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Helmut R. Brand and Harald Pleiner

Regular Article

Cubic and tetragonal liquid crystal phases composed of non-chiral molecules: Chirality and macroscopic properties

Helmut R. Brand[1,2,a] and Harald Pleiner[2,b]

[1] Department of Physics, University of Bayreuth, 95440 Bayreuth, Germany
[2] Max-Planck-Institute for Polymer Research, POBox 3148, 55021 Mainz, Germany

Abstract. We discuss the symmetry properties as well as the macroscopic behavior of the cubic liquid crystal phases showing large chiral domains of either hand in some non-chiral compounds reported recently in the group of Tschierske. These phases are tricontinuous. While they have O or $I432$ symmetry in each chiral domain, the overall symmetry is $Im\bar{3}m$ as there is no net chirality for compounds composed of non-chiral molecules. It turns out that a rather similar type of phase has also been reported for triblock copolymers. Here we analyze in detail the macroscopic static and dynamic behavior of such phases and we predict, among other results, that they show the analog of static and dissipative Lehmann-type effects in their chiral domains. A description of a cubic liquid crystalline phase of T_h symmetry, which has not yet been found experimentally, is also included. Suggestions for experiments are outlined to identify such a phase. In addition, we discuss tetragonal liquid crystalline phases of D_{4h} and D_4 ($I422$) symmetry as they have been reported last year experimentally in connection to the Q phase.

1 Introduction

Quite recently the group of Tschierske has described and analyzed the observation of chiral domains in several bicontinuous cubic phases composed of achiral molecules [1, 2]. The length scale on which these chiral domains are observed is $\sim 500\,\mu m$ [1,3,4], meaning large compared to all molecular length scales and therefore requiring a meso- or macroscopic description of these collective phenomena.

To put the work presented here into a the broader context of more frequent liquid crystalline phases we briefly summarize first the symmetry properties and selected macroscopic aspects of their behavior. For a more detailed exposition we suggest, for example, the book by de Gennes [5]. The most frequently investigated and physically well-understood liquid crystalline phase are uniaxial nematic liquid crystals. These are anisotropic fluid systems with fluidity in 3 dimensions and one preferred direction characterized by a unit vector, that does not distinguish between head and tail, the so-called director, n_i, which has length unity: $n_i^2 = 1$ with $n_i \rightarrow -n_i$ symmetry. It characterizes quadrupolar orientational order and renders all material properties of uniaxial nematics spatially anisotropic including dielectric properties, electric conductivity, thermal conductivity and viscous properties.

The deformation energy of this soft material is characterized by three elastic constants for director deformations: splay, twist and bend. Applied voltages of order $1\,V$ allow deformations of the director field and have been at the basis of applications in liquid crystal displays (LCDs).

When the molecules giving rise to a nematic phase are replaced by chiral molecules and/or when chiral molecules are added to the non-chiral molecules of a nematic phase, a cholesteric liquid crystalline (LC) phase with a helical superstructure with a characteristic repeat distance, the pitch, P_0, results. The cholesteric phase breaks parity symmetry, since in 3 dimensions it breaks mirror symmetry: left- and right-handed helices are clearly distinguishable from each other. On length scales large compared to the pitch cholesteric LCs are uniaxial [6]. Since many compounds arising in a biological context are chiral, the question of cholesteric phases also naturally comes up in systems as, for example, DNA solutions. We mention in passing that for both, nematic and cholesteric phases, there is a biaxial analogue with two rather than one preferred direction for the orientational order: biaxial nematics [5] and biaxial cholesterics [7].

Major other classes of liquid crystalline phases include layered phases (smectic phases) with varying degree of order within or from layer to layer, but always with positional order (denisty wave) in at least one direction and columnar phases with density waves in at least two dimensions. The simplest phase in this context is the smectic A

[a] e-mail: brand@uni-bayreuth.de
[b] e-mail: pleiner@mpip-mainz.mpg.de

phase with fluidity with isotropic fluidity in 2 dimensions. Depending on the field (for example micellar or biological systems) this type of phase is also called L_α or neat soap.

Approaching the liquid crystalline systems of interest here we refer to cubic liquid crystalline phases of various types including D phases [8], bicontinuous cubic phases in a lyotropic and biological context [9,10] and cholesteric blue phases [11–13]. The characteristic length scale of the unit cell varies from about 10 nm (D phases) to about 300 nm (cholesteric blue phases) reflecting the fact that the number of molecules in a unit cell is varying by about five orders of magnitude.

The observations of Tschierske's group are made for bicontinuous cubic liquid crystals of three networks and $Im\bar{3}m$ (O_h) symmetry. Within each chiral domain one encounters $I432$ or O symmetry. On the other hand, bicontinuous cubic liquid crystals of two networks and $Ia\bar{3}d$ (O_h) symmetry do not show chiral domains and are achiral [3,4].

In retrospect[1] it appears that also early work of about two decades ago [14] shows chiral domains of three networks in a bicontinuous cubic liquid crystal phase.

In the field of ABC triblock copolymers [15–17] cubic bicontinuous triblock copolymers have also been investigated ("double gyroid structures") and a tricontinuous double gyroid structure of $I432$ symmetry [17] has been identified. We note[2], however, that it appears not feasible to observe macroscopic chiral domains in the presently available triblock copolymer systems.

To describe cubic orientational order a fourth-rank traceless symmetric tensor, Q_{ijkl} [18–20] (also called unitary nonor [19]) has been introduced. For a detailed discussion of several of the physical properties and various phase transitions involving achiral cubic phases we refer to [18]. The implications of such an order parameter for transient cubic orientational order above the glass transition have been addressed in [20]. The group theoretical aspects and their physical implications of phases with O and O_h symmetry have been elucidated in refs. [21] and [22].

In contrast to other members of the cubic group such as the tetrahedral phases with T_d or T symmetry [19,22–27], the macroscopic structure of phases with O_h and O symmetry do not break parity (inversion symmetry) [19,21] via the presence of octupolar order. However, the O phase is chiral, possesses a pseudoscalar q_0, but does not show piezoelectricity. As has been noted in ref. [19], the fourth-rank symmetric and traceless order parameter Q_{ijkl} is suitable for the description of the structure of the achiral O_h phase as well as for the chiral O phase.

