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An elastic energy for the ferroelectric chiral smectic C* phase

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Abstract. An expression for the elastic energy of the ferroelectric, chiral smectic C* liquid crystal phase is presented. It is shown that due to the chirality of the system, compared with the elastic energy of the smectic C phase, the elastic energy of the smectic C* phase contains three additional terms, one of which is a surface term. Of the two remaining terms one is responsible for the helicoidal ordering of the c-director, while the other term will impose a tendency on the smectic layers to be non-uniform in space. The physical interpretation of the presence in the elastic energy of such a term is discussed.

1. Introduction

In order to describe a smectic C (SmC) liquid crystal one must specify [1] the spatial variation of the two unit vectors characterizing the layer normal and the projection of the director into the smectic planes (the c-director). We denote these two unit vectors $\boldsymbol{a}$ and $\boldsymbol{c}$, respectively. Assuming that the system studied is free from dislocations and of constant layer thickness, the layer normal $\boldsymbol{a}$ must fulfil [1] the constraint

$$\nabla \times \boldsymbol{a} = 0. \tag{1.1}$$

Often it is useful also to introduce a third unit vector $\boldsymbol{b}$ according to

$$\boldsymbol{b} = \boldsymbol{a} \times \boldsymbol{c}. \tag{1.2}$$

The definition of $\boldsymbol{b}$ is such that its direction corresponds to the direction of positive polarization [2] in the ferroelectric, chiral smectic C* (SmC*) phase. The elastic free-energy density of these tilted smectic systems has been presented in a number of different ways [1, 3-8] in the literature, but with few exceptions [1, 5] only the case of a non-chiral system has been considered. The original version of the SmC elastic energy which appeared in the literature was that of the Orsay Liquid Crystal Group [3]. In their work the elastic deformations of the system are expressed in terms of a rotation vector $\boldsymbol{\Omega}$. This formulation permits a straightforward, intuitive physical interpretation of the terms in the elastic energy, but is not very suitable as a starting point for performing calculations. For this it is more convenient to express the elastic energy in terms of vector operators acting on either two of the three unit vectors $\boldsymbol{a}$, $\boldsymbol{b}$ and $\boldsymbol{c}$.

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Using such an approach we have previously shown [7, 8] how the SmC elastic energy can be written as

$$g_0 = \tfrac{1}{2}A_{12}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})^2 + \tfrac{1}{2}A_{21}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b})^2 + A_{11}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b}) + \tfrac{1}{2}B_1(\nabla\cdot\boldsymbol{b})^2$$
$$+ \tfrac{1}{2}B_2(\nabla\cdot\boldsymbol{c})^2 + \tfrac{1}{2}B_3[\tfrac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b} + \boldsymbol{c}\cdot\nabla\times\boldsymbol{c})]^2$$
$$+ B_{13}(\nabla\cdot\boldsymbol{b})[\tfrac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b} + \boldsymbol{c}\cdot\nabla\times\boldsymbol{c})]$$
$$+ C_1(\nabla\cdot\boldsymbol{c})(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c}) + C_2(\nabla\cdot\boldsymbol{c})(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b}) \tag{1.3}$$

where each of the five vector operators appearing in (1.3) corresponds [7] to one of the five eigendeformations which a SmC liquid crystalline system can undergo.

When studying the ferroelectric SmC* phase the $\boldsymbol{ac}$-plane will not, in contrast to the case for the SmC phase, be a symmetry plane of the system. Thus the elastic free-energy density of this system will contain some additional terms to these given by (1.3). We show in this paper that the number of additional terms introduced by removing this symmetry is three, one of which is a surface term. We also show how one of the two remaining terms is responsible for ordering the c-director in a helicoidal way, while the presence of the other term imposes a tendency on the smectic layers to be non-uniform in space. While the helicoidal ordering of the c-director is a well established experimental fact, the implication of the second term is somewhat obscure. The physical interpretation of this term is discussed in sections 3 and 4.

