The Landau-de Gennes approach revisited: a minimal self-consistent microscopic theory for spatially inhomogeneous nematic liquid crystals

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We design a novel microscopic mean-field theory of inhomogeneous nematic liquid crystals formulated entirely in terms of the tensor order parameter field. It combines the virtues of the Landau-de Gennes approach in allowing both direction and magnitude of the local order to vary, with a self-consistent treatment of the local free-energy valid beyond the small order parameter limit. As a proof of principle, we apply this theory to the well-studied problem of a colloid dispersed in a nematic liquid crystal by including a tunable wall coupling term. For the two dimensional case, we investigate the organization of the liquid crystal and the position of the point defects as a function of the strength of the coupling constant.

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I. INTRODUCTION

Understanding how liquid crystals (LCs) behave when the spatial pattern of their preferred orientational axes is inhomogeneous is crucial for their application as optical elements in displays. A typical LCD cell has bounding surfaces, some of which have been treated to impose specific anchoring conditions on the LC molecules in order to produce a desired globally distorted state. It was realized very early on that, due to the presence of orientational order, an LC, although otherwise a freely flowing liquid, resists orientational distortions akin to a solid elastic medium. The simplest continuum theory to describe these effects is due to Oseen[1] and Frank[2]. It is a theory formulated in terms of the local nematic director $\hat{\mathbf{n}}\,(\mathbf{r})$. This unit vector describes the direction of the local preferred direction of order of the LC. The elastic energy due to the deformation of this vector field from a uniform state is then given by

$$E[\hat{\mathbf{n}}] = \frac{1}{2}\int d\mathbf{r}\ \left\{K_1(\nabla\cdot\hat{\mathbf{n}}\,(\mathbf{r}))^2 + K_2(\hat{\mathbf{n}}\,(\mathbf{r})\cdot\nabla\times\hat{\mathbf{n}}\,(\mathbf{r}))^2 + K_3(\hat{\mathbf{n}}\,(\mathbf{r})\times\nabla\times\hat{\mathbf{n}}\,(\mathbf{r}))^2\right\}\ , \qquad (1)$$

where the integration runs over the volume of the system and K_1, K_2 and, K_3 correspond to the so-called splay, twist and bend distortions respectively. The equilibrium state corresponds to the minimum of this energy subject to appropriate boundary conditions. Although the Oseen-Frank theory remains widely used for practical applications (see e.g.[3,4]), it does have an important limitation: It tacitly assumes that, while the preferred orientations may vary, the local degree of ordering does not. This means that the theory runs into problem in describing geometries where for purely topological reasons one expects the director patterns to develop singularities. Such singularities cause the free energy density to diverge. Although in 3d these singularities are integrable[5], they lead to logarithmic divergences in 2d. In practical applications, this problem is often circumvented by "excising" the singularities from the integration domain, replacing them by small domains with appropriate boundary conditions and adding an ad-hoc energy cost for each of them[6]. In reality, the core of a singularity is formed by a microscopic domain in which the net orientational order of the LC vanishes.

A well-known continuum theory that can explicitly address this shortcoming is the phenomenological Landau-De Gennes theory[7]. This theory is based on the generic Landau theory[8] of phase transitions, which posits that the local free energy of a system with a macroscopically broken symmetry can, at least in the neighbourhood of a phase transition point, be expanded as an invariant power series in an appropriate order parameter. In the standard case of nematic LCs in which the orientational symmetry is broken in an apolar fashion, the proper order parameter is a rank-2 symmetric and traceless tensor[9], conventionally denoted as $\mathbf{Q}(\mathbf{r})$. One can show[10] that the only independent invariants one can create from this tensor are $\mathbf{Q}(\mathbf{r}) : \mathbf{Q}(\mathbf{r}) = \mathrm{Tr}(\mathbf{Q}(\mathbf{r}) \cdot \mathbf{Q}(\mathbf{r})) = Q_{\mu\nu}(\mathbf{r}) Q_{\mu\nu}(\mathbf{r})$ and $\mathrm{Tr}(\mathbf{Q}(\mathbf{r}) \cdot \mathbf{Q}(\mathbf{r}) \cdot \mathbf{Q}(\mathbf{r})) = Q_{\mu\nu}(\mathbf{r}) Q_{\nu\sigma}(\mathbf{r}) Q_{\sigma\mu}(\mathbf{r})$, where here, as in the following, we adopt the Einstein convention that repeated indices are summed over. The local free energy density is then given by the expression

$$f_{\mathrm{loc}}(\mathbf{Q}(\mathbf{r})) = f_0 + \frac{1}{2} A Q_{\mu\nu}(\mathbf{r}) Q_{\mu\nu}(\mathbf{r}) - \frac{1}{3} B Q_{\mu\nu}(\mathbf{r}) Q_{\nu\sigma}(\mathbf{r}) Q_{\sigma\mu}(\mathbf{r}) + \frac{1}{4} C (Q_{\mu\nu}(\mathbf{r}) Q_{\mu\nu}(\mathbf{r}))^2, \quad (2)$$

where A is a function of the thermodynamic driving variable, either the temperature T or the density ρ, which changes sign at the critical point, and B and C are constants. Next to this local contribution, non-local contributions are constructed from derivatives $\partial_\alpha Q_{\mu\nu}(\mathbf{r})$ of the order parameter tensor. Since one is interested in the macroscopic, long wavelength, distortions only, these contributions are limited to the square gradient level. The only independent invariants one can construct of this type are $\partial_\alpha Q_{\mu\nu}(\mathbf{r}) \partial_\alpha Q_{\mu\nu}(\mathbf{r})$ and $\partial_\alpha Q_{\alpha\mu}(\mathbf{r}) \partial_\beta Q_{\beta\mu}(\mathbf{r})$. Contributions from a third possible invariant $\partial_\alpha Q_{\mu\nu}(\mathbf{r}) \partial_\nu Q_{\alpha\mu}(\mathbf{r})$ can be shown by partial integration to reduce to those of the second one up to a surface energy term (see Appendix D). This leads to a free energy of elastic distortions given by $f_{\mathrm{el}}(\nabla \otimes \mathbf{Q}(\mathbf{r})) = K_a \partial_\alpha Q_{\mu\nu}(\mathbf{r}) \partial_\alpha Q_{\mu\nu}(\mathbf{r}) + K_b \partial_\alpha Q_{\alpha\mu}(\mathbf{r}) \partial_\beta Q_{\beta\mu}(\mathbf{r})$. The final expression for the free energy is then simply

$$F[\mathbf{Q}] = \int d\mathbf{r} \, \{ f_{\mathrm{loc}}(\mathbf{Q}(\mathbf{r})) + f_{\mathrm{el}}(\nabla \otimes \mathbf{Q}(\mathbf{r})) . \quad (3)$$

In the Landau-de Gennes theory both the strength of the local order, characterized e.g. by an amplitude $Q \propto \sqrt{\mathrm{Tr}(\mathbf{Q} \cdot \mathbf{Q})}$, as well as the direction of ordering, encoded by the eigenvectors of $\mathbf{Q}$, can vary, opening up the way for a more realistic description of singularities. However, the theory still has a number of drawbacks. First, because it is based on a finite order expansion in the order parameter tensor, it does not correctly capture the fact that the strength of the local order is intrinsically bounded, and should saturate for states far away from phase boundaries. Next, all the parameters appearing in the theory are phenomenological, and therefore difficult to link consistently with an underlying molecular interaction-based models. Finally, it only considers two independent modes of elastic distortion, rather than the three correctly accounted for in the Oseen-Frank theory. This latter defect is known as the "elastic constant problem" and has a long history of theoretical attempts to address it[11–14], without a consensus solution apparent yet. In spite of its limitations, the Landau - de Gennes theory has become the workhorses of many studies on confined liquid crystals (for a review see e.g.[15]).

Arguably, the most powerful route to studying inhomogeneous LCs would be a fully microscopic approach based on classical density functional theory (DFT). Unfortunately, DFT can be both conceptually and computationally complex, and the state-of-the-art is limited to a semi-infinite fluid of hard rods in contact with a single planar hard wall[16].

Here, we propose an intermediate approach, which starts from a fully microscopic description in the form of a mean field free energy functional, with a judiciously chosen, but nevertheless very general, form of interparticle interaction. We show formally how to minimize this functional under the constraint that the local order parameter tensor $\mathbf{Q}\left(\mathbf{r}\right)$, which now appears as an ensemble average of a microscopic observable, is fixed throughout the system volume. This procedure yields a novel intermediate free energy, by construction now a functional solely of the order parameter field $\mathbf{Q}\left(\mathbf{r}\right)$. This functional thus shares the conceptual advantage of the Landau-de Gennes approach of describing the system with a limited number of readily interpreted degrees of freedom. However, this functional is fully parametrized in terms of the underlying microscopic interaction model, so in principle is free of phenomenological parameters. Moreover, we also formally show that minimizing this functional with respect to the field $\mathbf{Q}\left(\mathbf{r}\right)$, reproduces the ground state of the underlying microscopic density functional. This implies that the approach is fully thermodynamically consistent at any value of the thermodynamic driving variable. The latter property also ensures that order parameter field remains properly normalized. This in contrast to the Landau-de Gennes approach, which being a low-order expansion in powers of the order parameter, is strictly valid only in the neighbourhood of a critical point, and lacks the required self-consistency to properly bound the magnitude of $\mathbf{Q}\left(\mathbf{r}\right)$, allowing e.g. the scalar order parameter Q to take on values > 1, inconsistent with its microscopic definition. Finally, a small gradient expansion of the interaction term in our free energy functional allows the identification of the corresponding macroscopic elastic terms, with elastic constants explicitly defined in terms of the microscopic interaction potential. While arguably in the first place a conceptual advance, our approach can nevertheless also be put to practical use. The key observation is that the local non-linear self-consistency condition that enforces thermodynamic consistency in the passage from the full free energy functional to the reduced functional in terms of $\mathbf{Q}\left(\mathbf{r}\right)$ is readily implemented numerically. We illustrate this by applying our theory to a simple, yet non trivial, 2D example of a spherical colloid with homeotropic anchoring consitions, embedded in a nematic bulk. The appropriate reduced free energy functional is numerically minimized, reproducing the well-known phenomenology of this model[17].

