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

# On higher order variational analysis in one and three dimensions for soft boundaries

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**To cite this Article** Dahl, Ingolf and De Meyere, Arnout(1995) 'On higher order variational analysis in one and three dimensions for soft boundaries', Liquid Crystals, 18: 5, 683 — 692
**To link to this Article: DOI:** 10.1080/02678299508036677
**URL:** http://dx.doi.org/10.1080/02678299508036677

LIQUID CRYSTALS, 1995, VOL. 18, NO. 5, 683–692

# On higher order variational analysis in one and three dimensions for soft boundaries

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(*Received 15 April 1994; accepted 13 July 1994*)

For some problems in liquid crystal physics we need to use the Euler equation and the corresponding boundary equation in the three-dimensional case with soft boundaries. As a further complication the free energy expression, which should be minimized, might contain some second-order and third-order derivatives. These higher-order derivatives will cause the spatial derivatives of the boundary normal to appear in the boundary equation. Explicit formulae are given for the Euler equation and the corresponding surface equations for a general case. As an example, the theory is applied to nematic liquid crystals, where the general Euler equations and surface molecular fields are derived, including the effects of an imposed electric field.



## 1. Introduction

Variational analysis is used in different areas of physics to find the differential equation for the function that minimizes a given integral. An introduction to this subject can be found in for instance Arfken [1]. For nematic liquid crystals, variational analysis could be used to find the equilibrium conditions for the director field, if the boundary conditions and the expression for the deformational energy are known. Sometimes, however, the physical problems are a little too complicated to be solved by straight application of the textbook case. In this paper we are interested in including higher order derivatives of the function in the integral, and also in including variable boundary conditions. This enables us to solve some problems in the physics of liquid crystals. In the elasticity theory of nematics there are two terms involving second order derivatives in the volume free energy

$$K_{13}\nabla\cdot(\hat{\mathbf{n}}(\nabla\cdot\hat{\mathbf{n}})) - \tfrac{1}{2}(K_{22}+K_{24})\nabla\cdot(\hat{\mathbf{n}}(\nabla\cdot\hat{\mathbf{n}}) + \hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}})), \quad (1)$$

where $K_{13}$, $K_{22}$, and $K_{24}$ are elastic constants and $\hat{\mathbf{n}}$ is the nematic director, according to the notation of Nehring and Saupe [2]. Both these terms measure the divergence of some vector field. In the elasticity theory of smectic C, there are twelve similar terms, see [3]

$$\sum_{i=1}^{12} E_{vi}\nabla\cdot\mathbf{e}_i, \quad (2)$$

where $\mathbf{e}_i$ denote flexoelectric vector fields and $E_{vi}$ are the corresponding elastic constants. The interpretation of these divergence terms has for nematics caused a never-ending discussion [4–14]. Evidently we can use Gauss' theorem to rewrite these volume terms as surface terms, and for his reason we denote them 'gauge term' [15].

The surface energy will not contribute to the Euler equations for the internal volume elements, and thus these equations must in some way be invariant to our application of the Gauss' theorem. This could be achieved in a simple way if the gauge terms do not contribute to the Euler equations for the internal volume elements. Also the gauge terms should influence the boundary conditions in the same way if they are written as volume terms as if they are rewritten as surface terms. As surface energy terms, the gauge terms depend on the spatial derivatives of the orientation of the liquid crystal. Then we will not obtain the standard type of fixed boundary condition for the Euler equations. Some of these gauge terms involve second-order derivatives of the director or C-director, and this implies that we may have to use second-order variational theory [10, 11, 16]. In the elasticity theory of the smectic C phase, even third order derivatives might enter. These

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0267–8292/95 $10·00 

issues in the physics of liquid crystals justify an investigation of a mathematical problem: how to extend the variational calculus so that the extra complications of higher-order derivatives, three dimensions, and soft boundaries can be included.

In this article we will first look at a one-dimensional analogue to the free energy, and derive the Euler equation for the case of variable boundary conditions. The boundary conditions are controlled by a boundary energy expression, corresponding to the surface energy for the three-dimensional case. To simulate the action of the gauge terms, and to illustrate the difficulties introduced by these, we will let the boundary energy depend on the spatial derivative of the variable function. Some authors use the method of 'extremals' to solve such a variational problem. We will point out some difficulties in such an approach. Instead we have to introduce higher-order terms in the volume free energy, so that the variational problem becomes well stated. We solve then the corresponding variational problem for the three-dimensional case. The results are applied to the elasticity theory of nematic liquid crystals, and explicit expressions for the surface molecular fields are given in general vector notation.



## 2. Variation calculus at the surface in the one-dimensional case

For the sake of discussion, let us rederive the Euler equation for one dependent and one independent variable by calculus of variations, using the formalism of Arfken [1]. Instead of fixed boundary conditions, we shall assume variable boundary conditions and an energy contribution from these ('soft boundaries'). For instance Oldano and Barbero [17] have discussed this case in connection with liquid crystal problems. Here we are interested in the end conditions, since the arguments should be transferable to the boundary conditions for liquid crystals. Let us thus minimize the quantity

$$J = \int_{x_1}^{x_2} g_v(u, u_x, x)\, dx - g_s(u, u_x, x_1) + g_s(u, u_x, x_2). \quad (3)$$

We can see this as a one-dimensional analogue to the elastic free energy expression for liquid crystals. Here $g_v$ and $g_s$ are known functions of the indicated scalar variables $u$, $u_x = d_u/d_x$, and $x$, but the dependence of $u$ on $x$ is not fixed, so $u(x)$ is unknown. The signs for the end contributions $g_s$ have been chosen so that we obtain a convenient notation below. In the same way as in Arfken, we will use an arbitrary deformation $\eta(x)$ and a scale factor $\alpha$, and make a small variation around the path $u(x, \alpha = 0)$, which minimizes the quantity $J$. Thus,

$$u(x, \alpha) = u(x, 0) + \alpha\eta(x). \quad (4)$$

For our more general case we cannot assume as Arfken did that $\eta(x_1)$ and $\eta(x_2)$ are zero. The quantity $J$ now becomes a function of $\alpha$,

$$J = \int_{x_1}^{x_2} g_v(u(x, \alpha), u_x(x, \alpha), x)\, dx - g_s(u(x_1, \alpha), u_x(x_1, \alpha), x_1) + g_s(u(x_2, \alpha), u_x(x_2, \alpha), x_2). \quad (5)$$

The derivative of $J$ with respect to $\alpha$ should be zero for the extreme value at $\alpha = 0$. In the standard way by integration by parts we obtain

$$\frac{dJ}{d\alpha} = \int_{x_1}^{x_2} \eta(x) \left( \frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x} \frac{\partial g_v}{\partial u_x} \right) dx + \eta(x) \frac{\partial g_v}{\partial u_x} \Bigg|_{x_1}^{x_2} + \eta(x) \frac{\partial g_s}{\partial u} \Bigg|_{x_1}^{x_2} + \frac{\partial \eta}{\partial x} \frac{\partial g_s}{\partial u_x} \Bigg|_{x_1}^{x_2}, \quad (6)$$

which thus should be zero for $\alpha = 0$. In the interior of the interval $(x_1, x_2)$ we can use test functions $\eta(x)$, which vanish at the border, and also have a vanishing derivative at the border. Thus, the Euler equation,

$$\frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x} \frac{\partial g_v}{\partial u_x} = 0, \quad (7)$$

must be satisfied inside the interval. Due to the $\partial\eta/\partial x$ term, the end condition is more difficult to handle. Oldano and Barbero [17] assume that $\eta(x)$ and $\partial\eta/\partial x$ could be varied independently at the border, and in this way they get four boundary conditions, two at each border. Since the Euler equation, at least for their special case, depends only on two independent parameters, they obtain an overdetermined set of equations. Usually this set has no solution. In this way they found a kind of paradox.

