

Microscopic and macroscopic theories for the dynamics of polar liquid crystalsRaphael Wittkowski,¹ Hartmut Löwen,¹ and Helmut R. Brand²¹*Institut für Theoretische Physik II, Weiche Materie, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany*²*Theoretische Physik III, Universität Bayreuth, D-95540 Bayreuth, Germany*

(Received 20 July 2011; published 28 October 2011)

We derive and analyze the dynamic equations for polar liquid crystals in two spatial dimensions in the framework of classical dynamical density functional theory (DDFT). Translational density variations, polarization, and quadrupolar order are used as order-parameter fields. The results are critically compared with those obtained using the macroscopic approach of time-dependent Ginzburg-Landau (GL) equations for the analogous order-parameter fields. We demonstrate that, for both the microscopic DDFT and the macroscopic GL approach, the resulting dissipative dynamics can be derived from a dissipation function. We obtain microscopic expressions for all diagonal contributions and for many of the cross-coupling terms emerging from a GL approach. Thus, we establish a bridge between molecular correlations and macroscopic modeling for the dissipative dynamics of polar liquid crystals.

DOI: [10.1103/PhysRevE.84.041708](https://doi.org/10.1103/PhysRevE.84.041708)

PACS number(s): 64.70.mf, 61.30.Dk, 82.70.Dd

I. INTRODUCTION

Understanding the dynamic processes in liquid crystalline systems from a microscopic point of view is important for designing smart materials with novel optical and rheological properties. In fact, the key mechanisms of optical displays and switching devices are controlled by the dynamic response of liquid crystals to external stimuli [1–3]. Therefore, it is highly relevant to understand how these processes change in terms of the molecular interactions. As a first example, the switching dynamics of the nematic director upon a change in an external alignment field [4,5] is one of the basic functions in optical displays. Second, in microfluidic devices, micromixers [6] can be tailored by the response of a liquid crystalline system to a rotating external field.

Nonequilibrium dynamic processes in liquid crystals are also interesting from a more fundamental point of view since they involve a nontrivial coupling between translational and orientational degrees of freedom. Therefore, dynamic macroscopic Ginzburg-Landau (GL) approaches have been applied to liquid crystalline systems in order to obtain the basic dynamic equations on a phenomenological level. Apart from direct computer simulations of molecular systems [7,8], much less has been done in terms of a microscopic theory. Such a microscopic approach, which starts from the molecular interactions, is established by classical density functional theory (DFT) [9–13]. DFT can be generalized toward dynamics for colloidal particles in solution within the so-called dynamical density functional theory (DDFT) [14–16], which has been proven to be a realistic microscopic description for many phenomena, including liquid crystalline dynamics [5,17–19].

Static DFT constitutes a framework to derive GL equations from a microscopic level. The idea is to expand the microscopic one-particle density in terms of order-parameter fields and to perform a gradient expansion [20–25] of the free-energy functional. This leads to a GL-type theory, which can also be called a phase-field-crystal (PFC) model [24–28]. The phenomenological coupling constants of various terms can thereby be expressed in terms of generalized moments of molecular correlation functions.

In this paper, we perform a similar analysis for the *dynamics* of liquid crystals by using DDFT, on the one hand, and time-dependent GL equations, on the other hand. Therefore, we provide a microscopic basis for time-dependent GL theory and derive microscopic expressions for most of the coupling constants. We do this in two spatial dimensions for polar liquid crystals by including translational density variations, polarization, and quadrupolar order as the basic order-parameter fields. We further demonstrate that the resulting dissipative dynamics can be obtained from a dissipation function. This DDFT result holds for constant mobilities as well as for mobilities depending on the thermodynamic variables. Our work opens the way to study various dynamic processes of two-dimensional polar liquid crystals recently observed in experiments [29–31] by further numerical solution of the microscopically justified GL equations.

The paper is organized as follows: In Sec. II, we derive dynamic equations for polar liquid crystals in two spatial dimensions from DDFT. A macroscopic derivation in terms of time-dependent GL equations and its relation to the microscopic dynamics is presented in Sec. III. We conclude in Sec. IV.

II. MICROSCOPIC DERIVATION OF THE DYNAMICS

Our microscopic derivation of the dynamics uses the DDFT equation [17] for collective Brownian motion of anisotropic symmetric colloidal particles and is thus based on a static free-energy functional that can be derived from static DFT. A perturbative functional, which uses molecular fluid correlations as input, has recently been proposed in our previous work [25] for polar liquid crystalline particles in two spatial dimensions. In the following paragraph, we present this functional in a modified form that is more appropriate for our further considerations.

A. Static free-energy functional

A suitable quantity to describe the state of a system of many interacting anisotropic particles is the one-particle density $\rho(\vec{r}, \hat{u})$. It is proportional to the probability density to find a

particle with orientation \hat{u} at position \vec{r} , where $\hat{u} = (u_1, u_2) = (\cos(\varphi), \sin(\varphi))$ is the orientational unit vector that denotes the orientation of the symmetry axis of the respective particle and $\vec{r} = (x_1, x_2)$ is the center-of-mass position vector. In the following, slightly different from Ref. [25], we choose the parametrization

$$\rho(\vec{r}, \hat{u}) = \bar{\rho} [1 + \psi(\vec{r}) + P_i(\vec{r})u_i + u_i Q_{ij}(\vec{r})u_j] \quad (1)$$

with the mean number density $\bar{\rho}$ and the dimensionless order-parameter fields $\psi(\vec{r})$, $P_i(\vec{r})$, and $Q_{ij}(\vec{r})$, where Einstein's sum convention is used. The first of these order-parameter fields is the reduced translational density

$$\psi(\vec{r}) = \frac{1}{2\pi\bar{\rho}} \int_{S_1} d\hat{u} [\rho(\vec{r}, \hat{u}) - \bar{\rho}] \quad (2)$$

with S_1 denoting the unit circle. The field $\psi(\vec{r})$ measures translational deviations of $\rho(\vec{r}, \hat{u})$ from the mean density $\bar{\rho}$. The second order-parameter field is the polarization $\vec{P}(\vec{r})$. It has the components

$$P_i(\vec{r}) = \frac{1}{\pi\bar{\rho}} \int_{S_1} d\hat{u} \rho(\vec{r}, \hat{u}) u_i \quad (3)$$

and describes the averaged orientation. Finally, the traceless and symmetric nematic tensor with the components

