Theory of layer structure in ferroelectric liquid crystal devices in applied electric fields

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We propose a model for the free energy of a ferroelectric liquid crystal formed by cooling a sample from the smectic-*A* phase between parallel substrates. Under these circumstances the smectic layers may deform into V-shaped structures known as chevrons. Application of a strong electric field causes the layers to return to a flat shape, but this can occur in a number of ways. In the model presented here, it is a parameter related to the layer compression modulus that is the principal factor in determining the nature of the field-induced transition from chevrons to flat layers. When this parameter is large, the transition is sudden, but when it is small the chevron first takes on a rounded form before flattening. At intermediate values the tip of the chevron first flattens, and then this flat region gradually grows to encompass the entire layer.

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I. INTRODUCTION

When a ferroelectric liquid crystal in a device with planar anchoring is cooled from a smectic-*A* (Sm-A) phase to a smectic-*C* phase, a phenomenon known as ''chevronning'' generally occurs. This describes a transition from a geometry in which the smectic layers lie in planes containing the substrate normal (the so-called bookshelf geometry) to a state in which each layer is transformed to a V-shaped structure [1]. This is thought to occur because of some constraints on the motion of the liquid crystal molecules. The molecules in contact with the substrates are to some extent pinned to the positions they occupied in the Sm-*A* phase. This means that, unless defects are formed in the layer structure, the number of layers remains constant. If the volume of liquid crystal within the device is also constant, the volume of each layer must remain unchanged. This is a very restrictive condition on the layer shape.

The application of an electric field to a ferroelectric liquid crystal tends to restore the bookshelf geometry [2–5]. The way in which this happens, however, appears to depend on the type of phase and material involved. In this paper we explore the mechanism by which this transition might occur by studying the response of some model systems to an applied electric field.

After some remarks on the experimental observations and previous theoretical approaches we describe a model of the layer system in a ferroelectric liquid crystal, and list possible contributions to the free energy. Minimization of this free energy then yields the layer shape as a function of applied field and of the various parameters of the model. The results of this study suggest that it is one particular term in the free energy that is dominant in determining the way in which a sharp chevron transforms to a flat layer.

II. EXPERIMENTAL DETAILS

When a sufficiently large electric field is applied to a device containing an antiferroelectric (Sm-C^*_{AF}), intermediate (ferrielectric), or ferroelectric (Sm-C^*) liquid crystal, the chevron structure is transformed into a bookshelf geometry. This phenomenon has been reported in detail for surface stabilized ferroelectric liquid crystal (SSFLC) devices, and in such cases is known as high-field treatment [6]. A slightly different transformation is observed in liquid crystals that exhibit both Sm-C^* and Sm-C^*_{AF} phases, although such systems have been studied by relatively few authors [4,7,8]. Small-angle x-ray scattering experiments on devices have provided the most detailed information about both the initial layer geometry within a device and its transformation to a bookshelf structure as the applied electric field is gradually increased. In all of the systems that have been studied, the highest temperature liquid crystal phase is the Sm-*A* phase and the experiments involve cooling through this phase to the temperature of interest in order to achieve the best possible monodomain alignment. The shrinkage of the layer thickness as a consequence of the occurrence of molecular tilt at the Sm-*A* to Sm-C^* transition always resulted in the formation of a chevron structure that included a small bookshelf component, presumably arising as a flattening of the vertex of the chevron. The proportion of bookshelf structure in the device decreased as the temperature decreased, causing the layer structure in the Sm-C^*_{AF} phase to be almost purely chevron.

The transformation to a bookshelf structure occurs in different ways in the Sm-C^*, Sm-C^*_{AF}, and intermediate phases. In the Sm-C^* phase in some materials [4,7], application of an electric field causes a gradual growth of the bookshelf component within a layer at the expense of the chevron structure until finally a fully bookshelf geometry is achieved. No threshold is observed for this phenomenon, the bookshelf component increasing as soon as a field is applied. In the antiferroelectric phase, the transformation to a bookshelf structure occurs at a well-defined field with a sharp transition. In these same materials, layer structure associated with the intermediate phases (also known as ferrielectric or Sm-C^*_{FI1} and Sm-C^*_{FI2}) also deforms at fields greater than a distinct threshold, though in this case the chevron curves just above the transition, before a completely bookshelf geometry is reached. The fact that the layers curve during the transition is deduced from observation of an x-ray Bragg peak at all