## 3. The method of extremals

The method of extremals [7] has been suggested as a strategy to solve this paradox. By this method, we should not vary both $\eta(x)$ and $\partial\eta/\partial x$ independently at each border. Instead, we should restrict the variational problem to such functions that satisfy the Euler equation in the interior. These functions are denoted 'extremals'. For each set of boundary conditions for $u(x_1)$ and $u(x_2)$, the Euler equation for the interior will give us back a value for $\partial\eta/\partial x$ at the end points. This determines the relation between $\eta(x)$ and $\partial\eta/\partial x$. Thus, by varying the boundary conditions and by solving the Euler equation for each choice, it could be possible to minimize the $J$ expression, without any paradoxes. It is a bit discomforting that no local expression similar to the Euler equation is available, but anyway we may have to live with that. We could however find a global condition: Assume that $\alpha$ now is a general parameter such that $u(x, \alpha)$ satisfies the Euler equation for each choice of $\alpha$. With

$$J = \int_{x_1}^{x_2} g_v(u(x, \alpha), u_x(x, \alpha), x)\, dx - g_s(u(x_1, \alpha), u_x(x_1, \alpha), x_1) + g_s(u(x_2, \alpha), u_x(x_2, \alpha), x_2), \quad (8)$$

the derivative with respect to $\alpha$ is given by

$$\frac{dJ}{d\alpha} = \int_{x_1}^{x_2} \left( \frac{\partial g_v}{\partial u}\frac{\partial u}{\partial \alpha} + \frac{\partial g_v}{\partial u_x}\frac{\partial u_x}{\partial \alpha} \right) dx + \frac{\partial g_s}{\partial u}\frac{\partial u}{\partial \alpha}\Big|_{x_1}^{x_2} + \frac{\partial g_s}{\partial u_x}\frac{\partial u_x}{\partial \alpha}\Big|_{x_1}^{x_2}. \tag{9}$$

But since $u$ satisfies the Euler equation (7), we get

$$\int_{x_1}^{x_2} \frac{\partial g_v}{\partial u}\frac{\partial u}{\partial \alpha}\,dx = \int_{x_1}^{x_2} \frac{\partial u}{\partial \alpha}\left( \frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_x} \right) dx = \frac{\partial u}{\partial \alpha}\frac{\partial g_v}{\partial u_x}\Big|_{x_1}^{x_2} - \int_{x_1}^{x_2} \left( \frac{\partial}{\partial x}\frac{\partial u}{\partial \alpha} \right) \frac{\partial g_v}{\partial u_x}\,dx, \tag{10}$$

and thus

$$\frac{dJ}{d\alpha} = \int_{x_1}^{x_2} \frac{\partial g_v}{\partial u_x}\left( \frac{\partial u_x}{\partial \alpha} - \left( \frac{\partial}{\partial x}\frac{\partial u}{\partial \alpha} \right) \right) dx + \frac{\partial u}{\partial \alpha}\frac{\partial g_v}{\partial u_x}\Big|_{x_1}^{x_2} + \frac{\partial g_s}{\partial u}\frac{\partial u}{\partial \alpha}\Big|_{x_1}^{x_2} + \frac{\partial g_s}{\partial u_x}\frac{\partial u_x}{\partial \alpha}\Big|_{x_1}^{x_2}, \tag{11}$$

which should be zero for a function that gives an extreme value of $J$. The factor $\alpha$ could here be the end value $u(x_1)$ with $u(x_2)$ kept fixed or vice versa.

Thus one might expect to find extreme values to the quantity $J$ when the set of functions $u(x)$ is limited to the extremals that satisfy the Euler equation inside the interval $(x_1, x_2)$. But this set of functions has not been defined in the specification of the problem. It has instead been chosen as a part of the solution of the problem. If some function in this set is giving $J$ an extreme value, this method of extremals will find it. However, as usual with variational analysis, we are not guaranteed that any function found will be the one that we are looking for. It is even worse than that, since in the general case we could find other functions, outside the set of extremals, which give lower values of the integral $J$ than any candidate from the set of extremals. To illustrate this, let us assume that there is a minimizing function $u^*$ in the set of extremals. We could try to find a local end condition, by noting that it is possible to restrict the interval. Any solution $u^*$, minimizing $J$, also minimizes

$$\Delta J = \int_{x_1}^{x_1+\delta} g_v(u, u_x, x)\,dx - g_s(u, u_x, x_1) \tag{12}$$

for fixed boundary conditions $u^*(x_1 + \delta)$ and $u^*(x_1 + \delta)$ for $u$ and $u_x$ in the point $x = x_1 + \delta$. This is found by using a test function $\eta(x)$ with a continuous derivative and with $\eta(x) = 0$ for $x > x_1 + \delta$. If the function $u^*$ satisfies the Euler equation, the boundary conditions are strong enough to determine $u^*$ completely, and we have nothing to vary for each fixed boundary condition. However, if we allow a breakdown near the surface, we could check for stability. We can use a function $u^*(x, 0)$, satisfying the Euler equation, but add to it the test function

$$\eta(x) = (x - x_1 - \delta)^2, \tag{13}$$

multiplied by the scalar $\alpha$. In general, the sum $u(x, \alpha)$ will then not satisfy the Euler equation. This test function gives

$$\frac{d\Delta J}{d\alpha} = \int_{x_1}^{x_1+\delta} (x - x_1 - \delta)^2 \left( \frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_x} \right) dx - \delta^2 \frac{\partial g_v}{\partial u_x}\Big|_{x=x_1} - \delta^2 \frac{\partial g_s}{\partial u} + 2\delta \frac{\partial g_s}{\partial u_x}. \tag{14}$$

For $\alpha = 0$ the integral in equation (14) vanishes, thus

$$\frac{d\Delta J}{d\alpha}\Big|_{\alpha=0} = -\delta^2 \frac{\partial g_v}{\partial u_x}\Big|_{x=x_1} - \delta^2 \frac{\partial g_s}{\partial u} + 2\delta \frac{\partial g_s}{\partial u_x}. \tag{15}$$

If $\alpha = 0$ should give a minimum, this expression should be zero. But $\delta$ could be chosen in an arbitrary way, and by varying $\delta$ we get two conditions which must be satisfied simultaneously

$$\frac{\partial g_s}{\partial u_x}\Big|_{x=x_1} = 0, \tag{16}$$

and

$$\frac{\partial g_v}{\partial u_x}\Big|_{x=x_1} = -\frac{\partial g_s}{\partial u}\Big|_{x=x_1}. \tag{17}$$

If there are first order derivatives $u_x$ in the surface expression $g_s$, then these two conditions could not in general be satisfied simultaneously at both ends of the interval. Thus we have to look for functions $u(x)$ *not* satisfying the Euler equation to minimize $J$. But also, we know that any function minimizing $J$ should satisfy the Euler equation inside the interval. So we might get a contradiction from the assumption that a regular function is minimizing $J$.

