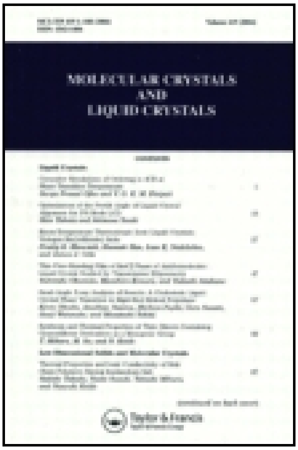


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# A Novel Method for Measuring Compression Constants in Smectics

Fiona Stewart [a] & Iain W. Stewart [a]
[a] Department of Mathematics, University of Strathclyde, Glasgow, Scotland, United Kingdom

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# A Novel Method for Measuring Compression Constants in Smectics

**Fiona Stewart**
**Iain W. Stewart**
Department of Mathematics, University of Strathclyde, Glasgow, Scotland, United Kingdom

*This article considers a refined analysis of the Helfrich–Hurault effect in smectic A when the layer normal* **a** *and the director* **n** *need not coincide. We demonstrate that the Helfrich-Hurault transition threshold decreases as* **a** *and* **n** *decouple. The dependence of this transition upon the elastic coupling constant* $B_1$ *is discussed and its qualitative universal influence upon the reduced threshold is depicted. The implications for the measurement of compression and coupling constants is discussed.*

**Keywords:** compression; Helfrich–Hurault effect; smectic A



## 1. INTRODUCTION

Smectic liquid crystals, in general, are layered structures of molecules which not only possess orientational order, as in nematics, but also positional order within the layers. The common preferred direction to which the molecules tend to align is denoted by the unit vector **n**, called the director. In the smectic A (SmA) phase the molecules generally arrange themselves in equidistantly spaced layers with the director commonly taken to coincide with the SmA layer normal **a**, as shown in Figure 1(a). Motivated by recent work by Auernhammer *et al.* [1–3] and Soddemann *et al.* [4], we consider extending this model by investigating the possibility of the director **n** and the unit layer normal **a** decoupling, which means they need not remain coincident as shown in Figure 1(b). To do this we will use the theory for SmA

Address correspondence to Fiona Stewart, Department of Mathematics, University of Strathclyde, Livingstone Tower, 26 Richmond Street, Glasgow, G1 1XH, Scotland, UK. E-mail: rs.fste@maths.strath.ac.uk

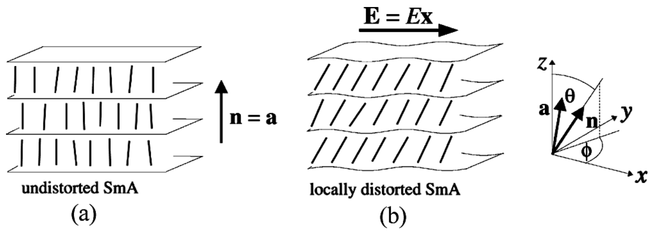


**FIGURE 1** Decoupling the director **n** and the layer normal **a** in the Helfrich–Hurault effect. (a) A planar aligned sample of SmA liquid crystal. The short bold lines are representative of the rod-like molecular structures within the layers. (b) Applying an electric field **E** in the $x$-direction. Beyond some critical electric field threshold the layers begin to distort and **n** and **a** need not precisely coincide. The angle $\theta$ measures the director tilt from the $z$-axis and $\phi$ is the orientation angle of the orthogonal projection of **n** onto the smectic layers.



developed by Stewart [5]. As we apply an electric field parallel to the smectic layers, assuming the dielectric anisotropy $\epsilon_a$ is positive, we expect that as the magnitude of $E$ is increased, there will be a critical threshold at which the director will desire to align itself with the electric field. As the director attempts to achieve this homogeneous alignment, the smectic layers and the director distort and undulate and it is this transition away from a planar alignment that is commonly referred to as the Helfrich–Hurault transition. An undulation indicates that the layers will adopt a wave-like form. Helfrich [6] and Hurault [7] were the first to study such phenomena in the 1970s when they investigated magnetic field effects in infinite samples of cholesteric liquid crystals. Following this work, the classical Helfrich–Hurault transition in SmA has since been reviewed by Chandrasekhar [8] and de Gennes and Prost [9] but only for samples of SmA where **n** and **a** coincide. Other work has been carried out by Geer *et al.* [10], Selinger *et al.* [11] and Singer [12], who examined SmA liquid crystals under the application of an electric field and considered the effects on the layers, such as undulation and buckling. The objective in this article is to analyse how the critical threshold for the onset of the Helfrich–Hurault transition is reduced as **n** and **a** separate.

