Why Does the Liquid Crystal Rotate?

Continuous Rotation of Achiral Nematic Liquid Crystal Droplets Driven by Heat Flux
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Recommended with a commentary by Jonathan V. Selinger, Kent State University

Liquid crystals are phases which have long-range order in the average molecular orientation, called the director, but which can still flow like liquids. If a material is composed of *achiral* molecules, which are equivalent to their mirror images, then it forms a *nematic* liquid crystal phase with a uniform director. If a material is composed of *chiral* molecules, not equivalent to their mirror images, it forms a *cholesteric* liquid crystal phase, with the director field twisted in a helix.

In a nematic phase, the molecular orientation can be influenced by many different physical mechanisms, including electric and magnetic fields, surface anchoring, and shear flow. In a cholesteric phase, the *Lehmann effect* is another remarkable mechanism to influence the orientation. In the Lehmann effect, a temperature gradient induces a continuous dynamic rotation of the director. This effect might be visualized as the rotation of chiral propellers as a current of heat flows through them.

Although the Lehmann effect was first discovered more than a century ago,[1] it has seen a resurgence of interest in recent years. Much of this interest arises from seeing the Lehmann effect in a more general context. Madhusudana and Pratibha found that the effect can be induced not only by a temperature gradient but also by a static electric field.[2] Tabe and Yokoyama found that it can further be induced by a gradient in humidity, which leads to a flow of water across a liquid-crystal film.[3] In the latter case, the flow-induced rotational force can compete with elastic forces on the director field, leading to complex spatial patterns. Hence, liquid crystals driven by the Lehmann effect are a simple case of materials driven out of equilibrium by molecular motors, and they can serve as examples of active matter with rotational driving. Furthermore, an inverse Lehmann effect might be used as a microscopic pump.[4]

Despite this resurgence of interest, important theoretical aspects of the Lehmann effect are still not clear. The standard textbook of de Gennes and Prost states that "a field [temperature gradient] (which is a polar vector) may induce a torque (an axial vector) in a cholesteric: this is possible only because the cholesteric differs from its mirror image."[5] But what needs to differ from its mirror image: the chemical structure of the liquid-crystal molecules, or the packing of the molecules into the phase, or the large-scale configuration of the director field? The early theory of Leslie discussed only a single chiral coupling, which presumably arises from the structure of the molecules.[6] However, further theoretical research showed that other types of coupling can occur: Rotation might be caused by the temperature gradient coupled with certain deformations of the director field,[7,8] or coupled with a spontaneous chiral order parameter arising from the packing of achiral banana-shaped molecules into a chiral structure.[9]

With all of these theoretical possibilities, we must ask: Which actually occurs in experiments? In a series of recent papers, Oswald and collaborators have addressed this question. In one study, they prepared a mixture of chiral liquid crystals with a compensation point, i.e. a special temperature at which the twisting effects of the chiral components cancel, so

that the cholesteric pitch goes to infinity.[10] This cancellation is just *accidental*, not caused by any symmetry, and the material is still made of chiral molecules. If the Lehmann effect were due to microscopic chirality in the molecular structure, then it should still occur at this compensation point. However, the experiments actually show that the Lehmann effect vanishes at the compensation point. This result implies that the Lehmann effect is not caused by the microscopic structure, but rather by the macroscopic chirality of the cholesteric helix.

In the current paper,[11] Oswald and collaborators do a complementary experiment. Instead of working with a cholesteric liquid crystal, they work with micron-scale droplets of a nematic liquid crystal of achiral molecules. In these droplets, the boundary conditions require a nonuniform configuration of the director field. Because the twist elastic constant K_2 of the material is particularly small, the director field in each droplet spontaneously breaks inversion symmetry and forms a twisted bipolar configuration, as shown in Fig. 1. In each droplet, the handedness of the configuration is random. The droplets are then subjected to a temperature gradient. Remarkably, they exhibit a Lehmann effect, rotating in the plane perpendicular to the temperature gradient, with right- and left-handed droplets rotating in opposite directions. This observation shows that the Lehmann effect does not require molecular chirality, but rather can be caused by the macroscopic twisted structure of the director field.

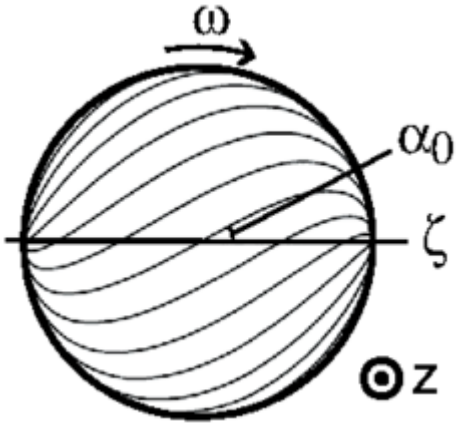

Fig. 1. Twisted bipolar configuration of the director field in a droplet, with the thermal gradient along the *z*-direction. From Ref. 11.

In future research, it might be interesting to connect these experimental studies of the Lehmann effect with the theory of orientation-dependent handedness by Efrati and Irvine.[12] This theoretical approach describes experimental phenomena like Lehmann rotation as a tensor relationship between stimulus and response. Here, the stimulus is the temperature gradient (which is a polar or proper vector), and the response is the rotation (which is an axial or pseudovector). These quantities must be related by a pseudotensor of rank 2. This pseudotensor represents the twisted director configuration of the droplet, but it carries more information than just the standard pseudoscalar twist; it also depends on the orientation of the droplet. Efrati and Irvine have already used this approach to describe the rotation of a macroscopic helicoidal propeller, which can spin in a right- or left-handed sense depending on its orientation with respect to the airflow. It should be possible to use a similar approach to predict how Lehmann rotation depends on the orientation of the droplet.[13] This theoretical approach may well be equivalent to existing theories,[6,7] but expressed in a way that highlights the pseudotensor structure of the twisted bipolar configuration.

Apart from these symmetry considerations, future research should also explore the microscopic origin of Lehmann rotation. What determines the magnitude of the effect? The existing macroscopic theories suggest that the effect arises from the differential dissipation as a current passes through a right- or left-handed rotating environment. How can we model this dissipation on the microscopic scale, or design molecules to have a greater Lehmann effect? Does the effect require a chiral conformation of the molecules (which might be induced by a chiral director field even in a nematic phase of achiral molecules), or would it occur even for rigid rod-like molecules?[14] Addressing these issues could bring together the liquid-crystal

physics explored in the current paper with new types of non-equilibrium theory for driven systems.

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[13] Reference 11 reports that droplets with their dipole axis parallel to the temperature gradient do not rotate, and only droplets with their axis perpendicular to temperature gradient are considered in the data analysis, so the rotation rate must depend significantly on droplet orientation.
[14] This issue was suggested by R. L. B. Selinger.