We have thus a dilemma and a choice: either we restrict the allowed set of functions to those satisfying the Euler equations, and could then eventually obtain a well-defined minimum, or we allow a larger set of functions, and will then probably have to look for functions with singular behaviour near the end points. The mathematics thus tells us to return to our physical problem and to study it more closely to be able to specify the mathematical problem in a better way. It is reasonable that this conclusion is also valid in the three-dimensional case.

## 4. The method of second-order derivatives

Another way to look at the expression

$$J = \int_{x_1}^{x_2} g_v(u, u_x, x)\,dx - g_s(u, u_x, x_1) + g_s(u, u_x, x_2) \tag{18}$$

is to see it as a special case of the integral

$$J_2 = \int_{x_1}^{x_2} g_v(u, u_x, u_{xx}, x)\,dx - g_s(u, u_x, x_1) + g_s(u, u_x, x_2), \tag{19}$$

thus involving also second-order derivatives. With the same kind of test function as in (4), we obtain

$$\frac{dJ_2}{d\alpha} = \int_{x_1}^{x_2} \eta(x) \left( \frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_x} + \frac{\partial^2}{\partial x^2}\frac{\partial g_v}{\partial u_{xx}} \right) dx + \eta(x)\frac{\partial g_v}{\partial u_x}\Big|_{x_1}^{x_2} + \frac{\partial \eta}{\partial x}\frac{\partial g_v}{\partial u_{xx}}\Big|_{x_1}^{x_2} - \eta(x)\frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_{xx}}\Big|_{x_1}^{x_2} + \eta(x)\frac{\partial g_s}{\partial u}\Big|_{x_1}^{x_2} + \frac{\partial \eta}{\partial x}\frac{\partial g_s}{\partial u_x}\Big|_{x_1}^{x_2}. \quad (20)$$

If the function $g_v$ contains terms of the type $u_{xx}^2$, the Euler equation

$$\frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_x} + \frac{\partial^2}{\partial x^2}\frac{\partial g_v}{\partial u_{xx}} = 0 \quad (21)$$

will be of fourth order, and thus we have four unknown constants to be determined to the boundary conditions. Thus we can allow two boundary conditions at each end of the interval, and can allow the independent variation of $\eta(x)$ and $\partial\eta/\partial x$. At the $x_1$ end we then get

$$\frac{\partial g_v}{\partial u}\Big|_{x=x_1} - \frac{\partial}{\partial x}\frac{\partial g_v}{\partial u_{xx}}\Big|_{x=x_1} + \frac{\partial g_s}{\partial u}\Big|_{x=x_1} = 0 \quad (22)$$

and

$$\frac{\partial g_v}{\partial u_{xx}}\Big|_{x=x_1} + \frac{\partial g_s}{\partial u_x}\Big|_{x=x_1} = 0. \quad (23)$$

Thus, the variational problem will have a well-defined mathematical solution. If we use a simple test function for $f$

$$g_v = Au_x^2 + Bu_{xx}^2, \quad (24)$$

we can easily see that a non-zero $B$ will introduce two extra solutions to the Euler equation. These solutions are

$$u = \exp\left( \pm x \sqrt{\frac{A}{B}} \right), \quad (25)$$

while the other solutions,

$$u = x \quad \text{and} \quad u = 1, \quad (26)$$

are not dependent on the size of $B$. In a given problem, we have to match a linear combination of these solutions to the boundary conditions. If the second-order elastic constant $B$ is small enough, it will only affect the boundary layers. In the interior of the volume the first-order elasticity will control the behaviour.

Thus the introduction of quadratic terms in the second-order derivatives essentially solves the dilemma introduced by gradient terms as boundary conditions. But for nematics, we then face the problem of using a full second-order elasticity theory in the sense of Barbero *et al.* [10], and that requires the introduction of a huge number of additional elastic constants. Maybe just a few of these are needed to control these gradient terms? It is also of interest to see under which geometrical conditions these gradient terms could be important. We then need the second order Euler equations in the 3D case.

## 5. Variation calculus at the surface in three dimensions

When we go to the three-dimensional case, and also want to include second-order terms and variation at the surface, we will encounter some new complications, which we do not have to worry about in simpler cases. Let us thus extend the integral $J$ to cover three dimensions

$$J = \int_V g_v(u, u_i, u_{ij}, x_i)\, dV + \int_S g_s(u, u_i, x_i)\, dS, \quad (27)$$

where the surface $S$ encloses the volume $V$ and where $u_i$ are the first order derivatives of $u$ with respect to the spatial derivatives, and $u_{ij}$ are the second order derivatives. By the standard procedure by adding a small disturbance $\alpha\eta(x_i)$, we obtain

$$\frac{dJ}{d\alpha} = \int_V \eta \left( \frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x_i}\frac{\partial g_v}{\partial u_i} + \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{ij}} \right) dV + \int_S \eta \left( s_i \left( \frac{\partial g_v}{\partial u_i} - \frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{ij}} \right) + \frac{\partial g_s}{\partial u} \right) dS + \int_S \eta_i \left( s_j \frac{\partial g_v}{\partial u_{ij}} + \frac{\partial g_s}{\partial u_i} \right) dS, \quad (28)$$

where $s_i$ are the components of the normalized outward surface normal $\hat{\mathbf{s}}$, $\eta i$ are the derivatives of $\eta$ with respect to the coordinate $x_i$, and repeated indices are summed over. Note in this expression that when we give the full functional form of $g_v$, we have to discriminate between terms containing $u_{12}$ and terms containing $u_{21}$, to avoid counting them twice when we take the derivatives of $g_v$ with respect to these two variables. We thus get the usual Euler equation for variation of $\eta$ inside the volume

$$\frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x_i}\frac{\partial g_v}{\partial u_i} + \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{ij}} = 0. \quad (29)$$

To find the corresponding surface expressions, we cannot simply let $\eta$ and $\eta_i$ vary independently at the surface. If we know the value of $\eta$ at the surface, we have also full information about how it varies along the surface, but no information about how it varies along the surface normal. This could be described mathematically by a theorem, similar to Gauss' theorem, but only involving surface integrals. To derive this theorem, we start with Stokes' theorem applied to a closed surface

$$\int_S \hat{\mathbf{s}} \cdot (\nabla \times \mathbf{V})\, dS = 0. \quad (30)$$

If we now make the substitution $\mathbf{V} \leftarrow \eta\mathbf{v} \times \hat{\mathbf{s}}$, where $\mathbf{v}$ is a

vector field, we get the theorem

$$\int_S (\nabla\eta)\cdot\mathbf{v}\,dS = -\int_S \eta\hat{\mathbf{s}}\cdot(\nabla\times(\hat{\mathbf{s}}\times\mathbf{v}))\,dS + \int_S (\hat{\mathbf{s}}\cdot\nabla\eta)(\hat{\mathbf{s}}\cdot\mathbf{v})\,dS. \quad (31)$$

This theorem can be applied to the last integral in equation (28) if we define $\mathbf{v}$ as the vector that has the components

$$v_i = s_j\frac{\partial g_v}{\partial u_{ij}} + \frac{\partial g_s}{\partial u_i}. \quad (32)$$

The surface part of $dJ/d\alpha$ becomes then

$$\int_S \eta\left(s_i\left(\frac{\partial g_v}{\partial u_i} - \frac{\partial}{\partial x_j}\left(\frac{\partial g_v}{\partial u_{ij}} + s_iv_j - s_jv_i\right)\right) + \frac{\partial g_s}{\partial u}\right)dS + \int_S s_i\eta_i s_j v_j\,dS. \quad (33)$$

