
**Liquid Crystals**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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Online publication date: 06 August 2010

**To cite this Article** Rey, Alejandro D.(1999) 'Nemato-capillarity theory and the orientation-induced Marangoni flow', Liquid Crystals, 26: 6, 913 — 917
**To link to this Article: DOI:** 10.1080/026782999204606
**URL:** http://dx.doi.org/10.1080/026782999204606

# Nemato-capillarity theory and the orientation-induced Marangoni flow

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(*Received 8 January 1999; accepted 13 January 1999*)

The macroscopic equations of nemato-capillarity, including the interfacial linear momentum balance equation and the interfacial director torque balance equation, are presented. The interfacial linear momentum balance equation for isotropic fluid–nematic liquid crystals involves the surface divergence of the surface stress tensor. It is shown that the surface stress tensor for isotropic fluid–nematic interfaces is, in most cases of interest, dominated by elastic modes. It is found that the anisotropic elastic contribution to the surface stress tensor gives rise to bending stresses, not observed in interfaces between isotropic fluids. In addition it is found that the anisotropic contribution to the surface elasticity also gives rise to tangential forces. Thus when the director orientation deviates from the easy axis of an isotropic fluid–nematic interface and the deviation has surface gradients, an orientation-driven Marangoni flow can exist. The strength of this novel effect is proportional to the anchoring energy of the interface, and the direction of flow is from low energy regions towards high energy regions, that is, from regions where the director is aligned along the easy axis towards regions where the director deviates from the easy axis.



## 1. Introduction

Capillary hydrodynamics in isotropic fluids is concerned with fluid flow phenomena in which interfacial tension is a significant effect [1]. The two important cases are flows with interfaces of finite curvature and with spatial gradients in the interfacial tension. For example, spatial gradients in the surface tension at the free surface of isotropic viscous fluids create a surface shear stress that can only be balanced by shear flow in the adjacent surface layers. The general phenomenon is known as Marangoni flow and the surface tension gradients driving the flow can be caused by temperature gradients (thermocapillary flows), surface concentration gradients (diffusocapillary flows), and electric charges (electrocapillary flows) [1, 2]. Applications of viscous flows driven by tangential stress caused by gradients in surface tensions are found in flow in porous media, damping of capillary waves, cleavage of biological cells, to name a few.

At the macroscopic level the theoretical framework to describe capillary hydrodynamics of isotropic fluids consists of the balance equations of mass, energy and momentum, and the necessary boundary conditions including the interfacial stress balance conditions, and the interface velocity conditions [3]. The interfacial stress boundary conditions play a foundational role in capillary hydrodynamics since they are involved in determining the shape (normal stress equation) of the interface as well as the presence of surface flows (tangential stress equations). In these two boundary conditions the term that balances the stresses in the bulk phases is the surface divergence of the surface stress tensor. As a consequence the surface stress tensor itself must describe the necessary deformation modes. There are many constitutive equations for the surface stress tensor such as elastic, viscoelastic, viscoplastic, etc [3]. Almost always the surface stress tensor is a $2\times 2$ symmetric tensor with normal elastic stress components and shear viscous stress components [4]. On the other hand, bending stresses are not present in isotropic fluids [4]. The presence of nematic ordering introduces anisotropic viscoelastic behaviour in the bulk nematic phase as well as anisotropic elastic modes in the interface of a nematic liquid crystal and an isotropic fluid. It will be shown below that the anisotropic surface elasticity of nematic liquid crystals introduces bending stresses not found in interfaces between isotropic fluids. Thus the conventional stress interface stress balance equations that are the foundations of capillary hydrodynamics [1, 3] need to be augmented with the nematic anisotropic viscoelastic modes.

Nematic liquid crystals are known to have a component of the surface tension that is orientation dependent [5]. A very well known expression that describes this property is the Rapini–Papoular surface free energy, which is widely used in the liquid crystals research field

*Journal of Liquid Crystals* ISSN 0267-8292 print/ISSN 1366-5855 online ©1999 Taylor & Francis Ltd
http://www.tandf.co.uk/JNLS/lct.htm
http://www.taylorandfrancis.com/JNLS/lct.htm

[6]. As shown below, nematic liquid crystals will also exhibit surface tension driven flow caused by tangential stresses that appear due to tangential surface orientation gradients. A novel phenomenon, the orientation-driven Marangoni flow, may thus appear only in the presence of weak anchoring, whenever the surface director orientation deviates from the easy axis of the surface. As in other Marangoni flows, the effect is important when the gradients in surface tension are comparable to the characteristic kinetic energy density.

