

Saddle-splay elasticity and interfacial nematostatics

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This Brief Report derives the generalized force balance equations of interfacial statics between nematic liquid crystals (NLC) and isotropic fluids (I), using the classical equations of liquid crystal physics, taking into account an important class of gradient surface elasticity, known as saddle-splay elasticity. The objective is to identify the exact nature of the saddle-splay contributions to the fundamental interfacial force balance equations, known as the Laplace-Young equation and the Marangoni force equation. General expressions for the dynamic generalization of these two equations were given by Shih, Mann, and Brown [Mol. Cryst. Liq. Cryst. **98**, 47 (1983)], but the specific form of the static terms appearing in these two equations were missing in the literature, and are now given in this paper. It is found that the tensorial order and functional form of the contributions of saddle-splay elasticity to the two force balance equations are congruent with those arising from the interfacial tension. Therefore, to generalize the interfacial equations of nematostatics by including saddle-splay energy, the interfacial tension must be renormalized with the saddle-splay energy contribution. In addition, saddle splay gives rise to distortion stresses, the two-dimensional analog to the bulk Ericksen stresses, which contribute to the tangential Marangoni force. Exact expressions for pressure jumps across NLC/I interfaces and for the tangential Marangoni force are derived and analyzed. These generalized results are expected to be useful in the characterization of nematocapillarity phenomena, such as wetting, spreading, and the mechanics of thin nematic films.

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The fundamental equation of interfacial hydrostatics for interfaces between two isotropic fluids in the absence of gravity and external fields is [2]

$$-\mathbf{k} \cdot (\mathbf{T}^+ - \mathbf{T}^-) = \nabla_s \cdot \mathbf{t}, \quad (1)$$

where $\mathbf{T}^{+/-}$ are the bulk stress tensors in the (+/-) phases, \mathbf{k} is the unit normal directed into the + phase, $\nabla_s = \mathbf{I}_s \cdot \nabla$ is the surface gradient operator, $\mathbf{I}_s = \mathbf{I} - \mathbf{k}\mathbf{k}$ is the surface idem factor, \mathbf{I} is the unit tensor, ∇ is the gradient vector, and \mathbf{t} is the surface stress tensor. For isotropic fluids the surface stress tensor is simply [2]

$$\mathbf{t} = \gamma \mathbf{I}_s, \quad (2)$$

and represents normal (tension) stresses within the surface plane. Thus the stress jump at isotropic interfaces is [1]

$$-\mathbf{k} \cdot (\mathbf{T}^+ - \mathbf{T}^-) = 2H\gamma\mathbf{k} + \nabla_s \gamma, \quad (3)$$

where H is the mean surface curvature, defined by $H = -\nabla_s \cdot \mathbf{k}/2$. The projection of Eq. (3) along \mathbf{k} gives the pressure jump across the interface, also known as the Laplace-Young equation. The projection of Eq. (3) along the surface gives the Marangoni force balance arising from the surface gradient term $\nabla_s \gamma$. Equation (3) fails to describe interfaces involving liquid crystals because the surface tension in such cases is anisotropic, and in addition to surface normal stresses liquid crystal interfaces generate bending stresses [3]. Theories [4–11] that take into account interfacial tension anisotropy have been formulated and used to explain or predict experimental phenomena. In these theories [4–11] the interfacial tension is given by

$$\gamma = \gamma_{\text{iso}} + \gamma_{\text{an}}(\mathbf{Q}, \mathbf{k}), \quad (4)$$

where γ_{iso} is the isotropic interfacial tension, γ_{an} is the anisotropic contribution known as the anchoring energy, \mathbf{Q} represents the interfacial order parameter, and \mathbf{k} is the unit normal. Thus the surface free energy density (4) takes into account only homogeneous contributions. To describe more accurately certain classes of capillary phenomena gradient terms have to be included in Eq. (4). By now the nature of gradient contributions to the surface free energy density is well established; see, for example, [4,12–14] and references therein. One well-established gradient contribution is the saddle-splay surface energy density [15,16] given by

$$f_{\text{SG}} = \mathbf{k} \cdot \mathbf{g}, \quad (5a)$$

$$\mathbf{g} = \frac{L_3}{2} (\mathbf{Q} : \nabla \mathbf{Q} - \mathbf{Q} \cdot \nabla \cdot \mathbf{Q}), \quad (5b)$$

where L_3 is an elastic modulus, and \mathbf{g} is the splay-bend surface energy vector. Higher order expressions than Eq. (5b) are given in the literature [13,14,17–20], but for the objectives of this report the simplified version will suffice. Here we wish to explore the contributions of f_{SG} , \mathbf{g} , and $\nabla \cdot \mathbf{g}$ to Eq. (1).