II. FORMALISM

A. Mean Field Theory and the effective potential

Our goal is to create a *microscopic* theory for a nematic liquid crystal that has as its fundamental degrees of freedom a quantity directly identifiable as the *macroscopic* order parameter $\mathbf{Q}$. Our starting point is the standard expression for the mean-field free energy density-functional[18] of a system of uniaxial particles, whose degrees of freedom are specified by the position of their center of mass $\mathbf{r}$ and a unit vector $\hat{\omega}$ pointing along their symmetry axis:

$$\beta F[\rho^{(1)}] = \int d\mathbf{r} d\hat{\omega} \rho^{(1)}(\mathbf{r},\hat{\omega}) \left\{ \log \mathcal{V}_T \rho^{(1)}(\mathbf{r},\hat{\omega}) - 1 \right\} + \frac{1}{2}\int d\mathbf{r}_1 d\hat{\omega}_1 \int d\mathbf{r}_2 d\hat{\omega}_2 \rho^{(1)}(\mathbf{r}_1,\hat{\omega}_1)\rho^{(1)}(\mathbf{r}_2,\hat{\omega}_2) W(\mathbf{r}_1,\hat{\omega}_1;\mathbf{r}_2,\hat{\omega}_2) \quad (4)$$

We now choose the interparticle interaction (for convenience expressed in units of $k_B T$), to be the simplest one expressible as a quadratic form in terms of the microscopic "molecular anisotropy" tensor. In three dimensions this tensor has the form

$$\mathbf{q}(\hat{\omega}) = \frac{1}{2}(3\hat{\omega}\otimes\hat{\omega} - \mathbb{I}), \quad (5)$$

whereas in 2D it reads

$$\mathbf{q}(\hat{\omega}) = (2\hat{\omega}\otimes\hat{\omega} - \mathbb{I}), \quad (6)$$

whre $\mathbb{I}$ denotes the unit matrix of the appropriate dimension. The normalization of these tensors is chosen such that their maximal eigenvalue is unity. Taking into account the requirement that the effective interparticle potential is invariant under global translations and should behave as a scalar under global rotations, we arrive at the following form

$$W(\mathbf{r}_1,\hat{\omega}_1;\mathbf{r}_2,\hat{\omega}_2) = W^{(0)}(\mathbf{r}_1-\mathbf{r}_2) + \mathbf{q}(\hat{\omega}_1):\mathbf{W^{(2)}}(\mathbf{r}_1-\mathbf{r}_2)+ \\ \mathbf{W^{(2)}}(\mathbf{r}_1-\mathbf{r}_2):\mathbf{q}(\hat{\omega}_2)+\mathbf{q}(\hat{\omega}_1):\mathbf{W^{(4)}}(\mathbf{r}_1-\mathbf{r}_2):\mathbf{q}(\hat{\omega}_2) \quad (7)$$

where the colon (:) denotes the double contraction of the symmetric tensors involved. We introduce the shorthand notation $\mathbf{s} = s\hat{\mathbf{s}} = \mathbf{r}-\mathbf{r}'$ for the interparticle separation vector. The general form of the tensors $\mathbf{W}^{(k)}$ then follows from a standard tensorial construction approach, the details of which we discuss in Appendix A:

$$W^{(0)}(\mathbf{s}) = w^{(0)}(s), \quad (8)$$

$$W^{(2)}_{\mu\nu}(\mathbf{s}) = w^{(2)}(s)\hat{s}_\mu\hat{s}_\nu, \quad (9)$$

$$\begin{aligned} W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = \; & w^{(4)}_0(s)(\delta_{\mu\sigma}\delta_{\nu\tau}+\delta_{\mu\tau}\delta_{\nu\sigma})+ \\ & w^{(4)}_2(s)(\hat{s}_\mu\delta_{\nu\sigma}\hat{s}_\tau+\hat{s}_\nu\delta_{\mu\sigma}\hat{s}_\tau+\hat{s}_\mu\delta_{\nu\tau}\hat{s}_\sigma+\hat{s}_\nu\delta_{\mu\tau}\hat{s}_\sigma)+ \\ & w^{(4)}_4(s)\hat{s}_\mu\hat{s}_\nu\hat{s}_\sigma\hat{s}_\tau. \end{aligned} \quad (10)$$

The explicit form of the distance dependence of the functions $w^{(l)}_k(s)$ turns out to be largely irrelevant. In section II C and D below, we show that only full-space integrals over these functions play a role. It suffices therefore to assume that these functions are sufficiently short-ranged, with a cut-off close to the origin, to ensure intergrability.

This tensorial form of the interaction potential in Eqs. (8)-(10) was first proposed by Ronis and coworkers, who employed it in a study of the phase behaviour of a lattice model of liquid crystals[19,20].We also note that in 3D this representation is equivalent to starting from a generic invariant expansion in spherical harmonics of the effective potential[21,22] of the form

$$W_{3D}(\mathbf{r}_1,\hat{\omega}_1;\mathbf{r}_2,\hat{\omega}_2) = \sum_{l_1,l_2,L}\sum_{m_1,m_2,M} w_{l_1,l_2,L}(s)\begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & M\end{pmatrix} Y^m_{l_1}(\hat{\omega}_1)\,Y^m_{l_1}(\hat{\omega}_2)\,Y^m_{l_1}(\hat{\mathbf{s}}) \quad (11)$$

and requiring that only terms with $l_1, l_2 = 0, 2$ are allowed, as first discussed in[23]. One checks that the angular momentum selection rules coded into the $3J$ symbol[24] than dictates that the only terms contributing (up to interchange of the two particles) are $(l_1, l_2, L) =$ $(0,0,0)$,$(2,2,0)$,$(2,0,2)$,$(2,2,2)$and $(2,2,4)$, yielding indeed 5 independent terms. However, in 2D the corresponding expansion is

$$W_{2D}(\mathbf{r}_1,\hat{\omega}_1;\mathbf{r}_2,\hat{\omega}_2) = \sum_{m_1,m_2,M} w_{m_1,m_2,M}(s)\,\delta(m_1+m_2+M)\,e^{im_1\theta_1}e^{im_2\theta_2}e^{iM\psi} \quad (12)$$

where θ_1, θ_2 and ψ are the angles the particle axis and their separation vector make with the reference frame respectively. Again one now restricts the orientation modes to be restricted to $m_1, m_2 = 0, 2$. This leaves as independent terms, up to particle interchange and global sign inversion (neccessary for the potential to be real-valued), $(m_1, m_2, M) = (0,0,0), (2,-2,0), (2,0,-2)$and $(2,2,-4)$, i.e. just 4 independent components. Indeed, one checks in this case that the double contractions of the molecular anisotropy tensors onto the terms labelled by $w_0^{(4)}$ and $w_2^{(4)}$ lead to the same result $\propto \mathbf{q}(\hat{\omega}_1) : \mathbf{q}(\hat{\omega}_2)$, allowing these two terms to be collected in to a single one (see Appendix B for details).

The utility of the tensorial form of the effective potential Eq. (7), as compared to the expansions in the appropriate harmonics, will become clear in the following, as it immediately allows us to write the non-ideal part of the free energy in terms of the macroscopic order parameter $\mathbf{Q}$.

B. The constrained free energy functional

We will confine ourselves to phases with a spatially homogeneous center of mass density distribution. We therefore introduce the local orientational distribution function $\psi(\mathbf{r}, \hat{\omega})$, which we will further abbreviate as ODF, through

$$\rho^{(1)}(\mathbf{r}, \hat{\omega}) = \rho\psi(\mathbf{r}, \hat{\omega}), \tag{13}$$

with $\rho = N/V$, where N is the number of particles and V the volume of the system, the constant number density. The ODF allows us to define the local orderparameter tensor as the average over the molecular anisotropy tensor

$$\mathbf{Q}(\mathbf{r}) = \int d\hat{\omega}\, \mathbf{q}(\hat{\omega})\psi(\mathbf{r}, \hat{\omega}). \tag{14}$$

The free energy as a functional of the ODF then reads

$$\beta F[\psi] = N \log \nu_T \rho + \frac{1}{2}\Omega_d N \rho \overline{w^{(0)}} + \rho \int d\mathbf{r} d\hat{\omega}\, \psi(\mathbf{r}, \hat{\omega}) \log \psi(\mathbf{r}, \hat{\omega}) + \frac{1}{2}\rho^2 \int d\mathbf{r} d\mathbf{r}'\, \mathbf{Q}(\mathbf{r}) : \mathbf{W}^{(4)}(\mathbf{r} - \mathbf{r}') : \mathbf{Q}(\mathbf{r}') \tag{15}$$

where Ω_d is the area of the unit ball of dimension d, and the overbar denotes radial averaging, i.e.

$$\overline{f} = \int_0^\infty dr\; r^{d-1} f(r). \tag{16}$$

Note that because of our assumptions of spatial homogeneity of the center of mass distribution the coupling of the individual particle orientations to the inter-particle separation vector, as described by the $\mathbf{W}^{(2)}$ terms in the interaction, does not play a role, since

$$\int d\mathbf{r}'\,\mathbf{W}^{(\mathbf{2})}(\mathbf{r}-\mathbf{r}') \propto \mathbb{I} \tag{17}$$

and $\mathbf{Q}(\mathbf{r}) : \mathbb{I} = Tr\left(\mathbf{Q}(\mathbf{r})\right) = 0$, as by definition $\mathbf{Q}$ is traceless.