The objectives of this paper are (1) to present the interfacial stress balance equations of capillary nematodynamics, and (2) to use the tangential stress balance equation of nematocapillarity to describe the orientation-induced Marangoni flow that arises in the presence of a nematic free surface or isotropic fluid–nematic interface. The main purpose of this paper is to present a new liquid crystal phenomenon that may have useful applications in material characterization and product use. To avoid lengthy repetitions of the well-known equations of nematodynamics in the bulk the reader is referred to Chapter 5 of [7].

The organization of this paper is as follows. Section 2 presents a derivation of the interface stress boundary balance equations and the surface stress tensor for a nematic liquid crystal in contact with an isotropic fluid. Section 3 derives the tangential surface force for a nematic–isotropic fluid interface and gives a simple example of the orientation driven Marangoni flow. Section 4 presents the conclusions.



## 2. Capillary nematodynamics equation

### 2.1. *Interfacial balance equations*

In this section we present the interfacial transport equations for an interphase between an isotropic viscous fluid and a uniaxial rod-like nematic liquid crystal of constant order parameter [7]. The system is isothermal, and both phases are incompressible. The interphase is assumed to be viscoelastic.

Assume that a nematic liquid crystal is undergoing flow with velocity $\mathbf{v}-$ and director $\mathbf{n}$ in region R−, and that an isotropic viscous fluid is undergoing flow with velocity $\mathbf{v}+$ in region R+. The interface between the two regions is characterized by a unit normal $\mathbf{k}$, directed from R− into R+. The flow in both regions satisfies the continuity equation and the momentum balance equation. In addition, in R− the liquid crystal satisfies the director torque balance equation. The kinematical interfacial boundary conditions are that the velocities are continuous: $\mathbf{v}^- = \mathbf{v}^+$. The dynamical boundary conditions are expressed by the interfacial linear momentum balance equations given by [3]:

$$-\mathbf{k}\cdot(\mathbf{t}^+ - \mathbf{t}^-) = \nabla_s \cdot \mathbf{t}^s \tag{1}$$

where $\mathbf{k}$ is the unit normal directed from the (−) phase towards the (+) phase, $\mathbf{t}^\pm$ is the total stress tensor in the two (±) bulk phases, $\nabla_s = \mathbf{I}_s\cdot\nabla$ is the surface gradient operator, $\mathbf{I}_s = \mathbf{I} - \mathbf{kk}$ is the surface idem factor, and $\mathbf{t}^s$ is the elastic surface stress tensor. The stress tensor in the isotropic bulk phase (+) is given by:

$$\mathbf{t}^+ = -p^+\mathbf{I} = 2\eta^+\mathbf{D}^+ \tag{2}$$

where $p^+$ is the pressure, $\eta^+$ the shear viscosity, $\mathbf{D}^+ = (\nabla\mathbf{v}^+ + \nabla\mathbf{v}^{+\mathrm{T}})/2$ is the deformation rate tensor and where the superscript T denotes the transpose. The stress tensor in the nematic phase (−) is given by [7]:

$$\mathbf{t}^- = -p^-\mathbf{I} - \frac{\partial F_b}{\partial \nabla\mathbf{n}}\cdot\nabla\mathbf{n}^{\mathrm{T}} + \alpha_1\mathbf{D}^-:\mathbf{nnnn} + \alpha_2\mathbf{nN}^- + \alpha_3\mathbf{N}^-\mathbf{n} + \alpha_4\mathbf{D}^- + \alpha_5\mathbf{nn}\cdot\mathbf{D}^- + \alpha_6\mathbf{D}^-\cdot\mathbf{nn} \tag{3}$$

