

Electric-field-induced transition between the anticlinic and the synclinc smectic-*C* surfaces in free-standing films

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Anticlinic smectic-*C* surfaces were found experimentally as ground state structures in free-standing films made of smectic liquid crystals with no anticlinic bulk phases. A mean-field interpretation of this observation is given within a discrete phenomenological model of antiferroelectric liquid crystals, which additionally considers the enhanced order present at the surfaces of the free-standing films. The temperature dependence of the critical electric field that drives the transition between the anticlinic and synclinc smectic-*C* surfaces is evaluated, and fair agreement with the experimental data is found.

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I. INTRODUCTION

Free-standing films of smectic liquid crystals are stacks of N parallel smectic layers, spanned in the air over a window of area approximately 1–5 cm². In the Sm-A phase on the average the molecules are perpendicular to the layers. Upon cooling toward the temperature of the transition from the Sm-A to the tilted phase T_c^b , the molecules in the surface layers tilt at the surface transition temperature T_c^{surf} , which is higher than T_c^b [1,2]. The difference between T_c^{surf} and T_c^b can be from ~ 1 K up to 10 K or even 20 K. The increase of the transition temperature in the surface layers is due to the surface tension, which quenches layer displacement fluctuations and enhances the smectic order at free surfaces [3].

At temperatures between T_c^{surf} and T_c^b , the molecules in layers close to the surfaces are tilted considerably more than those in the interior layers. Two distinct molecular configurations have been observed experimentally. The symmetric one, where the directions of the tilt in the upper and in the bottom surface layers are parallel, is called *synclinc smectic-*C* surfaces*, or the *S structure*, or just the *synclinc structure*, and the antisymmetric one, where the directions of the tilt at the two surfaces are antiparallel, is called *anticlinic smectic-*C* surfaces*, or the *C structure*, or simply the *anticlinic structure* [4–8]. It is surprising that, although a particular compound at temperatures below T_c^b in the bulk exhibits only the synclinc Sm-*C*=* structure, where the tilts in neighboring layers are almost parallel, it forms the *C* structure as the ground state in free-standing films at temperatures above T_c^b [4]. Upon applying an external electric field parallel to the smectic layers, a transition from the ground state *C* structure to the *S* structure was observed at the critical field E_c . The critical field was reported to increase upon cooling and to decrease with increase in the thickness of the film. When approaching T_c^b from above, a transition from the *C* to the *S* structure was observed and the critical field becomes zero.

In a recently proposed explanation for the stability of the *C* structure in compounds with high saturation values of the

transverse polarization, thermal fluctuations of the polarization field were recognized as an important source of the long-range interactions that stabilize the *C* structure [7]. The entropic contribution of the polarization fluctuations was shown to reduce the free energy of the *C* structure more than the free energy of the *S* structure. As we understand, this explanation rests on the assumption that both configurations present locally stable structures, and the contribution of the fluctuations merely lowers the minimum of the *C* structure below the minimum of the *S* structure. In this paper, we would like to show that the two minima actually appear in the mean-field treatment of the free-standing films so the assumption in Ref. [7] is justified. Moreover, with a properly chosen set of model parameters, the *C* structure happens to be preferred over the *S* structure even within the mean-field analysis. Fluctuations, however, can additionally enhance its stability.

There have also been observations of the opposite effect in other compounds, where a transition from the ground state *S* structure to the *C* structure was induced by an electric field [5–7]. This was explained successfully by the appearance of flexoelectric polarization and its coupling with the electric field, and we shall not discuss this case any further.

