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Two-Dimensional Modelling of a Freely Suspended Smectic Film

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We present a two-dimensional model for a smectic film suspended horizontally between two thin electrodes attached to glass plates. The film is bordered vertically by air and is subject to an electric field. Our two-dimensional model allows for spatial variations in the electric field, and predicts the existence of anticlinic or synclinic equilibrium director profiles. Governing equations describing the equilibrium behaviour of the electric potential and the director have been derived by minimizing the free energy of the system. The equations have been solved numerically, and for a fixed set of parameters we are able to obtain two different types of solution profile for our system, corresponding to anticlinic and synclinic configurations.

Keywords: numerical modeling; polarization; smectic films

INTRODUCTION

When the temperature of a free-standing smectic liquid crystal film is reduced towards the SmA^*-SmC^* phase transition, the layers at the free surfaces can transform into the low-temperature SmC^* phase well above the bulk transition [1,2]. Two contrasting director configurations can often be observed in these smectic films, namely synclinic and anticlinic structures [3–6]. An anticlinic structure exhibits a director tilt in opposing directions at the free surfaces, while in a synclinic configuration the film has surfaces aligned in the same direction. The presence of these structures has been attributed to the application of an electric field, although Link *et al.* [7] report that anticlinic surface structures can also occur when no field is present.

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We consider a two-dimensional model for a smectic film suspended horizontally between two thin electrodes attached to glass plates. The film is bordered vertically by air and is subject to an electric field. Previous analyses of polarization effects in smectic films [8,9] employed a continuum theory that allows variation in the smectic layer spacing and the molecular tilt [10]. Significantly, they also assumed that any electric field applied across the cell is constant in direction and magnitude. Our two-dimensional model incorporates a phenomenological description for the director tilt and allows for spatial variations in the electric field. By minimizing the system's total free energy, we are able to calculate equilibrium profiles for the electric potential and the director. For a fixed set of parameters the system exhibits two different types of equilibrium structure, corresponding to anticlinic and synclinic configurations. We determine the energetically preferred director structures as the applied voltage, polarization strengths and temperature are allowed to vary.

MODEL

We consider a thin SmA* liquid crystal film of width d and height h, as illustrated in Figure 1. The film is suspended horizontally between two thin electrodes of negligible thickness attached to glass plates of width d_g. The smectic layers are assumed to lie parallel to the free surfaces and a voltage is applied across the width of the film. The film and plates are bounded above and below by air regions of height h_a. The thickness of the film is much less than the helical pitch of the chiral molecules so that any inherent chirality may be neglected. We assume that the behaviour of the director is constrained to lie in

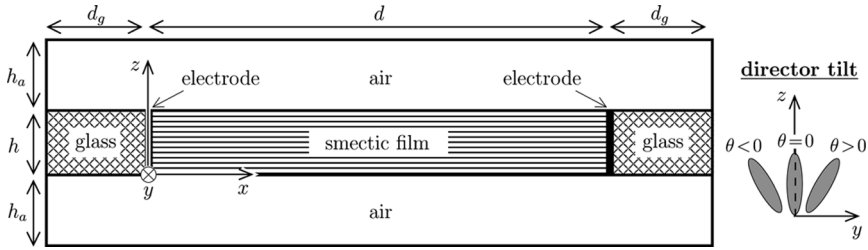

FIGURE 1 The smectic film of width d and height h is suspended between two glass plates of width d_g and air regions of height h_a. The director tilt is measured relative to the z-direction and may take negative values.