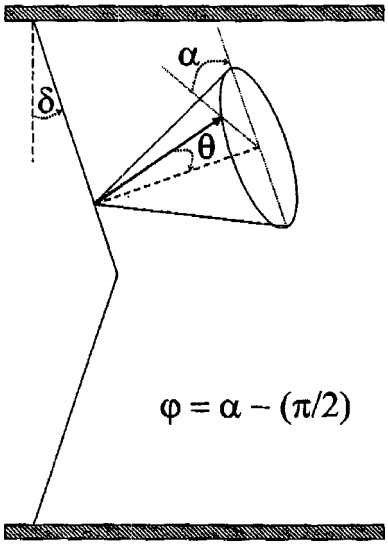

FIG. 1. The three angles that specify the orientation of the director.

rocking angles considered for fields slightly above the low-field threshold. In one other material, however, it has been reported that the characteristics in the ferroelectric and ferrielectric phases were reversed, with Sm-C^* exhibiting curvature and Sm-C^*_{FI2} showing flattening [5].

It is worth noting that in all cases the change in thickness of the layers, measured normal to the local layer surface, appears to be small, being limited to about one percent. This at first seems to be inconsistent with the principle that the volume of a layer should be conserved if the number of layers and the volume of the device remain constant. However, the expected thickness variation of a layer is also small, so this discrepancy may not be significant. Further, above the transition to a bookshelf structure, an in-plane chevron structure does not necessarily occur, though there is evidence of a broadening of the distribution of the layers in the plane of the device [4]. This latter point is in contrast to high-field treatment in SSFLC devices, where the transformation of the chevron to bookshelf structure always appears to occur together with the formation of an in-plane chevron (the so-called striped texture).

III. PRIOR THEORETICAL WORK

Previous theoretical studies of the effect of electric fields on the chevron structure in ferroelectric liquid crystals have generally been based on the concept of minimization of some type of free energy. This is expressed in terms of the angle δ between substrate and layer normal, the angle θ between layer normal and director **n**, and the azimuthal angle ϕ, as illustrated in Fig. 1.

The early work of Clark and co-workers [9,10] involved models in which δ was held at a fixed angle $\pm\delta_0$ in each part of the chevron, but in which the azimuthal angle ϕ varied with position while θ was constant; their system thus consisted of two flat planes meeting at an angle $2\delta_0$. Sabater *et al.* [11] developed a more general model by building on earlier work by Nakagawa and co-workers [12–14]. The angles δ and ϕ were allowed to vary with position, thus permitting a curved layer shape, but the cone angle θ remained constant in space. They took into account the interaction between the spontaneous polarization and the internal electric field, but did not discuss the influence of an external electric field. At about the same time, Limat [15] suggested a model in which two parameters were introduced that measured the degree of departure from the ''nematic'' or uniaxial approximation. In the limit in which the layer tilt angle δ was small in comparison with the cone angle θ, which was assumed to be a constant, this model gave the same results as Nakagawa's model, and in the purely nematic limit it reduced to that of Clark *et al.* This differed from the approach of De Meyere *et al.* [16] who had allowed both δ and θ to vary, but fixed the cosine of the cone angle to be proportional to the cosine of the tilt angle, putting $\cos\theta = \nu\cos\delta$ with ν being a constant less than unity. Čopič and co-workers [17] proposed a model in which the three angles were independent variables, and were not restricted to small values. They used a Landau-de Gennes free energy that contained three smectic and one nematic elastic constants to study the chevron structure, and found conditions for bistability in the chevron configuration. They also discussed the temperature dependence of the chevron shape and the threshold temperature for chevron formation. A later work [18] included the effect of a weak external electric field on the director position on the cone, and studied the switching dynamics between the two possible stable director states, but did not include the effects of electric fields sufficiently strong to modify the chevron shape or to destroy the chevrons altogether.

In the work that follows we investigate the effects of strong electric fields in modifying the shape of the chevron. We take a different approach from that used in many previous studies in that we take the existence of smectic layers as given, and concentrate on the equilibrium layer configuration. This differs from theories that start from an isotropic continuum and then observe the formation of smectic layers through the variation of a density variable $\rho(\mathbf{r})$. While such approaches are closer to first principles than our work, they are equilibrium theories, and as such cannot easily take into account the barrier that hinders the passage of molecules between layers. We find this barrier to be an essential feature in giving rise to a term in the effective free energy that attempts to conserve the volume of each individual smectic layer.

IV. MODEL

In attempting to model the behavior of a smectic liquid crystal, the first question to answer is the choice of variables in terms of which the system can be described. Ideally, these should all be measurable quantities, but this is rarely possible. For example, it may be feasible to measure the orientation of the smectic layer normal relative to the substrate, but the distribution of orientations of the liquid crystal molecules may not be accessible to measurement.

