## Dynamic interplay of nematic, magnetic and tetrahedral order in ferromagnetic nematic phases

Tilen Potisk, <sup>1,\*</sup> Harald Pleiner, <sup>2</sup> and Helmut R. Brand <sup>1</sup> Department of Physics, University of Bayreuth, 95440 Bayreuth, Germany <sup>2</sup>Max Planck Institute for Polymer Research, 55021 Mainz, Germany

(Received 30 August 2018; published 25 October 2018)

We consider the influence of tetrahedral/octupolar order on ferromagnetic nematic liquid-crystalline phases. The presence of tetrahedral order leads to broken parity symmetry in an achiral liquid-crystalline system, in addition to broken time-reversal symmetry associated with the existence of a spontaneous magnetization. As a consequence we find static as well as reversible and irreversible dynamic cross-coupling terms absent in usual ferromagnetic nematics. Several static and dynamic experiments are suggested to detect possible tetrahedral order. We predict that linear gradients terms in the generalized energy involving the ferromagnetic magnetization and the nematic director field lead to chiral domains of ambidextrous helicity. As a characteristic dissipative dynamic cross-coupling we point out that the rotation of the magnetization can be driven by temperature and/or concentration gradients. Conversely heat and concentration currents can be generated by rotations of the magnetization. As a characteristic example for reversible cross-coupling terms we analyze the consequences of the coupling between the molecular field of the nematic director and temperature and concentration gradients.

DOI: 10.1103/PhysRevE.98.042703 ©2018 American Physical Society

#### I. INTRODUCTION

Following the pioneering work of Fel [1, 2] on tetrahedral/octupolar order in liquid crystals the theoretical investigations of the physical consequences of this type of nonpolar order associated with broken parity symmetry focused on applications in liquid crystals: phase transitions [1, 3–5], microscopic models and phase diagrams [6-8] as well as the macroscopic properties of liquid-crystalline phases involving tetrahedral order [9–15]. Most of the experimental work on the question of tetrahedral order concentrated on the influence on phase transitions and on macroscopic properties of liquidcrystalline phases formed by bent-core molecules [16–28]. In addition, there were experimental reports indicating the presence of tetrahedral order in another class of compounds, namely ferrocenomesogens [29, 30]. Most of these observations and experimental results such as ambidextrous helicity and ambidextrous chirality [16, 19, 26] and unusual behavior near the isotropic - liquid crystal phase transitions including shifts of the phase transition temperature by up to 10 K linear in electric fields, two optically isotropic phases in magnetic fields and reentrant isotropic phases [17, 18, 20, 23, 25, 27, 28] could be interpreted successully in terms of the occurrence of tetrahedral order [12, 14, 15, 31, 32]. In parallel tetrahedral order has been incorporated in the dynamic description of movable and deformable active particles which are used as models for self-propelled microorganisms in biological applications [33-35]. Quite recently there is also growing interest in clarifying various mathematical aspects of tetrahedral order in two and three spatial dimensions [36–38]. Last year it has been pointed out [31] that the observed macroscopic chiral

An important issue so far not considered is the influence of a magnetization on tetrahedral liquid-crystalline phases. This influence is interesting from a symmetry point of view, because the magnetization is odd under time-reversal and in a ferromagnet with a spontaneous magnetization the ground state breaks time-reversal symmetry.

In the magnetic domain of soft matter physics Brochard and de Gennes predicted in their seminal work [42], the existence of ferromagnetic nematics and ferromagnetic cholesterics in the domain of liquid crystals. Simultaneously first experimental efforts along these lines started immediately [43], but they were not leading to ferromagnetic nematic phases, since suitably characterized and uniform magnetic nano-particles did not exist in 1970. Only about five years ago the group around Lisjak and Mertelj reported the successful synthesis and characterization of a homogeneous phase of a truly ferromagnetic nematic [44]. This is of particular interest, since this material represents the first liquid multi-ferroic system at room temperature. In addition to the director characterizing spontaneously broken rotational symmetry, a truly ferromagnetic phase breaks time-reversal symmetry and rotational symmetry in spin space. Several synthetic and static investigations also involving the phase transition to the isotropic phase followed quickly [45-48]. Biaxial ferromagnetic nematics have also been reported quite recently [49]. An earlier Landau investigation of the phase transitions involved [50] could be used to interpret some of the experimental results [44]. Building on the macroscopic dynamic work of Jarkova et al. [51, 52], the approach of macroscopic dynamics for truly ferromagnetic nematics has been used successfully recently to describe

domains in optically isotropic partially fluid systems [39–41] can be interpreted naturally in terms of tetrahedral order provided a transient network is assumed. For a recent review of tetrahedral order in liquid crystals we refer to [32].

<sup>\*</sup>tilen.potisk@uni-bayreuth.de

quantitatively dynamic experimental results [53, 54] and to make further experimentally testable predictions [55]. For a recent review on truly ferromagnetic nematics we refer to Ref. [56].

More recently ferromagnetic cholesterics, for which one has as a liquid-crystalline solvent a nematic containing chiral molecules, have been synthesized and characterized [57–59]. While ferromagnetic cholesterics turn out to have many different textures and defects depending on the ratio of cholesteric pitch and sample thickness, also simple textures could be obtained recently [59]. The latter observation will open the door to apply a recent macroscopic description of ferrocholesterics [60] to this rather complex system.

Our goal in the present paper is to analyze how to detect the possible presence of tetrahedral/octupolar order in ferromagnetic nematics, a system composed of nonchiral molecules. We focus our investigations on macroscopic properties in the static as well as in the dynamic domain.

The paper is organized as follows. In Sec. II we give a Landau analysis and determine the macroscopic variables. In Sec. III we present the thermodynamics and the static properties of ferromagnetic nematics followed in Sec. IV by the derivation of the macroscopic dynamic equations. In Sec. V we make suggestions how to detect the presence of tetrahedral order statically and dynamically followed by brief conclusions and a perspective.

# II. LANDAU ENERGIES AND MACROSCOPIC VARIABLES

In this section we discuss the properties of a phase, for which one allows for the additional presence of a tetrahedral order parameter in a ferromagnetic nematic phase. We use a Landau energy approach to discuss the possible ground states. We then identify all macroscopic variables for a selected ground state.

### A. Landau energy considerations

As variables in a Landau expansion we take into account, in addition to the magnetization,  $M_i$ , and the quadrupolar order parameter,  $Q_{ij}$ , the tetrahedral order parameter,  $T_{ijk}$ , a fully symmetric third rank tensor [1]

$$T_{ijk} = T_0 \sum_{\zeta=1}^{4} n_i^{\zeta} n_j^{\zeta} n_k^{\zeta}, \tag{1}$$

where the vectors  $\mathbf{n}^{\zeta}$  ( $\zeta=1,2,3,4$ ) span a tetrahedron and the order parameter  $T_0$  describes the strength of the tetrahedral order. We assume the strength of the tetrahedral order,  $T_0$ , as constant, which is a good approximation far away from a phase transition, where the tetrahedral order vanishes.

Tetrahedral order fully breaks rotational symmetry of isotropic space. However, in the absence of any orienting external field or boundary the actual orientation of the tetrahedron is arbitrary: Any homogeneous rotation of the tetra-

hedron leads to a distinct, but energetically identical equilibrium state. These are the three Goldstone modes that appear as (symmetry) variables in the hydrodynamic description. In that respect, tetrahedral order is analogous to the case of biaxial nematic liquid crystals [61, 62].

The nematic (quadrupolar) order parameter is described by a symmetric traceless second rank tensor  $Q_{ij}=\frac{1}{2}S(3n_in_j-\delta_{ij})$  [63]. The quantity S is a scalar order parameter, which describes the strength of the orientational ordering. It is zero in the isotropic phase, where the molecules are randomly oriented, while it is equal to 1 if on average all the molecules point in the same direction. The unit vector  ${\bf n}$  is the director field and describes the orientation of the nematic ordering. Without loss of generality one can assume  $n_in_i=1$ . It should be emphasized that due to the equivalence  ${\bf n}\to -{\bf n}$  all of the equations should be invariant with respect to this transformation.

The Landau energy has, in addition to the terms already present in a magnetic tetrahedral phase [64], also the Landau energy expressions for a pure nematic phase and the various coupling terms between the magnetization, the quadrupolar and the tetrahedral order parameter. These coupling terms read

$$F^{C} = F^{QM} + F^{QT} + F^{MT} + F^{QTM}, (2)$$

with  $F^{QM}$  being the same as for the ferromagnetic nematic phase, (see Ref. [50]):

$$F^{QM} = \frac{\gamma}{2} M_i M_j Q_{ij} + \frac{\delta_1}{2} M_k M_k Q_{ij} Q_{ji} + \frac{\delta_2}{2} M_i M_k Q_{ij} Q_{kj}.$$
(3)

The second term in Eq. (2) was investigated in Ref. [14]:

$$F^{QT} = d_1 Q_{il} Q_{jm} T_{ilk} T_{jmk} + \frac{d_2}{2} (Q_{im} Q_{jl} + Q_{ij} Q_{lm}) T_{ilk} T_{jmk}.$$
 (4)

There it was found that if  $d_1+d_2>0$  the phase is of  $D_{2d}$  symmetry where the director points along one of the improper  $\bar{4}$  axes, whereas if  $d_1+d_2<0$  the phase is of  $C_{3v}$  symmetry and the director points along one of the tetrahedral vectors. The cross coupling terms between the magnetization, the quadrupolar and the tetrahedral order parameter,  $F^{QTM}$ , are of quintic order:

$$F^{QTM} = c_1 Q_{il} M_j M_m T_{ilk} T_{jmk} + \frac{c_2}{2} (Q_{ij} M_m + Q_{im} M_j) M_l T_{ilk} T_{jmk}.$$
 (5)

One can see the similarities of Eqs. (5) and (4). This is due to the fact the free energy should be even in the magnetization to ensure invariance with respect to time-reversal symmetry.