the *yz*-plane. Millar [11] examines the alternative configuration where the director lies in the *xz*-plane. In both models the electric field has components in the x and z-directions, while the spontaneous polarization is perpendicular to the director. Therefore, spontanteous polarization does not play a role in the *xz*-director calculations found in [11]. Otherwise, the results for the two director configurations are qualitatively similar. The director tilt θ is measured as the variation in the director with respect to the vertical coordinate, z, therefore the tilt angle can take negative values, as illustrated in Figure 1. There is no variation of the director tilt in the y-direction, so the director $\underline{n}$ can be represented as

$$\underline{n} = (0, \sin\theta(x,z), \cos\theta(x,z)).$$

As mentioned previously, we allow for spatial variations in the electric field, $\underline{E}$. In doing so, the electric field and the associated displacement, $\underline{D}$, must satisfy Maxwell's equations in the absence of magnetic fields and free charges,

$$\nabla \times \underline{E} = \underline{0}, \qquad \nabla \cdot \underline{D} = 0. \tag{1}$$

We assume that any variation in the electric field will occur in the x and z-directions across the width of the film. Therefore, there exists an electric potential, $U(x, z)$, such that

$$\underline{E}(x,\ z) = -\nabla U = (-U_{,x}\,,\ 0,\ -U_{,z}),$$

where $U_{,x}$ represents $\partial U/\partial x$. The displacement and the electric field in a dielectric material can be related via $\underline{D} = \epsilon_0\ \underline{\underline{\hat{\epsilon}}}\ \underline{E}$, where ϵ_0 is the permittivity of free space and $\underline{\underline{\hat{\epsilon}}}$ is the appropriate relative dielectric tensor [12]. For our model we have three independent dielectric tensors, one for each medium (liquid crystal, glass and air).

The total free energy of our system, W, is given by

$$W = \int_0^h \int_0^d w_{\mathrm{LQ}}\ \mathrm{d}x\mathrm{d}z + \iint_{\text{glass}} w_{\mathrm{G}}\ \mathrm{d}x\mathrm{d}z + \iint_{\text{air}} w_{\mathrm{A}}\ \mathrm{d}x\mathrm{d}z + \int_0^d (w_{\mathrm{S}}|_{z=0} + w_{\mathrm{S}}|_{z=h})\mathrm{d}x, \tag{2}$$

where we have incorporated the two glass plates and air regions above and below the film. Energy densities w_{LQ}, w_{G}, w_{A} and w_{S}

corresponding to the smectic film, glass, air and the free surfaces, respectively, will be introduced in the following sections.

By minimizing W with respect to the director tilt θ and electric potential U, we can derive governing differential equations for the entire system. These Euler-Lagrange equations will involve θ and U in the smectic film region, but only the potential U in the glass and air. (The equations for the potential could also be obtained by substituting the appropriate electric displacement $\underline{D}$ for each region into Maxwell's equation $\nabla \cdot \underline{D} = 0$.) The system of differential equations will be solved subject to boundary conditions introduced in a later section.

SMECTIC FILM REGION

The bulk energy density for the liquid crystal, w_{LQ}, comprises three components: elastic (w_{elas}); thermotropic (w_{th}) and electric (w_{elec}). We consider a simplified energy density that incorporates elastic deformations in the x and z directions [15],

$$w_{\mathrm{elas}} = \frac{1}{2}\left(K_x(\theta_{,x})^2 + K_z(\theta_{,z})^2\right), \tag{3}$$

where K_x and K_z are (non-negative) elastic constants. This energy density is clearly minimized when the director tilt angle is constant throughout the cell. The thermotropic energy density is derived via a Landau expansion [16] (approximated to order 4) of the true thermotropic energy functional in the director tilt θ,

$$w_{\mathrm{th}} = \frac{1}{2}\alpha\Delta T\theta^2 + \frac{1}{4}b\theta^4. \tag{4}$$

The Landau coefficients α and b are positive, and $\Delta T = T - T_{AC}$ is the temperature measured from T_{AC}, the SmA* to SmC* transition temperature. As a first approximation, we have omitted any high-order terms that arise due to polarization. Given that α and b are strictly positive, when $\Delta T > 0$ (in the SmA* phase) or $\Delta T = 0$ (at the transition temperature) the only possible energy minimum for w_{th} is $\theta = 0$.