## 2. GEOMETRICAL SET-UP AND GOVERNING EQUATIONS

We consider a planar aligned sample of SmA liquid crystal where an electric field **E** is applied in the $x$-direction parallel to the layers, as shown in Figure 1(b).

The smectic layers are surfaces that can be described by the function

$$\Phi = z - u(x,y,z), \tag{1}$$

where $u(x,y,z)$ is the displacement of the layers from their original alignment at $\mathbf{a}_0 = (0,0,1)$. The general form of the unit layer normal is given by

$$\mathbf{a} = \frac{\nabla\Phi}{|\nabla\Phi|}, \tag{2}$$

and $\mathbf{n}$ and $\mathbf{a}$ must fulfil the constraints

$$\mathbf{n}\cdot\mathbf{n} = 1, \quad \mathbf{a}\cdot\mathbf{a} = 1. \tag{3}$$

Following [13], the form of $\nabla\Phi$ and $|\nabla\Phi|$ can be approximated to second order in $u$ by

$$\nabla\Phi = (-u_x, -u_y, 1-u_z), \quad |\nabla\Phi| = 1 - u_z + \frac{1}{2}(u_x^2 + u_y^2), \tag{4}$$

where the subscripts denote partial derivatives. Thus the unit layer normal $\mathbf{a}$ can be expressed as

$$\mathbf{a} = \left(-u_x(1+u_z), 0, 1 - \frac{1}{2}u_x^2\right). \tag{5}$$

Given that the unit layer normal and the director may differ, the director $\mathbf{n}$ need not remain parallel to the $z$-direction and so is able to tilt over, making an angle $\theta$ with the $z$-direction. Motivated by Figure 1(b), the form of the director in the distorted state is given by

$$\mathbf{n} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta), \tag{6}$$

where $\phi$ is the orientation angle of the orthogonal projection of $\mathbf{n}$ onto the smectic layers. Since we consider only small disturbances to the director, $|\theta| \ll 1$ and $|\phi| \ll 1$ and so Eq. (6) may be approximated by

$$\mathbf{n} = \left(\theta, 0, 1 - \frac{\theta^2}{2}\right), \tag{7}$$

where both $\mathbf{n}$ and $\mathbf{a}$ have been kept to second order for accuracy. Notice also that neither $\mathbf{a}$ nor $\mathbf{n}$ have any $y$-component to this order of approximation. The form for the electric field $\mathbf{E}$, applied in the $x$-direction, is given by

$$\mathbf{E} = E(1,0,0), \quad E = |\mathbf{E}|. \tag{8}$$

We now wish to consider the energy density given by [5]

$$w_A = \frac{1}{2}K_1^a(\nabla\cdot\mathbf{a})^2 + \frac{1}{2}K_1^n(\nabla\cdot\mathbf{n})^2 + \frac{1}{2}B_1(1-(\mathbf{n}\cdot\mathbf{a})^2) + \frac{1}{2}B_0(|\nabla\Phi| + (\mathbf{n}\cdot\mathbf{a}) - 2)^2 - \frac{1}{2}\epsilon_0\epsilon_a(\mathbf{n}\cdot\mathbf{E})^2, \tag{9}$$

where $K_1^a$ and $K_1^n$ are elastic constants related to layer orientation and splay of the director, respectively. The compression coefficient is $B_0$ and $B_1$ is a measure of the coupling strength between $\mathbf{n}$ and $\mathbf{a}$. Also, within Eq. (9), $\epsilon_0$ is the permittivity of free space and $\epsilon_a$ is the (unitless) dielectric anisotropy. We pay particular attention to the fact that the form for the energy density used in [5] separates out the effects of the director from the layer normal. It is also worth mentioning here that if we set $\mathbf{a}\equiv\mathbf{n}$, $K_1 \equiv K_1^a + K_1^n$ and ignore electric field effects, then the expression for the energy density collapses to the classical form [14]

$$w = \frac{1}{2}K_1(\nabla\cdot\mathbf{n})^2 + \frac{1}{2}B_0(|\nabla\Phi| - 1)^2. \tag{10}$$

Notice that use of this result with (4) and $\mathbf{a}\equiv\mathbf{n}$, as seen in (5), gives

$$w = \frac{1}{2}K_1 u_{xx}^2 + \frac{1}{2}B_0\left(u_z - \frac{1}{2}u_x^2\right)^2, \tag{11}$$

equivalent to the form given by de Gennes and Prost [9].

