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

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To cite this Article Nakagawa, Masahiro(1990) 'On the elastic properties of ferroelectric S_c* liquid crystals', Liquid Crystals, 8: 5, 651 — 675
To link to this Article: DOI: 10.1080/02678299008047378
URL: http://dx.doi.org/10.1080/02678299008047378

LIQUID CRYSTALS, 1990, VOL. 8, NO. 5, 651–675

On the elastic properties of ferroelectric S_C^* liquid crystals†

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(*Received 2 November 1988; accepted 22 March 1990*)

On the basis of the symmetry consideration of the S_C^* phase, a generalized elastic free energy of ferroelectric S_C^* liquid crystals is presented to account for the layer compression or dilatation and the layer distortion as well as **c** director deformation. The present elastic free energy is expressed in terms of three vectors, i.e. the **c** director and the wave vector and the spontaneous polarization vector. According to the present S_C^* model with C_2 symmetry, it is shown that there may exist 17 non-chiral, 4 chiral terms and 14 flexoelectric terms in the S_C^* phase. A few practical applications are also presented to elucidate some interesting elastic properties of S_C^* (or S_C) in simplifed geometries.

1. Introduction

Some elastic free energy expressions of S_C^* liquid crystals have been proposed to account for thier macroscopic properties. First of all Pikin and Indenbom have conducted a phenomenological S_C^* free energy to explain the thermoelastic property near the S_C^*–S_A phase transition point ($T = T_c$) introducing the vector order parameter defined by [1]

$$\mathbf{e} = (\mathbf{n} \times \nu)(\mathbf{n} \cdot \nu) = (n_2 n_3, -n_1 n_3, 0), \tag{1}$$

where $\mathbf{n} = (n_1, n_2, n_3)$ and $\nu = (0, 0, 1)$ denote the **n** director pointing an average direction of the long molecular axes and the layer normal unit vector, respectively. Supposing an undistorted flat layer structure, Pikin and Indenbom expanded the free energy in terms of **e**, $\nabla \mathbf{e}$ and the spontaneous polarization $\mathbf{P}_s$ as follows

$$\begin{aligned} F_R = {} & \frac{K}{2}(\nabla \times \mathbf{e})^2 + \Lambda(\mathbf{e} \cdot \nabla \times \mathbf{e}) \\ & + \tfrac{1}{2}\alpha(T - T_c)\mathbf{e}^2 + \tfrac{1}{4}\beta \mathbf{e}^4 \\ & + \mu_p \mathbf{e} \cdot \mathbf{P}_s + \mu_f (\nabla \times \mathbf{e}) \cdot \mathbf{P}_s + \frac{\mathbf{P}_s^2}{2\chi}, \end{aligned} \tag{2}$$

where K is an elastic constant, Λ is the Lifshitz invariant related to the molecular chirality, α and β are positive constants, μ_p and μ_f are the piezo and flexoelectric constants, respectively, and χ (> 0) is the dielectric susceptibility related to the local dipole–dipole interaction. In the approach of Piken and Indenbom, $\mathbf{P}_s$ is considered as a vector order parameter to be determined by minimizing the local free energy [1].

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From equation (2), one immediately derives $\mathbf{P}_s = -\chi\{\mu_p \mathbf{e} + \mu_f(\nabla \times \mathbf{e})\}$ which implies that the ferroelectricity in the S_C^* phase results from the piezoelectricity and the flexoelectricity. Applying equation (2) to practical problems, some thermoelastic properties of S_C^* have been understood qualitatively. It is noticeable that their free energy may allow us to account for a spatial variation of the tilt angle of $\mathbf{n}$ with respect to $\boldsymbol{v}$ as well as the variation of the azimuthal angle about $\boldsymbol{v}$. In general, however, since any spatially varying tilt angle perpendicular to $\boldsymbol{v}$ has to be accompanied with a certain layer distortion, a layer distortion energy coupled to $\nabla\mathbf{e}$ has to be included in eqation (2) as was discussed recently by Beresnev *et al.* [2].

To treat the layer distortions as well as the director deformation more systematically, the Orsay group deduced an elastic free energy expanded in terms of the first order spatial derivatives of an axial rotation vector $\boldsymbol{\Omega}(\mathbf{r})$ which relates the material deformation to the local frame. For the S_C^* phase with C_2 symmetry, ignoring some surface contributions which are to be equivalently transformed into a surface integration after certain integrations in part [4], the following elastic free energy expression was derived in the local frame of reference x–y–z as [3, 4]

$$F_d = \frac{A_{11}}{2}\left[\frac{\partial\Omega_x}{\partial x}\right]^2 + \frac{A_{21}}{2}\left[\frac{\partial\Omega_x}{\partial y}\right]^2 + \frac{A_{12}}{2}\left[\frac{\partial\Omega_y}{\partial x}\right]^2 + \frac{B_1}{2}\left[\frac{\partial\Omega_z}{\partial x}\right]^2 + \frac{B_2}{2}\left[\frac{\partial\Omega_z}{\partial y}\right]^2 + \frac{B_3}{2}\left[\frac{\partial\Omega_z}{\partial z}\right]^2 + B_{13}\left[\frac{\partial\Omega_z}{\partial x}\right]\left[\frac{\partial\Omega_z}{\partial z}\right] + C_1\left[\frac{\partial\Omega_x}{\partial x}\right]\left[\frac{\partial\Omega_z}{\partial x}\right] + C_2\left[\frac{\partial\Omega_x}{\partial y}\right]\left[\frac{\partial\Omega_z}{\partial y}\right] + D\frac{\partial\Omega_z}{\partial z} + D_1\frac{\partial\Omega_z}{\partial x} + D_2\frac{\partial\Omega_x}{\partial x} + \tfrac{1}{2}\underline{B}\gamma^2. \quad (3)$$

where Ω_x, Ω_y and Ω_z denote the infinitesimal rotation angles about the x, y and z axes, respectively, in the local frame varying from point to point in space [3, 4], γ denotes the relative local layer dilatation along the layer normal, which may exist not only in a distorted layer structure but also in a flat one with $\Omega_x = \Omega_y = 0$ [3, 4]. From this aspect, we have to take account of a non-elastic (or independent of the elastic deformation) part of the free energy, which consists of $\mathbf{a}$, $\mathbf{c}$ and $\mathbf{P}_s$ as well as the elastic part corresponding to $\gamma = \partial u/\partial z$ of Orsay's free energy equation (3) to construct a more generalized free energy; here $u(\mathbf{r})$ is the layer deviation, along the z axis, from the equilibrium position. In the Orsay expression, the z axis is taken as the layer normal, and $\mathbf{c}$ and $\boldsymbol{v}$ vectors lie in the x–z plane in the undistorted equilibrium state. Since a constant interlayer spacing is assumed in equation (3), we have the constraints for $\nabla\boldsymbol{\Omega}$, i.e. $\partial\Omega_x/\partial x + \partial\Omega_y/\partial y = 0$ and $\partial\Omega_x/\partial z = \partial\Omega_y/\partial z = 0$ from $\nabla \times \boldsymbol{v} = 0$ and $\boldsymbol{v} = (\Omega_y, -\Omega_x, 1)$ in the local frame [3, 4]. This local free energy expression was expressed in a laboratory frame by Rapin [5] (for S_C), and later by Dahl and Lagerwall [6] for (S_C^*) using a conventional vector notation. Linear terms of $\nabla\boldsymbol{\Omega}$, or D, D_1 and D_2 terms are pseudo-scalers and are concerned with the chirality, or broken symmetry with respect to the x–z plane of S_C^* [3]. Also the last $\underline{B}\gamma^2/2$ term represents a layer compression energy to be coupled to $\partial\Omega_x/\partial z$ and $\partial\Omega_y/\partial z$, which are set to 0 in equation (3) [3, 4]. In general such a spatial variation of the layer spacing has to be coupled to $\nabla\boldsymbol{\Omega}$. Apparently, however, these couplings are completely ignored in the Orsay expression (3) because the relation between γ and $\boldsymbol{\Omega}$ could not be specified in their framework. In addition, as can be easily seen from the conservation law of the layer numbers free from any layer dislocation [3] $\nabla \times \boldsymbol{v}$ may not vanish in general for compressible smectics. Consequently it is not plasuible to assume that $\partial\Omega_x/\partial z = \partial\Omega_y/\partial z = 0$ and $\partial\Omega_x/\partial x + \partial\Omega_y/\partial y = 0$ for compressible smectics in contrast to the assumption by the Orsay group to derive expression (3) [3]. From these respects the Orsay free energy for compressible S_C or S_C^* has to be reconsidered so as to take account consistently of the layer distortion accompanied with a compression or dilatation as well as the director deformation.

From the point of view mentioned previously we shall put forward a somewhat generalized expression of S_C^* free energy, which includes a non-elastic free energy as well as an elastic energy, to study a layer distortion with a compression or a dilatation, as well as the **c** director deformation. In the present approach, we shall introduce a wave vector **a**, whose magnitude is variable concerned with the layer compression, such that $\nabla \times \mathbf{a} = 0$ to assure the conservation of the layer numbers [3]. In §2 a theoretical formulation of free energy will be presented by means of a conventional tensor formalism to make a scalar (free energy) quantity instead of the symmetry-broken approach by the Orsay Group [3, 4]. Several applications of the present model will be given in §3. Finally §4 will be devoted to a short summary of the present study.

2. Theory

In this section, let us derive an S_C^* free energy, which includes an elastic part as well as a non-elastic part, based on the traditional tensor formalism [13]. The present vector field in the S_C^* phase is specified by three vector, i.e. the wave vector $\mathbf{a}(\mathbf{r})$, the **c** director, and the spontaneous polarization vector $\mathbf{P}_s$. While $\mathbf{c}(\mathbf{r})$ is assumed to be a unit vector, $\mathbf{a}(\mathbf{r})$ is defined as a vector whose magnitude depends on the interlayer spacing as will be mentioned later. In the present model, both $\nabla\mathbf{a}$ and $\nabla\mathbf{c}$ are assumed to be first order quantities, which vanish in the equilibrium uniform state. Since the spontaneous polarization $\mathbf{P}_s$ is induced by the flexoelectric effect related to $\nabla\mathbf{a}$ and $\nabla\mathbf{c}$ as well as by the peizoelectric effect [1], it is also regarded as a first order quantity. According to the conventional terminology of the ferroelectricity of S_C^* liquid crystals [1], let us call the total contribution of the piezo and flexoelectricities as ferroelectricity in the ferroelectric S_C^* liquid crystal. In this sense we have certainly to distinguish the ferroelectricity in liquid crystals from that in solids [1].

