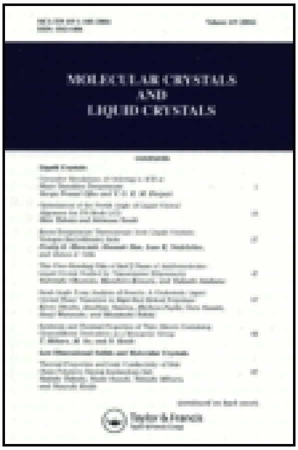

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Study of Liquid Crystal Structural Changes in Phase Under the Influence of Electric Field Using the Discrete Model

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Study of Liquid Crystal Structural Changes in SmC$_{\alpha}^{*}$ Phase Under the Influence of Electric Field Using the Discrete Model

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The Landau phenomenological free-energy expansion in its discrete form is used to describe the free energy of each layer of a ferroelectric (FE) and antiferroelectric (AF) liquid crystalline system. In order to study systematically the effect of electric field on phases, a simple free-energy model is considered, which includes only the usual Van der Waals, nearest-neighbour and next nearest-neighbour interactions. Numerical simulation based on the discrete-form of Landau-Khalatnikov equations of motion is performed to analyze the influence of electric field on the system. The resultant structural changes are presented.

Keywords: antiferroelectric liquid crystals; discrete model; free energy; phase structure

1. INTRODUCTION

In 1989, Chandani et al [1] discovered transverse antiferroelectricity properties in the chiral compound MHPOBC. In this compound, successive layers have anticlinic tilting and therefore antiparallel polarization. Detailed studies show that this novel compound exhibits a rich variety of other phases beside the antiferroelectric phase (SmC$_{A}^{*}$) as it is cooled from the isotropic phase according to the sequence: SmA-SmC$_{\alpha}^{*}$-SmC$_{\gamma}^{*}$-SmC$_{A}^{*}$ etc. A somewhat similar phase sequence is also observed recently in the thiobenzoate antiferroelectric liquid crystal [2].

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Much attention has been focused on the structure of SmC^*_α [3–5]. This phase as revealed by optical and X-ray measurements has a small tilt angle and a very short pitch [3,6], from a few layers to ten or more layers. It is ferrielectric in nature at low temperatures but becomes antiferroelectric in the proximity of the SmA-SmC^*_α transition temperature [7,8]. When an external electric field is applied, a number of interesting phenomena induced by the field is observed. For example, when exposed to external electric field the structure of the short-pitched incommensurate SmC^*_α phase deforms and gets locked to commensurate period [5]. In order to understand the structure and to explain the phenomena involved, several models have been proposed. The most successful one is the phenomenological theory of liquid crystal based on the discrete model first developed by Čepič *et al* [9]. This model takes into account the configuration of each layer which was first introduced by Sun and Orihara [10]. The phenomenological theory involves an expansion of the Landau free energy density in powers of suitable parameters. In general, the energy expansion contains terms that describe inter and intra layer interactions, chirality, electrostatic interactions within a layer, coupling of tilt and polarization and coupling to the external field.

A number of works has been devoted to studying the structure of the SmC^*_α phase using the discrete model [3,4,9,11,12]. However, there has been no systematic study undertaken to understand the role of each term in the free energy expansion in deforming and stabilizing the structure when exposed to an external applied field. We, therefore, are taking this challenge. In this paper we present our preliminary results of considering the simplest free energy expansions which include only the Van der Waal interactions within each layer, nearest neighbour (NN) and next-nearest neighbour (NNN) interactions and of course the coupling to the applied field.

2. THE DISCRETE MODEL OF THE FREE ENERGY EXPANSIONS

We consider the following free energy expansions in the SmA phase under the action of static external field, written as

$$F = \sum_{j=1}^{N} \left[\frac{A}{2}\xi_j^2 + \frac{B}{4}\xi_j^4 + \frac{J_1}{2}\left(\overrightarrow{\xi_j} \cdot \overrightarrow{\xi_{j+1}}\right) + \frac{J_2}{8}\left(\overrightarrow{\xi_j} \cdot \overrightarrow{\xi_{j+2}}\right) - \vec{\sigma} \cdot \overrightarrow{\xi_j} \right] \tag{1}$$

with $\vec{\sigma} = \varepsilon_0 c_p \vec{E}$ where c_p is the flexoelectric constant. In the above equation, the first two terms are the Van der Waal intra-layer interactions, the third and the fourth term is the NN and the NNN

