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K_{13} term and effective boundary condition for the nematic director

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We consider the problem of including the divergence term $K_{13}\nabla[\mathbf{n}(\nabla\mathbf{n})]$ in the macroscopic theory of a nematic liquid crystal. The orientation of the bulk director is shown to be determined by the standard Euler-Lagrange equation with an effective boundary condition which assumes a smooth vanishing of the nematic density at the surface and incorporates additional subsurface deformations. This boundary condition implies that, in three dimensions, the K_{13} term does not reduce to an anchoring term. [S1063-651X(98)50707-6]

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I. INTRODUCTION

Divergence terms have been introduced in the free energy (FE) of liquid crystals [1] and the B phase of liquid ^{3}He [2], and, generally speaking, can be important for any system that has surface or topological defects. Over recent years, the problem of divergence terms has been one of the central problems in the physics of liquid crystals [3,4].

The nematic order parameter is a unit vector $\mathbf{n}$ called director. The elastic energy of director deformations is given by the bulk integral $F_2=\int dVf_2$. The energy density f_2 is the sum $f_2=f_F-K_{24}f_{24}+K_{13}f_{13}$ of the terms quadratic in the differentiation operator ∂, i.e., [1],

$$f_F=\frac{K_{11}}{2}(\nabla\mathbf{n})^2+\frac{K_{22}}{2}(\mathbf{n}\cdot\nabla\times\mathbf{n})^2+\frac{K_{33}}{2}(\mathbf{n}\times\nabla\times\mathbf{n})^2,$$

$$f_{24}=\nabla[\mathbf{n}(\nabla\mathbf{n})-(\mathbf{n}\nabla)\mathbf{n}], \tag{1}$$

$$f_{13}=\boldsymbol{\nabla}[\mathbf{n}(\nabla\mathbf{n})].$$

The elastic constants $K_{\alpha\alpha}>0$, whereas K_{13} and K_{24} can have any sign. The last two terms, called the K_{13} and K_{24}, terms are total divergences and their contribution to F_2 can be converted to surface integrals of the form $\int dSKf_S$. If ν is a unit outer normal to the surface, the corresponding surface densities can be written as $f_{24,S}=(\nu\mathbf{n})(\nabla\mathbf{n})-\nu(\mathbf{n}\nabla)\mathbf{n}$ and $f_{13,S}=(\nu\mathbf{n})(\nabla\mathbf{n})$. The divergence terms do not alter the Euler-Lagrange equation (ELE) for the functional F_2 and can influence the director only through boundary conditions on the surface S.

In field theories, the problem of boundary conditions does not arise since all the fields rapidly vanish at infinity, and surface densities are absent. As a result, fields are solely determined by the ELE in which all sources are well-defined three-dimensional densities. In contrast, in the elastic theory of liquid crystals, surface densities and boundary conditions are involved because of the standard idea of an *ideal surface*, which assumes that the densities of all physical quantities vanish steplike at the edge S of the liquid crystal body. Consequently, the theory resorts to surface densities of anchoring W ($\mathbf{n}$-dependent part of the surface tension), surface elastic terms $K_{24}f_{24,S}$ and $K_{13}f_{13,S}$, surface polarity, and so on.

Effects associated with the K_{24} and K_{13} terms have been studied only over the recent decade (see reviews [3,4]). For instance, it was found that the K_{24} term gives rise to mechanisms of spontaneous translational and chiral symmetry breaking of the director field, which are responsible for the variety of patterns in thin nematic films [3]. In contrast, the very presence of the K_{13} term in the FE has been debated. The latest development of this problem has led to the idea that the ideal surface concept is not adequate for the macroscopic theory since surface effects are affected by this unrealistic assumption [5].