where the $\{\alpha_i\}$; $i = 1, \ldots, 6$, are the Leslie viscosity coefficients, $\mathbf{N}^- = \mathrm{d}\mathbf{n}/\mathrm{d}t - \mathbf{W}^-\cdot\mathbf{n}$ the director's Jauman derivative, $\mathbf{W}^- = (\nabla\mathbf{v}^- - \nabla\mathbf{v}^{-\mathrm{T}})/2$ the vorticity tensor in the nematic phase, $F_b$ is the bulk Frank free energy density [5, 7] given by:

$$2F_b = K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\cdot\operatorname{curl}\mathbf{n})^2 + K_{33}|\mathbf{n}\times\operatorname{curl}\mathbf{n}|^2 + (K_{22} - K_{24})(\operatorname{tr}(\nabla\mathbf{n})^2 - (\operatorname{div}\mathbf{n})^2) \tag{4}$$

and the Frank constants $\{K_{ii}\}$; $ii = 11, 22, 33, 24$, are the elastic moduli for splay, twist, bend and saddle–splay deformations, respectively.

The surface stress tensor $\mathbf{t}^s$ is given by the sum of the elastic $\mathbf{t}^{se}$ and the viscous $\mathbf{t}^{sv}$ contributions. The surface elastic stress tensor is given by [8, 9]:

$$\mathbf{t}^{se} = F_s\mathbf{I}_s - F_s'\mathbf{I}_s\cdot\mathbf{nk} \tag{5a}$$

$$F_s' = \frac{\mathrm{d}F_s}{\mathrm{d}(\mathbf{n}\cdot\mathbf{k})} \tag{5b}$$

where $F_s$ is the surface free energy density, here taken to be a function of $\mathbf{n}\cdot\mathbf{k}$. An example of a widely use constitutive equation for $F_s$ is the Rapini–Papoular expression [5, 6]:

$$F_s = \sigma[1 + \tau(\mathbf{n}\cdot\mathbf{k})^2] \tag{6}$$

where $\sigma$ is the isotropic interfacial tension and $\sigma_a = \sigma\tau$ is the anchoring energy [6, 7]. The surface elastic stress tensor $\mathbf{t}^{se}$ can be expressed as:

$$\mathbf{t}^{se} = F_s\mathbf{I}_s - F_s'(\mathbf{n}\cdot\mathbf{k})[\mathbf{i}_1\mathbf{k} + \mathbf{i}_2\mathbf{k}] \tag{7}$$

where $(\mathbf{i}_1, \mathbf{i}_2)$ are the surface unit orthonormal base vectors. In component form $\mathbf{t}^{se}$ is given as:

$$\mathbf{t}^{se} = \mathbf{i}_1\mathbf{i}_1 t_{11}^{se} + \mathbf{i}_2\mathbf{i}_2 t_{22}^{es} + \mathbf{i}_1\mathbf{k}t_{13}^{se} + \mathbf{i}_2\mathbf{k}t_{23}^{se}. \tag{8}$$

The surface elastic stress tensor contains the usual normal stresses (components 11 and 22) and surface bending stresses (components 13 and 23). These bending stresses are usually absent in fluids systems [4] but for nematics they arise when the surface orientation deviates from the easy axis. The magnitude of bending stresses is:

$$t_{13}^{se} = -F_s'(\mathbf{i}_1 \cdot \mathbf{K});\ t_{23}^{se} = -F_s'(\mathbf{i}_2 \cdot \mathbf{K}) \quad (3a,b) \tag{9}$$

Equation (8) shows that for a nematic liquid crystal in the presence of weak anchoring $\mathbf{t}^{se}$ is asymmetric and possesses at most four components. It is worth observing that including higher order terms in the classical Rapini–Papoular expression (6) will not modify the tensor structure of $\mathbf{t}^{se}$.