To obtain the free energy as a functional of the local order parameter tensor only, we now perform a constrained minimization keeping the value of the local order parameter fixed through a lagarange multiplier field that can also be interpreted as an external field. The idea is that the *intrinsic* free energy of system in an equilibrium with an external field that causes a certain deviation from the field-free equilibrium ODF can be interpreted as the non-equilibrium free energy of the distorted state. This procedure is simply an expression of the central tenet of density functional theory viz. the one-to-one correspondence between external field and equilibrium densities[25]. The average interaction energy in our mean field model already depends only on $\mathbf{Q}$, so that it is sufficient to perform the minimization of the orientational entropy term. We thus consider the following stationarity condition

$$\begin{aligned}\frac{\delta}{\delta\psi(\mathbf{r},\hat{\omega})}\left\{\rho\int d\mathbf{r}d\hat{\omega}\,\psi(\mathbf{r},\hat{\omega})\log\psi(\mathbf{r},\hat{\omega}) + \int d\mathbf{r}\mu(\mathbf{r})\left\{1-\int d\hat{\omega}\,\psi(\mathbf{r},\hat{\omega})\right\} - \right.\\ \left.\rho\int d\mathbf{r}\,\mathbf{\Lambda}(\mathbf{r}) : \left\{\mathbf{Q}(\mathbf{r}) - \int d\hat{\omega}\,\mathbf{q}(\hat{\omega})\psi(\mathbf{r},\hat{\omega})\right\}\right\} = 0\end{aligned} \tag{18}$$

Performing the variation and eliminating the field $\mu(\mathbf{r})$ which takes care of the normalization of the ODF we get

$$\psi(\mathbf{r},\hat{\omega}) = \frac{\exp\left\{-\mathbf{\Lambda}(\mathbf{r}) : \mathbf{q}(\hat{\omega})\right\}}{\zeta(\mathbf{r})}, \tag{19}$$

where the normalization factor is given by

$$\zeta(\mathbf{r}) = \int d\hat{\omega}\exp\left\{-\mathbf{\Lambda}(\mathbf{r}) : \mathbf{q}(\hat{\omega})\right\}. \tag{20}$$

The tensor field $\mathbf{\Lambda}(\mathbf{r})$ now has to be chosen such that the constraint

$$\mathbf{Q}(\mathbf{r}) = \int d\hat{\omega}\,\mathbf{q}(\hat{\omega})\frac{\exp\left\{-\mathbf{\Lambda}(\mathbf{r}) : \mathbf{q}(\hat{\omega})\right\}}{\zeta(\mathbf{r})} \tag{21}$$

is satisfied. Let us denote the solution to this problem by $\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r})$. Note that since any 2-tensor can be written as $\mathbf{A} = \frac{1}{d}\,\mathrm{Tr}\left(\mathbf{A}\right)\mathbb{I} + \mathbf{A}_0$, where $\mathrm{Tr}\left(\mathbf{A}_0\right) = 0$ and $\mathbb{I} : \mathbf{q}(\hat{\omega}) = \mathrm{Tr}\left(\mathbf{q}(\hat{\omega})\right) = 0$, it suffices to assume that $\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r})$ is also traceless.

The constrained free energy is then given by

$$\beta F[\mathbf{Q}] = -\rho \int d\mathbf{r} \log \zeta_{\mathbf{Q}}(\mathbf{r}) - \rho \int d\mathbf{r} \mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r}) : \mathbf{Q}(\mathbf{r}) + \\ \frac{1}{2}\rho^2 \int d\mathbf{r} d\mathbf{r}' \mathbf{Q}(\mathbf{r}) : \mathbf{W}^{(4)}(\mathbf{r}-\mathbf{r}') : \mathbf{Q}(\mathbf{r}') + \beta F_0, \quad (22)$$

where $\beta F_0 = N \log \nu_T \rho + \frac{1}{2}\Omega_d N \rho \overline{w^{(0)}}$. Note that the non-constant terms of the above expression of the free energy do not depend on explicitly on dimensionality, taking the same form for both two and three dimensions. It is now easy to verify that varying this functional with respect to $\mathbf{Q}$ yields the usual selfconsistency condition. Indeed,

$$\frac{\delta \beta F[\mathbf{Q}]}{\delta \mathbf{Q}(\mathbf{r})} = \rho \frac{\delta \mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r})}{\delta \mathbf{Q}(\mathbf{r})} : \left\{ \int d\hat{\omega}\, \mathbf{q}(\hat{\omega}) \frac{e^{-\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r}):\mathbf{q}(\hat{\omega})}}{\zeta_{\mathbf{Q}}(\mathbf{r})} - \mathbf{Q}(\mathbf{r}) \right\} \\ - \rho \left\{ \mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r}) - \rho \int d\mathbf{r}'\; \mathbf{W}^{(4)}(\mathbf{r}-\mathbf{r}') : \mathbf{Q}(\mathbf{r}') \right\} = 0$$

The first term is zero due to the constraint condition, leaving the standard prescription for the equilibrium mean-field

$$\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{r}) = \rho \int d\mathbf{r}'\, \mathbf{W}^{(4)}(\mathbf{r}-\mathbf{r}') : \mathbf{Q}(\mathbf{r}') \quad (23)$$

which could also have been obtained by minimizing the functional Eq. (15) directly with respect $\psi(\hat{\omega})$, showing that the constrained variation followed by a relaxation of the constraints yields the same result as expected. This result also clarifies the nature of the approximation implied by our approach; it provides a cumulant expansion of the ODF with the effective field truncated to terms linear in $\mathbf{Q}$. The ODF in turn can be used to predict the values of higher order tensorial order parameters, which are implicitly accounted for.

C. The homogeneous equilibrium state

If we assume that the state is fully homogeneous, Eq. (23) reduces to

$$\mathbf{\Lambda}_{\mathbf{Q}} = \rho \left\{ \int d\mathbf{s}\, \mathbf{W}^{(4)}(\mathbf{s}) \right\} : \mathbf{Q} \quad (24)$$

Using the results of Appendix C we find that in $3D$

$$\int d\mathbf{s}\, W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = 4\pi \overline{w_0^{(4)}} \left(\delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right) + \quad (25)$$

$$\frac{8\pi}{3} \overline{w_2^{(4)}} \left(\delta_{\mu\tau}\delta_{\nu\sigma} + \delta_{\mu\sigma}\delta_{\nu\tau}\right) + \quad (26)$$

$$\frac{4\pi}{15} \overline{w_4^{(4)}} \left(\delta_{\mu\nu}\delta_{\sigma\tau} + \delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right), \quad (27)$$

while in $2D$

$$\int d\mathbf{s}\, W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = 2\pi\overline{w_0^{(4)}}\left(\delta_{\mu\sigma}\delta_{\nu\tau}+\delta_{\mu\tau}\delta_{\nu\sigma}\right)+ \tag{28}$$

$$2\pi\overline{w_2^{(4)}}\left(\delta_{\mu\tau}\delta_{\nu\sigma}+\delta_{\mu\sigma}\delta_{\nu\tau}\right)+ \tag{29}$$

$$\frac{\pi}{4}\overline{w_4^{(4)}}\left(\delta_{\mu\nu}\delta_{\sigma\tau}+\delta_{\mu\sigma}\delta_{\nu\tau}+\delta_{\mu\tau}\delta_{\nu\sigma}\right), \tag{30}$$

In both cases this leads to

$$\mathbf{\Lambda}_{\mathbf{Q}} = \rho w_d \mathbf{Q}, \tag{31}$$

where w_d is a single temperature and dimension dependent coupling constant. The order-parameter itself now follows from the consistency equation

$$\mathbf{Q} = \int d\hat{\omega}\, \mathbf{q}(\hat{\omega}) \frac{e^{-\rho w_d \mathbf{Q}:\mathbf{q}(\hat{\omega})}}{\zeta_{\mathbf{Q}}} \tag{32}$$

The generic uniaxial solution to Eq. (32) is given by

$$\mathbf{Q}_0 = Q\mathbf{q}(\hat{\mathbf{n}}) \tag{33}$$

where $\hat{\mathbf{n}}$ is a unit vector, and Q the so-called scalar orderparameter. The selfconsistent equation for Q can be determined e.g. by taking the innerproduct of Eq. (32) with $\mathbf{q}(\hat{\mathbf{n}})$ and taking the trace. We note that $\mathrm{Tr}\left(\mathbf{q}(\hat{\mathbf{n}})\cdot\mathbf{q}(\hat{\omega})\right) = \frac{d}{d-1}P_2^{(d)}\left(\hat{\mathbf{n}}\cdot\hat{\omega}\right)$, where $P_2^{(2)}\left(\hat{\mathbf{n}}\cdot\hat{\omega}\right) = \cos 2\theta$ and $P_2^{(3)}\left(\hat{\mathbf{n}}\cdot\hat{\omega}\right) = \frac{1}{2}\left(3\cos^2\theta - 1\right)$. Thus

$$Q = \frac{\int d\hat{\omega}\, P_2^{(d)}\left(\hat{\mathbf{n}}\cdot\hat{\omega}\right) e^{-\frac{d}{d-1}\rho w_d Q P_2^{(d)}(\hat{\mathbf{n}}\cdot\hat{\omega})}}{\int d\hat{\omega}\, e^{-\frac{d}{d-1}\rho w_d Q P_2^{(d)}(\hat{\mathbf{n}}\cdot\hat{\omega})}} \tag{34}$$

which one recognizes as the standard Maier-Saupe theory results[26,27].

