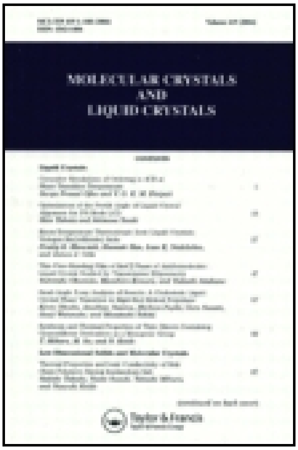

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DISTORTIONS INDUCED BY A MAGNETIC FIELD IN PLANAR ALIGNED SAMPLES OF SMECTIC-C LIQUID CRYSTALS

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Distortions Induced by a Magnetic Field in Planar Aligned Samples of Smectic-*C* Liquid Crystals

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This article presents some preliminary theoretical results for the onset of the Helfrich-Hurault transition in smectic-*C* liquid crystals induced by a magnetic field applied parallel to the smectic layers. An energy in terms of the smectic layer displacement u is minimized via averaging to enable the calculation of a critical field strength H_c for the onset of layer distortions.

Keywords: smectic *C*; layer distortions; Helfrich-Hurault instability

INTRODUCTION

This article exploits a model for layer distortions in smectic-C liquid crystals subjected to an applied magnetic field. There are well known results for determining a critical magnetic field magnitude H_c for the onset of layer distortions in infinite samples of smectic-A where the transition from uniformly aligned planar layers to undulated layers is known as the Helfrich-Hurault transition. To extend these results from the smectic-A case to smectic-C a novel possible energy formulation is used. A critical H_c value will be calculated in a simple case. The motivation for this work comes from that of Helfrich [1] and Hurault [2] who examined infinite samples of cholesteric liquid crystals under the influence of magnetic fields. The derivation of critical field magnitudes for infinite samples of smectic-A can be found in de Gennes and Prost [3] and Chandrasekhar [4].

Liquid crystals consist of elongated molecules whose average molecular axes locally align along a common direction in space which is usually denoted by the unit vector $\mathbf{n}$, called the director. Smectic-C liquid crystals are known to form equidistant parallel layers in which $\mathbf{n}$ makes an angle θ (the smectic tilt angle) with respect to the layer normal. Smectic-A occurs when $\theta \equiv 0$. Following de Gennes and Prost [3], smectic-C is described by the unit layer normal $\mathbf{a}$ and a vector $\mathbf{c}$ which is the unit orthogonal projection of $\mathbf{n}$ onto the smectic planes (see Fig.1). The director $\mathbf{n}$ is then given via the equation

$$\mathbf{n} = \mathbf{a}\cos\theta + \mathbf{c}\sin\theta. \tag{1}$$

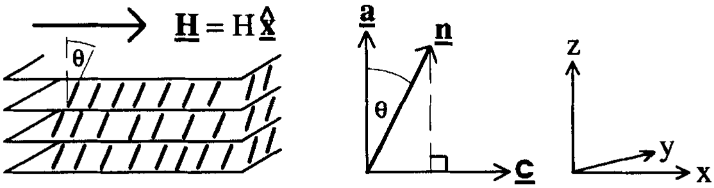

FIGURE 1 Planar samples of smectic C described in cartesian coordinates. The director $\mathbf{n}$ is tilted at an angle θ to the layer normal $\mathbf{a}$; $\mathbf{c}$ is the unit orthogonal projection of $\mathbf{n}$ onto the smectic planes. $\mathbf{H}$ is the magnetic field.

From their definitions the vectors $\mathbf{a}$ and $\mathbf{c}$ must satisfy the constraints

$$\mathbf{a}\cdot\mathbf{a} = \mathbf{c}\cdot\mathbf{c} = 1, \qquad \mathbf{a}\cdot\mathbf{c} = 0, \tag{2}$$

since these vectors are clearly unit and orthogonal. It is also mathematically convenient to introduce the unit vector $\mathbf{b}$ defined by

$$\mathbf{b} = \mathbf{a}\times\mathbf{c}. \tag{3}$$

In the absence of dislocations the layer normal $\mathbf{a}$ is subject to the additional constraint, due to Oseen [5],