Motivated by the observation of macroscopic chiral domains in cubic bicontinuous phases of $I432$ (O) symmetry, we present in the bulk of this paper the macroscopic dynamics of such a chiral phase. We find that it allows for the existence of static and dynamic Lehmann effects in each chiral domain separately while the overall symmetry

of the whole system is of $Im\bar{3}m$ symmetry and thus cubic, but achiral.

The existence of a pseudoscalar quantity q_0 in the O phase of chiral cubic liquid crystals can be traced back to the fact that one has three networks joined together, microscopically. This gives rise to a chiral arrangement locally even for non-chiral molecules [1–4], which in turn can lead to chiral domains of macroscopic size. This process has been called "propagation of homochiral twist across the entire networks through helix matching at network junctions" in ref. [1]. In the case of three networks there are two possibilities of joining them leading to either hand microscopically, thus defining q_0 and $-q_0$ as pseudoscalar quantities. A similar reasoning concerning the origin of chirality applies to triblockcopolymers [15–17] (see footnote [2]).

This situation should be contrasted to the type of structural chirality encountered, for example, for bent-core molecules in the smectic C_{B2} (B2) phase of bent-core liquid crystals [28–30]. In this case the way bent-core molecules are arranged macroscopically, *i.e.* the orientation of the polar direction and the tilt of the director with respect to the smectic layers, leads to chirality of either hand. This has been called "ambidextrous chirality" [30].

It is a hallmark of structurally chiral phases that both types of handedness are equal in the sense that there is no energetic preference for one or the other.

We also investigate the order parameter and the hydrodynamics of the T_h phase, the fifth member of the group of cubic phases (compare, for example, ref. [21]). The T_h phase is not breaking parity, and does not show any fourfold axes of rotation. We will discuss how the order parameter of T_h is different from O_h and suggest experiments how to distinguish O_h and T_h practically. We also mention some aspects regarding the chiral T phase in relation to the chiral O phase.

Among the tetragonal phases of interest are in particular D_{4h} and D_4, since phases of these symmetries have been identified recently [31] in connection with the exotic Q phase. Originally found several decades ago [32], for chiral materials [32,33], also a Q phase composed of achiral molecules exists [31]. For chiral compounds its symmetry has been identified as $I422$ (D_4) in ref. [34]. In the case of the Q phase of achiral components, it is shown experimentally that there are domains of both handedness, q_0 and $-q_0$. Here, the chirality of D_4 is brought out by the microscopic model discussed in ref. [31].

The paper is organized as follows. In sect. 2 we discuss in detail the symmetries and the hydrodynamic variables of the O_h and O phases as well as their differences, followed by the symmetry and the hydrodynamic variables of T_h and T in sect. 3 and of D_{4h} and of D_4 in sect. 4. In sect. 5 we derive hydrodynamic equations focusing in particular on the chiral O phase and its physical differences from the achiral O_h phase. In sect. 6 we discuss the hydrodynamics of the T_h (and partly the T) phase and in sect. 7 the hydrodynamics of the D_{4h} and D_4 phases. We conclude in sect. 8 by a summary of the main results and their connection to experiments.

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[1] It is a pleasure to thank Carsten Tschierske for correspondence.

[2] It is a pleasure to thank Volker Abetz for correspondence.

2 Symmetries and symmetry variables of the O_h and the O phase

The O_h phase in cubic liquid crystals can be viewed as a cubic biaxial nematic such that the (mutually orthogonal) directors $\boldsymbol{l}, \boldsymbol{m}, \boldsymbol{n}$ are along the edges of the cube. As directors they are normalized, e.g. $\boldsymbol{n}^2 = 1$, and show the equivalence of, e.g., $\boldsymbol{n} \rightarrow -\boldsymbol{n}$, etc. The three directors are all equivalent. It should be noted that those directors are not preferred directions in the nematic sense, since none of them can be detected by optical inspection. For the cubic order parameter our notation follows closely refs. [18,19]. Specifically, we have

$$Q_{ijkl}(\mathbf{r}) = Q_0[l_i l_j l_k l_l + m_i m_j m_k m_l + n_i n_j n_k n_l$$
$$-\frac{1}{5}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})] \quad (1)$$

with the cubic triad l_i, m_i n_i and the normalization $Q_0^2 = \frac{5}{6}Q_{ijkl}Q_{ijkl}$. The four-fold symmetry axes are given by $\boldsymbol{l}$, $\boldsymbol{m}$, $\boldsymbol{n}$ and $\boldsymbol{l/m}$, $\boldsymbol{m/n}$, and $\boldsymbol{n/l}$ are mirror planes. The three-fold symmetry axes are the space diagonals of the cube. It is not possible to construct out of Q_{ijkl} any tensor of rank 3 or lower, except for the isotropic Kronecker symbol δ_{ij}, thus excluding uniaxial nematic and polar order.

For the chiral O phase the mirror planes of the O_h phase are removed and only the symmetry axes remain. In the O phase the chirality is manifest by the existence of a pseudoscalar quantity q_0 that is of microscopic origin, either due to chiral molecules or due to local chiral arrangements of achiral molecules as discussed in the Introduction. We will adopt the description of ref. [6], where the hydrodynamics of the chiral state is that of the achiral one amended by all possible contributions allowed by q_0. This is similar to the case of a cholesteric liquid crystal arising from a nematic one, when molecular chirality is added.

In the O and the O_h phase the three independent rotations of the rigid structure are the hydrodynamic degrees of freedom (Goldstone modes), like in any other phase with spontaneously fully broken rotational symmetry, i.e. biaxial nematic phases [35,36] and the T_d phase [26,37]. In equilibrium Q_{ijkl} is constant. Changes of that structure, δQ_{ijkl}, generally contain internal deformations, in addition to the rigid rotations of the cubic structure. The former are non-hydrodynamic and will not be considered here. The latter are the symmetry variables given by

$$\delta\Theta_i \equiv \frac{5}{2Q_0^2} Q_{pjkl}\,\epsilon_{ipq}\,\delta Q_{qjkl}. \quad (2)$$

The definition (2) applies to any 1st-order differential δ and can be inverted, $\delta Q_{ijkl} = 2\epsilon_{iqp}Q_{pjkl}\delta\Theta_q$.