$$Q_{ij}(\vec{r}) = \frac{2}{\pi\bar{\rho}} \int_{S_1} d\hat{u} \rho(\vec{r}, \hat{u}) \left(u_i u_j - \frac{1}{2} \delta_{ij} \right) \quad (4)$$

and the Kronecker delta symbol δ_{ij} describes quadrupolar ordering. The free-energy functional $\mathcal{F}[\rho(\vec{r}, \hat{u})]$ is decomposed as

$$\mathcal{F}[\rho(\vec{r}, \hat{u})] = \mathcal{F}_{\text{id}}[\rho(\vec{r}, \hat{u})] + \mathcal{F}_{\text{exc}}[\rho(\vec{r}, \hat{u})] \quad (5)$$

into the ideal rotator gas functional

$$\beta \mathcal{F}_{\text{id}}[\rho(\vec{r}, \hat{u})] = \int_{\mathcal{A}} d\vec{r} \int_{S_1} d\hat{u} \rho(\vec{r}, \hat{u}) \{ \ln[\Lambda^2 \rho(\vec{r}, \hat{u})] - 1 \} \quad (6)$$

with the inverse thermal energy $\beta = 1/(k_B T)$, the two-dimensional domain \mathcal{A} , and the thermal de Broglie wavelength Λ as well as the excess free-energy functional $\mathcal{F}_{\text{exc}}[\rho(\vec{r}, \hat{u})]$, which in general is only known approximatively. Inserting the parametrization (1) into Eq. (6) and performing a Taylor expansion up to fourth order in the order-parameter fields yields to the approximation

$$\beta \mathcal{F}_{\text{id}}[\rho(\vec{r}, \hat{u})] = F_{\text{id}} + \pi\bar{\rho} \int_{\mathcal{A}} d\vec{r} f_{\text{id}}(\vec{r}) \quad (7)$$

with the local scaled ideal rotator gas free-energy density

$$\begin{aligned} f_{\text{id}}(\vec{r}) = & \frac{\psi}{4} (8 - 2P_i^2 + 2P_i Q_{ij} P_j - Q_{ij}^2) \\ & + \frac{\psi^2}{4} (4 + 2P_i^2 + Q_{ij}^2) - \frac{\psi^3}{3} + \frac{\psi^4}{6} \\ & + \frac{P_i^2}{8} (4 + Q_{kl}^2) - \frac{P_i Q_{ij} P_j}{4} + \frac{P_i^2 P_j^2}{16} \\ & + \frac{Q_{ij}^2}{4} + \frac{Q_{ij}^2 Q_{kl}^2}{64}, \end{aligned} \quad (8)$$

where

$$F_{\text{id}} = 2\pi\bar{\rho} A [\ln(\Lambda^2 \bar{\rho}) - 1] \quad (9)$$

is an irrelevant constant and

$$A = \int_{\mathcal{A}} d\vec{r} \quad (10)$$

is the total area of the domain \mathcal{A} . We further consider a functional Taylor expansion of $\mathcal{F}_{\text{exc}}[\rho(\vec{r}, \hat{u})]$ up to second order [32], also called *Ramakrishnan-Yussouff approximation* [33]

$$\begin{aligned} \beta \mathcal{F}_{\text{exc}}[\rho(\vec{r}, \hat{u})] = & -\frac{1}{2} \int_{\mathcal{A}} d\vec{r}_1 \int_{S_1} d\hat{u}_1 \int_{\mathcal{A}} d\vec{r}_2 \int_{S_1} d\hat{u}_2 \\ & \times c^{(2)}(\vec{r}_1 - \vec{r}_2, \hat{u}_1, \hat{u}_2) \\ & \times \Delta\rho(\vec{r}_1, \hat{u}_1) \Delta\rho(\vec{r}_2, \hat{u}_2), \end{aligned} \quad (11)$$

with the direct pair-correlation function [34]

$$c^{(2)}(\vec{r}_1, \vec{r}_2, \hat{u}_1, \hat{u}_2) = c^{(2)}(\vec{r}_1 - \vec{r}_2, \hat{u}_1, \hat{u}_2) \quad (12)$$

and the reduced one-particle density $\Delta\rho(\vec{r}, \hat{u}) = \rho(\vec{r}, \hat{u}) - \bar{\rho}$. By performing a gradient expansion up to second order, we obtain for the excess free-energy functional the approximation

$$\beta \mathcal{F}_{\text{exc}}[\rho(\vec{r}, \hat{u})] = -\frac{1}{2} \int_{\mathcal{A}} d\vec{r} f_{\text{exc}}(\vec{r}) \quad (13)$$

with the local scaled excess free-energy density

$$\begin{aligned} f_{\text{exc}}(\vec{r}) = & A_1 \psi^2 + A_2 (\partial_i \psi)^2 + A_3 (\partial_k^2 \psi)^2 + B_1 (\partial_i \psi) P_i \\ & + B_2 P_i (\partial_j Q_{ij}) + B_3 (\partial_i \psi) (\partial_j Q_{ij}) + C_1 P_i^2 \\ & + C_2 P_i (\partial_k^2 P_i) + C_3 (\partial_i P_i)^2 \\ & + D_1 Q_{ij}^2 + D_2 (\partial_j Q_{ij})^2. \end{aligned} \quad (14)$$

The various coefficients are given by

$$A_1 = 8 M_0^0(1), \quad (15)$$

$$A_2 = -2 M_0^0(3), \quad (16)$$

$$A_3 = \frac{1}{8} M_0^0(5), \quad (17)$$

$$B_1 = 4 [M_{-1}^1(2) - M_1^0(2)], \quad (18)$$

$$B_2 = 2 [M_1^1(2) - M_{-1}^2(2)], \quad (19)$$

$$B_3 = -M_{-2}^2(3) - M_0^0(3), \quad (20)$$

$$C_1 = 4 M_0^1(1), \quad (21)$$

$$C_2 = M_0^1(3) - \frac{1}{2} M_{-2}^1(3), \quad (22)$$

$$C_3 = -M_{-2}^1(3), \quad (23)$$

$$D_1 = 2 M_0^2(1), \quad (24)$$

$$D_2 = -M_0^2(3) \quad (25)$$

as linear combinations of the moments

$$M_l^m(\alpha) = \pi^3 \bar{\rho}^2 \int_0^\infty dR R^\alpha \tilde{c}_{l,m}^{(2)}(R) \quad (26)$$

of the Fourier coefficients

$$\tilde{c}_{l,m}^{(2)}(R) = \frac{1}{(2\pi)^2} \int_0^{2\pi} d\phi_R \int_0^{2\pi} d\phi c^{(2)}(R, \phi_R, \phi) e^{-i(l\phi_R + m\phi)} \quad (27)$$

of the direct pair-correlation function $c^{(2)}(R, \phi_R, \phi)$, for which the parametrization

$$c^{(2)}(\vec{r}_1 - \vec{r}_2, \hat{u}_1, \hat{u}_2) \equiv c^{(2)}(R, \phi_R, \phi) \quad (28)$$

with $\vec{r}_1 - \vec{r}_2 = R\hat{u}(\varphi_R)$, $\hat{u}_i = \hat{u}(\varphi_i)$ for $i = 1, 2$, $\phi_R = \varphi - \varphi_R$, and $\phi = \varphi_1 - \varphi_2$ was used. Equations (7) and (13) give a local functional of a polar liquid crystalline system reminiscent of a PFC model [19].