## 3. HELFRICH–HURAULT TRANSITION BY AVERAGING

We seek an expression for the critical electric field strength required for the Helfrich-Hurault transition to take place. To obtain this we need to consider averaging the energy density. Inserting $\mathbf{a}$, $\mathbf{n}$ and $\mathbf{E}$ from Eqs. (5), (7) and (8) into the energy density (9) gives

$$w_A = \frac{1}{2}K_1^a u_{xx}^2 + \frac{1}{2}K_1^n\theta_x^2 + \frac{1}{2}B_1(u_x+\theta)^2 + \frac{1}{2}B_0 u_z^2 - \frac{1}{2}\epsilon_0\epsilon_a E^2\theta^2. \tag{12}$$

Notice here that the coupling term $B_1$ between the director and the layer normal disappears when $\theta = -u_x$ and we recover the traditional expression for $w_A$. Furthermore, if additionally we set $\mathbf{a}\equiv\mathbf{n}$ and $K_1 \equiv K_1^a + K_1^n$ then Eq. (12) reduces to the classical case as given by (11), with the inclusion of the electric energy density. We wish to consider disturbances to the director and the layers by inserting sinusoidal ansatz of the form

$$u = u_0\sin(q_x x)\sin(q_z z), \quad \theta = \theta_0\cos(q_x x)\sin(q_z z), \quad q_z = \frac{\pi}{d}, \tag{13}$$

where $|u_0| << 1$ and $|\theta_0| << 1$ are constants, into Eq. (12). These forms take account of strong anchoring on the boundaries, that is, on the boundaries $z = 0$ and $z = d$, where $d$ is the sample thickness, there are no disturbances to the director or to the layers. Also, $q_z = \pi/d$ corresponds to the anticipated first mode between the boundary plates.

We wish to average $w_A$ and so we introduce the average $\langle f \rangle$ for a periodic function of period $P$ defined by [15, p. 288]

$$\langle f \rangle = \frac{1}{P} \int_0^P f(m)dm, \tag{14}$$

where we note that $\langle \sin^2 \rangle = \langle \cos^2 \rangle = \frac{1}{2}$. Having inserted the ansatz given in (13) into the energy density (12) and averaging the resulting expression by use of (14), we obtain

$$\langle w(u,\theta) \rangle = \frac{1}{8}[K_1^a u_0^2 q_x^4 + K_1^n \theta_0^2 q_x^2 + B_1(u_0 q_x + \theta_0)^2 + B_0 u_0^2 q_z^2 - \epsilon_0 \epsilon_a E^2 \theta_0^2]. \tag{15}$$

From this we are able to make a direct comparison between the distorted state, where the director has tilted and the layers have deformed, and the undistorted state, where the layers are unaffected and the tilt angle of the director relative to the layer normal is zero. To do this we consider the change in the averaged energy density $\triangle\langle w \rangle$ given by $\triangle\langle w \rangle = \langle w(u,\theta) \rangle - \langle w(u \equiv 0, \theta \equiv 0) \rangle = \langle w(u,\theta) \rangle$. Thus we see that, in this instance, the change in the averaged energy density is in fact equal to the averaged energy density given by (15). From this expression it is seen that as $E$ changes in magnitude, the distorted solution becomes energetically preferable when the averaged energy density $\triangle\langle w \rangle$ first becomes negative and so the critical electric field strength for the onset of any transition occurs at the value of $E$ for which $\triangle\langle w \rangle = 0$. Applying this to Eq. (15) leads to a critical field that must satisfy

$$\epsilon_0 \epsilon_a E^2 = K_1^n q_x^2 + B_1 + \frac{1}{\theta_0^2}[K_1^a u_0^2 q_x^4 + B_1(u_0^2 q_x^2 + 2u_0 q_x \theta_0) + B_0 u_0^2 q_z^2]. \tag{16}$$