The objectives of this Brief Report are (1) to formulate equations of interfacial nematostatics that take into account saddle-splay surface energy contributions, and (2) to identify the exact nature of these contributions to interfacial pressure jumps and to Marangoni tangential forces. Although the saddle-splay energy contributions have been widely investigated in interfacial torque balance equations, a similar analysis regarding force balance has not been systematically performed to our knowledge. Theories of nematocapillarity have been formulated [3,21–24], but none take into account saddle-splay elasticity in a systematic way.

The system considered here is a static interface between a nematic liquid crystal (NLC) and an isotropic fluid (I). The interface is assumed to be isothermal, and both phases are incompressible. The NLC occupies region R^N , and the isotropic fluid region R^I . The orientation of the interface between the R^N/R^I regions, denoted by NLC/I, is characterized by a unit normal \mathbf{k} , directed from R^N into R^I . The NLC structure is given by the symmetric, traceless, 3×3 tensor order parameter \mathbf{Q} , usually parametrized as follows [25]:

$$\mathbf{Q} = S(\mathbf{nn} - \mathbf{I}/3) + P(\mathbf{mm} - \mathbf{II})/3. \quad (6)$$

The total free energy of the NLC in the absence of external fields is given by [3,4,6,7,15,16,26]

$$F = F_H + F_{el} + F_{an} + F_{iso}, \quad (7)$$

where F_H is the homogeneous, F_{el} the elastic, F_{an} the anchoring, and F_{iso} the isotropic free energy. The homogeneous free energy is responsible for the nematic-isotropic phase transition and is given by

$$F_H = \int f_H(\mathbf{Q}) dV, \quad (8a)$$

$$f_H(\mathbf{Q}) = f_H(0) + a \text{tr} \mathbf{Q}^2 - b \text{tr} \mathbf{Q}^3 + c(\text{tr} \mathbf{Q}^2)^2, \quad (8b)$$

where a, b, c are the Landau coefficients. The elastic free energy F_{el} , also known as the Frank energy, contains long range gradient contributions and is given by

$$F_{el} = \int f_g dV, \quad (9)$$

where the gradient free energy density f_g is [15,16]

$$f_g(\nabla \mathbf{Q}) = \frac{L_1}{2} \text{tr} \nabla \mathbf{Q}^2 + \frac{L_2}{2} (\nabla \cdot \mathbf{Q}) \cdot (\nabla \cdot \mathbf{Q}) + \frac{L_3}{2} (\nabla \mathbf{Q}) : (\nabla \mathbf{Q}), \quad (10)$$

where $\{L_i\}$, $i = 1, 2, 3$, are the Frank elastic constants [15,16]. Using the identity

$$(\nabla \mathbf{Q}) : (\nabla \mathbf{Q}) = (\nabla \cdot \mathbf{Q}) \cdot (\nabla \cdot \mathbf{Q}) - \nabla \cdot [\mathbf{Q} \cdot (\nabla \cdot \mathbf{Q}) - \mathbf{Q} : \nabla \mathbf{Q}] \quad (11)$$

and the divergence theorem, the elastic free energy F_{el} becomes

$$F_{el} = \int f_{BG} dV + \int f_{SG} dS, \quad (12a)$$

$$f_{BG}(\nabla \mathbf{Q}) = \frac{L_1}{2} \text{tr} \nabla \mathbf{Q}^2 + \frac{L_2 + L_3}{2} (\nabla \cdot \mathbf{Q}) \cdot (\nabla \cdot \mathbf{Q}), \quad (12b)$$

where f_{BG} is the bulk gradient elastic free energy density and $f_{SG} = \mathbf{k} \cdot \mathbf{g}$ is the surface gradient free energy density given in Eq. (5). The anchoring energy F_{an} is given by [4]

$$F_{an} = \int \gamma_{an} dS, \quad (13a)$$

$$\gamma_{an} = \beta_{11} \mathbf{k} \cdot \mathbf{N} + \beta_{20} \mathbf{Q} \cdot \mathbf{Q} + \beta_{21} \mathbf{N} \cdot \mathbf{N} + \beta_{22} (\mathbf{k} \cdot \mathbf{N})^2, \quad \mathbf{N} = \mathbf{Q} \cdot \mathbf{k}. \quad (13b)$$