For an ideal surface, the theory predicts an intrinsic anchoring W_{id} two to three orders of magnitude larger than experimentally observed values [6–8]. As for the K_{13} term, for several years the problem was that for any nonzero K_{13} the functional F_2 is unbounded below [9,10]. However, this difficulty was recently shown to be actually irrelevant to the case of the ideal surface. Namely, Barbero, Gabbasova, and Osipov [11] showed that F_2 derived for the infinite medium ignores the additional elastic term f_1 induced by translational symmetry breaking at the surface. Later Faetti and Riccardi revealed [12] that, in the sum $f_1-K_{24}f_{24,S}+K_{13}f_{13,S}$, the term $\propto f_{13,S}$ is cancelled out. Recently, this cancellation was proven to be inherent for the elastic expansion if the surface is ideal [5]. Thus, the divergence K_{13} term is absent and the anchoring is too large for a nematic body with an ideal surface.

However, a liquid is known to never end abruptly. Rather, its density ρ vanishes smoothly along with its first derivative over some intermediate layer whose thickness l_S is of order of a few molecular lengths l_M [13]. This l_S layer represents *a nonideal surface* [5]. Consequences of replacing an ideal surface by a nonideal one turn out to be important.

The first consequence is that for a nonideal surface, intrinsic anchoring is much smaller than W_{id}, which naturally explains the mysterious smallness of the experimental values [14,5]. The second one is that if the order parameter at the surface is not constant, then the cancellation of $f_{13,S}$ is violated [5]. In the particular case when variation of the order parameter is smooth and monotonic, the value of K_{13} derived in [5] from a microscopic theory coincides with that found by Vissenberg, Stallinga, and Vertogen [15] in the Landau-de Gennes approach [16,17]. The third consequence is that the K_{13}term is no longer total divergence.

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In simple geometries where $\mathbf{n}$ depends on a single coordinate normal to S, the K_{13} term is known to induce strong subsurface deformations, while its effect on the observable bulk director reduces to anchoring-related phenomena [9,18–20,15]. However, when the tangential director derivatives do not vanish, the K_{13} term does not reduce to anchoring and can produce elastic deformations (Pergamenshchik [10]). Moreover, stripe domains in a thin nematic film can be quantitatively explained only for nonzero K_{13} [21]. The problem is that the boundary condition proposed in [10] to incorporate the K_{13} term and employed in Ref. [21] is shown to yield a nonextremum director, [22–24], and the role of the K_{13} term in three-dimensional effects remains debatable. In this Rapid Communication, a consistent boundary condition providing the missing director extremum is derived for a curved nonideal surface, including the subsurface deformations ignored in [10]. We show that the K_{13} term acts as an elastic term, and the subsurface mode effect reduces to additional anchoring with a tilted easy axis.

II. FE OF A NEMATIC BODY WITH A NONIDEAL SURFACE

The Landau-de Gennes theory can accommodate only smooth variations of the order parameter. It is shown in [5] that to incorporate arbitrary spatial variations both of the density and order parameter the constants $K_{\alpha\beta}$ should be replaced by their actual spatial-dependent equivalents $k_{\alpha\beta}(\mathbf{x})$, which vanish on S and reduce to $K_{\alpha\beta}$ in the bulk (at a distance l_S from S). In particular, since the surface-induced term f_1 renormalizes both K_{24} and K_{13} [12,5], the functions k_{24} and k_{13} can be considered as effective quantities attributed to the sum of f_1 and the bulk K_{24} and K_{13} terms. Since $k_{\alpha\beta}\propto\rho^2$, both $k_{\alpha\beta}$ and its normal derivative vanish at S along with ρ, and boundary conditions do not arise. Since even a thin intermediate layer is now described by ELE, one can think only of some effective ''boundary'' condition obtained by averaging this ELE over the surface layer.

For the ideal surface and an arbitrary geometry, any finite value of K_{13} makes the free energy (1) unbounded below [9,10]. Vanishing of the function k_{13} on S automatically brings a lower boundary to the FE. Indeed, consider the sum $\int dV[k_{11}(\boldsymbol{\nabla}\mathbf{n})^2+k_{13}(\mathbf{x})f_{13}]$. Integrating by parts,

$$\int dVk_{13}(\mathbf{x})\nabla[\mathbf{n}(\nabla\mathbf{n})]=-\int dV(\nabla\mathbf{n})(\mathbf{n}\nabla)k_{13}, \qquad (2)$$