It only remains to minimise the right-hand side of [16] with respect to $\theta_0$ and $q_x$. Minimising with respect to $\theta_0$ we are able to determine the value of $\theta_0$, in relation to the other parameters, at the critical threshold. This gives

$$\theta_0 = -\frac{(K_1^a u_0 q_x^4 + B_1 u_0 q_x^2 + B_0 u_0 q_z^2)}{B_1 q_x}, \tag{17}$$

which can then be substituted into [16], yielding the result

$$\epsilon_0 \epsilon_a E^2 = K_1^n q_x^2 + B_1 - \frac{B_1^2 q_x^2}{K_1^a q_x^4 + B_1 q_x^2 + B_0 q_z^2}. \tag{18}$$

If we now minimise this result with respect to $q_x$, we obtain the expression

$$\begin{aligned} K_1^n (K_1^a)^2 q_x^8 + 2K_1^n K_1^a B_1 q_x^6 + (K_1^n B_1^2 + 2K_1^n K_1^a B_0 q_z^2 + B_1^2 K_1^a) q_x^4 \\ + 2K_1^n B_1 B_0 q_z^2 q_x^2 = B_1^2 B_0 q_z^2 - K_1^n B_0^2 q_z^4, \end{aligned} \tag{19}$$

which clearly has non-zero real solutions for $q_x$ only when

$$B_1^2 B_0 q_z^2 - K_1^n B_0^2 q_z^4 > 0, \quad q_z = \frac{\pi}{d}. \tag{20}$$

This being the case, we are able to place a restriction on $B_1$ such that [18] subject to a solution for $q_x$ in [19] is valid, namely,

$$B_1 > \sqrt{K_1^n B_0}\,\frac{\pi}{d} \equiv B_1^c. \tag{21}$$

The left-hand side of [19] is a polynomial in $q_x$; it is clear that it is strictly increasing for $q_x > 0$ and equal to zero at $q_x = 0$. Hence, provided $B_1$ satisfies the inequality [21], we can conclude that there exists a unique, positive valued $q_x$, say $q_x^c$, such that $q_x^c$ satisfies [19]. Consequently, putting the value $q_x = q_x^c$ into [18] delivers the actual critical threshold $E_c$, i.e.

$$\epsilon_0 \epsilon_a E_c^2 = K_1^n (q_x^c)^2 + B_1 - \frac{B_1^2 (q_x^c)^2}{K_1^a (q_x^c)^4 + B_1 (q_x^c)^2 + B_0 q_z^2}. \tag{22}$$

In general, $q_x^c$ must be found numerically for a given set of material parameters $K_1^n$, $K_1^a$, $B_0$, $B_1$, given the complexity of the expression [19]. Consequently, solving [19] for $q_x^c$ delivers the final critical threshold when inserted into [22]. The threshold value $B_1^c$ defined in Eq. [21] is the minimum value of $B_1$ for which this process outlined above for obtaining $E_c$ is valid.

We have now obtained an expression for the critical electric field strength required for the Helfrich–Hurault transition to take place in an uncoupled system of SmA liquid crystal in terms of the elastic constants $K_1^a$, $K_1^n$, the coupling coefficient $B_1$, the layer compression $B_0$ and the wave vectors $q_x$ and $q_z$. It is worth mentioning here that standard Euler-Lagrange theory can be used for coupled second order systems and gives exactly the same results as above.

We first note that when $\theta = -u_x$ and $K_1^a + K_1^n = K_1$ in $w_A$, given by [12], the averaged energy [15] can be replaced by (cf. [15, p. 288])

$$\langle w(u) \rangle = \frac{u_0^2}{8}\left[K_1 q_x^4 + B_0 \frac{\pi^2}{d^2} - \epsilon_0 \epsilon_a E^2 q_x^2\right]. \tag{23}$$

which leads to the classical Helfrich-Hurault transition threshold, denoted by $E_{cc}$, via a minimisation over $q_x$. The result is ([15, p. 289] and [9, p. 363])

$$\epsilon_0 \epsilon_a E_{cc}^2 = 2\pi \frac{K_1}{\lambda d}, \quad \lambda = \sqrt{\frac{K_1}{B_0}}, \tag{24}$$

where $\lambda$ is a characteristic length scale, with the corresponding classical critical wave number given by

