

Lehmann effects and rotatoelectricity in liquid crystalline systems made of achiral moleculesHelmut R. Brand,^{1,2,*} Harald Pleiner,² and Daniel Svenšek³¹*Theoretische Physik III, Universität Bayreuth, 95440 Bayreuth, Germany*²*Max-Planck-Institute for Polymer Research, P.O. Box 3148, 55021 Mainz, Germany*³*Department of Physics, Faculty of Mathematics and Physics, University of Ljubljana, Jadranska 19, SI-1111 Ljubljana, Slovenia*

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We discuss Lehmann effects and rotato-electricity for liquid crystalline phases made of achiral molecules. We point out that for static and dynamic Lehmann effects to exist, it is not necessary to have chiral molecules provided the overall structure has macroscopic chirality. This question is of direct relevance for liquid crystalline phases formed by bent-core molecules provided they have a sufficiently low symmetry. This includes systems which break parity symmetry and have overall C_2 or C_1 symmetry. We point out that for liquid crystalline gels and elastomers one should be able to observe rotato-electricity for systems with macroscopic chirality. Rotatoelectricity is associated with the relative rotations between two subsystems, namely, between the network and the director, in an external electric field. Candidates include gels and even monolayers prepared from bent-core molecules with sufficiently low symmetry.

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I. INTRODUCTION

The first scientist to observe experimentally the coupling between an external force and the orientation of the director field in a cholesteric liquid crystal was Lehmann [1]. He observed the rotation of the director in a cholesteric droplet in an external temperature gradient. Several decades later Leslie [2] (compare also the textbooks by de Gennes [3] and Chandrasekhar [4]) described Lehmann effects in a continuum theory for cholesteric liquid crystals assuming a helical structure and a purely dynamic coupling.

After Lehmann's pioneering paper it took almost nine decades before an experiment of a similar nature was performed [5,6] for cholesteric droplets in an external electric field in the two-phase region near the isotropic-cholesteric phase transition. Stimulated by the experimental work described by Madhusudana and Pratibha [5], Lehmann-type effects were reexamined using the hydrodynamic approach [7]. It turns out that in general Lehmann effects in external fields have a static as well as a dissipative dynamic contribution [7]. In addition it was pointed out that Lehmann-type effects are not confined to cholesteric liquid crystals, but can also arise for chiral smectic liquid crystals such as smectic- C^* , I^* , and F^* [7]. It was also suggested already in Ref. [7] to use freely suspended smectic- C^* films to observe Lehmann-type effects. Indeed, only recently have Lehmann-type effects been reported experimentally for freely suspended smectic- C^* with a layer thickness of about 3, . . . , 15 layers in a concentration gradient by Tabe's group [8]. In these experiments phase winding patterns of Lehmann-type due to concentration gradients have been documented in detail [8]. The observation of these patterns complements nicely the studies on pattern formation in freely suspended smectic- C and smectic- C^* films due to a mechanical torque [9] and due to rotating electric fields [10–12].

Inspired by biophysical work on synthetic molecular motors [13–15], Tabe and Yokoyama [16] did the next decisive

experimental step by studying Lehmann-type effects in Langmuir monolayers and the associated pattern formation due to a concentration gradient near an air-liquid interface. They demonstrated experimentally that effectively one layer of a chiral smectic- C^* phase was enough to generate Lehmann-type effects [16], which were also shown to be absent when nonchiral molecules of the same type were used. These experimental results are put on a firm basis from a modeling point of view [17] by introducing the concept of macroscopic chirality. The latter means the existence of a pseudoscalar quantity, q_0 , that changes sign under spatial inversion and that allows the discrimination of the inverted state from the original one. The existence of q_0 is not necessarily based on the presence of chiral molecules, and macroscopic chirality can occur in systems made of achiral molecules, if the symmetry of the phase is low enough.

In the realm of biophysical applications we also suggested that inverse Lehmann-type effects can be used as a microscopic pump [18].

Stimulated by recent progress on liquid crystalline phases formed by banana-shaped or bent-core molecules [19–22] we suggest here a further generalization of Lehmann-type effects. We point out that for sufficiently low-symmetry liquid crystalline phases (e.g., C_2 or C_1) formed by achiral molecules, Lehmann-type effects can occur. In Sec. II we briefly summarize the analysis of what are now considered to be classical Lehmann effects. In Sec. III we generalize these Lehmann-type effects to systems formed by achiral molecules and discuss the pertinent pseudoscalar quantities involved. In Sec. IV we analyze rotato-electric effects for this type of system and present a dissipative rotato-electric contribution not considered before.