In the liquid crystal region the electric field induces a polarization, $\underline{P}$. Therefore, we have to account for contributions from both the electric field and the polarization in the electric displacement [12]. By transforming the (diagonal) dielectric tensor in the molecular frame [11,12], we derive the dielectric tensor for the liquid crystal in

the Cartesian frame,

$$\underline{\underline{\epsilon}}_{\mathrm{LQ}} = \begin{bmatrix} \epsilon_1 & 0 & 0 \\ 0 & \epsilon_2 + (\epsilon_3 - \epsilon_2)\sin^2\theta & (\epsilon_3 - \epsilon_2)\sin\theta\cos\theta \\ 0 & (\epsilon_3 - \epsilon_2)\sin\theta\cos\theta & \epsilon_3 + (\epsilon_2 - \epsilon_3)\sin^2\theta \end{bmatrix} \tag{5}$$

The coefficients ϵ_1 and ϵ_2 are the relative dielectric permittivities perpendicular to the long axis of the molecule, while ϵ_3 is the dielectric permittivity parallel to the long axis. The dielectric anisotropies $\epsilon_3 - \epsilon_1$ and $\epsilon_3 - \epsilon_2$ determine the behaviour of the director under the influence of an electric field.

An effective polarization, $\underline{P} = \underline{P}_{\mathrm{sp}} + \underline{P}_{\mathrm{fl}}$, is introduced as the combination of a spontaneous polarization $\underline{P}_{\mathrm{sp}}$ and $\underline{P}_{\mathrm{fl}}$, the contribution due to flexoelectricity. We adopt the spontaneous polarization [13]

$$\underline{P}_{\mathrm{sp}} = P_s(\underline{n}\cdot\underline{\hat{a}})\underline{n}\times\underline{\hat{a}} = (P_s\cos\theta\sin\theta, 0, 0), \tag{6}$$

where $\underline{\hat{a}} = (0,0,1)$ is a unit vector normal to the smectic layers and P_s is the magnitude of the spontaneous polarization. We choose a flexoelectric polarization resembling that for a tilted uniaxial,

$$\begin{aligned} \underline{P}_{\mathrm{fl}} &= e_{11}\underline{n}\,(\nabla\cdot\underline{n}) + e_{33}(\nabla\times\underline{n})\times\underline{n} \\ &= -\theta_{,z}\,(0, e_{11}\sin^2\theta + e_{33}\cos^2\theta,\ [e_{11}+e_{33}]\sin\theta\cos\theta), \end{aligned} \tag{7}$$

where e_{11} and e_{33} are coefficients for splay and bend similar to those found in the study of nematics [14]. Although not given explicitly here, we can now combine (5), (6) and (7) in order to form the electric displacement for the liquid crystal,

$$\underline{D}_{\mathrm{LQ}} = \epsilon_0\,\underline{\underline{\epsilon}}_{\mathrm{LQ}}\underline{E} + \underline{P}.$$

The electric energy density for the smectic film is now [14]

$$\begin{aligned} w_{\mathrm{elec}} = -\int_{\underline{0}}^{\underline{E}} \underline{\epsilon}_{\mathrm{LQ}}\cdot\mathrm{d}\underline{E} = -\frac{\epsilon_0}{2}\left(\epsilon_1(U_{,x})^2 + [\epsilon_3 + (\epsilon_2-\epsilon_3)\sin^2\theta](U_{,z})^2\right) \\ + U_{,x}P_s\cos\theta\sin\theta - U_{,z}\,(e_{11}+e_{33})\theta_{,z}\sin\theta\cos\theta. \end{aligned} \tag{8}$$

Finally, the total bulk energy density for the liquid crystal film is $w_{\mathrm{LQ}} = w_{\mathrm{elas}} + w_{\mathrm{th}} + w_{\mathrm{elec}}$, i.e., the sum of the elastic, thermotropic and electric energy densities derived in (3), (4) and (8), respectively.