II. THE MODEL

In the following we shall show that the reported phenomena can be interpreted within the discrete phenomenological model of antiferroelectric liquid crystals [9]. The order parameter is a set of two-dimensional layer-tilt vectors $\xi_i = (\xi_{i,x}, \xi_{i,y})$, which are the projections of the layer directors n_i onto the plane xy parallel to the smectic layers. The index i stands for the i th layer. The free energy G_0 is written as a sum of terms describing the intralayer and interlayer interactions up to the second neighboring layers:

$$G_0 = \sum_i \left[\frac{1}{2} a_0 \xi_i^2 + \frac{1}{4} b_0 \xi_i^4 + \frac{1}{2} a_1 (\xi_i \cdot \xi_{i+1}) + \frac{1}{8} a_2 (\xi_i \cdot \xi_{i+2}) \right]. \quad (1)$$

Intralayer interactions are described by the first two terms, where $a_0 = \alpha(T - T_0)$. T_0 would be the Sm-A \leftrightarrow tilted phase transition temperature if there were no interaction between different layers and assuming a second order transition from the Sm-A to the tilted phase, α and b_0 are positive constants. The lowest order terms describing interactions between neighboring layers are the a_1 term, describing the nearest layers' interactions, and the a_2 term, describing the next-nearest layers' interactions. The signs of parameters a_1 and a_2 determine the preferred relative orientation of tilts in interacting layers. If a_1 and a_2 are positive, anticlinic tilts are favored, and if they are negative, synclinic tilts are favored.

Where a_2 is negative or positive but smaller than $|a_1|$, the structure of the tilted phase is determined by the sign of the parameter a_1 . If a_1 is negative, the tilted phase at temperatures below the Sm-A phase is the synclinic Sm-C phase, and if a_1 is positive, it is the anticlinic Sm- C_A phase. For positive a_2 , which is also larger than $|a_1|$, a balance between competing nearest layer and next-nearest layer interactions is achieved in the short pitch structure of the Sm- C_α phase. We wish to explain the observed stability of the anticlinic C structure in a film made of the bulk synclinic material—therefore we shall choose the values of the model parameters so as to obtain the synclinic Sm-C phase just below the Sm-A phase in the bulk. Thus a_1 should be negative. But we shall also permit a certain competition between the nearest and the next-nearest layer interactions by setting a_2 positive but smaller than $|a_1|$. The competition should be weak enough not to disturb the synclinic structure in the bulk, but should result in the appearance of not completely synclinic structures in free-standing films.

In order to obtain the surface transition at temperature higher than the bulk transition temperature T_c^b , which in our case equals $T_c^b = T_0 + (-a_1 + a_2/4)/\alpha$, we shall follow a simple phenomenological approach [1]. The parameters a_0 and b_0 , which describe intralayer interactions, will be the same in all the layers in the bulk but we will take a different \tilde{a}_0 in the surface layers of the free-standing film. Since the smectic order is larger in the surface layers, we assume \tilde{T}_0 is higher than T_0 and the temperature dependent parameter $\tilde{a}_0 = \alpha(T - \tilde{T}_0)$ for the surface layer is smaller than a_0 for interior layers. The difference $\tilde{T}_0 - T_0 = (a_0 - \tilde{a}_0)/\alpha = \Delta a_0/\alpha$ is a positive measure of the excess order at the surfaces and will be temperature independent.

We have described elsewhere in detail [10] how the stability analysis of the high temperature Sm-A phase is performed and how the structure of the tilted phase below the Sm-A phase in free-standing films is determined numerically within this model. The Sm-A phase is stable as long as the matrix of the second derivatives of the free energy (1) with respect to the set of two-dimensional order parameters (i.e., the inverse susceptibility matrix) is positive definite. The main part of the analysis is to find the temperature where the smallest eigenvalue of the inverse susceptibility matrix in the Sm-A phase becomes zero. The corresponding eigenvector is related to the structure of the tilted phase. We have shown that in films with a finite number of smectic layers the tran-

sition from the Sm-A phase first goes to a uniplanar structure, and that a spatially modulated structure may only evolve at a lower temperature. We call a structure or a fluctuation uniplanar if the molecules in all the layers tilt within a single plane. The same procedure was done here and we will only review the results.