For infinitesimal rotations, where in the definition eq. (2) Q_{pjkl} can be replaced by its equilibrium value, the three angles Θ_i, describing infinitesimal rotations about a direction d_i, form an axial vector. In the general case of finite rotations consequent rotations about different direc-

tions do not commute, and the conditions

$$(\delta_1\delta_2 - \delta_2\delta_1)\Theta_i = \frac{5}{Q_0^2}\,\epsilon_{ipq}(\delta_1 Q_{pjkl})(\delta_2 Q_{qjkl})$$
$$= 2\epsilon_{ipq}(\delta_1\Theta_p)(\delta_2\Theta_q) \quad (3)$$

apply [35,38] indicating Θ_i not being a vector.

There is an alternative way of describing the hydrodynamics of the O_h and O phase [36] by using the rotations of the individual directors. One possible representation is

$$\delta\Theta_i = \epsilon_{ijk}(l_j\delta l_k + n_j\delta n_k + m_j\delta m_k), \quad (4)$$

where the equivalence of the rotations of $\boldsymbol{l}$, $\boldsymbol{m}$, and $\boldsymbol{n}$ is apparent. The expression (4) can only be used together with the conditions that guarantee orthogonality among the directors. In order to discuss differences between an isotropic state and the O_h phase, which is optically isotropic, the use of $\delta\Theta_i$, however, is more suitable.

3 Symmetry and symmetry variables of the T_h and the T phase

While the O_h-phase and more recently also the O-phase have become of experimental relevance as symmetries in the field of liquid crystals, the third phase of the cubic family, which does not break parity symmetry, namely the T_h phase, has not been reported as yet experimentally. One of the goals of the present paper is therefore to find out how one could distinguish a liquid crystalline phase with O_h symmetry from a phase with T_h symmetry experimentally. To do this we first analyze how the order parameter for the T_h phase must be different form the order parameter Q_{ijkl} used to characterize the O and the O_h phases discussed above. The difference in symmetry is related to the fact that T_h has no 4-fold axes of rotations. We are therefore looking for an order parameter which accounts for this lower symmetry when compared to O_h and O, but nevertheless has cubic symmetry and does not break parity such as T_d and T. Inspecting the structure of Q_{ijkl} one realizes that one can construct easily the fourth-rank tensor Q_{ijkl}^T

$$Q_{ijkl}^T = Q_0^T[l_i l_j(m_k m_l - n_k n_l) + m_i m_j(n_k n_l - l_k l_l)$$
$$+ n_i n_j(l_k l_l - m_k m_l)]. \quad (5)$$

Note that Q_{klij}^T is antisymmetric, when $\{kl\}$ and $\{ij\}$ are interchanged.

By construction, Q_{ijkl}^T still has $\boldsymbol{l}$, $\boldsymbol{m}$, $\boldsymbol{n}$ as symmetry axes, which are now twofold axes of rotation, rather than fourfold ones, as they are for O_h. Thus we can reduce the symmetry from O_h to T_h by adding Q_{ijkl}^T as order parameter in the T_h phase. Therefore Q_{ijkl}^T can serve as the order parameter for describing the $O_h \rightarrow T_h$ phase transition.

The order parameter Q_{ijkl}^T does not introduce any new variables, and the three rotations of the cube of eq. (2) can here be written as $\delta\Theta_i \sim Q_{pjkl}^T\epsilon_{ipq}\delta Q_{qjkl}^T$. Although

the variables in the O_h and the T_h phase are the same, there are differences in the form of the material tensors, regarding, *e.g.*, Frank elasticity, and electro- and magnetostriction (for liquid crystalline elastomers and gels). The terms electrostriction and magnetostriction refer to coupling terms in the energy, which are linear in the strain and quadratic in the electric or magnetic field. They are of the form

$$\varepsilon_{ES} = \chi^E_{ijkl}\varepsilon_{ij}E_kE_l, \qquad (6)$$

where ε_{ij} denotes the tensor of the strain field and E_i the electric field. Thus electrostriction and magnetostriction are associated with a fourth-rank material tensor, which is symmetric in the first pair of indices (symmetry of the strain tensor) as well as in he second pair of indices separately. Applying an electric field thus leads to an elongation/shortening of the sample, which is quadratic in the electric field.

If a T_h phase gets chiralized, *e.g.*, by the presence of a pseudoscalar of microscopic origin, the mirror planes are removed and the phase has T symmetry. The same phase is obtained, when a tetrahedral T_d phase gets chiralized. The hydrodynamics of such a chiral T phase has been described earlier [27, 37] and will not be repeated here. Only a few remarks on the differences between the T_h and the T phase will be given.

4 Symmetry and symmetry variables of the D_{4h} and D_4 phase

Both, the D_{4h} and the D_4 phase are of tetragonal symmetry. Therefore both can be characterized by a preferred direction, which we represent by a director n_i. It is also associated with uniaxial nematics and can be extracted from the quadrupolar order parameter

$$Q_{ij} = S\left(n_in_j - \frac{1}{3}\delta_{ij}\right). \qquad (7)$$

This observation allows us to use as invariants in both phases, D_{4h} and D_4, the director n_i and the transverse Kronecker delta, $\delta^\perp_{ij} = \delta_{ij} - n_in_j$. This is all what is needed to describe the rank-2 material tensors.

For the rank-4 material tensors the four-fold symmetry in the plane perpendicular to n_i requires a suitable rank-4 order parameter. In principle, one could use Q_{ijkl} familiar from the O_h, and O phases. A more specific and convenient choice is the projection of the cubic order parameter onto the plane $\perp n_i$

$$\begin{aligned}Q^{tr}_{ijkl} = Q_0\big[&l_il_jl_kl_l + m_im_jm_km_l \\ &-1/4\left(\delta^\perp_{ij}\delta^\perp_{kl} + \delta^\perp_{ik}\delta^\perp_{jl} + \delta^\perp_{il}\delta^\perp_{jk}\right)\big], \qquad (8)\end{aligned}$$

where l_i and m_i are directors describing a square in the plane $\perp n_i$ with the normalization $Q_0^2 = 2Q^{tr}_{ijkl}Q^{tr}_{ijkl}$. With the help of the transverse directors, the transverse Kronecker delta can also be written as $\delta^\perp_{ij} = m_im_j + l_il_j$, reflecting the tetragonal symmetry, *i.e.* invariance under a transformation $\boldsymbol{m} \to \boldsymbol{l} \wedge \boldsymbol{l} \to -\boldsymbol{m}$.

