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Quasi-uniaxial treatment of elasticity in smectic C* liquid crystals

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An approximate description of the elastic behaviour of smectic C* liquid crystals is introduced in order to facilitate the experimental and numerical analysis of these materials. In this way the original number of 14 independent elastic parameters is reduced to six in the approximate description.

1. Introduction

In recent years a lot of attention has been paid to smectic C* liquid crystals. The description of the elastic behaviour of these materials is quite complicated, as their local orientation must be specified by three orthonormal vectors. The elastic free energy involves 11 bulk elastic constants and three surface elastic constants [1–5]. Until now experimental methods for determining all these constants do not exist [6]. In view of this fact and the mathematical complexity of the free energy expression a simplified description may be quite fruitful.

In this paper an approximate description of the elastic behaviour of the smectic C* phase is introduced. The basic ingredient of this approximation is an additional symmetry requirement. The result is a free energy expression with five bulk elastic constants and only one surface elastic constant. The approximation is formulated mathematically in § 2. Then a simple illustration of the approximation is given in § 3. Next, in § 4, the approximation is applied to the full expression of the smectic C* phase. Finally a summary of the main conclusions is presented in § 5.

2. Mathematical formulation of the approximation

The local orientation of a biaxial liquid crystal is specified by three orthonormal vectors, the directors **a**, **b** and **c**. They are represented in terms of the Eulerian angles $\theta(\mathbf{r})$, $\phi(\mathbf{r})$ and $\psi(\mathbf{r})$ by

$$\mathbf{a} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta), \tag{1 a}$$

$$\mathbf{b} = \cos\psi\,(\cos\theta\cos\phi, \cos\theta\sin\phi, -\sin\theta) + \sin\psi(-\sin\phi, \cos\phi, 0), \tag{1 b}$$

$$\mathbf{c} = -\sin\psi(\cos\theta\cos\phi, \cos\theta\sin\phi, -\sin\theta) + \cos\psi(-\sin\phi, \cos\phi, 0). \tag{1 c}$$

The angle $\psi(\mathbf{r})$ describes the orientation of **b** and **c** in the plane perpendicular to **a**, i.e. is connected with the biaxiality of the liquid crystal. The general expression of the free energy density due to elastic deformation reads [7]

$$f_a = \sum_{i,j=1,2,3} k_{ij}D_{ij} + \frac{1}{2}\sum_{i,j,k,l=1,2,3} K_{ijkl}D_{ij}D_{kl} + \sum_{i,j,k=1,2,3} L_{ijk}S_{ijk}, \tag{2}$$

where the invariants D_{ij} are given by

$$D_{11} = \tfrac{1}{2}[\mathbf{c}\cdot(\nabla\times\mathbf{c}) + \mathbf{a}\cdot(\nabla\times\mathbf{a}) - \mathbf{b}\cdot(\nabla\times\mathbf{b})], \tag{3 a}$$

$$D_{12} = -\mathbf{c}\cdot(\nabla\times\mathbf{b}), \tag{3 b}$$

$$D_{13} = -\mathbf{a}\cdot(\nabla\times\mathbf{b}), \tag{3 c}$$

$$D_{21} = -\mathbf{b}\cdot(\nabla\times\mathbf{c}), \tag{3 d}$$

$$D_{22} = \tfrac{1}{2}[\mathbf{a}\cdot(\nabla\times\mathbf{a}) + \mathbf{b}\cdot(\nabla\times\mathbf{b}) - \mathbf{c}\cdot(\nabla\times\mathbf{c})], \tag{3 e}$$

$$D_{23} = -\mathbf{a}\cdot(\nabla\times\mathbf{c}), \tag{3 f}$$

$$D_{31} = -\mathbf{b}\cdot(\nabla\times\mathbf{a}), \tag{3 g}$$

$$D_{32} = -\mathbf{c}\cdot(\nabla\times\mathbf{a}), \tag{3 h}$$

$$D_{33} = \tfrac{1}{2}[\mathbf{b}\cdot(\nabla\times\mathbf{b}) + \mathbf{c}\cdot(\nabla\times\mathbf{c}) - \mathbf{a}\cdot(\nabla\times\mathbf{a})], \tag{3 i}$$

and the surface terms S_{ijk} by

$$S_{11k} = 2\nabla\cdot(\mathbf{b}D_{1k}), \tag{4 a}$$

$$S_{22k} = 2\nabla\cdot(\mathbf{c}D_{2k}), \tag{4 b}$$

$$S_{33k} = 2\nabla\cdot(\mathbf{a}D_{3k}), \tag{4 c}$$

$$S_{12k} = S_{21k} = \nabla\cdot(\mathbf{b}D_{2k} + \mathbf{c}D_{1k}), \tag{4 d}$$

$$S_{23k} = S_{32k} = \nabla\cdot(\mathbf{c}D_{3k} + \mathbf{a}D_{2k}), \tag{4 e}$$

$$S_{31k} = S_{13k} = \nabla\cdot(\mathbf{a}D_{1k} + \mathbf{b}D_{3k}), \tag{4 f}$$