As a first step we assume the director is fixed with respect to the tetrahedral structure and points along the z axis,  $\mathbf{n} = \hat{\mathbf{e}}_z$ . To find the orientation of  $\mathbf{M}$  in the ground state, we vary the azimuthal and the polar angle, defined by  $\mathbf{M} = M_0(\cos\varphi\sin\psi,\sin\varphi\sin\psi,\cos\psi)$ . There are five different solutions of the angles, that correspond to a minimum of the free energy. The first solution is where the magnetization points along the director field  $\mathbf{M} \parallel \mathbf{n}$ .

Next also the energy term coupling the magnetization and the tetrahedral order parameter enters the picture

$$F^{MT} = aT_{ilk}T_{jmk}M_iM_lM_jM_m. (6)$$

Two of the other solutions correspond to the magnetization lying in the plane perpendicular to the director. One of these solutions is stable if a>0 and the magnetization points along one of the other two improper  $\bar{4}$  axes. The other solution is stable if a<0 and the magnetization lies within one of the mirror planes. For the last two solutions the angle  $\psi$  depends on the value of coefficients in the expression for the free energy, Eq. (2).

In the following we focus on the solution where the director and the magnetization are parallel in the ground state as depicted in Fig.1.

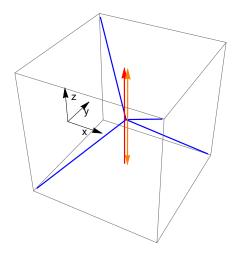


FIG. 1: The ground state of the system showing the magnetization (red) and the director (shown as a double-headed arrow in orange) along one of the improper  $\bar{4}$  axes of the tetrahedron (blue).

## B. Macroscopic variables

To derive the macroscopic equations of a particular macroscopic system one must first identify the relevant macroscopic variables based on a specific ground state as input. In addition to the conserved variables characteristic of an isotropic fluid – the mass density  $\rho$ , the energy density  $\varepsilon$  and the density of linear momentum  $\mathbf{g}$  – one must address the issue of variables associated with spontaneously broken continuous symmetries and of macroscopic variables, which relax on a long, but finite time scale [65–67].

Inspired by the experimental results available on ferromagnetic nematic liquid-crystalline phases [44–48, 53, 54, 56], we will assume that in the ground state the director  ${\bf n}$  and the magnetization  ${\bf M}$  are parallel. In addition, we assume that a>0 and that the magnetization points along one of the improper  $\bar{4}$  axes of the tetrahedron. Thus the situation considered in the following is that of a nematic phase with  $D_{2d}$  symmetry [14] with an additional spontaneous magnetization parallel to the nematic director in the ground state.

Rotations of the tetrahedral structure can be described by a projection,  $\delta\Gamma_i$ , with [14]

$$\delta\Gamma_i = \frac{1}{4\tilde{\alpha}} \epsilon_{ipq} T_{pkl} \delta T_{qkl}, \tag{7}$$

where  $\delta T_{qkl}$  is the deviation of the tetrahedral order parameter from the equilibrium one,  $\delta T_{qkl} = T_{qkl} - T_{qkl}^{\rm eq}$ . We use the normalization [14, 32]  $T_{ikl}T_{jkl} = \tilde{\alpha}\delta_{ij}$ , with  $\tilde{\alpha} = (32/27)T_0^2$ . This relation can be inverted [14, 32] to yield

$$\delta T_{alk} = 2\epsilon_{ipq} T_{pkl} \delta \Gamma_i. \tag{8}$$

In this paper we focus on the importance and influence of tetrahedral order on a ferromagnetic nematic phase. The hydrodynamic orientational degrees of freedom associated with the director are characterized by the variations of the director field,  $\delta n_i$ , with  $n_i \delta n_i = 0$ . The magnetic order is due to the existence of a spontaneous magnetization,  $\mathbf{M}$ . It describes the strength of magnetic order by the order parameter  $M \equiv |\mathbf{M}|$ , and its orientation by the unit vector  $\mathbf{m} = \mathbf{M}/M$ . The former is neither connected to a Goldstone mode, nor to a conservation law, and therefore does not give rise to a genuine hydrodynamic variable. Nevertheless, its relaxation time can be large enough to be relevant in the hydrodynamic regime, and we will keep  $\delta M \equiv M - M_0$ , with  $M_0$  the equilibrium magnetization, as a macroscopic variable.

Since we will assume a rigid coupling between the director, the tetrahedral order and the magnetization in the ground state we have as hydrodynamic variables the director variations,  $\delta n_i$  and the quantity  $\delta\Omega\equiv n_i\delta\Gamma_i$ , with  $\delta\Gamma_i$  given by Eq. (7), which describes a rotation of the tetrahedral structure about the equilibrium director and thus also about the magnetization in equilibrium. In addition we have as macroscopic variables  $\delta m_i$  and  $\delta M$ .

### III. THERMODYNAMICS AND STATIC PROPERTIES

To describe the statics of the tetrahedral ferromagnetic nematic phase we proceed along the same lines as for ferronematics [51, 52] and ferromagnetic nematics [53–55]. We use the conservation laws for density  $\rho$ , energy density  $\varepsilon$ , density of linear momentum g and particle concentration c. For the magnetic degrees of freedom we have the variation of the modulus  $\delta M$  and the variations of the magnetic unit vector m,  $\delta m_i$ . In addition we have the director degrees of freedom,  $\delta n_i$ . As discussed above there is now the additional variable  $\delta \Omega$  describing rotations of the tetrahedral structure about the equilibrium director. To satisfy Maxwell's equations the magnetic induction  $\mathbf{B}$  must be considered as well.

Throughout this paper we assume local thermodynamic equilibrium. Changes of the macroscopic variables listed above are then related to changes of the total energy density via the Gibbs relation,

$$df = T d\sigma + \mu d\rho + v_i dg_i + \mu_c dc + h^M dM + h_i^{m'} dm_i$$

$$+ \Psi_{ij}^m d\nabla_j m_i + h^{\Omega'} d\Omega + \Psi_i^{\Omega} d\nabla_i \Omega + h_i^{n'} dn_i$$

$$+ \Phi_{ii}^n d\nabla_j n_i$$
(9)

which is the local formulation of the first law of thermodynamics. In Eq. (9)  $h_i^{m'}$ ,  $h^{\Omega'}$  and  $h_i^{n'}$  are the thermodynamic conjugate forces to  $m_i$ ,  $\Omega$  and  $n_i$  and are given explicitly in eqs.(37), (42) and (39).

In the static behavior only the combinations

$$\begin{array}{lll} h_i^M &=& h_i^{M'} - \nabla_j \Psi_{ij}^M, & h_i^n = h_i^{n'} - \nabla_j \Phi_{ij}^n, & \text{and} \\ h^\Omega &=& h^{\Omega'} - \nabla_i \Psi_i^\Omega \end{array} \tag{10}$$

enter the picture. In addition, in the absence of boundaries or orienting fields,  $h_i^{m'} = h_i^{n'} = h^{\Omega\prime} = 0$  to guarantee that changes in the orientation do not change the energy.

The thermodynamic conjugates are prefactors of the differentials in Eq. (9), it i.e. temperature T, chemical potential  $\mu$ , velocity  $v_i$ , osmotic pressure (divided by the pressure)  $\mu_c$ , so-called molecular fields of the magnetic order  $h^M$ , of the magnetization rotations  $h^m_i$ , of rotations about the director  $h^\Omega$ , and of director rotations  $h^n_i$ . They (or their gradients) act as thermodynamic forces in the dynamics (depending whether they are zero or finite in equilibrium).

Rotational invariance leads for eq. (9) to the additional requirement

$$0 = \epsilon_{ijk} (h_i^{m'} m_j + h_i^{n\prime} n_j + \Psi_i^{\Omega} \nabla_j \Omega + \Psi_{li}^{m} \nabla_j m_l + \Psi_{il}^{m} \nabla_l m_j + \Phi_{li}^{n} \nabla_i n_l + \Phi_{il}^{n} \nabla_l n_j) - h^{\Omega} m_k$$
(11)

where the last term is due to the fact that  $\Omega$  is not a scalar quantity and is not invariant under rotations. For details cf. [14, 64].