the integrand reduces to $k_{11}(\nabla\mathbf{n})^2-(\nabla\mathbf{n})(\mathbf{n}\nabla)k_{13}\geqslant-1/4k_{11}|\boldsymbol{\nabla}k_{13}|^2$, and thus the FE has a lower boundary. The term $(\mathbf{n}\nabla k_{13})(\nabla\mathbf{n})$ is seen to be a kind of Lifshitz term which produces director deformations in the l_S layer [9]. Since elastic constants vary fast only along the normal ν to S, one has $(\mathbf{n}\nabla k_{13})\simeq(\upsilon\mathbf{n})(\nu\nabla)k_{13}$, and these deformations imply large normal-to-surface director derivatives $\mathbf{n}'$ in the l_S layer [9] (normal derivatives will be labeled by the prime).

The standard density of deformation sources in a nematic phase is of order K/d^2, where d is a macroscopic length (typically $d\sim1\,\mu$m), and results in deformations $\partial n\sim1/d$. The elastic resistance to such deformations is linear (Hook law), i.e., higher order terms f_h are negligible compared to the leading terms (1). However, since $k'\sim K/l_S\gg K/d$, the source $(\mathbf{n}\nabla k_{13})(\nabla\mathbf{n})\gg K/d^2$, the subsurface mode n_S' can be much stronger than $1/d$, and the nonlinear elasticity f_h can come into play in the l_S layer.

The term f_h is an infinite sum of terms with director derivatives of all orders and can be studied only numerically. Such a study was recently performed by Skačej *et al.* [8] for quite a general molecular interaction in the form of a superposition of the Maier-Saupe and dipole-induced–dipole-induced pairwise potentials. It was found that the role of f_h is mainly that the resistance to a source K/l_M^2 is about an order of magnitude larger than the linear one. This can be expressed as $n_S'\sim(K/K_{ef})1/l_M$ where the effective elastic constant $K_{ef}\sim10K$. We emphasize that this assumption does not imply $n_S'\sim1/d$ and is the weakest one necessary for the applicability of elastic approach. Indeed, otherwise $K_{ef}\sim K$, $n_S'\sim1/l_M$, the leading terms and all terms in f_h are of the same order of magnitude, and restricting FE to any finite number of terms is meaningless [10].

This picture of director deformations makes appropriate the separation $\mathbf{n}=\mathbf{m}+\mathbf{s}$ of the director into surface mode $\mathbf{s}$ which vanishes outside the l_S layer and the bulk director $\mathbf{m}$ which alone is observable. The hierarchy of deformation magnitudes can be derived from the above consideration. We have

$$|\partial\mathbf{m}|\sim|\partial_t\mathbf{s}|\sim1/d\ll|\mathbf{s}'|\ll1/l_S\sim k', \qquad (3)$$

where ∂_t stands for tangential-to-S derivatives. This hierarchy means that only the normal derivative of the surface mode is connected to the fast variations across the l_S layer whereas all others are connected to standard macroscopic lengths d. In particular, Eq. (3) in the form $l_S|\mathbf{s}'|\ll1$ implies that in spite of a large derivative s', the total surface variation of $\mathbf{s}$ is negligible compared to $\mathbf{m}$, and hence one can set $\mathbf{n}=\mathbf{m}$ in the expressions that do not contain $\partial\mathbf{n}$.

Consider all possible FE terms. Making use of integration by parts (2), the divergence terms can be reduced to $-f_{13,S}(\nu\nabla)k_{13}-f_{24,S}(\nu\nabla)k_{24}$. Further, to be consistent with our approach, the anchoring W has to be represented by its bulk density $f_a(\mathbf{n},\mathbf{x})\sim W/l_S\sim K/(l_Sd)$, defined as $W=\int d(\nu\mathbf{x})f_a$ [10].