γ_{an} is the anchoring energy density, and $\{\beta_{ij}\}$, $ij = 11, 20, 22, 22$, are the anchoring coefficients (energy per area). Discussion and different uses of Eq. (13b) can be found in the literature [4–11]. The isotropic free energy F_{iso} is the surface integral of the usual isotropic interfacial tension γ_{iso} . The total surface free energy F_s and its density γ are given in terms of the following sum of isotropic, anchoring, and gradient contributions:

$$F_s = \int \gamma dS, \quad (14a)$$

$$\gamma(\mathbf{Q}, \mathbf{k}, \nabla \mathbf{Q}) = \gamma_{iso} + \gamma_{an} + f_{SG} = \gamma_{iso} + \gamma_{an} + \mathbf{k} \cdot \mathbf{g}, \quad (14b)$$

which now depends on $\nabla \mathbf{Q}$ as well as \mathbf{k} and \mathbf{Q} . By decomposing the gradient vector as $\nabla(*) = \mathbf{k} \mathbf{k} \cdot \nabla(*) + \nabla_s(*)$, it is possible to show that only surface gradients enter in Eq. (14), and $\gamma = \gamma(\mathbf{Q}, \mathbf{k}, \nabla_s \mathbf{Q})$.

The specific characteristic nature of the hydrostatics of NLC/I interfaces resides in the constitutive equations for \mathbf{T}^N , \mathbf{T}^I , and \mathbf{t}_s . The total stress tensor in the isotropic phase \mathbf{T}^I is just

$$\mathbf{T}^I = -p^I \mathbf{I}, \quad (15)$$

where p^I is the hydrostatic pressure. The total stress tensor in the NLC phase \mathbf{T}^N is given by

$$\mathbf{T}^N = -p^N \mathbf{I} + \mathbf{T}^E, \quad (16)$$

where p^N is the pressure and \mathbf{T}^E is the Ericksen stress [27] given by

$$\begin{aligned} \mathbf{T}^E &= - \frac{\partial f_{BG}}{\partial \nabla \mathbf{Q}} : (\nabla \mathbf{Q})^T \\ &= -L_1 \nabla \mathbf{Q} : (\nabla \mathbf{Q})^T - (L_2 + L_3) (\nabla \mathbf{Q})^T \cdot (\nabla \mathbf{Q}). \end{aligned} \quad (17)$$

Following the procedures shown in [23,25], the pressure in the NLC phase is given by

$$p^N = -(f_{BG} + f_H) + \Phi, \quad (18)$$

where Φ is the hydrostatic component of the pressure [23], is a function of density and temperature, $\Phi(\rho, T)$, and is space independent, $\nabla \Phi = \mathbf{0}$.

The surface anchoring energy gives rise to an additional contribution to the surface stress tensor \mathbf{t}_s of isotropic materials [3]. For an interface between an isotropic substrate and a NLC, the most general surface elastic stress tensor \mathbf{t} is a 2×3 tensor given by the sum of the normal (tension) \mathbf{t}^n , bending \mathbf{t}^b stresses [3], and shear $\mathbf{t}^{\text{shear}}$ stresses. According to the principle of virtual work, the variation of the surface energy due to a displacement u of the interface is given by

$$\delta F_s = \int \mathbf{t} : (\nabla_s \mathbf{u})^T dS = \int \mathbf{I}_s \cdot \mathbf{t} : (\nabla_s \mathbf{u})^T dS, \quad (19)$$

where the second equality follows from the fact that \mathbf{t} is a 2×3 tensor. To derive the expression of the surface stress tensor in terms of the surface free energy density γ , we compute the variation of F_s :

$$\delta F_s = \int \left[\mathbf{I}_s \gamma : (\nabla_s \mathbf{u})^T + \frac{\partial \gamma}{\partial \mathbf{k}} \cdot \delta \mathbf{k} + \frac{\partial \gamma}{\partial \nabla_s \mathbf{Q}} : \delta (\nabla_s \mathbf{Q})^T \right] dS. \quad (20)$$