The material tensors will be constructed using the invariants  $n_i$ ,  $\delta_{ij}^{\perp n} = \delta_{ij} - n_i n_j$ ,  $\epsilon_{ijk}$  and  $T_{ijk}$ . The magnetization  $M_i$  does not define an extra, independent preferred direction, and will occur only, when its specific time-reversal behavior is crucial. Since all material parameters can be arbitrary functions of  $M^2$ , only linear contributions of  $M_i$  will explicitly show up in the material tensors. This is in the same spirit as for the case without tetrahedral order [51–54].

The thermodynamic conjugates are defined as partial derivatives of the total energy density with respect to the appropriate variable. Thus they follow from a total energy functional that can be written as

$$f = f_0 + f_{\rm el} + f_M + f_{Marad} + f_{\rm lin},$$
 (12)

where  $f_0$  is the total energy of an isotropic liquid mixture,  $f_{\rm el}$  contains the gradient terms associated with the director, the orientation of the magnetization and with  $\Omega$ ,  $f_M$  is the spatially homogeneous magnetic energy including external magnetic fields and  $f_{Mgrad}$  contains gradients of M while  $f_{\rm lin}$  is linear in gradients.

When constructing the explicit forms of the various energy contributions one can make use of the totally antisymmetric symbol,  $\epsilon_{ijk}$ , the tetrahedral structure  $T_{ijk}$  and the director  $n_i$ . One has to note that  $T_{ijk}$  is odd under spatial inversion and  $n_i$  is even under time-reversal, while  $m_i$  is odd under time-reversal. In particular we find [67]

$$f_0 = \frac{T}{2C_V} (\delta \sigma)^2 + \frac{1}{2\rho^2 \kappa_s} (\delta \rho)^2 + \frac{\gamma}{2} (\delta c)^2 + \frac{1}{\rho \alpha_s} (\delta \sigma) (\delta \rho)$$

+ 
$$\beta_{\sigma}(\delta c)(\delta \sigma) + \beta_{\rho}(\delta c)(\delta \rho) + \frac{g_i^2}{2\rho}$$
. (13)

containing the standard thermodynamic susceptibilities, such as specific heat  $C_V$ , compressibility  $\kappa_s$ , thermal expansion  $\alpha_s$  etc

In general, inhomogeneous rotations of  $n_i$ ,  $m_i$  and  $\Omega$  must increase the total energy

$$f_{\text{el}} = \frac{1}{2} K_{ijkl}^{m} (\nabla_{j} m_{i}) (\nabla_{l} m_{k}) + \frac{1}{2} K_{ijkl} (\nabla_{j} n_{i}) (\nabla_{l} n_{k})$$

$$+ K_{ijkl}^{nm} (\nabla_{i} n_{j}) (\nabla_{k} m_{l}) + \frac{1}{2} K_{ij}^{\Omega} (\nabla_{i} \Omega) (\nabla_{j} \Omega)$$

$$+ C_{ijk}^{n\Omega} (\nabla_{i} \Omega) (\nabla_{k} n_{j}) + C_{ijk}^{M_{i}\Omega} (\nabla_{i} \Omega) (\nabla_{k} m_{j})$$

$$+ \Pi_{ijk}^{c} (\nabla_{i} c) (\nabla_{k} n_{j}) + \Pi_{ijk}^{\sigma} (\nabla_{i} \sigma) (\nabla_{k} n_{j})$$

$$+ \Pi_{ijk}^{\rho} (\nabla_{i} \rho) (\nabla_{k} n_{j}) + \Pi_{ijk}^{M_{i}c} (\nabla_{i} c) (\nabla_{k} m_{j})$$

$$+ \Pi_{ijk}^{M_{i}\sigma} (\nabla_{i} \sigma) (\nabla_{k} m_{j}) + \Pi_{ijk}^{M_{i}\rho} (\nabla_{i} \rho) (\nabla_{k} m_{j})$$

$$+ (\nabla_{i} \Omega) (C_{ij}^{c} \nabla_{j} c + C_{ij}^{\sigma} \nabla_{j} \sigma + C_{ij}^{\rho\Omega} \nabla_{j} \rho)$$
 (14)

with the rotational stiffness (or rotational elastic) tensors

$$K_{ijkl}^{m} = K_1^{m} \delta_{ij}^{\perp} \delta_{kl}^{\perp} + K_2^{m} n_p n_q \varepsilon_{ijp} \varepsilon_{klq}$$

$$+ K_3^{m} n_j n_l \delta_{ik}^{\perp} + K_4^{m} n_p n_q T_{ijp} T_{klq}$$

$$(15)$$

$$K_{ijkl} = K_1 \delta_{ij}^{\perp} \delta_{kl}^{\perp} + K_2 n_p n_q \varepsilon_{ijp} \varepsilon_{klq}$$
$$+ K_3 n_i n_l \delta_{ik}^{\perp} + K_4 n_p n_q T_{iip} T_{klq}$$

$$+K_{3}n_{j}n_{l}\delta_{ik}^{\perp} + K_{4}n_{p}n_{q}T_{ijp}T_{klq}$$

$$K_{ijkl}^{mn} = K^{mn}\delta_{il}^{\perp}(n_{i}M_{k} + n_{k}M_{i})$$
(16)

$$K_{ijkl}^{\Omega} = K^{\Omega} \delta_{jl}^{1} (h_{i}M_{k} + h_{k}M_{i}) \tag{17}$$

$$K_{ij}^{\Omega} = K_{i}^{\Omega} \delta_{ij}^{1} + K_{\parallel}^{\Omega} n_{i} n_{j} \tag{18}$$

$$C_{ijk}^{n\Omega} = C_{\perp}(\varepsilon_{jkp}n_i + \varepsilon_{jip}n_k)n_p \tag{19}$$

$$C_{ijk}^{M_i\Omega} = C_2^{M_i\Omega}(\varepsilon_{jkp}n_i + \varepsilon_{jip}n_k)M_p$$

$$+C_3^{M_i\Omega}(\varepsilon_{jkp}M_i + \varepsilon_{jip}M_k)n_p \tag{20}$$

$$\Pi_{ijk}^{\lambda} = \Pi^{\lambda} (n_i \delta_{jk}^{\perp} + n_k \delta_{ij}^{\perp})$$
 (21)

$$\Pi_{ijk}^{M_i\lambda} = \Pi^{M_i\lambda}(M_i\delta_{jk}^{\perp} + M_k\delta_{ij}^{\perp})$$
 (22)

$$C_{ij}^{\lambda\Omega} = C_{\perp}^{\lambda} n_k T_{kps} (\varepsilon_{irs} T_{jpr} + \varepsilon_{jrs} T_{ipr})$$
 (23)

where  $\lambda \in \{\sigma, \rho, c\}$ .

The structure of  $f_{\rm el}$  bears some similarity with the gradient energy in the  ${\rm D}_{2d}$  phase [68] and contains four coefficients each related to bending distortions of the orientation of the magnetization and the director. In addition there are two coefficients related to inhomogeneous rotations about the director and one mixed one. We emphasize that there is only one gradient term coupling the gradients of the director with those of  $m_i$ . In addition, there are cross-couplings of the inhomogeneous rotations of  $\Omega$  with gradients of the scalar conserved variables. Also note that the contribution  $\sim C_{\perp}$  [69], which couples gradient of  $n_i$  and of  $\Omega$  is associated with  $\nabla \times {\bf n}$ .

The magnetic part of the free energy homogeneous in the magnetization in Eq. (12) reads

$$f_M = -M_i H_i - \frac{1}{2} A_1 (m_i n_i)^2 + \frac{1}{2} \alpha M^2 + \frac{1}{4} \beta M^4.$$
 (24)

This expression is derived taking into account the static magnetic Maxwell equations.  $\alpha$  and  $\beta$  are expansion coefficients

in a Landau expansion for M and where the contribution  $\sim A_1$  describes the coupling between  $m_i$  and  $n_i$ . The derivation parallels very closely that given in Ref.[52] and quite recently in Ref.[64].  $f_M$  is the Legendre transformed magnetic energy containing the magnetic field  $\mathbf{H}$ . The ferromagnetic coupling in  $f_M$  leads to the parallel equilibrium orientation of the magnetization along an external magnetic field. As a result a homogeneous external field is compatible with a homogeneous combined magnetization/tetrahedral structure in the phase considered here: ferromagnetic nematic with additional tetrahedral order. However, the degeneracy of the (combined) orientation of the magnetization and the tetrahedral structure is partially lifted and only the orientation of the structure perpendicular to the field (and  $\mathbf{m}$ ) is still arbitrary.