B. Dynamic equations

We now derive dynamic equations for $\mathcal{A} = \mathbb{R}^2$ for the order-parameter fields $\psi(\vec{r}, t)$, $P_i(\vec{r}, t)$, and $Q_{ij}(\vec{r}, t)$ from dynamical density functional theory. DDFT is constructed to describe the Brownian dynamics of colloidal particles in a viscous solvent [38,39] via a time-dependent one-particle density field $\rho(\vec{r}, \hat{u}, t)$. This theory was recently extended to anisotropic Brownian particles with orientational degrees of freedom [17,40,41]. It provides as a starting point for the case of symmetric uniaxial particles without translational-rotational coupling in two spatial dimensions the DDFT equation [17]

$$\frac{\partial \rho}{\partial t}(\vec{r}, \hat{u}, t) = \beta \vec{\nabla} \cdot \left(\mathbf{D}_T(\hat{u}) \rho(\vec{r}, \hat{u}, t) \vec{\nabla} \frac{\delta \mathcal{F}[\rho(\vec{r}, \hat{u}, t)]}{\delta \rho(\vec{r}, \hat{u}, t)} \right) + \beta D_R \frac{\partial}{\partial \varphi} \left(\rho(\vec{r}, \hat{u}, t) \frac{\partial}{\partial \varphi} \frac{\delta \mathcal{F}[\rho(\vec{r}, \hat{u}, t)]}{\delta \rho(\vec{r}, \hat{u}, t)} \right) \quad (29)$$

with the translational short-time diffusion tensor

$$\mathbf{D}_T(\hat{u}) = D_{\parallel} \hat{u} \otimes \hat{u} + D_{\perp} (\mathbf{1} - \hat{u} \otimes \hat{u}). \quad (30)$$

Here, D_{\parallel} and D_{\perp} are the translational diffusion coefficients for translation parallel and perpendicular to the orientation \hat{u} , respectively, D_R is the rotational diffusion coefficient, \otimes is the dyadic product, and the symbol $\mathbf{1}$ denotes the two-dimensional unit matrix. The two terms on the right-hand side of this DDFT equation for uniaxial particles correspond to pure translation and pure rotation, respectively. Translational-rotational coupling terms, which are especially relevant for screwlike particles, do not appear in this DDFT equation. Additional terms in the DDFT equation, which regard a possible translational-rotational coupling, would have the same structure as the present terms, but with only one gradient and one angular derivative each instead of two gradients or two angular derivatives, respectively [41].

Following the analysis of Ref. [19], the functional derivative $\delta \mathcal{F} / \delta \rho$ in the DDFT equation (29) has to be expressed by functional derivatives of the free-energy functional with respect to the order-parameter fields $\psi(\vec{r}, t)$, $P_i(\vec{r}, t)$, and $Q_{ij}(\vec{r}, t)$

since we parametrized the one-particle density $\rho(\vec{r}, \hat{u}, t)$ as well as the free-energy functional $\mathcal{F}[\psi, P_i, Q_{ij}]$ with these order-parameter fields. In the following equations, a large number of functional derivatives of the free-energy functional appear. Therefore, we shorten the notation by defining the *conjugated order-parameter fields* or *thermodynamic forces*

$$\Xi^{\natural} = \frac{\delta \mathcal{F}}{\delta \Xi} \quad \text{with} \quad \Xi \in \{\rho, \psi, P_i, Q_{ij}\}. \quad (31)$$

Using this notation, the equation

$$\rho^{\natural} = \frac{1}{2\pi\bar{\rho}} \psi^{\natural} + \frac{u_i}{\pi\bar{\rho}} P_i^{\natural} + \frac{u_i u_j}{\pi\bar{\rho}} Q_{ij}^{\natural} \quad (32)$$

follows by functional differentiation. When performing functional derivatives with respect to Q_{ij} or Q_{ij}^{\natural} , one has to notice that Q_{ij} as well as Q_{ij}^{\natural} are symmetric and traceless. The interdependence of the elements of these tensors leads to more complicated derivatives that respect the symmetry properties of these tensors. A very useful equation in this context is

$$\frac{\delta Q_{kl}}{\delta Q_{ij}^{\natural}} = \frac{\delta Q_{kl}^{\natural}}{\delta Q_{ij}^{\natural}} = \delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il} - \delta_{ij} \delta_{kl}. \quad (33)$$

Together with the parametrization (1) of the one-particle density, the relation (32) can now be inserted into the DDFT equation (29). The dynamic equations for the order-parameter fields are then obtained by an orthogonal projection that separates the evolution equations for the particular order-parameter fields from each other. This projection is achieved by a multiplication of Eq. (29) with $\mathbf{1}$, u_i , and $u_i u_j - \delta_{ij}/2$, respectively, with a subsequent integration over the orientation \hat{u} . In doing so, the translational density $\psi(\vec{r}, t)$ appears to be conserved, while $P_i(\vec{r}, t)$ and $Q_{ij}(\vec{r}, t)$ are not conserved due to their association with orientational degrees of freedom. The dynamic equations can thus be written in the form

$$\dot{\psi} + \partial_i J_i^{\psi} = 0, \quad (34)$$

$$\dot{P}_i + \Phi_i^P = 0, \quad (35)$$

$$\dot{Q}_{ij} + \Phi_{ij}^Q = 0 \quad (36)$$

with $\dot{\Xi} = \frac{\partial \Xi}{\partial t}$ denoting the partial time derivative of the field $\Xi \in \{\psi, P_i, Q_{ij}\}$ and with the current J_i^{ψ} and the quasicharacteristics Φ_i^P and Φ_{ij}^Q . These *dissipative currents* and quasicharacteristics are given by the expressions

