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The Neumann and Young equations for nematic contact lines

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The Neumann and Young equations for three-phase nematic contact lines have been derived using the momentum balance equation and classical liquid crystal physics theories. The novel finding is the presence of bending forces, originating from the anchoring energy of nematic interfaces, and acting on the contact line. The classical Neumann triangle or tensile force balance becomes in the presence of a nematic phase the Neumann pentagon, involving the usual three tensile forces and two additional bending forces. The Young equation that describes the static contact angle of a fluid in contact with a rigid solid is again a tensile force balance along the solid, but for nematics it also involves an additional bending force. The effects of the bending forces on contact angles and wetting properties of nematic liquid crystals are thoroughly characterized. It is found that in terms of the spreading coefficient, bending forces enlarge the partial wetting window that exists between dewetting and spontaneous spreading. Bending forces also affect the behaviour of the contact angle, such that spreading occurs at contact angles greater than zero and dewetting at values greater than π. Finally, the contact angle range in the partial wetting regime is always less than π.

1. Introduction

Wetting and spreading phenomena arise during manufacturing and characterization of many material systems. Manufacturing examples include film coatings, paints, fibre finishings, and multiphase materials [1]. Material characterization includes contact angle measurements, and the Whilhelmy balance [2]. The surface properties of liquid crystals are also important for many display applications [3], and in material systems such as mesophase carbon–carbon fibre composites where the adhesion between the two phases strongly affects the mechanical properties. Despite the importance of surface phenomena in liquid crystals [3, 4] their theoretical description is often based on models for isotropic fluids, neglecting certain aspects of the anisotropic nature of liquid crystals. Thus there is a strong need to develop mechanical models that describe anisotropic surface thermodynamics and transport phenomena. The present paper is concerned with deriving macroscopic equations that model the wetting properties of nematic liquid crystals (NLCs). A previous paper [5] considered the interfacial transport phenomena in nematic interfaces.

A frequent phenomenon that arises in surface-dominated problems is the presence of a contact line at which three different phases intersect. The three-phase contact line arises in many practical problems, including flotation, fluid displacement, and wetting of solid surfaces [1, 2, 6, 7]. The theoretical description of the momentum balance equation at the contact line is known as the Neumann equation or Neumann triangle [1, 7], which in the absence of inertia and mass transfer gives the balance of tensile forces that exist at the three intersecting phases. The surface stress tensors involved in the classical Neumann equation contain only normal components, that is, tensile stresses. Since the surface stress tensor of NLC contains bending stresses arising from the anisotropic surface anchoring energy [5], the classical Neumann equation is incomplete for NLC. When one of the phases is a solid, the projection of the tensile forces along the three intersecting phases onto the solid phase gives the well known Young equation [8]. Given its origin, the Young equation is also incomplete when NLCs are involved. Wetting properties of fluids modelled by the Young equation are based on interfacial tensions, but as shown below, for NLCs they will also involve the anisotropic surface anchoring energies.

The effect of bending stresses may appear to be modest in most circumstances and material systems, since for low molar mass NLCs the isotropic surface tension is usually three orders of magnitude greater than the anchoring energy. Important exceptions are cases involving interfaces between a NLC and its isotropic phase, since interfacial tensions are comparable to anchoring energies [9, 10]. Thus when describing the nematic–isotropic phase transition next to a solid surface the bending stresses may be as important as the tensile stresses. Other nematics, such as nematic polymers and carbonaceous mesophases, may reveal anchoring energies comparable

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to interfacial tensions. Thus there is a need to know the nature and importance of the terms that are neglected when using the classical Neumann and Young equations for isotropic fluids.

The objectives of this paper are: (1) to derive the Neumann equation at a three-phase contact line, when one of the phases is a NLC; (2) to derive the Young equation for NLCs; (3) to characterize the wetting regimes for nematic liquid crystals. Dynamical wetting phenomena [8], curvature line tension effects [8], and scalar order parameter effects [11] are beyond the scope of this paper.

The organization of this paper is as follows. Section 2 presents a derivation of the surface stress tensor for a nematic liquid crystal. Section 3 contains the derivation of the Neumann equation for a three phase contact line involving a NLC. Section 4 presents a derivation of the Young equation for NLCs. Section 5 discusses the wetting regimes of NLCs, and gives thresholds for the onset of dewetting, partial wetting, and spreading. Section 6 gives the conclusions.