$$q_x^{cc} = \sqrt{\frac{\pi}{\lambda d}}. \tag{25}$$

The result given by Eq. (22) can be interpreted more effectively by plotting the dependence of $E_c$ upon $B_1$, especially in relation to the classical threshold $E_{cc}$. This is best accomplished by non-dimensionalising $E_c$ and $B_1$. The non-dimensionalisation of the electric field takes place via the classical threshold $E_c$ by setting $\overline{E}_c = E_c/E_{cc}$: the classical threshold then corresponds to $\overline{E}_c = 1$, allowing a comparison of our results with those from the classical setting. Similarly, $B_1$ can be non-dimensionalised (cf. Eq. (18)) by introducing $\overline{B}_1 = B_1/\epsilon_0\epsilon_a E_{cc}^2 = B_1 \lambda d / 2\pi K_1$ by Eq. (24). The result is shown in Figure 2 where it is clear that, when considering the Helfrich–Hurault transition in SmA for decoupled **n** and **a**, the coupling term $B_1$ between the director and the layer normal must, from a theoretical perspective, be greater than $\overline{B}_1^c$, obtained via [21], before any critical electric field effects can take place. The material parameters used to obtain the result displayed in Figure 2 are given by ([15, p. 330] and [9, p. 420])

$$K_1^n = K_1^a = 2.5 \times 10^{-12}\,\mathrm{N}, \quad B_0 = 5 \times 10^7\,\mathrm{Nm}^{-2}, \quad d = 10^{-4}\,\mathrm{m}. \tag{26}$$

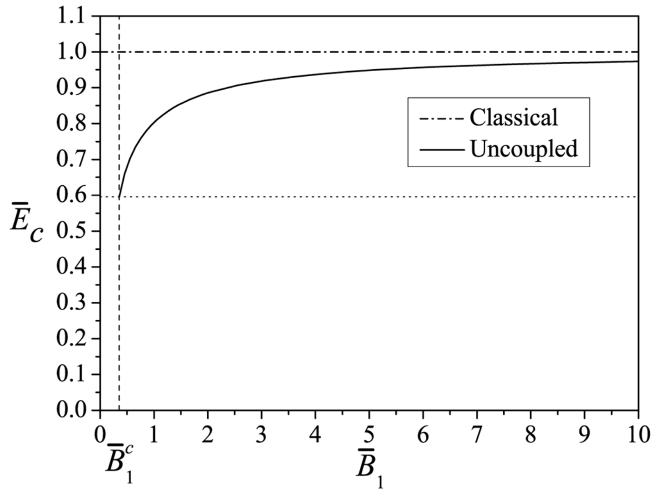


**FIGURE 2** The graph obtained from Eq. (22) showing the influence of the coupling term $\overline{B}_1$ between the director and the layer normal normalised upon the critical electric field strength $\overline{E}_c$, as defined in the text. The bars represent the non-dimensionalised terms, adopted to produce a 'universal' curve. It is clear from the bold curve that having identified a particular critical electric field strength, $\overline{E}_c$ say, the corresponding coupling term, $\overline{B}_1$ is easily identified. The material paremeters used are stated in Eq. (26). $B_1^c$ is as stated at Eq. (21) and $\overline{B}_1^c = B_1^c \lambda d / 2\pi K_1$.

For these parameters, it is clear that the necessary inequality [21] is satisfied. Thus, if $\overline{E}_c$ is obtained experimentally by observation, then $\overline{B}_1$ can be determined from Figure 2 and therefore a novel method for finding $B_1$ experimentally can be achieved.

We first note that for the data in [26], $E_{cc}$ can be calculated from Eq. (24) to be $E_{cc} \approx 12.66\ \mathrm{V\,\mu m^{-1}}$ where we have set $\epsilon_0 = 8.854 \times 10^{-12}\ \mathrm{F\ m^{-1}}$ and chosen $\epsilon_a = 0.7$. As examples, using Eq. (22), the values stated in [26] and the aforementioned $\epsilon_0$ and $\epsilon_a$, when $\overline{B}_1 = 1$, $E_c \approx 10.16\ \mathrm{V\,\mu m^{-1}}$ and when $\overline{B}_1 = 10$, $E_c \approx 12.32\ \mathrm{V\,\mu m^{-1}}$. (These results can be confirmed in Figure 2, noting that $\overline{E}_c \approx 0.80$ and $\overline{E}_c \approx 0.97$ when $\overline{B}_1 = 1$ and $\overline{B}_1 = 10$, respectively.) From Figure 2 we also notice that for lower values of $\overline{B}_1$, the actual magnitude of the critical electric field strength required for the Helfrich–Hurault transition to take place in a decoupled sample of SmA is considerably less in comparison with the classical case. This is to be expected since if we decouple **n** and **a**, we allow the director **n** more independence, that is, it is easier for it to tilt in an attempt to align with the direction of the applied electric field. Thus, the critical threshold for the electric field at which this change in the director's position will occur, causing the layers to distort and undulate, will be lower than that given when **n** and **a** are fixed and parallel to each other. It is also clear from Figure 2 that $\overline{E}_c \to 1$ as $\overline{B}_1 \to \infty$, i.e. $E_c$ tends to the classical threshold (where **n** and **a** coincide) as the coupling strength $B_1 \to \infty$.