$$J_i^{\psi} = -\alpha_1 [2(1 + \psi)(\partial_i \psi^{\natural}) + Q_{kl}(\partial_i Q_{kl}^{\natural})] - \alpha_2 P_j(\partial_i P_j^{\natural}) - \alpha_3 [2(1 + \psi)(\partial_j Q_{ij}^{\natural}) + P_i(\partial_j P_j^{\natural}) + P_j(\partial_j P_i^{\natural}) + Q_{ij}(\partial_j \psi^{\natural})], \quad (37)$$

$$\begin{aligned} \Phi_i^P = & -2\alpha_1 \partial_k [Q_{ij}(\partial_k P_j^{\natural}) + P_j(\partial_k Q_{ij}^{\natural})] - \alpha_2 \partial_k [2(1 + \psi)(\partial_k P_i^{\natural}) + P_i(\partial_k \psi^{\natural})] \\ & - \alpha_3 \{2\partial_i [(1 + \psi)(\partial_j P_j^{\natural})] + 2\partial_j [(1 + \psi)(\partial_i P_j^{\natural})] + \partial_i [P_j(\partial_j \psi^{\natural})] + \partial_j [P_i(\partial_i \psi^{\natural})] \\ & + 2\partial_j [P_i(\partial_k Q_{jk}^{\natural}) + Q_{jk}(\partial_k P_i^{\natural})] + \alpha_4 [2(1 + \psi)P_i^{\natural} + 2P_j Q_{ij}^{\natural} - Q_{ij} P_j^{\natural}], \end{aligned} \quad (38)$$

$$\begin{aligned} \Phi_{ij}^Q = & -2\alpha_1 \partial_k [2(1 + \psi)(\partial_k Q_{ij}^{\natural}) + P_i(\partial_k P_j^{\natural}) + P_j(\partial_k P_i^{\natural}) - \delta_{ij} P_l(\partial_k P_l^{\natural}) + Q_{ij}(\partial_k \psi^{\natural})] \\ & - \frac{\alpha_3}{2} \{4\partial_i [(1 + \psi)(\partial_j \psi^{\natural})] + 4\partial_j [(1 + \psi)(\partial_i \psi^{\natural})] - 4\delta_{ij} \partial_l [(1 + \psi)(\partial_l \psi^{\natural})] \\ & + 4\partial_i [P_k(\partial_j P_k^{\natural})] + 4\partial_j [P_k(\partial_i P_k^{\natural})] - 4\delta_{ij} \partial_l [P_k(\partial_l P_k^{\natural})] + \partial_i [Q_{kl}(\partial_j Q_{kl}^{\natural})] + \partial_j [Q_{kl}(\partial_i Q_{kl}^{\natural})] - \delta_{ij} \partial_l [Q_{km}(\partial_l Q_{km}^{\natural})] \\ & + 2\partial_k [Q_{ij}(\partial_l Q_{kl}^{\natural})] + 2\partial_k [Q_{kl}(\partial_l Q_{ij}^{\natural})] + 2\alpha_4 [4(1 + \psi)Q_{ij}^{\natural} + P_i P_j^{\natural} + P_j P_i^{\natural} - \delta_{ij} P_l P_l^{\natural}], \end{aligned} \quad (39)$$

where ∂_i are the components of the gradient $\vec{\nabla} = (\partial_1, \partial_2)$. Four positive coefficients, of which three are independent, appear in these equations. With the abbreviation $\lambda = \pi \bar{\rho} / \beta$, they are defined as

$$\begin{aligned} \alpha_1 &= \frac{D_{\parallel} + D_{\perp}}{8\lambda}, & \alpha_2 &= \frac{D_{\parallel} + 3D_{\perp}}{8\lambda}, \\ \alpha_3 &= \frac{D_{\parallel} - D_{\perp}}{8\lambda}, & \alpha_4 &= \frac{D_R}{2\lambda}. \end{aligned} \quad (40)$$

Note that $D_{\parallel} \geq D_{\perp}$ holds for all types of uniaxial particles if the vector \hat{u} for the orientation of the symmetry axis is chosen properly [42].

At this stage, we emphasize that the DDFT approach (29) *a priori* contains only three independent mobility coefficients, namely, the two translational diffusion coefficients D_{\parallel} and D_{\perp} and the rotational diffusion coefficient D_R . Therefore, all other mobility coefficients for the order-parameter fields can be expressed in terms of these three basic coefficients. In general, the diffusion coefficients in DDFT are always related to translational or orientational degrees of freedom and not to certain order parameters, which appear only with the parametrization of the one-particle density. The parametrization of the one-particle density (1) in turn does not involve further dissipation coefficients. This is in sharp contrast to GL theory, where every additional order parameter involves at least one new dissipative coefficient, as will be discussed in Sec. III in more detail.

Since Eqs. (37)–(39) are rather complicated, for numerical calculations, a simpler version of these equations might be desirable. Such a simplification is the *constant-mobility approximation* (CMA), where the one-particle density in the translational and rotational mobility terms of the DDFT equation (29) is approximated by its mean value $\bar{\rho}$:

$$\begin{aligned} \frac{\partial \rho}{\partial t}(\vec{r}, \hat{u}, t) &= \beta \bar{\rho} \vec{\nabla} \cdot \left(\mathbf{D}_T(\hat{u}) \vec{\nabla} \frac{\delta \mathcal{F}[\rho(\vec{r}, \hat{u}, t)]}{\delta \rho(\vec{r}, \hat{u}, t)} \right) \\ &+ \beta \bar{\rho} D_R \frac{\partial^2}{\partial \varphi^2} \frac{\delta \mathcal{F}[\rho(\vec{r}, \hat{u}, t)]}{\delta \rho(\vec{r}, \hat{u}, t)}. \end{aligned} \quad (41)$$

With Eq. (41) instead of the DDFT equation (29), the following dissipative currents and quasicurrents are obtained:

$$J_i^{\psi} = -2\alpha_1(\partial_i \psi^{\natural}) - 2\alpha_3(\partial_j Q_{ij}^{\natural}), \quad (42)$$

$$\Phi_i^P = -2\alpha_2(\partial_k^2 P_i^{\natural}) - 4\alpha_3(\partial_i \partial_j P_j^{\natural}) + 2\alpha_4 P_i^{\natural}, \quad (43)$$

$$\begin{aligned} \Phi_{ij}^Q &= -4\alpha_1(\partial_k^2 Q_{ij}^{\natural}) - 2\alpha_3[2(\partial_i \partial_j \psi^{\natural}) - \delta_{ij}(\partial_k^2 \psi^{\natural})] \\ &+ 8\alpha_4 Q_{ij}^{\natural}. \end{aligned} \quad (44)$$