GLASS AND AIR REGIONS

In the (homogeneous) glass, the dielectric tensor is given by $\underline{\underline{\hat{\epsilon}}}_{\mathrm{G}} = \epsilon_G \underline{\underline{I}}$ where ϵ_G is the relative dielectric permittivity of the glass and $\underline{\underline{I}}$ is the identity tensor. Similarly, the dielectric tensor for the air region is $\underline{\underline{\hat{\epsilon}}}_{\mathrm{A}} = \epsilon_A \underline{\underline{I}}$ where ϵ_A is the relative dielectric permittivity of air. Thus, the electric displacement in the glass and air may be written as, respectively,

$$\underline{D}_{\mathrm{G}} = \epsilon_0 \epsilon_G (-U_{,x},\ 0,\ -U_{,z}), \qquad \underline{D}_{\mathrm{A}} = \epsilon_0 \epsilon_A (-U_{,x},\ 0,\ -U_{,z}). \tag{9}$$

The electric energy densities for the glass, w_{G}, and air, w_{A}, can now be expressed as

$$w_{\mathrm{G}} = -\int_0^{\underline{E}} \underline{D}_{\mathrm{G}} \cdot d\underline{E} = -\frac{1}{2}\epsilon_0 \epsilon_G (U_{,x}^2 + U_{,z}^2), \tag{10}$$

$$w_{\mathrm{A}} = -\int_0^{\underline{E}} \underline{D}_{\mathrm{A}} \cdot d\underline{E} = -\frac{1}{2}\epsilon_0 \epsilon_A (U_{,x}^2 + U_{,z}^2). \tag{11}$$

BOUNDARY CONDITIONS

There are numerous boundaries in our geometry where we must impose appropriate conditions. To begin, we consider boundary conditions for the director tilt. When the temperature of a free standing liquid crystal film is reduced towards the SmA*-SmC* phase transition, the layers at the free surface transform into the low temperature SmC* phase at a temperature well above the bulk transition. To model this expected behaviour, we introduce a Rapini-Papoular type [17] surface energy density $w_{\mathrm{S}} = \omega \sin^2(\theta^2 - \theta_w^2)$, where ω is the anchoring strength and θ_w is a prescribed pretilt angle. This surface energy is clearly minimized when $\theta = \pm\theta_w$. Therefore, if the chosen pretilt angle is non-zero the director alignment at the free surfaces may be non-homeotropic. The weak anchoring boundary conditions that we impose on θ at the free surfaces are [12]

$$\frac{\partial w_{\mathrm{LQ}}}{\partial \theta_{,z}} = \frac{\partial w_{\mathrm{S}}}{\partial \theta} \quad (z = 0) \qquad \text{and} \qquad \frac{\partial w_{\mathrm{LQ}}}{\partial \theta_{,z}} = -\frac{\partial w_{\mathrm{S}}}{\partial \theta} \quad (z = h).$$

At the interface between the glass/electrodes and the liquid crystal film, for a temperature above the SmA*-SmC* transition, we expect the director tilt angle to be close to zero across most of the film. Thus,

at the intereface between the liquid crystal and the glass/electrodes, we impose the conditions

$$\theta(0,\ z) = \theta(d,\ z) = 0, \qquad z \in (0,\ h). \tag{12}$$

The potential difference across the liquid crystal film is determined by the conditions set at the smectic-glass interfaces. We have chosen a positive difference by introducing the boundary conditions

$$U(0,\ z) = -V/2, \qquad U(d,\ z) = V/2, \qquad z \in (0,h), \tag{13}$$

for $V > 0$. At the remaining internal boundaries between the glass, air and liquid crystal we assume continuity of the normal component of the electric displacement $\underline{D}$.