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for $k = 1, 2, 3$. The parameters k_{ij}, $K_{ijkl} = K_{klij}$ and L_{ijk} are the elastic constants. In the most general case there are 9 chiral elastic constants k_{ij}, 45 elastic constants K_{ijkl} and 18 elastic constants L_{ijk}. In order to avoid confusion, it should be noted that the elastic constants k_{ij}, K_{ijkl} and L_{ijk} as well as the invariants D_{ij} are scalars, although the use of Latin indices to label them may suggest that they are tensors.

The expression (2) contains, apart from the 18 surface terms S_{ijk}, an additional number of 9 surface terms [7]. Three of these surface terms can be expressed as a linear combination of terms linear in the invariants D_{ij}, whereas the remaining six surface terms can be written as a linear combination of terms quadratic in the invariants D_{ij}. This means that three terms linear in D_{ij} can be replaced by surface terms and the remaining 6 terms linear in D_{ij}, whereas six terms quadratic in D_{ij} can be replaced by surface terms and the remaining 39 terms quadratic in D_{ij}. The 18 surface terms S_{ijk} are of a different nature. They can be expressed as a linear combination of terms quadratic in the invariants D_{ij} and terms linear in the spatial derivatives of the invariants D_{ij}.

In the following it is important to realize that symmetry of the phase entails equivalent choices of the directors. For example, if the liquid crystal is symmetric with respect to a reflection in a plane perpendicular to **a**, the set of directors $\{\mathbf{a}, \mathbf{b}, \mathbf{c}\}$ is equivalent to the set of directors $\{-\mathbf{a}, \mathbf{b}, \mathbf{c}\}$. In general, the set $\{\mathbf{a}, \mathbf{b}, \mathbf{c}\}$ is related to an equivalent set $\{\mathbf{a}', \mathbf{b}', \mathbf{c}'\}$ by

$$\mathbf{a}' = T_{33}\mathbf{a} + T_{32}\mathbf{b} + T_{32}\mathbf{c}, \tag{5a}$$

$$\mathbf{b}' = T_{13}\mathbf{a} + T_{11}\mathbf{b} + T_{12}\mathbf{c}, \tag{5b}$$

$$\mathbf{c}' = T_{23}\mathbf{a} + T_{21}\mathbf{b} + T_{22}\mathbf{c}, \tag{5c}$$

where the numbers T_{ij} are the matrix elements of the symmetry transformation. It should be remarked that equations (5) hold for all space-fixed coordinate frames. Thus the numbers T_{ij} are scalars. All physical properties, such as the elastic properties, remain unchanged under such a transformation of the directors. Consequently the elastic constants satisfy the relations [7]

$$k_{ij} = \sum_{m,n=1,2,3} T T_{mi} T_{nj} k_{mn}, \tag{6a}$$

$$K_{ijkl} = \sum_{m,n,p,q=1,2,3} T_{mi} T_{nj} T_{pk} T_{ql} K_{mnpq}, \tag{6b}$$

$$L_{ijk} = \sum_{m,n,p=1,2,3} T T_{mi} T_{nj} T_{pk} L_{mnp}, \tag{6c}$$

for all transformations belonging to the symmetry group of the particular phase under consideration. Here $T = \pm 1$ is the determinant of the transformation matrix. These symmetry relations reduce the number of independent elastic constants.