$$\nabla\times\mathbf{a} = \mathbf{0}. \tag{4}$$

This constraint is known to restrict the available types of equilibrium structures consisting of undistorted parallel layers which form planes, concentric cylinders, spheres and the more complex structures consisting of concentric circular tori, Dupin cyclides [3,6,7] and parabolic cyclides [8–11]. It is expected therefore that when layer undulations or distortions occur then the above Oseen constraint in equation (4) will be broken. The construction of a suitable energy which incorporates small layer distortions is based upon relaxing the constraints (2) and (4) and examining the consequent changes to the usual (undistorted layer) smectic-C bulk energy. This approach has been employed in the planar aligned smectic-A phase in references [3, 4] and in a more general setting for smectic-A by Kleman and Parodi [12], whose methods have been adapted for our purposes in the next section.

The bulk elastic energy integrand w_b for a non-chiral smectic-C liquid crystal can be written in terms of the derivatives of $\mathbf{a}$ and $\mathbf{c}$ as [13]

$$\begin{aligned} w_b = {} & \tfrac{1}{2}A_{21}(\nabla\cdot\mathbf{a})^2 + \tfrac{1}{2}B_2(\nabla\cdot\mathbf{c})^2 + \tfrac{1}{2}B_1(\mathbf{a}\cdot\nabla\times\mathbf{c})^2 + \tfrac{1}{2}B_3(\mathbf{c}\cdot\nabla\times\mathbf{c})^2 \\ & + \tfrac{1}{2}(2A_{11}+A_{12}+A_{21}+B_3)(\mathbf{b}\cdot\nabla\times\mathbf{c})^2 \\ & - \tfrac{1}{2}(2A_{11}+2A_{21}+B_3)(\nabla\cdot\mathbf{a})(\mathbf{b}\cdot\nabla\times\mathbf{c}) \\ & - B_{13}(\mathbf{a}\cdot\nabla\times\mathbf{c})(\mathbf{c}\cdot\nabla\times\mathbf{c}) \\ & + (C_1+C_2-B_{13})(\nabla\cdot\mathbf{c})(\mathbf{b}\cdot\nabla\times\mathbf{c}) - C_2(\nabla\cdot\mathbf{a})(\nabla\cdot\mathbf{c}), \end{aligned} \tag{5}$$

where the elastic constants A_i, B_i and C_i are related to those introduced by the Orsay Group [14], the minor modification being that $A_{11} = -\frac{1}{2}A_{11}^{Orsay}$ and $C_1 = -C_1^{Orsay}$. Other equivalent formulations can be found in [13] while a physical interpretation of the elastic constants has been discussed by Carlsson *et al* [15]. It is known that these constants obey the inequalities, among others [15],

$$A_{12},\ A_{21},\ B_1,\ ,B_2,\ B_3 > 0. \tag{6}$$

These constants are estimated to be of the order 10^{-7} to 10^{-6} dynes (*cgs*), as in nematics [3]. Further *a priori* inequalities have been derived by Atkin and Stewart [16]: one of particular relevance here is

$$B_2 + A_{21} \pm 2C_2 > 0. \tag{7}$$

It will also be relevant later to note that the smectic tilt angle dependence of the elastic constants can be approximated for small θ

by [15]

$$A_{12} = K + \overline{A}_{12}\theta^2, \quad A_{21} = K + \overline{A}_{21}\theta^2, \quad A_{11} = -K + \overline{A}_{11}\theta^2, \tag{8}$$

$$B_1 = \overline{B}_1\theta^2, \quad B_2 = \overline{B}_2\theta^2, \quad B_3 = \overline{B}_3\theta^2, \tag{9}$$

$$B_{13} = \overline{B}_{13}\theta^3, \quad C_1 = \overline{C}_1\theta, \quad C_2 = \overline{C}_2\theta, \tag{10}$$

where K, $\overline{A}_i$, $\overline{B}_i$ and $\overline{C}_i$ are assumed only to be weakly temperature dependent. The elastic constant $K > 0$ is the usual splay constant which arises in the smectic-A phase elastic energy, given by [3, 4]

$$w_A = \tfrac{1}{2}K(\nabla \cdot \mathbf{n})^2. \tag{11}$$

The magnetic energy density is generally given by [3, p.287]

$$w_m = -\tfrac{1}{2}\chi_a(\mathbf{n} \cdot \mathbf{H})^2 \tag{12}$$

where $\mathbf{H}$ is the magnetic field and χ_a is the magnetic anisotropy of the liquid crystal. When $\chi_a > 0$ the director prefers to align parallel with the magnetic field; $\mathbf{n}$ will tend to align perpendicular to the field when $\chi_a < 0$. We shall assume that $\chi_a > 0$ and that $\mathbf{H}$ is applied in the x direction as shown in Fig.1. In this case, the director will be attracted by the field in such a way that the layers can be expected to distort when $H = |\mathbf{H}|$ is greater than some critical value H_c.

