University of Strathclyde Glasgow

Helfrich–Hurault Effects in Smectic A Liquid Crystals

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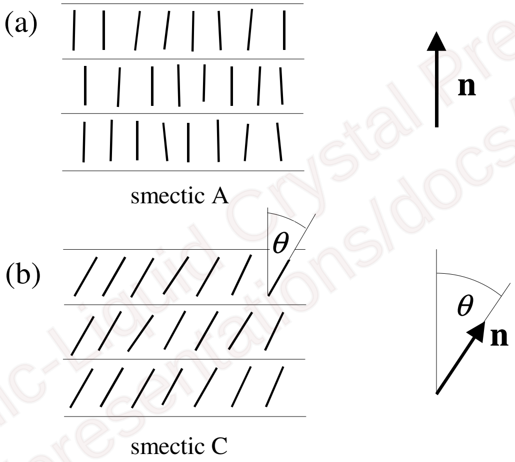
Outline of Talk

- Introduction and Motivation
- Classical Helfrich–Hurault transition
- A novel investigation of the Helfrich–Hurault transition
- Conclusions and Future Plans

Smectic Liquid Crystals



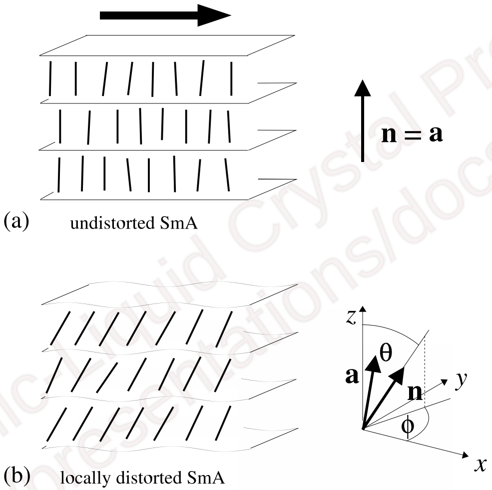
Motivation

G.K. AUERNHAMMER, H. BRAND and H. PLEINER
Shear-induced instabilities in layered liquids, Phys. Rev. E, **66**, 061707, (2002)

An important reference since

- **n** and **a** have been decoupled (no longer the same)
- $\nabla \times \mathbf{a}$ is no longer forced to be zero
- The linearised equations coincide with those of the more extensive nonlinear dynamic theory of Stewart (*Dynamic theory for smectic A liquid crystals*, Cont. Mech. and Thermodyn., **18**(6), (2007))

Set-up of Auernhammer's Problem



$$\Phi = z - u(x,y,z,t), \quad \mathbf{a} = \frac{\nabla \Phi}{|\nabla \Phi|}$$

$$\mathbf{n} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta), \quad \mathbf{a} = (-u_x, -u_y, 1)$$

$$\mathbf{v} = (\dot{\gamma} z, 0, 0)$$

$\dot{\gamma}$: shear rate

u : displacement of the layers

Governing dynamic equations (IWS)

$$n_i n_i = 1, \quad a_i a_i = 1, \quad v_{i,i} = 0$$

$$\rho \dot{v}_i = -\tilde{p}_{,i} + \tilde{g}_k^n n_{k,i} + \tilde{t}_{ij,j} + |\nabla \Phi| a_i J_{j,j}$$

$$\left(\frac{\partial w}{\partial n_{i,j}} \right)_{,j} - \frac{\partial w}{\partial n_i} + \tilde{g}_i^n + \mu n_i = 0$$

$$\dot{\Phi} = -\lambda_p (\nabla \cdot \mathbf{J})$$

$$\tilde{g}_i^n = -2(\lambda_1 D_i^a + \lambda_4 A_i)$$

$$\begin{aligned}\mathbf{J} = &K_1^a \frac{\nabla(\nabla\cdot\mathbf{a})}{|\nabla\Phi|} - K_1^a[\nabla(\nabla\cdot\mathbf{a})\cdot\mathbf{a}]\frac{\mathbf{a}}{|\nabla\Phi|} + B_1\frac{(\mathbf{n}\cdot\mathbf{a})}{|\nabla\Phi|}[\mathbf{n}-(\mathbf{n}\cdot\mathbf{a})\mathbf{a}] \\ &- B_0[|\nabla\Phi| + (\mathbf{n}\cdot\mathbf{a}) - 2]\left[\mathbf{a} + \frac{\mathbf{n}}{|\nabla\Phi|} - \frac{(\mathbf{n}\cdot\mathbf{a})}{|\nabla\Phi|}\mathbf{a}\right]\end{aligned}$$

$$\begin{aligned}w = \frac{1}{2}K_1^a(\nabla\cdot\mathbf{a})^2 + \frac{1}{2}K_1^n(\nabla\cdot\mathbf{n})^2 + \frac{1}{2}B_1(1-(\mathbf{n}\cdot\mathbf{a})^2) \\ + \frac{1}{2}B_0(|\nabla\Phi| + (\mathbf{n}\cdot\mathbf{a}) - 2)^2 - \frac{1}{2}\epsilon_0\epsilon_a(\mathbf{n}\cdot\mathbf{E})^2\end{aligned}$$