II. CLASSICAL LEHMANN EFFECTS

To set the stage for the analysis given in the next section we briefly summarize the description of Lehmann-type effects as it applies to cholesteric and chiral smectic liquid crystal phases. Since the investigations presented in Ref. [7] it is clear that

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Lehmann-type contributions come in two groups, namely, as static and as dissipative dynamic contributions. Using the local approach [23–26], that is, the director \mathbf{n} for the description of cholesterics and the in-plane director \mathbf{c} [3] for tilted chiral smectics, we have for the generalized energy

$$F = F_{\text{nem}} + L_2 q_0 [\mathbf{n} \cdot (\nabla \times \mathbf{n}) + \frac{1}{2} q_0] - q_0 [\mathbf{n} \cdot (\nabla \times \mathbf{n}) + q_0] (\tau_c \delta c_1 + \tau_\sigma \delta \sigma + \tau_\rho \delta \rho), \quad (1)$$

where we have used macroscopic variations of the mass density ρ , of the concentration c_1 , and of the entropy density σ , as well as the director \mathbf{n} (or \mathbf{c} in the chiral smectic case). Here q_0 is classically associated with the chirality of the molecules. The helical axis \mathbf{p} is perpendicular to the rotation plane of the director in the cholesteric case and parallel to the layer normal ($\mathbf{k} \parallel \mathbf{p}$) for chiral smectics.

L_2 is the elastic constant associated with linear twist, and F_{nem} includes all terms already present in the corresponding achiral liquid crystal system. The distinction of the elastic constant for the linear twist term ($\sim L_2$) and of the elastic constant ($\sim K_2$) associated with the contribution to the deformation energy quadratic in the twist goes back to Frank [27]. The difference between L_2 and K_2 has attracted attention recently for the case of compensated cholesterics [28] for which a thermomechanical Lehmann effect has been observed in the absence of a helical structure [29]. In Ref. [28] we have shown that for $K_2 \neq L_2$ the divergence of the pitch of the helix at the helix inversion point does not necessarily mean that the pseudoscalar q_0 vanishes, thus making a Lehmann effect possible at the helix inversion point.

The dissipative part of the dynamics can be discussed most succinctly in terms of the dissipation function R [23,26]. It is constructed in terms of the thermodynamic forces, which are either the conjugate quantities or their gradients

$$R = R_0 + \kappa_{ij}^E E_i \nabla_j T + D_{ij}^E E_i \nabla_j \mu_c + \frac{1}{2} \sigma_{ij} E_i E_j + q_0 \epsilon_{ijk} h_j n_k (\psi_c \nabla_i \mu_c + \psi_\sigma \nabla_i T + \psi_e E_i), \quad (2)$$

where σ_{ij} , κ_{ij}^E , and D_{ij}^E are the electric conductivity, thermoelectric, and concentration-electric tensors, respectively (replace n_i by c_i in the chiral smectic case). R_0 contains all the terms not associated with electric terms or the dissipative Lehmann-type effects. In Eq. (2) the thermodynamic forces are the gradients of temperature T and of the chemical potential associated with the concentration μ_c , the electric field \mathbf{E} , and the molecular field h_i , conjugate to the director (the in-plane director in the chiral smectic case). The second line of Eq. (2) contains all the dissipative Lehmann-type effects as made explicit by their proportionality $\sim q_0$. In contrast to the static case, where there is no coupling to an electric field [7], such a contribution arises naturally in the dissipation function. There is no dissipative coupling between density gradients and the molecular field of the director, since there is no dissipative current associated with density (particle conservation).

As usual in the symmetry-based approach to hydrodynamics and macroscopic dynamics [23,26], the dissipative part of the currents and quasicurrents is obtained by taking variational

derivatives of the dissipation function with respect to the corresponding forces.

III. LEHMANN-TYPE EFFECTS IN SYSTEMS MADE OF ACHIRAL MOLECULES

In a pioneering experiment on Langmuir monolayers [16] it has been demonstrated that a helical structure is not a necessary requirement to observe Lehmann-type effects. These experiments were performed on compounds composed of chiral molecules. Using achiral compounds of the same structure, Lehmann-type effects were not observed. Since the molecules were tilted in these Langmuir monolayers, they effectively formed one layer of a chiral smectic- C^* phase. In Ref. [17] we have argued that a necessary condition to observe Lehmann-type effects is, indeed, not a helical structure but rather a system that is macroscopically chiral. In Ref. [18] we have demonstrated that inverse Lehmann-type effects can be used as a pump, both in the presence of a helical superstructure or, more generally, for systems showing macroscopic chirality.