We consider a cell of thickness L in which a ferroelectric

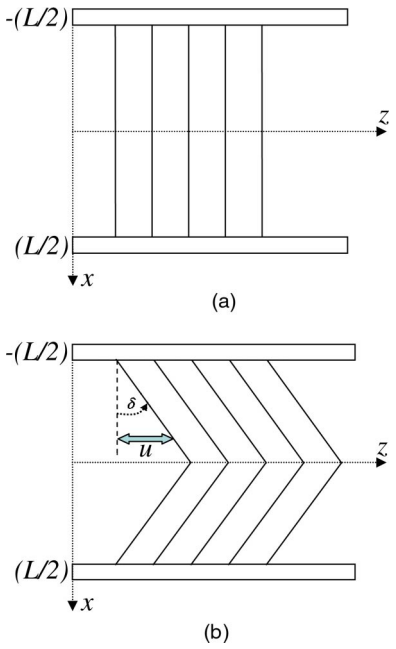

FIG. 2. (a) The bookshelf geometry. (b) The chevron geometry.

liquid crystal is contained between surfaces at $x=\pm L/2$. We assume that in the Sm-*A* phase the smectic layers lie in the *x*-*y* plane, and thus have their normals in the *z* direction. This is the bookshelf geometry, as shown in Fig. 2(a). On passing into other smectic phases the layers in general distort. We take the position of the boundary between two given adjacent layers to be a well-defined quantity, and denote it by $u(x,y)$. The angle δ between the layer and the *x*-*y* plane is then given by $\tan\delta=-\partial u/\partial x$, as in Fig. 2(b).

To describe the internal structure of a smectic layer is a harder task. For rigid molecules the distribution $f(\theta,\phi,\psi)$ of the three Eulerian angles would suffice, but for flexible molecules having internal degrees of freedom the picture is more complicated. In order to develop a tractable model to form the basis of computations we need to condense this complicated picture into a single quantity which we identify as the director. This might be the direction of the time-averaged orientation of the principal axis of inertia that corresponds to the smallest moment of inertia of the molecule. The local order is then partially described by the declination angle θ and the azimuthal angle ϕ.

This, however, is not sufficient to describe the interaction of an electric field with the material. The transition from Sm-*A* to Sm-C^* is accompanied by a loss of cylindrical symmetry in the rotation of a molecule about its axis. A third quantity, related to the remaining Euler angle ψ, is required to describe the preferred orientation of the molecule, and an additional quantity is required to indicate the magnitude of this asymmetry. In some previous work it has been assumed that the electric dipole moment resulting from this asymmetry is proportional to $\sin\theta$, the sine of the tilt angle. We note that the dipole strength will also be affected by the strength *E* of the applied electric field. In the present work we assume that the dipole moment is oriented perpendicularly to both the director and the layer normal.

We now specialize to the case of a system in which the positions of the layers at the surface of the substrates are not altered by the transition from Sm-*A* to Sm-C^* or the application of a modest electric field. This implies that the system does not exhibit any chevrons in the horizontal *y*-*z* plane. The layer displacement in the *z* direction, $u(x)$, is then a function only of *x*. The variables in terms of which we describe the state of a layer are now the displacement $u(x)$ and its derivative $\partial u/\partial x=-\tan\delta$, the tilt angle $\theta(x)$, and the azimuthal angle $\phi(x)$. The fact that there is a single function *u* for all smectic layers ensures that the layer volume is conserved, and thus implicitly assumes no passage of molecules between layers.

The model as described above has been derived from a physical picture of the ferroelectric phase, and is not obviously applicable to antiferroelectric or ferrielectric phases. One may, however, interpret the ''layers'' in the above model as being composite layers consisting of two actual layers in the antiferroelectric phase and three or four in the ferrielectric phases. The physical justification for the model is then less compelling, but there may still be some validity as a phenomenological model of these phases.

V. CONTRIBUTIONS TO THE FREE ENERGY

We can divide the contributions to the free energy into two broad categories. The first category contains no spatial derivatives, but represents the stress induced when two or more conflicting forces act on a molecule, and prevent it from reaching the orientation that would minimize the energy of each contribution individually. The second category represents various forms of elastic energy, and all terms in it contain some sort of spatial derivative of at least one of the variables.