A further reduction of the number of elastic parameters is possible in case of uniaxial symmetry around a certain axis, say **a**. Then the deformation free energy density is invariant with respect to an arbitrary rotation of the directors **b** and **c** around **a**. This means that neither the Eulerian angle $\psi(\mathbf{r})$ nor the gradient $\nabla\psi(\mathbf{r})$ may appear in the deformation free energy density. The dependence on $\psi(\mathbf{r})$ is eliminated by means of the relations (6). However, the dependence on $\nabla\psi(\mathbf{r})$ must be eliminated by means of additional symmetry relations; that is, the invariants D_{ij} depending on $\nabla\psi(\mathbf{r})$ must be excluded. It appears that only the invariants D_{i3} depend on $\nabla\psi(\mathbf{r})$. Thus the additional symmetry relations read

$$k_{i3} = 0, \tag{7a}$$

$$K_{i3jk} = K_{jki3} = 0, \tag{7b}$$

$$L_{ij3} = 0, \tag{7c}$$

for $i, j, k = 1, 2, 3$.

In order to obtain the approximate description of the smectic C* phase, it must be realized that the biaxial character of that phase is expressed by $\psi(\mathbf{r})$ as well as $\nabla\psi(\mathbf{r})$. Further the smectic plane is identified with the plane spanned by the vectors **b** and **c**. The approximation boils down to neglecting the dependence of the free energy density on $\psi(\mathbf{r})$. Thus the biaxiality is only taken into account by $\nabla\psi(\mathbf{r})$. This means that the elastic constants are required to satisfy the symmetry relations (6) that hold for uniaxial symmetry around the smectic layer normal **a**. Therefore the present approximation can be called a quasi-uniaxial approximation. For the sake of clearness, a similar approximation for uniaxial liquid crystals, the quasi-isotropic approximation, is treated first.

3. The quasi-isotropic approximation

The gist of the quasi-isotropic approximation is the requirement that the elastic constants satisfy the relations (6) for all possible rotations. Then it follows that the elastic constants must be expressed in terms of the Kronecker-symbol δ_{ij}, i.e.

$$k_{ij} = \gamma \delta_{ij}, \tag{8a}$$

$$K_{ijkl} = \beta_1 \delta_{ij} \delta_{kl} + \beta_2 \delta_{ik} \delta_{jl} + \beta_3 \delta_{il} \delta_{jk}, \tag{8b}$$

$$L_{ijk} = 0. \tag{8c}$$

As to the elastic constants K_{ijkl}, this result has also been derived by Liu [8]. The elastic constant γ equals zero for a non-chiral nematic. Further, the uniaxial character of the nematic must be taken into account. Consequently, choosing **a** as the uniaxial axis, all invariants D_{i3} must be put equal to zero. Then the quasi-isotropic approximation gives rise to the following free energy density of a nematic:

$$f_d = \tfrac{1}{2}\beta_2(\nabla \cdot \mathbf{a})^2 + \tfrac{1}{2}(\beta_1 + \beta_2 + \beta_3)(\mathbf{a} \cdot (\nabla \times \mathbf{a}))^2 + \tfrac{1}{2}\beta_2(\mathbf{a} \times (\nabla \times \mathbf{a}))^2 + \tfrac{1}{2}(\beta_2 + \beta_3)\nabla \cdot [(\mathbf{a} \cdot \nabla)\mathbf{a} - \mathbf{a}(\nabla \cdot \mathbf{a})]. \quad (9)$$

The full deformation free energy density reads [9–11]

$$f_d = \tfrac{1}{2}K_{11}(\nabla \cdot \mathbf{a})^2 + \tfrac{1}{2}K_{22}(\mathbf{a} \cdot (\nabla \times \mathbf{a}))^2 + \tfrac{1}{2}K_{33}(\mathbf{a} \times (\nabla \times \mathbf{a}))^2 + \tfrac{1}{2}(K_{22} + K_{24})\nabla \cdot [(\mathbf{a} \cdot \nabla)\mathbf{a} - \mathbf{a}(\nabla \cdot \mathbf{a})] + K_{13}\nabla \cdot [\mathbf{a}(\nabla \cdot \mathbf{a})]. \quad (10)$$

Consequently it holds that

$$K_{11} = K_{33} = \beta_2, \quad (11\,a)$$

$$K_{22} = \beta_1 + \beta_2 + \beta_3, \quad (11\,b)$$

$$K_{24} = -\beta_1, \quad (11\,c)$$

$$K_{13} = 0. \quad (11\,d)$$

Clearly the quasi-isotropic approximation for uniaxial liquid crystals boils down to neglecting the splay–bend surface elastic constant as well as the difference between the elastic constants for splay and bend.