While D_{4h}, also called $\frac{4}{m}$ mm or $\frac{4}{m}\frac{2}{m}\frac{2}{m}$, has horizontal mirror planes, they are absent in the chiral D_4 phase (also called 422), due to the presence of a pseudoscalar q_0. Particularly, it is well known [22] that the D_4 phase shows piezoelectricity requiring a parity-breaking third-rank tensor, which can be constructed using q_0 and $n_in_p\epsilon_{pjl}\delta^\perp_{kl}$.

The piezoelectric effect is, in general, a static coupling in the energy between the strain field and an electric field, which takes the form

$$\varepsilon_p = \zeta^p_{ijk}\varepsilon_{ij}E_k. \qquad (9)$$

It requires a ground state that breaks parity (inversion) symmetry such as, for example, quartz [22]; ζ^p_{ijk} is symmetric in i and j due to the symmetry of the strain field. In contrast to electrostriction, piezoelectricity is an effect in an external electric field. Conversely, applying a strain to a piezoelectric material generates an electric field. The piezoelectric effect has no direct analogue in the energy in the magnetic domain, since the magentic field is odd under time reversal and even under parity, while the electric field is even under time reversal and odd under parity.

The symmetry variables are the three rotations of the rigid director structure, $\delta\Theta_i$, eq. (4). Since there is a preferred direction, $\boldsymbol{n}$, which is visible in the microscope, it is appropriate to split the three rotation angles into two rotations of, and one rotation about, $\boldsymbol{n}$

$$\delta n_i \equiv \epsilon_{ikj}\,n_j\delta\Theta_k \qquad \text{and} \qquad \delta\Omega \equiv n_i\,\delta\Theta_i \qquad (10)$$

where the non-commutativity relations, eq. (3), apply to a nonlinear theory with finite rotations, appropriately. Note, both variables are odd under the replacement $n_i \to -n_i$ and read in terms of the directors $\delta n_i = \delta^\perp_{ij}\delta n_j$ and $\delta\Omega = -l_i\delta m_i + m_i\delta l_i$.

5 Hydrodynamics of the O_h and O phases

5.1 Statics

We will concentrate on the hydrodynamic effects specific to the O_h, and particularly the O phase, when compared to the isotropic state.

The Gibbs relation, connecting changes of the total energy density $d\varepsilon$ with those of all hydrodynamic variables (entropy density σ, mass density ρ, momentum density g_i, concentration c, and the symmetry variables) [39,40] can be written as

$$d\varepsilon = Td\sigma + \mu d\rho + v_idg_i + \mu_cdc + h^{\Theta'}_i d\Theta_i + \Psi^\Theta_{ij}d\nabla_j\Theta_i \quad (11)$$

thereby defining the conjugate quantities (temperature T, chemical potential μ, velocity v_i, relative chemical potential μ_c, and the "molecular fields" $h^{\Theta'}_i$ and Ψ^Θ_{ij}) as partial derivatives of the total energy.

Since rigid rotations of the cubic structure must not cost energy, $h^{\Theta'}_i$ is zero (in the absence of any orienting field) and only gradients of Θ_i (space dependent rotations

of the cube structure) enter the total energy. In the general case the "molecular fields" can be combined into

$$h_i^\Theta = h_i^{\Theta\prime} - \nabla_j \Psi_{ij}^\Theta - 2\epsilon_{ikl}\Psi_{kj}^\Theta \nabla_j \Theta_l, \qquad (12)$$

where the last contribution is due to relation (3).

The form of the quadratic, Frank-type gradient energy of inhomogeneous rotations of the cube is not affected by the chirality. Therefore, the expression for ordinary cubic biaxial nematics [36,41] is valid for the O phase, as well. In our notation it has the form

$$\varepsilon_g = \frac{1}{2} K_{ijkl}(\nabla_j \Theta_i)(\nabla_l \Theta_k) \qquad (13)$$

with $K_{ijkl} = K_1 \delta_{jl}\delta_{ik} + K_2(\delta_{jk}\delta_{il} + \delta_{lk}\delta_{ij}) + K_3 Q_{ijkl}$ containing 3 independent Frank parameters.

On the other hand, linear gradient terms, absent in the O_h phase are possible in the O phase, due to the existence of q_0 (as in the cholesteric phase) and we get one contribution

$$d\varepsilon_{lg} = q_0 k_2 \nabla_i \Theta_i = q_0 k_2 \epsilon_{ijk}(n_i \nabla_j n_k + m_i \nabla_j m_k + l_i \nabla_j l_k). \qquad (14)$$

This expression is the analogue of the well-known linear twist term of chiral uniaxial nematics (cholesterics). There, the linear gradient term of the director results in a helical director structure as defect-free energetic minimum state. In biaxial cholesterics helical rotations of the different directors are independent and lead to frustration [7]. In the O phase rotations of the directors are not independent, but have to preserve the cubic structure. Therefore eq. (14) is minimized by a single helical rotation of the cubic structure about any of the directors —which one does not matter, since all are indistinguishable. However, this rotation of the cubic structure cannot be seen directly by optical means. Only if the orientation of the cube matters for a definite experiment, the helical structure might be detected indirectly.

If there is a linear gradient term, there are also static Lehmann-type energy contributions [42,43], bilinear in linear director gradients and variations of the scalar variables, $S \in \{\sigma, \rho, c\}$

$$d\varepsilon_F = q_0(\nabla_i \Theta_i)\sum_S K^S(\delta S)$$
$$= q_0 \epsilon_{ijk}(n_i \nabla_j n_k + m_i \nabla_j m_k + l_i \nabla_j l_k)\sum_S K^S(\delta S), \qquad (15)$$

where the sum over S comprises all scalar variables. All these terms are due to the chirality of the O phase.