Now we shall first note that **c** and $\boldsymbol{\nu}$ are related to the **n** director by the following relation:

$$\mathbf{n} = \boldsymbol{\nu}\cos\Theta + \mathbf{c}\sin\Theta, \tag{4}$$

where Θ is the molecular tilt angle with respect to the layer normal and is assumed to be constant throughout the material in the present model (see figure 1). Here it is convenient to introduce an auxiliary unit vector **p** defined by

$$\mathbf{p} = \boldsymbol{\nu} \times \mathbf{c}. \tag{5}$$

then **c**–**p**–$\boldsymbol{\nu}$ make a right handed orthogonal triad. Since $\boldsymbol{\nu}$ and **c** are assumed to be genuine vectors as well as $\mathbf{P}_s$, **p** is axial. It is noticeable that $\mathbf{P}_s$ is introduced as a vector independent of $\mathbf{p} = \boldsymbol{\nu} \times \mathbf{c}$ in the present model although $\mathbf{P}_s$ is often assumed to be always parallel to **p** in a simple treatment [1]. Now a scaler quantity $\kappa(\mathbf{r})$ is assumed to be related to $\mathbf{a}(\mathbf{r})$ and $\boldsymbol{\nu}(\mathbf{r}) = \mathbf{a}(\mathbf{r})/|\mathbf{a}(\mathbf{r})|$ in the following manner:

$$\mathbf{a}(\mathbf{r}) = \{1 + \kappa(\mathbf{r})\}\boldsymbol{\nu}(\mathbf{r}), \tag{6a}$$

or

$$\kappa(\mathbf{r}) = \mathbf{a}(\mathbf{r})\cdot\boldsymbol{\nu}(\mathbf{r}) - 1, \tag{6b}$$

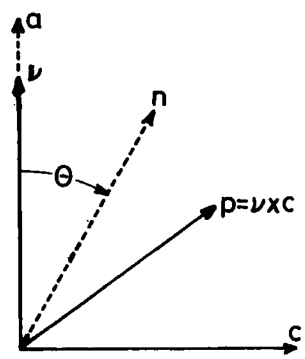

Figure 1. Definition of the vectors in the present model. Here $\mathbf{n}$ is the director along the average direction of the long molecular axes. $\boldsymbol{\nu}$ and $\mathbf{c}$ are the layer normal unit vector and the $\mathbf{c}$ director, respectively. $\mathbf{p}$ is defined by $\boldsymbol{\nu} \times \mathbf{c}$. $\mathbf{a}$ denotes the wave vector.

where $\mathbf{a}(\mathbf{r})$ is assumed to satisfy the curl-less, or solenoidal condition for the dislocation free layers [3] as follows:

$$\nabla \times \mathbf{a}(\mathbf{r}) = 0. \tag{7}$$

It should be borne in mind here that $\nabla \times \boldsymbol{\nu}(\mathbf{r}) \neq 0$ for a compressed or dilated layer structure [3]. At this point, one may find that the Orsay expression (3) cannot be extended straightforwardly to a compressible case as was noted in the previous section. $\kappa - \kappa_0$ is also assumed to be a first order quantity; here κ_0 is the equilibrium value of κ. In a dimensionless form, $|\mathbf{a}(\mathbf{r})|$ can be set to [3]

$$|\mathbf{a}(\mathbf{r})| = d_A/d_C(\mathbf{r}), \tag{8}$$

where d_A and $d_C(\mathbf{r})$ are the interlayer thicknesses in the S_A and S_C phases, respectively. In the equilibrium state, since $d_C(\mathbf{r}) = d_C =$ constant, we have the following equilibrium value of κ, or κ_0:

$$\begin{aligned}\kappa_0 &= d_A/d_C - 1\\ &= \sec\Theta - 1\\ &\simeq \Theta^2/2 \quad (\text{for } |\Theta| \ll 1),\end{aligned} \tag{9}$$

where we replaced d_C by $d_A \cos\Theta$. The scalar, κ is an isotropic scalar, which is independent of the frame of reference and its handedness. In general the non-derivative of κ has to be included in the non-elastic part of the free energy because the free energy depends on the interlayer distance even in the homogeneous flat layers with an equi-interlayer distance as was previously noted by Ribotta and Durand [14].

Now on the basis of the S_C^* symmetry, we shall express the free energy as follows:

$$\begin{aligned}F &= F(\mathbf{a}, \mathbf{c}, \mathbf{p}, \mathbf{P}_s, \nabla\mathbf{a}, \nabla\mathbf{c})\\ &= F_L(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}) + F_c(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{c}) + F_{Lc}(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) + F^*(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c})\\ &\quad + F_f(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \mathbf{P}_s, \nabla\mathbf{a}, \nabla\mathbf{c}) + F_{ne}(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s) + F^*_{ne}(\boldsymbol{\nu}, \mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s),\end{aligned} \tag{10}$$

where F_L is the layer distortion energy density accompanied with a compression or dilatation, F_c is for the $\mathbf{c}$ director deformation, F_{Lc} is a coupling energy between them, F^* and F_f represent the chiral and the ferroelectric free energies, respectively, and F_{ne} and F^*_{ne} are the non-chiral and the chiral contributions of the non-elastic part of the total free energy, respectively. They include the layer compression energy as well as the dipole–dipole interaction between spontaneous polarization. Now, supposing the monoclinic structure with C_2 symmetry in the S_C^* phase, symmetry properties of $F_{non} \equiv F_L + F_c + F_{Lc}$, F^*, F_f, F_{ne} and F_{ne}^* are defined by

$$\begin{aligned} F_{non}(\mathbf{v}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) &= F_{non}(-\mathbf{v}, -\mathbf{c}, \mathbf{p}, -\nabla\mathbf{a}, -\nabla\mathbf{c}) \\ &= F_{non}(\mathbf{v}, \mathbf{c}, -\mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) \\ &= F_{non}(-\mathbf{v}, -\mathbf{c}, -\mathbf{p}, -\nabla\mathbf{a}, -\nabla\mathbf{c}), \end{aligned} \tag{11}$$

$$\begin{aligned} F^*(\mathbf{v}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) &= F^*(-\mathbf{v}, -\mathbf{c}, \mathbf{p}, -\nabla\mathbf{a}, -\nabla\mathbf{c}) \\ &= -F^*(\mathbf{v}, \mathbf{c}, -\mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) \\ &= -F^*(-\mathbf{v}, -\mathbf{c}, -\mathbf{p}, -\nabla\mathbf{a}, -\nabla\mathbf{c}), \end{aligned} \tag{12}$$

$$\begin{aligned} F_f(\mathbf{v}, \mathbf{c}, \mathbf{p}, \mathbf{P}_s, \nabla\mathbf{a}, \nabla\mathbf{c}) &= F_f(-\mathbf{v}, -\mathbf{c}, \mathbf{p}, \mathbf{P}_s, -\nabla\mathbf{a}, -\nabla\mathbf{c}) \\ &= F_f(\mathbf{v}, \mathbf{c}, -\mathbf{p}, \mathbf{P}_s, \nabla\mathbf{a}, \nabla\mathbf{c}) \\ &= -F_f(\mathbf{v}, \mathbf{c}, \mathbf{p}, -\mathbf{P}_s, \nabla\mathbf{a}, \nabla\mathbf{c}), \end{aligned} \tag{13}$$

$$\begin{aligned} F_{ne}(\mathbf{v}, \mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s) &= F_{ne}(-\mathbf{v}, -\mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s) \\ &= F_{ne}(\mathbf{v}, \mathbf{c}, -\mathbf{p}, \kappa, \mathbf{P}_s) \\ &= F_{ne}(\mathbf{v}, \mathbf{c}, \mathbf{p}, \kappa, -\mathbf{P}_s), \end{aligned} \tag{14}$$

$$\begin{aligned} F_{ne}^*(\mathbf{v}, \mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s) &= F_{ne}^*(-\mathbf{v}, -\mathbf{c}, \mathbf{p}, \kappa, \mathbf{P}_s) \\ &= -F_{ne}^*(\mathbf{v}, \mathbf{c}, -\mathbf{p}, \kappa, \mathbf{P}_s) \\ &= -F_{ne}^*(\mathbf{v}, \mathbf{c}, \mathbf{p}, \kappa, -\mathbf{P}_s). \end{aligned} \tag{15}$$

Here $(\mathbf{c}, \mathbf{p}, \mathbf{v}) \to (-\mathbf{c}, \mathbf{p}, -\mathbf{v})$ and $(\mathbf{c}, \mathbf{p}, \mathbf{v}) \to (\mathbf{c}, -\mathbf{p}, \mathbf{v})$ represent π rotation about the $\mathbf{p}$ axis (or the two-fold axis) and the reflection with respect to the $\mathbf{c}$–$\mathbf{v}$ plane, respectively. In equation (13) the ferroelectricity is defined as the non-equivalence between $\mathbf{P}_s$ and $-\mathbf{P}_s$ but not as the non-equivalence between $\mathbf{p}$ and $-\mathbf{p}$, i.e. as non equivalence under reflection. Therefore the chirality of the material is not necessarily related with the ferroelectricity of materials as was first noted by Dahl and Lagerwall [6]. That is, even non-chiral terms may result in the ferroelectricity as will be elucidated later.

Now our aim is to express the free energy F in terms of $\nabla\mathbf{a}$ and $\nabla\mathbf{c}$ as well as $\mathbf{v}$, κ, $\mathbf{c}$, $\mathbf{p}$, and $\mathbf{P}_s$ satisfying the previously mentioned symmetry property, or equations (11)–(15). It should be noted hereafter that κ is related to the wave vector, $\mathbf{a}(\mathbf{r})$, through equation (6 *b*). The present free energy will be constructed to include up to second power of the first-order quantities $\nabla\mathbf{a}$, $\nabla\mathbf{c}$, $\kappa - \kappa_0$, and $\mathbf{P}_s$.

First, let us consider the elastic part of the S_C^* free energy. Noting that $a_{i,j} = a_{j,i}$ (or $\nabla \times \mathbf{a} = 0$) and $\mathbf{c} \cdot \mathbf{c} = 1$, the basic scalar quantities expressed in terms of the first order quantities $a_{i,j}$, $c_{i,j}$, $\mathbf{P}_s$, and $\kappa - \kappa_0$ in terms of the $\mathbf{c}$–$\mathbf{p}$–$\mathbf{v}$ triad are given by

$$\left.\begin{array}{lll} (\mathbf{c} \cdot \nabla\mathbf{a} \cdot \mathbf{c}), & (\mathbf{p} \cdot \nabla\mathbf{a} \cdot \mathbf{c})^*, & (\mathbf{p} \cdot \nabla\mathbf{a} \cdot \mathbf{p}), \\ (\mathbf{c} \cdot \nabla\mathbf{a} \cdot \mathbf{v}), & (\mathbf{p} \cdot \nabla\mathbf{a} \cdot \mathbf{v})^*, & (\mathbf{v} \cdot \nabla\mathbf{a} \cdot \mathbf{v}), \end{array}\right\} \tag{16a}$$

$$(\mathbf{c} \cdot \nabla\mathbf{c} \cdot \mathbf{p})^*, \quad (\mathbf{p} \cdot \nabla\mathbf{c} \cdot \mathbf{p}), \quad (\mathbf{v} \cdot \nabla\mathbf{c} \cdot \mathbf{p})^*, \tag{16b}$$

$$(\mathbf{c} \cdot \mathbf{P}_s), \quad (\mathbf{p} \cdot \mathbf{P}_s)^*, \quad (\mathbf{v} \cdot \mathbf{P}_s), \tag{16c}$$

$$\kappa - \kappa_0 = (\mathbf{a} \cdot \mathbf{v}) - 1 - \kappa_0, \tag{16d}$$

where we used the notations $(\mathbf{X}\cdot\nabla\mathbf{Y}\cdot\mathbf{Z})$ for $X_i Y_{j,i} Z_j$ and $(\mathbf{X}\cdot\mathbf{Y})$ for $X_i Y_i$ for abbreviations [6], and * stands for the chiral (or the pseudo-scaler) quantity which is invariant under π rotation about $\mathbf{p}$, or $(\mathbf{c},\mathbf{p},\mathbf{\nu}) \rightarrow (-\mathbf{c},\mathbf{p},-\mathbf{\nu})$, and changes its sign under reflection operation with respect to the $\mathbf{c}$–$\mathbf{\nu}$ plane, or $(\mathbf{c},\mathbf{p},\mathbf{\nu}) \rightarrow (\mathbf{c},-\mathbf{p},\mathbf{\nu})$. Noting that the product of two chiral terms results in a non-chiral term, from equation (16 *a*) and (16 *b*), we can easily count up the maximum combination numbers up to the second power of the first order derivatives.

Non-chiral

$F_{\mathrm{L}}(\nu,\mathbf{c},\mathbf{p},\nabla\mathbf{a})$: 13 (combinations of (16 *a*)),

$F_{\mathrm{c}}(\nu,\mathbf{c},\mathbf{p},\nabla\mathbf{c})$: 4 (combinations of (16 *b*)),

$F_{\mathrm{Lc}}(\nu,\mathbf{c},\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c})$: 8 (combinations between (16 *a*) and (16 *b*).

Chiral

$F^*(\nu,\mathbf{c},\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c})$: 4 (chosen from (16 *a*) and (16 *b*)).

Ferro

$F_{\mathrm{f}}(\nu,\mathbf{c},\mathbf{p},\mathbf{P}_{\mathrm{s}},\nabla\mathbf{a},\nabla\mathbf{c})$: 14 (combinations between (16 *a*) and (16 *c*), between (16 *b*) and (16 *c*)).