D. Expansion around the homogeneous equilibrium state

We are now going to look at the free energy cost of perturbations around the uniaxially aligned nematic equilibrium state. We first note that the first two terms of the constrained free energy are already entirely local, and the final constant term does not depend on the order parameter. We therefore only need to consider the interaction term

$$
\begin{aligned}
\beta F_{int}[\mathbf{Q}] &= \frac{1}{2}\rho^2 \int d\mathbf{r}d\mathbf{r}'\,\mathbf{Q}(\mathbf{r}) : \mathbf{W}^{(4)}(\mathbf{r}-\mathbf{r}') : \mathbf{Q}(\mathbf{r}') \\
&= \frac{1}{2}\rho^2 \int d\mathbf{R}d\mathbf{s}\,\mathbf{Q}(\mathbf{R}+\frac{1}{2}\mathbf{s}) : \mathbf{W}^{(4)}(\mathbf{s}) : \mathbf{Q}(\mathbf{R}-\frac{1}{2}\mathbf{s}) \qquad (35)\\
&\equiv \frac{1}{2}\rho^2 \int d\mathbf{R}\,\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] \qquad (36)
\end{aligned}
$$

where $\mathbf{R} = \frac{1}{2}\left(\mathbf{r}+\mathbf{r}'\right)$ is the center of mass coordinate and $\mathbf{s}$ the previously defined separation vector. As we are primarily interested in case where the orientational distortions vary only slowly on the molecular scale, we can simplify the effective interaction (36) by expanding $\mathbf{Q}$ in to second order in its derivatives as

$$
\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] \simeq \int d\mathbf{s}\left(\mathbf{Q}(\mathbf{R}) + \frac{1}{2}\left(\mathbf{s}\cdot\nabla\right)\mathbf{Q}(\mathbf{R})\right) : \mathbf{W}^{(4)}(\mathbf{s}) : \left(\mathbf{Q}(\mathbf{R}) - \frac{1}{2}\left(\mathbf{s}\cdot\nabla\right)\mathbf{Q}(\mathbf{R})\right). \qquad (37)
$$

Recalling that the expansion of the fourth rank tensor given by Eq. (10) only contains terms involving products of an even number of compeonents of the the unit vector $\hat{\mathbf{s}}$ and that by symmetry all intergrals over products of an odd number of components of $\hat{\mathbf{s}}$ vanish, this can be reduced to

$$
\begin{aligned}
\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] &= \mathbf{Q}(\mathbf{R}) : \int d\mathbf{s}\,\mathbf{W}^{(4)}(\mathbf{s}) : \mathbf{Q}(\mathbf{R}) - \frac{1}{4}\int d\mathbf{s}\,\left(\mathbf{s}\cdot\nabla\right)\mathbf{Q}(\mathbf{R}) : \mathbf{W}^{(4)}(\mathbf{s}) : \left(\mathbf{s}\cdot\nabla\right)\mathbf{Q}(\mathbf{R}) \\
&= \left\{\int d\mathbf{s}\,W^{(4)}_{\mu\nu\tau\sigma}(\mathbf{s})\right\} Q_{\mu\nu}\left(\mathbf{R}\right) Q_{\tau\sigma}\left(\mathbf{R}\right) - \frac{1}{4}\left\{\int d\mathbf{s}\,s_\alpha s_\beta W^{(4)}_{\mu\nu\tau\sigma}(\mathbf{s})\right\} \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\tau\sigma}\left(\mathbf{R}\right)
\end{aligned}
$$

where in the component representation we have adopted the Einstein summation convention. The remaining integrals over the separation vector can be worked out analytically both in 2 dimensions as well as in 3 dimensions. The relevant details are given in Appendix C and D

III. APPLICATION

As a proof-of-principle application of our theory we will consider the case of a 2D system with an embedded circular colloidal particle, that couples to the liquid crystal through an explicit anchoring interaction. This system has been studied using standard continuum theory, supplemented with ad hoc prescriptions for dealing with the inevitable singularities in the director field[5,28–30]. Later, in the context of directed assembly of colloids dispersed in a nematic host, Landau- de Gennes theory has been used to study the defects formed around the colloids and how these can be used for engineering the self-assembly of more complex structures[31,32]. We first derive a more explicit form of the 2D functional, which will be used in our numerical calculations. We then introduce the anchoring conditions on the colloid. Next, we briefly discuss our numerical procedure and present results for a number parameter values.

A. Details of the 2D free energy functional

The explicit form for the 2D free energy functional is given by

$$\beta F[\mathbf{Q}] = -\rho \int d\mathbf{R} \log \zeta_{\mathbf{Q}}(\mathbf{R}) - \rho \int d\mathbf{R}\, \mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) : \mathbf{Q}(\mathbf{R}) \tag{38}$$
$$-\frac{1}{2}\rho U \int d\mathbf{R}\, \mathbf{Q}\left(\mathbf{R}\right) : \mathbf{Q}\left(\mathbf{R}\right) + \frac{1}{2}\rho K \int d\mathbf{R}\, \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right),$$

where the explicit form of the coupling constant U and the single elastic constant K are discussed in Appendix D (Eq. (D3) and (D4) respectively). We now choose a convenient representation for the orderparameter tensor

$$\mathbf{Q}(\mathbf{R}) = Q\left(\mathbf{R}\right)\left\{2\hat{\mathbf{n}}\left(\mathbf{R}\right) \otimes \hat{\mathbf{n}}\left(\mathbf{R}\right) - \mathbb{I}\right\} \tag{39}$$

where $Q\left(\mathbf{R}\right)$ is the local scalar order parameter, and the local director is given by $\hat{\mathbf{n}}\left(\mathbf{R}\right) = \left(\cos\alpha\left(\mathbf{R}\right), \sin\alpha\left(\mathbf{R}\right)\right)$, with α the angle with repect to the x-axis of the reference frame.

Since $\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R})$ is traceless, we can parametrize is as $\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) = \Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)\left\{2\hat{\mathbf{m}}\left(\mathbf{R}\right) \otimes \hat{\mathbf{m}}\left(\mathbf{R}\right) - \mathbb{I}\right\}$, where $\hat{\mathbf{m}}\left(\mathbf{R}\right)$ is an as yet undetermined unit vector. We now show that it must be equal to the local director $\hat{\mathbf{n}}\left(\mathbf{R}\right)$. Consider the contraction

$$\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) : \mathbf{Q}(\mathbf{R}) = 2\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right) Q\left(\mathbf{R}\right)\left\{2\left(\hat{\mathbf{m}}\left(\mathbf{R}\right) \cdot \hat{\mathbf{n}}\left(\mathbf{R}\right)\right)^2 - 1\right\} \tag{40}$$

On the other hand, using the selfconsistency condition Eq. (21), we have

$$\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) : \mathbf{Q}(\mathbf{R}) = \int d\hat{\omega}\, \mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) : \mathbf{q}(\hat{\omega}) \frac{\exp\left\{-\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R}) : \mathbf{q}(\hat{\omega})\right\}}{\zeta(\mathbf{R})} \tag{41}$$

The integrand on the right hand side is readily seen to be a function only of the innerproduct $\hat{\mathbf{m}}\left(\mathbf{R}\right) \cdot \hat{\omega}$. Since we are integrating over all angles, this integral does not depend on the choice of $\hat{\mathbf{m}}\left(\mathbf{R}\right)$. This is only consistent with Eq. (40) if $\hat{\mathbf{m}}\left(\mathbf{R}\right) = \hat{\mathbf{n}}\left(\mathbf{R}\right)$, showing that $\mathbf{\Lambda}_{\mathbf{Q}}(\mathbf{R})$ has the same principal directions as $\mathbf{Q}(\mathbf{R})$. To obtain the scalar amplitude $\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)$, we use Eq. (41) to obtain

$$Q\left(\mathbf{R}\right) = \frac{\int_0^{2\pi} d\theta \cos 2\theta e^{-2\Lambda_{\mathbf{Q}}(\mathbf{R})\cos 2\theta}}{\int_0^{2\pi} d\theta e^{-2\Lambda_{\mathbf{Q}}(\mathbf{R})\cos 2\theta}} = \frac{I_1\left(2\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)\right)}{I_0\left(2\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)\right)}, \tag{42}$$

where $\cos\theta = \hat{\mathbf{n}}\left(\mathbf{R}\right)\cdot\hat{\omega}$, $I_k\left(z\right)$ is a modified Bessel function, and we have used entry (9.6.19) of Abramovitz & Stegun[33] to evaluate the integrals. While this relation cannot be inverted analytically, a simple non-linear root finding procedure will provide an accurate numerical determination of $\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)$ given $Q\left(\mathbf{R}\right)$.