The conditions imposed on the potential U on the outer boundaries of the computational region are more difficult to choose. Our calculations are carried out on a finite domain, therefore we need to choose appropriate outer boundary conditions that model the unbounded space surrounding the liquid crystal. We adopt a method involving perfectly matched layers (PMLs), artificial boundaries that avoid the necessity of meshing large, empty regions where a solution is relatively unimportant. Millar [11] adopts a more simplistic approach where the normal component of the electric field is assumed to be zero on the outer boundaries. However, it is also shown in [11] that when the size of the glass and air regions are sufficiently large, the predicted behaviour of the smectic film is largely unaffected by the choice of outer boundary conditions.

EQUILIBRIUM DIRECTOR PROFILES AND ENERGY COMPARISONS

As mentioned previously, minimization of the total energy W defined in (2) leads to a system of ordinary differential equations in the tilt θ and potential U. These we solve, in conjunction with the appropriate boundary conditions, using the numerical package COMSOL Multiphysics [18]. (More details about the Euler-Lagrange equations and the computational method can be found in Millar [11].) The following parameter values are fixed in all our calculations:

$$\epsilon_0 = 8.854 \times 10^{-12}\mathrm{C}^2\mathrm{N}^{-1}\mathrm{m}^{-2},\ \epsilon_G = 4,\ \epsilon_A = 1,\ \epsilon_1 = 10,\ \epsilon_2 = 15,$$
$$\epsilon_3 = 5,\ \theta_w = 25^\circ, K_x = 10^{-11}\mathrm{N},\ K_z = 10^{-11}\mathrm{N},\ \alpha = 10^3\mathrm{N}\mathrm{K}^{-1}\mathrm{m}^{-2},$$
$$b = 10^6\mathrm{N}\mathrm{m}^{-2}, \omega = 0.005\mathrm{N}\mathrm{m}^{-1}.$$

Constants ϵ_G, ϵ_A, ϵ_1, ϵ_2 and ϵ_3 are unitless as they are measured relative to ϵ_0, while the magnitude of ω invokes relatively weak anchoring at the boundaries. For brevity, all length and height measurements that follow are assumed to be in microns, μm. For the computational region we choose $d = 1$, $h = 1000$, $d_g = 250$, $h_a = 10$.

By varying the "initial guess" for the director tilt provided to COMSOL while keeping all parameter values fixed, we are able to obtain two very different types of solution for the differential equations that govern the entire system. The two director configurations are demonstrated in Figure 2 where we have taken a cross-section of the film at $x = 500$. The choice of temperature ΔT ensures that we are in the SmA phase with $\theta \approx 0^\circ$ in the majority of the film for the anticlinic profile, but with SmC ordering close to the free surfaces. The boundary angle differs from the prescribed pretilt angle ($\theta_w = 25^\circ$) due to the weak anchoring strength chosen. The tilt is also close to, although not exactly, zero in the majority of the film in the synclinic case.

In Figure 3 we have taken the vertical cross-section for the electric potential U at $x = 100$ for an anticlinic director profile, and the horizontal cross-section along the centre of the film at $z = 0.5$. In and around the film the potential is generally constant in the vertical direction for both director structures, except close to the free surfaces. The regions of significant vertical variation in U close to the free surfaces $z = 0$ and $z = h$ also coincide with large director tilt gradients,