B_0: Layer compression

B_1: Coupling between director and layer normal

K_1: Bending modulus of the layers

Dynamic Equations

$\mathbf{v} = (\dot{\gamma} z, 0, 0)$ satisfies linear momentum equations and enables us to determine the pressure, p.

The remaining equations for angular momentum and permeation reduce to the following:

$$\left(\frac{\lambda + 1}{2} - \lambda \sin^2(\theta_0) \right) \dot{\gamma} = \frac{B_1}{\gamma_1} \sin(\theta_0) \cos(\theta_0) + \frac{B_0}{\gamma_1} \sin(\theta_0)[1 - \cos(\theta_0)],$$

Dynamic Equations cont.

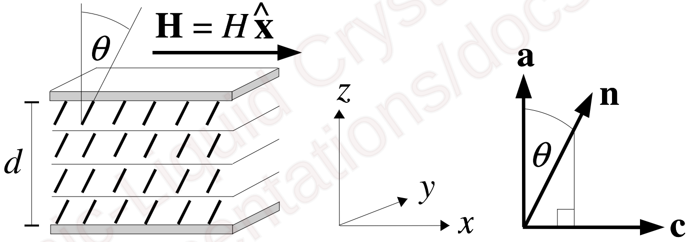
$$A_\theta \left[2\dot{\gamma}\lambda \sin(\theta_0)\cos(\theta_0) + \frac{B_0}{\gamma_1}\left[\sin^2(\theta_0) - \cos^2(\theta_0) + \cos(\theta_0)\right] - \frac{B_1}{\gamma_1}[\sin^2(\theta_0) - \cos^2(\theta_0)] \right] - A_u \frac{B_0}{\gamma_1}\sin(\theta_0) q_z = 0,$$

$$A_\phi \frac{1}{2}\dot{\gamma}(\lambda + 1) - A_u \left[q_x \frac{B_0}{\gamma_1}\frac{1-\cos(\theta_0)}{\cos(\theta_0)} + \frac{B_1}{\gamma_1} q_x \right] = 0,$$

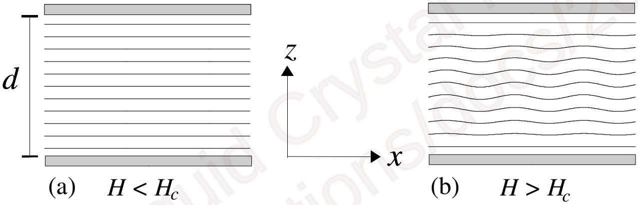
$$A_\theta \lambda_p B_0 \sin(\theta_0) q_z$$
$$+ A_\phi \lambda_p [B_1 q_x \sin(\theta_0)\cos(\theta_0) + B_0 q_x \sin(\theta_0)(1 - \cos(\theta_0))]$$
$$+ A_u \lambda_p [B_0 q_x^2 (1 - \cos(\theta_0))^2 - B_1 q_x^2 \cos^2(\theta_0) - K_1 q_x^4 - B_0 q_z^2] = 0.$$

Helfrich–Hurault transition

The transition from planar layers to undulating (or distorted) layers

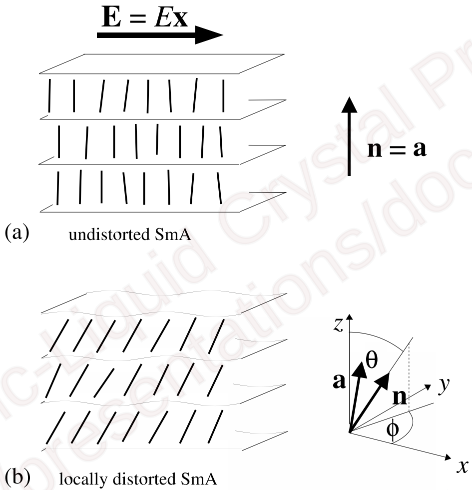


Beyond a particular threshold, the smectic layers desire to undulate (or distort). This is called the critical threshold



Notice that below this threshold, the smectic layering remains unchanged.