For both the general Eqs. (37)–(39) and the much simpler constant-mobility Eqs. (42)–(44), the explicit forms of the conjugated order-parameter fields $\psi^{\natural}(\vec{r}, t)$, $P_i^{\natural}(\vec{r}, t)$, and $Q_{ij}^{\natural}(\vec{r}, t)$ result directly from the functional derivatives of Eqs. (7) and (13) with respect to the order-parameter fields. These functional derivatives are given by

$$\begin{aligned} \frac{1}{\lambda} \frac{\delta \mathcal{F}_{\text{id}}}{\delta \psi}(\vec{r}, t) &= 2 - \frac{P_i^2}{2} + \frac{P_i Q_{ij} P_j}{2} - \frac{Q_{ij}^2}{4} \\ &+ \frac{\psi}{2}(4 + 2P_i^2 + Q_{ij}^2) - \psi^2 + \frac{2}{3}\psi^3, \end{aligned} \quad (45)$$

$$\begin{aligned} \frac{1}{\lambda} \frac{\delta \mathcal{F}_{\text{id}}}{\delta P_i}(\vec{r}, t) &= -\psi(P_i - Q_{ij} P_j) + \psi^2 P_i \\ &+ \frac{P_i}{4}(4 + Q_{kl}^2) - \frac{Q_{ij} P_j}{2} + \frac{P_i P_j^2}{4}, \end{aligned} \quad (46)$$

$$\begin{aligned} \frac{1}{\lambda} \frac{\delta \mathcal{F}_{\text{id}}}{\delta Q_{ij}}(\vec{r}, t) &= \frac{\psi}{2}(2P_i P_j - \delta_{ij} P_l^2 - 2Q_{ij}) \\ &+ \psi^2 Q_{ij} + \frac{P_k^2}{2} Q_{ij} - \frac{1}{4}(2P_i P_j - \delta_{ij} P_l^2) \\ &+ Q_{ij} + \frac{Q_{ij} Q_{kl}^2}{8} \end{aligned} \quad (47)$$

and

$$\begin{aligned} -2\beta \frac{\delta \mathcal{F}_{\text{exc}}}{\delta \psi}(\vec{r}, t) &= 2A_1 \psi - 2A_2(\partial_k^2 \psi) + 2A_3(\partial_k^2 \partial_l^2 \psi) \\ &- B_1(\partial_i P_i) - B_3(\partial_i \partial_j Q_{ij}), \end{aligned} \quad (48)$$

$$\begin{aligned} -2\beta \frac{\delta \mathcal{F}_{\text{exc}}}{\delta P_i}(\vec{r}, t) &= B_1(\partial_i \psi) + B_2(\partial_j Q_{ij}) + 2C_1 P_i \\ &+ 2C_2(\partial_k^2 P_i) - 2C_3(\partial_i \partial_j P_j), \end{aligned} \quad (49)$$

$$\begin{aligned} -2\beta \frac{\delta \mathcal{F}_{\text{exc}}}{\delta Q_{ij}}(\vec{r}, t) &= -B_2[\partial_i P_j + \partial_j P_i - \delta_{ij}(\partial_l P_l)] \\ &- B_3[2(\partial_i \partial_j \psi) - \delta_{ij}(\partial_l^2 \psi)] + 4D_1 Q_{ij} \\ &- 2D_2 \partial_k [\partial_i Q_{kj} + \partial_j Q_{ki} - \delta_{ij}(\partial_l Q_{kl})]. \end{aligned} \quad (50)$$

C. Dissipation function

In the field of linear irreversible thermodynamics [43–45], the dissipative parts of the currents and quasicurrents arising in the balance equations for the thermodynamic variables (including, for example, hydrodynamic and macroscopic variables) can be derived from a *dissipation function* \mathfrak{R} , which is quadratic in the thermodynamic forces. Frequently, one uses equivalently the *entropy production* \mathfrak{R}/T with T denoting the absolute temperature [43,46]. The entropy production emerges as a source term in the balance equation

$$\dot{\sigma} + \partial_i j_i^{\sigma} = \frac{\mathfrak{R}}{T} \quad (51)$$

for the entropy density σ , where \vec{j}^{σ} is the entropy current density. Both dissipation function and entropy production are maximized close to local thermodynamic equilibrium and, thus, are a useful tool in the determination of the dissipative currents and quasicurrents. This approach has been applied to a large number of hydrodynamic and macroscopic systems [43–47], but is in general not applicable for active systems and for systems driven far from equilibrium (compare, for example, Refs. [45,47]). In this more general case, a number of additional conditions must be satisfied [48–52] in order to obtain a Ljapunov functional. Far away from equilibrium, the Ljapunov functional is the analog of the dissipation function of linear irreversible thermodynamics.

More precisely, the dissipative currents and quasicurrents of the dynamic equations (34)–(36) (which are, in linear irreversible thermodynamics, by construction linear in the thermodynamic forces [43–46]) are given by the variational

derivatives of the dissipation function with respect to the thermodynamic forces:

$$J_i^\psi = -\frac{\delta \mathfrak{R}}{\delta (\partial_i \psi^{\text{b}})}, \quad (52)$$

$$\Phi_i^P = \frac{\delta \mathfrak{R}}{\delta P_i^{\text{b}}}, \quad (53)$$

$$\Phi_{ij}^Q = \frac{\delta \mathfrak{R}}{\delta Q_{ij}^{\text{b}}}. \quad (54)$$