III. RESULTS

Important ingredients of our analysis are the temperatures where the lowest eigenvalues of the inverse susceptibility matrix in the Sm-A phase become zero. In Fig. 1(a) the largest three among these temperatures denoted by T_{c1} , T_{c2} , and T_{c3} are plotted as a function of $\Delta a_0/\alpha$. Calculations were done for a nine-layer-thick film and an appropriate set of model parameters. The temperature of the Sm-A \leftrightarrow tilted phase transition is the highest among them: T_{c1} for $\Delta a_0/\alpha < \sim 6$ K and the higher of T_{c1} and T_{c2} for larger $\Delta a_0/\alpha$, where the two temperatures almost coincide. This part is reproduced in Fig. 1(b), where the difference $T_{c1} - T_{c2}$ is plotted in dependence on $\Delta a_0/\alpha$. Later we shall recall that there exists a region of values of $\Delta a_0/\alpha$ such that T_{c2} is higher than T_{c1} .

To understand the influence of large $\Delta a_0/\alpha$ on the small difference between T_{c1} and T_{c2} let us look at the shape of the uniplanar fluctuations that they belong to. Due to the symmetry of the film, all the eigenfluctuations are either symmetric or antisymmetric with respect to the midpoint. In Fig. 1(c) the first two fluctuation modes are shown schematically at temperatures such that the fluctuation modes are (or would be) critical and where the corresponding relaxation frequency (the eigenvalue of the inverse susceptibility matrix) is zero. The first one corresponds to T_{c1} and is symmetric and the second one corresponds to T_{c2} and is antisymmetric with respect to the midpoint.

For small $\Delta a_0/\alpha$, both fluctuations are characterized by small amplitudes in the surface layers compared to the amplitudes in the interior layers. This means that the tilt vector fluctuations are harder at the surfaces and softer inside the film. Upon cooling, a tilted structure with a small tilt in the surface layers and a large tilt in the interior layers should evolve.

For large $\Delta a_0/\alpha$, the situation is reversed. The amplitude of the first two fluctuation modes is large at the surfaces and small inside the film and both modes become unstable at almost the same temperature. Since mostly the surface layers are involved in this process for large $\Delta a_0/\alpha$, the critical temperature (higher between T_{c1} and T_{c2}) is called the surface transition temperature T_c^{surf} . It is much higher than the bulk transition temperature T_c^b .

At temperatures below T_c^{surf} , the critical fluctuation mode induces a uniplanar tilted structure with nonzero tilt in the surface layers and either symmetric or antisymmetric tilt profile. A schematic representation of both possible structures is shown in Fig. 2. The symmetric one is the S structure and the antisymmetric one is the C structure. They both resemble the shapes of the fluctuations they evolve from.