Uncoupling n and a in the Helfrich–Hurault effect



Set-up of problem

$$\Phi = z - u(x,y,z), \quad \mathbf{a} = \frac{\nabla\Phi}{|\nabla\Phi|}, \quad \mathbf{n}\cdot\mathbf{n} = 1, \quad \mathbf{a}\cdot\mathbf{a} = 1$$

$$\boxed{\mathbf{a} = (-u_x - u_x u_z, 0, 1 - \frac{1}{2}u_x^2), \quad \mathbf{n} = \left(\theta, 0, 1 - \frac{\theta^2}{2}\right), \quad \mathbf{E} = E(1,0,0)}$$

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

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

Euler Lagrange equations

The energy equation is of the form:

$$W = \int_V w_A(\theta, \theta_x, u_x, u_z, u_{xx}) dV$$

We minimise the energy via the first Gateaux variation

$$\frac{dW}{d\epsilon}((\theta, u) + \epsilon(h_1, h_2))\bigg|_{\epsilon=0} = 0$$

Integrating using first principles gives us

$$\boxed{\frac{\partial}{\partial x}\left(\frac{\partial w_A}{\partial \theta_x}\right) - \frac{\partial w_A}{\partial \theta} = 0}$$

$$\boxed{\frac{\partial^2}{\partial x^2}\left(\frac{\partial w_A}{\partial u_{xx}}\right) - \frac{\partial}{\partial x}\left(\frac{\partial w_A}{\partial u_x}\right) - \frac{\partial}{\partial z}\left(\frac{\partial w_A}{\partial u_z}\right) = 0}$$

Euler Lagrange equations for θ and u

$$K_1^n \theta_{xx} - B_1 u_{xx} - B_1 \theta + \epsilon_0 \epsilon_a E^2 \theta = 0$$
$$K_1^a u_{xxxx} - B_1 u_{xx} - B_1 \theta_x - B_0 u_{zz} = 0$$

Using $\theta = \theta_0 e^{i(q_x x + q_z z)}$ and $u = u_0 i e^{i(q_x x + q_z z)}$ gives us

$$-K_1^n \theta_0 q_x^2 + B_1 u_0 q_x - B_1 \theta_0 + \epsilon_0 \epsilon_a E^2 \theta_0 = 0$$
$$K_1^a u_0 q_x^4 + B_1 u_0 q_x^2 - B_1 \theta_0 q_x + B_0 u_0 q_z^2 = 0$$

From the first equation,

$$u_0 = \frac{\theta_0}{B_1 q_x}(K_1^n q_x^2 + B_1 - \epsilon_0 \epsilon_a E^2).$$

Substituting into the other equation and some re-arranging gives

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

an expression for the critical electric field strength

Averaging the energy

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

Using $u = u_0\sin(q_x x)\sin(q_z z)$, $\theta = \theta_0\cos(q_x x)\sin(q_z z)$ $(q_z = \frac{\pi}{d})$ and averaging the energy gives us

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

(since $\langle\sin^2\rangle = \langle\cos^2\rangle = \frac{1}{2}$)

$$\begin{aligned}\triangle\langle w\rangle &= \langle w(u,\theta)\rangle - \langle w(u\equiv 0,\theta\equiv 0)\rangle\\ &= \langle w\rangle\\ &= \frac{1}{8}\Big[K_1^a u_0^2 q_x^4 + K_1^n \theta_0^2 q_x^2 + B_1(u_0 q_x + \theta_0)^2\\ &\qquad + B_0 u_0^2 q_z^2 - \epsilon_0\epsilon_a E^2\theta_0^2\Big]\end{aligned}$$

As E changes in magnitude, the distorted solution becomes energetically preferable when $\triangle\langle w\rangle < 0$ and so the critical electric field strength occurs at $\triangle\langle w\rangle = 0$. Using this in the above equation we obtain

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

Recall the result obtained using the Euler Lagrange approach:

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

The result determined so far by averaging:

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

Minimising this expression w.r.t θ_0 determines the minimum to occur when

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

The critical electric field strength is given by

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

where $q_z = \frac{\pi}{d}$.