ENERGIES

It is possible to develop the energies given above in terms of the layer displacement $u = u(x, y, z)$. From the work of Kléman and Parodi [12], the usual smectic layer compression energy w_L is given by

$$w_L = \tfrac{1}{2}Bu_z^2. \tag{13}$$

The layer compression constant B describes the elastic resistance to changes in the smectic layer thickness: it is estimated to be of the order 10^7 dynes/cm^2 (*cgs*) in smectic-A, for example [12, 17]. This term remains valid for smectic C also.

In the general derivation of the bulk elastic energy there is a need to retain up to second order in u and its derivatives when working with $\mathbf{a}$ and $\mathbf{c}$ to guarantee that all the correct terms to second order appear in the final (quadratic) energy: this is especially true for geometries other than planar as is the case in the work of Kleman and Parodi [12] for smectic-A. In the planar aligned smectic-A case working to first order in u is sufficient, but in smectic-C more care is needed in, for example, the construction of the magnetic energy density because of the additional symmetries inherent in this phase. Further, this approach lends itself to adaption in later work for smectic-C in non-planar geometries.

Working to second order in u and its derivatives we can set, as in previous work on SmC* liquid crystals subjected to electric fields [18],

$$\mathbf{a} = (-u_x(1+u_z), -u_y(1+u_z), 1-\tfrac{1}{2}(u_x^2+u_y^2)), \quad (14)$$
$$\mathbf{c} = (1-\tfrac{1}{2}(u_x^2+u_y^2), u_y(1+u_z), u_x(1+u_z)+u_y^2), \quad (15)$$

which, from (3), leads to

$$\mathbf{b} = (-u_y(1+u_x+u_z), 1-u_y^2, u_y(1-u_x+u_z)). \quad (16)$$

It is a simple exercise to verify that the smectic-C constraints in equation (2) are then satisfied to second order in u. The quantity $\nabla\times\mathbf{a}$ is small and does not vanish; as mentioned above, this is as anticipated and is analogous to the situation in [12]. The choice of $\mathbf{c}$ is driven by the following observation. Given the geometry of Fig.1, the director appears more likely to align further with the applied magnetic field along the x axis rather than the y axis in the initial stages of reorientation. Looking locally in the xz plane, an initial small perturbation in $\mathbf{c}$ ought to be like $(1,0,u_x)$ when $\mathbf{a}\approx(-u_x,0,1)$ if y dependence is ignored.

Straightforward calculations involving the above versions of $\mathbf{a}$, $\mathbf{b}$ and $\mathbf{c}$ lead to the bulk energy w_b in (5) being reinterpreted as

$$\begin{aligned} w_b = {} & \tfrac{1}{2}A_{12}u_{xx}^2 + \tfrac{1}{2}(B_2+A_{21}+2C_2)u_{yy}^2 + \tfrac{1}{2}B_2u_{zz}^2 + \tfrac{1}{2}B_3u_{yz}^2 \\ & + \tfrac{1}{2}(B_1+B_3-2B_{13})u_{xy}^2 + \tfrac{1}{2}(2B_{13}-B_3-2A_{11}-2C_1)u_{xx}u_{yy} \\ & + (B_2+C_2)u_{yy}u_{zz} + (B_{13}-B_3)u_{xy}u_{yz} + (B_{13}-C_1)u_{xx}u_{xz}. \end{aligned} \quad (17)$$