2. The nematic surface stress tensor

In this section we derive the equation for the nematic surface stress tensor for an interface involving a uniaxial rod-like nematic liquid crystal of constant order parameter [11]. The system is isothermal, and both phases are incompressible. The interface is assumed to be elastic. The orientation in the NLC is given by the unit vector **n**, or director.

The static limit of the interfacial linear momentum balance equation for an interface between a uniaxial rod-like nematic liquid crystal and an isotropic fluid was first presented by Jenkins and Barratt [12, equation (3.40)]. The same equation was later derived by Ericksen [13, equation (72)] in his review paper on the equilibrium theory of liquid crystals, where it is indicated that the equation expresses balancing stress with surface tension. Later Virga [14, equation (2.29)] investigating the shape of droplets in contact with isotropic fluids derived the same equation that describes the static limit of the interfacial linear momentum balance equation for an interface between a uniaxial rod-like nematic liquid crystal and an isotropic fluid. Recently Rey [5] identified the origin, nature, and physical significance of the surface stress tensor that appears in the static limit of the interfacial linear momentum balance equation, for an interface between a uniaxial rod-like nematic liquid crystal and an isotropic fluid. In particular, it was shown that for such interfaces the surface stress tensor is not in general a 2 × 2 tangential symmetric tensor as for isotropic interfaces, and a clear distinction of the nature and origin of tension stresses and bending stresses was established. The results obtained in [5] are necessary in order to solve and understand practical problems in interfacial phenomena involving nematic liquid crystals. One such practical problem, the wetting properties of nematic liquid crystals, is the topic of this paper.

Assume that a nematic liquid crystal is in region R_N, and that another material α is in region R_α. The interface between the two regions is characterized by a unit normal $\mathbf{k}^{N\alpha}$, directed from the R_N phase towards the R_α phase. The surface stress tensor $\mathbf{t}^{N\alpha}$ is given by the sum of the normal (tension) $\mathbf{t}_N^{N\alpha}$ and the bending $\mathbf{t}_B^{N\alpha}$ contributions [5]:

$$\mathbf{t}_N^{N\alpha} = F_s^{N\alpha}\mathbf{I}_s^{N\alpha}; \quad \mathbf{t}_B^{N\alpha} = -\frac{dF_s^{N\alpha}}{d(\mathbf{n}\cdot\mathbf{k}^{N\alpha})}\mathbf{I}_s\cdot\mathbf{n}\mathbf{k}^{N\alpha} \qquad (1\,a,b)$$

where $\mathbf{I}_s$ is the surface idem factor for the N–α interface, and $F_s^{N\alpha}$ is the surface free energy density, here taken to be a function of $\mathbf{n}\cdot\mathbf{k}^{N\alpha}$. An example of a simple and widely used constitutive equation for $F_s^{N\alpha}$ is the Rapini–Papoular expression [15, 16]:

$$F_s^{N\alpha} = \gamma_{is}^{N\alpha} + \gamma_{an}^{N\alpha}(\mathbf{n}\cdot\mathbf{k}^{N\alpha})^2 \qquad (2)$$

where $\gamma_{is}^{N\alpha}$ is the isotropic interfacial tension and $\gamma_{an}^{N\alpha}$ is the anchoring energy [3, 4, 16]. The sign of $\gamma_{an}^{N\alpha}$ is positive (negative) when the orientation of the interface's easy axis is planar (homeotropic). For low molar mass NLCs the isotropic surface interfacial tension $\gamma_{is}^{N\alpha}$ is the order 10 erg cm^{-2}, while the anchoring energy $\gamma_{an}^{N\alpha}$ varies in the range 10^{-4}–1 erg cm^{-2} [3]. The nematic–isotropic interface of low molar mass materials has a $\gamma_{is}^{N\alpha}$ of the order 10^{-2} erg cm^{-2} which appears to be of the same order of magnitude as the anchoring energy of the same interface [3, 9, 10]. It is also interesting that the anchoring energy in the isotropic phase of 5CB is zero only when the temperature is several degrees above the nematic–isotropic transition temperature [3].