To find an expression for the surface extra stress tensor $\mathbf{t}^{sv} = \mathbf{t}^{ssv} + \mathbf{t}^{sav}$ we identify the forces and fluxes that contribute to the surface rate of entropy production $\Delta$, as follows:

$$\Delta = \mathbf{t}^{ssv} : \mathbf{D}^s + \mathbf{t}^{sav} : \mathbf{W}^s + \mathbf{h}^{v\parallel} \cdot \frac{d\mathbf{n}^{\parallel}}{dt} + \mathbf{h}^{v\perp} \cdot \frac{d\mathbf{n}^{\perp}}{dt} \tag{10}$$

where $\mathbf{t}^{ssv}$ is the surface symmetric extra stress tensor, $\mathbf{D}^s$ is the surface rate of deformation tensor,

$$2\mathbf{D}^s = (\nabla \mathbf{v}^s \cdot \mathbf{I}_s + \mathbf{I}_s \nabla \mathbf{v}^{sT}) \tag{11}$$

where $\mathbf{v}^s$ is the surface velocity vector, $\mathbf{t}^{sav}$ is the surface asymmetric extra stress tensor, $\mathbf{W}^s$ is the surface vorticity tensor,

$$2\mathbf{W}^s = (\nabla \mathbf{v}^s \cdot \mathbf{I}_s - \mathbf{I}_s \cdot \nabla \mathbf{v}^{sT}) \tag{12}$$

where $\mathbf{h}^{v\parallel}$ is the parallel component of the surface viscous molecular field, $d\mathbf{n}^{\parallel}/dt$ is the total derivative of the parallel director component,

$$\frac{d\mathbf{n}^{\parallel}}{dt} = \frac{\partial \mathbf{n}^{\parallel}}{\partial t} + \mathbf{v}^s \cdot \nabla_s \mathbf{n}^{\parallel};\quad \mathbf{n}^{\parallel} = \mathbf{I}_s \cdot \mathbf{n} \tag{13a,b}$$

where $\mathbf{h}^{v\perp}$ is the normal component of the surface viscous molecular field, $d\mathbf{n}^{\perp}/dt^s$ is the total derivative of the normal director component, and

$$\frac{d\mathbf{n}^{\perp}}{dt} = \frac{\partial \mathbf{n}^{\perp}}{\partial t} + \mathbf{v}^s \cdot \nabla_s \mathbf{n}^{\perp};\quad \mathbf{n}^{\perp} = \mathbf{kk} \cdot \mathbf{n}. \tag{14a,b}$$

Expressing $\mathbf{t}^{sav}$ in terms of the parallel components of the molecular field and director, $\Delta$ becomes

$$\Delta = \mathbf{t}^{ssv} : \mathbf{D}^s + \mathbf{h}^{v\parallel} \cdot \mathbf{N}^{\parallel} + \mathbf{h}^{v\perp} \cdot \frac{d\mathbf{n}^{\perp}}{dt};\quad \mathbf{N}^{\parallel} = \frac{d\mathbf{n}^{\parallel}}{dt} - \mathbf{W}^s \cdot \mathbf{n}^{\parallel}. \tag{15}$$

Expanding the fluxes ($\mathbf{t}^{ssv}, \mathbf{h}^{v\parallel}, \mathbf{h}^{v\perp}$) in terms of the forces ($\mathbf{D}^s, \mathbf{N}^{\parallel}, d\mathbf{n}^{\perp}/dt$) we find that the symmetric surface viscous stress tensor and molecular fields are given by:

$$\mathbf{t}^{ssv} = \alpha_1^s \mathbf{D}^s : \mathbf{n}^{\parallel}\mathbf{n}^{\parallel}\mathbf{n}^{\parallel}\mathbf{n}^{\parallel} + \frac{1}{2}(\alpha_2^s + \alpha_3^s)(\mathbf{N}^{\parallel}\mathbf{n}^{\parallel} + \mathbf{n}^{\parallel}\mathbf{N}^{\parallel})$$
$$+ \alpha_4^s \mathbf{D}^s + \frac{1}{2}(\alpha_5^s + \alpha_6^s)(\mathbf{D}^s \cdot \mathbf{n}^{\parallel}\mathbf{n}^{\parallel} + \mathbf{n}^{\parallel}\mathbf{n}^{\parallel} \cdot \mathbf{D}^s) \tag{16a}$$

$$\mathbf{h}^{v\parallel} = \gamma_2^s \mathbf{D}^s \cdot \mathbf{n}^{\parallel} + \gamma_1^s \mathbf{N}^{\parallel} \tag{16b}$$