Non-elastic

$$\left.\begin{array}{ll} F_{\mathrm{non}}(\nu,\mathbf{c},\mathbf{p},\mathbf{P}_{\mathrm{s}}) & :5 \\ F^*_{\mathrm{non}}(\nu,\mathbf{c},\mathbf{p},\mathbf{P}_{\mathrm{s}}) & :2 \end{array}\right\} \text{(combinations between (16}c\text{) and (16}d\text{)).}$$

Those combinations are explicitly given as follows:

F_{L}

$$\left.\begin{array}{l}
(1)\ (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2, \quad (2)\ (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2, \quad (3)\ (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2, \\
(4)\ (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)^2, \quad (5)\ (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)^2, \quad (6)\ (\nu\cdot\nabla\mathbf{a}\cdot\nu)^2, \\
(7)\ \underline{(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})} \rightarrow (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 = (2), \\
(8)\ \underline{(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\nu\cdot\nabla\mathbf{a}\cdot\nu)} \rightarrow (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)^2 = (4), \\
(9)\ (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu) \\
(10)\ \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\nu\cdot\nabla\mathbf{a}\cdot\nu)} \rightarrow (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)^2 = (5), \\
(11)\ (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu) \\
(12)\ (\nu\cdot\nabla\mathbf{a}\cdot\nu)\,(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu), \\
(13)\ \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)} \rightarrow (\nu\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) = (11).
\end{array}\right\} \quad (17a)$$

F_{c}

$$\left.\begin{array}{l}
(1)\ (\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2, \quad (2)\ (\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2, \quad (3)\ (\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2, \\
(4)\ (\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\,(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p}).
\end{array}\right\} \quad (17b)$$

$$
\left.\begin{array}{ll}
F_{\mathrm{Lc}} & \\
(1) & (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),\\
(2) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),\\
(3) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),\\
(4) & (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),\\
(5) & \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})} \rightarrow (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (1),\\
(6) & \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p})} \rightarrow (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (4),\\
(7) & \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})} \rightarrow (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (4),\\
(8) & \underline{(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p})} \rightarrow (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (3).
\end{array}\right\} \qquad (17c)
$$

$$
\left.\begin{array}{llll}
F^* & & & \\
(1) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c}), & (2) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu}),\\
(3) & (\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}), & (4) & (\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p}).
\end{array}\right\} \qquad (17d)
$$

Here $\rightarrow$ means the equivalence after partial integration ignoring a surface contribution as is proved in Appendix A. Therefore, the underlind terms are reduced to construct the bulk elastic free energy. Thus 17 non-chiral and 4 chiral terms are found to be allowed in the bulk elastic free energy. Here if we put $\mathbf{a}\cdot\mathbf{a} = (1 + \kappa(\mathbf{r}))^2 = $ constant, or $\kappa(\mathbf{r}) = \kappa_0$, as well as $\mathbf{c}\cdot\mathbf{c} = 1 = $ constant, then the present model can be reduced to the Orsay model expressed by equation (3) as shown in Appendix B.

Next let us consider the ferroelectric part F_f. Up to the second power of $\mathbf{P}_s$ and $\nabla\mathbf{a}$ or $\nabla\mathbf{c}$, referring to equation (13), the possible combinations are given by,

$$
\left.\begin{array}{llll}
F_f & & & \\
(1) & (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\boldsymbol{\nu}\cdot\mathbf{P}_s), & (2) & (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{c}\cdot\mathbf{P}_s),\\
(3) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{p}\cdot\mathbf{P}_s), & (4) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\boldsymbol{\nu}\cdot\mathbf{P}_s),\\
(5) & (\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\mathbf{c}\cdot\mathbf{P}_s), & (6) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\boldsymbol{\nu}\cdot\mathbf{P}_s),\\
(7) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\boldsymbol{\nu})\,(\mathbf{c}\cdot\mathbf{P}_s), & (8) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\,(\mathbf{p}\cdot\mathbf{P}_s),\\
(9) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\boldsymbol{\nu}\cdot\mathbf{P}_s), & (10) & (\boldsymbol{\nu}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\,(\mathbf{c}\cdot\mathbf{P}_s),\\
(11) & (\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\,(\mathbf{p}\cdot\mathbf{P}_s), & (12) & (\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\,(\boldsymbol{\nu}\cdot\mathbf{P}_s),\\
(13) & (\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\,(\mathbf{c}\cdot\mathbf{P}_s), & (14) & (\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\,(\mathbf{p}\cdot\mathbf{P}_s).
\end{array}\right\} \qquad (18)
$$

All other combinations between equations (16c) and (16a) (or (16b)) as $(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})$ $(\mathbf{c}\cdot\mathbf{P}_s)$ are of course ruled out because of C_2 symmetry in the S_C^* phase specified by equation (13). Therefore we have 14 ferroelectric terms related to $\mathbf{P}_s$ and the $\mathbf{c}$ director deformation or the layer distortion.

Finally let us consider the non-elastic parts of the free energy, or F_{ne} (non-chiral) and F_{ne}^* (chiral) which must be closely related to the above derived elastic part. Up to the second order of $\mathbf{P}_s$ and $\kappa - \kappa_0$ from equations (16c) and (16d), referring equation (14) and (15) with C_2 symmetry, we have the following possible combinations:

$$
\left.\begin{array}{l}
F_{ne}\\
(1)\quad (\kappa - \kappa_0)^2 = \{(\mathbf{a}\cdot\boldsymbol{\nu}) - 1 - \kappa_0\}^2\\
(2)\quad (\mathbf{c}\cdot\mathbf{P}_s)^2, \quad (3)\quad (\mathbf{p}\cdot\mathbf{P}_s)^2, \quad (4)\quad (\boldsymbol{\nu}\cdot\mathbf{P}_s)^2, \quad (5)\quad (\mathbf{c}\cdot\mathbf{P}_s)\,(\boldsymbol{\nu}\cdot\mathbf{P}_s).
\end{array}\right\}
\qquad (19)
$$

F^*_{ne}
(1) $(\mathbf{p}\cdot\mathbf{P}_s)$, (2) $(\mathbf{p}\cdot\mathbf{P}_s)(\kappa-\kappa_0).\}$ (20)

Summarizing the results and classifying them again for convenience, we may write down the free energy as follows:

$$F = F_s + F_a + F_c + F_{sc} + F_{as} + F_{ca} + F^* + F_p, \tag{21}$$

where the components are defined by

$$F_s = \frac{A_{11}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + \frac{A_{21}}{2}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + \frac{A_{12}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2, \tag{22}$$

$$F_a = \frac{L_1}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)^2 + \frac{L_2}{2}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)^2 + \frac{L_3}{2}(\nu\cdot\nabla\mathbf{a}\cdot\nu)^2 + L_{13}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)(\nu\cdot\nabla\mathbf{a}\cdot\nu) + \frac{L_0}{2}(\kappa-\kappa_0)^2, \tag{23}$$

$$F_c = \frac{B_1}{2}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + \frac{B_2}{2}(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + \frac{B_3}{2}(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + B_{13}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p}), \tag{24}$$

$$F_{sc} = -C_1(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - C_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}), \tag{25}$$

$$F_{as} = -M_1(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + M_2(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}), \tag{26}$$

$$F_{ca} = N_1(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu)(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + N_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p}), \tag{27}$$

$$F^* = D(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + D_1(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - D_2(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + D_3(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu), \tag{28}$$

$$\begin{aligned} F_p = {} & d^*(\mathbf{P}_2\cdot\mathbf{p})(\nu\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + d_1^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - d_2^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\ & + d_3^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu) - d_1(\mathbf{P}_s\cdot\nu)(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\ & - d_2(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + d_3(\mathbf{P}_s\cdot\nu)(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + d_4(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\ & + d_5(\mathbf{P}_s\cdot\nu)(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + d_6(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + d_7(\mathbf{P}_s\cdot\nu)(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu) \\ & + d_8(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\nu) + d_9(\mathbf{P}_s\cdot\nu)(\nu\cdot\nabla\mathbf{a}\cdot\nu) + d_{10}(\mathbf{P}_s\cdot\mathbf{c})(\nu\cdot\nabla\mathbf{a}\cdot\nu) \\ & + \tfrac{1}{2}d_{01}(\mathbf{c}\cdot\mathbf{P}_s)^2 + \tfrac{1}{2}d_{02}(\mathbf{p}\cdot\mathbf{P}_s)^2 + \tfrac{1}{2}d_{03}(\nu\cdot\mathbf{P}_s)^2 + d_{013}(\mathbf{c}\cdot\mathbf{P}_s)(\nu\cdot\mathbf{P}_s) \\ & + d_0^*(\mathbf{p}\cdot\mathbf{P}_s) + d_{01}^*(\kappa-\kappa_0)(\mathbf{p}\cdot\mathbf{P}_s) \end{aligned} \tag{29}$$

where some symbols and the signs of each term are chosen to make comparison with earlier works [3–6] easy. Here F_s, F_c, and F_{sc} are the same as those derived by the Orsay group if we set $\mathbf{a}\cdot\nu = (1+\kappa(\mathbf{r}))$ = constant, which corresponds to $L_0\to\infty$ in the present model. (See the last term in equation (23).) Apparently $L_0\,(>0)$ in equation (23) suppresses a spontaneous layer compression or dilatation analogous to $\underline{B}$ in equation (3). A similar layer compression energy was introduced by Ribotta and Durand to study the mechanical instabilities from S_A to S_C under an external stress [14]. The C_1 and C_2 terms in F_{sc} are for the coupling between the layer distortion and the $\mathbf{c}$ director deformations, whose effects will be studied in the next section. F_a, F_{as}, and F_{ca} are additional contributions related to the layer compression or dilatation. F^* consists of pseudo-scalars, which are reduced to the result of Dahl and Lagerwall with

$\mathbf{a} \cdot \boldsymbol{\nu} = (1 + \kappa(\mathbf{r})) = \text{constant}$ [6]. The flexoelectric terms d^*, $d_1^* - d_3^*$ are concerned with the chirality of molecules and result in the spontaneous polarization components parallel to $\mathbf{p}$. On the other hand, the other possible terms proportional to $d_1 - d_{10}$ result from the non-chiral contributions and the components parallel to $\boldsymbol{\nu}$ or $\mathbf{c}$. Therefore the latter contributions may exist not only in S_C^* but also in the S_C phase as was previously noted. The terms d_{01}, d_{02}, d_{03} and d_{013} in equation (29) are introduced to express the locally anisotropic dipole–dipole interactions suppressing the modulus of $\mathbf{P}_s$ or $|\mathbf{P}_s|$. The D and D_1 terms represent the inherent twist and bend in the S_C^* phase, respectively. D may result in a spontaneous helicoidal structure along the layer normal as observed in the S_C^* phase. On the other hand, D_1 has a tendencey to cause a bend deformation of the $\mathbf{c}$ director along the layers. Therefore the D_1 term may destablize the uniformly twisted helicoidal structure [3]. It should be noted here that the D_1 term can be converted into a surface term in terms of the vector identity $(\boldsymbol{\nu} \cdot \nabla \times \mathbf{c}) = -\nabla \cdot \mathbf{p}$ if $\mathbf{a} \cdot \mathbf{a} = \text{constant}$ for the incompressible case studied by the Orsay Group [4]. The D_2 term may result in a twisted ribbon-like layer structure as was first pointed out by de Gennes [3]. The D_3 term is related to a layer compression and has not been derived so far. This term may result in a spontaneous layer compression or dilatation along $\mathbf{p}$ and has a tendency to make the uniformly spaced layer structure unstable. On the other hand, the L_2 (>0) term in equation (23) plays a role to suppress such an instability concerned with the layer compression or dilatation. Therefore a competition between them defines an equilibrium layer spacing as will be discussed in the next section. This relation just corresponds to that between the D_2 term in equation (28) and A_{11} in equation (22). In any case, only D is considered to be compatible with a uniformly twisted helicoidal structure.