Now it should be possible to vary $\eta$ and $\hat{\mathbf{s}}\cdot\nabla\eta = s_i\eta_i$ (the derivative of $\eta$ along the surface normal) independently at the surface, and in this way we obtain the surface conditions corresponding to the Euler equation. For $\eta$ we get what we will call 'the first surface relation'

$$s_i\left(\frac{\partial g_v}{\partial u_i} - \frac{\partial}{\partial x_j}\left(\frac{\partial g_v}{\partial u_{ij}} + s_iv_j - s_jv_i\right)\right) + \frac{\partial g_s}{\partial u} = 0, \quad (34)$$

and for $s_i\eta_i$ we get 'the second surface relation'

$$s_jv_j = 0. \quad (35)$$

The expressions obtained are not fully symmetric with regard to interchange of the indices in $u_{ij}$. To obtain such symmetry, we can require that the second order derivatives $\eta_{ij}$ of the variation $\eta$ should be independent of the derivation order:

$$\eta_{ij} = \eta_{ji}. \quad (36)$$

Then we can interchange the order in which we do the partial integrations in the derivation of equation (28), and that will result in expressions similar to equations (29), (32), (34), and (35), just with $u_{ij}$ replaced by $u_{ji}$. Thus, by subtracting and adding these new equations with the original ones, we conclude that $g_v$ must satisfy the relation

$$\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{ij}} = \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{ji}}. \quad (37)$$

and that in the Euler equation (29), in the definition (32) of $v_i$, and in the two surface relations (34) and (35) we should make the replacement

$$\frac{\partial g_v}{\partial u_{ij}} \leftarrow \frac{1}{2}\left(\frac{\partial g_v}{\partial u_{ij}} + \frac{\partial g_v}{\partial u_{ji}}\right) \quad \text{which we denote} \quad \frac{\partial g_v}{\partial u_{(ij)}}. \quad (38)$$

If instead we consider $u_{ij}$ and $u_{ji}$ as the same variable, we can define a functional derivative, which we might denote

$$\left.\frac{\partial g_v}{\partial u_{ij}}\right|_{\text{commute}}. \quad (39)$$

If we would prefer to use this derivative in the expressions, we also have to change the summing range of $i$ and $j$ or to introduce an inconvenient numeric factor, since

$$\left.\frac{\partial g_v}{\partial u_{ij}}\right|_{\text{commute}} = 2\frac{\partial g_v}{\partial u_{(ij)}} \quad \text{if} \quad i\neq j,$$

but

$$= \frac{\partial g_v}{\partial u_{(ij)}} \quad \text{if} \quad i = j. \quad (40)$$

To avoid this factor in the expressions, we keep to the notation introduced in equation (38).

To make the definition of the mixed functional derivate completely clear, let us have an example. If

$$g_v = u_{11}^2 + u_{12}^2 + u_{22}^2 \quad (41)$$

we will get

$$\frac{\partial g_v}{\partial u_{(11)}} = \left.\frac{\partial g_v}{\partial u_{11}}\right|_{\text{commute}} = 2u_{11} \quad (42)$$

but

$$\frac{\partial g_v}{\partial u_{(12)}} = \frac{\partial g_v}{\partial u_{(21)}} = u_{12} \quad (43)$$

while

$$\left.\frac{\partial g_v}{\partial u_{(12)}}\right|_{\text{commute}} = 2u_{12}. \quad (44)$$

To simplify the surface expressions, we can assume that we are studying a regular region of the surface. As a regularity requirement we can demand that we define an environment to the surface region such that the surface can be seen as a member of a set of parallel surfaces. Then this set defines the direction of the surface normal, not only exactly at the surface, but also in an environment of the surface. The normal $\hat{\mathbf{s}}$ in a point slightly outside the surface is then simply defined as the direction of that unique surface normal that emanates from a surface point in the neighbourhood and passes through the point of study. Then the surface normal can be seen as the gradient of a potential function, measuring the distance from the surface, defined so that the rotation of $\hat{\mathbf{s}}$ will vanish. Thus $\nabla\hat{\mathbf{s}}$ in all regular surface points will be symmetric in any coordinate system, with

$$\frac{\partial}{\partial x_j}s_i = \frac{\partial}{\partial x_i}s_j. \quad (45)$$

Moreover, the unit length of the surface normal gives us the relations

$$s_is_i = 1 \quad \text{and} \quad s_i\frac{\partial s_i}{\partial x_j} = 0. \quad (46)$$

By inserting the definition of $v_i$ and carrying out the

differentiation in the first surface relation, using the symmetry of the $\nabla\hat{\mathbf{s}}$ tensor and the normalization of $\hat{s}$, we could rewrite the first surface relation in a form where the dependence on the surface topography is clearer

$$s_i \frac{\partial g_v}{\partial u_i} - s_i \left(2\frac{\partial}{\partial x_j} - s_j s_k \frac{\partial}{\partial x_k}\right)\frac{\partial g_v}{\partial u_{(ij)}} + \frac{\partial g_s}{\partial u} - \left(\frac{\partial}{\partial x_i} - s_i s_j \frac{\partial}{\partial x_j}\right)\frac{\partial g_s}{\partial u_i} - \left(\frac{\partial s_i}{\partial x_j} - s_i s_j \frac{\partial s_k}{\partial x_k}\right)\frac{\partial g_v}{\partial u_{(ij)}} + s_i \frac{\partial s_k}{\partial x_k}\frac{\partial g_s}{\partial u_i} = 0. \quad (47)$$

We see from this equation how derivatives of the surface normal get involved if second-order derivatives are present in $g_v$ or if first order derivatives are present in $g_s$. The last term in equation (47) gives surface derivative terms connected to the surface energy $g_s$, but more such contributions could appear if $g_s$ is dependent on the surface normal.

The second surface relation can be rewritten in the same way as

$$s_i s_j \frac{\partial g_v}{\partial u_{(ij)}} + s_i \frac{\partial g_s}{\partial u_i} = 0. \quad (48)$$

This equation could be used to simplify somewhat the first surface relation

$$s_i \frac{\partial g_v}{\partial u_i} - s_i \left(2\frac{\partial}{\partial x_j} - s_j s_k \frac{\partial}{\partial x_k}\right)\frac{\partial g_v}{\partial u_{(ij)}} + \frac{\partial g_s}{\partial u} - \left(\frac{\partial}{\partial x_i} - s_i s_j \frac{\partial}{\partial x_j}\right)\frac{\partial g_s}{\partial u_i} - \frac{\partial s_i}{\partial x_j}\frac{\partial g_v}{\partial u_{(ij)}} = 0, \quad (49)$$

In the first surface relation, it may look peculiar that we have to take the spatial derivatives of the vector $\mathbf{v}$, which should only be defined at the surface. However, the term $\hat{\mathbf{s}}\cdot(\nabla\times(\hat{\mathbf{s}}\times\mathbf{v}))$, visible in equation (31), will only pick out the variation along the surface, and this property is inherited by the later equations.

We can note that Barbero *et al.* [11], for the case of the nematic director, overlooked the necessity of differentiating between variation of $\eta$ along and normal to the surface, and thus arrived at four boundary conditions for each component of the nematic director, given by their equation (15) and (16), instead of our two equations (48) and (49).