The integrand expressions in w_b can be simplified following the methods outlined in [3, p.343]. Integrating by parts with respect to x and again with respect to z shows that for any volume Ω

$$\int_\Omega u_{xz}^2\, d\Omega = \int_\Omega u_{xx}u_{zz}\, d\Omega + S_1, \quad (18)$$

where S_1 is a surface contribution to the total energy. Similar terms can also be calculated. Terms which only ever enter the energy when evaluated at a boundary surface can be considered as not influencing the bulk layer orientation: their presence, as noted in [12], would merely shift the total energy by a constant, of no physical significance when the boundary conditions are ignored. Terms involving u_{zz} can be ignored since they will be dominated by Bu_z^2 in w_L. Applying formulae such as the above, omitting the u_{zz} terms and neglecting surface contributions gives

$$\begin{aligned} w_b = {} & \tfrac{1}{2}A_{12}u_{xx}^2 + \tfrac{1}{2}(B_2 + A_{21} + 2C_2)u_{yy}^2 + \tfrac{1}{2}(B_1 - 2(A_{11} + C_1))u_{xx}u_{yy} \\ & + (B_2 - B_3 + B_{13} + C_2)u_{yy}u_{xz} + (B_{13} - C_1)u_{xx}u_{xz}. \end{aligned} \tag{19}$$

Putting $\mathbf{a}$, $\mathbf{c}$ and $\mathbf{H} = (H, 0, 0)$ into (1) and (12) gives, to second order,

$$w_m = -\tfrac{1}{2}\chi_a H^2 \left(\cos(2\theta)u_x^2 + \sin^2\theta(1 - u_y^2) - \sin(2\theta)u_x u_z\right), \tag{20}$$

the contribution in u_x obviously being integrated to an evaluation on the boundaries. Although w_m has an unfamiliar appearance, it is particularly worthwhile to draw attention to the special case for w_m when $\theta = 0$. For $\theta = 0$ the magnetic energy density ought to collapse to that for the smectic-A phase, and indeed

$$w_m = -\tfrac{1}{2}\chi_a H^2 u_x^2, \tag{21}$$

in this case, exactly the form used by de Gennes and Prost [3, p.363] when considering the geometry in Fig.1 for smectic-A with $\chi_a > 0$. The expression for w_m in equation (20) for smectic-C is therefore a natural and meaningful extension to that for smectic-A; further, it is necessary to include the second order terms in $\mathbf{a}$ and $\mathbf{c}$ above, otherwise the special cases just mentioned do not collapse to the smectic-A cases because an additional quadratic contribution would have been overlooked.

To summarize, the total energy density for planar aligned smectic-C in the geometry of Fig.1, ignoring surface contributions, is given via (13), (19) and (20) as

$$\begin{aligned} w &= w_L + w_m + w_b, \\ &= \tfrac{1}{2}Bu_z^2 - \tfrac{1}{2}\chi_a H^2 \left(\cos(2\theta)u_x^2 + \sin^2\theta(1 - u_y^2) - \sin(2\theta)u_x u_z\right) \\ &\quad + \tfrac{1}{2}A_{12}u_{xx}^2 + \tfrac{1}{2}(B_2 + A_{21} + 2C_2)u_{yy}^2 + \tfrac{1}{2}(B_1 - 2(A_{11} + C_1))u_{xx}u_{yy} \\ &\quad + (B_2 - B_3 + B_{13} + C_2)u_{yy}u_{xz} + (B_{13} - C_1)u_{xx}u_{xz}. \end{aligned} \tag{22}$$

The results from (6) can be inserted into w in (22) to find that in the limit $\theta \to 0$ we obtain the related smectic-A bulk energy, namely,

$$w_{SmA} = \tfrac{1}{2}Bu_z^2 - \tfrac{1}{2}\chi_a H^2 u_x^2 + \tfrac{1}{2}K(u_{xx} + u_{yy})^2, \qquad (23)$$

which is discussed in [3, 4, 19]. Equation (23) is also the same energy discussed by de Gennes [20] when changes in density are ignored (for the magnetic field direction considered here). This further highlights w to be a meaningful extension to smectic-C from the smectic-A theory for layer distortions.

We aim to employ periodic solutions which will be averaged over the sample volume, thereby determining critical behaviour via a minimization process: this is the elementary approach adopted in [3,4].