$$\mathbf{h}^{v\perp} = \gamma_1^s \frac{d\mathbf{n}^{\perp}}{dt} \tag{16c}$$

where the $\{\alpha_i^s\}$; $i = 1, \ldots, 6$, are the surface viscosity coefficients, with units of energy × time/area, and the $\{\gamma_i^s\}$; $i = 1, 2$ are the torque coefficients, with units of energy × time/area, and due to Onsager reciprocal relations the coefficients are related as follows: $\gamma_1^s = \alpha_3^s - \alpha_2^s$, $\gamma_2^s = \alpha_3^s + \alpha_2^s = \alpha_6^s + \alpha_5^s$, as found in the bulk case [7]. The antisymmetric surface viscous stress tensor is given by: $\mathbf{t}^{sav} = (\mathbf{h}^{v\parallel}\mathbf{n}^{\parallel} - \mathbf{n}^{\parallel}\mathbf{h}^{v\parallel})/2$.

Finally the interfacial director torque balance equation is given by the balance of the surface elastic torque $\Gamma^{se}$ and the surface viscous torque $\Gamma^{sv}$:

$$\Gamma^{se} + \Gamma^{sv} = 0 \tag{17a}$$

$$\Gamma^{se} = \mathbf{n} \times \mathbf{h}^{se};\quad \Gamma^{sv} = -\mathbf{n} \times (\mathbf{h}^{v\parallel} + \mathbf{h}^{v\perp}) \tag{17b,c}$$

where the surface elastic molecular field $\mathbf{h}^{se}$ is given by [5, 8]:

$$\mathbf{h}_i^{se} = -\frac{\partial F_s}{\partial n_i} - \frac{\partial F_b}{\partial n_{i,j}} k_j \tag{18}$$

The relative ratio of the characteristic viscous stresses to elastic stresses and the relative ratio of the characteristic viscous torques to elastic torques, gives, respectively:

$$\frac{\|\mathbf{t}^{sv}\|}{\|\mathbf{t}^{se}\|} = Ca\frac{h}{L};\quad Ca = \frac{\alpha_4 U}{\sigma} \tag{19a}$$

$$\frac{\|\mathbf{h}^{sv}\|}{\|\mathbf{h}^{se}\|} = E_r\frac{h}{L};\quad E_r = \frac{\gamma_1 U}{\sigma_a} \tag{19b}$$

where $Ca$ is the capillary number, $E_r$ the surface Ericksen number, $\sigma_a$ is the anchoring energy, $U$ is a characteristic velocity, $L$ is a characteristic macroscopic length, and $h$ is the microscopic thickness of the surface layer. The value of $h$ is of the order $10^{-8}$–$10^{-9}$ m [10]. Thus for most practical applications:

$$\frac{\|\mathbf{t}^{sv}\|}{\|\mathbf{t}^{se}\|} = Ca\frac{h}{L} \ll 1;\quad \frac{\|\mathbf{h}^{sv}\|}{\|\mathbf{h}^{se}\|} = E_r\frac{h}{L} \ll 1 \tag{20a,b}$$

and the interface can be considered to be purely elastic. In such cases we can neglect the surface viscous stresses and torques, $\mathbf{h}^{sv} = \mathbf{t}^{sv} = 0$.

### 2.2. *Normal and tangential force balances*

The normal force balance equation reflects the shape effects of the interface and involves the role of surface tension due to surface curvature effects. It is obtained by projecting the vector equation (2) along the unit normal $\mathbf{k}$:

$$\begin{aligned} -\mathbf{k}\;\; (\mathbf{t}^{+}-\mathbf{t}^{-})\;\; \mathbf{kk} &= F_{s}[2H]\mathbf{k}+F_{s}'[\mathbf{kk}:\nabla_{s}\mathbf{n}^{T}-\nabla_{s}\;\;\mathbf{n}-2H(\mathbf{n}\;\;\mathbf{k})]\mathbf{k} \\ &\quad -F_{s}''[\mathbf{kn}:\nabla_{s}\mathbf{n}+\mathbf{nn}:\nabla_{s}\mathbf{k}]\mathbf{k}+\nabla_{s}\;\;\mathbf{t}^{sv}\;\;\mathbf{kk} \qquad (21) \end{aligned}$$