## 6. Relation between this work and the papers by Toupin, Ericksen, and Hinov

The results in this paper up to the volume part of equation (48), had already been derived by Ericksen [18] quite a long time ago, based on work by Toupin [19]. However, neither Toupin nor Ericksen mentions the difficulty in the definition of the second-order mixed functional derivative. Since the equations are suitable as a starting point for future work, it is important that this point is clear.

There also seems to be a sign difference between equation (87) of the Ericksen article and equation (47) of the present paper. If Ericksen's expression for $F^3$ reads

$$F^3 = p_k v_k - b_{mm}\bar{\tau}_{ik}v_i v_k - D_i(\bar{\tau}_{ik}v_k) \quad (50)$$

in his notation, with a minus instead of a plus before the $b_{mm}$ term, it would agree with the expression here. If we proceed as above, and use the second surface relation, the $b_{mm}$ term can be eliminated anyway.

The integral identity

$$\int D_i(f v_j)\, dS = -\int (b_{kk}v_i v_j) f\, dS, \quad (51)$$

between equations (85) and (86) in the same article by Ericksen is correct, and agrees with our equation (31), but does not agree with the 'simplified and corrected version' in equation (22) in Hinov [6]. The integral relation might be checked by using a test function

$$f = xz \quad (52)$$

integrated over the surface of a unit sphere for $i = 1$ and $j = 3$. If this relation is wrong in the Hinov paper, this may have caused secondary errors in his further calculations.

## 7. Third-order variational calculus

If we also include third-order derivatives, we could do this by the same principles, but we of course obtain a larger volume for the mathematical formulae. First the integral $J$ has to be extended to

$$J = \int_V g_v(u, u_i, u_{ij}, u_{ijk}, x_i)\, dV + \int_S g_s(u, u_i, u_{ij}, x_i)\, dS, \quad (53)$$

where $u_{ijk}$ are the third-order derivatives. The third-order volume contributions and the second-order surface contributions to $dJ/d\alpha$ are

$$-\int_V \eta \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}\frac{\partial g_v}{\partial u_{ijk}}\, dV + \int_S \eta s_i \frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}\frac{\partial g_v}{\partial u_{ijk}}\, dS - \int_S \eta_i s_j \frac{\partial}{\partial x_k}\frac{\partial g_v}{\partial u_{ijk}}\, dS + \int_S \eta_{ij}\left(s_k \frac{\partial g_v}{\partial u_{ijk}} + \frac{\partial g_s}{\partial u_{ij}}\right) dS \quad (54)$$

where $\eta_{ij}$ are the second order derivatives of $\eta$ with respect to the coordinate $x_i$. We have still the same kind of trouble in discriminating between variables with different differentiation order, for example, $u_{123}$ and $u_{312}$, as we had for the second-order terms, but now we have more variants to monitor. But the Euler equation for variation of $\eta$ inside the volume does not contain any surprises:

$$\frac{\partial g_v}{\partial u} - \frac{\partial}{\partial x_i}\frac{\partial g_v}{\partial u_i} + \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial g_v}{\partial u_{(ij)}} - \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}\frac{\partial g_v}{\partial u_{(ijk)}} = 0. \quad (55)$$

$$\frac{\partial g_v}{\partial u_{(ijk)}} = \frac{1}{6}\left(\frac{\partial g_v}{\partial u_{ijk}} + \frac{\partial g_v}{\partial u_{ijk}} + \frac{\partial g_v}{\partial u_{jik}} + \frac{\partial g_v}{\partial u_{jki}} + \frac{\partial g_v}{\partial u_{kij}} + \frac{\partial g_v}{\partial u_{kji}}\right). \quad (56)$$

In the derivation of the surface expressions, we derived a theorem (see equation (31)) from Stokes' theorem. We can use that theorem again to rewrite the third integral in equation (54). To take care of the last integral in equation (54), we have to supplement this theorem by one that is a bit more spacious. The idea is to reduce the expression so that they contain only the variation $\eta$ and its derivatives along the surface normal. Let us substitute $\mathbf{V}$ in Stokes' theorem by

$$V_i \leftarrow \varepsilon_{ijk} s_j v_{km} \eta_m \tag{57}$$

in tensor notation, with $\varepsilon_{ijk}$ as the Levi–Civita symbol, and $v_{km}$ as a symmetric tensor. For a regular surface, with symmetric tensor $\nabla\hat{\mathbf{s}}$, we get

$$\begin{aligned}\int_S \eta_{ij} v_{ij}\, dS = \int_S \eta \Big( & \frac{\partial}{\partial x_i}\frac{\partial v_{ij}}{\partial x_j} - 2s_j \frac{\partial v_{ij}}{\partial x_i}\frac{\partial s_k}{\partial x_k} + 2s_i s_j s_k \frac{\partial v_{ij}}{\partial x_k}\frac{\partial s_m}{\partial x_m} \\ & - s_k \frac{\partial v_{ij}}{\partial x_k}\frac{\partial s_j}{\partial x_i} - 2s_j s_k \frac{\partial}{\partial x_i}\frac{\partial v_{ij}}{\partial x_k} + s_i s_j s_k s_m \frac{\partial}{\partial x_k}\frac{\partial v_{ij}}{\partial x_m} \\ & + v_{ij}\frac{\partial s_k}{\partial x_i}\frac{\partial s_j}{\partial x_k} - v_{ij}\frac{\partial s_j}{\partial x_i}\frac{\partial s_k}{\partial x_k} + s_i s_j v_{ij}\frac{\partial s_k}{\partial x_k}\frac{\partial s_m}{\partial x_m} \\ & - s_i s_j v_{ij}\frac{\partial s_m}{\partial x_k}\frac{\partial s_m}{\partial x_k}\Big) dS + \int_S s_k \frac{\partial \eta}{\partial x_k}\Big( - 2s_j \frac{\partial v_{ij}}{\partial x_i} \\ & + 2s_i s_j s_m \frac{\partial v_{ij}}{\partial x_m} - v_{ij}\frac{\partial s_j}{\partial x_i} + 2s_i s_j v_{ij}\frac{\partial s_m}{\partial x_m}\Big) dS \\ & + \int_S \Big( s_k \frac{\partial}{\partial x_k}\Big( s_m \frac{\partial \eta}{\partial x_m}\Big)\Big)(s_i s_j v_{ij})\, dS. \end{aligned} \tag{58}$$

To apply this relation to equation (54), $v_{ij}$ should be chosen as

$$v_{ij} = s_k \frac{\partial g_v}{\partial u_{(ijk)}} + \frac{\partial g_s}{\partial u_{(ij)}}, \tag{59}$$

where we use the symmetric form of the derivatives. We might now apply the theorems derived in order to reduce the surface integrals, so that they will only contain one of factors

$$\eta, \quad s_k \frac{\partial \eta}{\partial x_k}, \quad \text{or} \quad s_k \frac{\partial}{\partial x_k}\Big( s_m \frac{\partial \eta}{\partial x_m}\Big). \tag{60}$$

These quantities could be varied independently, and will give us the first, second and third surface equation. After permutation of the indices in all possible ways, we obtain the third surface equation

$$s_i s_j s_k \frac{\partial g_v}{\partial u_{(ijk)}} + s_i s_j \frac{\partial g_s}{\partial u_{(ij)}} = 0. \tag{61}$$