Finally we shall rewrite equations (22)–(29) in vector form utilizing some vector formulae given in Appendix C.

$$F_a = \frac{A_{11}}{2}(1+\kappa)^2\{(\mathbf{p}\cdot\nabla\times\mathbf{p} - \mathbf{c}\cdot\nabla\times\mathbf{c})/2\}^2 + \frac{A_{21}}{2}(1+\kappa)^2(\mathbf{c}\cdot\nabla\times\mathbf{p})^2 + \frac{A_{12}}{2}(1+\kappa)^2(\mathbf{p}\cdot\nabla\times\mathbf{c})^2, \tag{22a}$$

$$F_a = \frac{L_1}{2}(\mathbf{c}\cdot\nabla\kappa)^2 + \frac{L_2}{2}(\mathbf{p}\cdot\nabla\kappa)^2 + \frac{L_3}{2}(\boldsymbol{\nu}\cdot\nabla\kappa)^2 + L_{13}(\mathbf{c}\cdot\nabla\kappa)(\boldsymbol{\nu}\cdot\nabla\kappa) + \frac{L_0}{2}(\kappa-\kappa_0)^2, \tag{23a}$$

$$F_c = \frac{B_1}{2}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c})^2 + \frac{B_2}{2}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p})^2 + \frac{B_3}{2}\{(\mathbf{c}\cdot\nabla\times\mathbf{c} + \mathbf{p}\cdot\nabla\times\mathbf{p})/2\}^2 - B_{13}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c})(\mathbf{c}\cdot\nabla\times\mathbf{c} + \mathbf{p}\cdot\nabla\times\mathbf{p})/2, \tag{24a}$$

$$F_{sc} = -C_1(1+\kappa)\{(\mathbf{p}\cdot\nabla\times\mathbf{p} - \mathbf{c}\cdot\nabla\times\mathbf{c})/2\}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c}) + C_2(1+\kappa)(\mathbf{c}\cdot\nabla\times\mathbf{p})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}), \tag{25a}$$

$$F_{as} = +M_1(1+\kappa)(\mathbf{c}\cdot\nabla\kappa)(\mathbf{c}\cdot\nabla\times\mathbf{p}) + M_2(1+\kappa)(\mathbf{c}\cdot\nabla\kappa)(\mathbf{p}\cdot\nabla\times\mathbf{c}), \tag{26a}$$

$$F_{ca} = N_1(\mathbf{c}\cdot\nabla\kappa)(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}) - N_2(\mathbf{p}\cdot\nabla\kappa)\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\}, \tag{27a}$$

$$F^* = -D\{(\mathbf{p}\cdot\nabla\times\mathbf{p}+\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}+D_1(\nu\cdot\nabla\times\mathbf{c})$$
$$-D_2(1+\kappa)\{(\mathbf{p}\cdot\nabla\times\mathbf{p}-\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}+D_3(\mathbf{p}\cdot\nabla\kappa), \quad (28\,a)$$

$$F_p = d_0^*(\mathbf{P}_s\cdot\mathbf{p})-d^*(\mathbf{P}_s\cdot\mathbf{p})\{(\mathbf{p}\cdot\nabla\times\mathbf{p}+\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}$$
$$+d_1^*(\mathbf{P}_s\cdot\mathbf{p})(\nu\cdot\nabla\times\mathbf{c})-d_2^*(\mathbf{P}_s\cdot\mathbf{p})(1+\kappa)\{(\mathbf{p}\cdot\nabla\times\mathbf{p}-\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}$$
$$+d_3^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\kappa)+d_1(\mathbf{P}_s\cdot\nu)(1+\kappa)(\mathbf{c}\cdot\nabla\times\mathbf{p})$$
$$+d_2(\mathbf{P}_s\cdot\mathbf{c})(1+\kappa)(\mathbf{c}\cdot\nabla\times\mathbf{p})+d_3(\mathbf{P}_s\cdot\nu)(\nu\cdot\nabla\times\mathbf{p})$$
$$+d_4(\mathbf{P}_s\cdot\mathbf{c})(\nu\cdot\nabla\times\mathbf{p})+d_5(\mathbf{P}_s\cdot\nu)(1+\kappa)(\mathbf{p}\cdot\nabla\times\mathbf{c})$$
$$+d_6(\mathbf{P}_s\cdot\mathbf{c})(1+\kappa)(\mathbf{p}\cdot\nabla\times\mathbf{c})+d_7(\mathbf{P}_s\cdot\nu)(1+\kappa)\{(\mathbf{p}\cdot\nabla\times\mathbf{p})$$
$$-(\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}+d_8(\mathbf{P}_s\cdot\mathbf{c})(1+\kappa)\{\mathbf{p}\cdot\nabla\times\mathbf{p})-(\mathbf{c}\cdot\nabla\times\mathbf{c})/2\}$$
$$+d_9(\mathbf{P}_s\cdot\nu)(\nu\cdot\nabla\kappa)+d_{10}(\mathbf{P}_s\cdot\mathbf{c})(\nu\cdot\nabla\kappa)+\tfrac{1}{2}d_{01}(\mathbf{c}\cdot\mathbf{P}_s)^2+\tfrac{1}{2}d_{02}(\mathbf{p}\cdot\mathbf{P}_s)^2$$
$$+\tfrac{1}{2}d_{03}(\nu\cdot\mathbf{P}_s)^2+d_{013}(\mathbf{c}\cdot\mathbf{P}_s)(\nu\cdot\mathbf{P}_s)+d_{01}^*(\kappa-\kappa_0)(\mathbf{p}\cdot\mathbf{P}_s). \quad (29\,a)$$

3. Applications

In this section we present some applications of the present model. For simplicity, neglecting the anisotropy of the local dipole–dipole interaction, we shall assume hereafter $d_{01} = d_{02} = d_{03} = d_0$, $d_{013} = 0$, $d_{01}^* = 0$. The d_0 and d_0^* just correspond to χ^{-1} and μ_p, respectively, in Equation (2) as proposed by Pikin and Indenbom [1].

3.1. *Layer compression in the surface stabilized geometry*

In this subsection, we shall treat the problem of a spontaneous layer compression which may exist even in a uniformly aligned S_C^* thin sample, or in the surface stabilized sample.

First let us assume that $\nabla\mathbf{c} = \nabla\mathbf{p} = \nabla\nu = 0$ setting the triad $\mathbf{c}$–$\mathbf{p}$–ν to

$$\mathbf{c} = (1,0,0), \quad (30\,a)$$
$$\mathbf{p} = (0,1,0), \quad (30\,b)$$
$$\nu = (0,0,1). \quad (30\,c)$$

In addition $\kappa(\mathbf{r})$ is assumed only to be a function of y. The geometry is schematically depicted in figure 2. In this case, the free energy density F is simply given by

$$F = \frac{L_2}{2}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)^2+\frac{L_0}{2}\{(\mathbf{a}\cdot\nu)-1-\kappa_0\}^2+D_3(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)+d_0^*(\mathbf{P}_s\cdot\mathbf{p})$$
$$+d_3^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)+\tfrac{1}{2}d_0\mathbf{P}_s^2. \quad (31)$$

Now, minimizing equation (31) with respect to $\mathbf{P}_s$, one has

$$\mathbf{P}_s = -d_0^{-1}\{d_0^*+d_3^*(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\nu)\}\mathbf{p}. \quad (32)$$

Substituting this into equation (31) and putting $\kappa = \mathbf{a}\cdot\nu - 1 = \mathbf{a}_z - 1$ and noting $\nabla\mathbf{a}\cdot\nu = \nabla\kappa$ given in equation (C4) of Appendix C, we have

$$F = \frac{L_2}{2}\left[\frac{d\kappa}{dy}\right]^2+\frac{L_0}{2}(\kappa-\kappa_0)^2+\underline{D}_3\frac{d\kappa}{dy}-\frac{(d_0^*)^2}{2d_0}, \quad (33)$$

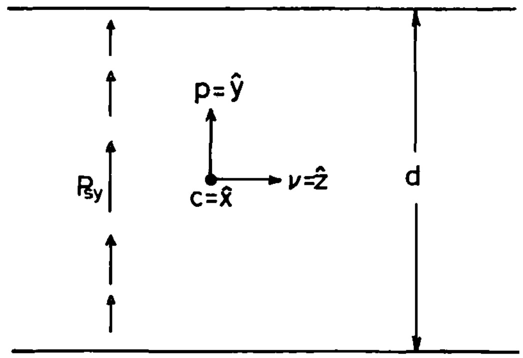

Figure 2. Definition of the coordinates. Here **c**, **p** and **ν** are assumed to be coincident with the x, y, and z axes, respectively. P_{sy} denotes the y component of the spontaneous polarization vector $\mathbf{P}_s$. Here d is the sample thickness.

where $\underline{L}_2$ and $\underline{D}_3$ are defined by

$$\underline{L}_2 = L_2 - \frac{(d_3^*)^2}{d_0}, \tag{34a}$$

$$\underline{D}_3 = D_3 - \frac{d_0^* d_3^*}{d_0}. \tag{34b}$$

Then the Euler–Lagrange equation reads

$$\frac{d^2\kappa}{dy^2} - \frac{1}{\lambda_L^2}(\kappa - \kappa_0) = 0, \tag{35}$$

where $\lambda_L = (\underline{L}_2/L_0)^{1/2}$ is a relaxation length of the layer compression. Therefore a possible solution is simply given by

$$\kappa(y) - \kappa_0 = A\cosh(y/\lambda_L) + B\sinh(y/\lambda_L). \tag{36}$$

Substituting this into equation (33), the averaged free energy density g can be given by

$$\begin{aligned} g &= \frac{1}{d}\int_{-d/2}^{+d/2} dy\, F \\ &= \frac{L_0}{2}(\lambda_L/d)\sinh(d/\lambda_L)A^2 + \frac{L_0}{2}(\lambda_L/d)\sinh(d/\lambda_L)B^2 \\ &\quad + 2\frac{\underline{D}_3}{\lambda_L}(\lambda_L/d)\sinh(d/2\lambda_L)B - \frac{(d_0^*)^2}{2d_0}. \end{aligned} \tag{37}$$

Minimizing equation (37) with respect to the unknown coefficients, A and B, one readily finds

$$A = 0, \tag{38a}$$

$$B = -\frac{\underline{D}_3}{(L_0\underline{L}_2)^{1/2}\cosh(d/2\lambda_L)}. \tag{38b}$$

Hence we have the following solution which represents a spontaneous layer compression or dilatation,

$$\kappa(y) - \kappa_0 = -\frac{\underline{D}_3 \sinh(y/\lambda_L)}{(L_0 \underline{L}_2)^{1/2} \cosh(d/2\lambda_L)}. \tag{39}$$

This result implies that the spontaneous layer compression ($\underline{D}_3 y < 0$) or dilatation ($\underline{D}_3 y > 0$) can be induced by the chiral coefficient $\underline{D}_3$. Finally, from equation (32), the y component of spontaneous polarization $P_{sy}(y)$ is given by

$$P_{sy}(y) = d_0^{-1}\left[-d_0^* + \frac{d_3^* \underline{D}_3}{\underline{L}_2}\frac{\cosh(y/\lambda)}{\cosh(d/2\lambda)}\right]. \tag{40}$$

In the equation (40) the first term is concerned with the piezoelectric effect, which has so far been investigated [1], and the second term d_3^*, stands for the flexoelectric contribution related with the layer compression introduced in this paper. Therefore even if there is no spatial variation of $\mathbf{c}$, $\mathbf{p}$ and $\boldsymbol{\nu}$ in a surface stabilized S_C^* sample, the spontaneous polarization may vary spatially along the sample thickness due to the possible chiral d_3^* and $\underline{D}_3$ terms.

3.2. *Layer distortions in the surface stabilized geometry*

In this subsection, let us discuss the layer distortion in the π twisted state of the surface stabilized S_C^* and S_C samples based on numerical computation. While the significance of the coupling effect between the layer distortion and the director deformation may be inferred as seen from equation (25 *a*), there has been no report so far for such a coupling effect. In this respect, it seems to be worth while to study such a coupling effect on a distorted layer structure based on equations (22)–(29) or equations (22 *a*)–(29 *a*).