Static Lehmann contributions are associated in general with chiral phases for which either the building blocks (for example, the molecules) are chiral [42], or for which the chirality is due to the low symmetry of the spatial arrangement of the units [43].

The other energy contributions regarding the scalar variables in the O_h and O phase are the same as in the isotropic state and comprise the appropriate (scalar) static susceptibilities.

5.2 Dynamics

The dynamics is described by conservation laws for the conserved variables and balance equations for the non-conserved ones [35]. The former contain (the divergence of) the currents (*e.g.*, the stress tensor, σ_{ij}, in the momentum conservation law, the heat and the concentration current), while in the latter case a quasi-current balances the temporal changes of the variable

$$\frac{\partial \Theta_i}{\partial t} + v_j \nabla_j \Theta_i - \omega_i + J_i^{\Theta R} + J_i^{\Theta D} = 0 \qquad (16)$$

with the vorticity $\omega_i = \epsilon_{ijk}\nabla_j v_k$. Both, currents and quasi-currents, are additively split into a reversible (superscript R) and a dissipative part (superscript D). The irreversible part of the dynamics can be derived from the dissipation function (or the entropy production R, which acts as the source term in the entropy balance), while the reversible part is non-potential, requires $R = 0$ (conserved entropy), and often (but not always) follows from general invariance principles [40].

The dissipative dynamics of the O_h and O phases is similar to that of the isotropic phase regarding second-rank material tensors (*e.g.*, describing heat conduction, diffusion, and thermo-diffusion) containing one material coefficient each [36]. The viscosity tensor, relating the stress tensor σ_{ij} with symmetrized flow, $2A_{ij} = \nabla_j v_i + \nabla_i v_j$, has the same form as K_{ijkl} in eq. (13) containing three flow viscosities, one more than in the isotropic phase. There is one rotational viscosity, γ_1, describing relaxation of inhomogeneous cube rotations, $J_i^{\Theta D} = (1/\gamma_1)h_i^\Theta$.

In addition, in the O phase there are dissipative Lehmann-type terms due to chirality (as in cholesterics)

$$2R_L = q_0 h_i^\Theta \sum_P \psi_P \nabla_i P, \qquad (17)$$

where the sum over P comprises $\nabla_k P \in \{\nabla_k T, \nabla_k \mu_c, E_k\}$, the thermodynamic forces related to the thermal degree of freedom, the concentration and the electric field, with one dissipative coefficient ψ_P for each force.

The dissipative parts of the Lehmann effect has been modeled first by Leslie [44] for classical cholesterics in the framework of a continuum-type description. Using the hydrodynamic approach [42] it became clear that such effects not only arise for cholesterics, but also for chiral smectic liquid crystals. Dissipative Lehmann effects are thus complementing the static Lehmann effects discussed in sect. 5.1.

As a result there are contributions to the dissipative quasi-current, $J_i^{\Theta D} = q_0 \sum_P \psi_P \nabla_i P$ indicating rotations of the cubic structure about the direction of, *e.g.*, an applied temperature gradient, which in nematics is called Lehmann rotation [42,43]. In the cubic O phase, however, this rotation cannot be seen directly. Variational derivatives of R with respect to $\nabla_i P$ lead to, *e.g.* dissipative heat current, $j_i^{\sigma D} = q_0 \psi_T h_i^\Theta$. In nematics, where the appropriate force h_i^n can be generated by an external rotating magnetic field, this is called the inverse Lehmann effect [45]. How to excite a finite h_i^Θ and therefore a heat current will

be discussed in the next section. In ref. [45] it has been shown that measurements of the inverse Lehmann effect and of the direct Lehmann effects discussed so far can be combined to disentangle static and dynamci contributions of the Lehmann effect.

The part of the reversible dynamics related to transport ($v_j \nabla_i \Theta_i$) or convection ($-\omega_i$) in eq. (16) is due to general invariance principles, like Galilean invariance and rotational covariance [39,40]. For biaxial nematics this has been investigated in detail in ref. [35].

In many nematic phases there is a phenomenological reversible coupling between director reorientation and symmetrized flow known as "flow alignment" and "back flow". In general, flow alignment takes in the stress tensor the form

$$\sigma_{ij} = \lambda_{ijk} h_k \qquad (18)$$

where σ_{ij} is the stress tensor and h_k is the molecular field associated with variables characterizing spontaneously broken rotational symmetries, such as, for example, deviations of the director from its preferred direction. λ_{ijk} does not lead to dissipation or the generation of heat, since h_k is even under time reversal [46,47].

In uniaxial nematics there is one phenomenological parameter (related to the flow alignment angle under shear flow). In the O_h and O phases, however, there is none due to the cubic symmetry [36].

5.3 External fields

Generally, external fields can orient liquid crystals. In uniaxial nematics, electric fields E_i and magnetic fields H_i reorient the nematic director either along the field or perpendicular to it, depending on the sign of the dielectric or magnetic anisotropy. In cubic biaxial nematics (both, in the O_h and in the O phase) there is no anisotropic quadratic term in the energy due to the high symmetry of the cubic phase. There are also no third-rank tensors. Naturally anisotropic terms of fourth order in external fields are possible for both, the O_h and the O phase; they are of the structure $\sim E_i E_j E_k E_l$, $\sim H_i H_j H_k H_l$ or $\sim E_i E_j H_k H_l$ and are well documented in the literature for cubic liquid crystalline systems [41,48,49].

Taking, *e.g.*, the magnetic field energy

$$\varepsilon_H = -\chi_H Q_{ijkl} H_i H_j H_k H_l \qquad (19)$$

it is easy to see that the energetic minimum for $\chi_H > 0$ is obtained, when the cube is oriented with one of its edges along the field ($\boldsymbol{l}$, $\boldsymbol{m}$, or $\boldsymbol{n}$), while for $\chi_H < 0$ the field is along the space diagonal of the cube. For deviations from the equilibrium orientation, a finite energy occurs giving rise to a torque on the cube $h_i^\Theta = (|\chi_H|/3)\epsilon_{iqp}Q_{pjkl}H_q H_j H_k H_l$ using the inverse of eq. (2).