Within the next-nearest layer interaction model the two

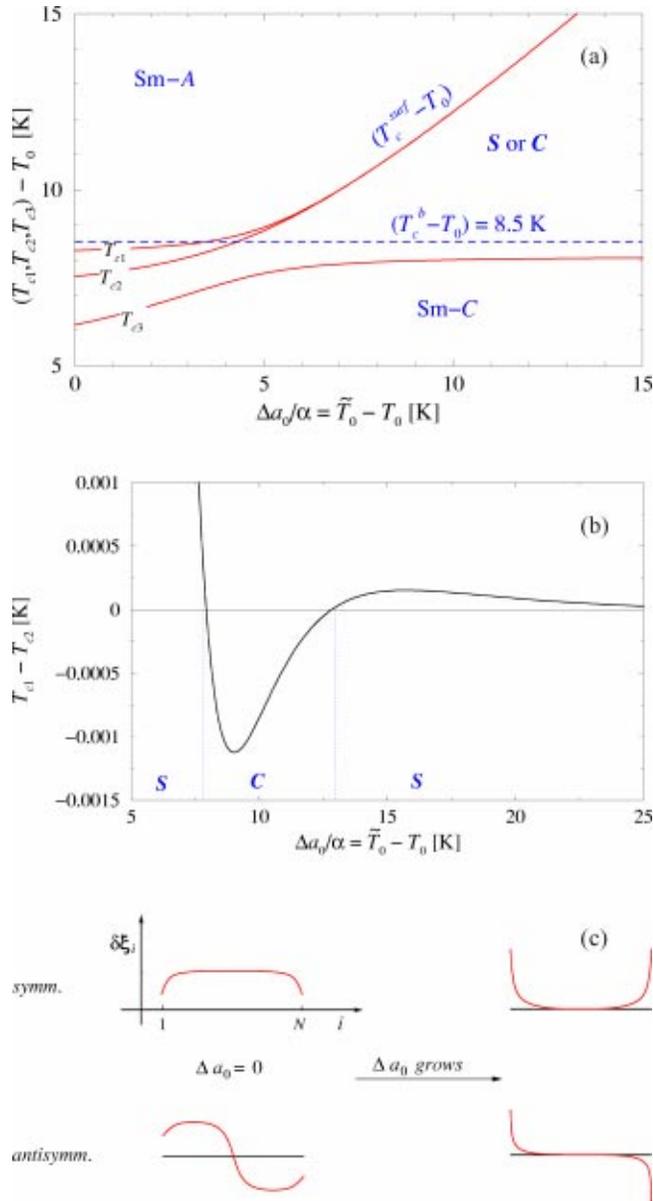


FIG. 1. (a) Temperatures where the first, the second, and the third eigenvalues of the inverse susceptibility matrix in the Sm-A phase become zero are plotted as functions of the surface order $\Delta a_0/\alpha = \tilde{T}_0 - T_0$. The Sm-A phase is stable at temperatures above T_{c1} and T_{c2} , whichever is higher. Calculations were done for a nine-layer film made of a ferroelectric liquid crystal described by parameters $a_1/\alpha = -10$ K and $a_2/\alpha = 6$ K. (b) The difference between T_{c1} and T_{c2} in the region where they are almost the same. The unstable fluctuation mode corresponds to the symmetric S fluctuation for $\Delta a_0/\alpha < 7.9$ K and for $\Delta a_0/\alpha > 12.8$ K, whereas for $\Delta a_0/\alpha \in (7.9, 12.8)$ K the unstable mode is the antisymmetric C fluctuation. (c) Schematic representation of the symmetric and antisymmetric uniplanar fluctuations at temperatures where they become unstable in the Sm-A free-standing film.

structures will have the same free energy if the tilt is nonzero only in a few surface layers and exactly zero in more than two neighboring layers inside the film. In such a case the directions of the tilt at the two surfaces of the film are not related. But, in fact, their free energies are not quite the

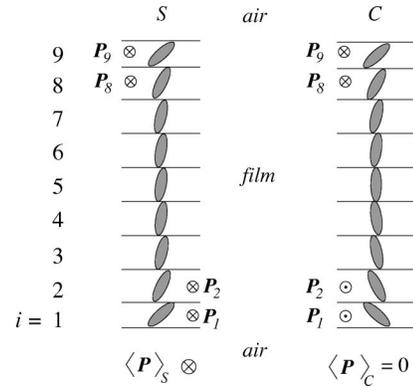


FIG. 2. Schematic representation of the uniplanar S and C structures, which evolve from the symmetric and the antisymmetric critical uniplanar fluctuations below the transition from the Sm-A phase as the surface order is enhanced in a nine-layer film. The slightly tilted molecules in the interior layers suppress the degeneracy among the two structures, since they transmit interactions between the two surface sides of the film. The parameters are chosen to correspond to the region C in Fig. 1(b), at some temperature between the surface transition temperature T_c^{surf} and the bulk transition temperature T_c^b .

same, since the tilt in the middle layers of the film is not exactly zero and the interaction between the surfaces is transmitted across the film. Therefore only one of them describes the ground state structure. Which one is it?

Let us look once more at Fig. 1(b), where the difference of the temperatures $T_{c1} - T_{c2}$ is plotted as a function of $\Delta a_0/\alpha$. There is a region of $\Delta a_0/\alpha$ between 7.9 K and 12.8 K where T_{c2} is larger than T_{c1} and where the antisymmetric fluctuation mode is critical. For the values of $\Delta a_0/\alpha$ outside this region the critical mode is the symmetric fluctuation mode. In the bulk the parameter $\Delta a_0/\alpha$ has no significance and the structure of the tilted phase is completely determined by α , b_0 , a_1 , and a_2 . But for an appropriate set of model parameters the tilted structure in a free-standing film just below the T_c^{surf} can be either symmetric or antisymmetric about the midpoint, and the choice between them depends essentially on $\Delta a_0/\alpha$, the excess order at the surfaces of the film.