The second surface equation, after a simplification by the third surface equation, becomes

$$\begin{aligned} & s_i s_j \frac{\partial g_v}{\partial u_{(ij)}} - s_i s_j \Big( 3\frac{\partial}{\partial x_k} - 2s_k s_m \frac{\partial}{\partial x_m}\Big)\frac{\partial g_v}{\partial u_{(ijk)}} \\ & + s_i \frac{\partial g_s}{\partial u_i} - 2s_i \Big( \frac{\partial}{\partial x_j} - s_j s_k \frac{\partial}{\partial x_k}\Big)\frac{\partial g_s}{\partial u_{(ij)}} \\ & - 3s_i \frac{\partial s_j}{\partial x_k}\frac{\partial g_v}{\partial u_{(ijk)}} - \frac{\partial s_i}{\partial x_j}\frac{\partial g_s}{\partial u_{(ij)}} = 0. \end{aligned} \tag{62}$$

The first surface equation, after simplification by both the second and the third surface equations, becomes

$$\begin{aligned} & s_i \frac{\partial g_v}{\partial u_i} - s_i \Big( 2\frac{\partial}{\partial x_j} - s_j s_k \frac{\partial}{\partial x_k}\Big)\frac{\partial g_v}{\partial u_{(ij)}} + s_i \Big( 3\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k} \\ & \quad - 3s_j s_m \frac{\partial}{\partial x_k}\frac{\partial}{\partial x_m} + s_j s_k s_m s_p \frac{\partial}{\partial x_m}\frac{\partial}{\partial x_p}\Big)\frac{\partial g_v}{\partial u_{(ijk)}} \\ & \quad + \frac{\partial g_s}{\partial u} - \Big( \frac{\partial}{\partial x_i} - s_i s_j \frac{\partial}{\partial x_j}\Big)\frac{\partial g_s}{\partial u_i} \\ & \quad + \Big( \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j} - 2s_i s_k \frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k} + s_i s_j s_k s_m \frac{\partial}{\partial x_k}\frac{\partial}{\partial x_m}\Big)\frac{\partial g_s}{\partial u_{(ij)}} \\ & \quad - \frac{\partial s_i}{\partial x_j}\frac{\partial g_v}{\partial u_{(ij)}} + \Big( s_i \frac{\partial s_m}{\partial x_j}\frac{\partial s_k}{\partial x_m} + \frac{\partial^2 s_k}{\partial x_i \partial x_j} + 3\frac{\partial s_j}{\partial x_k}\frac{\partial}{\partial x_i} \\ & \quad - 3s_i s_m \frac{\partial s_j}{\partial x_k}\frac{\partial}{\partial x_m} - s_i s_j s_k \frac{\partial s_p}{\partial x_m}\frac{\partial s_p}{\partial x_m}\Big)\frac{\partial g_v}{\partial u_{(ijk)}} \\ & \quad + \Big( \frac{\partial s_k}{\partial x_j}\frac{\partial s_i}{\partial x_k} - s_k \frac{\partial s_i}{\partial x_j}\frac{\partial}{\partial x_k} - s_i s_j \frac{\partial s_m}{\partial x_k}\frac{\partial s_m}{\partial x_k}\Big)\frac{\partial g_s}{\partial u_{(ij)}} = 0. \end{aligned} \tag{63}$$

## 8. Nematic liquid crystals as an example

To be able to apply these results, we will look at the elasticity theory of nematic liquid crystals, as an example. The Euler equation for nematics, including the now conventional elastic terms, are available in, for instance, de Gennes [20], but the corresponding surface equations are not equally easily found. The energy expression, which we will use, is composed by elastic and electric energy terms,

$$\begin{aligned} g_v = & \tfrac{1}{2}K_{11}(\nabla\cdot\hat{\mathbf{n}})^2 + \tfrac{1}{2}K_{22}(\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}}))^2 \\ & + \tfrac{1}{2}K_{33}(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}}))^2 + K_{22}q_0\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}}) \\ & + K_{13}\nabla\cdot\mathbf{v}_1 - \tfrac{1}{2}(K_{22}+K_{24})\nabla\cdot(\mathbf{v}_1-\mathbf{v}_3) \\ & + K^*(\nabla^2\hat{\mathbf{n}})^2 - \tfrac{1}{2}\mathbf{E}\cdot\boldsymbol{\varepsilon}\mathbf{E} - e_+(\mathbf{v}_1+\mathbf{v}_3)\cdot\mathbf{E} \\ & - e_-(\mathbf{v}_1-\mathbf{v}_3)\cdot\mathbf{E}, \end{aligned} \tag{64}$$

where

$$\mathbf{v}_1 = \hat{\mathbf{n}}(\nabla\cdot\hat{\mathbf{n}}) \quad \text{and} \quad \mathbf{v}_3 = -\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}}). \tag{65}$$

and $K_{11}$, $K_{22}$, and $K_{33}$ are the ordinary Frank elastic constants. $K_{13}$ and $K_{24}$ are the elastic constants for the 'gauge' terms, which could be transformed to surface terms by Gauss' theorem. The $K^*$ term was introduced by Barbero *et al.* [10], to avoid the paradox introduced by the $K_{13}$ term. The influence of the local value of an external electric field $\mathbf{E}$ is determined by the dielectric tensor $\boldsymbol{\varepsilon}$ and the flexoelectric coefficients $e_+$ and $e_-$. The electric field

is assumed to be due to a regulated voltage source. The volume free energy density should, in the case of soft boundary conditions, be supplemented by a surface free energy

$$g_s = -W_{nem}(\mathbf{r})(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}}_s)^2 + E_{s1}(\mathbf{r})\hat{\mathbf{s}}\cdot\mathbf{v}_1 + E_{s2}(\mathbf{r})\hat{\mathbf{s}}\cdot(\mathbf{v}_1-\mathbf{v}_3) - e_s(\mathbf{r})(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})(\hat{\mathbf{n}}\cdot\mathbf{E}) + K_c(\mathbf{r})(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})(\hat{\mathbf{s}}\cdot(\mathbf{E}\times\hat{\mathbf{n}})). \quad (66)$$

Here $\hat{\mathbf{s}}$ is the surface normal, pointing out from the liquid crystal. $W_{nem}(\mathbf{r})$ is the interaction coefficient determining the degree of alignment to some preferred direction $\hat{\mathbf{n}}_s$, attached to the surface. A more general interaction could be modelled by the introduction of two different preferred directions, but the generalization to that case is trivial. $E_{s1}(\mathbf{r})$ and $E_{s2}(\mathbf{r})$ are the two surface constants connected to the elastic gauge-term volume constants $K_{13}$ and $(K_{22}+K_{24})$. The constant $e_s(\mathbf{r})$ determines the strength of the surface flexoelectric effect [21], and the $K_c(\mathbf{r})$ term describes the simplest possible surface electroclinic behaviour, of chiral nature [14]. All the surface constants here could be dependent on the nature of the substrate material, and thus they could vary in size between different points at the surface.