To make the present problem mathematically tractable we shall ignore the spatial change of $\kappa(\mathbf{r})$ or the corresponding terms with $\nabla\kappa$ in the present free energy as a first approximation and also assume that $|\mathbf{a}| \simeq 1$ and $|\kappa| \ll 1$ under a soft layer compression or dilatation. In this case the total free energy density F can be given by

$$\begin{aligned}
F = {} & \frac{B_1}{2}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c})^2 + \frac{B_2}{2}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p})^2 + \frac{B_3}{2}\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\}^2 \\
& - B_{13}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c})\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\} + \frac{A_{11}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 \\
& + \frac{A_{21}}{2}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + \frac{A_{12}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 - C_1(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c})) \\
& - C_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p})) + \frac{L_0}{2}\{(\mathbf{a}\cdot\boldsymbol{\nu}) - 1 - \kappa_0\}^2 \\
& - D\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\} + D_1(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c}) - D_2(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\
& + d_0^*(\mathbf{P}_s\cdot\mathbf{p}) - d^*(\mathbf{P}_s\cdot\mathbf{p})\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\} \\
& + d_1^*(\mathbf{P}_s\cdot\mathbf{p})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c}) - d_2^*(\mathbf{P}_s\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) - d_1(\mathbf{P}_s\cdot\boldsymbol{\nu})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\
& - d_2(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + d_3(\mathbf{P}_s\cdot\boldsymbol{\nu})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}) + d_4(\mathbf{P}_s\cdot\mathbf{c})(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}) \\
& + d_5(\mathbf{P}_s\cdot\boldsymbol{\nu})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + d_6(\mathbf{P}_s\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + \tfrac{1}{2}d_0\,\mathbf{P}_s^2.
\end{aligned} \tag{41}$$

First, minimizing F with respect to $\mathbf{P}_s$, one finds

$$\begin{aligned}\mathbf{P}_s = & -d_0^{-1}\,[d_0^*\mathbf{p} - d^*\mathbf{p}\,\{(\mathbf{p}\cdot\nabla\times\mathbf{p} + \mathbf{c}\cdot\nabla\times\mathbf{c})/2\} + d_1^*\mathbf{p}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{c}) \\ & - d_2^*\mathbf{p}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) - d_1\boldsymbol{\nu}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) - d_2\mathbf{c}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + d_3\boldsymbol{\nu}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}) \\ & + d_4\mathbf{c}(\boldsymbol{\nu}\cdot\nabla\times\mathbf{p}) + d_5\boldsymbol{\nu}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + d_6\mathbf{c}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})]. \qquad (42)\end{aligned}$$

It may be noticeable here that there exists three components of $\mathbf{P}_s$ along $\mathbf{p}$, $\mathbf{c}$, and $\boldsymbol{\nu}$ in general. That is, the direction of $\mathbf{P}_s$ is not necessarily coincident with $\mathbf{p} = \boldsymbol{\nu}\times\mathbf{c}$ as has first been pointed out by Dahl and Lagerwall [6]. Then, ignoring the trivial constant $-(d_0^*)^2/(2d_0)$, the free energy density F reads,

$$\begin{aligned}F = & \frac{\underline{B}_1}{2}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + \frac{\underline{B}_2}{2}(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + \frac{\underline{B}_3}{2}(\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 \\ & - \underline{B}_{13}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + \frac{\underline{A}_{11}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + \frac{\underline{A}_{21}}{2}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 \\ & + \frac{\underline{A}_{12}}{2}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 - \underline{C}_1(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - \underline{C}_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\ & + \underline{D}(\boldsymbol{\nu}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - \underline{D}_1(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - \underline{D}_2(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + \frac{L_0}{2}(\kappa-\kappa_0)^2, \qquad (43)\end{aligned}$$

where the coefficients with underbars are defined by

$$\underline{B}_1 = B_1 - \frac{(d_1^*)^2}{d_0}, \qquad (44a)$$

$$\underline{B}_2 = B_2 - \frac{d_3^2 + d_4^2}{d_0}, \qquad (44b)$$

$$\underline{B}_3 = B_3 - \frac{(d^*)^2}{d_0}, \qquad (44c)$$

$$\underline{B}_{13} = B_{13} - \frac{d^* d_1^*}{d_0}, \qquad (44d)$$

$$\underline{A}_{11} = A_{11} - \frac{(d_2^*)^2 - 2(d_1 d_5 + d_2 d_6)}{d_0}, \qquad (44e)$$

$$\underline{A}_{12} = A_{12} - \frac{d_5^2 + d_6^2}{d_0}, \qquad 44f)$$

$$\underline{A}_{21} = A_{21} - \frac{d_1^2 + d_2^2}{d_0}, \qquad (44g)$$

$$\underline{C}_1 = C_1 - \frac{d_1^* d_2^* - d_3 d_5 - d_4 d_6}{d_0}, \qquad (44h)$$

$$\underline{C}_2 = C_2 - \frac{d_1 d_3 + d_2 d_4}{d_0}, \qquad (44i)$$

$$\underline{D} = D - \frac{d_0^* d^*}{d_0}, \qquad (44j)$$

$$\underline{D}_1 = D_1 - \frac{d_0^* d_1^*}{d_0}, \tag{44k}$$

$$\underline{D}_2 = D_2 - \frac{d_0^* d_2^*}{d_0}. \tag{44l}$$

To simplify equation (43) further, we shall put $\underline{B}_1 = \underline{B}_2 = \underline{B}_3 = \underline{B}$, $\underline{B}_{13} = 0$. $(\underline{A}_{11} - \underline{B})/2 = \underline{A}_{12} - \underline{B} = \underline{A}_{21} = \underline{A}$, $\underline{C}_1 = \underline{C}_2 = \underline{C}$, and $\underline{D} + \underline{D}_2 = 0$, noting the following relations, together with the previously noted assumptions $\nabla \mathbf{a} \cdot \nu = \nabla \kappa \simeq 0$ and $|\mathbf{a}| \simeq 1$ or $|\kappa| \ll 1$,

$$\begin{aligned}
&(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p})^2 + (\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p})^2 + (\nu \cdot \nabla \mathbf{c} \cdot \mathbf{p})^2 \\
&= c_{i,j} p_i c_{k,j} p_k \\
&= c_{i,j} c_{i,j} - c_{i,j} \nu_i c_{k,j} \nu_k \\
&= c_{i,j} c_{i,j} - \nu_{i,j} c_i \nu_{k,j} c_k \\
&= c_{i,j} c_{i,j} - c_i a_{i,j} c_k a_{k,j} / |\mathbf{a}|^2 \\
&= (c_{i,i})^2 + (\nabla \times \mathbf{c})^2 + (c_{i,j} c_j - c_{j,j} c_i)_{,i} - c_i a_{i,j} c_k a_{k,j} / (1 + \kappa)^2 \\
&\simeq (c_{i,i})^2 + (\nabla \times \mathbf{c})^2 + (c_{i,j} c_j - c_{j,j} c_i)_{,i} - c_i a_{i,j} c_k a_{k,j},
\end{aligned} \tag{44m}$$

$$\begin{aligned}
c_i a_{i,j} c_k a_{k,j} &= (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\nu \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 \\
&= (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\mathbf{c} \cdot \nabla \kappa)^2 \\
&\simeq (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2,
\end{aligned} \tag{44n}$$

$$\begin{aligned}
&(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + 2(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})^2 \\
&= \{(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c}) + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\}^2 + 2\{(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\} \\
&= \{(\nabla \cdot \mathbf{a}) - (\nu \cdot \nabla \mathbf{a} \cdot \nu)\}^2 + 2\{(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\} \\
&= \{(\nabla \cdot \mathbf{a}) - (\nu \cdot \nabla \kappa)\}^2 + 2\{(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\} \\
&\simeq (\nabla \cdot \mathbf{a})^2 + 2\{(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2 - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\},
\end{aligned} \tag{44o}$$

$$\begin{aligned}
&(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{p})(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) \\
&= \{(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c}) + (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p})\}(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) \\
&\quad + \{(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{p})(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p})\} \\
&= \{(\nabla \cdot \mathbf{a}) - (\nu \cdot \nabla \mathbf{a} \cdot \nu)\}\{(\nabla \cdot \mathbf{c}) - (\nu \cdot \nabla \mathbf{c} \cdot \nu)\} \\
&\quad + \{(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{p})(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p})\} \\
&= \{(\nabla \cdot \mathbf{a}) - (\nu \cdot \nabla \kappa)\}\{(\nabla \cdot \mathbf{c}) + (\nu \cdot \nabla \nu \cdot \mathbf{c})\} \\
&\quad + \{(\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{p})(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p})\} \\
&\simeq (\nabla \cdot \mathbf{a})(\nabla \cdot \mathbf{c}) \\
&\quad + \{(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) - (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c})(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p})\},
\end{aligned} \tag{44p}$$

$$(\nu \cdot \nabla \mathbf{c} \cdot \mathbf{p}) = (\mathbf{p} \cdot \nabla \mathbf{c} \cdot \nu) - (\mathbf{c} \cdot \nabla \times \mathbf{c}). \tag{44q}$$

$$(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\nu) + (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) = (\mathbf{p}\cdot\nabla\mathbf{c}\cdot\nu) - (\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{a})$$
$$= -(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\nu)\kappa$$
$$\simeq 0. \tag{44r}$$

Then, ignoring such surface contributions as $(c_{i,j}c_j - c_{j,j}c_i)_{,i}$ and $(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 - (\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})$, etc., one finally has

$$F = \frac{\underline{A}}{2}(\nabla\cdot\mathbf{a})^2 + \frac{\underline{B}}{2}\{(\nabla\cdot\mathbf{c})^2 + (\nabla\times\mathbf{c})^2\} - \underline{D}\mathbf{c}\cdot\nabla\times\mathbf{c} + \underline{D}_1\nu\cdot\nabla\times\mathbf{c}$$
$$- \underline{C}(\nabla\cdot\mathbf{a})(\nabla\cdot\mathbf{c}) + \frac{L_0}{2}(\kappa-\kappa_0)^2. \tag{45}$$

Here it should be borne in mind again that the constraints for $\mathbf{a}(\mathbf{r})$ and $\mathbf{c}(\mathbf{r})$ are given by $\nabla\times\mathbf{a}(\mathbf{r}) = 0$ and $\mathbf{a}(\mathbf{r})\cdot\mathbf{c}(\mathbf{r}) = 0$, and $\mathbf{c}(\mathbf{r})\cdot\mathbf{c}(\mathbf{r}) = 1$. In equation (45) the $\underline{C}$ term stands for the coupling effect between splay deformations of $\mathbf{a}(\mathbf{r})$ and $\mathbf{c}(\mathbf{r})$. Therefore, although the spontaneous splay of the layer structure due to the term proportional to $(\nabla\cdot\mathbf{a})$ cannot be allowed in the S_C^* or the S_C phase, a splayed layer structure may be caused by a possible splay of $\mathbf{c}(\mathbf{r})$ or by the existence of $(\nabla\cdot\mathbf{c})$. In fact such an induced splay layer structure will be shown to be stabilized by the coupling effect between $(\nabla\cdot\mathbf{a})$ and $(\nabla\cdot\mathbf{c})$.

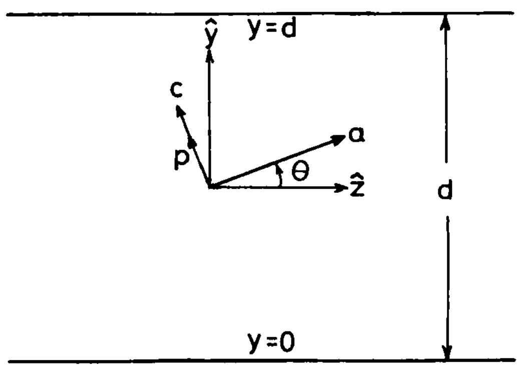

Figure 3. Definition of the coordinates. Here the wave vector $\mathbf{a}$ is assumed to be in the y–z plane. d is the sample thickness.