The dissipation function that corresponds to the dissipative currents and quasicurrents (37)–(39) of the general phase-field-crystal (PFC) model is found to be

$$\begin{aligned} \mathfrak{R}^{\text{(PFC)}} = \int_{\mathcal{A}} d\vec{r} \left\{ \alpha_1 \{ (1 + \psi) [(\partial_i \psi^{\text{b}})^2 + (\partial_k Q_{ij}^{\text{b}})^2] + Q_{ij} (\partial_k \psi^{\text{b}}) (\partial_k Q_{ij}^{\text{b}}) + Q_{ij} (\partial_k P_i^{\text{b}}) (\partial_k P_j^{\text{b}}) \right. \\ + 2P_i (\partial_k P_j^{\text{b}}) (\partial_k Q_{ij}^{\text{b}}) \} + \alpha_2 [P_i (\partial_j \psi^{\text{b}}) (\partial_j P_i^{\text{b}}) + (1 + \psi) (\partial_j P_i^{\text{b}})^2] \\ + \alpha_3 \left[(\partial_i \psi^{\text{b}}) \left(\frac{1}{2} Q_{ij} (\partial_j \psi^{\text{b}}) + P_i (\partial_j P_j^{\text{b}}) + P_j (\partial_j P_i^{\text{b}}) + 2(1 + \psi) (\partial_j Q_{ij}^{\text{b}}) \right) \right. \\ + (1 + \psi) (\partial_i P_i^{\text{b}})^2 + (\partial_j P_i^{\text{b}}) [(1 + \psi) (\partial_i P_j^{\text{b}}) + Q_{jk} (\partial_k P_i^{\text{b}}) + 2P_i (\partial_k Q_{jk}^{\text{b}})] \\ + \frac{1}{4} Q_{ij} (\partial_i Q_{kl}^{\text{b}}) (\partial_j Q_{kl}^{\text{b}}) + \frac{1}{2} Q_{ij} (\partial_k Q_{ij}^{\text{b}}) (\partial_l Q_{kl}^{\text{b}}) \left. \right] \\ + \alpha_4 \left((1 + \psi) [(P_i^{\text{b}})^2 + 2(Q_{ij}^{\text{b}})^2] - \frac{1}{2} Q_{ij} P_i^{\text{b}} P_j^{\text{b}} + 2P_i P_j Q_{ij}^{\text{b}} \right) \left. \right\}. \quad (55) \end{aligned}$$

Together with the dissipative currents and quasicurrents (37)–(39), this dissipation function constitutes the basic result of this paper. The dissipation function that corresponds to the currents and quasicurrents (42)–(44) of the constant-mobility approximation is much simpler and given by

$$\begin{aligned} \mathfrak{R}^{\text{(CMA)}} = \int_{\mathcal{A}} d\vec{r} \left(\alpha_1 [(\partial_i \psi^{\text{b}})^2 + (\partial_k Q_{ij}^{\text{b}})^2] \right. \\ + \alpha_2 (\partial_k P_i^{\text{b}})^2 + \alpha_4 [(P_i^{\text{b}})^2 + 2(Q_{ij}^{\text{b}})^2] \\ + 2\alpha_3 [(\partial_i \psi^{\text{b}}) (\partial_j Q_{ij}^{\text{b}}) + (\partial_i P_i^{\text{b}})^2] \left. \right). \quad (56) \end{aligned}$$

By construction, both dissipation functions (55) and (56) are positive. This is obvious for Eq. (56), but not manifest for Eq. (55).

III. MACROSCOPIC APPROACH: GINZBURG-LANDAU DYNAMICS

In this section, we investigate the Ginzburg-Landau dynamics in the vicinity of the phase transitions isotropic to polar nematic and isotropic to polar smectic. In analogy to the previous section, the GL dynamics is discussed for three types of macroscopic variables. These are the smectic density variation ρ_ψ , which is closely related to the complex scalar ψ often used to describe smectic layering [53,54], the macroscopic polarization P_i , which becomes important when polar nematic and/or polar smectic phases are considered [55–57], and the quadrupolar nematic order parameter Q_{ij} , which is characteristic of the usual nematic ordering [58,59].

We assume that the local formulation of the first law of thermodynamics, the Gibbs-Duhem relation, is valid [44–46].

It can be written in the form

$$T d\sigma = d\varepsilon - \mu d\rho - \rho_\psi d\mu_\psi - h_i^P dP_i - Q_{ij} dS_{ij} \quad (57)$$

with the absolute temperature T , the entropy density σ , the energy density ε , the chemical potential μ , the number density ρ , the chemical potential μ_ψ associated with the layering ρ_ψ , the thermodynamic force h_i^P associated with the macroscopic polarization P_i , and the thermodynamic conjugate S_{ij} of the nematic order parameter Q_{ij} .

Throughout the following, we focus entirely on the dissipative dynamics of the variables associated with the additional degrees of ordering, i. e., layering ρ_ψ , polar order P_i , and quadrupolar orientational order Q_{ij} . For the associated dynamic balance equations, we have one dynamic equation each for every hydrodynamic or macroscopic variable. These dynamic equations take the form of a conservation law for conserved quantities and are of balance equation type for hydrodynamic variables associated with spontaneously broken continuous symmetries and for macroscopic variables such as order parameters close to a phase transition. The dynamic balance equations thus take the form [43,46,47,58,60,61]

$$\dot{\rho}_\psi + \partial_i X_i^\psi = 0, \quad (58)$$

$$\dot{P}_i + Y_i^P = 0, \quad (59)$$

$$\dot{Q}_{ij} + Z_{ij}^Q = 0. \quad (60)$$

The currents and quasicurrents X_i^ψ , Y_i^P , and Z_{ij}^Q are introduced via Eqs. (58)–(60). Further below, the dissipative part of their structure will be determined from the dissipation function $\mathfrak{R}^{\text{(GL)}}$. There are no reversible currents and quasicurrents throughout this paper since flow effects associated with a

velocity field \vec{v} or with a density of linear momentum \vec{g} are generally not considered for the completely overdamped Brownian dynamics described by DDFT. We note that the dynamic equation associated with the smectic layering is of conservation-law type, while the equations for polar and nonpolar orientational order are balance laws.

In the spirit of linear irreversible thermodynamics, we expand the dissipation function $\mathfrak{R}^{(\text{GL})}$ quadratically in the thermodynamic forces μ_ψ , h_i^P , and S_{ij} . Those in turn have to be determined by taking variational derivatives

$$\mu_\psi = \frac{\delta \mathcal{F}}{\delta \rho_\psi}, \quad (61)$$

$$h_i^P = \frac{\delta \mathcal{F}}{\delta P_i}, \quad (62)$$

$$S_{ij} = \frac{\delta \mathcal{F}}{\delta Q_{ij}} \quad (63)$$

of the suitably chosen generalized potential \mathcal{F} with respect to the variables, where \mathcal{F} has been discussed in detail in Refs. [24,25].