The variations $\delta \mathbf{k}$ and $\delta (\nabla_s \mathbf{Q})^T$ in terms of the surface displacement gradient $(\nabla_s \mathbf{u})^T$ are simply

$$\delta \mathbf{k} = -\mathbf{k} \cdot (\nabla_s \mathbf{u})^T, \quad \delta (\nabla_s \mathbf{Q})^T = -(\nabla_s \mathbf{Q})^T \cdot (\nabla_s \mathbf{u})^T, \quad (21)$$

which yield

$$\delta F_s = \int \mathbf{I}_s \cdot \left[\mathbf{I}_s \gamma - \frac{\partial \gamma}{\partial \mathbf{k}} \mathbf{k} - \frac{\partial \gamma}{\partial \nabla_s \mathbf{Q}} : (\nabla_s \mathbf{Q})^T \right] : (\nabla_s \mathbf{u})^T dS. \quad (22)$$

The factor \mathbf{I}_s in front of the brackets appears because only a 2×3 tensor performs work. Thus it follows that the most general surface stress tensor for the NLC/I interface is

$$\mathbf{t} = \mathbf{I}_s \gamma - \mathbf{I}_s \cdot \frac{\partial \gamma}{\partial \mathbf{k}} \mathbf{k} - \mathbf{I}_s \cdot \frac{\partial \gamma}{\partial \nabla_s \mathbf{Q}} : (\nabla_s \mathbf{Q})^T. \quad (23)$$

The surface stress tensor can naturally be decomposed into the following physically significant contributions.

(a) *Normal surface stresses* \mathbf{t}^n ,

$$\mathbf{t}^n(\mathbf{Q}, \mathbf{k}, \nabla_s \mathbf{Q}) = \gamma \mathbf{I}_s. \quad (24)$$

These are the classical 2×2 tension stresses arising in all interfaces. For the NLC/I interface, the tension stresses are a function of \mathbf{Q} , \mathbf{k} , and $\nabla_s \mathbf{Q}$, in addition to the usual temperature dependence. In particular, surface gradients of the tensor order parameter $\nabla_s \mathbf{Q}$ affect \mathbf{t}^n .

(b) *Bending stresses* \mathbf{t}^b ,

$$\mathbf{t}^b(\mathbf{Q}, \mathbf{k}, \nabla \mathbf{Q}) = -\mathbf{I}_s \cdot \left(\frac{\partial \gamma}{\partial \mathbf{k}} \mathbf{k} \right) = -\mathbf{I}_s \cdot \left(\frac{\partial \gamma_{\text{an}}}{\partial \mathbf{k}} \mathbf{k} \right) - \mathbf{I}_s \cdot \mathbf{g} \mathbf{k}. \quad (25)$$

These are nonclassical 2×3 bending stresses. For the NLC/I interface, the bending stresses are a function of \mathbf{Q} , \mathbf{k} , and $\nabla \mathbf{Q}$, in addition to the usual temperature dependence. In particular, gradients of the tensor order parameter $\nabla \mathbf{Q}$ at the interface, including surface gradients $\nabla_s \mathbf{Q}$ and normal gradients $\mathbf{k} \mathbf{k} \cdot \nabla \mathbf{Q}$ at the interface affect \mathbf{t}^b .

(c) *Tension and shear distortion stresses* \mathbf{t}^d ,

$$\begin{aligned} \mathbf{t}^d(\mathbf{Q}, \mathbf{k}, \nabla_s \mathbf{Q}) &= -\mathbf{I}_s \cdot \frac{\partial \gamma}{\partial \nabla_s \mathbf{Q}} : (\nabla_s \mathbf{Q})^T \\ &= -\mathbf{I}_s \cdot \frac{\partial (\mathbf{g} \cdot \mathbf{k})}{\partial \nabla_s \mathbf{Q}} : (\nabla_s \mathbf{Q})^T. \end{aligned} \quad (26)$$