As a simple example we shall restrict ourselves to a one dimensional geometry as shown in figure 3, where d denotes the samples thickness. Here we assume that $\mathbf{a}(y)$ is in the y–z plane, and that the y axis is normal to the two bounding plates. Normalizing the free energy density F and denoting y/d as τ, we find

$$\underline{F} = Fd^2/B$$
$$= \frac{A^*}{2}\left[\frac{da_y}{d\tau}\right]^2 + \frac{1}{2}\left[\left[\frac{dc_x}{d\tau}\right]^2 + \left[\frac{dc_y}{d\tau}\right]^2 + \left[\frac{dc_z}{d\tau}\right]^2\right] - C^*\left[\frac{da_y}{d\tau}\right]\left[\frac{dc_y}{d\tau}\right]$$
$$+ D^*\left[c_z\left[\frac{dc_x}{d\tau}\right] - c_x\left[\frac{dc_z}{d\tau}\right]\right] - D_1^*\frac{1}{(1+a_y^2)^{1/2}}\left[\frac{dc_x}{d\tau}\right] + \frac{L_0^*}{2}(\kappa-\kappa_0)^2, \tag{46}$$

where $A^* = \underline{A}/\underline{B}$, $C^* = \underline{C}/\underline{B}$, $D^* = \underline{D}d/\underline{B}$, $D_1^* = \underline{D}_1 d/\underline{B}$, and $L_0^* = L_0 d^2/\underline{B}$, and we put a_z = constant = 1 because of the constraint $\nabla \times \mathbf{a}(\tau) = 0$. Therefore the tilt angle $\theta(\tau)$ of the wave vector $\mathbf{a}(\tau)$ measured from the z axis can be given by

$$\theta(\tau) = \tan^{-1}\{a_y(\tau)\}. \tag{47}$$

In addition $\kappa(\tau)$ can be determined by

$$\kappa(\tau) = \{1 + a_y(\tau)^2\}^{1/2} - 1. \tag{48}$$

Minimizing $\underline{F}$ of equation (46) under the following constraints

$$\mathbf{a}(\tau) \cdot \mathbf{c}(\tau) = 0, \tag{49 a}$$

and

$$\mathbf{c}(\tau) \cdot \mathbf{c}(\tau) = 1, \tag{49 b}$$

we find the following set of ordinary differential equations:

$$\begin{aligned} A^* \frac{d^2 a_y}{d\tau^2} - C^* \frac{d^2 c_y}{d\tau^2} & \\ - D_1^* \frac{a_y}{(1 + a_y^2)^{3/2}} \frac{dc_x}{d\tau} & \\ - L_0^* \{(1 + a_y^2)^{1/2} - 1 - \kappa_0\} \frac{a_y}{(1 + a_y^2)^{1/2}} & = \omega c_y, \end{aligned} \tag{50 a}$$

$$\frac{d^2 c_x}{d\tau^2} + 2D^* \frac{dc_z}{d\tau} + D_1^* \frac{a_y}{(1 + a_y^2)^{3/2}} \frac{da_y}{d\tau} = \mu c_x, \tag{50 b}$$

$$\frac{d^2 c_y}{d\tau^2} - C^* \frac{d^2 a_y}{d\tau^2} = \mu c_y + \omega a_y, \tag{50 c}$$

$$\frac{d^2 c_z}{d\tau^2} - 2D^* \frac{dc_x}{d\tau} = \mu c_z + \omega, \tag{50 d}$$

where $\mu(\tau)$ and $\omega(\tau)$ are the Lagrange multipliers to be determined simultaneously so as to satisfy the constraints (49 a) and (49 b). Assuming the π twist state between two plates, the boundary values of $\mathbf{a}(\tau)$ and $\mathbf{c}(\tau)$ may be assumed simply as follows

$$\left.\begin{aligned} \mathbf{a}(0) &= (0,0,1), \\ \mathbf{a}(1) &= (0,0,1), \end{aligned}\right\} \tag{51 a}$$

and

$$\left.\begin{aligned} \mathbf{c}(0) &= (\ \ 1,0,0), \\ \mathbf{c}(1) &= (-1,0,0), \end{aligned}\right\} \tag{51 b}$$

respectively. (see figure 3.) Then the total free energy $\underline{F}_{tot}$ per unit area can be given by

$$\underline{F}_{tot} = \int_0^1 d\tau \underline{F}. \tag{52}$$

It may be noticeable here that $\underline{F}_{tot}$ is equal to $\pi^2/2$ for the uniformly twisted state without any layer distortion, or for $\mathbf{a}(\tau) = (0,0,1)$ and $\mathbf{c}(\tau) = (\cos(\pi\tau), \sin(\pi\tau), 0)$.

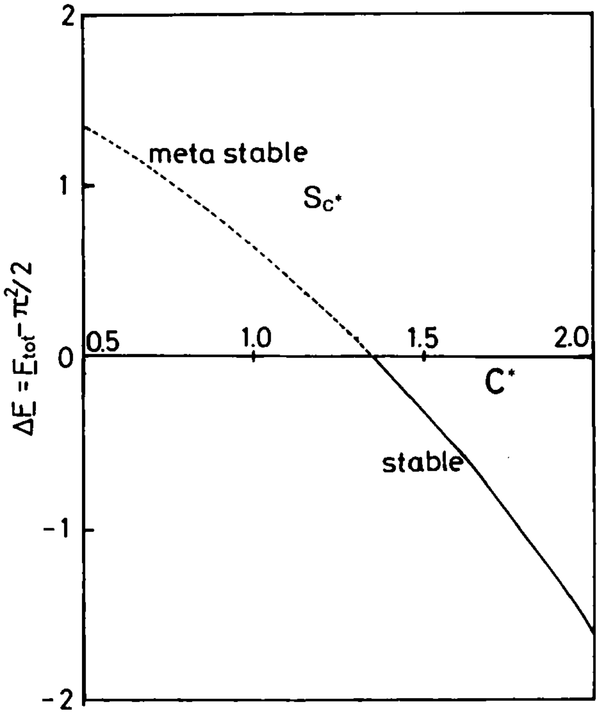

Figure 4. Dependence of the free energy on the coupling constant C^* between the splay deformations of **a** and **c** for S_C^*. Here the splayed layer structure is metastable for $C^* < 1{\cdot}35$, and stable for $C^* > 1{\cdot}35$.

We solved numerically the above non-linear ordinary differential equations (50 *a*)–(50 *d*) by means of the conventional finite difference method combined with an iteration scheme. As an example, let us show here the numerical results for $A^* = 4$ and $L_0^* = 10$, $\kappa_0 = 0$. First we shall present the result for S_C^* assuming that $D^* = -1$ and $D_1 = 1$. Varying the coupling constant C^*, we found that the splayed layer structure for $C^* > 1{\cdot}35$ corresponds to the stable state rather than the uniform π twist state without any layer distortion as shown in figure 4. Therein the splay layer structure for $C^* < 1{\cdot}35$ corresponds to the meta-stable state which is energetically unfavourable relative to the uniform π twist. The corresponding layer tilt angle $\theta(\tau)$ and the layer compression $\kappa(\tau)$, determined by equation (47) and (48), respectively, are shown in figure 5. From these results the coupling between $\mathbf{a}(\mathbf{r})$ and $\mathbf{c}(\mathbf{r})$ is found to play a significant role for the layer structure in the surface stabilized geometry of S_C^*. Next, supposing an S_C phase or putting $D^* = D_1^* = 0$ with the previously chosen parameters, we solved again equations (50 *a*)–(50 *d*) in the same manner for various C^* values. Remarkably, in this case, it is found that the distorted layer structure can *always* be more stable for any $C^* \neq 0$ than the π-twist state without any layer distortion as seen in figures 6 and 7. Therefore it may be considered that this kind of layer distortion can be observed not only in S_C^* but also more often in the S_C phase.

4. Discussions and conclusions

In this paper we have derived an extended elastic energy of ferroelectric S_C^* liquid crystals introducing the variable wave vector related with the interlayer spacing. From

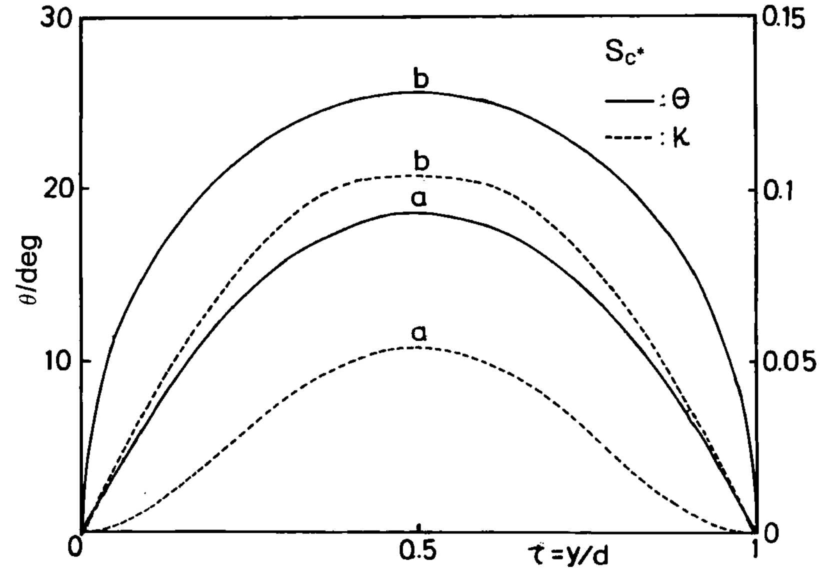

Figure 5. Dependences of $\theta(\tau)$ (solid curves) and $\kappa(\tau)$ (dashed curves) on τ for S_C^*. Here curves a and b are for $C^* = 1$ and $C^* = 2$, respectively.

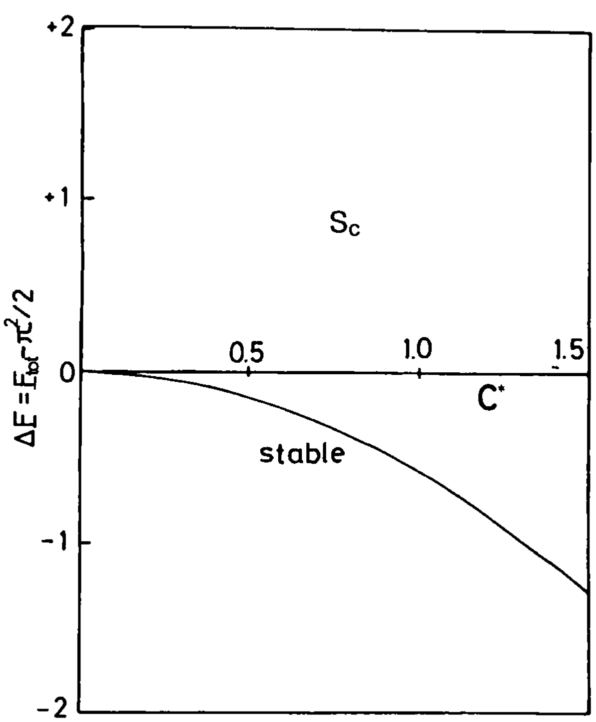

Figure 6. Dependence of the free energy on the coupling constant C^* between the splay deformations of **a** and **c** for S_C. Here the splayed layer structure is stable for any C^*.

