. Then we get in this case instead of Eq. (4)

$$\omega = q_0 \psi_E . \qquad (5)$$

This is the configuration studied by Madhusudana and Pratibha,[2] thus providing directly from experiment a value for the dissipative cross-coupling ψ_E.

Finally, we come to the case of an external density gradient (at constant temperature), for which there is no dissipative dynamic cross-coupling. We obtain

$$\omega = q_0 \zeta_1(\tau_\rho - \tau_\sigma C_V T_0^{-1}\beta) , \qquad (6)$$

where $\beta \equiv \partial T/\partial \rho$. Equation (6) represents a rather intriguing prediction, since it says that a small pressure gradient applied to a freely suspended cholesteric droplet will also produce a constant rotation velocity of the director (in which case also the dynamic cross-coupling ψ_T enters the expression for the rotation velocity).

Since the concentration and the chemical potential difference in mixtures enter the free energy and the dissipation function analogously to the entropy density and the

temperature, it is obvious that an external gradient in the chemical potential difference leads to a Lehmann-type effect, which has a static and a dynamic component as in the temperature gradient case. However, the actual formula for the rotation velocity due to a chemical potential gradient [and Eq. (4) for mixtures] is rather complex, because of the additional cross-couplings between the concentration and the other degrees of freedom.

To get an upper bound of this velocity we make use of the requirement of thermostatic stability, and thus obtain from the generalized free energy the inequality

$$\tau_\rho^2 q_0^2 < K_2 \frac{\partial^2 F}{\partial \rho^2} . \qquad (7)$$

In the same spirit, one can estimate the static contributions induced by temperature variations

$$\tau_\sigma^2 q_0^2 < K_2 T_0 C_V^{-1} . \qquad (8)$$

Similarly, we get from the positivity of the entropy production the following upper bounds for the dynamic coefficients:

$$\psi_T^2 q_0^2 < \zeta_1 \kappa_\perp, \quad \psi_E^2 q_0^2 < \zeta_1 \sigma_\perp , \qquad (9)$$

where $\kappa_\perp$ and $\sigma_\perp$ are the perpendicular components (in nematic notation) of the heat conduction and the electric conduction, respectively. Unfortunately, lower bounds —even approximate ones—require extensive microscopic calculations.

Clearly, the prediction of a constant rotation velocity of the orientation of the molecules, as a consequence of an applied density or pressure gradient, is experimentally the most challenging one. Although this experiment might not be easily done in cholesteric liquid crystals, for which it is close to impossible to produce stable thick freely suspended films, there seems to be a way out. Everything we have said above for the cross-couplings of the director in cholesterics to variations in density, concentration, temperature, and to an external homogeneous electric field applied equally well to the in-plane director in chiral smectic C^* showing a helix. Provided that the coupling to bond-orientational order is weak, the same can be expected to hold for the other chiral smectic phases showing a helical structure, namely, F^* and I^* (cf. Ref. 12 for a macroscopic description of these phases). It is well known that in chiral smectics perfectly stable, thick freely suspended films can be produced fairly easily.[13] And it should be possible to apply to such a film a pressure gradient—e.g., by blowing air at the film statically or at a small frequency. Then the only problem left to be overcome to observe visually the rotation of the in-plane director would be to find a chiral smectic liquid crystal with a fairly large pitch (of order 10 μm or more) or, alternatively, to choose a material which has a weakly first order C^*-cholesteric transition, in which case the chiral smectic helix is known to unwind as the phase-transition temperature is approached. It seems more difficult experimentally, but not impossible, to apply gradients of temperature or chemical potential to chiral smectic films, which would also show under these conditions Lehmann-type effects.

In conclusion, we have discussed the Lehmann effect originally observed for a temperature gradient applied along the helical axis of a cholesteric droplet and we have pointed out that static contributions are as important as dissipative dynamic ones for the understanding of this effect. Generalizations of this analysis to other external forces such as homogeneous electric fields and gradients of density and concentration on one hand as well as to other types of liquid crystals, namely chiral smectics, have been given and a number of novel predictions has been made. Finally, we point out that the same effects will arise in cholesterics II,[14] the cholesteric phase, which is locally biaxial nematiclike.

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