In order to examine the merits of the simplified description of nematics, the optical transmission of a non-twisted nematic liquid crystal cell filled with the liquid crystal 4′-*n*-pentyl-4-cyanobiphenyl (5CB) is considered. To that end the result of the quasi-isotropic approximation is compared with the result based upon the measured K_{11} and K_{33} values. These values are at room temperature: $K_{11} = 7{\cdot}05\,\mathrm{pN}$, $K_{33} = 9{\cdot}75\,\mathrm{pN}$ [12]. The average value $K_{11} = K_{33} = 8{\cdot}40\,\mathrm{pN}$ is taken in the quasi-isotropic approximation. The results of the calculations are shown in figure 1. It appears that the approximation works reasonably well, bearing in mind that the relative difference between the splay and bend elastic constants amounts to 30 per cent. The same conclusion can be drawn for commercially available liquid crystal mixtures in view of the values of their elastic constants, i.e. $K_{33}/K_{11} \approx 1{\cdot}4$ and $K_{33}/K_{22} \approx 2{\cdot}5$, respectively [13].

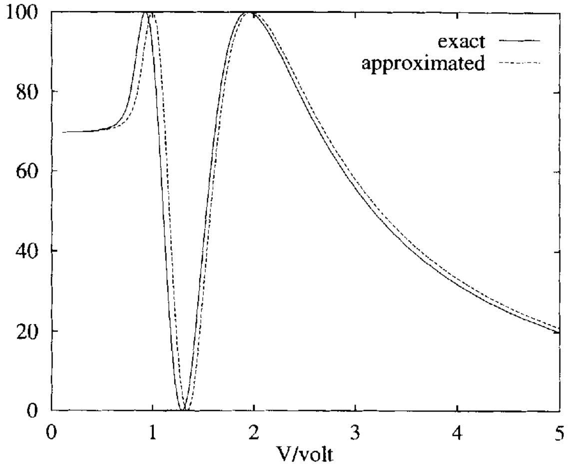

Figure 1. Exact and approximate optical transmission of a 3 μm thick cell filled with 5CB between crossed polarizers for 632·8 nm light. The polarizers are at 45° with the rubbing direction. The dielectric constants are $\varepsilon_\perp = 5{\cdot}5$ and $\varepsilon_\parallel = 16{\cdot}1$ [14] and the refractive indices are $n_e = 1{\cdot}709$ and $n_0 = 1{\cdot}531$ [15]. The surface tilt was taken to be 2° independent of the applied voltage, i.e. strong anchoring conditions are assumed.

4. The quasi-uniaxial approximation for smectic C* liquid crystals

In order to apply the quasi-uniaxial approximation, the director **a** is chosen perpendicular to the smectic layers. Further the director **b** is taken along the axis of twofold rotation symmetry. In addition to orientational deformations, smectics allow for positional deformations. The general form of the deformation free energy density of a smectic C* liquid crystal has been given in [1–5]. According to [5] this deformation free energy density reads

$$f_d = \tfrac{1}{2}\lambda\gamma^2 + \tfrac{1}{2}A_{11}D_{11}^2 + \tfrac{1}{2}A_{12}D_{21}^2 + \tfrac{1}{2}A_{21}D_{12}^2 + \tfrac{1}{2}B_1D_{23}^2 + \tfrac{1}{2}B_2D_{13}^2 + \tfrac{1}{2}B_3D_{33}^2 + B_{13}D_{23}D_{33} + C_1D_{11}D_{23} + C_2D_{12}D_{13} - DD_{33} + E_5(D_{12}D_{21} + D_{11}^2) + E_6D_{11}D_{33} + E_7(D_{13}D_{21} - D_{23}D_{11}). \quad (12)$$

The positional deformations of the smectic are taken into account by the layer compression term $\lambda\gamma^2/2$, where λ is the positional elastic constant and γ is given by

$$\gamma = \partial_z u_z - \tfrac{1}{2}\{(\partial_x u_z)^2 + (\partial_y u_z)^2 + (\partial_z u_z)^2\}. \quad (13)$$