It is thus possible that in the bulk only the completely synclitic tilted Sm-C structure is stable, whereas in the film the ground state just below T_c^{surf} is the C structure. Its appearance depends on the enhanced order in the surface layers assumed at large $\Delta a_0/\alpha$ and on a positive value of a_2 , which does not favor synclitic tilts in next-nearest layers. Let us illustrate the possible stability of the C structure by two examples dealing with films with odd and even numbers of layers, respectively.

In the next-nearest layer interaction model, a difference between the S and the C structure emerges mainly in the few middle layers of the film. In the first approximation, we can neglect the minor differences in the tilt profile of both structures in other layers. If we look at the middle layers and their contribution to the interlayer interaction energy, we should compare the symmetric S and the antisymmetric C configurations. In Fig. 3(a) the four middle layers of a film with an

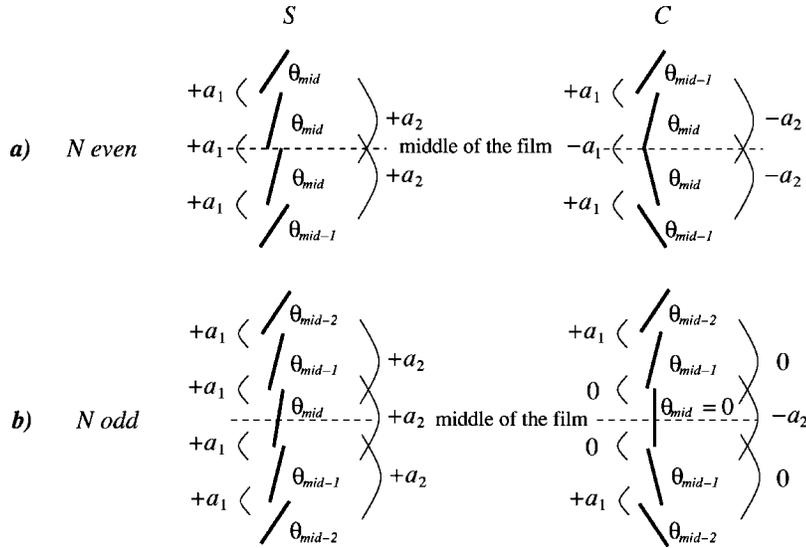


FIG. 3. Schematic representations of the S and C tilt configurations in (a) the four middle layers in a film with an even number of layers and (b) the five middle layers in a film with an odd number of layers. The pairs of layers that contribute to the interlayer interaction energy and the sign of their contributions are denoted by $\pm a_{1,2}$ [see Eq. (1)].

even number of layers are shown. Their interlayer ($i-l$) interaction contribution to the free energy is

$$G_{i-l, N \text{ even}, S} = \frac{1}{2} a_1 (2 \theta_{mid-1} \theta_{mid} + \theta_{mid}^2) + \frac{1}{8} a_2 (2 \theta_{mid-1} \theta_{mid})$$

for the S configuration and

$$G_{i-l, N \text{ even}, C} = \frac{1}{2} a_1 (2 \theta_{mid-1} \theta_{mid} - \theta_{mid}^2) - \frac{1}{8} a_2 (2 \theta_{mid-1} \theta_{mid})$$

for the C configuration, where θ_{mid} is the magnitude of the tilt in the two middle layers and θ_{mid-1} is the magnitude of the tilt in the neighboring two layers [see Fig. 3(a)]. If $G_{i-l, N \text{ even}, C} < G_{i-l, N \text{ even}, S}$, the C structure is preferred over the S structure:

$$a_2 > -2a_1 \frac{\theta_{mid}}{\theta_{mid-1}} \Rightarrow G_{i-l, N \text{ even}, C} < G_{i-l, N \text{ even}, S} \Rightarrow C \text{ stable.} \quad (2)$$

The magnitude of the tilt decreases toward the midpoint due to a large $\Delta a_0/\alpha$, so that $\theta_{mid} < \theta_{mid-1}$ and the approximate condition (2) for the stability of the C structure is satisfied ($a_1 < 0$, $a_2 > 0$).