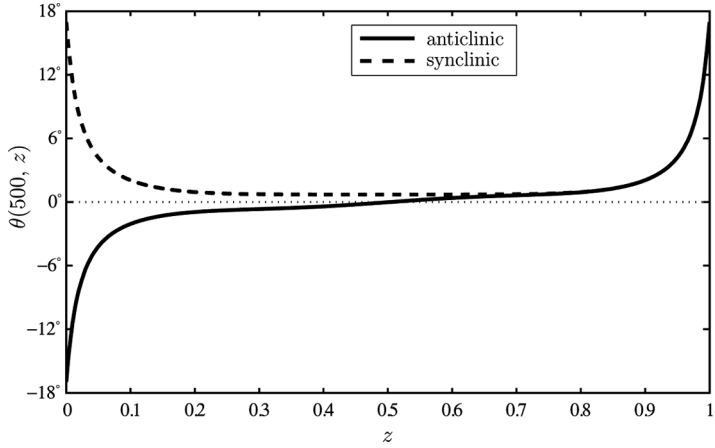

FIGURE 2 Cross-section of the director tilt at $x = 500$ illustrating the anticlinic and synclinic director tilt profiles. $P_s = 5 \times 10^{-5}\mathrm{Cm}^{-2}$, $e_{33} = 3 \times 10^{-11}\,\mathrm{Cm}^{-1}$, $e_{11} = 2e_{33}$, $V = 10\,\mathrm{V}$ and $\Delta T = 1\,\mathrm{K}$.

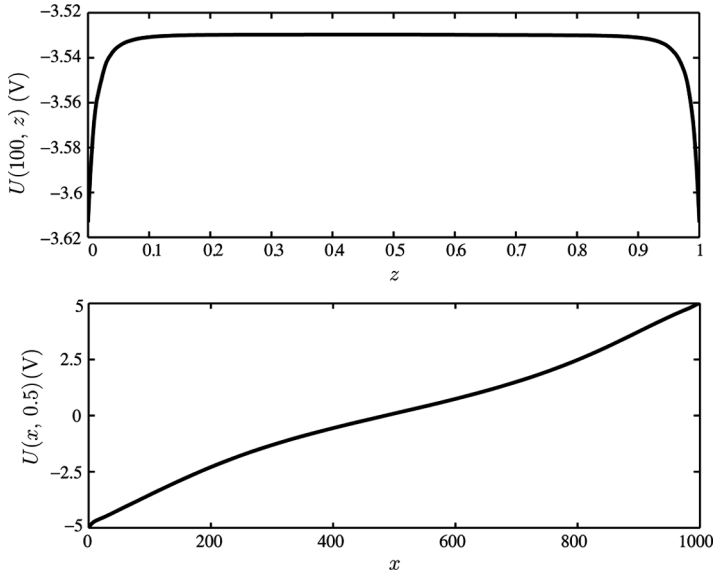

FIGURE 3 Vertical cross-section at $x = 100$ and horizontal cross-section at $z = 0.5$ illustrating spatial variations of the electric potential U for an anticlinic director profile. $P_s = 5 \times 10^{-5}\,\mathrm{Cm}^{-2}$, $e_{33} = 3 \times 10^{-11}\,\mathrm{Cm}^{-1}$, $e_{11} = 2e_{33}$, $V = 10\,\mathrm{V}$ and $\Delta T = 1\,\mathrm{K}$.

as seen in Figure 3. The localized changes in U are a result of the interaction between the electric field and the director tilt, especially via the polarization components in electric energy density (8). We also observe from Figure 3 that the electric potential is non-linear in the horizontal direction.

To find which director configuration is preferred energetically for our chosen parameters, we can compute the total free energy (2) associated with each structure. Generally, it is the competition between the spontaneous and flexoelectric contributions to the electric energy density for the smectic film (8) that determines which structure is lower in energy. If we increase the magnitude of the spontaneous polarization while keeping the applied voltage, temperature and flexoelectric coefficients fixed, the nature of the energetically preferred director structure changes from anticlinic to synclinic. Alternatively, for a fixed spontaneous polarization there is a transition at a critical voltage, V. The preferred director configuration can also change as the film temperature is reduced towards the phase transition at $\Delta T = 0\,\mathrm{K}$ or as the flexoelectric coefficients vary. Figures 4 and 5 represent phase diagrams illustrating the energetically preferred director