Here we take the argument one step further by pointing out that the molecules making up the liquid crystalline phase need not be chiral to show Lehmann-type effects in the bulk or in Langmuir monolayers. All that is, indeed, necessary is a structure with macroscopic chirality or, equivalently, collective chirality. This effect can be generated by at least two different mechanisms: either via the by now well-established way using chiral molecules as building blocks, which are then assembled into superstructures showing macroscopic chirality, or, and this possibility seems to have been overlooked so far, one can arrange achiral molecules, for example, onto layers, such that the overall symmetry of the collective arrangement has sufficiently low symmetry as to allow macroscopic chirality. This can be done for truly three-dimensional (3D) or for quasi-2D systems as in the case of Langmuir monolayers. As a prerequisite for such a behavior one only needs a low enough symmetry for such a collective arrangement, e.g., C_2 or C_1 symmetry.

In fact, liquid crystalline phases of such a low symmetry have been predicted and found experimentally for liquid crystalline phases made of banana-shaped or bent-core molecules [19–22]. For phases of such low symmetry it was shown in Refs. [19,30] that one can construct a pseudoscalar quantity for both cases, C_2 and C_1 (these phases were denoted by C_{B2} and C_G in Refs. [19,30]). Denoting by \mathbf{m} the polar direction of these banana-shaped molecules and by \mathbf{n} the nonpolar direction tilted with respect to the layer normal \mathbf{k} , a pseudoscalar q_0 arising naturally is of the form [19,30]

$$q_0 = \mathbf{m} \cdot (\mathbf{k} \times \mathbf{n}). \quad (3)$$

In the C_{B2} phase the polar direction \mathbf{m} is in the plane of the layers (C_2 symmetry), while if also \mathbf{m} is tilted a C_1 symmetric C_G phase is obtained.

All the arguments for analyzing Lehmann-type effects then go through completely in parallel to the classical static and dissipative dynamic analysis of Lehmann effects [7]. In particular, Eqs. (1) and (2) apply, if \mathbf{n} is replaced by \mathbf{m} or its in-plane projection for the C_{B2} and C_G phase, respectively. We thus predict that even for an arrangement made of achiral bent-core molecules in a sufficiently low symmetry configuration

in Langmuir monolayers Lehmann-type effects of the nature demonstrated in Ref. [16] should be observable.

Generally speaking, in the bulk (that is in 3D) one might not even need a layered system. In Ref. [31] it was pointed out that in biaxial polar nematics phases of C_1 symmetry are possible based on general symmetry considerations. One has to keep in mind, however, that biaxial nematics may show a competition between helices in different directions and subsequent frustration [32]. In the case where one is able to synthesize compounds showing such polar biaxial nematic phases of C_1 symmetry, one expects those to reveal Lehmann-type effects. One approach could be as follows. For nematic polymers [33] biaxial nematic phases with the very low, but nonchiral, C_{1h} symmetry have already been identified. If one is now able to remove the last horizontal mirror plane, for example, by a slight redesign of the molecules, then one could have C_1 and thus a chiral system of the type we are interested in.

Another class of systems for which the analysis given above is relevant are liquid crystalline phases for which one has the simultaneous presence of conventional quadrupolar orientational order [3] and tetrahedric (octupolar) orientational order, which has been pioneered for the field of liquid crystals by Fel [34]. Such liquid crystalline phases have been discussed over the last few years macroscopically [35–39], microscopically [40,41], and, quite recently for achiral ferrocene derivatives, experimentally [42]. In Refs. [36] and [39] it has been pointed out that for nematic phases of (nonchiral) D_{2d} symmetry a linear gradient term coupling quadrupolar and octupolar order leads to the spontaneous formation of regions of opposite helicity that show Lehmann-type effects. Should such domains become large enough, the predictions regarding Lehmann-type effects could be tested experimentally.

IV. ROTATO-ELECTRICITY IN LIQUID CRYSTALLINE GELS AND ELASTOMERS WITH MACROSCOPIC CHIRALITY

Liquid crystalline elastomers and gels are fascinating materials in the macroscopic domain since they couple two subsystems, the network associated with the gel or elastomer and a liquid crystalline phase [43,44]. As has been pointed out by de Gennes [45], this combination of subsystems allows for a novel type of macroscopic variables, namely, relative rotations between the two subsystems. These relative rotations couple to mechanical strains and therefore can have a profound influence on the behavior of liquid crystalline elastomers under mechanical external fields such as extensional and shear forces leading in nematic elastomers to a reorientation of the director field [46–50] above a threshold. The combined rotation of the two subsystems does not contribute to the macroscopic behavior, since the elastic energy must be invariant under rigid rotations [23,26].