For the inverse Lehmann effect, a magnetic field rotating with angular velocity ω (*e.g.*, in x/y plane) drags the cube also into a rotation in this plane. There is, however, a time lag, Δ, due to the rotational viscosity γ_1, ensuring that the orientation of the cube (for $\chi_H < 0$) is never in the equilibrium position parallel to the field. For the

time lag we get $\sin(4\Delta) = -(12/|\chi_H|H_0^4)\gamma_1\omega$, which leads to a heat current $j_z^{\sigma D} = q_0 \psi_T h_z^\Theta = -q_0 \psi_T \gamma_1 \omega$ independent of the magnetic field strength, similar to the nematic case [43]. Although the cube rotation cannot be seen directly, the heat current due to this inverse Lehmann effect is detectable by standard means.

In a gel with O or O_h symmetry, permanent or transient elasticity is prone to electro- and magnetostriction, when external fields are present. This effect is governed by a fourth-rank tensor of the structure, eq. (13), with 3 independent material coefficients.

6 Hydrodynamics of the T_h phase

The hydrodynamic variables in T_h phases are the same as for the O_h and O phase, respectively: three independent rotation of the cubic structure, eq. (2).

In the following we will focus on the changes of the hydrodynamics in the T_h phase when compared to the hydrodynamics of the O_h phase.

For the static behaviour related to director deformations we find one additional Frank constant, which is associated with the contribution

$$\varepsilon_{T_h} = K_4 Q_{ijkl}^T (\nabla_i \Theta_k)(\nabla_j \Theta_l). \qquad (20)$$

Thus instead of having three Frank constants as for O_h, eq. (13), we have now four for T_h.

There exists a third-rank tensor,

$$d_{ijk} \equiv \epsilon_{ipq} Q_{qjpk}^T = Q_0 \big(n_i[l_j m_k + l_k m_j] \\ + m_i[l_j n_k + l_k n_j] + l_i[n_j m_k + n_k m_j]\big) \qquad (21)$$

which is symmetric in j, k and is invariant under spatial inversion. By its definition it is clear that d_{ijk} fulfils all symmetry requirements of the T_h phase, since Q_{ijkl}^T does. In the representation in terms of directors one has to take into account that $\boldsymbol{l}$ is a shorthand for $\boldsymbol{n} \times \boldsymbol{m}$ thereby leading to expressions that are manifestly symmetric under $\boldsymbol{n} \to -\boldsymbol{n}$ and $\boldsymbol{m} \to -\boldsymbol{m}$. This tensor allows to describe flexoelectricity

$$\varepsilon_{fl} = e_1 d_{ijk} E_i \nabla_j \Theta_k \qquad (22)$$

containing one flexoelectric coefficient. Flexoelectric contributions are, in general, static contributions coupling electric fields to gradients of the variables associated with spontaneously broken rotational symmetries. They have been introduced first for uniaxial nematics by Meyer [50] under the notation of "piezoelectricity".

All static material properties associated with second-rank tensors are the same as for O_h and contain one parameter each due to the cubic symmetry.

For liquid crystalline gels and elastomers, where elasticity is described by the strain tensor u_{ij}, there are two other static material properties associated with fourth-rank property tensors, which differ in the T_h phase from the O_h phase, namely electrostriction and magnetostriction, for which we have one additional contribution each

$$\varepsilon_{T_h EM} = Q_{ijkl}^T u_{kl} (\chi^E E_i E_j + \chi^H H_i H_j). \qquad (23)$$

This contribution comes in addition to the usual three contributions from the O_h phase (compare ref. [22] for solid state physics). It can be investigated experimentally, for example by applying an electric field in the x-direction. Then the strain fields induced in the two directions perpendicular to the electric field, namely the y- and z-directions, would be different. It is easily checked that in this case the strain difference

$$u_{yy} - u_{zz} \sim Q_0^T \chi^E E_x^2 \qquad (24)$$

arises.

Other static fourth-rank property tensors are identical in the O_h and T_h phase, [22], in particular those associated with the energetic contributions $\chi_{ijkl}^E E_i E_j E_k E_l$, $\chi_{ijkl}^H H_i H_j H_k H_l$, and the Hookean elasticity, $c_{ijkl} u_{ij} u_{kl}$, since the antisymmetric $Q_{ijkl}^T = -Q_{klij}^T$ cannot contribute. On the other hand, $\chi_{ijkl}^{EH} H_i H_j E_k E_l$ has a fourth coefficient due to Q_{ijkl}^T.

In the dynamics there is, in the O_h and the T_h phase, one rotational viscosity and three flow viscosities, the latter because the viscosity tensor has the same symmetry as the elastic one. In contrast to the O_h phase, there is flow alignment and back flow in the T_h phase

$$J_i^{\Theta R} = \lambda^T d_{ijk} A_{jk}, \qquad (25)$$
$$\sigma_{ij}^R = -\lambda^T d_{kij} h_k^{\Theta}, \qquad (26)$$

indicating *e.g.* orientation of the cubic structure by shear flow, which is, however, difficult to detect optically.

When the T_h phase is chiralized, a chiral T phase results. It shows piezoelectricity $\varepsilon_p \sim q_0 d_{ijk} E_i u_{jk}$ and non-linear dielectric orientation $\varepsilon_d \sim q_0 d_{ijk} E_i E_j E_k$, but also characteristic reversible dynamic couplings between flow and $\nabla_k P \in \{\nabla_k T, \nabla_k \mu_c, E_k\}$, *e.g.* $\sigma_{ij}^R \sim q_0 d_{kij} \nabla_k T$ and $j_i^{\sigma R} \sim q_0 d_{ijk} A_{jk}$. The full hydrodynamics of the T phase has been presented before in refs. [27, 37], in a slightly different language.

7 Hydrodynamics of the D$_{4h}$ and D$_4$ phases

We focus on the hydrodynamics of the symmetry variables δn_i and $\delta \Omega$ and their couplings to other variables involving various material tensors.