For the dissipation function associated with the three types of order considered here, we have to lowest order in the gradients

$$\begin{aligned} \mathfrak{R}_0^{(\text{GL})} = \int_{\mathcal{A}} d\vec{r} & \left(\frac{1}{2} \gamma_{ijkl} S_{ij} S_{kl} + \frac{1}{2} \alpha_{ij} (\partial_i \mu_\psi) (\partial_j \mu_\psi) \right. \\ & + \frac{1}{2} b_{ij} h_i^P h_j^P + \beta_{ikl} (\partial_i \mu_\psi) S_{kl} \\ & \left. + \tilde{\alpha}_{ij}^P (\partial_i \mu_\psi) h_j^P + \beta_{ikl}^P h_i^P S_{kl} \right). \end{aligned} \quad (64)$$

It is usual to consider at first only the lowest-order gradient terms in the dissipation function. Then, one inspects whether contributions containing more gradients are physically relevant. For example, one can always add a term containing two more gradients for diagonal terms. This was done for the diagonal term $\sim \gamma_{ijkl}$ in Eq. (64), which leads to a relaxation of the nematic order parameter close to the phase transition [58], by the term $\sim \tilde{\gamma}_{ijklmn}$ in Eq. (70) further below, which contains two more gradients and is the dissipative analog of the gradient energy of the order parameter Q_{ij} .

From the dissipation function (64), we obtain for the dissipative currents and quasicurrents the expressions

$$X_i^\psi = -\frac{\delta \mathfrak{R}_0^{(\text{GL})}}{\delta (\partial_i \mu_\psi)} = -\alpha_{ij} (\partial_j \mu_\psi) - \tilde{\alpha}_{ij}^P h_j^P - \beta_{ikl} S_{kl}, \quad (65)$$

$$Y_i^P = \frac{\delta \mathfrak{R}_0^{(\text{GL})}}{\delta h_i^P} = \tilde{\alpha}_{ij}^P (\partial_j \mu_\psi) + b_{ij} h_j^P + \beta_{ikl}^P S_{kl}, \quad (66)$$

$$\begin{aligned} Z_{ij}^Q = \frac{\delta \mathfrak{R}_0^{(\text{GL})}}{\delta S_{ij}} & = (\beta_{kij} + \beta_{kji} - \delta_{ij} \beta_{kll}) (\partial_k \mu_\psi) \\ & + (\beta_{kij}^P + \beta_{kji}^P - \delta_{ij} \beta_{kll}^P) h_k^P \\ & + (\gamma_{klij} + \gamma_{klji} - \delta_{ij} \gamma_{klmm}) S_{kl}. \end{aligned} \quad (67)$$

In a truly isotropic phase, one has only two invariants: the Kronecker delta δ_{ij} and the totally antisymmetric symbol ϵ_{ijk} . To preserve the symmetries of such a system, all the diagonal terms in Eqs. (64)–(67) contribute, while all off-diagonal coupling terms except for one ($\sim \tilde{\alpha}_{ij}^P$) vanish. Correspondingly,

the property tensors take the form

$$\gamma_{ijkl} = \gamma (\delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il}) \quad (68)$$

(compare also Ref. [62]), $\alpha_{ij} = \alpha \delta_{ij}$, and $b_{ij} = b \delta_{ij}$ for the diagonal terms, and $\tilde{\alpha}_{ij}^P = \tilde{\alpha}^P \delta_{ij}$ for the nonvanishing off-diagonal term.

When comparing this result to Eqs. (55) and (56), we thus arrive at the conclusion that in a time-dependent GL approach, we have, even to lowest order in the gradients, one diagonal dissipative coefficient for each variable entering the dynamics. This has to be contrasted to the DDFT approach outlined in the last section, where one has only three independent dissipative transport coefficients in total (even to higher order in the gradients, compare the discussion below). In addition, we find here one off-diagonal contribution $\sim \tilde{\alpha}^P$, which has no analog in Eqs. (37)–(39). By direct comparison, we find explicitly $\gamma = 2\alpha_4$, $\alpha = 2\alpha_1$, and $b = 2\alpha_4$.

As always, all the dissipative transport coefficients can depend on all scalar variables in the system including ρ_ψ and the temperature T . This general dependence on scalar quantities arises partially in the general DDFT result (55) via the factors $(1 + \psi)$ instead of 1, when compared to the CMA. If one allows also for a dependence on vector- and tensor-valued variables, such as the polarization P_i and the quadrupolar order Q_{ij} , thus giving up the assumption of strict isotropy, the picture outlined above changes as follows: the coupling terms between the force associated with S_{ij} and the forces associated with $\partial_i \mu_\psi$ and h_i^P can be mediated by the presence of a macroscopic polarization P_i . In this case, the property tensors β_{ikl} and β_{ikl}^P take the form

$$\begin{aligned} \beta_{ikl} & = \beta (\delta_{ik} P_l + \delta_{il} P_k), \\ \beta_{ikl}^P & = \beta^P (\delta_{ik} P_l + \delta_{il} P_k). \end{aligned} \quad (69)$$

Thus, these dissipative cross-coupling terms can only contribute in the presence of a macroscopic polarization. Furthermore, they bring along two additional dissipative coefficients in a dynamic GL description, while this is not the case for the DDFT [compare Eqs. (37)–(39) of Sec. II B]. Actually, there is no analog of the contribution $\sim \beta_{ikl}$ in DDFT, while for the contribution $\sim \beta_{ikl}^P$ we find $\beta^P = \alpha_4$.

One can also take into account terms containing more gradients in the dissipation function, as it has been done in the DDFT approach (55) and even for the CMA (56). To the next order in the gradients, we obtain

$$\begin{aligned} \mathfrak{R}_1^{(\text{GL})} = \int_{\mathcal{A}} d\vec{r} & \left(\frac{1}{2} \tilde{\gamma}_{ijklmn} (\partial_m S_{ij}) (\partial_n S_{kl}) + \frac{1}{2} \tilde{b}_{ijkl} (\partial_k h_i^P) (\partial_l h_j^P) \right. \\ & + \tilde{\beta}_{iklm} (\partial_i \mu_\psi) (\partial_m S_{kl}) + \tilde{\zeta}_{ijk}^P (\partial_i \mu_\psi) (\partial_k h_j^P) \\ & \left. + \tilde{\xi}_{iklmn}^P (\partial_n h_i^P) (\partial_m S_{kl}) + \tilde{\beta}_{iklm}^P h_i^P (\partial_m S_{kl}) \right). \end{aligned} \quad (70)$$