We are now in a position to give the form of the functional we will actually use in our calculations

$$\begin{aligned}\beta F[\mathbf{Q}] = &-\rho\int d\mathbf{R}\,\log\left(2\pi I_0\left(2\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)\right)\right) - 2\rho\int d\mathbf{R}\,\Lambda_{\mathbf{Q}}\left(\mathbf{R}\right)Q\left(\mathbf{R}\right) - \rho U\int d\mathbf{R}\,Q^2\left(\mathbf{R}\right) \\ &+\rho K\int d\mathbf{R}\,\left\{\left(\partial_x Q\left(\mathbf{R}\right)\right)^2 + \left(\partial_y Q\left(\mathbf{R}\right)\right)^2 + 2Q^2\left(\mathbf{R}\right)\left(\left(\partial_x\alpha\left(\mathbf{R}\right)\right)^2 + \left(\partial_y\alpha\left(\mathbf{R}\right)\right)^2\right)\right\}.\end{aligned} \tag{43}$$

B. Bulk behaviour compared to standard Landau-de Gennes theory

In order to illustrate how the present approach produces consistent results for all values of the temperature, we compare its predictions for the behaviour of the spatially homogeneous bulk phase to that of the appropriate Landau- de Gennes theory. The latter is obtained within our model by expanding the ideal part of the free energy

$$\frac{\beta F_{id}\left[\mathbf{Q}\right]}{N} = -\log\left(2\pi I_0\left(2\Lambda_{\mathbf{Q}}\right)\right) - 2\Lambda_{\mathbf{Q}}Q, \tag{44}$$

in powers of the scalar order parameter Q. This is readily done by expanding the effective field $\Lambda_{\mathbf{Q}}$ as follows

$$\Lambda\left(Q\right) = \lambda_0 + \lambda_1 Q + \lambda_2 Q^2 + \lambda_3 Q^3 + \lambda_4 Q^4 + \lambda_5 Q^5 + \cdots, \tag{45}$$

substituting this expansion into the selfconsistency relations Eq. (42), and constructing the order-by-order solution. We find

$$\frac{\beta F_{id}\left[Q\right]}{N} = -\log 2\pi + Q^2 + \frac{1}{4}Q^4 + \frac{5}{36}Q^6 + O\left(Q^8\right). \tag{46}$$

The bulk Landau- de Gennes free energy is thus

$$\frac{\beta F_{LdG}\left[Q\right]}{N} = -\log 2\pi + Q^2 + \frac{1}{4}Q^4 + \frac{5}{36}Q^6 - UQ^2. \tag{47}$$

Here, the dimensionless coupling constant U plays the role of the inverse temperature. We note that this expression is also often already truncated at the fourth order term, which is

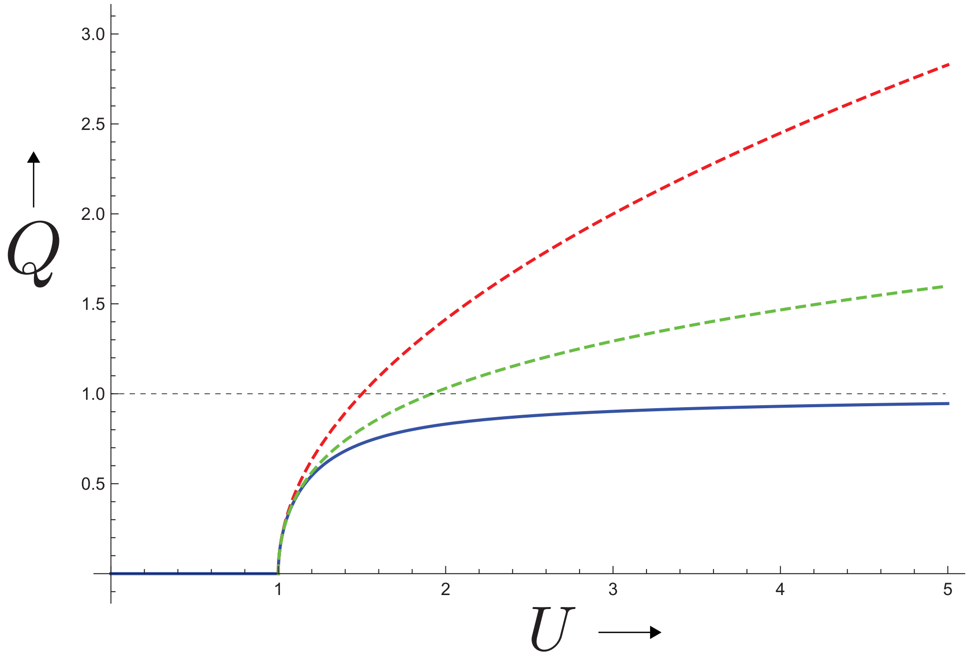

FIG. 1. Scalar order parameter in the homogeneous bulk as function of the coupling constant U for our theory (solid blue line) compared to the corresponding Landau- de Gennes theory with up to 4th order terms (dashed red line), and up to 6th order terms (dashed green line). The thin dashed line marks the upper bound $Q = 1$.

sufficient to ensure that one obtains bounded positive solutions. The solutions of the free energy minimization problem are found to be

$$Q = \begin{cases} 0 & & U < 1 \\ \sqrt{2(U-1)} & \text{4th order approximation} & \\ \sqrt{\frac{1}{5}\left(\sqrt{60U-51}-3\right)} & \text{6th order approximation} & U \geq 1 \end{cases} . \tag{48}$$

These results should be compared to the results of the full theory, which are obtained by considereing Eq. (34) for the 2D case, yielding the self-consistency equation

$$Q = \frac{\int_0^{2\pi} d\theta\ \cos 2\theta e^{-2UQ\cos 2\theta}}{\int_0^{2\pi} d\theta\, e^{-2UQ\cos 2\theta}} = \frac{I_1\left(2UQ\right)}{I_0\left(2UQ\right)}, \tag{49}$$

which needs to be solved numerically. The results are shown in Figure 1 Clearly, both versions of the Landau- de Gennes theory start deviating from the solution of the full theory just beyond the common critical point $U_* = 1$, and in fact rapidly violate the upper bound $Q = \langle cos2\theta\rangle \leq 1$.

C. The anchoring interaction

The free energy functional of our model (43) only describes the interactions within the bulk of the liquid crystal. To mimic the preferential orientation of the nematic induced by surface of the colloid, we need to add an anchoring term to this free energy. We choose this anchoring free energy to be of the form

$$\beta F_S[\rho^{(1)}(\mathbf{r}),\hat{\omega}] = \oint_{\partial A} \mathrm{d}s\, f_S(\hat{\mathbf{b}}(\mathbf{R}),\mathbf{Q}(\mathbf{R})), \tag{50}$$

where the local anchoring energy density is chosen to reproduce the classic Rapini-Papoular expression[34],

$$\begin{aligned} f_S(\hat{\mathbf{n}}(\mathbf{R}),\mathbf{Q}(\mathbf{R})) &= \frac{1}{2\sqrt{2}}\frac{W\mathbf{Q}(\mathbf{R}) : \hat{\mathbf{b}}(\mathbf{R})\otimes\hat{\mathbf{b}}(\mathbf{R})}{\sqrt{\mathbf{Q}(\mathbf{R}) : \mathbf{Q}(\mathbf{R})}} \\ &= \frac{W}{2}\left(\hat{\mathbf{b}}(\mathbf{R})\cdot\hat{\mathbf{n}}(\mathbf{R})\right)^2 + \ \mathit{constant}. \end{aligned} \tag{51}$$

Here $\hat{\mathbf{b}}(\mathbf{R})$ is the outward unit normal to the surface of the colloid and $\hat{\mathbf{n}}(\mathbf{R})$ the local director. For $W < 0$ this term favors radial anchoring to the surface of the colloid. Note that, due to the normalisation, this anchoring term only controls the orientation direction of the nematic but does not lead to an increase in the degree of order around the colloid. Although one can argue about the merits of this form of anchoring potential, it has the virtue of simplicity, and we have also used it in previous work[35] thus facilitating comparisons across systems.

D. Numerical procedure

Our free energy functional cannot be minimized analytically. We therefore proceed to minimize it numerically by discretizing the tensor order parameter on a square lattice covering a square region (of side lenght L) which is centered on the colloidal particle of radius R_{coll}. Initially each point on our lattice is attributed a random orientation and random value of the scalar order parameter with the exception of the lattice points at the exterior border of the simulation box, which are set to a chosen direction . At the surface of the colloid the nematic is coupled by the interaction given in the previous subsection, whereas at the exterior edges the tensor order parameter is set to the expected bulk value, obtained from a separate calculation, and the direction is fixed along the y axis. Since our free energy features a number of local minima in addition to the global one, we make use of an adaptive simulated annealing technique[36], rather than employing a standard Monte Carlo technique. The simulated annealing technique allows us to cool the system progressively. In this way, in the beginning of the relaxation procedure non energetically favorable moves are more frequent, allowing the system to escape from local minima. The temperature is then progressively lowered during the course of the minimization, and the number of moves that increase the energy steadily decreases. We use an adaptive technique, where, in addition to progressively cooling the system, we also decrease the amplitude of the Monte Carlo trial moves as the system approaches its minimum. This latter approach significantly speeds up to the relaxation procedure. The initial configuration does not influence the final state of the system. Different initial conditions can, however, influence the time needed for the system to equilibrate.

E. Results

Using our theory we investigate the patterns formed by the liquid crystal in the vicinity a colloidal particle of fixed radius R_{coll} with radial anchoring as a function of the anchoring strength W. We choose to carry out the minimization for $U = 1.25$ and $K = 0.00005$, which corresponds to a bulk value of the scalar order parameter of $Q \approx 0.6$. For high values of the surface coupling constant W, we find that the nematic in the close proximity of the colloid is arranged radially and forms, at a distance from the surface, two point defects of topological charge 1/2 located in the equatorial plane (see figure 2: (a) - (c)). These points represent the planar projection of the "Saturn ring" configuration and feature not only a discontinuity in the nematic director but also, as expected, a decrease in the scalar order parameter. By lowering the coupling constant, we see that the ring contracts directly onto the surface of the colloid (see figure 2: (d) - (f)). Here as well we observe the co-localized decrease of the scalar order parameter with the orientation discontinuity. If the coupling strength is further lowered the two point defects retract inside the the colloid and only a bend in the nematic orientation is observed as can be seen in figure 2: (g) - (i).