Parametrizing the interface with orthonormal unit surface base vectors $(\mathbf{i}_1, \mathbf{i}_2)$ the normal and bending stresses become:

$$\mathbf{t}_N^{N\alpha} = F_s^{N\alpha}\mathbf{I}_s^{N\alpha}; \quad \mathbf{t}_B^{N\alpha} = -\frac{dF_s^{N\alpha}}{d(\mathbf{n}\cdot\mathbf{k}^{N\alpha})}\mathbf{I}_s\cdot\mathbf{n}\mathbf{k}^{N\alpha}$$

$$\mathbf{t}_N^{N\alpha} = [\gamma_{is}^{N\alpha} + \gamma_{an}^{N\alpha}(\mathbf{n}\cdot\mathbf{k}^{N\alpha})^2]\,[\mathbf{i}_1\mathbf{i}_1 + \mathbf{i}_2\mathbf{i}_2] \qquad (3\,a)$$

$$\mathbf{t}_B^{N\alpha} = B_{13}^{N\alpha}\mathbf{i}_1\mathbf{k}^{N\alpha} + B_{23}^{N\alpha}\mathbf{i}_2\mathbf{k}^{N\alpha} \qquad (3\,b)$$

where the bending coefficients, $B_{13}^{N\alpha}$, $B_{23}^{N\alpha}$, are given by:

$$B_{13}^{N\alpha} = -2\gamma_{an}^{N\alpha}(\mathbf{n}\cdot\mathbf{k}^{N\alpha})(\mathbf{n}\cdot\mathbf{i}_1);$$

$$B_{23}^{N\alpha} = -2\gamma_{an}^{N\alpha}(\mathbf{n}\cdot\mathbf{k}^{N\alpha})(\mathbf{n}\cdot\mathbf{i}_2). \qquad (4\,a,b)$$

The bending coefficients are proportional to the anchoring energy and to the director's projections along the unit normal and along the surface base vectors. In the Rapini–Papoular model the largest magnitudes of the bending coefficients arise at $\pi/4$ angles from the interface, and they vanish at the planar and homeotropic orientations. In matrix form, the 2×3 surface stress tensor $\mathbf{t}^{N\alpha}$ is:

$$\mathbf{t}^{N\alpha} = \begin{bmatrix} \gamma^{N\alpha} & 0 & B_{13}^{N\alpha} \\ 0 & \gamma^{N\alpha} & B_{23}^{N\alpha} \end{bmatrix}. \tag{5}$$

Clearly the surface stress tensor is asymmetric. In the presence of interfacial flows shear stresses arise, but this paper is restricted to statics. Bending stresses are intrinsic to nematic interfaces and result in forces normal to the interface even in the absence of curvature.

3. The Neumann equation for three-phase nematic contact lines

The macroscopic equilibrium contact angles at which three fluid phases meet is determined by the balance of forces acting on the contact line. The Neumann equation for three-phase contact lines is the expression of the force balance equation at the contact line, neglecting mass transfer and inertia of the contact line [7, 17]. Figure 1 shows a typical example of a three phase (A–B–C) contact line (L). The contact line is the common line formed by the intersection of the A–B, B–C, and C–A dividing surfaces. The unit vectors $(\mathbf{v}^{AB}, \mathbf{v}^{BC}, \mathbf{v}^{CA})$ are normal to the contact line and are tangent to the A–B, B–C, and C–A dividing surfaces, and pointing into them. The unit vectors $(\mathbf{k}^{AB}, \mathbf{k}^{BC}, \mathbf{k}^{CA})$ are orthogonal to the A–B, B–C, and C–A dividing surfaces and oriented such that when turning a right handed screw in the sense defined by them it advances in the positive direction of the unit tangent vector $\boldsymbol{\tau}$ to the contact line. With this choices the tangent vectors are given by: $\mathbf{v}^{ij} = -\boldsymbol{\tau} \times \mathbf{k}^{ij}$; ij = AB, BC, CA. The contact angle measured through phase C is defined by: $\cos\theta = \mathbf{v}^{BC} \cdot \mathbf{v}^{CA}$.