Here u_z is the displacement of the smectic layer perpendicular to the undistorted flat equilibrium layer, which is assumed to lie in the *xy*-plane. Further, the coupling between positional and orientational deformations leads to the Oseen-condition [9]

$$\nabla \times \mathbf{a} = 0. \quad (14)$$

Application of the requirement that the symmetry relations (6) hold for all possible rotations around **a** results in

$$A_{11} = A_{12} = A_{21} = A, \quad (15\,a)$$

$$B_1 = B_2 = B_\parallel, \quad (15\,b)$$

$$B_3 = B_\perp, \quad (15\,c)$$

$$B_{13} = 0, \quad (15\,d)$$

$$C_1 = C_2 = 0, \quad (15\,e)$$

$$D = -B_{\perp} q_0, \tag{15f}$$

$$E_5 = A_s - A, \tag{15g}$$

$$E_6 = E_7 = 0. \tag{15h}$$

Consequently the number of independent elastic parameters is reduced from 14 to six in the quasi-uniaxial approximation. The resulting expression for f_d can be rewritten in terms of the usual vector notation with the aid of the Oseen-condition (14) and the relations

$$D_{21} - D_{12} = \nabla \cdot \mathbf{a}, \tag{16a}$$

$$D_{33} = \tfrac{1}{2}[\mathbf{b} \cdot (\nabla \times \mathbf{b}) + \mathbf{c} \cdot (\nabla \times \mathbf{c}) - \mathbf{a} \cdot (\nabla \times \mathbf{a})], \tag{16b}$$

$$D_{13} = \nabla \cdot \mathbf{c} - \mathbf{b} \cdot (\nabla \times \mathbf{a}), \tag{16c}$$

$$D_{23} = -\nabla \cdot \mathbf{b} - \mathbf{c} \cdot (\nabla \times \mathbf{a}), \tag{16d}$$

$$D_{12}D_{21} - D_{11}D_{22} = \tfrac{1}{2}\nabla \cdot [(\mathbf{a} \cdot \nabla)\mathbf{a} - \mathbf{a}(\nabla \cdot \mathbf{a})]. \tag{16e}$$

Then the deformation free energy density of a smectic C* liquid crystal in the quasi-uniaxial approximation can be expressed as

$$\begin{aligned} f_d = {} & \tfrac{1}{2}\lambda\gamma^2 + \tfrac{1}{2}A(\nabla \cdot \mathbf{a})^2 \\ & + \tfrac{1}{2}B_{\perp}[\tfrac{1}{2}\{(\mathbf{b} \cdot (\nabla \times \mathbf{b}) + \mathbf{c} \cdot (\nabla \times \mathbf{c})\}]^2 \\ & + B_{\perp} q_0 [\tfrac{1}{2}\{(\mathbf{b} \cdot (\nabla \times \mathbf{b}) + \mathbf{c} \cdot (\nabla \times \mathbf{c})\}] \\ & + \tfrac{1}{2}B_{\parallel}[(\nabla \cdot \mathbf{b})^2 + (\nabla \cdot \mathbf{c})^2] \\ & + \tfrac{1}{2}A_s \nabla \cdot [(\mathbf{a} \cdot \nabla)\mathbf{a} - \mathbf{a}(\nabla \cdot \mathbf{a})]. \end{aligned} \tag{17}$$

Three terms in (17) correspond to deformations of the smectic layers. The λ term is associated with the compression and dilatation of the smectic layers, the A term with the splay of the normal to the smectic layers and the A_s surface term with the saddle–splay of the normal to the smectic layers. The local shape of the smectic layers for these last two deformations is shown in figure 2. The other three terms in (17) correspond to deformations of the directors **b** and **c**. The $B_{\perp}$ terms involve the twist of **b** and **c**, and the $B_{\parallel}$ term involves the splay of **b** and **c**. These deformations can be conveniently shown using the director **n**, which is defined by

$$\mathbf{n} = \cos\omega\, \mathbf{a} + \sin\omega\, \mathbf{c}, \tag{18}$$

where ω is the so-called tilt angle. From a molecular point of view, the long axes of the molecules are on average directed along **n**. Then the **c** director corresponds to the projection on the smectic layers of the average direction of the long axes of the molecules. The director **n** is constrained to a cone with a top angle ω around **a**, in the case that the smectic layer structure is fixed. Both the $B_{\perp}$ terms and the $B_{\parallel}$ term describe spatially dependent rotations of **n** over the smectic cone. The $B_{\perp}$ terms give the spatial dependence in a direction perpendicular to the smectic layers, whereas the $B_{\parallel}$ term gives the spatial dependence parallel to the smectic layers. These deformations are shown in figure 3. The director fields **b**(**r**) and **c**(**r**) are twisted in equilibrium, i.e. the director **n** precesses around **a** in the direction perpendicular to the smectic layers. The pitch of this helix deformation is given by $2\pi/q_0$.