Since both, the D_{4h} and the D_4 phase do not have a polar direction, there are no non-vanishing property tensors of first rank. Second-rank property tensors in both, the D_{4h} and the D_4 phase take typically the uniaxial form

$$\xi_{ij} = \xi_{\parallel} n_i n_j + \xi_{\perp} \delta_{ij}^{\perp}, \qquad (27)$$

where n_i denotes the director associated with the preferred direction. Property tensors of this type include the dielectric tensor and the tensor of magnetic susceptibilities, in hydrostatics and in dissipative dynamics the tensors of heat conduction, electric conductivity, and diffusion.

For fourth-rank property tensors, the differences to the case of uniaxial nematics can all be traced back to the

existence of the additional variable $\delta \Omega_i$, and of the order parameter Q_{ijkl}^{tr}, which is characteristic for tetragonal symmetry. For the analogue of Frank's free energy of deformations of the orientational order,

$$\varepsilon_g = \frac{1}{2} K_{ikjl} (\nabla_i n_k)(\nabla_j n_l) + K_7 \delta_{ik}^{tr} \epsilon_{lij} (\nabla_l \Omega)(\nabla_j n_k)$$
$$+ (K_5 n_i n_j + K_6 \delta_{ij}^{tr})(\nabla_i \Omega)(\nabla_j \Omega). \qquad (28)$$

There are four Frank-type orientational elastic coefficients related to distortions of the director, *i.e.* one more than in uniaxial nematics, since K_{ijkl} contains additionally $K_4 Q_{ijkl}^{\perp}$, totaling the number of coefficients to seven.

Similarly, there are seven coefficients for the electro- and magnetostriction tensor (in the case of gels). Another class of rank-4 tensors are the viscosity tensor η_{ijkl} and the elastic tensor c_{ijkl} (in the case of gels), which have six coefficients. This is one more than in the uniaxial case [39, 46] and the additional coefficient, *e.g.* $\eta_6 Q_{ijkl}^{\perp}$ again is due to the tetragonal transverse order. Thus, the number of coefficients in rank-4 tensors in the D_{4h} and D_4 phase is the same as in the D_{2d} phase [26, 37], in accordance with [22].

There is no rank-3 tensor, which is odd under the $\boldsymbol{n} \to -\boldsymbol{n}$ transformation. Therefore, the flexoelectric energy $\varepsilon_{fl} = f_{ijk} E_i \nabla_j n_k$ has the same form as in the uniaxial nematic phase [5] and contributions of the form $E_i \nabla_i \Omega$ are ruled out. Similarly in the dynamics of the symmetry variables

$$\frac{\partial}{\partial t} n_i + v_j \nabla_j n_i + (\boldsymbol{\omega} \times \boldsymbol{n})_i - \lambda_{ijk} A_{jk} + X_i = 0, \qquad (29)$$

$$\frac{\partial}{\partial t} \Omega + v_j \nabla_j \Omega - n_i \omega_i + Y^{\Omega} = 0, \qquad (30)$$

the flow alignment tensor, λ_{ijk}, is as in the uniaxial nematic case [46], and there is no flow alignment regarding Ω.

All features discussed above apply to the D_4 phase as well, but there are additional chiral effects, which we will discuss in the following. First we inspect linear gradient terms in the generalized energy as well as static and dissipative Lehmann-type contributions. For these the arguments for the D_4 phase are quite in parallel to those for the O phase, with differences originating in the different symmetries, tetragonal and cubic, involved.

For deformations of the director fields we find two linear gradient terms

$$d\varepsilon_{lg} = q_0 (\epsilon_{ijk} k_{\perp} n_i \nabla_j n_k + k_{\parallel} n_i \nabla_i \Omega). \qquad (31)$$

Minimizing the first contribution leads to a helical rotation of $\boldsymbol{n}$ about any of the two (indistinguishable) transverse preferred directions, $\boldsymbol{m}$ or $\boldsymbol{l}$, while the second one gives a helical rotation of the transverse structure about $\boldsymbol{n}$. This latter type of helical state actually arises, if $|k_{\parallel}| > |k_{\perp}|$.

For the static Lehmann contributions we have, with $P \in \{\sigma, \rho, c\}$ denoting all the scalar variables,

$$d\varepsilon_F = q_0 \sum_P \delta P (K_{\perp}^P \epsilon_{ijk} n_i \nabla_j n_k + K_{\parallel}^P n_i \nabla_i \Omega) \qquad (32)$$

again with two coefficients for each variable δP.

Table 1. We summarize the physical properties of the various phases considered here. We have also included the T_d and T phases, which show octupolar order for comparison purposes. The macroscopic physical properties listed include chirality, Lehmann effects, number of electrostrictive coefficients, piezoelectricity and flow alignment.

Phase	Parity	Chiral	Lehmann	Electrostriction	Piezoelectric	Flow alignement
O (432)	-1	yes	yes	3	no	no
O_h ($m3m$)	$+1$	no	no	3	no	no
T_h ($m3$)	$+1$	no	no	4	no	yes
T_d ($\overline{4}3m$)	-1	no	no	3	yes	no
T (23)	-1	yes	yes	4	yes	yes
D_{4h} ($\frac{4}{m}mm$)	$+1$	no	no	7	no	yes
D_4 (422)	-1	yes	yes	7	yes	yes

For the corresponding dissipative Lehmann contributions in the D_4 phase we get

$$2R_L = q_0 \sum_S (\nabla_k S) \left(\psi_\perp^S h_k^n + \psi_\parallel h_k^\Omega \right), \qquad (33)$$

where the thermodynamic forces $\nabla_i S \in \{\nabla_i T, \nabla_i \mu_c, E_i\}$ and the conjugates, h_i^n and h_i^Ω, follow from the appropriate part of the energy density

$$\varepsilon = h_i^{n'} dn_i + \Psi_{ij}^n d\nabla_j n_i + h_i^\Omega d\nabla_i \Omega + T d\sigma + \mu_c dc + \dots \quad (34)$$

In the absence of external fields there is $h_i^{n'} = 0$, but in the general case the full conjugate of dn_i can be written as

$$h_i^n = h_i^{n'} - \nabla_j \Psi_{ij}^n + 2n_k \Psi_{kj}^n \nabla_j n_i \qquad (35)$$

where the last contribution is due to non-commutativity of finite rotations, eq. (3). There is no external field that couples to $\delta\Omega$ and only gradients of Ω enter the energy.