In the first category we start with the energy cost for the tilt angle θ of a molecule to deviate from its preferred value θ_0. In previous work this has sometimes been taken to vary as $(\cos\theta-\cos\theta_0)^2$ or some related polynomial expression in θ and θ_0 [19]. The difficulty here is that in order for such a potential to give a reasonable energy difference between $\theta=0$ and $\theta=\theta_0$, an enormous energy difference between $\theta=0$ and $\theta=\pi/2$ would be required. The reason for this is that when $\theta_0=18°$, for example, and $\theta=0$ then $(\cos\theta-\cos\theta_0)^2=0.002$. The energy at $\theta=\pi/2$ would thus be about 500 times greater than at $\theta=0$. Because liquid crystal molecules have large aspect ratios, it is to be expected that there is a significant energy barrier to rotation of even a few degrees. Once the rotation exceeds this value, there will be little further contribution to the energy, as the molecular axis is now not oriented at any small angle to the liquid crystal director. We thus require a form for the potential that will rise rapidly as a molecule is first rotated away from the director orientation, but will then flatten out, and not rise very much more. The potential $-a\exp[-c(\cos\theta-\cos\theta_0)^2]$ satisfies this criterion, as it varies little once the exponent has become significantly greater than unity. To make the potential equal to $-0.1a$, for example, when $\theta_0=14°$ and $\theta=0$ requires *c* to take on the surprisingly large value of 2600. In our calculations we set *c* equal to 2000. We thus have for this

contribution to the free energy

$$\mathcal{F}_1 = -a\int_{-L/2}^{L/2}\exp[-c(\cos\theta-\cos\theta_0)^2]dx$$

with a and c being constants.

The next term arises from the stress caused by the constraint that a layer must maintain constant volume. In the Sm-A phase, both δ and θ vanish, and thus $\delta=\theta$. As the system enters the Sm-C^* phase, θ becomes nonzero, which reduces the layer thickness. Chevron formation then increases the effective length of the layer to compensate for the reduced thickness. For a uniform chevron angle δ, the length is increased by a factor of sec δ, while the thickness is correspondingly reduced by a factor of cos δ. In the absence of other forces, the director tilt angle θ would take on some equilibrium value $\tilde{\delta}$. (For the case of slender rod-shaped molecules we would have $\tilde{\delta}=\delta$, but for molecules having a larger girth this will not be exactly true.) Any deviation of θ from $\tilde{\delta}$ will add a contribution to the free energy, as it will tend to alter the volume of the layer by some amount ΔV. Restoring the volume to its constrained value will thus add an energy that will be proportional to the layer compression modulus B and to $(\Delta V)^2$. For small deviations, ΔV will be linear in $(\theta-\tilde{\delta})$ when θ and $\tilde{\delta}$ are constant across a layer.

A complication that arises at this point is the fact that the liquid crystal can flow within a layer when δ varies with position. If θ and $\tilde{\delta}$ are more unequal in some region within a layer than in some other region, then material will flow to equalize the pressure. The free-energy contribution will thus not be proportional to an integral over the layer of the form $B\int[\Delta V(x)]^2dx$, but will instead be proportional to $(B/L)[\int\Delta V(x)dx]^2$. In our model we make the approximation that $\tilde{\delta}=\delta$, and assume that ΔV takes on the form

$$\Delta V\propto\int_{-L/2}^{L/2}\{1-\exp[-c(\cos\theta-\cos\delta]^2\}dx.$$

If we now compress the layer to restore its original volume, we will add to the free energy a term proportional to $(\Delta V)^2$, which we write as

$$\mathcal{F}_2 = b\left(\int_{-L/2}^{L/2}\{1-\exp[-c(\cos\delta-\cos\theta)^2]\}dx\right)^2.$$

The coefficient b will be related to the layer compression modulus B that has been measured by Takezoe and coworkers [20–22]. Frustration occurs when $\delta\neq\theta_0$ because the effect of $\mathcal{F}_1$ is to make θ equal to θ_0, while $\mathcal{F}_2$ tends to make θ equal to δ.

The third term comes from the interaction of an applied electric field E with the polarization of the sample. Each molecule carries both an intrinsic and an induced dipole moment. The measured polarization is a consequence of both the induced dipole moment and the hindered rotation about its axis of a molecule carrying a permanent dipole moment. The transition from Sm-A to Sm-C^* is itself accompanied by an asymmetry in the distribution of the angle ψ describing rotation about the molecular axis, and so it has become common to assume a polarization proportional to sin θ in ferroelectrics in the absence of an applied field [18]. Application of a field then increases the polarization by further hindering rotation and by distorting the charge distribution in the molecules. The free energy then contains terms linear in E from the zero-field polarization and quadratic in E from field-induced polarization. In the present work we neglect the linear terms, which are most important in weak applied fields, and represent the electric-field contribution to the free energy as

$$\mathcal{F}_E = -\int_{-L/2}^{L/2}\chi E^2\cos\delta\cos\phi\,dx,$$

where χ is a constant and the trigonometric factors are present to extract only the polarization component normal to the substrates.