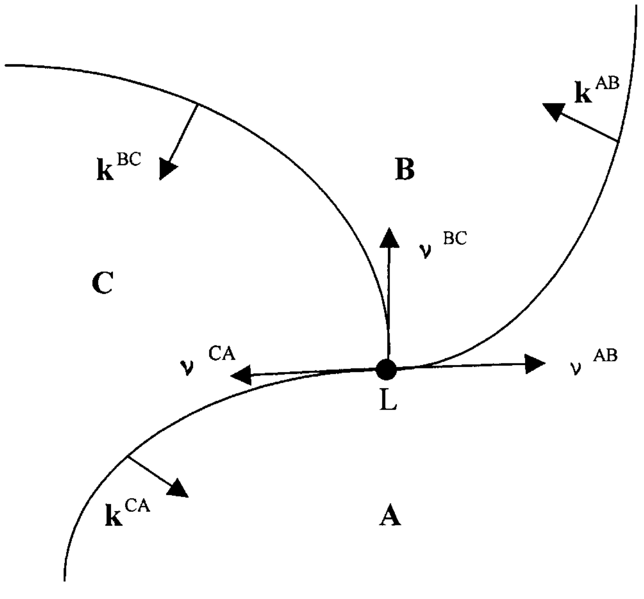

Figure 1. Schematic representing a three-phase (A–B–C) contact line (L) geometry. The contact line is the common line formed by the intersection of the A–B, B–C, and C–A dividing surfaces. The unit vectors $(\mathbf{v}^{AB}, \mathbf{v}^{BC}, \mathbf{v}^{CA})$ are normal to the contact line and are tangent to the A–B, B–C, and C–A dividing surfaces, and pointing into them. The unit vectors $(\mathbf{k}^{AB}, \mathbf{k}^{BC}, \mathbf{k}^{CA})$ are orthogonal to the A–B, B–C, and C–A dividing surfaces, and to the contact line.

The momentum balance equation at the contact line is given by [1, 7]:

$$\sum_{i=AB,BC,CA} \mathbf{v}^i \cdot \mathbf{t}^i = 0 \tag{6}$$

A derivation of equation (6) is given in the Appendix. The classical Neumann equation takes into account only isotropic normal surface stresses (i.e., $\mathbf{t}^i = \gamma^i \mathbf{I}_s^i$; i = AB, BC, CA) and its expression is:

$$\gamma^{AB}\mathbf{v}^{AB} + \gamma^{BC}\mathbf{v}^{BC} + \gamma^{CA}\mathbf{v}^{CA} = 0 \tag{7}$$

where $(\gamma^{AB}, \gamma^{BC}, \gamma^{CA})$ are the interfacial tensions for the three interfaces. This equation is invalid when one of the phases is a nematic liquid crystal. The correct equation is derived below.

Let C now denote a uniaxial NLC. In this case the director orientation at the contact line is defined and given by $\mathbf{n}$. The appropriate surface stresses are given by equations (3, 4) with α = B, C. Replacing these expressions into the momentum balance equation we get:

$$\gamma^{AB}\mathbf{v}^{AB} + \gamma^{BN}\mathbf{v}^{BN} + \gamma^{NA}\mathbf{v}^{NA} + B^{NA}\mathbf{k}^{NA} + B^{BN}\mathbf{k}^{BN} = 0 \tag{8}$$

where the interfacial coefficients involving the nematic phase are given by:

$$\gamma^{BN} = \gamma_{is}^{BN} + \gamma_{an}^{BN}(\mathbf{n} \cdot \mathbf{k}^{BN})^2;$$

$$\gamma^{NA} = \gamma_{is}^{NA} + \gamma_{an}^{NA}(\mathbf{n} \cdot \mathbf{k}^{NA})^2 \tag{9a,b}$$

$$B^{NA} = -2\gamma_{an}^{NA}(\mathbf{n} \cdot \mathbf{k}^{NA})(\mathbf{n} \cdot \mathbf{v}^{NA});$$

$$B^{BN} = -2\gamma_{an}^{NB}(\mathbf{n} \cdot \mathbf{k}^{NB})(\mathbf{n} \cdot \mathbf{v}^{NB}). \tag{9c,d}$$

The Neumann equation for three-phase contact lines involving a NLC contains new contributions in the tension forces and additional terms accounting for bending forces. At the contact line the tension forces are orthogonal to their respective bending forces. The total number of forces has increased from three to five, and the Neumann triangle for isotropic fluids has become for NLCs the Neumann pentagon.

4. The Young equation for nematic contact angles

When phase A is a rigid solid, the projection of the Neumann equation onto the tangent to the solid surface is known as the Young equation [2, 8]. Figure 2 defines the geometry, where θ is the contact angle through the

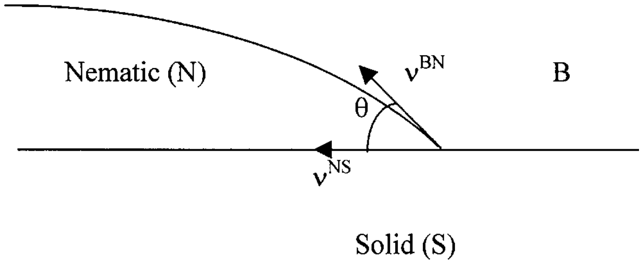

Figure 2. Schematic of a three-phase contact line, involving a nematic liquid crystal (N), a rigid flat solid (S), and an isotropic fluid B. θ is the equilibrium contact angle through the nematic liquid crystalline phase.