2. The chiral operators in the elastic energy

The possible candidates for chiral terms in the elastic energy of the SmC* phase are those which are linear in the gradients of $\boldsymbol{b}$ or $\boldsymbol{c}$. Using the constraints (1.1) and (1.2) it is simple to show, in Cartesian component form, that the only non-zero possible terms are

$$b_{i,i} = c_i b_{i,j} c_j \qquad a_i b_{i,j} c_j \qquad c_i b_{i,j} a_j \tag{2.1}$$

$$c_{i,i} = b_i c_{i,j} b_j \qquad a_i b_{i,j} b_j \qquad a_i c_{i,j} c_j. \tag{2.2}$$

Since the energy terms must be invariant to the simultaneous changes in sign, $\boldsymbol{a}\to-\boldsymbol{a}$ and $\boldsymbol{c}\to-\boldsymbol{c}$, we see that only the terms in (2.1) are relevant terms. The corresponding operators may be written in vector form as

$$O_1^* = c_i b_{i,j} a_j = \tfrac{1}{2}[\boldsymbol{b}\cdot\nabla\times\boldsymbol{b} + \boldsymbol{c}\cdot\nabla\times\boldsymbol{c}] \tag{2.3}$$

$$O_2^* = a_i b_{i,j} c_j = \tfrac{1}{2}[\boldsymbol{c}\cdot\nabla\times\boldsymbol{c} - \boldsymbol{b}\cdot\nabla\times\boldsymbol{b}] \tag{2.4}$$

$$O_3^* = b_{i,i} = \nabla\cdot\boldsymbol{b}. \tag{2.5}$$

3. The bulk elastic energy

In order to write down the elastic energy of the ferroelectric chiral SmC* phase one adds the operators of (2.3)-(2.5) to the expression of the SmC elastic energy of (1.3). We thus can write the elastic energy of the SmC* phase as $g = g_0 + g^*$, where g^* is the chiral part of g which can be written as

$$g^* = \Lambda[\tfrac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b} + \boldsymbol{c}\cdot\nabla\times\boldsymbol{c})] + 2\tau[\tfrac{1}{2}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{c} - \boldsymbol{b}\cdot\nabla\times\boldsymbol{b})] + \sigma\nabla\cdot\boldsymbol{b}. \tag{3.1}$$

In (3.1) we have introduced three chiral elastic constants Λ, τ and σ. Being interested in the bulk energy of the system only, the term $\sigma\nabla\cdot\boldsymbol{b}$ can be neglected, since it has the form of a divergence and thus can be transformed into a surface term.

Concerning the τ-term in (3.1) we reason as follows. Due to the identity [8]

$$-\tfrac{1}{2}\,\mathrm{div}[(\mathrm{div}\,\boldsymbol{a})\boldsymbol{a}]=(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b})+[\tfrac{1}{2}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{c}-\boldsymbol{b}\cdot\nabla\times\boldsymbol{b})]^2 \tag{3.2}$$

we can, neglecting a surface term and a shift in the zero level of g, write the sum of the A_{11}- and τ-terms of the SmC* elastic energy as

$$g_{A_{11}}+g^*_\tau=-A_{11}\left[\frac{1}{2}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{c}-\boldsymbol{b}\cdot\nabla\times\boldsymbol{b})-\frac{\tau}{A_{11}}\right]^2. \tag{3.3}$$

Thus a non-zero τ leads to a ground state of the system for which $(\boldsymbol{c}\cdot\nabla\times\boldsymbol{c}-\boldsymbol{b}\cdot\nabla\times\boldsymbol{b})$ is non-zero, or equivalently apart from a surface term (cf (3.2)), $(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b})$ is non-zero. The physical implication [7] of this operator being non-zero is that the smectic layer normal is non-uniform in space. Thus, if we assume that flat, undistorted layers represent the ground state of the system, we must demand $\tau=0$.