For the magnetic gradient energy we find

$$f_{Mgrad} = \frac{1}{2} K_{ij}^{M}(\nabla_{i}M)(\nabla_{j}M) + C_{ij}^{M\Omega}(\nabla_{i}M)(\nabla_{j}\Omega)$$

$$+ K_{ijk}^{Mm}(\nabla_{i}M)(\nabla_{j}m_{k}) + K_{ijk}^{Mn}(\nabla_{i}M)(\nabla_{j}n_{k})$$

$$+ (\nabla_{i}M)(\Pi_{ij}^{cM}\nabla_{j}c + \Pi_{ij}^{\sigma M}\nabla_{j}\sigma + \Pi_{ij}^{\rho M}\nabla_{j}\rho)$$
 (25)

with

$$K_{ij}^{M} = K_{\parallel}^{M} \delta_{ij}^{\perp} + K_{\parallel}^{M} n_{i} n_{j}$$
 (26)

$$C_{ij}^{M\Omega} = C_{\parallel} n_k T_{kps} (\epsilon_{irs} T_{jpr} + \epsilon_{jrs} T_{ipr})$$
 (27)

$$K_{ijk}^{Mm} = K^{Mm} (M_j \delta_{ik}^{\perp} + M_i \delta_{jk}^{\perp})$$
 (28)

$$K_{ijk}^{Mn} = K^{Mn}(n_i \delta_{ik}^{\perp} + n_i \delta_{ik}^{\perp})$$
 (29)

$$\Pi_{ij}^{\lambda M} = \Pi_{\perp}^{\lambda M} \delta_{ij}^{\perp} + \Pi_{\parallel}^{\lambda M} n_i n_j \tag{30}$$

where  $\lambda \in \{\sigma, \rho, c\}$ . There are two stiffness coefficients  $(K_{\perp}^{M}, K_{\parallel}^{M})$  related to distortions of M. cross-couplings

between distortions of M and inhomogeneous rotations of, and about the director, are described by one coefficient each  $(K^{Mn})$  and  $C_{\parallel}$  respectively), while there are in total six coefficients  $(\Pi_{\perp,\parallel}^{\lambda M})$  connected to the coupling of gradients of M with gradients of the scalar conserved variables. Finally we note that we have kept in Eq. (25) one term linear in the magnetization  $M_i$ :  $K^{Mm}$ .

The last energy contribution we are discussing here is the linear gradient energy

$$f_{\text{lin}} = \xi^M T_{ijk} M_i(\nabla_j m_k) + \xi^n T_{ijk} n_i(\nabla_j n_k). \tag{31}$$

This expression is identical to the linear gradient term in the  $D_{2d}$  phase [14] for the director  $n_i$ . In addition it also contains the analogous linear gradient term, when one uses  $m_i$  instead of the director  $n_i$ . These two linear gradient terms are allowed due to the presence of tetrahedral order, which breaks parity. The present system appears to be the first one for which two of these linear gradient terms exist: one associated with the nematic director and one associated with the direction of the magnetization. As a consequence, the ground state might not be homogeneous, resembling the case of added chirality to nematic liquid crystals. In fact these terms are well-known by now to give rise to ambidextrous helicity [14, 15, 31, 32]. In case one can obtain sufficiently large domains of either hand in a ferromagnetic nematic liquid crystal composed of nonchiral constituents, this would form a rather obvious evidence of the presence of tetrahedral order. Naturally an observation in the visible range would be most attractive.

For completeness we list the expressions for the thermodynamic conjugates that follow from the energy contributions introduced above

$$v_i = \frac{1}{\rho} g_i \tag{32}$$

$$\delta T = \frac{T}{C_V} \delta \sigma + \frac{1}{\rho \alpha_s} \delta \rho + \beta_\sigma \delta c - \nabla_i \left( \Pi_{ijk}^\sigma \nabla_k n_j + \Pi_{ij}^{\sigma M} \nabla_j M + C_{ij}^{\sigma \Omega} \nabla_j \Omega - \Pi_{ijk}^{M_i \sigma} \nabla_k m_j \right)$$
(33)

$$\delta\mu = \frac{1}{\rho^2 \kappa_s} \delta\rho + \frac{1}{\rho \alpha_s} \delta\sigma + \beta_\rho \delta c - \nabla_i \left( \Pi_{ijk}^\rho \nabla_k n_j + \Pi_{ij}^{\rho M} \nabla_j M + C_{ij}^{\mu \Omega} \nabla_j \Omega - \Pi_{ijk}^{M_i \rho} \nabla_k m_j \right)$$
(34)

$$\delta\mu_c = \gamma\delta c + \beta_\sigma\delta\sigma + \beta_\rho\delta\rho - \nabla_i \left( \Pi^c_{ijk} \nabla_k n_j + \Pi^{cM}_{ij} \nabla_j M + C^{c\Omega}_{ij} \nabla_j \Omega - \Pi^{M_ic}_{ijk} \nabla_k m_j \right)$$
(35)

$$h^{M} = -m_{i}H_{i} + \alpha M + \beta M^{3} + \left(C_{2}^{M_{i}\Omega}[\varepsilon_{jkp}n_{i} + \varepsilon_{jip}n_{k}]m_{p} + C_{3}^{M_{i}\Omega}[\varepsilon_{jkp}m_{i} + \varepsilon_{jip}m_{k}]n_{p}\right)(\nabla_{i}\Omega)(\nabla_{k}m_{j})$$
$$-\nabla_{i}\left(K_{ij}^{M}\nabla_{j}M + C_{ij}^{M\Omega}\nabla_{j}\Omega + K_{ijk}^{Mm}\nabla_{j}m_{k} - K_{ijk}^{Mn}\nabla_{j}n_{k} - \Pi_{ij}^{\sigma M}\nabla_{j}\sigma + \Pi_{ij}^{\rho M}\nabla_{j}\rho + \Pi_{ij}^{cM}\nabla_{j}c\right)$$

$$+(m_{i}\delta_{jk}^{\perp}+m_{k}\delta_{ij}^{\perp})\left(\Pi^{M_{i}\rho}\nabla_{i}\rho+\Pi^{M_{i}\sigma}\nabla_{i}\sigma+\Pi^{M_{i}c}\nabla_{i}c\right)\left(\nabla_{k}m_{j}\right)$$
(36)

$$h_i^{m'} = -MH_i - A_1(m_i n_j) n_i (37)$$

$$\Psi_{ij}^{m} = K_{ijkl}^{m} \nabla_{l} m_{k} + K_{jikl}^{nm} \nabla_{l} n_{k} + K_{kji}^{Mm} \nabla_{k} M + \Pi_{kji}^{M_{i}\sigma} \nabla_{k} \sigma + \Pi_{kji}^{M_{i}\rho} \nabla_{k} \rho + \Pi_{kji}^{M_{i}c} \nabla_{k} c + C_{kji}^{M_{i}\Omega} \nabla_{k} \Omega$$

$$(38)$$

$$h_i^{n'} = -A_1(m_j n_j) m_i (39)$$

$$\Phi_{ij}^{n} = K_{ijkl} \nabla_{l} n_{k} + C_{kij}^{n\Omega} \nabla_{k} \Omega + K_{kji}^{Mn} \nabla_{k} M + \Pi_{kij}^{\sigma} \nabla_{k} \sigma + \Pi_{kij}^{\rho} \nabla_{k} \rho + \Pi_{kij}^{c} \nabla_{k} c + K_{jikl}^{nm} \nabla_{k} m_{l}$$

$$\tag{40}$$

$$\Psi_{i}^{\Omega} = K_{ij}^{\Omega} \nabla_{j} \Omega + C_{ijk}^{\Omega\Omega} \nabla_{k} n_{j} + C_{ij}^{M\Omega} \nabla_{j} M + C_{ij}^{\sigma\Omega} \nabla_{j} \sigma + C_{ij}^{\mu\Omega} \nabla_{j} \mu + C_{ij}^{c\Omega} \nabla_{j} c + C_{ijk}^{Mi\Omega} \nabla_{k} m_{j}$$

$$\tag{41}$$

$$h^{\Omega'} = 0 (42)$$

Since the  $\delta$ 's in Eqs. (33) - (35) describe deviations from the constant equilibrium values of the appropriate variable, all expressions on the left hand side of Eqs. (32) - (42) are zero in equilibrium and can act as thermodynamic forces that drive the dynamics of the system. On the other hand, the right hand sides of all these equations have to be zero in equilibrium (Euler conditions). Note that the energy  $f_{\rm lin}$  does not enter any Euler condition (except for  $\nabla_l T_{ijk} \neq 0$ ), since it is linear in gradients of  $m_i$ .

# IV. DYNAMICS OF FERROMAGNETIC NEMATICS WITH TETRAHEDRAL ORDER

#### A. Dynamic equations

The hydrodynamic variables can be put into two different classes. There are conserved variables, like the mass density, energy density and momentum density  $\mathbf{g}$ , which are governed by conservation laws. The second class of variables corresponds to the variables associated with spontaneously broken continuous symmetries. Their dynamics is governed by balance laws. In our case we have from this class the director variations,  $\delta n_i$ , and the rotation around the director,  $\delta \Omega$ . There are some macroscopic variables, that relax on a finite but very long time scale and it is therefore sensible to include them into the macroscopic description, Ref. [67]. In our case we will consider the magnitude of the magnetization,  $M_i$ :  $\delta m_i$ .