PERIODIC SOLUTIONS

To simplify matters we assume that $u = u(x, z)$, in which case the energy density in equation (22) becomes

$$\begin{aligned} w &= \tfrac{1}{2}Bu_z^2 - \tfrac{1}{2}\chi_a H^2\left[\cos(2\theta)u_x^2 + \sin^2(\theta) - \sin(2\theta)u_x u_z\right] \\ &\quad + \tfrac{1}{2}A_{12}u_{xx}^2 + (B_{13} - C_1)u_{xx}u_{xz}. \end{aligned} \qquad (24)$$

Now let

$$u = u_0 \sin(kx)\sin(qz), \qquad q = \frac{\pi}{d}, \qquad (25)$$

where u_0 is a small constant. For periodic functions f, define the average $\langle f \rangle$ by

$$\langle f \rangle = \frac{1}{P}\int_0^P f(m)\, dm, \qquad (26)$$

where P is the period of f. After averaging over a sample of unit length in y, we have

$$\langle w \rangle = \tfrac{1}{8}u_0^2 k^2\left[B\left(\frac{q}{k}\right)^2 + A_{12}k^2 - \chi_a H^2\cos(2\theta)\right] - \tfrac{1}{2}\chi_a H^2 \sin^2(\theta). \qquad (27)$$

Also, for the undistorted state, $u \equiv 0$ and

$$\langle w(u \equiv 0) \rangle = -\tfrac{1}{2}\chi_a H^2 \sin^2(\theta), \qquad (28)$$

and so a comparison of energies gives

$$\begin{aligned}\Delta\langle w\rangle &= \langle w(u)\rangle - \langle w(u\equiv 0)\rangle \\ &= \tfrac{1}{8}u_0^2k^2\left[B\left(\frac{q}{k}\right)^2 + A_{12}k^2 - \chi_a H^2\cos(2\theta)\right]. \qquad (29)\end{aligned}$$

The above form for $\Delta\langle w\rangle$ is reminiscent of that in [4, p.314]. The critical field H_c is found by minimizing $\Delta\langle w\rangle$ over values of k and then determining the least value of H above which $\Delta\langle w\rangle$ will become negative, indicating the system's preference for adopting the distorted variable solution u rather than the zero solution. The square bracket in (29) can be minimized with respect to k to find that its minimum occurs when $k = k_x$ given by

$$k_x^2 = \sqrt{\frac{B}{A_{12}}}\,q. \qquad (30)$$

(Recall that $A_{12} > 0$ by (6)). For this value of k we have

$$\Delta\langle w\rangle = \tfrac{1}{8}u_0^2k_x^2\left[2\sqrt{BA_{12}}\,q - \chi_a H^2\cos(2\theta)\right]. \qquad (31)$$

The critical magnetic field strength is then given by, using $q = \frac{\pi}{d}$,

$$\chi_a H_c^2\cos(2\theta) = 2\sqrt{BA_{12}}\,\frac{\pi}{d} = 2\pi\frac{A_{12}}{\lambda d}, \qquad (32)$$

where

$$\lambda = \sqrt{\frac{A_{12}}{B}}, \qquad (33)$$

can be introduced as a characteristic length scale to allow comparison with known results for smectic-A.

The results from equations (8) and (9) can be inserted into (32) and (33) to find that as $\theta \to 0$ we have

$$\chi_a H_c^2 = 2\pi\frac{K}{\lambda d}, \qquad \lambda = \sqrt{\frac{K}{B}}, \qquad (34)$$

which is precisely the result in de Gennes and Prost [3] for the critical field at the onset of the Helfrich-Hurault transition in smectic-A, K being the usual splay constant in (11). This shows that the threshold derived in (32) naturally extends the result from the smectic-A case to smectic-C.

DISCUSSION

The results presented here are consistent extensions of the results which are known for the smectic-A phase, as indicated above: the novel energy w introduced in (22) for layer distortions collapses to the known energy for smectic-A given by equation (23), while the critical threshold for the onset of layer distortions in smectic-C, given by equation (32), collapses to the well known result for smectic-A as $\theta \to 0$. The indications are that the onset of layer distortions in smectic-C essentially involves a one-dimensional effect when an observer looks at collections of layers: this is in contrast to the general two-dimensional effects which can occur in the smectic-A phase [3,21]. Comparisons of these results with the work of McKay and Leslie [22], Pleiner *et al* [23] and Blake and Virga [24] are currently in progress while some preliminary results for smectic C^* have been reported recently [18].

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