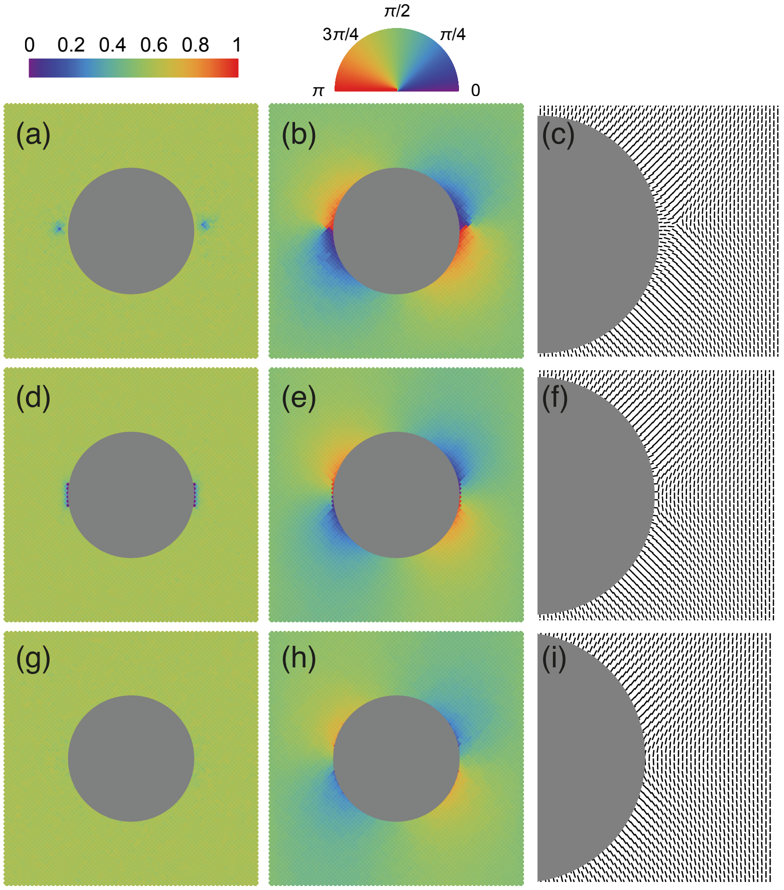

FIG. 2. Scalar order parameter (panels (a), (d), and (g)), corresponding nematic director (panels (b), (e), and (h)), and enlarged image the nematic director (panels (c), (f), and (i)) around a colloid with radius $R_{coll}/L = 0.25$ for different anchoring strengths: $W = -1$ (panels (a)- (c)), $W = -0.05$ (panels (d) - (f)), and $W = -0.01$ (panels (g) -(i)) . All results were obtained for $U = 1.25$ and $K = 0.00005$.

IV. DISCUSSION

The theory we have presented here provides a systematic microscopic underpinning to the Landau-de Gennes theory of inhomogeneous liquid crystals. It achieves this by "projecting" a microscopic mean-field density functional onto the subspace of solutions with fixed values for the field of local macroscopic order parameter tensors $\mathbf{Q}(\mathbf{r})$. Combined with a specific choice of the interaction potential, this leads to a tractable theory with a local free energy density that now correctly captures the saturating behaviour of the degree of order at large values of the thermal coupling constant. In this sense it goes beyond the limitations implicit in the Landau expansion, which strictly speaking is only valid sufficiently close to the isotropic high temperature state. The latter property is certainly relevant for the isotropic-nematic transition, which, albeit a weak first order transition[37], does transition into a state with a significant degree of order.

Obviously, the theory has a number of intrinsic limitations. Being a mean-field theory it strongly overestimates the first order nature of the bulk phase transition, as it neglects relevant fluctuations. However, this limitation it basically shares with all microscopic theoretical approaches to liquid crystals, which invariably rely on strongly approximated free energy expressions. The theory also neglects the effects of changes in the local particle density. In principle, the present approach could be extended to deal with spatially varying densities, by conditioning the microscopic functional on a fixed number density field $\rho(\mathbf{r})$. This involves the introduction of an effective field conjugate to this density as well as a significant jump in complexity, as the coupling between the density and orientational inhomogeneities comes in to play. Moreover, to properly account for the compressibility of the system, the simple mean-field interaction term in the free energy would have to be replaced by a more involved one. A suitable candidate in this respect would be a classical Parsons/Lee type approximation[38], which factorises the density dependence and the orientational dependence of the excess free energy, in a way that remains consistent with our approach. Finally, by construction, the free energy density of inhomogeneous distortions of the homogeneous uniaxial equilibrium state, reproduces the results of the Landau-de Gennes approach, including its failure to generate three independent elastic constants. This defect could in principle be remedied by adding additional terms in the elastic free energy involving higher powers[12] in $\mathbf{Q}(\mathbf{r})$, which, however, introduces additional generalized elastic constants that are not connected to the underlying microscopic model.

We have also shown that the non-linear inversion required to eliminate the field that enforces the constraint on the solution space of the free-energy minimization problem is computationally tractable in the 2D example treated here. The theory itself, however, is already fully formulated in 3D. It is easy to see by counting degrees of freedom that the 3D case will involve no more than the local elimination of two scalars that characterize the constraint-field, one corresponding to fixing the degree of order with respect to the preferred axis and the other to the degree of local biaxiality around this axis. We plan to return to this issue in future work, which would allow e.g. the study of the temperature dependence of the intriguing interactions between colloidal induced by entanglement with disclination lines[32,39].

Finally, it will be interesting to extend the present approach to more complex molecular geometries, such as biaxiality and chirality. Especially the latter case would allow the theoretical exploration of the undoubtedly subtle interplay between confinement geometries, boundary conditions and chiral phases (see e.g.[40]).

Appendix A: Derivation of the effective interaction

The most general interaction expressible as a quadratic in the microscopic polarizability tenor $\mathbf{q}$ is conveniently written as

$$\begin{aligned} W(\mathbf{r},\hat{\omega};\mathbf{r}',\hat{\omega}') = \mathbf{W}^{(0)}(\mathbf{r}-\mathbf{r}') + &\\ \mathbf{q}(\hat{\omega}) : \mathbf{W}_1^{(2)}(\mathbf{r}-\mathbf{r}') + \mathbf{W}_2^{(2)}(\mathbf{r}-\mathbf{r}') : \mathbf{q}(\hat{\omega}') + \mathbf{q}(\hat{\omega}) : \mathbf{W}^{(4)}(\mathbf{r}-\mathbf{r}') : \mathbf{q}(\hat{\omega}') \end{aligned}$$

This expression already takes care of translation invariance. We still need to insure that it is properly symmetric under the interchange of the two sets of particle coordinates and that the $\mathbf{W}^{(k)}$ actually are tensors of the proper rank. First, we need to introduce a shorthand notation for the separation vector $\mathbf{s} = s\hat{\mathbf{s}} = \mathbf{r}_1 - \mathbf{r}_2$. The tensors can now be built using the standard isotropic tensors $\delta_{\mu\nu}$ and $\epsilon_{\mu\nu\tau}$, the unit vector pointing along the separation vector $\hat{\mathbf{s}}$ and scalars which can be a function of the magnitude s of the interparticle separation. The basis for the zero'th rank tensors is trivially given by a constant. A basis for the second rank tensors is given by

$$\delta_{\mu\nu} \tag{A1}$$

$$\hat{s}_\mu \hat{s}_\nu \tag{A2}$$

A basis for the fourth rank tensors constructed from these ingredients is given by

$$\delta_{\mu\nu}\delta_{\sigma\tau} \tag{A3}$$

$$\hat{s}_\mu \epsilon_{\nu\sigma\tau} \tag{A4}$$

$$\hat{s}_\mu \hat{s}_\nu \delta_{\sigma\tau} \tag{A5}$$

$$\hat{s}_\mu \hat{s}_\nu \hat{s}_\sigma \hat{s}_\tau \tag{A6}$$

and all other inequivalent tensors obtained by permuting the indices of the above. Since all contractions are performed on the symmetric tensors $\mathbf{q}$ and $\mathbb{I}$, the relevant tensors can, without loss of generality, be taken to be symmetric in the indices that are contracted. Particle interchange symmetry implies that:

(i) $\mathbf{W}_1^{(2)} = \mathbf{W}_2^{(2)} = \mathbf{W}^{(2)}$,

(ii) $\mathbf{W}^{(4)}$ must be symmetric under the interchange of the first two indices with the second two,

(iii) all tensors must be invariant under the substitution $\hat{\mathbf{s}} \to -\hat{\mathbf{s}}$.