The last term in the first category of free-energy contributions is due to the anchoring. Molecules located at the boundary tend to retain the orientation that they had in the Sm-A phase, and so we include a term

$$\mathcal{F}_s = -w(\sin\delta\sin\theta\sin\phi+\cos\theta\cos\delta)^2$$

to account for this tendency to lie either parallel or antiparallel to the z axis. This expression is evaluated at each surface, where $x=\pm L/2$.

The second category of terms starts with the usual elastic energy due to spatial variations in the director orientation. In the one-elastic-constant approximation, this is just $\frac{1}{2}k(d\mathbf{n}/dx)^2$ where $\mathbf{n}$ is the director, which can be written as

$$\mathbf{n}=(\cos\delta\sin\theta\sin\phi-\sin\delta\cos\theta)\hat{\mathbf{x}}+\sin\delta\cos\theta\hat{\mathbf{y}}$$
$$+(\sin\delta\sin\theta\sin\phi+\cos\theta\cos\delta)\hat{\mathbf{z}}.$$

The free-energy contribution is then

$$\mathcal{F}_3=\int_{-L/2}^{L/2}f_3\,dx$$

with

$$f_3=\tfrac{1}{2}k[\,\delta'^2(1-\sin^2\theta\cos^2\phi)+\phi'^2\sin^2\theta+\theta'^2$$
$$-2\phi'\delta'\sin\theta\cos\theta\cos\phi-2\delta'\theta'\sin\phi],$$

where we have written δ' for $\partial\delta/\partial x$, θ' for $\partial\theta/\partial x$, and ϕ' for $\partial\phi/\partial x$. To this we add an elastic energy of bending of the layer itself, which will be of the form

$$\mathcal{F}_4=\frac{1}{2}g\int_{-L/2}^{L/2}\delta'^2\,dx$$

with g being another constant.

We thus have identified six different contributions to the free energy of a ferroelectric liquid crystal in its chevron geometry. All are approximations built on assumptions that greatly oversimplify the complex structure of these materials. Our hope is that some of the essential features of the real substance will be retained in this description. The problem being obviously beyond the reach of analytical solution, we now turn to a sample numerical calculation of the properties that result from this form of the free energy.

VI. MINIMIZING THE FREE ENERGY

Calculations were performed for a range of values of the various parameters of the model. It quickly became apparent that the crucial variable determining the nature of the flattening transition is the ratio $\gamma \equiv bL/a$ that specifies the strength of the force driving θ to be equal to δ relative to the force driving θ to be equal to θ_0. We accordingly present the results of varying γ while holding the other parameters fixed at values prescribed as follows.

(1) The value of θ_0 was taken from Ref. [4], in which the steric tilt angle was found to be in the range $15°-18°$ in the compounds studied. In the absence of an applied field we expect the tilt angle to be equal to the chevron angle, and so we chose θ_0 to be about 18°, making $\cos\theta_0 = 0.95$.

(2) Because the results depend only on the relative energies of different layer shapes, the absolute energy values need not be stipulated, and so the value of a is left unspecified.

(3) The relative values of the two elastic constants g, which refers to bending of a smectic layer, and k, which refers to the bending of the director within a layer, are hard to estimate. Because they are of the same order of magnitude, we took them to be equal.

(4) The magnitude of the elastic constant k will be related to the energy density a by a factor having the dimensions of $(\text{length})^2$, with this length of the order of the layer thickness. Because the layer thickness does not enter our model, we specify this factor in terms of the only relevant length, which is the cell thickness L. Our choice of $k = aL^2/160$ was thus effectively a specification of the cell thickness as being large in comparison to the layer thickness.

(5) Finally, the anchoring strength w is related to a by a factor having the dimension of a length. This length was chosen to be sufficiently small to permit the director some motion at the cell wall by putting $w = La/400$. It was found that increasing w by a factor of 100 above this value did not appear to change our results in any noticeable way.

(6) As discussed in the preceding section, the value of c was fixed at 2000.

In addition, it was assumed that $\delta(x)$ and $\phi(x)$ are odd functions, while $\theta(x)$ is an even function and $\theta'(0) = 0$.