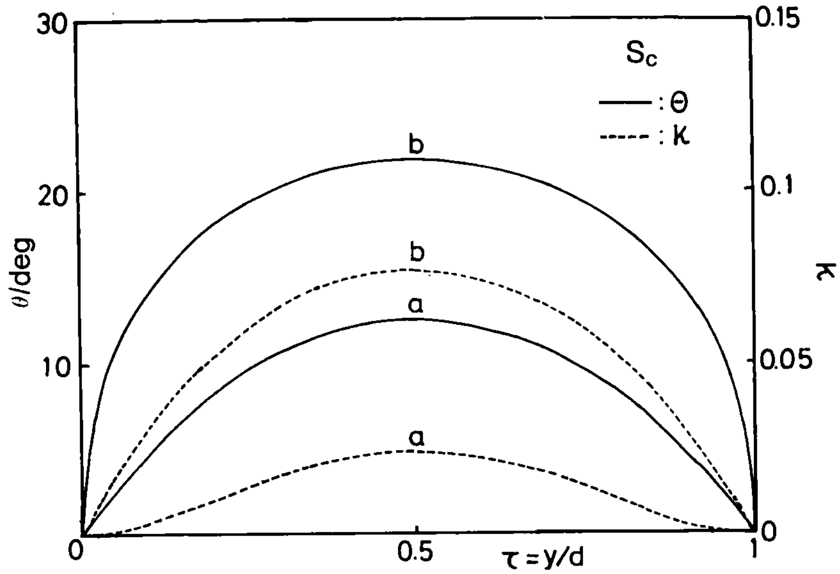

Figure 7. Dependence of $\theta(\tau)$ (solid curves) and $\kappa(\tau)$ (dashed curves) on τ for S_C. Here curves *a* and *b* are for $C^* = 1$ and $C^* = 2$, respectively.

the symmetry argument of the S_C^* phase, we have found 17 non-chiral and 4 chiral elastic coefficients in the bulk free energy related to the layer compression or dilatation and the layer distortion as well as the **c** director deformations. Furthermore we have derived 14 flexoelectric constants. If we suppose an incompressible S_C^* phase or $\mathbf{a} \cdot \mathbf{a} = (1 + \kappa(\mathbf{r}))^2 =$ constant as well as $\mathbf{c} \cdot \mathbf{c} = 1$ in the present model, then we have 9 non-chiral terms and 3 chiral terms consistent with the Orsay expression (3). This result can also be easily derived assuming $\nabla \times$(*one of the directors in the orthogonal triad*) $= 0$ in the result derived by Liu for the monoclinic biaxial nematics [12]. (See Appendix D.) As examples of applications some simplified problems were discussed under the surface stabilized geometry. First we have studied the layer compression as can be observed in a uniformly aligned S_C^* film sample. In this case the spontaneous polarization may vary along the sample thickness direction without any layer distortion and any **c** director deformation. In practice such a layer compression may be accompanied with the line dislocations parallel to the bounding plates and perpendicular to the layer normal. Next the energetical stability of the distorted layer structure in the π twist state in the S_C and S_C^* phases was numerically investigated. From the present results, an appropriate strength of the coupling between the splay deformations of the wave vector **a** and the **c** director was found to make a splayed layer structure with the π twist of the **c** director in the surface stabilized S_C^* sample stable. In addition, it was concluded that this kind of distorted layer structure may be observed not only in an S_C^* phase but also in an S_C phase consistent with the experimental observation [10]. Remarkably it was also shown that such a distorted layer structure in an S_C phase is always more stabilized than the π twist state without any layer distortion. An application of the present theory to the chevron structure observed in the surface stabilized geometry [7–11, 15, 16] will be reported in a later paper [11, 18, 19].

The author would like to thank Professor F. M. Leslie at the University of Strathclyde for the valuable discussions on the continuum theory of liquid crystals. He is also grateful to Professor T. Akahane at the Nagaoka University of Technology in Japan for encouragement through the present work. The sincere thanks are due to the present referee of the journal for the critical reading of the manuscript and the valuable comments. Thanks are also due for finanical support from the Ministry of Education of Japan during the author's stay in Glasgow as a Visiting Researcher.

Appendix A

As an example let us prove the following relation with the assumptions of the partial derivatives such that $a_{k,ij} = a_{k,ji}$ and $c_{k,ij} = c_{k,ji}$.

$$\int_V d\mathbf{r} v_i a_{j,i} p_j c_k c_{l,k} p_l = \int_V d\mathbf{r} c_i a_{j,i} p_j v_k c_{l,k} p_l + I_s, \tag{A 1}$$

where I_s is a surface integral which will be derived later. To prove equation (A 1), let us first note the following identity.

$$\begin{aligned} v_i c_k - c_i v_k &= \varepsilon_{plm} v_l c_m \varepsilon_{pik} \\ &= p_p \varepsilon_{pik}. \end{aligned} \tag{A 2}$$

Then, from equation (A 1) and (A 2), I_s can be expressed as

$$I_s = \int_V d\mathbf{r} p_p \varepsilon_{pik} a_{j,i} p_j c_{l,k} p_l. \tag{A 3}$$

Now let us divide the whole volume V, covered by the surface S, into N cells (e.g. simple cubes or more generally polygons) with volume $\Delta V^{(\alpha)}$ ($\alpha = 1, 2, \ldots, N$). Then (A 3) becomes

$$I_s = \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dx_1 dx_2 dx_3 p_p \varepsilon_{pik} a_{j,i} p_j c_{l,k} p_l; \tag{A 4}$$

here the symbol $\Delta V^{(\alpha)}$ denotes the volume integral in the αth cell. Since one can choose the coordinate axes, $X_1^{(\alpha)} - X_2^{(\alpha)}$–$X_3^{(\alpha)}$ to the vector traid $\mathbf{c}$–$\mathbf{p}$–$\mathbf{v}$ at any point in each cell, one can put $\mathbf{c} = (1,0,0)$, $\mathbf{p} = (0,1,0)$ and $\mathbf{v} = (0,0,1)$ at a point in each cell. In other words, there exists a proper rotation matrix $Q_{ij}^{(\alpha)}$ such that $Q_{ij}^{(\alpha)} c_j = \delta_{i1}$, $Q_{ij}^{(\alpha)} p_j = \delta_{i2}$ and $Q_{ij}^{(\alpha)} a_j = \delta_{i3}$ at a point in each cell. Since we take such a limitation as $\Delta V^{(\alpha)} \to 0$, $Q_{ij}^{(\alpha)}$ can be treated as a constant matrix in each cell with the volume $\Delta V^{(\alpha)}$ when we carry out the integration (A 4) over $\Delta V^{(\alpha)}$ in each cell. It should be noted, however, that a gradient of vectors such as $a_{i,j}$ or $c_{i,j}$ may not vanish in the cell and should be transformed as tensors in the following manner,

$$\left.\begin{aligned} a^*_{i,j} &= Q_{ik}^{(\alpha)} Q_{jl}^{(\alpha)} a_{k,l}, \\ c^*_{i,j} &= Q_{ik}^{(\alpha)} Q_{jl}^{(\alpha)} c_{k,l}. \end{aligned}\right\} \tag{A 5}$$

Then equation (A 4) reads

$$\begin{aligned} I_s &= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{\delta_{w2} Q_{wp}^{(\alpha)} \varepsilon_{pik} Q_{rj}^{(\alpha)} Q_{qi}^{(\alpha)} a^*_{r,q} \delta_{m2} Q_{mj}^{(\alpha)} Q_{nl}^{(\alpha)} Q_{uk}^{(\alpha)} c^*_{n,u} \delta_{V2} Q_{Vl}^{(\alpha)}\} \\ &= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{\delta_{w2} Q_{wp}^{(\alpha)} \varepsilon_{pik} \delta_{rm} Q_{qi}^{(\alpha)} a^*_{r,q} \delta_{m2} \delta_{nV} Q_{uk}^{(\alpha)} c^*_{n,u} \delta_{V2}\} \end{aligned}$$

$$= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{\delta_{w2} Q_{wp}^{(\alpha)} \varepsilon_{pik} \delta_{r2} Q_{qi}^{(\alpha)} a^*_{r,q} \delta_{n2} Q_{uk}^{(\alpha)} c^*_{n,u}\}$$

$$= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{Q_{2p}^{(\alpha)} \varepsilon_{pik} Q_{qi}^{(\alpha)} a^*_{2,q} Q_{uk}^{(\alpha)} c^*_{2,u}\}. \quad (A\,6)$$

Then, from equation (A 2), we have

$$Q_{3i}^{(\alpha)} Q_{1k}^{(\alpha)} - Q_{1i}^{(\alpha)} Q_{3k}^{(\alpha)} = Q_{2p}^{(\alpha)} \varepsilon_{pik}. \quad (A\,7)$$

Thus

$$I_s = \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{Q_{3i}^{(\alpha)} Q_{1k}^{(\alpha)} - Q_{1i}^{(\alpha)} Q_{3k}^{(\alpha)}\} Q_{qi}^{(\alpha)} a^*_{2,q} Q_{uk}^{(\alpha)} c^*_{2,u}$$

$$= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} (a^*_{2,3} c^*_{2,1} - a^*_{2,1} c^*_{2,3})$$

$$= \lim_{N\to\infty} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{(a^*_2 c^*_{2,1})_{,3} - a^*_2 c^*_{2,13} - (a^*_2 c^*_{2,3})_{,1} + a^*_2 c^*_{2,31}\}$$

$$= \lim_{N\to 1} \sum_{\alpha=1}^{N} \int_{\Delta V^{(\alpha)}} dX_1^{(\alpha)} dX_2^{(\alpha)} dX_3^{(\alpha)} \{(a^*_2 c^*_{2,1})_{,3} - (a^*_2 c^*_{2,3})_{,1}\}, \quad (A\,8)$$

where we noted the continuity of the derivatives, or $c^*_{2,13} = c^*_{2,31}$. Then integrating the first and second integrands with respect to $X_3^{(\alpha)}$ and $X_1^{(\alpha)}$, respectively, we have

$$I_s = \lim_{N\to\infty} \sum_{\alpha=1}^{N} \left[\int_{\Delta S^{(\alpha)}} dS_3^{(\alpha)} (a^*_2 c^*_{2,1}) - \int_{\Delta S^{(\alpha)}} dS_1^{(\alpha)} (a^*_2 c^*_{2,3}) \right]. \quad (A\,9)$$

where $(dS_1^{(\alpha)}, dS_2^{(\alpha)}, dS_3^{(\alpha)}) = (dX_2^{(\alpha)} dX_3^{(\alpha)}, dX_3^{(\alpha)} dX_1^{(\alpha)}, dX_1^{(\alpha)} dX_2^{(\alpha)})$, the symbol $\Delta S^{(\alpha)}$ denotes the surface integral on the αth cell; here the sign of the surface integration was selected to be positive towards the outward surface of the cell. Since the surface integrals on the interfaces between neighbouring cells cancel each other out, the summation over α over the cells in equation (A5) can be reduced to the cells which have the common surface with the outer surface S of the volume V, or

$$I_s = \lim_{N\to\infty} \sum_{\alpha:\,\text{outer cell}} \left[\int_{(\Delta S^{(\alpha)} \Lambda S)} dS_3^{(\alpha)} (a^*_2 c^*_{2,1}) - \int_{(\Delta S^{(\alpha)} \Lambda S)} dS_1^{(\alpha)} (a^*_2 c^*_{2,3}) \right]$$

$$= \text{surface contribution}. \quad (A\,10)$$

where $(\Delta S^{(\alpha)} \Lambda S)$ represents the common surface between $\Delta S^{(\alpha)}$ and S. Of course the proof of equation (A 10) is not unique in showing that I_s is the surface contribution. Generally in a similar manner we have the following relations

$$(\mathbf{u}_1 \cdot \nabla\mathbf{U} \cdot \mathbf{u}_2)(\mathbf{u}_3 \cdot \nabla\mathbf{V} \cdot \mathbf{u}_4)$$
$$= (\mathbf{u}_3 \cdot \nabla\mathbf{U} \cdot \mathbf{u}_2)(\mathbf{u}_1 \cdot \nabla\mathbf{V} \cdot \mathbf{u}_4) + \text{surface integral}$$
$$\to (\mathbf{u}_3 \cdot \nabla\mathbf{U} \cdot \mathbf{u}_2)(\mathbf{u}_1 \cdot \nabla\mathbf{V} \cdot \mathbf{u}_4)$$
$$(\{\mathbf{u}_1, \mathbf{u}_2, \mathbf{u}_3, \mathbf{u}_4\} = \{\mathbf{c}, \mathbf{p}, \boldsymbol{\nu}\})$$
$$(\{\mathbf{U}, \mathbf{V}\} = \{\mathbf{a}, \mathbf{c}\}).$$

If we put $\mathbf{a} \cdot \mathbf{a} = 1$ (or $\mathbf{a} = \nu$) as well as $\mathbf{c} \cdot \mathbf{c} = 1$, these results will be immediately reduced to those utilized in the Orsay approach [3–5].