In odd-layer films the magnitude of the tilt in the middle layer is zero for the C structure and small but nonzero for the S structure. In Fig. 3(b) the symmetric S and antisymmetric C configurations in five layers in the middle of the film are shown. A similar reasoning as for the even films leads to a similar condition for the stability of the C structure:

$$a_2 > -4a_1 \frac{\theta_{mid} \theta_{mid-1}}{\theta_{mid} \theta_{mid-2} + \theta_{mid-1}^2} \Rightarrow G_{i-l, N \text{ odd}, C} < G_{i-l, N \text{ odd}, S} \Rightarrow C \text{ stable.} \quad (3)$$

Upon cooling the tilt increases in the interior layers as well and at some temperature T_{CS} near T_c^b a completely synclinc structure is preferred, as is observed experimentally [4]. We can easily reproduce such behavior: We present it in Fig. 4, where the curve corresponding to $E=0$ represents the difference of the free energies of the symmetric S and antisymmetric C structures $\Delta G_0 = G_{0,S} - G_{0,C}$.

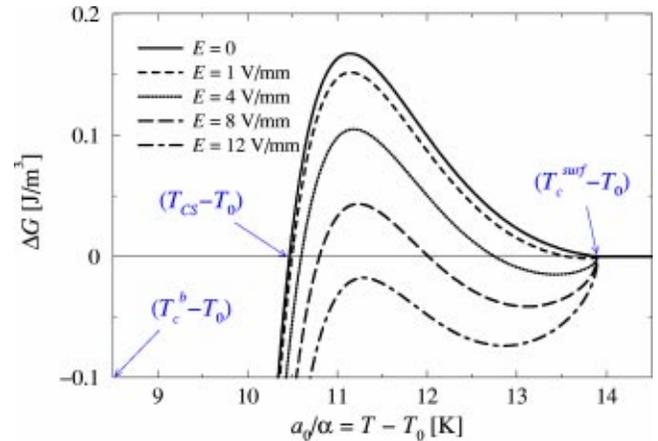


FIG. 4. The difference between the free energies of the S and C structures as a function of temperature for a nine-layer film. The typical values of material parameters are chosen such as to obtain meaningful values of the energy: the smectic layer thickness d_0 is 3 nm and α , which corresponds to the specific heat, is $4 \times 10^4 \text{ J/m}^3 \text{ K}$ [11]. The ratio of the polarization and the tilt depends on a compound. We have chosen a value that is twice as large as in DOBAMBC (*p*-decyloxybenzylidene *p*-amino 2-methyl butyl cinnamate), $P = 8 \text{ nC/cm}^2$ at $\theta = \pi/9$. The other model parameters are the same as before, $a_1/\alpha = -10 \text{ K}$, $a_2/\alpha = 6 \text{ K}$, $b_0/\alpha = 100 \text{ K}$, and $\Delta a_0/\alpha = 12 \text{ K}$.

At temperatures where $\Delta G_0 > 0$, below T_c^{surf} and above T_{CS} , the C structure is the ground state since it has a lower free energy than the S structure. At T_{CS} their free energies are identical and the first order transition between the two structures changes the character of the ground state below T_{CS} . As expected, the synclinic S or Sm- C structures are preferred at low temperatures, where the tilt becomes larger through the film.

Until now we have not discussed the influence of an external electric field, which is commonly used in experiments to align the sample. If the field is weak (and usually it is) it does not distort the structure, but it can lift a degeneracy between the various structures or increase the free energy difference between them. We will consider the coupling between the electric field parallel to the smectic layers and the transverse ferroelectric polarization \mathbf{P}_i of separate layers, which also lies in the smectic plane and is perpendicular to the tilt vector $\boldsymbol{\xi}_i$.