Finally, introducing $\tilde{f}_F=f_F(k)+\frac{1}{2}\lambda\mathbf{n}^2$, where $f_F(k)$ is f_F (1) with the constants $K_{\alpha\alpha}$ replaced by the functions $k_{\alpha\alpha}(\mathbf{x})$ and λ is the Lagrange multiplier, the true FE functional of nematic body with a nonideal surface can be written in the form $F_{true}=\int dVf$, where

$$f=\tilde{f}_F-f_{13,S}(\boldsymbol{\nu}\nabla)k_{13}+f_{24,S}(\nu\nabla)k_{24}+f_a+f_h. \qquad (4)$$

The quantities entering f satisfy the following conditions: on S, $k_{\alpha\beta}(\mathbf{x})=k'_{\alpha\beta}(\mathbf{x})=f_h=f_a=0$; in the bulk outside the l_S layer, $f_a=f_h=0$ and $k_{\alpha\beta}(\mathbf{x})=K_{\alpha\beta}$. By virtue of these conditions, functional F_{true} has a minimum determined by its ELE alone. The magnitude hierarchy (3) enables one to treat this ELE by separating terms of different orders in the small quantities l_S/d and $s'l_S\sim(K/K_{ef})$. Thus, obtained equations of the two leading orders will be considered below.

III. SEPARATION OF THE SURFACE MODE AND OBSERVABLE DIRECTOR

To consider an arbitrary geometry of a liquid crystal we introduce the curvilinear orthogonal coordinate system (x_1,x_2,x_3) with metric tensor g_{ij} [10]. Let the geometrical boundary S coincide with the coordinate surface $x_3=\text{const}=S_0$. Then x_1 and x_2 are the orthogonal coordinates on S and on other surfaces $x_3=$const, and the outer normal to each of these ''parallel'' surfaces is directed along the coordinate line x_3, i.e., $\nu(\mathbf{x})=(0,0,1)$; the differentials of the area of any surface $x_3=$const and of the volume are, respectively, $dS=\sqrt{g_S}dx_1dx_2$ and $dV=\sqrt{g}dx_1dx_2dx_3=dSdx_3\sqrt{g_{33}}$, where $g_S=g_{11}g_{22}$ and $g=g_{11}g_{22}g_{33}$. The intermediate l_S layer is sandwiched between the surfaces $x_3=S_0$ and $x_3=S_0-l_S$.

The general ELE for the total director $\mathbf{n}$ both in the bulk and in the l_S layer is

$$\frac{\partial f}{\partial n_k}-\frac{1}{\sqrt{g}}\partial_i\left(\frac{\partial}{\partial(\partial_i n_k)}-\Lambda_{i,n_k}\right)\sqrt{g}f=0, \tag{5}$$

where the operator

$$\Lambda_{i,n_k}=\partial_j\frac{\partial}{\partial(\partial_i\partial_j n_k)}-\partial_j\partial_{j'}\frac{\partial}{\partial(\partial_i\partial_j\partial_{j'}n_k)}+\cdots$$

incorporates higher order director derivatives in f_h; $i,j,k=1,2,3$ and summation over repeating indices is implied. At the same time, the bulk director $\mathbf{m}$ is determined by Eq. (5) in which all the subsurface sources and $\mathbf{s}$ are set to zero and $k_{\alpha\beta}=K_{\alpha\beta}$, i.e.,

$$\frac{\partial f_F}{\partial m_k}-\frac{1}{\sqrt{g}}\partial_i\frac{\partial(\sqrt{g}f_F)}{\partial(\partial_i m_k)}+\lambda m_k=0. \tag{6}$$

Here we recognize the standard ELE for the bulk director $\mathbf{m}$, which has been the basis of the continuum theory before the problem of divergence terms came into light. Now we will consider the surface mode $\mathbf{s}$ which is coupled to $\mathbf{m}$ in the l_S layer.

In view of Eq. (3), one can expect $\mathbf{s}$ to be determined by the leading terms in Eq. (5). Obviously, these terms contain normal derivatives k' of the elastic ''constants'' and $\mathbf{s}'$ (the prime now stands for the x_3 derivative). For further consideration it is convenient to introduce the polar angle Θ between ν and $\mathbf{n}$ and the azimuthal angle ϕ, i.e., $\mathbf{n}=(\sin\Theta\cos\phi,\sin\Theta\sin\phi,\cos\Theta)$. Direct calculation yields the leading part of the Θ equation in the form

$$k's'+\frac{k''_{13}}{2\sqrt{g_{33}}}\sin 2\Theta+\Lambda_{3,\Theta}f'_h=0, \tag{7}$$