In a truly isotropic phase, the following picture emerges when Eq. (70) is analyzed: the contribution $\sim \tilde{\gamma}_{ijklmn}$ is the dissipative analog of the gradient energy for the nematic order parameter. It contains one independent material parameter in two spatial dimensions and two parameters in three spatial dimensions. In two spatial dimensions, we have $\tilde{\gamma} \sim \alpha_1$. The tensor \tilde{b}_{ijkl} has two independent parameters via

$$\tilde{b}_{ijkl} = \tilde{b}_1 \delta_{ik} \delta_{jl} + \tilde{b}_2 (\delta_{ij} \delta_{kl} + \delta_{jk} \delta_{il}), \quad (71)$$

while the tensor $\tilde{\beta}_{iklm}$ contains one independent parameter

$$\tilde{\beta}_{iklm} = \tilde{\beta}(\delta_{ik}\delta_{lm} + \delta_{il}\delta_{km}). \quad (72)$$

The same applies to the tensor $\tilde{\beta}_{iklm}^P$:

$$\tilde{\beta}_{iklm}^P = \tilde{\beta}^P(\delta_{ik}\delta_{lm} + \delta_{il}\delta_{km}). \quad (73)$$

All other contributions in Eq. (70) vanish in a truly isotropic phase. Making now an explicit comparison with DDFT, we find $\tilde{b}_1 = 4\alpha_3$, $\tilde{b}_2 = \alpha_2$, and $\tilde{\beta} = \alpha_3$, while the contribution $\sim \tilde{\beta}^P$ has no analog in DDFT. The reason for the absence of this term and two other terms mentioned further up in our DDFT result (55) lies in the fact that all missing terms contain only one gradient, while in the DDFT equation (29), from which Eq. (55) is derived, the gradients appear only quadratically. To obtain also the terms with only one gradient, one would have to generalize the DDFT equation (29) by the addition of appropriate terms that contain only one gradient and one angular derivative. As discussed below Eq. (30), such terms take into account a possible translational-rotational coupling and make the DDFT equation applicable to systems of Brownian particles with an arbitrary shape [41].

The contributions $\sim \tilde{\zeta}_{ijk}^P$ and $\sim \tilde{\xi}_{iklmn}^P$ in Eq. (70) start to contribute as soon as one allows a dependence of the property tensors on the polarization P_i . These contributions can also be associated with the general DDFT result given by Eq. (55). We note that all the contributions found in Eq. (55) can also be found in the dynamic GL approach when one allows for a dependence of the property tensors on the vector- and tensor-valued variables P_i and Q_{ij} used here. However, in contrast to DDFT, these dependencies bring along numerous additional independent coefficients.

Thus, we arrive at the conclusion that the DDFT equation (29) involves three independent dissipative coefficients in both the general case and the CMA. The corresponding terms obtained in the GL framework are associated with nine independent coefficients for the analog of the CMA, i. e., for property tensors that do not depend on the variables. We also note that there are three cross-coupling terms [see Eqs. (64) and (70)], which do not exist in the current DDFT picture.

The overall picture that emerges is therefore the following. In a dynamic GL approach, there is at least one independent dissipative coefficient for every dissipation channel (every order-parameter field) entering the description. In addition, one finds frequently dissipative cross-coupling terms that bring along further coefficients, in particular, if one considers a dependence of the dissipative property tensors on the macroscopic variables. This can be contrasted to the current DDFT picture, where one has only two dissipation channels of diagonal nature, namely, translational and rotational diffusion. In the present version of DDFT, there are also no independent dissipative cross-coupling terms. These observations clearly call for a generalization of the current DDFT equation to incorporate processes that allow for additional dissipation on a microscopic level. Since dissipative constants appear in the DDFT equation only as diffusion coefficients, such a generalization can be obtained in two steps. First of all, terms that take into account a possible translational-rotational coupling and involve further diffusion coefficients should be added to

the current form of the DDFT equation [41]. In addition, a more complicated expression for the mobility can be used [16]. This generalized mobility depends on the order-parameter fields and includes a number of additional dissipative constants. It is a necessary generalization if the hydrodynamic interaction between the colloidal particles, which is entirely neglected by Eq. (29), shall be taken into account.

IV. CONCLUSIONS AND POSSIBLE EXTENSIONS

In conclusion, we have proposed both microscopic and macroscopic theoretical descriptions for the dynamics of polar liquid crystals in two spatial dimensions. The microscopic theory is derived from DDFT, while the macroscopic formulation is based on time-dependent GL theory. We have done this by including translational density variations, polarization, and quadrupolar order as the basic order-parameter fields. Most but not all phenomenologically possible couplings of GL theory occur also in the DDFT approach. These couplings are derived from a microscopic approach, and the associated coupling parameters can be expressed as generalized moments of a molecular correlation function. We further demonstrated that the whole dynamics can be obtained from a dissipation function.

Our theoretical framework can be used for a further exploration of various dynamic processes of polar liquid crystals. This requires numerical solutions of the microscopically justified GL equations following numerical schemes proposed earlier [63,64].

For future work, it is challenging to construct a generalized DDFT that explicitly contains the momentum field as appropriate for molecular dynamics or systems in flow fields [65]. This turns out to be much more difficult than the traditional DDFT approach for simple overdamped Brownian dynamics. But, in principle, the way of generalization was explored by Tarazona, Marconi, and Melchionna [66–68] and by Archer [69,70] for molecular dynamics. An alternative derivation is based on projector techniques [71] leading to a hydrodynamical density functional theory [16,72]. Additional dynamic expressions for a colloidal liquid under shear flow were recently discussed in Ref. [65]. Furthermore, a phase-field-crystal model coupled to flow was considered by Voigt and co-workers [73] (see also Ref. [74]). In these extensions, one will presumably obtain nonvanishing microscopic expressions for phenomenological dynamic terms caused by the existence of a momentum or velocity field. These come macroscopically mainly in two groups, namely, contributions leading to a flow alignment associated with reversible currents coupling extensional flow (symmetrized velocity gradients) to orientational degrees of freedom [46,57,61,75–81] and coupling terms between extensional flow and variations of the moduli of nematic, smectic, and columnar order [46,58,59,61,82,83].

ACKNOWLEDGMENTS

We thank M. Schmiedeberg for helpful discussions. H.L. acknowledges support from the Deutsche Forschungsgemeinschaft within the science priority program SPP 1296. H.R.B. thanks the Deutsche Forschungsgemeinschaft for partial support of his work through the Forschergruppe FOR 608 ‘Nichtlineare Dynamik komplexer Kontinua’.

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