Piezoelectricity has a rather specific form in the D_4 phase [22]. Taking the preferred direction $\boldsymbol{n}$ as the z-axis, and applying a transverse electric field in the x-direction, leads to a strain in the y-z plane, while a field in y-direction leads to a strain of opposite sign in the x-z plane. Defining $s_{ijk} = n_i n_p \epsilon_{pjk}$ we can write the piezoelectric energy as

$$\varepsilon_{piezo} = \frac{1}{2} q_0 \zeta_p (s_{ijk} + s_{kji}) E_j u_{ik} \qquad (36)$$

which has one piezocoefficient ζ_p, since the strain tensor u_{ik} is symmetric. The other rank-3 tensor $s_{ijk}^\perp = \delta_{ip}^\perp \epsilon_{pjk}$ does not lead to an additional contribution in the symmetrized case, eq. (36). The sign of ζ_p is not fixed (instead of s_{ijk} one could have defined a $\tilde{s}_{ijk}$ with the opposite sign), but the two contributions for $\boldsymbol{E} \| \boldsymbol{l}$ and $\boldsymbol{E} \| \boldsymbol{m}$ always come with opposite signs.

There is a third-order orienting energy, linear in the electric field, $\boldsymbol{E}$, and quadratic in the magnetic one, $\boldsymbol{H}$,

$$\varepsilon_f^{cub} = \frac{1}{2} q_0 \zeta_f^{cub} (s_{ijk} + s_{kji}) E_j H_i H_k \qquad (37)$$

of the same form as piezoelectricity. Taking $\boldsymbol{E} \perp \boldsymbol{H}$, the electric field orients one of the transverse directions and the magnetic fields defines a direction tilted $45°$ to the

second transverse direction and $45°$ to $\boldsymbol{n}$. This fits to a negative dielectric anisotropy that favours $\boldsymbol{n} \perp \boldsymbol{E}$, but is always in conflict with the diamagnetic anisotropy that favours either $\boldsymbol{n} \perp \boldsymbol{H}$ or $\boldsymbol{n} \| \boldsymbol{H}$.

There are no physical effects in D_{4h} related to the parity-conserving and n-symmetric tensor s_{ijk}.

The fourth-order field energies are, for D_{4h} and D_4, of the same form as for the O and O_h phase, eq. (19), when Q_{ijkl} is replaced by Q_{ijkl}^{tr}. They allow to orient $\boldsymbol{m}$ or $\boldsymbol{l}$, when the appropriate coefficient is positive, and $(1/\sqrt{2})(\boldsymbol{m} \pm \boldsymbol{l})$ if it is negative.

8 Summary

In this paper we have analyzed cubic and non-polar tetragonal liquid crystals and gels, which do not possess octupolar order, in contrast to the systems reviewed in ref. [37]. Systems of interest therefore included O, O_h and T_h from the cubic class and D_4 and D_{4h} from the tetragonal class of symmetries. The physical properties of these phases including Lehmann effects, piezoelectricity, flow alignment and number of electrostrictive/magnetostrictive coefficients is summarized in table 1. For completeness as well as for comparison purposes we have also included in this table the cubic phases T_d and T, which have octupolar order.

We have investigated the symmetry properties and the macroscopic behavior of cubic liquid crystals with O and O_h symmetry as they have been found recently experimentally for bicontinuous cubic liquid crystalline systems with three networks. Since the domains with one hand can reach up to 1 mm in size [1], our prediction that static and dissipative dynamic Lehmann type should be observable in these chiral domains in the O phase. In particular, the inverse Lehmann effect can be tested experimentally: A rotating magnetic field drags along the cubic structure to also rotate. Although the latter rotation is not visible in the microscope, the temperature gradient induced by this rotation due to the inverse Lehmann effect is detectable.

For the T_h phase, which has no longer four-fold axes of rotation, in contrast to the O_h phase, we predict the occurrence of additional electrostrictive and magnetostrictive effects associated with the static coupling terms linear in the strain and quadratic in the external

fields. For example, when an electric field is applied parallel to an axis of the cube, there is a strain difference induced in the two directions perpendicular to the applied field. The experimental detection of such an effect would help to identify the cubic T_h phase, which has not yet been reported in the field of liquid crystals. As a reversible dynamic effect not present in the O and O_h phases, we find that flow alignment, reflecting the coupling between the director orientation and extensional flow involving symmetrized velocity gradients, is possible in T_h phases. We also find that, when a T_h phase is chiralized, a phase of T symmetry, which breaks parity symmetry, in contrast to the T_h phase, results.

We emphasize that all cubic phases discussed (O, O_h and T_h) are optically isotropic just like an isotropic liquid phase. While in some cases one can see experimentally the growth habits of a cubic phase growing in the isotropic liquid phase, our analysis provides additional possibilities to distinguish the cubic phases from the isotropic phase as well as methods to identify the three cubic phases O, O_h and T_h separately.

Comparing the macroscopic properties of the D_{4h} and the D_4 phase, it emerges that for the chiral D_4 phase static and dynamic Lehmann effects are expected to be detectable experimentally provided the domain size of domains of either hand can be made sufficiently large. It should be noted that there two types of helical structures are possible in D_4, either the preferred direction is helical, or one of the transverse direction winds about the (constant) preferred direction. The former case is detectable in the microscope, while the latter is not and such a domain looks like an achiral D_{4h} one. In crossed magnetic and electric fields the orientation of the preferred direction is always frustrated due to a cubic field-orienting energy. For liquid crystalline gels of D_4 symmetry a rather specific piezoelectric effect —the static cross-coupling between strain and electric fields, which is linear in the electric field— is predicted: taking the z-axis as the preferred direction, and applying a transverse electric field in the x-direction, leads to a strain in the y-z plane, while a field in y-direction leads to a strain of opposite sign in the x-z plane.

It thus emerges that cubic and non-polar tetragonal liquid crystalline systems can be rather varied in mechanisms generating mesoscopic and macroscopic chiral domains in systems composed of achiral molecules. For chiral domains generated in optically isotropic systems made of achiral molecules only one such mechanism has been suggested so far [51]. In addition, interesting electro- and magneto-mechanical effects emerge.

Author contribution statement

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