Transverse polarization of each smectic layer is induced by piezoelectric coupling between the tilt and the polarization, $P_i = \epsilon C_{pie} \theta_i$. Where the tilt is zero, there is no polarization. In the S and C structures, tilt and polarization are present mostly in the surface layers. Tilts in the upper and in the bottom surface layers are parallel in the S structure and the average polarization $\langle \mathbf{P} \rangle_S = (1/N) \sum_{i=1}^N \mathbf{P}_i$ of the film is maximal, whereas in the antisymmetric C structure tilts and polarizations in the surface layers are antiparallel and the average polarization $\langle \mathbf{P} \rangle_C$ of the film is zero (see Fig. 2). A weak external electric field \mathbf{E} parallel to the smectic layers aligns the S structure so that $\langle \mathbf{P} \rangle_S$ is parallel to \mathbf{E} and does not have any influence on the C structure.

When an external electric field is applied, an additional linear term must be added to the free energy G_0 . The total free energy is now

$$G_E = G_0 - \mathbf{E} \cdot \langle \mathbf{P} \rangle. \quad (4)$$

The free energy $G_{E,C}$ of the C structure is not changed by this linear coupling term, whereas the free energy of the S structure $G_{E,S}$ is decreased, $G_{E,S} < G_{0,S}$. If the S structure is the ground state when $E = 0$, it remains the ground state even when $E \neq 0$. But if the C structure is the ground state for $E = 0$, there exists a critical field E_c where the free energies of both structures are the same, $G_{E_c,S} = G_{E_c,C}$. If E is larger than E_c , the S structure is stable. In Fig. 4 the temperature dependence of the free energy difference $\Delta G_E = G_{E,S} - G_{E,C}$ in the presence of an electric field is shown for the same set of model parameters as before.

Our understanding of the effect is summarized in Fig. 5, where the critical field E_c is shown in dependence on temperature for a few films of different thicknesses. The curves showing the temperature dependence of the critical field separate the stability regions of the S , C , and Sm- C structures. For $E_c = 0$, the S structure is stable at temperatures up to T_{CS} , where the transition to the C structure takes place, and the C structure is replaced by the Sm- A phase at temperatures above T_c^{surf} . At temperatures around and below T_c^b the S structure is more like Sm- C , with synclinic and con-

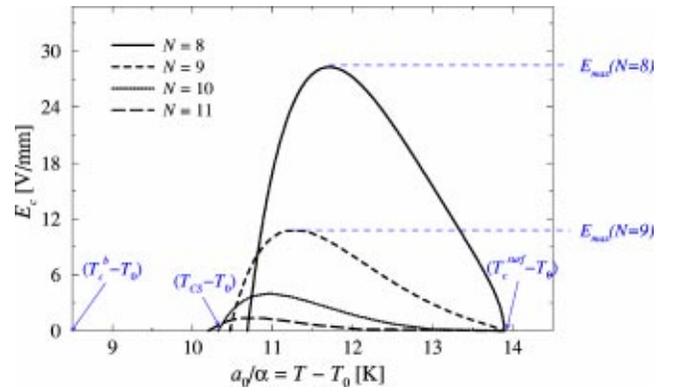


FIG. 5. The critical electric field that induces the transition between the C and the S structures in films with 8, 9, 10, and 11 layers. At high temperatures, the C structure is stable in the regions below the $E_c(a_0/\alpha)$ curves and the S structure is stable above them. At low temperatures or strong electric fields, the stable structure corresponds to the Sm- C phase. For $T > T_c^{surf}$ and $E = 0$ the Sm- A phase is stable. The material and model parameters are the same as in the previous figure.

siderable tilt in all layers. The transition temperature T_{CS} depends on the number of layers N in the film, whereas the surface transition temperature T_c^{surf} does not depend on N for films thicker than approximately seven layers. In the Sm- A phase there exists some nonzero tilt in the same direction in all the layers if $E \neq 0$. This is the linear response due to the coupling between \mathbf{E} and \mathbf{P} , which stabilizes the S structure at high temperatures. When cooling at constant E we first reach a certain temperature where the C structure becomes preferred over the field-induced S structure, and at some lower temperature the C structure becomes stable again. The corresponding electric field is thus the critical field E_c which induces the transition from the C to the S structure at the two temperatures. The temperature region where the C structure is stable shrinks as the electric field is increased. If E is larger than E_{max} , the C structure is driven out of the phase sequence. This effect was reported first in Ref. [8]: The authors experimentally observed the reentrant appearance of the S structure when the Sm- A film was cooled in a moderate electric field of around 10 V/mm. When the field was even higher, the structure remained synclinic in the whole temperature region, and the C structure was not observed at all.