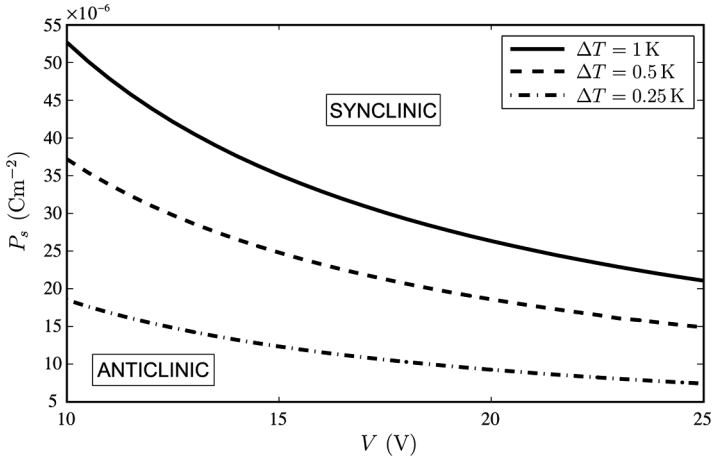

FIGURE 4 Critical spontaneous polarization magnitudes as the applied voltage and sample temperature are allowed to vary. The flexoelectric coefficients are $e_{33} = 10^{-12}\,\mathrm{Cm}^{-1}$ and $e_{11} = 2e_{33}$.

profiles in various parameter regimes. In both figures we have chosen three temperatures above the SmA*-SmC* transition, $\Delta T = 0.25$, 0.5 and 1 K. When the flexoelectric or spontaneous polarization coefficients

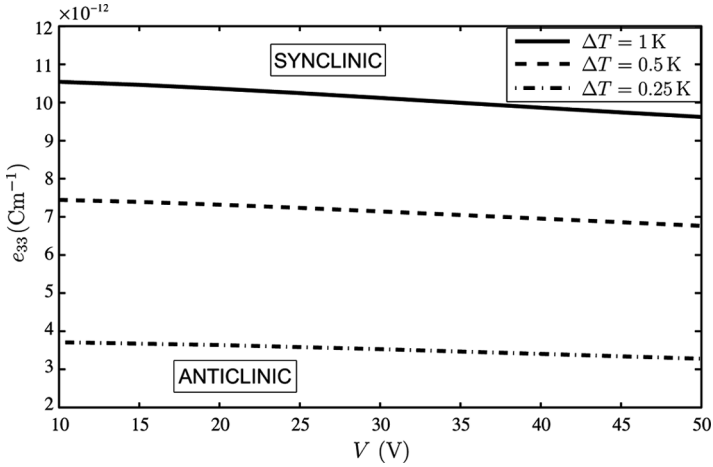

FIGURE 5 Critical magnitudes for the flexoelectric polarization e_{33} as the applied voltage and sample temperature are allowed to vary. The flexoelectric coefficient $e_{11} = 2e_{33}$ and $P_s = 5 \times 10^{-5}\,\mathrm{Cm}^{-2}$.

are relatively small, we find that the anticlinic director structures are preferred energetically over the synclinic structures. Anticlinic structures are also favoured at small voltages. Conversely, synclinic director profiles prevail for larger voltages and polarization strengths. We also find that if we reduce the temperature towards the transition temperature of $\Delta T = 0\,\mathrm{K}$, synclinic structures dominate over a wider range of polarizations and voltages.

DISCUSSION

We have modelled a two-dimensional, freely suspended smectic film. The film was suspended horizontally between two thin electrodes attached to glass plates and surrounded by air. Our model also incorporated a spatially varying electric field. Governing equations to describe the electric potential and director behaviour in the glass, air and liquid crystal film were derived via minimization of the total free energy. We found that varying the key parameters within the system led to a transition in the energetically favoured director alignment. In particular, when the magnitude of the spontaneous polarization and the applied voltage were relatively small, the anticlinic regime exhibited the lowest energy. However, by reducing the sample temperature or the potential difference it was seen that synclinic structures dominated over a wider range of spontaneous polarization strengths.

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