The dimensionless free energy per unit length can be written in terms of the appropriate dimensionless units as

$$
\begin{aligned}
F/aL = \gamma \Bigg(& \int_{-1/2}^{1/2} \{1 - \exp[-c(\cos\delta - \cos\theta)^2]\} dX \Bigg)^2 \\
& + \int_{-1/2}^{1/2} \{-\exp[-c(\cos\theta - 0.95)^2] \\
& + (1/320)[\delta'^2(2 - \sin^2\theta\cos^2\phi) + \phi'^2 \sin^2\theta + \theta'^2 \\
& - 2\phi'\delta' \sin\theta\cos\theta\cos\phi - 2\delta'\theta' \sin\phi] \\
& - (E/E_0)^2 \cos\delta\cos\phi\} dX - (1/200) \\
& \times (\sin\delta\sin\theta\sin\phi + \cos\theta\cos\delta)^2_{x=L/2}
\end{aligned}
$$

with $X = x/L$, $\gamma = bL/a$, and $E_0 = (a/\chi)^{1/2}$. Note that now δ', for example, means $d\delta/dX$.

As expected, minimizing the free energy shows that applying a weak electric field $[(E/E_0) \ll 1]$ does not significantly change the tilt angle and the cone angle, but the azimuthal angle becomes smaller as the electric field becomes larger, and at the chevron-bookshelf transition point ϕ is almost equal to zero throughout the cell. The parameter γ is proportional to the layer compression modulus. It measures the relative importance of the force tending to keep θ equal to δ, and thus preserve layer volume, with the term that tends to keep θ equal to θ_0. Calculations were performed for values of γ ranging from 0.1 to 10. In the case of large γ, the layer tilt angle has to remain close to θ (which in turn is close to θ_0), and so increasing the electric field does not significantly change δ. However when the electric field reaches a certain value, δ goes to zero very rapidly, as shown in Fig. 3(a). When $\gamma = 1$ the two terms are comparable, and applying a sufficiently high electric field forms a flat region $(\delta = 0)$ in the middle of the cell, while δ remains almost constant elsewhere, as in Fig. 3(b). Increasing the electric field makes the flat region larger and larger, and finally a bookshelf geometry is reached. Figure 3(c) shows the transition when γ is much smaller $(\gamma = 0.1)$. In this case layers begin to transform at lower electric fields. Although once again flat regions are formed, the layers are more rounded.

VII. DISCUSSION

We have presented a theory of the chevron-bookshelf transition in a ferroelectric Sm-C^* liquid crystal when a strong electric field is applied. This model may be helpful in understanding the different types of behavior experimentally observed when this transition occurs. The theory is based on numerical minimization of a rather complicated free energy, which contains various terms due to different interactions including a term due to the fact that layer volume must remain constant during the transition. The strength of this term in comparison with the others (the parameter γ in the dimensionless free energy) seems to be the crucial factor that determines the way in which the layer shape transforms. Depending on the value of this parameter, our model predicts three different types of behavior as the transition is ap-