In the derivation of the Euler equation we must take care of the condition

$$\hat{\mathbf{n}}\cdot\hat{\mathbf{n}} = 1, \quad (67)$$

and that is done by subtracting the Lagrange multiplier term

$$\lambda(\mathbf{r})(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}} - 1) \quad (68)$$

from the volume energy density $g_v$, and the corresponding term

$$\lambda_s(\mathbf{r})(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}} - 1) \quad (69)$$

from the surface energy density. We will get one Euler equation for each component $n_i$ of $\hat{\mathbf{n}}$, and these can in traditional way [20] be added to give a vector relation

$$\mathbf{h} = -\lambda(\mathbf{r})\hat{\mathbf{n}}, \quad (70)$$

where $\mathbf{h}$ is called the molecular field. Thus at equilibrium, the $\mathbf{h}$ field should be directed along the director. Any component of $\mathbf{h}$ parallel to the director will only serve the purpose of determining $\lambda(\mathbf{r})$, which is of no interest to us. We are only interested in the two relations we obtain when we specify the two components normal to the director to be zero. Ignoring parts of $\mathbf{h}$ that are always parallel to the director, and assuming the relation $\nabla\times\mathbf{E} = 0$, we could write $\mathbf{h}$ explicitly. The required calculations, component for component, are quite extensive, and the program *Mathematica* has been of great help in the derivations below. The *Mathematica* files may be obtained from Ingolf Dahl.

$$\begin{aligned}\mathbf{h} = {} & K_{11}\nabla(\nabla\cdot\hat{\mathbf{n}}) - K_{22}\{(\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}}))(\nabla\times\hat{\mathbf{n}}) \\ & + \nabla\times(\hat{\mathbf{n}}(\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}})))\} + K_{33}\{(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}})) \\ & \times(\nabla\times\hat{\mathbf{n}}) + \nabla\times(\hat{\mathbf{n}}\times(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}})))\} \\ & - 2K_{22}q_0\nabla\times\hat{\mathbf{n}} - K^*(\nabla\cdot\nabla)^2\hat{\mathbf{n}} + \Delta\varepsilon(\hat{\mathbf{n}}\cdot\mathbf{E})\mathbf{E} \\ & - 2e_+(\hat{\mathbf{n}}\cdot\nabla)\mathbf{E} + e_-(\mathbf{E}(\nabla\cdot\hat{\mathbf{n}}) - \nabla(\mathbf{E}\cdot\hat{\mathbf{n}}) \\ & + \nabla\times(\mathbf{E}\times\hat{\mathbf{n}}) - \mathbf{E}\times(\nabla\times\hat{\mathbf{n}})), \end{aligned} \quad (71)$$

where $\Delta\varepsilon$ is the dielectric anisotropy,

$$\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}. \quad (72)$$

The $e_-$ term here agrees with the flexoelectric term derived by Fan [22]. To simplify the mathematics he assumed that the $e_+$ constant was zero.

We might ask here: what happens if we add a variation of equation (68) to the free energy? We might for instance subtract

$$\lambda_{lap}(\mathbf{r})\nabla^2(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}} - 1) \quad (73)$$

from the volume free energy. The answer is that this term only gives another contribution along the director, and thus does not change the situation for the volume Euler equation, with the molecular field aligned along the director. The situation will change, however, when we change over to look at the corresponding surface equations. The volume Lagrange multiplier $\lambda(\mathbf{r})$ does not contribute to the first or second surface equations, while the term equation (73) contributes to both, fortunately in a manageable way, as we will see below. We have looked for other terms, similar to equation (73), but have not found any contributing in any non-trivial way to the theory.

The surface Lagrange multiplier $\lambda_s(\mathbf{r})$ will enter the first surface equation in the same way as the Lagrange multiplier entered the Euler equation, and thus we can define a surface molecular field $\mathbf{h}_1$, such that

$$\mathbf{h}_1 = -\lambda_s(\mathbf{r})\hat{\mathbf{n}} \quad (74)$$

that at equilibrium should be directed along the director. This molecular field is explicitly

$$\begin{aligned}\mathbf{h}_1 = {} & -K_{11}(\nabla\cdot\hat{\mathbf{n}})\hat{\mathbf{s}} - K_{22}(\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}}))(\hat{\mathbf{n}}\times\hat{\mathbf{s}}) \\ & + K_{33}(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}}))(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}}) - K_{22}q_0(\hat{\mathbf{n}}\times\hat{\mathbf{s}}) \\ & + (e_+ + e_s(\mathbf{r}))\{(\hat{\mathbf{n}}\cdot\mathbf{E})\hat{\mathbf{s}} + (\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\mathbf{E}\} + e_-\{(\hat{\mathbf{n}}\cdot\mathbf{E})\hat{\mathbf{s}} \\ & - (\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})E\} + \tfrac{1}{2}(K_{13} + E_{s1}(\mathbf{r}))\{-2\hat{\mathbf{s}}(\hat{\mathbf{s}}\cdot(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}}) \\ & - \hat{\mathbf{s}}(\nabla\cdot\hat{\mathbf{n}}) + \nabla(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}}) - \nabla\times(\hat{\mathbf{s}}\times\hat{\mathbf{n}}) + \hat{\mathbf{s}}\times(\nabla\times\hat{\mathbf{n}}) \\ & + 2(\hat{\mathbf{n}}\cdot\nabla)\hat{\mathbf{s}}\} - (\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\hat{\mathbf{s}}\times(\hat{\mathbf{s}}\times\nabla E_{s1}(\mathbf{r})) \\ & + \tfrac{1}{2}(K_{22} + K_{24} - 2E_{s2}(\mathbf{r}))\{\hat{\mathbf{s}}(\nabla\cdot\hat{\mathbf{n}}) - \nabla(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})\end{aligned}$$

$$+\nabla\times(\hat{\mathbf{s}}\times\hat{\mathbf{n}})-\hat{\mathbf{s}}\times(\nabla\times\hat{\mathbf{n}})+(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}}\}$$
$$-\hat{\mathbf{n}}\times(\hat{\mathbf{s}}\times\nabla E_{s2}(\mathbf{r}))+2K^*(\hat{\mathbf{s}}\cdot\nabla)\nabla^2\hat{\mathbf{n}}$$
$$-2\lambda_{lap}(\mathbf{r})(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}}+2W_{nem}(\mathbf{r})(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}}_s)\hat{\mathbf{n}}_s$$
$$+K_c(\mathbf{r})\{\hat{\mathbf{n}}\times\mathbf{E}-(\hat{\mathbf{s}}\cdot\mathbf{E})\hat{\mathbf{n}}\times\hat{\mathbf{s}}$$
$$+2(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\mathbf{E}\times\hat{\mathbf{s}}\}, \tag{75}$$

where

$$(\nabla\hat{\mathbf{n}})\hat{\mathbf{s}}=n_{j,i}s_j \tag{76}$$

and

$$(\hat{\mathbf{s}}\cdot(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}})=s_is_kn_{j,k}. \tag{77}$$

The three second surface equations, one for each component of $\hat{\mathbf{n}}$, could also be added to give a vector relation. We thus get a second surface molecular field $\mathbf{h}_2$, defined by

$$\mathbf{h}_2=-(K_{13}+E_{s1}(\mathbf{r}))(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\hat{\mathbf{s}}+\tfrac{1}{2}(K_{22}+K_{24}$$
$$-2E_{s2}(\mathbf{r}))\hat{\mathbf{n}}-2K^*\nabla^2\hat{\mathbf{n}}-2\lambda_{lap}(\mathbf{r})\hat{\mathbf{n}}. \tag{78}$$

This vector field should vanish completely at equilibrium. We could use this relation to solve for $\lambda_{lap}(\mathbf{r})$ at the surface by taking the component along $\hat{\mathbf{n}}$

$$2\lambda_{lap}(\mathbf{r})=-(K_{13}+E_{s1}(\mathbf{r}))(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})^2+\tfrac{1}{2}(K_{22}+K_{24}$$
$$-2E_{s2}(\mathbf{r}))-2K^*\hat{\mathbf{n}}\cdot\nabla^2\hat{\mathbf{n}}. \tag{79}$$