The Λ-term in (3.1) can be incorporated into the expression of (1.3) in a similar way. Apart from a constant contribution, the sum of the B_3- and Λ-terms of the SmC* elastic energy can be written

$$g_{B_3}+g^*_\Lambda=\frac{1}{2}B_3\left[\frac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b}+\boldsymbol{c}\cdot\nabla\times\boldsymbol{c})+\frac{\Lambda}{B_3}\right]^2. \tag{3.4}$$

The ground state of a system with the Λ-term present is a system for which $-\frac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b}+\boldsymbol{c}\cdot\nabla\times\boldsymbol{c})=\Lambda/B_3$. The operator $-(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b}+\boldsymbol{c}\cdot\nabla\times\boldsymbol{c})/2$ corresponds [7] to the c-director rotating in a positive sense as we are moving in the direction of the layer normal $\boldsymbol{a}$. Thus the Λ-term is responsible for the helicoidal ordering of the c-director which appears in the SmC* phase. Introducing the wavevector q of the pitch

$$q=\Lambda/B_3 \tag{3.5}$$

we thus conclude that, with the assumption that the system with flat layers is the stable one, the only difference between the SmC and SmC* elastic energies is the incorporation of $q=\Lambda/B_3$ into the B_3-term of (1.3) according to (3.4).

4. Discussion

Extending the expression of the elastic free-energy density of the SmC phase to that of the SmC* phase we have to introduce the three additional terms of (3.1) into the elastic energy (1.3). As one of these three terms is written as a divergence, only two terms are of importance for the bulk elastic energy which in the general case can be written as

$$\begin{aligned}g=&\frac{1}{2}A_{12}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})^2+\frac{1}{2}A_{21}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b})^2-A_{11}\left[\frac{1}{2}(\boldsymbol{c}\cdot\nabla\times\boldsymbol{c}-\boldsymbol{b}\cdot\nabla\times\boldsymbol{b})-\frac{\tau}{A_{11}}\right]^2\\&+\frac{1}{2}B_1(\nabla\cdot\boldsymbol{b})^2+\frac{1}{2}B_2(\nabla\cdot\boldsymbol{c})^3+\frac{1}{2}B_3\left[\frac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b}+\boldsymbol{c}\cdot\nabla\times\boldsymbol{c})+\frac{\Lambda}{B_3}\right]^2\\&+B_{13}(\nabla\cdot\boldsymbol{b})[\tfrac{1}{2}(\boldsymbol{b}\cdot\nabla\times\boldsymbol{b}+\boldsymbol{c}\cdot\nabla\times\boldsymbol{c})]+C_1(\nabla\cdot\boldsymbol{c})(\boldsymbol{b}\cdot\nabla\times\boldsymbol{c})\\&+C_2(\nabla\cdot\boldsymbol{c})(\boldsymbol{c}\cdot\nabla\times\boldsymbol{b}).\end{aligned} \tag{4.1}$$

From (1.3) and (4.1) we see that the difference between the SmC and SmC* elastic energies is taken care of by a slight modification of the A_{11}- and the B_3-terms. Concerning the B_3-term this modification resembles the corresponding difference between the nematic and cholesteric elastic energies [1]. In a similar way as in the cholesteric case, the ground state of the chiral SmC* system is the one for which the c-director rotates in space with the wavevector $q = \Lambda/B_3$ as we transverse the medium perpendicular to the smectic layers.

The presence of the τ-term in (4.1) leads to a ground state of the system for which the smectic layers are distorted. There are two different ways of handling this term. If we assume that the flat, uniform layers represent the ground state of the SmC* phase then we are forced to impose the condition $\tau = 0$, since allowing τ to be non-zero would lead to distortions of the smectic layers. Such distortions could of course be suppressed by surface forces, especially in the case of thin samples. As was also pointed out by de Gennes [1], the distortions caused by a non-zero τ would either lead to a change of the constant layer thickness or be accompanied by a series of dislocations. Thus, if the energy which is associated with the compression of the smectic layers or with dislocation formation is too large, the system consisting of flat, uniform layers can still represent the ground state of the system even for a non-zero τ. In such a system the smectic layers will possess an inherent tendency of twist, a tendency which however is suppressed by energetical reasons. Thus we would like to raise the question whether the experimental difficulties in making SmC* samples with a well defined ordering of the smectic layers could be due to the presence of the τ-term in the SmC* elastic energy. In the end, however, the question whether the τ-term should be incorporated into the SmC* elastic energy or not must be answered by a careful experimental investigation of the layer structure of the SmC* phase.

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