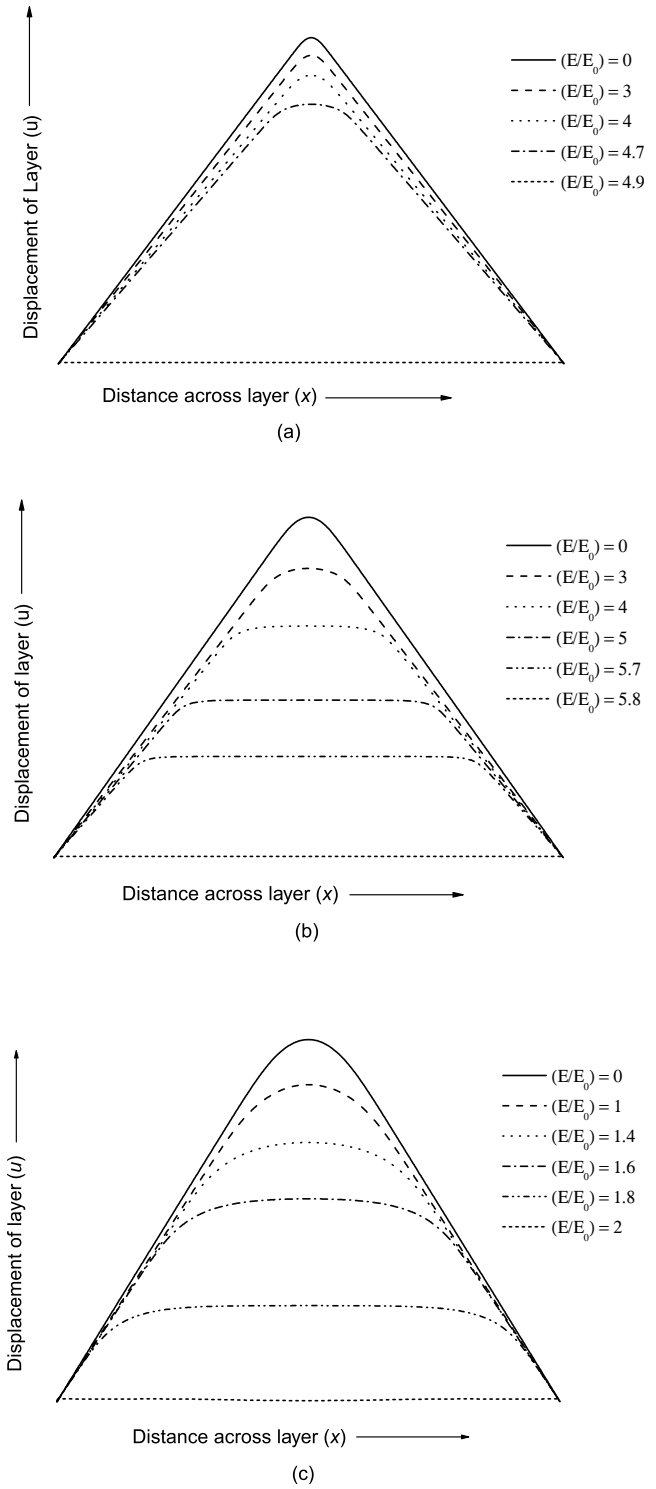

FIG. 3. This figure shows how the chevron shape transforms under the influence of an electric field for (a) $\gamma=10$, (b) $\gamma=1$, and (c) $\gamma=0.1$. The arbitrary scale for the dimensionless layer displacement u has been exaggerated relative to the scale for the distance x across the layer in order to show more clearly the difference between the ''snap,'' ''flatten,'' and ''bend'' types of transition.

proached. We refer to these as ''snap,'' ''flatten,'' and ''bend.''

These three different modes of layer deformation have indeed been experimentally observed in different Sm-C^* phases. This suggests that our model, which was derived from physical considerations of only a ferroelectric phase, may have a wider applicability. A fairly sharp transition (snap), corresponding to the case of large γ in our model, has been reported in the Sm-C^*_{AF} phase by Gleeson and co-workers [4,7] and by Barois and co-workers [5]. They find a distinct threshold field at which the transition from chevron to bookshelf occurs. That this happens only in the large-γ range in the model implies an energy cost to change layer volume that is large in comparison to the cost of moving the tilt angle θ from its preferred value θ_0. This is consistent with the fact that increased layer ordering is often observed on the transition to the Sm-C^*_{AF} phase, where layers become better defined, less ''interlaced,'' and therefore presumably harder to compress [23].

If γ is comparable to unity, the proposed model shows gradual growth of a flat region formed in the middle of the cell (flatten). This is similar to what has been observed by the Barois group in the intermediate ferrielectric phase of one material, where a continuous growth of the bookshelf component was seen, with no sharp field threshold [5]. Work on several different materials by the Gleeson group produced evidence for this type of transformation in the ferroelectric phase [4].

The third type of behavior, which the model displays when γ is appreciably less than unity, is characterized by a gradual transformation from a sharp chevron to a smooth curve (bend). This has been seen in the ferroelectric phase in one material by the Barois group [5]. A similar behavior was observed in ferrielectric phases of different materials by the Gleeson group, although there the rounding was formed over a restricted range of field values, and thus could also have been described as snap. The experimental results in this regime are comparatively sparse, and so further measurements would be desirable.

We now attempt to give a physical picture of the mechanisms underlying these different modes of transition from the chevron to the bookshelf structure. Unfortunately, the full free-energy functional is too complex to permit a simple visualization, and so we illustrate the mechanism by considering a special case in which θ and δ are held constant, and the anchoring energy is omitted. We can then show the variation of the free energy $\mathcal{F}(\delta,\theta,\gamma,E)$ by drawing contour plots of $\mathcal{F}(\delta,\theta)$ for various γ and E. These are shown in Fig. 4.

The first pair of plots, Fig. 4(a), shows the free energy for large γ before and after application of an electric field. When $\gamma=10$, the tendency of θ to equal δ is dominant, and there is a narrow valley in the free energy along the line $\theta=\delta$, with a minimum at $\theta=\theta_0$. Application of an electric field creates a new minimum at small values of θ and δ. Because there is an energy penalty associated with values between these two regions, intermediate values of θ tend not to occur. The transition is thus sudden (snap).