In Ref. [51], using the global description of cholesteric liquid crystals [23,24], the property of rotato-electricity has been predicted to arise for cholesteric and chiral smectic elastomers. The rotato-electric contribution to the generalized energy takes the form

$$F_{RE} = q_0 \zeta^R p_j D_j [\delta\phi - p_i \Omega_i], \quad (4)$$

where p_i denotes the helical axis [3], D_j is the electric displacement vector, ϕ , the phase variable characterizes the rotation of the director, and Ω_i is the network rotation vector, $\Omega_i = \frac{1}{2} \varepsilon_{ijk} \nabla_j u_k$ with u_k the network displacement. By minimizing the appropriate terms in the generalized energy one finds that an electric field applied parallel to the helical axis leads to a relative rotation between the director and the network [51]. This prediction has been studied in detail [52] using the local description of cholesterics [2–4] for various boundary conditions clarifying further under which experimental conditions rotato-electricity is best observed. In the local description Eq. (4) takes the form

$$F_{RE} = q_0 \zeta^R \varepsilon_{ijk} D_i n_j \tilde{\Omega}_k \quad (5)$$

with $n_i(\mathbf{r})$ the helical director orientation, $\delta n_i(\mathbf{r})$ its local variation, and $\tilde{\Omega}_k = \delta n_k - n_j \Omega_{ji}$ the relative rotations, with $\Omega_{ij} = \frac{1}{2} (\nabla_i u_j - \nabla_j u_i)$. In principle, there exist additional static couplings between relative rotations and temperature, density, and concentration gradients. They are of higher gradient order [53,54] and will not be discussed here.

We now generalize this scenario to systems made of achiral molecules that nevertheless have macroscopic chirality in the form of a pseudoscalar quantity q_0 . For example, for smectic C_{B2} and smectic C_G liquid crystalline gels these pseudoscalars take the form (compare also Refs. [19,30])

$$q_0 = \mathbf{m} \cdot (\mathbf{k} \times \mathbf{n}) \quad (6)$$

with \mathbf{n} the tilted nonpolar direction and where the polar vector \mathbf{m} resides in the layer plane for C_{B2} , while for C_G it has also a component perpendicular to the layer planes. In that case the rotato-electric energy contribution, Eq. (4), can be taken over with \mathbf{k} as helical axis (replacing \mathbf{p}) and ϕ describing rigid rotations of the \mathbf{n}/\mathbf{m} structure about the layer normal. Similarly, Eq. (5) applies with \mathbf{n} replaced by \mathbf{m} or $\mathbf{m}^{(\perp)}$ for the C_{B2} and C_G phase, respectively ($m_i^{(\perp)} \sim m_i - k_i k_j m_j$).

Finally, we would like to point out that there are hitherto undetected dynamic counterparts to the static rotato-electric effect discussed above. The dissipation function characterizing all effects leading to the generation of heat (entropy production) shows the additional contributions

$$R = R_0 + q_0 \varepsilon_{ijk} n_j L_k^\perp (\psi_c^R \nabla_i \mu_c + \psi_\sigma^R \nabla_i T + \psi_e^R E_i), \quad (7)$$

where L_k^\perp is the thermodynamic conjugate force to relative rotations. Equation (7) describes (dissipative) dynamic changes of relative rotations ($\partial_t \tilde{\Omega}_i$) due to concentration and temperature gradients and due to electric fields, and vice versa, dissipative contributions to concentration, heat, and electric currents due to conjugate force of relative rotations (L_k^\perp). We stress that these dissipative contributions occur for all condensed matter systems with macroscopic chirality including cholesteric and chiral smectic gels and elastomers. For the latter class of systems it has not been discussed before, since the general macroscopic dynamic equations for cholesterics and chiral smectic gels have, in contrast to the case of nematic elastomers [55], not yet been presented.

We close this section by pointing out that there is no direct static or dissipative dynamic analog of rotato-electricity in the magnetic domain, for example, for uniaxial magnetic gels [56], because of the different behavior under parity and time reversal

of the electric polarization \mathbf{P} and the magnetization \mathbf{M} . The most closely related cross-coupling between relative rotations and the magnetization is, in fact, a reversible current coupling the temporal variations of the relative rotations to the internal magnetic field.

V. CONCLUSION

We have shown for liquid crystals and closely related soft matter systems such as Langmuir monolayers, that macroscopic chirality of the structure is sufficient to generate Lehmann-type effects and rotato-electricity. These phenomena were previously thought to be intrinsically related to the chirality of the building blocks or molecules. It turns out, however, that collective arrangements, even of achiral building blocks, with sufficiently low symmetry (C_2 and C_1) are sufficient to generate macroscopic chirality. This result has

direct implications for several of the chiral liquid crystalline phases formed by bent-core achiral molecules [20,22]. A bulk sample or a thin film (for example, a Langmuir monolayer) of such materials should show Lehmann effects. In bulk materials these could be, for example, examined by the influence of an external temperature gradient on chiral domains as they are observed in a number of liquid crystalline phases formed by bent-core molecules.

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