These are nonclassical 2×2 shear and tension stresses. These stresses are the two-dimensional analog of the 3×3 bulk Ericksen stresses. Since \mathbf{t}^d is not traceless, it contains both shear (i.e., components 12 and 21) and tension components (i.e., components 11 and 22). For the NLC/I interface, the distortion stresses are a function of \mathbf{Q} , \mathbf{k} , and $\nabla_s \mathbf{Q}$, in addition to the usual temperature dependence. In particular, surface gradients of the tensor order parameter $\nabla_s \mathbf{Q}$ affect \mathbf{t}^d . To check the validity of the expression for the surface stress tensor \mathbf{t} given in Eq. (23), assume uniaxiality [set $P = 0$ in Eq. (6)], and neglect saddle splay [set $\mathbf{g} = \mathbf{0}$ and $f_{\text{SG}} = 0$ in Eqs. (5)] to obtain

$$\mathbf{t} = \mathbf{I}_s (\gamma_{\text{iso}} + \gamma_{\text{an}}) - \mathbf{I}_s C (\mathbf{n} \cdot \mathbf{k}) \cdot \mathbf{n} \mathbf{k}, \quad (27)$$

in perfect agreement with the surface stress tensor expression previously derived by Ericksen [22], Jenkins and Barrat [24], and Virga [23]. Here C is a constant.

Saddle splay is the source of the following contributions to the normal, bending, and distortion surface stresses:

$$\mathbf{t}_{\text{SS}}^n = +(\mathbf{g} \cdot \mathbf{k}) \mathbf{I}_s = +\mathbf{I}_s \left[\frac{L_3}{2} (\mathbf{Q} : \nabla_s \mathbf{Q} - \mathbf{Q} \nabla_s : \mathbf{Q}) \cdot \mathbf{k} \right], \quad (28a)$$

$$\mathbf{t}_{\text{SS}}^b = -\mathbf{I}_s \cdot \mathbf{g} \mathbf{k} = -\mathbf{I}_s \cdot \left[\frac{L_3}{2} (\mathbf{Q} : \nabla \mathbf{Q} - \mathbf{Q} \nabla : \mathbf{Q}) \mathbf{k} \right], \quad (28b)$$

$$\mathbf{t}_{\text{SS}}^d = -\frac{\partial \gamma}{\partial \nabla_s \mathbf{Q}} : (\nabla_s \mathbf{Q})^T = \mathbf{I}_s \cdot \mathbf{\Pi},$$

$$\mathbf{\Pi} = -\left[\frac{L_3}{2} (\mathbf{Q} \mathbf{k} - \mathbf{I} \mathbf{k} \cdot \mathbf{Q}) \mathbf{k} \right] : (\nabla_s \mathbf{Q})^T. \quad (28c)$$

The interfacial tangential force $\mathbf{f}_{\text{SS}\parallel}$ and normal force $\mathbf{f}_{\text{SS}\perp}$ generated by the saddle-splay stresses are obtained by calculating the surface divergence of \mathbf{t}_{SS} , and the following results are obtained:

$$\mathbf{f}_{\text{SS}\parallel} = [\nabla_s (\mathbf{g} \cdot \mathbf{k})] \cdot \mathbf{I}_s + \mathbf{b} \cdot \mathbf{g} + \nabla_s \cdot \mathbf{\Pi}_s, \quad (29a)$$

$$\mathbf{f}_{\text{SS}\perp} = -(\nabla_s \cdot \mathbf{g}) \mathbf{k}, \quad (29b)$$

where $\mathbf{b} = -\nabla_s \mathbf{k}$ is the symmetric surface curvature dyadic, and $\mathbf{\Pi}_s = \mathbf{I}_s \cdot \mathbf{\Pi}$.

The interfacial tangential saddle-splay force $\mathbf{f}_{\text{SS}\parallel}$ is a function of \mathbf{Q} , \mathbf{k} , and $\nabla \mathbf{Q}$, and has contributions from tension, bending, and distortion stresses; $\mathbf{f}_{\text{SS}\parallel}$ is zero if the gradients of the tensor order parameter at the interface are zero. The interfacial normal saddle-splay force $\mathbf{f}_{\text{SS}\perp}$ is a function of \mathbf{Q} , \mathbf{k} , and $\nabla \mathbf{Q}$, and has contributions from bending; $\mathbf{f}_{\text{SS}\perp}$ is zero if the gradients of the tensor order parameter at the interface are zero.