Further information can be obtained by solving Eq. (19) for $q_x$, the result of which is depicted in Figure 3, where the effect of $B_1$ on the wave number $q_x$ is shown. Again the terms have been non-dimensionalised: $B_1$ as before using $\overline{B}_1 = B_1\lambda d/2\pi K_1$ by Eq. (24) and $q_x$ from [19] by introducing $\bar{q}_x^c = q_x^c/q_x^{cc} = q_x\sqrt{\lambda d/\pi}$ by Eq. (25). Similar to before, the classical threshold then corresponds to $\bar{q}_x^c = 1$. Thus, if $q_x^c$ may be obtained experimentally then $B_1$ can be determined from Figure 3. This curve exhibits similar features to Figure 2. For lower values of $\overline{B}_1$, the actual magnitude of the critical wave numbers observed in the $x$-direction when the Helfrich–Hurault transition takes place in a decoupled sample of SmA is found to be considerably less than that of the classical case. It is also clear that $\bar{q}_x^c \to 1$ as

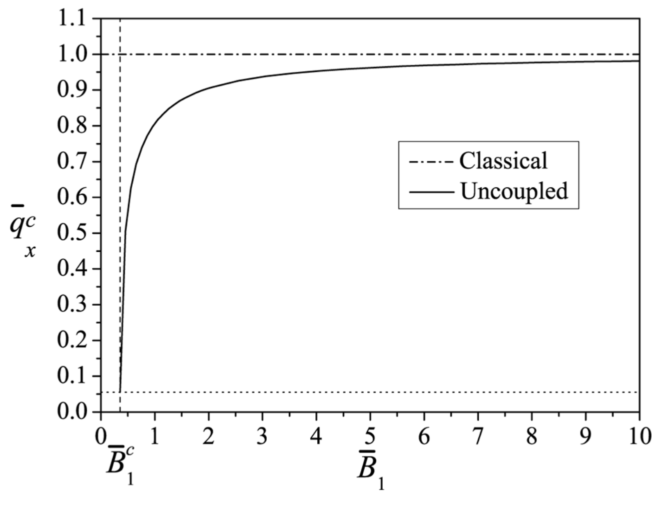


**FIGURE 3** The graph obtained from Eq. (19) having solved it for $q_x$. This result shows the influence of the normalised coupling term $\overline{B}_1$ between the director and the layer normal upon the dimensionless critical wave number $\overline{q}_x^c = q_x^c/q_x^{cc}$, $q_x^{cc}$ is as stated at Eq. (25). The bars represent the non-dimensionalised terms, adopted to produce a 'universal' curve. It is clear from the bold curve that having identified a particular critical wave number, $\overline{q}_x^c$ say, the corresponding coupling term, $\overline{B}_1$, is easliy identified. The material parameters used are stated in Eq. (26).

$\overline{B}_1 \to \infty$, i.e. $q_x^c$ tends to the classical threshold (where $\mathbf{n}$ and $\mathbf{a}$ coincide) as the coupling strength $B_1 \to \infty$.

## 4. DISCUSSION

In relation to the problem considered here there are two things which are physically relevant from an experimental perspective: the applied electric field strength and the periodicity $P$ of the undulations observed, related to the wave number by $P = 2\pi/q_x^c$. These quantities, in relation to the effect the coupling term $B_1$ has on them, are discernable from the results within this article and are depicted in Figures 2 and 3. In future work we will consider the dynamical aspects of this problem by incorporating flow into the set-up. The Helfrich–Hurault effect is generally transient: the layers will not remain distorted indefinitely but will desire to return to the undistorted state. The relaxation time-dependence of this problem is to be investigated. Ben-Abraham and Oswald [16] considered a similar situation when they examined the time dependence of an undulation instability in SmA. We will consider the ideas developed within their article by use of the more general dynamic theory for SmA introduced by Stewart [5].



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