Appendix B

Here we show the equivalence between the present theory and the Orsay theory when we put $\mathbf{a} \cdot \mathbf{a} = (1 + \kappa(\mathbf{r}))^2 =$ constant. First of all we have to note the following relation between the triad $\mathbf{c}$–$\mathbf{p}$–$\boldsymbol{\nu}$ and the triad $\mathbf{e}_1$–$\mathbf{e}_2$–$\mathbf{e}_3$ in the local coordinate x–y–z.

$$\begin{bmatrix} \mathbf{c} \\ \mathbf{p} \\ \boldsymbol{\nu} \end{bmatrix} = \begin{bmatrix} 1 & \Omega_z & -\Omega_y \\ -\Omega_z & 1 & \Omega_x \\ \Omega_y & -\Omega_x & 1 \end{bmatrix} \begin{bmatrix} \mathbf{e}_1 \\ \mathbf{e}_2 \\ \mathbf{e}_3 \end{bmatrix}, \tag{B 1}$$

where Ω_x, Ω_y, and Ω_z are the infinitesimal rotation angles about the x, y, and z axes, respectively [3]. From this we have the relations between the tensor notation and the symmetry broken expression by the Orsay Group [6]

$$(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) = \partial \Omega_z / \partial x, \tag{B 2a}$$

$$(\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) = \partial \Omega_z / \partial y, \tag{B 2b}$$

$$(\boldsymbol{\nu} \cdot \nabla \mathbf{c} \cdot \mathbf{p}) = \partial \Omega_z / \partial z, \tag{B 2c}$$

$$(\mathbf{c} \cdot \nabla \boldsymbol{\nu} \cdot \mathbf{p}) = -\partial \Omega_x / \partial x, \tag{B 2d}$$

$$(\mathbf{p} \cdot \nabla \boldsymbol{\nu} \cdot \mathbf{p}) = -\partial \Omega_x / \partial y, \tag{B 2e}$$

$$(\mathbf{c} \cdot \nabla \boldsymbol{\nu} \cdot \mathbf{c}) = \partial \Omega_y / \partial x. \tag{B 2f}$$

Substituting these relations into equations (22), (24), (25) and (28), we can readily find equation (3) as derived by the Orsay Group [4].

Appendix C

We shall summarize here some vector formulae derived under $\mathbf{a} \cdot \mathbf{c} = \mathbf{c} \cdot \mathbf{p} = \mathbf{p} \cdot \mathbf{a} = 0$, $\mathbf{p} = \boldsymbol{\nu} \times \mathbf{c}$, $\mathbf{c} \cdot \mathbf{c} = \mathbf{p} \cdot \mathbf{p} = \boldsymbol{\nu} \cdot \boldsymbol{\nu} = 1$, $\mathbf{a} = (1 + \kappa)\boldsymbol{\nu}$ as well as $\nabla \times \mathbf{a} = 0$.

$$\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{p} = \boldsymbol{\nu} \cdot \nabla \times \mathbf{c} = -\nabla \cdot \mathbf{p} + \boldsymbol{\nu} \cdot \nabla \mathbf{p} \cdot \boldsymbol{\nu}, \tag{C 1}$$

$$\mathbf{p} \cdot \nabla \mathbf{c} \cdot \mathbf{p} = \boldsymbol{\nu} \cdot \nabla \times \mathbf{p} = \nabla \cdot \mathbf{c} - \boldsymbol{\nu} \cdot \nabla \mathbf{c} \cdot \boldsymbol{\nu}, \tag{C 2}$$

$$\begin{aligned} \boldsymbol{\nu} \cdot \nabla \mathbf{c} \cdot \mathbf{p} &= -\mathbf{c} \cdot \nabla \times \mathbf{c} + \mathbf{p} \cdot \nabla \mathbf{c} \cdot \boldsymbol{\nu} = -\mathbf{p} \cdot \nabla \times \mathbf{p} - \mathbf{c} \cdot \nabla \mathbf{p} \cdot \boldsymbol{\nu} \\ &= -\mathbf{c} \cdot \nabla \times \mathbf{c} - \mathbf{p} \cdot \nabla \boldsymbol{\nu} \cdot \mathbf{c} = -\mathbf{p} \cdot \nabla \times \mathbf{p} + \mathbf{c} \cdot \nabla \boldsymbol{\nu} \cdot \mathbf{p} \\ &= -(\mathbf{c} \cdot \nabla \times \mathbf{c} + \mathbf{p} \cdot \nabla \times \mathbf{p})/2, \end{aligned} \tag{C 3}$$

$$\begin{aligned} \kappa_{,j} &= a_{i,j} \nu_i + a_i \nu_{i,j} \\ &= a_{i,j} \nu_i, \end{aligned} \tag{C 4}$$

$$\begin{aligned} (\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{p}) &= -(\mathbf{p} \cdot \nabla \mathbf{p} \cdot \mathbf{a}) = (\mathbf{p} \times \nabla \times \mathbf{p}) \cdot \mathbf{a} = (\mathbf{a} \times \mathbf{p}) \cdot (\nabla \times \mathbf{p}) \\ &= -(1 + \kappa)(\mathbf{c} \cdot \nabla \times \mathbf{p}), \end{aligned} \tag{C 5}$$

$$\begin{aligned} (\mathbf{c} \cdot \nabla \mathbf{a} \cdot \mathbf{c}) &= -(\mathbf{c} \cdot \nabla \mathbf{c} \cdot \mathbf{a}) = (\mathbf{c} \times \nabla \times \mathbf{c}) \cdot \mathbf{a} = (\mathbf{a} \times \mathbf{c}) \cdot \nabla \times \mathbf{c} \\ &= (1 + \kappa)(\mathbf{p} \cdot \nabla \times \mathbf{c}), \end{aligned} \tag{C 6}$$

$$\begin{aligned} 2(\mathbf{p} \cdot \nabla \mathbf{a} \cdot \mathbf{c}) &= (\mathbf{a} \times \mathbf{c}) \cdot \nabla \times \mathbf{p} - (\mathbf{p} \times \mathbf{a}) \cdot \nabla \times \mathbf{c} \\ &= (1 + \kappa)(\mathbf{p} \cdot \nabla \times \mathbf{p} - \mathbf{c} \cdot \nabla \times \mathbf{c}). \end{aligned} \tag{C 7}$$

In the derivation of equation (C 3), we noted the relation $(\mathbf{p}\cdot\nabla\boldsymbol{\nu}\cdot\mathbf{c}) = (\mathbf{c}\cdot\nabla\boldsymbol{\nu}\cdot\mathbf{p})$ with $\nu_i = a_i/|\mathbf{a}|$.

Appendix D

Let us show that the Orsay free energy equation (3) can easily be derived from Liu's free energy for the biaxial nematics [12]. First we shall write explicitly the free energy for the monoclinic nematics derived from Liu's result for the triclinic nematics in a similar way to the present text [12].

$$\begin{aligned}2F = {} & A_{cc}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{pc}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{ac}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{cp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2\\
&+ A_{pp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + A_{ap}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + 2A_{accc}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})\\
&+ 2A_{acpp}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + 2A_{ccpp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\\
&+ 2A_{pccp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2A_{pcap}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\\
&+ 2A_{cpap}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + B_{cp}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + B_{pp}(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2\\
&+ B_{ap}(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + 2B_{apcp}(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\\
&+ 2C_{apap}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{apcp}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\\
&+ 2C_{cpap}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{cpcp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\\
&+ 2C_{pcap}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{pccp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\\
&+ 2C_{pppp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{acpp}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})\\
&+ 2C_{ccpp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),\end{aligned} \tag{D 1}$$

$$\begin{aligned}F^* = {} & D(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + D_1(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - D_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) + D_3(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})\\
&+ D_4(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p}),\end{aligned} \tag{D 2}$$

where $\mathbf{A}_s$, $\mathbf{B}_s$, $\mathbf{C}_s$, and $\mathbf{D}_s$ are appropriate elastic constants. These energy densities satisfy the monoclinic C_2 symmetry such that

$$\begin{aligned}F(\mathbf{a},\mathbf{c},\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c}) &= F(-\mathbf{a},-\mathbf{c},\mathbf{p},-\nabla\mathbf{a},-\nabla\mathbf{c})\\
&= F(\mathbf{a},\mathbf{c},-\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c}),\end{aligned} \tag{D 3}$$

$$\begin{aligned}F^*(\mathbf{a},\mathbf{c},\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c}) &= F^*(-\mathbf{a},-\mathbf{c},\mathbf{p},-\nabla\mathbf{a},-\nabla\mathbf{c})\\
&= -F^*(\mathbf{a},\mathbf{c},-\mathbf{p},\nabla\mathbf{a},\nabla\mathbf{c}).\end{aligned} \tag{D 4}$$

Therefore the total numbers of the non-chiral and chiral elastic terms are 25 and 5, respectively, consistent with Kini's result for the monoclinic biaxial nematics [13]. Then, to eliminate the surface contributions as shown in Appendix A, the following relations can be utilized:

$$(\mathbf{X}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{Y}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) = (\mathbf{Y}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{X}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + \text{surface contribution}, \tag{D 5a}$$

$$(\mathbf{X}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\mathbf{Y}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (\mathbf{Y}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\mathbf{X}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + \text{surface contribution}, \tag{D 5b}$$

$$(\mathbf{X}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{Y}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (\mathbf{Y}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{X}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + \text{surface contribution}, \tag{D 5c}$$

$$(\mathbf{X}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{Y}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) = (\mathbf{Y}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{X}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + \text{surface contribution} \tag{D 5d}$$

where $(\mathbf{X}, \mathbf{Y}) = (\mathbf{c}, \mathbf{p}, \mathbf{a})$. Now we derive the following bulk energy F_B.

$$
\begin{aligned}
2F_B = {} & A_{cc}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{pc}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{ac}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{cp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 \\
& + A_{pp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + A_{ap}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + 2A_{accc}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c}) \\
& + 2(A_{acpp} + A_{pcap})(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\
& + 2(A_{ccpp} + A_{pccp})(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) + 2A_{cpap}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p}) \\
& + B_{cp}(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + B_{pp}(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + B_{ap}(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 \\
& + 2B_{apcp}(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{apap}(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\
& + 2(C_{apcp} + C_{cpap})(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{cpcp}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\
& + 2(C_{pcap} + C_{acpp})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\
& + 2(C_{pccc} + C_{ccpc})(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + 2C_{pppp}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}).
\end{aligned}
\tag{D 6}
$$

Then we have 20 non-chiral elastic terms for the bulk free energy [17]. If we assume further $F(\mathbf{a}, \mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c}) = F(\pm\mathbf{a}, \mathbf{c}, \mathbf{p}, \pm\nabla\mathbf{a}, \nabla\mathbf{c}) = F(\mathbf{a}, \pm\mathbf{c}, \mathbf{p}, \nabla\mathbf{a}, \pm\nabla\mathbf{c}) = F(\mathbf{a}, \mathbf{c}, \pm\mathbf{p}, \nabla\mathbf{a}, \nabla\mathbf{c})$, we can readily obtain the bulk free energy of the orthorhombic biaxial nematics with 12 non-chiral elastic coefficients [12, 13]. Finally, assuming that there exists no layer dislocation, or $a_{i,j} = a_{j,i}$, i.e. $(\mathbf{a}\cdot\nabla\mathbf{a}\cdot\mathbf{X}) = (\mathbf{X}\cdot\nabla\mathbf{a}\cdot\mathbf{a}) = 0$ [3], we can derive the following results after some rearrangements for the elastic constants:

$$
\begin{aligned}
2F_B = {} & A_{11}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{12}(\mathbf{c}\cdot\nabla\mathbf{a}\cdot\mathbf{c})^2 + A_{21}(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})^2 + B_1(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 \\
& + B_2(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p})^2 + B_3(\mathbf{a}\cdot\nabla\mathbf{p}\cdot\mathbf{c})^2 + 2B_{13}(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) \\
& - 2C_1(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c})(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - 2C_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{p})(\mathbf{p}\cdot\nabla\mathbf{c}\cdot\mathbf{p}),
\end{aligned}
\tag{D 7}
$$

$$
F^* = D(\mathbf{a}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) + D_1(\mathbf{c}\cdot\nabla\mathbf{c}\cdot\mathbf{p}) - D_2(\mathbf{p}\cdot\nabla\mathbf{a}\cdot\mathbf{c}). \tag{D 8}
$$

Now, together with the previous results, equations (B 2 *a*)–(B 2 *f*), we can find the Orsay expression for the incompressible S_C^* given by equation (3). These results are obviously the same as equation (22), (24), (25) and (28) if we put $\mathbf{a}\cdot\mathbf{a} = (1 + \kappa(\mathbf{r}))^2 =$ constant, or $\kappa(\mathbf{r}) = \kappa_0$, therein.

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