The second pair of plots, Fig. 4(b), shows the free energy for intermediate γ. When $\gamma=1$, there is initially a deep minimum near $\theta=\delta=\theta_0$. Application of an electric field creates a new broad minimum at small values of δ and θ and

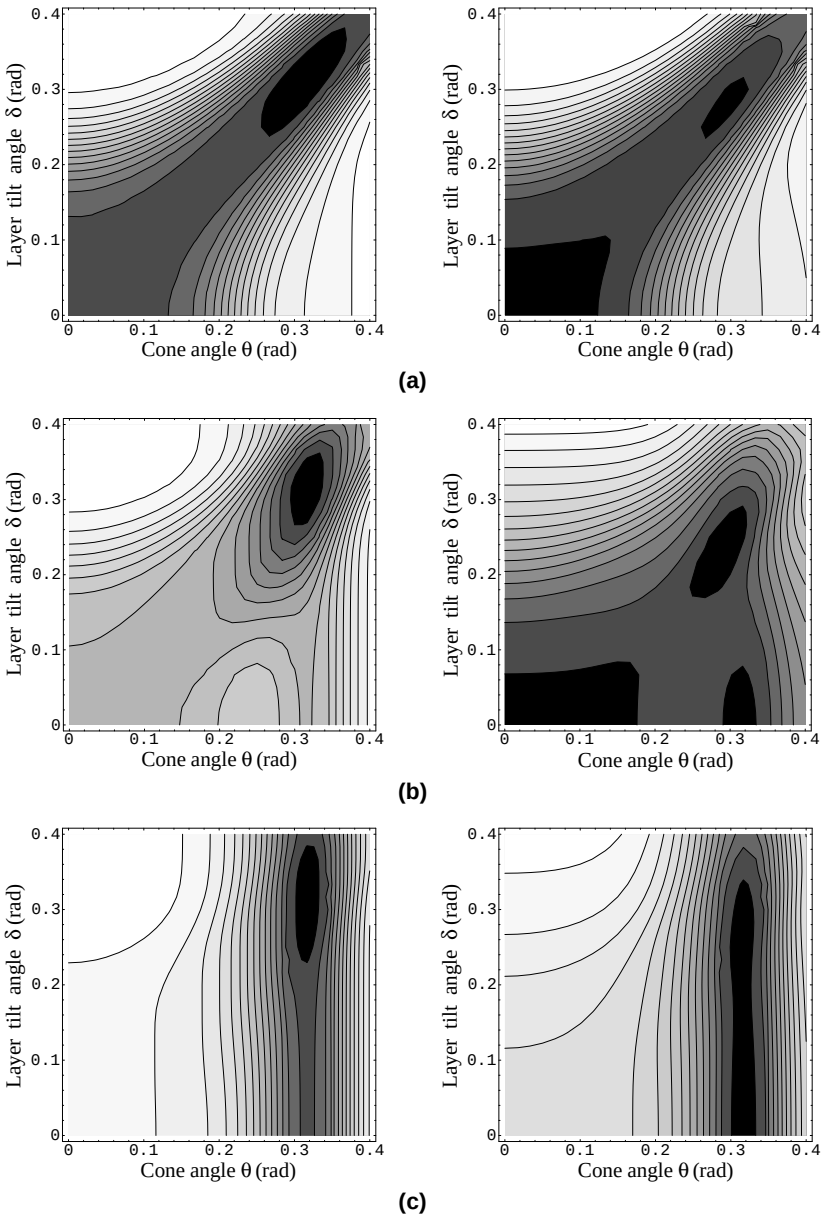

FIG. 4. These contour plots show a simplified version of the free energy as a function of layer tilt angle δ and cone angle θ before and after the application of an electric field. (a) $\gamma=10$, (b) $\gamma=1$, (c) $\gamma=0.1$.

a slightly deeper one near $\delta=0$ and $\theta=\theta_0$. Flattening of the chevron tip occurs when part of the layer makes a transition from $\delta=\theta_0$ to $\delta=0$ with θ remaining initially close to θ_0. This would normally be a sudden transition, but the fact that θ can change from θ_0 to zero at very little energy cost means that the compressibility of the liquid crystal is high. This allows intralayer flow, which extends the range of applied field over which the flattening transition occurs.

The third pair of plots, Fig. 4(c), shows the free energy for small γ. When $\gamma=0.1$, there is again initially a minimum near $\theta=\delta=\theta_0$, but now application of an electric field creates a long narrow valley extending from $\delta=\theta_0$ to $\delta=0$ with θ remaining constant at θ_0. There is now little energy penalty when δ varies continuously within the layer, and the layer curves smoothly (bend).

In summary, our model appears capable of describing the three main types of layer deformation observed experimentally in the ferroelectric, antiferroelectric, and ferrielectric phases. The theoretical basis for the model, however, lies in only the ferroelectric phase, and so it must at the moment be regarded principally as a phenomenological model until the theory of deformation of a multilayer system is better developed.

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