The dynamic equations read (including the dynamic equations already given in Ref.[52]):

$$\frac{\partial}{\partial t}f + \nabla_i([f+p]v_i + j_i^f) = 0, \qquad (43)$$

$$\frac{\partial}{\partial t}\rho + \nabla_i g_i = 0, \qquad (44)$$

$$\frac{\partial}{\partial t}g_i + \nabla_j(g_iv_j + p\delta_{ij} + \sigma_{ij}^{th} + \sigma_{ij}) = 0, \qquad (45)$$

$$\frac{\partial}{\partial t}\sigma + \nabla_i(\sigma v_i + j_i^{\sigma}) = \frac{2R}{T},$$
 (46)

$$\rho \left( \frac{\partial}{\partial t} + v_j \nabla_j \right) c + \nabla_i j_i^c = 0, \qquad (47)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j\right) M + X^M = 0, \qquad (48)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j\right) m_i - \epsilon_{ijk} \omega_j m_k + X_i^m = 0, \quad (49)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j\right) \Omega - m_i \omega_i + Z = 0, \quad (50)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j\right) n_i - \epsilon_{ijk} \omega_j n_k + Y_i^n = 0, \quad (51)$$

with the vorticity  $\omega_i = (1/2)\epsilon_{ijk}\nabla_j v_k$  and the pressure p. The vorticity contributions are due to the fact that  $m_i$  and  $n_i$  transform under spatial rotations as a vector, and  $\Omega$  as a special component of a vector [32]. These terms ensure that only those rotations enter hydrodynamics that go beyond the global rotation (e.g., of the coordinate system).

In Eq. (45) we have explicitly written down the non-phenomenological part of the stress tensor,  $\sigma_{ij}^{th}$ , which is given by

$$\sigma_{ij}^{th} = \frac{1}{2} (m_i h_j^m - m_j h_i^m) + \Psi_{kj}^m \nabla_i m_k + \Psi_j^{\Omega} \nabla_i \Omega$$
$$- h^{\Omega} \epsilon_{ijk} m_k + \frac{1}{2} (n_i h_j^n - n_j h_i^n) + \Phi_{kj}^n \nabla_i n_k. \quad (52)$$

Using the condition of a rotational invariant free energy, Eq. (11), it can be brought into the form, [67]

$$2\sigma_{ij}^{th} = \Psi_{kj}^{m} \nabla_{i} m_{k} + \Psi_{ki}^{m} \nabla_{j} m_{k} + \Psi_{j}^{\Omega} \nabla_{i} \Omega + \Psi_{i}^{\Omega} \nabla_{j} \Omega + \Phi_{ki}^{n} \nabla_{j} n_{k} + \Phi_{kj}^{n} \nabla_{i} n_{k} + \nabla_{k} (m_{j} \Psi_{ik}^{m} - m_{i} \Psi_{ik}^{m} + n_{j} \Phi_{ik}^{n} - n_{i} \Phi_{ik}^{n})$$
 (53)

that guarantees angular momentum conservation [65].

The source term in the dynamic evolution equation for the entropy density, Eq. (46), is proportional to the dissipation function R representing (half of) the rate at which the heat is transferred to the microscopic degrees of freedom. The second law of thermodynamics requires R>0 for dissipative processes, while R=0 holds for the reversible parts of the currents, in which case Eq. (46) is a conservation law. Splitting the phenomenological currents  $(j_i^f,\sigma_{ij},j_i^\sigma,j_i^c,X^M,X_i^m,Z,Y_i^n)$  into the dissipative part (superscript D) and the reversible one (superscript R) the Gibbs relation Eq. (9) then leads to the condition

$$2R = -\nabla_{i}j_{i}^{fD} - j_{i}^{\sigma D}\nabla_{i}T - j_{i}^{cD}\nabla_{i}\mu_{c} - \sigma_{ij}^{D}A_{ij}$$

$$+X^{MD}h^{M} + X_{i}^{mD}h_{i}^{m} + Z^{D}h^{\Omega} + Y_{i}^{nD}h_{i}^{nD} > 0$$
(54)

for dissipative processes, where only the symmetrized velocity gradient  $2A_{ij} \equiv \nabla_i v_j + \nabla_j v_i$  enters, in order to prevent solid body rotations to produce entropy. For reversible currents, the condition

$$-\nabla_{i}j_{i}^{fR} - j_{i}^{\sigma R}\nabla_{i}T - j_{i}^{cR}\nabla_{i}\mu_{c} - \sigma_{ij}^{R}A_{ij}$$

$$+X^{MR}h^{M} + X_{i}^{mR}h_{i}^{m} + Z^{R}h^{\Omega} + Y_{i}^{nR}h_{i}^{nR} = 0$$
(55)

applies. Possible pure divergence contributions (surface terms) are put into  $j_i^f$ , but are not needed in the following. The various transport contributions in the time derivatives of Eqs. (43)-(50) are all reversible. Their zero entropy production is ensured by the non-phenomenological parts of the stress tensor  $\sigma_{ij}^{th}$  and by the pressure p.

A current is reversible, if it transforms under time-reversal in the same way as the time derivative of the appropriate variable, while the dissipative part of a current has the opposite time-reversal behavior. In the following we will discuss the dissipative and reversible dynamics separately.

To derive the dissipative parts of the phenomenological currents one first writes the dissipation function as a positive quadratic form in the thermodynamic forces taking into account that R has to be a time-reversal symmetric, scalar quantity. By taking the variational derivative of this function with respect to the chosen thermodynamic force one gets the corresponding dissipative current.

(67)

#### B. Dissipation function and dissipative currents

The dissipation function reads

$$R = \frac{1}{2} \kappa_{ij} (\nabla_{i} T) (\nabla_{j} T) + \frac{1}{2} D_{ij} (\nabla_{i} \mu_{c}) (\nabla_{j} \mu_{c}) + D_{ij}^{T} (\nabla_{i} T) (\nabla_{j} \mu_{c}) + \frac{1}{2} \nu_{ijkl}^{D} A_{ij} A_{kl} + \frac{1}{2} b_{ij}^{D} h_{i}^{m} h_{j}^{m} + \frac{1}{2} b^{M} h^{M} h^{M}$$

$$+ \frac{1}{2} b^{\Omega} h^{\Omega} h^{\Omega} + \frac{1}{2 \gamma_{1}} h_{i}^{n} \delta_{ij}^{\perp} h_{j}^{n} + \tau_{ij} A_{ij} h^{\Omega} + c_{ij}^{M} A_{ij} h^{M} + \chi_{ij}^{D} h_{j}^{m} h_{i}^{n} + c_{ijk}^{D} A_{jk} h_{i}^{m} + \lambda_{ijk}^{D} A_{jk} h_{i}^{n} + \Gamma_{ijk}^{(2)} A_{ij} \nabla_{k} T$$

$$+ \Gamma_{ijk}^{(3)} A_{ij} \nabla_{k} \mu_{c} + T_{ijk} M_{j} (\tilde{\psi}^{TD} \nabla_{k} T + \tilde{\psi}^{cD} \nabla_{k} \mu) h_{i}^{m} + T_{ijk} n_{j} \delta_{iq}^{\perp} (\psi^{TD} \nabla_{k} T + \psi^{cD} \nabla_{k} \mu) h_{q}^{n}$$

$$(56)$$

and the dissipative parts of the currents are

$$j_i^{\sigma D} = -\kappa_{ij} \nabla_j T - D_{ij}^T \nabla_j \mu_c - \tilde{\psi}^{TD} M_j T_{kji} h_k^m - \psi^{TD} n_j \delta_{kq}^{\perp} T_{kji} h_q^n - \Gamma_{kji}^{(2)} A_{kj}, \tag{57}$$

$$j_i^{cD} = -D_{ij}\nabla_j \mu_c - D_{ii}^T \nabla_j T - \tilde{\psi}^{cD} M_j T_{kji} h_k^m - \psi^{cD} n_j \delta_{ka}^\perp T_{kji} h_a^n - \Gamma_{kii}^{(3)} A_{kj}, \tag{58}$$

$$\sigma_{ij}^{D} = -\nu_{ijkl}^{D} A_{kl} - c_{kji}^{D} h_{k}^{m} - c_{ij}^{M} h^{M} - \tau_{ij} h^{\Omega} - \Gamma_{ijk}^{(2)} \nabla_{k} T - \Gamma_{ijk}^{(3)} \nabla_{k} \mu_{c} - \lambda_{kij}^{D} h_{k}^{n}, \tag{59}$$

$$X_{i}^{mD} = b_{\perp}^{D} \delta_{ij}^{\perp} h_{j}^{m} + c_{ijk}^{D} A_{jk} + \chi_{ji}^{D} h_{i}^{n} + M_{j} T_{ijk} (\tilde{\psi}^{TD} \nabla_{k} T + \tilde{\psi}^{cD} \nabla_{k} \mu_{c}), \tag{60}$$

$$X^{MD} = b^M h^M + c_{ij}^M A_{ij}, (61)$$

$$Z^{D} = b^{\Omega}h^{\Omega} + \tau_{ij}A_{ij}, \tag{62}$$

$$Y_i^{nD} = \frac{1}{\gamma_1} \delta_{ij}^{\perp} h_j^n + \chi_{ij}^D h_j^m + \lambda_{ijk}^D A_{jk} + T_{qjk} n_j \delta_{iq}^{\perp} (\psi^{TD} \nabla_k T + \psi^{cD} \nabla_k \mu)$$