The two components orthogonal to $\hat{\mathbf{n}}$ could be separated out by taking the vector cross product by $\hat{\mathbf{n}}$, and so we could define a third surface molecular field

$$\mathbf{h}_3=\hat{\mathbf{n}}\times\mathbf{h}_2=-(K_{13}+E_{s1}(\mathbf{r}))(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\hat{\mathbf{n}}\times\hat{\mathbf{s}}$$
$$-2K^*\hat{\mathbf{n}}\times\nabla^2\hat{\mathbf{n}}, \tag{80}$$

and thus this vector should also vanish at equilibrium. When the Lagrange multipliers have been eliminated, we thus end with two 2D boundary conditions in the form of two surface molecular fields $\mathbf{h}_1$ and $\mathbf{h}_3$ that both should have vanishing components orthogonal to the director. When $\lambda_{lap}(\mathbf{r})$ is eliminated from $\mathbf{h}_1$, it will be of the form

$$\mathbf{h}_1=-K_{11}(\nabla\cdot\hat{\mathbf{n}})\hat{\mathbf{s}}-K_{22}(\hat{\mathbf{n}}\cdot(\nabla\times\hat{\mathbf{n}}))(\hat{\mathbf{n}}\times\hat{\mathbf{s}})$$
$$+K_{33}(\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}}))(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})-K_{22}q_0(\hat{\mathbf{n}}\times\hat{\mathbf{s}})$$
$$+(e_++e_s(\mathbf{r}))\{(\hat{\mathbf{n}}\cdot\mathbf{E})\hat{\mathbf{s}}+(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\mathbf{E}\}+e_-\{(\hat{\mathbf{n}}\cdot\mathbf{E})\hat{\mathbf{s}}$$
$$-(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})E\}+\tfrac{1}{2}(K_{13}+E_{s1}(\mathbf{r}))\{-2(\hat{\mathbf{s}}\cdot(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}})\hat{\mathbf{s}}$$
$$-\hat{\mathbf{s}}(\nabla\cdot\hat{\mathbf{n}})+\nabla(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})-\nabla\times(\hat{\mathbf{s}}\times\hat{\mathbf{n}})+\hat{\mathbf{s}}\times(\nabla\times\hat{\mathbf{n}})$$
$$+2(\hat{\mathbf{n}}\cdot\nabla)\hat{\mathbf{s}}+2(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})^2(\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}}\}-(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\hat{\mathbf{s}}$$
$$\times(\hat{\mathbf{s}}\times\nabla E_{s1}(\mathbf{r}))+\tfrac{1}{2}(K_{22}+K_{24}$$
$$-2E_{s2}(\mathbf{r}))\{\hat{\mathbf{s}}(\nabla\cdot\hat{\mathbf{n}})-\nabla(\hat{\mathbf{s}}\cdot\hat{\mathbf{n}})+\nabla\times(\hat{\mathbf{s}}\times\hat{\mathbf{n}})$$
$$-\hat{\mathbf{s}}\times(\nabla\times\hat{\mathbf{n}})\}-\hat{\mathbf{n}}\times(\hat{\mathbf{s}}\times\nabla E_{s2}(\mathbf{r}))$$
$$+2K^*\{(\hat{\mathbf{s}}\cdot\nabla)\nabla^2\hat{\mathbf{n}}+((\hat{\mathbf{s}}\cdot\nabla)\hat{\mathbf{n}})\hat{\mathbf{n}}\cdot\nabla^2\hat{\mathbf{n}}\}$$
$$+2W_{nem}(\mathbf{r})(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}}_s)\hat{\mathbf{n}}_s+K_c(\mathbf{r})\{\hat{\mathbf{n}}\times\mathbf{E}$$
$$-(\hat{\mathbf{s}}\cdot\mathbf{E})\hat{\mathbf{n}}\times\hat{\mathbf{s}}+2(\hat{\mathbf{n}}\cdot\hat{\mathbf{s}})\mathbf{E}\times\hat{\mathbf{s}}\}. \tag{81}$$

In this approach we of course miss all non-conservative forces that could be present at a real surface. Our ambition here was to obtain the correct and explicit form of the conservative forces.

## 9. The nematic $K_{13}$ and $K_{24}$ terms

Barbero *et al.* [11], have discussed the elastic constant $K_{13}$. They consider the local free energy density as a function of deformation sources: $g_v=g_v(n_{i,j};\ n_{i,jk};\ldots)$, where $n_i$ are the components of the nematic director and $n_{i,j}$ and $n_{i,jk}$ denote the first- and second-order spatial derivatives. They assume as deformation sources these first- and second-order derivatives $n_{i,j}$ and $n_{i,jk}$. To simplify the arguments by Barbero *et al.*, they ignore the second-order deformation sources in a first-order theory, and in a full second-order theory they have to include such terms as squares of the variables $n_{i,jk}$, normally ignored in the elasticity theory. If these squares of the variables $n_{i,jk}$, normally ignored in the elasticity theory. If these squares are included, the Euler equation becomes of fourth-order. What we have tried to achieve for the nematic case, is to find a correct, reasonably general, and reasonably simple form of these equations. From the definition of the vanishing $\mathbf{h}_3$ field in equation (80), we easily see that a non-zero $K_{13}$ elastic constant *either* requires that the director is parallel or orthogonal to the surface *or* requires a non-zero second-order elastic constant, such as $K^*$. One could define more anisotropic second-order elastic constants to complicate the mathematics further, but we doubt that this is worthwhile.

From the first surface molecular field $\mathbf{h}_1$, we can see that the $K_{13}$ term, as the only term, involves the surface topography via the spatial derivatives of the surface normal.

Both the nematic gauge terms are *nilpotent* according to the terminology of Ericksen [4], and vanish from the Euler equations. Ericksen showed that the nematic $K_{24}$ term is nilpotent, but it is quite easy to verify that the $K_{13}$ term is also nilpotent if second-order deformation sources are allowed in the free energy. It looks rather peculiar that the $K_{24}$ term appears in the second surface molecular field $h_2$, but that is connected with the fact that we have to use the condition

$$\nabla^2(\hat{\mathbf{n}}\cdot\hat{\mathbf{n}}-1)=0 \tag{82}$$

to prove the nilpotency of the $K_{24}$ term. It is then logical that this contribution to the molecular field will be removed by the appropriate choice of the Lagrange multiplier $\lambda_{lap}(\mathbf{r})$.

## 10. Summary

In order to find the correct boundary conditions for liquid crystals, the variational analysis with soft boundaries is discussed. The gauge terms is the volume free energy, which could be rewritten by Gauss' theorem as surface terms, are conceptually difficult to handle. In the one-dimensional case, such terms could cause instabilities near the surfaces. To prevent instabilities, we could use a restricted set of functions when we do the variational analysis, but the restrictions have to be imposed in an unsatisfactory way. A better method is to introduce some higher-order terms in the volume free energy. Then the three-dimensional case becomes a non-trivial, but solvable extension of the one-dimensional case, since in three dimensions we have to differentiate between normal and tangential derivatives at the boundary. The equilibrium equations in a volume element and at the surface are derived, and the result is applied to the case of nematic liquid crystals, where explicit expression are given.

This work has been supported by the Swedish National Board for Industrial and Technical Development (NUTEK) and by the Belgian National Fund for Scientific Research (NFWO).

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