Together these requirements lead to the following general form

$$W^{(0)}(\mathbf{s}) = w^{(0)}(s) \tag{A7}$$

$$W^{(2)}_{\mu\nu}(\mathbf{s}) = w_0^{(2)}(s)\delta_{\mu\nu} + w_2^{(2)}(s)\hat{s}_\mu\hat{s}_\nu \tag{A8}$$

$$\begin{aligned} W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = {} & w_a^{(4)}(s)\delta_{\mu\nu}\delta_{\sigma\tau} + w_b^{(4)}(s)\left\{\delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right\} + \\ & w_c^{(4)}(s)\left\{\hat{s}_\mu\hat{s}_\nu\delta_{\sigma\tau} + \delta_{\mu\nu}\hat{s}_\sigma\hat{s}_\tau\right\} + \\ & w_d^{(4)}(s)\left\{\hat{s}_\mu\delta_{\nu\sigma}\hat{s}_\tau + \hat{s}_\mu\delta_{\nu\tau}\hat{s}_\sigma + \hat{s}_\nu\delta_{\mu\sigma}\hat{s}_\tau + \hat{s}_\nu\delta_{\mu\tau}\hat{s}_\sigma\right\} \\ & + w_e^{(4)}(s)\hat{s}_\mu\hat{s}_\nu\hat{s}_\sigma\hat{s}_\tau \end{aligned} \tag{A9}$$

Finally, because $\mathbf{q}$ is traceless, all contractions with the unit tensor vanish operating either on the first two, or the second two indices of $W^{(4)}_{\mu\nu\sigma\tau}$ vanish, so that we can set $w_0^{(2)}(s) = w_a^{(4)}(s) = w_c^{(4)}(s) = 0$, leaving the five independent contributions shown in Eqs. (8)-(10):

$$W^{(0)}(\mathbf{s}) = w^{(0)}(s), \tag{A10}$$

$$W^{(2)}_{\mu\nu}(\mathbf{s}) = w^{(2)}(s)\hat{s}_\mu\hat{s}_\nu, \tag{A11}$$

$$\begin{aligned} W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = {} & w_0^{(4)}(s)(\delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}) + \\ & w_2^{(4)}(s)(\hat{s}_\mu\delta_{\nu\sigma}\hat{s}_\tau + \hat{s}_\nu\delta_{\mu\sigma}\hat{s}_\tau + \hat{s}_\mu\delta_{\nu\tau}\hat{s}_\sigma + \hat{s}_\nu\delta_{\mu\tau}\hat{s}_\sigma) + \\ & w_4^{(4)}(s)\hat{s}_\mu\hat{s}_\nu\hat{s}_\sigma\hat{s}_\tau. \end{aligned} \tag{A12}$$

Appendix B: Identity of terms in effective potential for $D = 2$

Consider the $2D$ molecular anisotropy tensor $\mathbf{q}(\hat{\omega}) = 2\hat{\omega} \otimes \hat{\omega} - \mathbb{I}$. Recalling the expansion formula for characteristic polynomials we find

$$\det\left(\mathbf{q}(\hat{\omega})\right) = 1 - 2\,\mathrm{Tr}\left(\hat{\omega} \otimes \hat{\omega}\right) + 4\det\left(\hat{\omega} \otimes \hat{\omega}\right) = -1 \tag{B1}$$

Now consider the contraction with any of the terms labelled by $w_d^{(4)}(s)$, e.g.

$$q_{\mu\nu}\left(\hat{\omega}_1\right)\hat{s}_\mu\delta_{\nu\sigma}\hat{s}_\tau q_{\sigma\tau}\left(\hat{\omega}_2\right)=q_{\mu\nu}\left(\hat{\omega}_1\right)q_{\nu\tau}\left(\hat{\omega}_2\right)\hat{s}_\mu\hat{s}_\tau=\mathbf{q}\left(\hat{\omega}_1\right)\cdot\mathbf{q}\left(\hat{\omega}_2\right):\hat{\mathbf{s}}\otimes\hat{\mathbf{s}} \tag{B2}$$

Now the product $\mathbf{R}\left(\hat{\omega}_1,\hat{\omega}_2\right)=\mathbf{q}\left(\hat{\omega}_1\right)\cdot\mathbf{q}\left(\hat{\omega}_2\right)$, has determinant $\det(\mathbf{q}\left(\hat{\omega}_1\right)\cdot\mathbf{q}\left(\hat{\omega}_2\right))=(-1)^2=1$, so it is a $2D$ rotation. The value of the innerproduct $\left(\mathbf{R}\left(\hat{\omega}_1,\hat{\omega}_2\right)\cdot\hat{\mathbf{s}}\right)\cdot\hat{\mathbf{s}}$, therefore does not depend on the direction $\hat{\mathbf{s}}$, and is easily evaluated to be $R_{11}=R_{22}=\frac{1}{2}\mathbf{q}\left(\hat{\omega}_1\right):\mathbf{q}\left(\hat{\omega}_2\right)$.

Appendix C: Integrals over the orientation of the separation vectors

Here we consider integrals over products of components of the orientation of the separation vector. These integrals are of the form

$$S_{\alpha\beta\cdots\sigma\tau}=\int d\hat{\mathbf{s}}\;\hat{s}_\alpha\hat{s}_\beta\cdots\hat{s}_\sigma\hat{s}_\tau \tag{C1}$$

Each integral is identified by its index set $I=\{\alpha,\beta,\cdots,\sigma,\tau\}$. Because the integrals are rotationally invariant, and totally symmetric in all their indices, their value must be a totally symmetric expansion in products of Kronecker delta's $\delta_{\mu\nu}$ connecting pairs of indices. This already implies that all integrals with an odd number of indices vanish identically. Now consider partitioning the index set I into d disjoint subsets $I_i, i=1,\ldots,d$ whose union is I. Within the subset I_i all indices have the value i. Thus, each partition is characterized by d integers $n_i=|I_i|$, where n_i is even, such that $n_1+n_2+\cdots+n_d=|I|$. Clearly, the value of the integral is independent on the specific distribution of indices over these subsets, but solely by the size of the subsets, hence

$$S_{\alpha\beta\cdots\sigma\tau}=S\left(n_1,n_2,\ldots,n_d\right) \tag{C2}$$

Moreover, since the labelling of the orthogonal axes is arbitrary $S\left(n_1, n_2, \ldots, n_d\right)$ is also totally symmetric in its arguments. This leads to the following representation

$$S = S_0 \tag{C3}$$

$$S_{\mu\nu} = S_2 \delta_{\mu\nu} \tag{C4}$$

$$S_{\mu\nu\sigma\tau} = S_4 \left(\delta_{\mu\nu}\delta_{\sigma\tau} + \delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right) \tag{C5}$$

$$S_{\alpha\beta\mu\nu\sigma\tau} = S_6(\delta_{\alpha\beta}(\delta_{\mu\nu}\delta_{\sigma\tau} + \delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}) + \tag{C6}$$

$$\delta_{\alpha\mu}(\delta_{\beta\nu}\delta_{\sigma\tau} + \delta_{\beta\sigma}\delta_{\nu\tau} + \delta_{\beta\tau}\delta_{\nu\sigma}) + \tag{C7}$$

$$\delta_{\alpha\nu}(\delta_{\beta\mu}\delta_{\sigma\tau} + \delta_{\beta\sigma}\delta_{\mu\tau} + \delta_{\beta\tau}\delta_{\mu\sigma}) + \tag{C8}$$

$$\delta_{\alpha\sigma}(\delta_{\beta\mu}\delta_{\nu\tau} + \delta_{\beta\nu}\delta_{\mu\tau} + \delta_{\beta\tau}\delta_{\mu\nu}) + \tag{C9}$$

$$\delta_{\alpha\tau}(\delta_{\beta\mu}\delta_{\nu\sigma} + \delta_{\beta\nu}\delta_{\mu\sigma} + \delta_{\beta\sigma}\delta_{\mu\nu})). \tag{C10}$$

where the values of the coefficients $S_{|I|}$ depend on dimension.

1. 2D case

In two dimension the unit separation vector has the form $\hat{\mathbf{s}} = (\cos\theta, \sin\theta)$. Hence we need to consider the integrals

$$S\left(n_1, n_2\right) = \int d\theta \, \cos^{n_1}(\theta) \sin^{n_2}(\theta) = 2B\left(\frac{n_1+1}{2}, \frac{n_2+1}{2}\right), \tag{C11}$$

where $B\left(x, y\right)$ is the beta function (see entry 3.191.3 in[41]).

We thus find

$\lvert I\rvert$			$S_{\lvert I\rvert}$
0	$S\left(0,0\right) = 2\pi$		2π
2	$S\left(2,0\right) = \pi$		π
4	$S\left(4,0\right) = \frac{3\pi}{4}$	$S\left(2,2\right) = \frac{\pi}{4}$	$\frac{\pi}{4}$
6	$S\left(6,0\right) = \frac{5\pi}{8}$	$S\left(4,2\right) = \frac{\pi}{8}$	$\frac{\pi}{8}$

(C12)

2. 3D case

In 3 dimension the unit separation vector has the form $\hat{\mathbf{s}} = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta)$. Hence we need to consider the integrals

$$S\left(n_1, n_2, n_3\right) = \int_0^{\pi} d\theta\, \sin^{1+n_1+n_2}\theta \cos^{n_3}\theta \times \int_0^{2\pi} d\varphi \cos^{n_1}\varphi \sin^{n_2}\varphi \tag{C13}$$

$$= 2B\left(\frac{n_1+1}{2}, \frac{n_2+1}{2}\right) B\left(1 + \frac{n_1+n_2}{2}, \frac{n_3+1}{2}\right) \tag{C14}$$

These yield

$\lvert I\rvert$				$S_{\lvert I\rvert}$
0	$S\left(0,0,0\right) = 4\pi$			4π
2	$S\left(2,0,0\right) = \frac{4\pi}{3}$			$\frac{4\pi}{3}$
4	$S\left(4,0,0\right) = \frac{4\pi}{5}$	$S\left(2,2,0\right) = \frac{4\pi}{15}$		$\frac{4\pi}{15}$
6	$S\left(6,0,0\right) = \frac{4\pi}{7}$	$S\left(4,2,0\right) = \frac{4\pi}{35}$	$S\left(2,2,2\right) = \frac{4\pi}{105}$	$\frac{4\pi}{105}$

(C15)