$$(63)$$

where the tensors  $\kappa_{ij}, D_{ij}, D_{ij}^T$  and  $b_{ij}^D$  are of the usual uniaxial form

$$\zeta_{ij}^D = \zeta_1^D \delta_{ij}^{\perp} + \zeta_2^D n_i n_j. \tag{64}$$

while the others read

$$\Gamma_{ijk}^{(2)} = \Gamma_{21}^D \epsilon_{kpr} T_{ijp} M_r + \Gamma_{22}^D (\epsilon_{ipr} T_{kjp} M_r + \epsilon_{jpr} T_{kip} M_r)$$

$$(65)$$

$$\Gamma_{ijk}^{(3)} = \Gamma_{31}^D \epsilon_{kpr} T_{ijp} M_r + \Gamma_{32}^D (\epsilon_{ipr} T_{kjp} M_r + \epsilon_{jpr} T_{kip} M_r)$$

$$(66)$$

$$\nu_{ijkl}^{D} = \nu_{1}\delta_{ij}^{\perp}\delta_{kl}^{\perp} + \nu_{2}(\delta_{jl}^{\perp}\delta_{ik}^{\perp} + \delta_{il}^{\perp}\delta_{jk}^{\perp}) + \nu_{3}n_{i}n_{j}n_{k}n_{l} + \nu_{4}(\delta_{ij}^{\perp}n_{k}n_{l} + \delta_{kl}^{\perp}n_{i}n_{j}) \\
+ \nu_{5}(\delta_{ik}^{\perp}n_{i}n_{l} + \delta_{ik}^{\perp}n_{i}n_{l} + \delta_{il}^{\perp}n_{i}n_{k} + \delta_{il}^{\perp}n_{i}n_{k}) + \nu_{6}n_{n}n_{a}T_{ijn}T_{kla}$$

$$c_{ijk}^D = c_1^D (\epsilon_{imk} n_j + \epsilon_{imj} n_k) n_m \tag{68}$$

$$\tau_{ij} = \tau(n_i M_i + n_j M_i) \tag{69}$$

$$c_{ij}^{M} = c_2^{D} (\epsilon_{irs} T_{ipr} + \epsilon_{jrs} T_{ipr}) M_k T_{kps}$$

$$(70)$$

$$\chi_{ij}^D = \chi_2^D \delta_{ij}^{tr} M_k n_k \tag{71}$$

$$\lambda_{ijk}^{D} = \lambda_{1}^{D} \delta_{iq}^{tr} (\epsilon_{pjq} M_{p} n_{k} + \epsilon_{pkq} M_{p} n_{j}) + \lambda_{2}^{D} n_{p} (M_{j} \epsilon_{ipk} + M_{k} \epsilon_{ipj}) + \lambda_{3}^{D} M_{q} n_{q} n_{p} (n_{j} \epsilon_{ipk} + n_{k} \epsilon_{ipj})$$

$$(72)$$

#### C. Reversible currents

The reversible parts of the currents do not follow from any potential, but can be derived by requiring that the entropy production R in Eq. (54) is zero

$$j_{i}^{\sigma R} = -\kappa_{ij}^{R} \nabla_{j} T - D_{ij}^{TR} \nabla_{j} \mu_{c} + \psi_{ij}^{T} h_{j}^{m} + \Gamma_{kji}^{T} A_{jk} + \xi_{ij}^{Tn} h_{j}^{n}$$
(73)

$$j_{i}^{cR} = -D_{ij}^{R} \nabla_{j} \mu_{c} + D_{ij}^{TR} \nabla_{j} T + \psi_{ij}^{c} h_{j}^{m} + \Gamma_{kji}^{c} A_{jk} + \xi_{ii}^{cn} h_{j}^{n}$$
(74)

$$\sigma_{ij}^{R} = -\frac{1}{2}\lambda_{kji}h_{k}^{n} - \nu_{ijkl}^{R}A_{kl} - c_{kij}^{R}h_{k}^{m} - c_{ij}^{R}h^{M}$$
$$-\Gamma_{ijk}^{T}\nabla_{k}T - \Gamma_{ijk}^{c}\nabla_{k}\mu_{c} - \tau_{ij}^{R}h^{\Omega}$$
(75)

$$X_i^{mR} = b_{ij}^R h_j^m - c_{ijk}^R A_{jk} + \psi_{ji}^T \nabla_j T$$

$$+\psi_{ji}^{c}\nabla_{j}\mu_{c} + \chi^{R}(\mathbf{n} \times \mathbf{h}^{n})_{i}$$

$$X^{MR} = -c_{ij}^{R}A_{ij}$$
(76)

$$Z^R = -\tau_{ij}^R A_{ij} \tag{78}$$

$$Y_i^{nR} = (\gamma_1^{-1})_{ij}^R h_j^n + \lambda_{ijk} A_{jk}$$
  
+  $\xi_{ji}^{Tn} \nabla_j T + \xi_{ji}^{cn} \nabla_j c + \chi^R (\mathbf{n} \times \mathbf{h}^m)_i$  (79)

where the tensors  $\kappa^R_{ij}$ ,  $D^{TR}_{ij}$ ,  $D^R_{ij}$ ,  $b^R_{ij}$  and  $(\gamma^{-1}_1)_{ij}$  are all of the form

$$\kappa_{ij}^R = \kappa_1^R \epsilon_{ijk} M_k + \kappa_2^R \epsilon_{ijk} n_k n_p M_p \tag{80}$$

and the other tensors read

$$\psi_{ij}^{c,T} = \psi^{c,T} \epsilon_{ipr} T_{jpk} n_k n_r \tag{81}$$

$$c_{ijk}^{R} = c_{1}^{R}(M_{i}\delta_{ik}^{\perp} + M_{k}\delta_{ij}^{\perp}),$$
 (82)

$$c_{ij}^R = c_{\perp}^R \delta_{ij}^{\perp} + c_{\parallel}^R n_i n_j \tag{83}$$

$$\tau_{ij}^{R} = \tau^{R} (\epsilon_{irs} T_{jpr} + \epsilon_{jrs} T_{ipr}) n_k T_{kps}$$
 (84)

$$\xi_{ij}^{Tn} = \xi^{Tn} M_k n_r \varepsilon_{ipr} T_{ipk} \tag{85}$$

$$\xi_{ii}^{cn} = \xi^{cn} M_k n_r \varepsilon_{ipr} T_{ipk} \tag{86}$$

$$\lambda_{ijk} = \lambda(\delta_{ij}^{\perp} n_k + \delta_{ik}^{\perp} n_j) \tag{87}$$

$$\Gamma_{kii}^{T,c} = T_{qjk} (\Gamma_1^{TR,cR} \delta_{qi}^{\perp} + \Gamma_2^{TR,cR} n_q n_i)$$
 (88)

It is straightforward to check that there is no linearly independent reversible coupling of  $n_i$  and  $m_i$  containing  $T_{ijk}$  quadratically in addition to the contribution  $\sim \chi^R$ . We note that  $\xi_{ij}^{Tn}$  and  $\xi_{ij}^{cn}$  are odd under parity, time-reversal and  $\mathbf{n} \to -\mathbf{n}$  symmetry. This type of coupling has not been given before and its possible experimental consequences will be discussed in the next section.

The reversible analog of the viscosity tensor has five components

$$\nu_{ijkl}^{R} = \nu_{1}^{R} [\epsilon_{ikp} n_{j} n_{l} + \epsilon_{ilp} n_{j} n_{k} 
+ \epsilon_{jlp} n_{i} n_{k} + \epsilon_{jkp} n_{i} n_{l}] n_{p} n_{m} M_{m} 
+ \nu_{2}^{R} [\epsilon_{ikp} n_{j} n_{l} + \epsilon_{ilp} n_{j} n_{k} 
+ \epsilon_{jlp} n_{i} n_{k} + \epsilon_{jkp} n_{i} n_{l}] M_{p} 
+ \nu_{3}^{R} [\epsilon_{ikp} \delta_{jl} + \epsilon_{ilp} \delta_{jk} + \epsilon_{jlp} \delta_{ik} + \epsilon_{jkp} \delta_{il}] n_{p} n_{m} M_{m} 
+ \nu_{4}^{R} [\epsilon_{ikp} \delta_{jl} + \epsilon_{ilp} \delta_{jk} + \epsilon_{jlp} \delta_{ik} + \epsilon_{jkp} \delta_{il}] M_{p} 
+ \nu_{5}^{R} [\epsilon_{ikp} (M_{j} n_{l} + M_{l} n_{j}) + \epsilon_{ilp} (M_{j} n_{k} + M_{k} n_{j}) 
+ \epsilon_{jlp} (M_{i} n_{k} + M_{k} n_{i}) + \epsilon_{jkp} (M_{i} n_{l} + M_{l} n_{i})] n_{p}$$
(89)

This fourth order tensor is antisymmetric in the exchange of the first pair of indices with the second one, thus guaranteeing zero entropy production.