where $k=k_{11}+(k_{33}-k_{11})\cos^2\Theta$. Equation (3) implies $\Theta\simeq\theta$, where θ is the angle between ν and the bulk director $\mathbf{m}$, whereas $\Theta'\simeq s'=(\Theta-\theta)'$. Then Eq. (7) reduces to following relation between the surface mode and surface value of the observable angle θ:

$$s'\simeq-\frac{k'_{13}}{2\sqrt{g_{33}}K_{ef}}\sin 2\theta(x_3=S_0). \tag{8}$$

Here the effective elastic resistance $K_{ef}=K+K_h$ where K_h is the higher order contribution; $K_{ef}\sim 10K_{11}$ as was indicated above. The obtained θ dependence of s is similar to that for the ideal surface [18]; the difference is that, as is seen from Eq. (8), at the nonideal surface s oscillates. At the same time, the K_{13} term does not induce the surface ϕ-mode.

IV. EFFECTIVE BOUNDARY CONDITIONS

ELE (6) does not determine the observable director $\mathbf{m}$ uniquely, since its solution still depends on arbitrary functions (constants in the simplest cases). Now we can derive the effective boundary condition to these equations and make the description in terms of $\mathbf{m}$ closed. To this end we integrate Eq. (5) over $\sqrt{g_{33}}dx_3$ from S_0-l_S to S_0 within the context of hierarchy (3). This yields the desired boundary condition in the form

$$\sqrt{g_{33}}\frac{\partial f_F}{\partial(\partial_3 m_k)}-\frac{\partial f_\perp}{\partial m_k}-\frac{1}{\sqrt{g_S}}\partial_s\left(\sqrt{g_S}\frac{\partial f_\parallel}{\partial(\partial_s m_k)}\right)+\frac{\partial(f_\parallel+W+W_{13})}{\partial m_k}+\lambda_S m_k=0, \tag{9}$$

where the subscript s can take only values 1 and 2. The surface densities $f_\parallel=-K_{24}f_{24,S}+K_{13}f_{13,S}-f_\perp$ and $f_\perp=K_{13}(\nu\mathbf{n})(\nu\nabla)(\nu\mathbf{n})$ introduced in [10] contain the normal-to-surface and tangential director derivatives, respectively; $\mathbf{m}$ and g_{ij} are the surface values of the observable director and metric tensor of S. The quantity W_{13} is additional, a K_{13} connected anchoring term. It is induced by the surface mode $\mathbf{s}$ in the intermediate layer and appears only for nonvanishing K_{13}. Its origin follows from its form, i.e.,

$$\frac{\partial W_{13}}{\partial\theta}=-\frac{1}{\sqrt{g_{33}}}\int_{S_0}^{S_0-l_S}dz k'_{13}s'(\theta)\cos 2\theta. \tag{10}$$

Simple estimate with s' (8) yields

$$W_{13}\approx\frac{K_{11}}{16l_S}\left(\frac{K_{13}}{K_{ef}}\right)^2\cos 4\theta$$

with easy axis $\theta_{13}=\pi/4$. For plausible values $l_S=5l_M$, $K_{13}/K_{ef}\sim 0.1$, its magnitude $\sim K_{11}/(10^4 l_M)$ which is about ten times smaller that the standard anchoring strength. If, however, K_{13}/K_{ef} was about 1 then $W_{13}\sim K_{11}/(10^2 l_M)$, which for smooth surfaces is larger than the observed anchoring (see also [18]). Since usually surfaces have an easy axis at $\theta=0$ or $\theta=\pi/2$, the effect of strong W_{13} orienting the director at $\pi/4$ would have been observed. Thus, experimental data support the inequality $K_{13}/K_{ef}\ll 1$.