Armed with these results we can now evaluate

$$\begin{aligned}\int d\mathbf{s}\, W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = \overline{w^{(4)}_0} S\left(\delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right) + &\\ \overline{w^{(4)}_2}\left(S_{\mu\tau}\delta_{\nu\sigma} + S_{\nu\tau}\delta_{\mu\sigma} + S_{\mu\sigma}\delta_{\nu\tau} + S_{\nu\sigma}\delta_{\mu\tau}\right) + &\\ \overline{w^{(4)}_4} S_{\mu\nu\sigma\tau} &\end{aligned} \tag{C16}$$

Likewise

$$\begin{aligned}\int d\mathbf{s}\, s_\alpha s_\beta W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s}) = \overline{s^2 w^{(4)}_0} S_{\alpha\beta}\left(\delta_{\mu\sigma}\delta_{\nu\tau} + \delta_{\mu\tau}\delta_{\nu\sigma}\right) + &\\ \overline{s^2 w^{(4)}_2}\left\{\delta_{\nu\sigma}S_{\alpha\beta\mu\tau} + \delta_{\mu\sigma}S_{\alpha\beta\nu\tau} + \delta_{\nu\tau}S_{\alpha\beta\mu\sigma} + \delta_{\mu\tau}S_{\alpha\beta\nu\sigma}\right\} + &\\ \overline{s^2 w^{(4)}_4} S_{\alpha\beta\mu\nu\sigma\tau} &\end{aligned} \tag{C17}$$

where we have defined the integral over functions depending only on the radial coordinate

$$\overline{f} = \int_0^{\infty} ds\, s^{d-1} f\left(s\right) \tag{C18}$$

Appendix D: Expansion of the non-ideal part of the free energy

Here we apply the results of the previous appendix C to obtain the expansion of the non-ideal part of the free energy. Starting point is Eq. (38)

$$\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] = \left\{\int d\mathbf{s}\, W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s})\right\} Q_{\mu\nu}\left(\mathbf{R}\right) Q_{\sigma\tau}\left(\mathbf{R}\right) - \frac{1}{4}\left\{\int d\mathbf{s}\, s_\alpha s_\beta W^{(4)}_{\mu\nu\sigma\tau}(\mathbf{s})\right\} \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \partial_\beta Q_{\sigma\tau}\left(\mathbf{R}\right).$$

Applying Eqs. (C16) and (C17) we find

$$\begin{aligned}
&\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] = \{2S\overline{w_0^{(4)}} + 4S_2\overline{w_2^{(4)}} + 2S_4\overline{w_4^{(4)}}\}Q_{\mu\nu}\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right) \\
&-\frac{1}{4}\left\{\begin{array}{c}
2S_2\overline{s^2w_0^{(4)}}\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \\
+4S_4\overline{s^2w_2^{(4)}}\left(\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) + \partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) + \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right)\right) \\
+2S_6\overline{s^2w_4^{(4)}}\left(\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) + 2\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) + 2\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right)\right)
\end{array}\right\}
\end{aligned} \tag{D1}$$

1. 2D case

In the $2D$ case we have already seen that the number of independent terms in the effective interaction is one less than in the $3d$ case. Specifically, this implies that $\beta\varphi_{int}$ can only depend on a single linear combination of the terms involving $w_0^{(4)}\left(s\right)$ and $w_2^{(4)}\left(s\right)$. The observation that $2S = 4S_2$ indicates that this combination is actually one-on-one. Looking at the second term in Eq. (D1) this suggests the existence of an additional identity linking the contractions over the derivatives of $\mathbf{Q}$. Using just the fact that $\mathbf{Q}$ is a traceless and symmetric 2×2 matrix, we can indeed show that

$$\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) + \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) = \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right). \tag{D2}$$

Using this latter identity yields the final result

$$\begin{aligned}
\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] = &\left\{4\pi\left(\overline{w_0^{(4)}} + \overline{w_2^{(4)}}\right) + \frac{\pi}{2}\overline{w_4^{(4)}}\right\}Q_{\mu\nu}\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right) \\
&- \frac{1}{4}\left\{2\pi\left(\overline{s^2w_0^{(4)}} + \overline{s^2w_2^{(4)}}\right) + \frac{3\pi}{4}\overline{s^2w_4^{(4)}}\right\}\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)
\end{aligned}$$

Note that the $2D$ theory therefore possesses only a single elastic constant. It is useful to define the coupling constant, adding a conventional minus sign,

$$U = -\rho\left\{4\pi\left(\overline{w_0^{(4)}} + \overline{w_2^{(4)}}\right) + \frac{\pi}{2}\overline{w_4^{(4)}}\right\}, \tag{D3}$$

and the elastic constant

$$K = -\frac{1}{4}\rho\left\{2\pi\left(\overline{s^2w_0^{(4)}} + \overline{s^2w_2^{(4)}}\right) + \frac{3\pi}{4}\overline{s^2w_4^{(4)}}\right\} \tag{D4}$$

so that the non ideal term becomes

$$\beta F_{int}[\mathbf{Q}\left(\mathbf{R}\right)] = -\frac{1}{2}\rho U\int d\mathbf{R}\,Q_{\mu\nu}\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right) + \frac{1}{2}\rho K\int d\mathbf{R}\,\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \tag{D5}$$

2. 3D case

In the $3D$ case we directly find

$$\varphi_{int}[\mathbf{Q}\left(\mathbf{R}\right)] = \{8\pi\overline{w_0^{(4)}} + \frac{16\pi}{3}\overline{w_2^{(4)}} + \frac{8\pi}{15}\overline{w_4^{(4)}}\}Q_{\mu\nu}\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right)$$

$$-\frac{1}{4}\left\{\begin{array}{c} \frac{8\pi}{3}\overline{s^2w_0^{(4)}}\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \\ +\frac{16\pi}{15}\overline{s^2w_2^{(4)}}\left(\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) + \partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) + \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right)\right) \\ +\frac{8\pi}{105}\overline{s^2w_4^{(4)}}\left(\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) + 2\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) + 2\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right)\right) \end{array}\right\} \tag{D6}$$

For convenience, we introduce the shorthand

$$\kappa_0 = \frac{2\pi}{3}\overline{s^2w_0^{(4)}}, \tag{D7}$$

$$\kappa_2 = \frac{4\pi}{15}\overline{s^2w_2^{(4)}}, \tag{D8}$$

$$\kappa_4 = \frac{2\pi}{105}\overline{s^2w_4^{(4)}}, \tag{D9}$$

allowing us to define

$$\varphi_{el}[\mathbf{Q}\left(\mathbf{R}\right)] \equiv \left(\kappa_0 + \kappa_2 + \kappa_4\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \tag{D10}$$

$$+ \left(\kappa_2 + 2\kappa_4\right)\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) \tag{D11}$$

$$+ \left(\kappa_2 + 2\kappa_4\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) \tag{D12}$$

We now recall the identity

$$\begin{aligned} \int_\Omega d\mathbf{R}\,\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) &= \int_\Omega d\mathbf{R}\,\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) \\ &+ \int_{\partial\Omega} d\sigma\ \left\{b_\alpha\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) - b_\nu\left(\mathbf{R}\right)Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\alpha\mu}\left(\mathbf{R}\right)\right\} \\ &= \int_\Omega d\mathbf{R}\,\partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) \\ &+ \int_\Omega d\mathbf{R}\partial_\alpha\left\{Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) - Q_{\alpha\mu}\left(\mathbf{R}\right)\partial_\nu Q_{\nu\mu}\left(\mathbf{R}\right)\right\}, \end{aligned} \tag{D13}$$

where $b_\alpha\left(\mathbf{R}\right)$ are the components of the outward normal vector to the bounding surface. Introducing

$$\lambda_1 = \kappa_0 + \kappa_2 + \kappa_4, \tag{D14}$$

$$\lambda_2 = 2\left(\kappa_2 + 2\kappa_4\right), \tag{D15}$$

we have

$$\begin{aligned}\varphi_{el}[\mathbf{Q}\left(\mathbf{R}\right)] &= \lambda_1 \partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\alpha Q_{\mu\nu}\left(\mathbf{R}\right) \\ &+ \lambda_2 \partial_\alpha Q_{\alpha\nu}\left(\mathbf{R}\right)\partial_\beta Q_{\beta\nu}\left(\mathbf{R}\right) \\ &+ \frac{1}{2}\lambda_2 \partial_\alpha\left\{Q_{\mu\nu}\left(\mathbf{R}\right)\partial_\nu Q_{\mu\alpha}\left(\mathbf{R}\right) - Q_{\alpha\mu}\left(\mathbf{R}\right)\partial_\nu Q_{\nu\mu}\left(\mathbf{R}\right)\right\}\end{aligned} \tag{D16}$$

showing that, as expected, the $3D$ case only has two independent *bulk* elastic constants. It is a tedious exercise (see e.g.[42,43]) to compare this result to the classical Oseen-Frank elastic theory, which is achieved by specializing to the case were the system has local uniaxial order throughout and a constant value of the scalar order parameter, i.e. $Q_{\mu\nu}\left(\mathbf{R}\right) \propto \frac{1}{2}\left(3n_\mu\left(\mathbf{R}\right)n_\nu\left(\mathbf{R}\right) - \delta_{\mu\nu}\right)$ where $\mathbf{n}\left(\mathbf{R}\right)$ is the local nematic director. The result is that $K_{11} = K_{33} \propto \lambda_1 + \frac{1}{2}\lambda_2$ and $K_{22} \propto \lambda_1$, where K_{11}, K_{22} and K_{33} are the splay, twist and bend elastic constants respectively. Moreover, additional surface terms are generated which then fix the splay-bend and saddle-splay constants $K_{13} = 0$ and $K_{24} \propto 2\lambda_1 + \frac{1}{2}\lambda_2$ explicitly as functions of λ_1 and λ_2.

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