Due to the presence of a tetrahedral order parameter one has dissipative dynamic cross-couplings of the temperature and the concentration gradients with the magnetization or the director field. This is in principle also possible in the ferromagnetic cholesteric phase.

If one applies a temperature or a concentration gradient to the sample of a tetrahedral ferromagnetic nematic phase, one can induce flow via both the dissipative and reversible currents.

#### V. SUGGESTIONS FOR EXPERIMENTS

In this section we discuss various experimental set-ups that can reveal selected static and dynamic cross-coupling effects due to the presence of tetrahedral order in ferromagnetic nematics.

#### A. Ambidextrous helical domains

In Sec. III we already briefly discussed the linear gradient energy

$$f_{\text{lin}} = \xi^M T_{ijk} M_i(\nabla_j m_k) + \xi^n T_{ijk} n_i(\nabla_j n_k).$$
 (90)

We note that these two terms can only arise for a system with broken parity. In addition,  $\xi^M$  and  $\xi^n$  can have either sign, since they are linear gradient terms. To study their consequences we perform an analysis, which closely resembles that for  $D_{2d}$  nematics given in Ref.[14]. That is we look for a helical state, which has lower energy than the homogeneous state. As a result of this analysis we obtain an energy reduction due to the two linear gradient terms, which takes the form

$$\Delta f = \frac{8}{27} T_0^2 \frac{(\xi^n + \xi^M M_0)^2}{K_2 + K_2^m}.$$
 (91)

which yields for the helical wave vector

$$q_0 = -\frac{4}{3\sqrt{3}} T_0 \frac{(\xi^n + \xi^M M_0)}{K_2 + K_2^m}.$$
 (92)

We point out that the cross-coupling term  $\sim K^{mn}$  between gradients of the director and the magnetization does not enter the picture, since the components of the director and the magnetization along the helical axis are zero. From Eqs.(91) and (92) two important conclusions follow immediately. First of all the system can gain energy by generating a helical state. Surely the system will also generate defects, which cost energy. Provided the helical domains obtained are large enough this result leads to a straightforward way to detect the presence of octupolar order in a ferromagnetic nematic: the optical observation of domains of opposite handedness. The other conclusion is closely tied to the fact that we have two linear gradient terms. The sign and magnitudes of  $\xi^n$  and  $\xi^M$  are material properties that are fixed. In the case in which the signs of  $\xi^n$ and  $\xi^M$  are opposite, but their magnitude is comparable, the expectation is to have a small value of the net wave vector or a large wavelength for the ambidextrous helical domains.

## B. Temperature gradients can drive reversible director rotations

As a reversible cross-coupling term characteristic of ferromagnetic nematics with octupolar order we consider coupling terms involving temperature, concentration and the director field. For heat and concentration currents we get a coupling to the molecular field of the director (compare IV C):

$$j_i^{\sigma R} = \dots + \xi_{ij}^{Tn} h_i^n \tag{93}$$

$$j_i^{cR} = \dots + \xi_{ij}^{cn} h_j^n \tag{94}$$

or, explicitly for  $\sigma$  and for  $m_i \parallel \hat{z}$  and  $n_i \parallel \hat{z}$ :

$$j_{\pi}^{\sigma R} = \dots + \xi^{Tn} M_0 \tilde{T}_0 h_{\pi}^n \tag{95}$$

$$j_y^{\sigma R} = \dots - \xi^{Tn} M_0 \tilde{T}_0 h_y^n \tag{96}$$

$$j_z^{\sigma R} = \dots + 0 \tag{97}$$

where  $\tilde{T}_0 = \frac{4}{3\sqrt{3}}T_0$ .

From inspection of Eqs. (85) and (86) we see that the cross-coupling  $\sim \xi_{ij}^{Tn}$  and  $\sim \xi_{ij}^{cn}$  is linear in  $\mathbf{M}$ ,  $T_{ijk}$  and  $\mathbf{n}$ . Thus the coupling is mediated by making use of the odd behavior under parity and time reversal of the ground state. And the physics is quite apparent: director rotations drive heat and concentration currents without generating entropy.

As a complement we find that temperature gradients and concentration gradients applied externally generate director rotations

$$\dot{n}_i \sim Y_i^{nR} = \dots + \xi_{ii}^{Tn} \nabla_j T + \xi_{ii}^{cn} \nabla_j c \tag{98}$$

or, explicitly for  $m_i \parallel \hat{z}$  and  $n_i \parallel \hat{z}$ :

$$Y_r^{nR} = \dots + \xi^{Tn} M_0 \tilde{T}_0 \nabla_x T + \xi^{cn} M_0 \tilde{T}_0 \nabla_x c \qquad (99)$$

$$Y_u^{nR} = \cdots - \xi^{Tn} M_0 \tilde{T}_0 \nabla_u T - \xi^{cn} M_0 \tilde{T}_0 \nabla_u c \quad (100)$$

$$Y_z^{nR} = \dots + 0 \tag{101}$$

#### C. Magnetization rotations can drive heat currents

Here we present an example of a dissipative effect, which requires a magnetization as well as tetrahedral order. From Sec. IV B we have for the parts of the heat and concentration current coupling to magnetization rotations

$$j_i^{\sigma D} = \dots - \tilde{\psi}^{TD} M_i T_{kii} h_k^m \tag{102}$$

$$j_i^{cD} = \dots - \tilde{\psi}^{cD} M_i T_{kji} h_k^m \tag{103}$$

or, explicitly for the concentration c and for  $m_i \parallel \hat{z}$  and  $n_i \parallel \hat{z}$ :

$$j_x^{cD} = \cdots - \tilde{\psi}^{cD} M_0 \tilde{T}_0 h_y^m \tag{104}$$

$$j_y^{cD} = \cdots - \tilde{\psi}^{cD} M_0 \tilde{T}_0 h_x^m \tag{105}$$

$$j_z^{cD} = \dots + 0 \tag{106}$$

Inspecting Eqs. (102) and (103) we see that heat currents as well as concentration currents are induced by rotations of the magnetization for ferromagnetic nematics with tetrahedral order, since such a rather unique system breaks both, time-reversal and parity symmetry. Conversely temperature gradi-

ents as well as concentration gradients drive the dynamics of the magnetization via

$$\dot{m}_i \sim X_i^{mD} = \dots + M_j T_{ijk} (\tilde{\psi}^{TD} \nabla_k T + \tilde{\psi}^{cD} \nabla_k \mu_c)$$
 (107)

or, explicitly for  $m_i \parallel \hat{z}$  and  $n_i \parallel \hat{z}$ :

$$X_x^{mD} = \cdots + M_0 \tilde{T}_0 (\tilde{\psi}^{TD} \nabla_y T + \tilde{\psi}^{cD} \nabla_y \mu_c)$$
 (108)

$$X_y^{mD} = \cdots + M_0 \tilde{T}_0 (\tilde{\psi}^{TD} \nabla_x T + \tilde{\psi}^{cD} \nabla_x \mu_c)$$
 (109)

$$X_z^{mD} = \dots + 0 ag{110}$$

#### VI. SUMMARY AND PERSPECTIVE

In this paper we have analyzed how the macroscopic properties of ferromagnetic nematic liquid crystals are influenced by the presence of parity breaking octupolar order. It turns out that many additional cross-coupling terms arise in statics and dynamics, since now one has a ground state that breaks both, time-reversal and inversion symmetry. Clearly the hallmark for the presence of octupolar order will be the detection of chiral domains of both hands in a ferromagnetic nematic compound composed of nonchiral molecules: ambidextrous helicity. Since there are two linear gradient terms in the system investigated here, one associated with the nematic director and one associated with ferromagnetic order, one can tune the helical pitch by changing the magnitude of the spontaneous magnetization,  $M_0$ .

As a perspective it will be most interesting to investigate how tetrahedral order will influence ferromagnetic cholesteric liquid crystals, since in such a system parity symmetry breaking is achieved by two different mechanisms: a pseudoscalar quantity associated with the chirality of the molecules of at least one of the constituents as well as with octupolar order. Such a system represents also a challenge for its mathematical description in three spatial dimensions, when both parity breaking mechanisms are at work.

## ACKNOWLEDGMENTS

Partial support of this work by H.R.B., H.P. and T.P. through the Schwerpunktprogramm SPP 1681 'Feldgesteuerte Partikel-Matrix-Wechselwirkungen: Erzeugung, skalenübergreifende Modellierung und Anwendung magnetischer Hybridmaterialien' of the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

<sup>[1]</sup> L. G. Fel, Phys. Rev. E **52**, 702 (1995).

<sup>[2]</sup> L. G. Fel, Phys. Rev. E 52, 2692 (1995).

<sup>[3]</sup> L. Radzihovsky and T.C. Lubensky, Europhys. Lett. **54**, 206 (2001).

<sup>[4]</sup> T. C. Lubensky and L. Radzihovsky, Phys. Rev. E 66, 031704 (2002).

<sup>[5]</sup> B. Mettout, Phys. Rev. E 74, 041701 (2006).