In terms of the angles θ and ϕ, Eq. (9) reads

$$\sqrt{g_{33}}\frac{\partial f_F}{\partial(\partial_3\theta)}-\frac{K_{13}}{\sqrt{g_{33}}}(\partial_3\theta)\cos 2\theta-\frac{1}{\sqrt{g_S}}\partial_s\left(\sqrt{g_S}\frac{\partial f_\parallel}{\partial(\partial_s\theta)}\right)+\frac{\partial(W+W_{13}+f_\parallel)}{\partial\theta}=0, \tag{11}$$

$$\sqrt{g_{33}}\frac{\partial f_F}{\partial(\partial_3\phi)}-\frac{1}{\sqrt{g_S}}\partial_s\left(\sqrt{g_S}\frac{\partial f_\parallel}{\partial(\partial_s\phi)}\right)+\frac{\partial(W+f_\parallel)}{\partial\phi}=0. \tag{12}$$

These boundary conditions along with the ''naïve'' bulk Euler-Lagrange equations (6) unambiguously determine the observable director $\mathbf{m(x)}$. Let us briefly describe some basic properties of the general boundary conditions.

If θ, ϕ, and g_{ij} depend on the single coordinate x_3, both the K_{13} and K_{24} terms reduce to additional anchoring terms and do not manifest their elastic nature. This can be briefly illustrated for $g_{ij}=1$ (plane layer). In this case, $f_\parallel\equiv 0$ and the θ equation (11) reduces to $K\theta'+\partial W_{ef}/\partial\theta=0$, where $K=K_{11}+(K_{33}-K_{11})\cos^2\theta$, and

$$W_{ef}(\theta)=\int\frac{d\theta}{\left(1-\frac{K_{13}}{K}\cos 2\,\theta\right)}\frac{\partial(W+W_{13})}{\partial\theta}. \qquad (13)$$

However, if $\mathbf{m}$ depends on two or three coordinates (Cartesian or curvilinear) and the tangential derivatives $\partial_t\mathbf{m}$ do not vanish, then $f_\parallel$ enters the θ-boundary condition (11), and both the K_{13} and K_{24} terms cannot be reduced to anchoring terms. Therefore, in general, the K_{13} term whose contribution to the boundary condition contains the normal director derivative produces elasticity that can be conventionally referred to as surfacelike.

V. THE ROLE OF F_2

For $K_{13}=0$, the observable director satisfies the equations which minimize the naïve functional F_2. In contrast, for $K_{13}\neq 0$, the only meaningful minimization can be performed for F_{true}. Nonetheless, F_2 has certain important implications in finding the minimizers of F_{true}. Namely, if $\mathbf{n}_{eq}$ is a solution of the exact equations (5) while $\mathbf{m}_{eq}$ satisfies the systems (6) and (9), then up to small terms $O(l_S s')$,

$$F_{true}\{\mathbf{n}_{eq}\}\simeq F_2\{\mathbf{m}_{eq}\}+\int dSW(\mathbf{m}_{eq}), \qquad (14)$$

where W is the total anchoring. We see that the meaning of F_2 (1) is that $F_2\{\mathbf{m}_{eq}\}$ is equal to the *equilibrium* FE $F_{true}\{\mathbf{n}_{eq}\}$ plus certain anchoring terms. This enables one to compare FEs for different solutions $\mathbf{m}_{eq}$ of the systems (6) and (9) and select the one with lower FE. At the same time, calculating $F_2\{\mathbf{m}_{neq}\}$ for *arbitrary nonequilibrium* function $\mathbf{m}_{neq}$ has *no meaning*. In particular, comparing values $F_2\{\mathbf{m}_{neq}\}$ for two different $\mathbf{m}_{neq}$ or for $\mathbf{m}_{neq}$ and $\mathbf{m}_{eq}$ is meaningless, since $\mathbf{m}_{neq}$ is not a minimizer of the true FE and is unstable. It is this forbidden procedure that has resulted in the extra contribution $\propto K_{13}$ to the boundary condition of Ref. [10] and, consequently, in a nonextremum director criticized in [22–24]. In contrast, boundary condition (9), which is derived from the ELE, automatically provides the extremity.

In spite of the difference, boundary condition (9) and the one proposed in [10] have important common properties: they do not explicitly depend on higher order terms, and the K_{13} contribution corresponds to elasticity which, in general, cannot be reduced to anchoring. The elastic nature of the K_{13} term was crucial to the conclusion of that fitting the experimental data on stripe domains in a thin nematic film is possible for nonzero K_{13} [21]. Of course, this should be reexamined in the context of the corrected boundary condition (9).

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