where $H$ is the mean surface curvature: $H=-1/2\nabla_{s}\;\;\mathbf{k}$. In the absence of flow the equation reduces to the static normal force balance equation that can be considered as the Laplace equation for nematic–isotropic systems. The tangential force balance equation involves gradients in the surfance free energy density and is obtained by projecting equation (2) along the tangent direction:

$$-\mathbf{k}\;\;(\mathbf{t}^{+}-\mathbf{t}^{-})\;\;\mathbf{I}_{s}=F_{s}'[\mathbf{k}\;\;\nabla_{s}\mathbf{n}^{T}]\;\;\mathbf{I}_{s}+\nabla_{s}t^{sv}\;\;\mathbf{I}_{s} \qquad (22)$$

where the first term on the right hand side is the tangential force due to surface orientation gradients.

## 3. Orientation-induced Marangoni flow

In capillary hydrodynamics the effect produced by tangential forces due to interfacial tension gradients is known as the Marangoni effect [1, 2, 3]. The Marangoni flows are known as thermocapillary (thermal gradients), difussocapillary (concentration gradients) and electrocapillary (electric charge gradients) flows. To this list one may add nematocapillary flows, which are flows driven by orientation gradients. The orientation-induced Marangoni effect for nematic liquid crystals is given by the following tangential force:

$$\mathbf{f}_{\|}=F_{s}'[\mathbf{k}\;\;\nabla_{s}\mathbf{n}^{T}]\;\;\mathbf{I}_{s}. \qquad (23)$$

For the Rapini–Papoular constitutive equation (6) the tangential force $\mathbf{f}_{\|}$ becomes:

$$\mathbf{f}_{\|}=2\sigma\tau(\mathbf{n}\;\;\mathbf{k})(\mathbf{k}\;\;\nabla_{s}\mathbf{n}^{T})\;\;\mathbf{I}_{s}. \qquad (24)$$

Thus to observe this effect the surface director orientation must deviate from the easy axis of the nematic–isotropic surface. If the scalar order parameter [7] is not constant, Marangoni flow can also occur due to surface gradients of the tensor order parameter; this more general case will be treated in another publication. The orientation-induced Marangoni effect creates a flow from the region of low interfacial tension to the region of high interfacial tension. In our case the flow will be from regions where the director is along the easy axis to regions where the director deviates from the easy axis.

As a simple example of this novel phenomenon, consider a nematic free standing film with a splay–bend inversion wall [7] along the $z$-direction due to an imposed magnetic field along the $x$-direction: $\mathbf{H}=(H,0,0)$. The director field is assumed to be given by $\mathbf{n}=[\sin\alpha(x),0,\cos\alpha(x)]$. The easy axis and the outward unit normal are along $z$: $\mathbf{k}=(0,0,1)$. At the centre of the wall ($x=0$) the director is along the easy axis ($\alpha=0$), and sufficiently far ($x\to\pm\infty$) from the centre of the inversion wall the director is aligned along the magnetic field ($\alpha=\pm\pi/2$). The tangential force is in the $x$-direction and is given by:

$$f_{\|x}=\sigma|\tau|\sin(2\alpha)\frac{d\alpha}{dx} \qquad (25)$$

showing that the flow is from the low energy ($x=0$) region to the high energy regions ($x\to\pm\infty$), as in all Marangoni flows. The magnitude of the flow will be proportional to the surface anchoring energy $\sigma|\tau|$, where $\tau$ is a negative constant for homeotropic anchoring.

## 4. Conclusions

The interfacial stress balance equation for liquid crystals involves the surface divergence of the surface stress tensor. For many common cases the surface stress tensor can be considered purely elastic. The anisotropic elastic contribution to the surface stress tensor gives rise to bending stresses, not observed in isotropic materials. The anisotropic contribution to the surface elasticity also gives rise to tangential forces. Thus when the director orientation deviates from the easy axis of an isotropic fluid–nematic interface and the deviation has surface gradients, an orientation-driven Marangoni flow can exist. The strength of the effect is proportional to the anchoring energy characteristic of the interface, and the direction of flow is from low energy regions towards high energy regions, that is, from regions where the director is aligned along the easy axis towards regions where the director deviates from the easy axis.

Financial support of the Natural Sciences and Engineering Research Council (NSERC) of Canada is gratefully acknowledged.

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