A phase. In the absence of anchoring energies, projecting the classical Neumann equation (7) in the $\mathbf{v}^{SB}$ direction gives the famous Young equation [8]:

$$\gamma^{SB} - (\gamma^{BA}\cos\theta + \gamma^{AS}) = 0. \tag{10}$$

If phase A is a NLC, Young's equation obtained by projecting equation (8) in the $\mathbf{v}^{SB}$ direction becomes:

$$\gamma^{SB} - (\gamma^{BN}\cos\theta + \gamma^{NS}) - B^{BN}\sin\theta = 0. \tag{11}$$

The Young equation for NLC contains additional contributions in the tension terms and a new term originating from bending forces. When the anchoring energies are zero we recover the classical Young equation.

It is useful to discuss contact angles and wetting properties using the spreading coefficient S [2, 7, 8]. For two isotropic fluids (A, B) in contact with a solid the spreading coefficient S is:

$$S = \gamma^{SB} - \gamma^{BA} - \gamma^{AS}. \tag{12}$$

If $S > 0$ fluid A will spread spontaneously over the solid displacing fluid B; if $S < -2\gamma^{BA}$ a thin film of A will spontaneously dewet the solid by displacement A by fluid B; and if $-2\gamma^{BA} < S < 0$ fluid A partially wets the solid. The contact line at which phases A and B meet will move spontaneously over the solid if $S > 0$ or if $S < -2\gamma^{BA}$. The common line is stationary if $-2\gamma^{BA} < S < 0$. As shown below for nematic liquid crystals bending stresses modify these results.

The spreading coefficient for a nematic liquid crystal (N) and an isotropic fluid (B) in contact with a solid surface is:

$$S = \gamma^{SB} - \gamma^{BN} - \gamma^{NS}. \tag{13}$$

As explained above, the spreading coefficient captures the ability of N to spread over the solid, displacing fluid B. A discussion of the spreading coefficient for liquid crystals can be found in the textbook of Sonin [3, see page 64]. In terms of the spreading coefficient, Young's equation becomes:

$$S = \gamma^{BN}(\cos\theta - 1) + B^{BN}\sin\theta \tag{14a}$$

whose solution in terms of the contact angle θ is:

$$\tan\left(\frac{\theta}{2}\right) = \frac{2B^{BN} + [(2B^{BN})^2 - 4S(S + 2\gamma^{BN})]^{1/2}}{2(S + 2\gamma^{BN})}. \tag{14b}$$

When $B^{BN} = 0$, equation (14b) properly reduces to the classical result: $\cos\theta = S/\gamma^{BN} + 1$. For NLC the contact angle is a function of the bending coefficient B^{BN} in addition to S and γ^{BN}. Since the bending coefficient depends on the director orientation at the surface it follows that the equilibrium contact angle for NLC is not given in terms of thermodynamic parameters as in the case of isotropic fluids. By setting $B^{BN} = 0$ in equation (14), it is seen that a contact angle exists when $-2\gamma^{BN} < S < 0$, but for finite bending coefficients the range of existence of the contact angle is now a function of B^{BN}.

5. Wetting transitions

For two isotropic fluids (A, B) Young's equation (10) in conjunction with equation (12) predicts three wetting regimes, according to the magnitude of the spreading coefficient S:

$$S = \gamma^{AB}(\cos\theta - 1). \tag{15}$$

When discussing wetting regimes by considering the contact angle, the wetting regimes for isotropic fluids are found to be [18]:

(a) If $S > 0$, there is no contact angle θ that satisfies equation (15) and, as stated above, fluid A spreads over the solid displacing fluid B.
(b) If $-2\gamma^{AB} < S < 0$, there is a finite contact angle θ that satisfies equation (15), the contact line is stationary and, as stated above, A partially wets the solid.
(c) If $S < -2\gamma^{AB}$, there is no value of θ that satisfies equation (15) and, as stated above, fluid A will dewet the solid, being displaced by fluid B.