Where else do our predictions meet the experimental observations? The critical field calculated within our model is of the same order of magnitude as the measured fields (5–15 V/mm) [4,7,8]. With increasing number of layers N , the critical field E_c decreases as reported [4,8]. At higher temperatures E_c decreases with increasing temperature, as was observed experimentally [4]. We also predict that at low temperatures close to T_c^b the critical field E_c should decrease with decreasing temperature. This was not observed yet to the best of our knowledge, but such behavior can be expected since the synclinic Sm- C structure is stable in the bulk at low temperatures and therefore the critical field should go to zero.

A word should be said also about the parameters we have used in our calculations and their influence on the critical

field. A large piezoelectric coupling constant C_{pie} leads to a large spontaneous transverse polarization and to small critical fields. Strong interlayer interactions (large a_1 and a_2) in general increase the difference of the free energies of the various structures, and so the critical field becomes higher. On the contrary, a large $\Delta a_0/\alpha$ indicates a small magnitude of the tilt in interior layers, a less important contribution to the difference of the free energies of the S and the C structures, and therefore a small critical field. The explanation we propose here is meaningful for substances that have a suitable combination of different properties: Strong but weakly competing interlayer interactions increase the acceptable difference of the transition temperatures $T_c^{surf} - T_c^b$.

IV. CONCLUSION

To summarize, a mean-field interpretation of the electric-field-induced transition from the ground state C structure to the S structure in free-standing films was proposed for compounds with no antclinic bulk phases. The model proposed is a discrete phenomenological model of antiferroelectric liquid crystals, which explicitly includes interactions up to next-nearest layers. The basic assumption of our explanation is the existence of a minimal competition between nearest layer interactions, which favor synclitic tilts, and next-nearest layer interactions, which favor anticlinic tilts. In spite of this the only tilted structure in the bulk is synclitic, whereas in films with enlarged surface order and increased transition temperature the balance between the nearest and the next-nearest layer interactions can restore the partially anticlinic C structure. A necessary condition for this to happen is that the interior molecules are tilted as well. A nonzero tilt in the interior layers ensures that the influence of competing interactions extends through the whole film.

With a reasonable choice of the model and material pa-

rameters, the estimated values of the critical electric field for the transition from the C to the S structure are macroscopic and of the same order of magnitude as observed experimentally [4,7,8]. The temperature dependence of the critical field at high temperatures also agrees with observations; upon cooling the critical field increases. The explanation we offered involves a strong dependence of the critical field on the number of layers in the film. With increasing number of layers the critical field becomes smaller, which is the same functional dependence as observed [8].

Finally, we must not forget that physical mechanisms other than those included in our model can be important in these systems. Due to a large tilt and the polarization in the surface layers, the dipolar interaction between the upper and the bottom surface layers can be important; this was already partially analyzed [12]. We have shown that a direct dipolar interaction promotes antiparallel orientation of the transversal polarizations of both surfaces and thus stabilizes the C structure. In an alternative explanation, the stability of the C structure was proposed to result from the interaction between the polarization fluctuations at the two surfaces [7,13].

But, as already suggested, further studies of the structure as a function of thickness are needed to confirm the validity of the proposed models. Nevertheless, we believe that our explanation contributes to a general understanding of this intriguing phenomenon, which has been much studied recently.

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 - [13] For a comparison with our results we have tried to reproduce the calculations of Ref. [7] with the same set of material parameters and we were not able to obtain the same results. However, we have found that the difference in the mean-field free energies of the S and C structures contributes 40 times more to the critical field than the polarization fluctuations. Therefore we conclude that the transition between the S and C structures is governed mostly by the mean-field mechanisms.