As to the usefulness of the quasi-uniaxial approximation of the free energy density of the smectic C* phase, the following remarks should be made. First of all the quasi-uniaxial expression reduces to the one used by Glogarová *et al.* [16, 17], and by van Haaren [6] in the analysis of their experiments for the special case of rigid smectic layers, i.e. $\gamma = 0$ and **a** is spatially independent. The resulting expression involves only three parameters, namely the helical pitch $2\pi/q_0$ and the elastic constants $B_{\parallel}$ and $B_{\perp}$. The experiments of Willis *et al.* [18], suggest that the approximation of rigid smectic layers is reasonable. From a theoretical point of view it would be interesting to

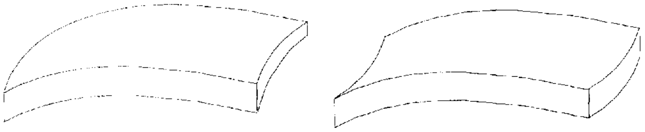

Figure 2. Curved smectic layers in the case of splay deformation of the layer normal and a saddle–splay deformation of the layer normal.

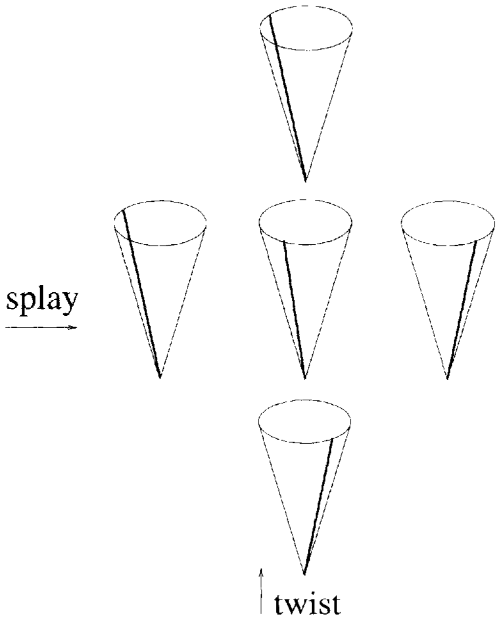

Figure 3. Twist deformation of the directors **b** and **c** (rotation of the **n** director over the smectic cone perpendicular to the smectic layers) and splay deformation of the directors **b** and **c** (rotation of the **n** director over the smectic cone parallel to the smectic layers).

apply the quasi-uniaxial approximation to the study of the influence of an electric field on a smectic C* cell, analogous to the work of De Meyere *et al.* [19, 20]. Further the quasi-uniaxial approximation could be useful in treating the dynamical behaviour of displays based on the smectic C* phase, because it can be expected that the number of dynamical parameters, for example, viscosity coefficients, is substantially reduced as well. In general, the usefulness of the quasi-uniaxial approximation must appear from its adequacy to describe experimental results. Unfortunately the elastic constants of smectic C* liquid crystals are unknown, i.e. a definite assessment cannot be made. However, the adequacy of the analogous quasi-isotropic approximation seems to point out that the quasi-uniaxial approximation may be relevant as well. Finally it should be mentioned that the present approximation differs essentially from the approximation of Nakagawa [21] as to the invariants that appear.

5. Conclusion

In order to reduce the large number of independent parameters that appear in the description of the behaviour of biaxial liquid crystals, the free energy density is approximated by only taking into account a part of the biaxial character of the phase. In this way a simplified expression of the free energy density is obtained that may be useful for experimental and numerical analysis of complex liquid crystalline phases such as the smectic C* phase. The resulting, so-called quasi-uniaxial, approximation requires that the elastic constants satisfy a part of the symmetry relations of the uniaxial phase. A similar approximation, the quasi-isotropic approximation, can be made for the uniaxial phase. The quasi-uniaxial approximation for describing smectic C* liquid crystals involves six elastic parameters instead of the 14 parameters appearing in the exact theory. These six parameters consist of three parameters that are related to deformations of the smectic layers. These deformations are the compression and dilatation of the smectic layers, the splay of the smectic layer-normal and the saddle–splay of the layer normal. The contribution due to the saddle–splay deformation is a surface term. The remaining three parameters are related to deformations of the directors **b** and **c**. Two parameters are associated with the twist of **b** and **c**, and one parameter is associated with the splay of **b** and **c**. One of the two parameters related to the twist deformation is the pitch of the equilibrium helix deformation.

We are indebted to J. A. M. M. van Haaren of the Philips Research Laboratories for pointing out the discrepancy between the theoretical analysis of the elastic free energy of smectic C* and the development of measurement methods for the elastic constants of this phase, which motivated this work.

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