The presence of bending forces in nematics modifies all these results. To analyse the parametric dependence of the spreading, partial wetting, and dewetting regimes we consider the discriminant in equation (14):

$$D(S) = (2B)^2 - 4S(S + 2\gamma^{BN}). \tag{16}$$

The transitions between the three regimes occur at two thresholds values of S (S_s, S_d), for which $D = 0$. The predicted regimes and threshold values of S are as follows:

(1) Spontaneous spreading of N over the solid displacing fluid B occurs if $S > S_s > 0$:

$$S_s = -\gamma^{BN} + [(\gamma^{BN})^2 + (B^{BN})^2]^{1/2} > 0. \tag{17}$$

(2) Dewetting of N from the solid by displacement by fluid B occurs if $S < S_{\mathrm{d}} < -2\gamma^{\mathrm{BN}}$:

$$S_{\mathrm{d}} = -\gamma^{\mathrm{BN}} - [(\gamma^{\mathrm{BN}})^2 + (B^{\mathrm{BN}})^2]^{1/2} < -2\gamma^{\mathrm{BN}}. \tag{18}$$

(3) Partial wetting occurs if $S_{\mathrm{d}} < S < S_{\mathrm{s}}$.

In terms of the spreading coefficient, the effect of the bending forces is to enlarge the partial wetting window, such that S has to be positive for spontaneous spreading and less than $-2\gamma^{\mathrm{BN}}$ for dewetting. In addition, the contact angles at the transitions are also affected by the bending stresses. For isotropic fluids the contact angle predicted at the partial wetting–spreading transition is zero [2, 8] but for NLC is:

$$\cos\theta_{\mathrm{s}} = \frac{\{\gamma^{\mathrm{BN}} + [(\gamma^{\mathrm{BN}})^2 + (B^{\mathrm{BN}})^2]^{1/2}\}^2 - (B^{\mathrm{BN}})^2}{\{\gamma^{\mathrm{BN}} + [(\gamma^{\mathrm{BN}})^2 + (B^{\mathrm{BN}})^2]^{1/2}\}^2 + (B^{\mathrm{BN}})^2} \tag{19}$$

which shows that at the threshold the contact angle θ_{s} is positive definite: $\theta_{\mathrm{s}} > 0$. At the partial wetting–dewetting transition the contact angle θ_{d} for isotropic fluids is π [2, 8]. For NLC the contact angle θ_{d} at this transition is:

$$\cos\theta_{\mathrm{d}} = \frac{\{\gamma^{\mathrm{BN}} - [(\gamma^{\mathrm{BN}})^2 + (B^{\mathrm{BN}})^2]^{1/2}\}^2 - (B^{\mathrm{BN}})^2}{\{\gamma^{\mathrm{BN}} - [(\gamma^{\mathrm{BN}})^2 + (B^{\mathrm{BN}})^2]^{1/2}\}^2 + (B^{\mathrm{BN}})^2} \tag{20}$$

which for a finite bending coefficient shows that θ_{d} is always greater less than π.

Bending forces affect the parametric envelopes of the spreading–partial wetting–dewetting regimes, and the transition threshold contact angles and spreading coefficient values. In general terms bending forces increase the partial wetting spreading parameter window and reduce the range of possible contact angles. How noticeable these effects are depends on the relative magnitudes between the isotropic interfacial tension and the anchoring strength.

6. Conclusions

The Neumann and Young equilibrium equations for three-phase contact lines have been developed using the momentum balance equation and classical liquid crystal physics, when one of the three intersecting phases is a nematic liquid crystal. The force balance at the three-phase contact line involves three tensile and two bending forces, and the well known Neumann triangle for isotropic fluids becomes, in the presence of one nematic phase, the Neumann pentagon. The presence of bending forces modifies the three contact angles at which the three phases intersect. The Young equation for contact angles on solid substrates also includes a bending force term that influences the wetting behaviour of such nematic liquid crystals surfaces. In terms of the spreading parameter, bending forces increase the window of partial wettability by delaying the onset of spontaneous spreading and dewetting. For spontaneous spreading the spreading coefficient of nematics has to be greater than a positive value, instead of zero for isotropic fluids. For dewetting, the spreading coefficient of a nematic is always smaller than for the isotropic fluid. The contact angles at these transitions are also modified by the bending forces. At the spreading-partial wetting transition the contact angle for the nematic case is positive. The contact angle at the dewetting–partial wetting transition is always less than π radians. Thus the total range of possible contact angles in the partial wetting regimes is less than π radians.