Minimising the expression for the electric field w.r.t. q_x gives us

$$K_1^n (K_1^a)^2 q_x^8 + 2K_1^n K_1^a B_1 q_x^6 + (K_1^n B_1^2 + 2K_1^n K_1^a B_0 q_z^2 + B_1^2 K_1^a) q_x^4 + 2K_1^n B_1 B_0 q_z^2 q_x^2 = B_1^2 B_0 q_z^2 - K_1^n B_0^2 q_z^4,$$

which has non-zero real solutions only if

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

thus giving a restriction on B_1 such that

$$\boxed{B_1 > \sqrt{K_1^n B_0} \frac{\pi}{d} \equiv B_1^c}$$

Provided B_1 satisfies the inequality, we can conclude that there exists a unique, positive valued q_x, say q_x^c, such that q_x^c satisfies the polynomial for q_x. Consequently, putting the value $q_x = q_x^c$ into the expression for the critical electric field delivers the actual critical threshold E_c, i.e.

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

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.

Observations on the energy density

We first note that when $\theta = -u_x$ and $K_1^a + K_1^n = K_1$ in w_A, recall the form of which given by

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

the averaged energy can be replaced by

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

which leads to the classical Helfrich–Hurault transition threshold, denoted by E_{cc}, via a minimisation over q_x.

The result (given by Stewart and de Gennes) is

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

where λ is a characteristic length scale.

The corresponding classical critical wave number is given by

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

Non-dimensionalisation

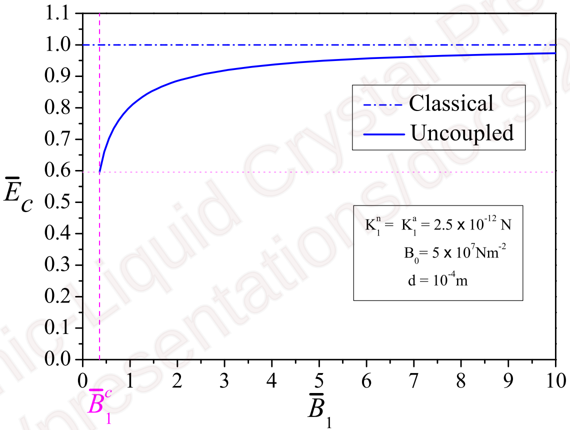
The non-dimensionalisation of the electric field takes place via the classical threshold E_c by setting

$$\boxed{\overline{E}_c = E_c/E_{cc}}$$

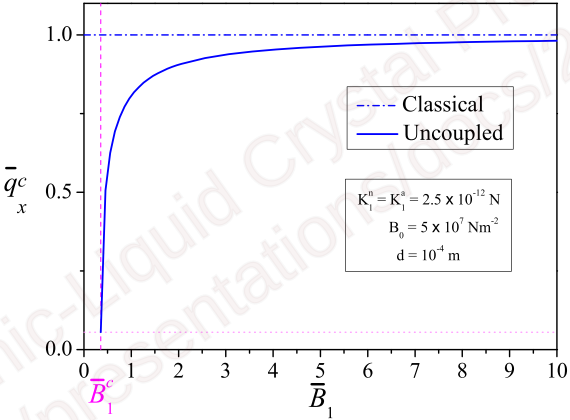
the classical threshold then corresponds to $\overline{E}_c = 1$, allowing a comparison of our results with those from the classical setting. Similarly, B_1 can be non-dimensionalised by introducing

$$\boxed{\overline{B}_1 = B_1/\epsilon_0\epsilon_a E_{cc}^2 = B_1\lambda d/2\pi K_1}$$

Universal Curve 1: $\overline{B}_1$ against $\overline{E}_c$



Universal Curve 2: $\overline{B}_1$ against $\overline{q}_x^c$



Conclusions

- The graphs provide a mechanism for identifying and measuring B_1 experimentally.
- For the Helfrich–Hurault transition to take place in a decoupled sample of SmA

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

- For low values of $\overline{B}_1$, the actual magnitude of the critical electric field strength and critical wave number are considerably less in comparison to their respective classical cases.

- E_c and q_x^c tend to the classical electric field threshold and classical critical wave number as the coupling strength $B_1 \to \infty$ when **n** and **a** coincide.
  i.e. $\overline{E}_c \to 1$ and $\overline{q}_x^c \to 1$ as $\overline{B}_1 \to \infty$.
- 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 and are depicted in universal curves shown.

Possible Future Work

- Consider the dynamical aspects of this problem by incorporating flow and/or transverse flow into the set-up
- Use of a more extensive viscous stress
- Helfrich–Hurault is generally a transient effect: the layers will not remain distorted indefinitely but will desire to return to undistorted state. What is the relaxation time dependence of this problem?

Acknowledgements

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Thank you for your attention