<sup>[6]</sup> L. Longa, G. Pajak, and T. Wydro, Phys. Rev. E 79, 040701 (2009).

<sup>[7]</sup> K. Trojanowski, G. Pajak, L. Longa, and T. Wydro, Phys. Rev. E 86, 011704 (2012).

<sup>[8]</sup> L. Longa and K. Trojanowski, Acta. Phys. Polonica B 44, 1201 (2013).

<sup>[9]</sup> H. R. Brand, H. Pleiner, and P. E. Cladis, Eur. Phys. J. E 7, 163 (2002).

<sup>[10]</sup> P.E. Cladis, H. Pleiner and H.R. Brand, Eur. Phys. J. E 11, 283 (2003).

- [11] H.R. Brand, P.E. Cladis and H. Pleiner, Ferroelectrics, **315**, 165 (2005)
- [12] H.R. Brand, H. Pleiner and P.E. Cladis, Physica A 351, 189 (2005).
- [13] H. Pleiner, P.E. Cladis, and H.R. Brand, Eur. Phys. J. E 20, 257 (2006).
- [14] H. R. Brand and H. Pleiner, Eur. Phys. J. E 31, 37 (2010).
- [15] H. Pleiner and H. R. Brand, Eur. Phys. J. E 37, 11 (2014).
- [16] G. Pelzl, A. Eremin, S. Diele, H. Kresse and W. Weissflog, J. Mat. Chem. 12, 2591 (2002).
- [17] V. Bourny, V. Lorman, J. Pavel, B. Mettout, and H.T. Nguyen, Ferroelectrics 276, 127 (2002).
- [18] W. Weissflog, M.W. Schröder, S. Diele and G. Pelzl, Adv. Mater. 15, 630 (2003).
- [19] T. Niori, J. Yamamoto and H. Yokoyama, Mol. Cryst. Liq. Cryst., 409, 475 (2004).
- [20] M.W. Schroeder, S. Diele, G. Pelzl, and W. Weissflog, ChemPhysChem 5, 99 (2004).
- [21] D. Wiant, J.T. Gleeson, N. Eber, K. Fodor-Csorba, A. Jakli, and T. Toth-Katona, Phys. Rev. E 72, 041712 (2005).
- [22] J. Harden, B. Mbanga, N. Eber, K. Fodor-Csorba, S. Sprunt, J.T. Gleeson, and A. Jakli, Phys. Rev. Lett. 97, 157802 (2006).
- [23] D. Wiant, S. Stojadinovic, K. Neupane, S. Sharma, K. Fodor-Csorba, A. Jakli, J.T. Gleeson, and S. Sprunt, Phys. Rev. E 73, 030703 (2006).
- [24] D. Wiant, K. Neupane, S. Sharma, J.T. Gleeson, S. Sprunt, A. Jakli, N. Pradhan, and G. Iannacchione, Phys. Rev. E 77, 061701 (2008).
- [25] T. Ostapenko, D.B. Wiant, S.N. Sprunt, A. Jakli, and J.T. Gleeson, Phys. Rev. Lett. 101, 247801 (2008).
- [26] Y. Jang, R. Balachandran, C. Keith, A. Lehmann, C. Tschierske, and J.K. Vij, Soft Matter 8, 10479 (2012).
- [27] F. Vita, I.F. Placentino, C. Ferreo, G. Singh, E.T. Samulski, and O. Francescangeli, Soft Matter 9, 6475 (2013).
- [28] M. Jasinski, D. Pociecha, H. Monobe, J. Szczytko, and P. Kaszynski, J. Am. Chem. Soc. 136, 14658 (2014).
- [29] O.N. Kadkin, E.H. Kim, Y.J. Rha, S.Y. Kim, J. Taem and M.-G. Choi, Chem. Eur. J. 15, 10343 (2009).
- [30] E.H. Kim, O.N. Kadkin, S.Y. Kim, J. Taem and M.-G. Choi, Eur. J. Inorg. Chem. 2011, 2933 (2011).
- [31] H. R. Brand and H. Pleiner, Eur. Phys. J. E 40, 34 (2017).
- [32] H. Pleiner and H. R. Brand, Braz. J. Phys. 46, 565 (2016).
- [33] T. Ohta, T. Ohkuma, and K. Shitara, Phys. Rev. E 80, 056203 (2009).
- [34] M. Tarama and T. Ohta, Phys. Rev. E 87, 062912 (2013).
- [35] T. Hiraiwa, M.Y. Matsuo, T. Ohkuma, T. Ohta, and M. Sano, EPL **91**, 20001 (2010).
- [36] E.G. Virga, Eur. Phys. J. E 38, 63 (2015)
- [37] G. Gaeta and E.G. Virga, Eur. Phys. J. E 39, 113 (2016)
- [38] Y. Chen, L.Qi, and E.G. Virga, J. Phys. A **51**, 025206 (2018)
- [39] C. Dressel, T. Reppe, M. Prehm, M. Brautzsch, and C. Tschierske, Nat. Chem. 6, 971 (2014).
- [40] C. Dressel, W. Weissflog, and C. Tschierske, Chem. Comm. 51, 15850 (2015).
- [41] M. Alaasar, M. Prehm, Y. Cao, F. Liu, and C. Tschierske, Angew. Chem., Int. Ed. 55, 312 (2016).

- [42] F. Brochard and P. G. de Gennes, J. Phys. (France) 31, 691 (1970).
- [43] J. Rault, P. E. Cladis, and J. Burger, Phys. Lett. A 32, 199 (1970).
- [44] A. Mertelj, D. Lisjak, M. Drofenik, and M. Čopič, Nature (London) 504, 237 (2013).
- [45] A. Mertelj, N. Osterman, D. Lisjak, and M. Čopič, Soft Matter 10, 9065 (2014).
- [46] R. Sahoo, M. V. Rasna, D. Lisjak, A. Mertelj, and S. Dahra, Appl. Phys. Lett. 106, 161905 (2015).
- [47] A. J. Hess, Q. Liu, and I. I. Smalyukh, Appl. Phys. Lett. 107, 071906 (2015).
- [48] M. Shuai et al., Nat. Commun. 7, 10394 (2016).
- [49] Q. Liu, P. J. Ackerman, T. C. Lubensky, and I. I. Smalyukh, Proc. Natl. Acad. Sci. USA 113, 10479 (2016).
- [50] H. Pleiner, E. Jarkova, H.-W. Müller, and H.R. Brand, Magnetohydrodynamics 37, 254 (2001).
- [51] E. Jarkova, H. Pleiner, H.-W. Müller, A. Fink, and H. R. Brand, Eur. Phys. J. E 5, 583 (2001).
- [52] E. Jarkova, H. Pleiner, H.-W. Müller, and H. R. Brand, J. Chem. Phys. 118, 2422 (2003).
- [53] T. Potisk, D. Svenšek, H. R. Brand, H. Pleiner, D. Lisjak, N. Osterman, and A. Mertelj, Phys. Rev. Lett. 119, 097802 (2017).
- [54] T. Potisk, A. Mertelj, N. Sebastián, N. Osterman, D. Lisjak, H.R. Brand, H. Pleiner, and D. Svenšek, Phys. Rev. E 97, 012701 (2018).
- [55] T. Potisk, H. Pleiner, D. Svenšek, and H. R. Brand, Phys. Rev. E 97, 042705 (2018).
- [56] A. Mertelj and D. Lisjak, Liquid Crystals Reviews 5, 1 (2017).
- [57] Q. Zhang, P. J. Ackerman, Q. Liu, and I. I. Smalyukh, Phys. Rev. Lett 115, 097802 (2015).
- [58] P. J. Ackerman and I. I. Smalyukh, Nat. Mater. 16, 426 (2017).
- [59] P. Medle Rupnik, D. Lisjak, M. Čopič, S. Čopar, and A. Mertelj, Science Advances 3, e1701336 (2017).
- [60] H. R. Brand, A. Fink, and H. Pleiner, Eur. Phys. J. E 38, 65 (2015).
- [61] H. Brand and H. Pleiner, Phys. Rev. A 24, 2777 (1981).
- [62] M. Liu, Phys. Rev. A 24, 2720 (1981).
- [63] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1995).
- [64] T. Potisk, H. Pleiner and H.R. Brand (unpublished).
- [65] P.C. Martin, O. Parodi, and P.S. Pershan, Phys. Rev. A 6, 2401 (1972).
- [66] D. Forster, Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions, (Benjamin, Reading, MA, 1975).
- [67] H. Pleiner and H.R. Brand, Hydrodynamics and Electrohydrodynamics of Liquid Crystals, Pattern Formation in Liquid Crystals, edited by A. Buka and L. Kramer (Springer, New York, 1996) p.15 ff.
- [68] W.P. Mason, Physical Acoustics and the Properties of Solids (D. Van Nostrand, New York, 1958).
- [69] We note that the term  $\sim C_{\perp}$  term is different from the  $K_7$  term in Ref.[32], Eq. (74): the  $C_{\perp}$  is correctly symmetrized in the gradients, while the  $K_7$  term is antisymmetric and therefore does not exist.