Next we derive the complete Laplace-Young equation and the Marangoni force balance equation for a NLC/I interface, and characterize the saddle-splay contributions in these two basic equations. The generalized Laplace-Young equation, obtained using Eqs. (15)–(18) and (22b) and taking the projection of Eq. (1) along \mathbf{k} , is given by

$$p^I - \Phi = \left\{ -(f_{BG} + f_H) - \mathbf{k}\mathbf{k} : \mathbf{T}^E \right\} + \left\{ 2H\gamma - 2H \left(\frac{\partial \gamma}{\partial \mathbf{k}} \cdot \mathbf{k} \right) + \nabla_s \cdot \left(\frac{\partial \gamma}{\partial \mathbf{k}} \right) \right\}, \quad (30)$$

where the first curly bracket represents the bulk contributions at the interface, and the second the surface contributions. We note that gradients of the tensor order parameter contribute to the pressure jumps in the bulk terms as well as in the surface terms. The contributions from saddle splay are perfectly incorporated in the surface energy density. The net saddle-splay contribution to the Laplace-Young equation then is

$$p^I - \Phi = p^I - \Phi|_{\text{saddle splay}=0} - \nabla_s \cdot \mathbf{g} = p^I - \Phi|_{\text{saddle splay}=0} - \nabla_s \cdot \mathbf{g}_s + 2H(\mathbf{g} \cdot \mathbf{k}), \quad (31)$$

where $\mathbf{g}_s = \mathbf{I}_s \cdot \mathbf{g}$. In the absence of curvature ($H=0$, $\mathbf{b}=0$) the total pressure jump $p^I - \Phi$ across a NLC/I interface is

$$p^I - \Phi = \left\{ -(f_{BG} + f_H) - \mathbf{k}\mathbf{k} : \mathbf{T}^E \right\} + \nabla_s \cdot \left(\frac{\partial \gamma}{\partial \mathbf{k}} \right), \quad (32)$$

showing that all the nematic energy densities f_{BG} , f_H , \mathbf{g} , and γ_{AN} may contribute to pressure discontinuities across a flat NLC/I interface. In other words, interfacial pressure jumps are functions of \mathbf{Q} , \mathbf{k} , and $\nabla \mathbf{Q}$.

The tangential projection of Eq. (1) is a balance between the bulk stress jump and the Marangoni force:

$$-\mathbf{k} \cdot (\mathbf{T}^I - \mathbf{T}^N) \cdot \mathbf{I}_s = \left[\nabla_s(\gamma) + \mathbf{b} \cdot \frac{\partial \gamma}{\partial \mathbf{k}} + (\nabla_s \cdot \mathbf{I}_s) \right] \cdot \mathbf{I}_s. \quad (33)$$

Thus, in contrast to isotropic interfaces, NLC/I interfaces display the additional curvature dependent Marangoni force ($\mathbf{b} \cdot d\gamma/d\mathbf{k}$), as well as shear forces ($\nabla_s \cdot \mathbf{I}_s$). Saddle splay is therefore absorbed into the surface energy contributions to the Marangoni force, with congruent tensorial and functional form. Finally, we conclude that the saddle-splay contributions to tangential forces are

$$-\mathbf{k} \cdot (\mathbf{T}^I - \mathbf{T}^N) \cdot \mathbf{I}_s = -\mathbf{k} \cdot (\mathbf{T}^I - \mathbf{T}^N) \cdot \mathbf{I}_s|_{\text{saddle splay}=0} + \nabla_s(\mathbf{g} \cdot \mathbf{k}) + \mathbf{b} \cdot \mathbf{g} + \nabla_s \cdot \mathbf{I}_s, \quad (34)$$

which persists even for planar interfaces.

In summary, the analysis identifies the exact nature of the contributions of saddle-splay energy to the interfacial stress balance equations between nematic liquid crystals and isotropic fluids. The tensorial order and functional form of the saddle-splay contributions to the Laplace-Young equation and to the Marangoni force are congruent with the bulk gradient elasticity and anchoring energy terms. These findings will be needed to understand and evaluate liquid crystal interfacial phenomena.

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