The ability of bending forces to modify the wetting and spreading abilities of nematics depends on the relative magnitude of the anchoring energy with respect to the isotropic interfacial tension. For low molar mass nematics typical anchoring energies on many interfaces are significantly smaller than isotropic interfacial tensions, and bending effects may be ignored. Modelling and interpreting experimental data for other materials and experimental conditions in which the isotropic and anisotropic contributions to surface tension are comparable, require the use of the governing equations presented here.

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Appendix

The purpose of this Appendix is to derive equation (6). Here we follow the derivation of Slattery [7] given in section 2.1.9, equation 9-8. Equation (6) is the static limit of the momentum balance equation at the common line, also known as Euler's first law at the common line [7]. In the static limit, and in the absence of external body forces, Euler's first law for a body B that occupies region R is:

$$\int_{\mathrm{S}} \mathbf{k} \;\; \mathbf{T}\,\mathrm{d}A + \int_{\mathrm{C}} \boldsymbol{\mu} \;\; \mathbf{t}\,\mathrm{d}s = 0 \qquad \text{(A.1)}$$

where S is a closed surface that bounds the region R, C is the closed curve that bounds any dividing surface Σ within R, $\mathbf{k}$ is the outward unit normal to S, $\boldsymbol{\mu}$ is the outward unit normal to C and tangent to S, $\mathbf{T}$ is the stress tensor and $\mathbf{t}$ is the surface stress tensor. Using Green's theorem the first integral in equation (A.1) can be written as:

$$\int_{\mathrm{S}} \mathbf{k} \;\; \mathbf{T}\,\mathrm{d}A = \int_{\mathrm{R}} \operatorname{div} \mathbf{T}\,\mathrm{d}V + \int_{\Sigma} [\xi \;\; \mathbf{T}]\,\mathrm{d}A \qquad \text{(A.2)}$$

where $[\xi \;\; \mathbf{T}] = \xi^{(i,j)}\,(\mathbf{T}^i - \mathbf{T}^j)$ and $\xi^{(i,j)}$ is the unit normal directed from phase '*j*' into phase '*i*'. Using the surface divergence theorem the second integral in equation (A.1) becomes:

$$\int_{\mathrm{C}} \boldsymbol{\mu} \;\; \mathbf{T}\,\mathrm{d}A = \int_{\Sigma} \operatorname{div}_{\mathrm{s}} \mathbf{t}\,\mathrm{d}A + \int_{\mathrm{C(cl)}} (\mathbf{v} \;\; \mathbf{t})\,\mathrm{d}s \qquad \text{(A.3)}$$

where $\mathrm{C}^{(\mathrm{cl})}$ denotes the intersection of dividing surfaces at the common or contact line. For three (i, j, k) phases we have

$$(\mathbf{v} \;\; \mathbf{t}) = \mathbf{v}^i \;\; \mathbf{t}^i + \mathbf{v}^j \;\; \mathbf{t}^j + \mathbf{v}^k \;\; \mathbf{t}^k = \sum_{\alpha = i,j,k} \mathbf{v}^{\alpha} \;\; \mathbf{t}^{\alpha} \qquad \text{(A.4)}$$

where $\mathbf{v}^i$ is the unit normal to $\mathrm{C}^{(\mathrm{cl})}$ tangent to and pointing into Σ^i. Substituting equations (A.2) and (A.3) into (A.1) it is found:

$$\int_{\mathrm{R}} \operatorname{div} \mathbf{T}\,\mathrm{d}V + \int_{\Sigma} [\xi \;\; \mathbf{T}]\,\mathrm{d}A + \int_{\Sigma} \operatorname{div}_{\mathrm{s}} \mathbf{t}\,\mathrm{d}A + \int_{\mathrm{C}} (\mathbf{v} \;\; \mathbf{t})\,\mathrm{d}s = 0. \qquad \text{(A.5)}$$

Thus the balance of forces at a three-phase contact line is given by:

$$(\mathbf{v} \;\; \mathbf{t}) = \mathbf{v}^i \;\; \mathbf{t}^i + \mathbf{v}^j \;\; \mathbf{t}^j + \mathbf{v}^k \;\; \mathbf{t}^k = \sum_{\alpha = i,j,k} \mathbf{v}^{\alpha} \;\; \mathbf{t}^{\alpha} = 0 \qquad \text{(A.6)}$$

which is equation (6).