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Reply

Reply to the Comment by H. Pleiner and H. R. Brand

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In their comment [1], Pleiner and Brand claimed to resolve the apparent contradiction between experiments and theory regarding the existence of a Lehmann effect in compensated cholesteric liquid crystals. Their article contains however some ambiguities that deserve further comments.

Apart from racemic mixtures, materials made of chiral molecules are intrinsically chiral, whatever the orientation of these molecules. For instance, an aqueous solution of chiral molecules is chiral and this can be demonstrated experimentally by measuring chiral physical properties such as its optical rotatory power. Recently, we emphasized that the same held for a compensated cholesteric, which is intrinsically chiral even though the molecules are not arranged in a helical structure [2]. This chirality is even experimentally observable and was first shown by Éber and Jánossy [3] and confirmed by our group more recently [2].

Chiral properties that may occur in chiral materials are theoretically described by pseudo-scalars, *i.e.* scalars which change sign under parity. Rotatory power in chiral solutions, spontaneous helical twist and Lehmann-type coefficients in cholesterics are such pseudo-scalars. From Curie's symmetry principle, chiral properties cannot appear in achiral phases, and accordingly the pseudo-scalar coefficients have to be zero in achiral phases. But the converse is not true: chiral properties of chiral phases may accidentally individually vanish. This is what happens in compensated cholesterics when the helical twist vanishes. Nevertheless the phase remains intrinsically chiral at the macroscopic scale at the compensation point where Lehmann-type couplings can be evidenced.

A system is either equivalent to its image in a mirror or chiral. Chirality is thus a 0/1 property. A finer description might be desired to quantify how much the material is chiral. For that reason, Pleiner and Brand proposed to introduce a new quantity q_0 in order to quantify chirality. This notation is quite confusing because q_0 is not the equilibrium twist, also denoted by q_0 in the introduction of the comment. They then proposed that every pseudo-scalar coefficients characterizing the cholesteric phase be proportional to q_0 with a prefactor that can possibly vanish at a certain temperature. In this framework, the linear twist coefficient k_2 in the Frank elastic free energy reads $k_2 = L_2 q_0$, where L_2 vanishes at the compensation temperature (as a consequence the equilibrium twist q_0^{helix}, equal to $-k_2/K_2$, also vanishes at this temperature, as expected). The problem, in our opinion, is that the definition of q_0 is too vague. In addition, we do not see any justification to explicitly write that all pseudo-scalar coefficients are proportional to q_0.

Another problem concerns the scalar quantities Δ used to describe the (density, entropy, etc.) deviations with respect to the unwound configuration of the director field. We believe that it is incorrect and misleading to describe this configuration as an achiral state of the system, because, as we previously emphasized, the unwound cholesteric (being compensated or not) is still chiral.

In conclusion, it is currently admitted that the Lehmann coefficient does not vanish usually at the compensation temperature of a cholesteric phase. This result is due to the fact that, at the compensation temperature, the unwound cholesteric is still chiral at the macroscopic scale in spite of its "nematic-like" structure (its symmetry group is D_∞ and not $D_{\infty h}$ as in usual nematics). For that reason, it is simply incorrect to write, as theorists proposed before [4,5], that the Lehmann coefficient is proportional

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to the equilibrium twist q_0^{helix} in the vicinity of the compensation point. If one admits this result, there is, from our point of view, no real, nor even apparent, contradiction between the classical theory and the observations. Pleiner and Brand introduced a new quantity q_0 to quantify chirality (we highly recommend to use another notation, χ for instance, since q_0 is not the equilibrium twist). This idea could indeed be interesting but the definition of q_0 needs to be better clarified and its proportionality to all pseudo-scalar coefficients proven.

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