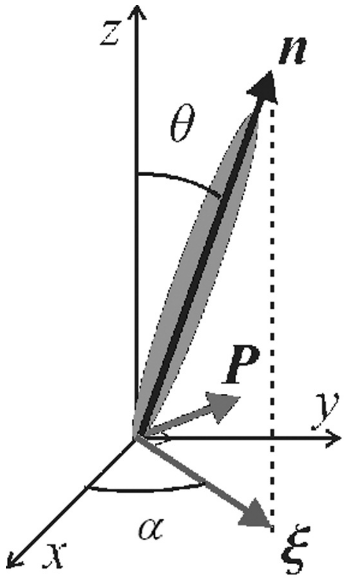

FIGURE 1 Geometry showing the director, $\boldsymbol{n}$, the order parameter, ξ, and the induced polarization $\boldsymbol{P}$ of a smectic layer. ξ_0 is the tilt angle of molecule with respect to the z-axis (the layer normal). α is phase angle of the tilt in the $x-y$ plane.

interactions respectively, and the last is the coupling to the external field. ξ_j is the order parameter for the jth smectic layer which is a set of two-dimensional layer-tilt as shown in Figure 1 where they are the projections of the layer directors n_j on to the plane $x-y$ parallel to the smectic layers. The order parameter is defined by $\vec{\xi}_j = \left(\vec{\xi}_{j,x}, \vec{\xi}_{j,y}\right)$. The constants A and B are associated with the magnitude of the intra-layer interactions where $A = a_0(T - T_0)$. B is a material-dependent parameter; and it is either positive or negative depending on whether the transition to the tilted phase is continuous or discontinuous. J_1 describes the strength of the NN interaction and the parameter J_2 represents the NNN interactions.

In order to obtain the stable solutions of (1), we assumed the structure is helicoidally modulated without applied electric field ($E = 0$). We use the Constant Amplitude Approximation, where the amplitude of tilt angle, ξ_0, is assumed constant in all layers and the phase difference of tilts in neighboring layers, α, is a constant. The equilibrium structure may be given by

$$\xi_j = \xi_0(\cos j\alpha, \sin j\alpha) \tag{2}$$

Substituting (2) in the free energy expression (1), we have

$$\frac{F}{N} = \frac{A}{2}\xi_0^2 + \frac{B}{4}\xi_0^4 + \frac{J_1}{2}\xi_0^2\cos\alpha + \frac{J_2}{8}\xi_0^2\cos 2\alpha \tag{3}$$

as the average free-energy in each layer. Minimizing the free energy with respect to tilt and phase angle gives the following solutions

$$\alpha = 0, \pi \text{ or } \cos^{-1}\left(-\frac{J_1}{J_2}\right) \tag{4}$$

In the absence of external field the solution $\alpha = 0$ corresponds to SmC* phase which is stable for $J_1 < 0$ and $|J_1| > J_2$. The solution $\alpha = \pi$ corresponds to antiferroelectric phase, $\mathrm{SmC}_\alpha{}^*$, which is stable for $J_1 > 0$ and $J_1 > J_2$. The non-trivial solution $Cos\alpha = -(J_1/J_2)$ corresponds to the structure proposed for SmC_α^* phase. However, when the free-energies given by the three solutions are compared, we find that the SmC_α^* phase is most stable and we therefore focus on this solution from now on.

3. THE NUMERICAL SIMULATION

In order to investigate how the structure of the SmC_α^* phase changes with variation in static external field, we consider a bulk liquid crystal system, in equilibrium, consisting of an infinite number of smectic layers with a pitch of three layers. We assume the electric field $\vec{E}$ is applied in the x direction and set a scenario where the static field is slowly varied in small step. With each increment, the system is allowed to settle to its new equilibrium. The dynamic of how the tilt in each layer changes may be described by the Landau–Khalatnikov equation which for the jth layer, is given by

$$\gamma\frac{\partial\xi_{j,x}}{\partial t} = -\frac{\partial F}{\partial\xi_{j,x}} \tag{5a}$$

$$\gamma\frac{\partial\xi_{j,y}}{\partial t} = -\frac{\partial F}{\partial\xi_{j,y}} \tag{5b}$$

for the x and y component respectively. Since $\overrightarrow{\xi_j} = (\xi_{j,x}, \xi_{j,y})$, $\vec{E} = (E_x, 0, 0)$ and $\vec{P} = (-\xi_{j,y}, \xi_{j,x})$ and by noting that $\overrightarrow{\xi_j}\cdot\vec{\xi}_{j+1} = \xi_{j,x}\xi_{j+1,x} + \xi_{j,y}\xi_{j+1,y}$, (1) becomes

$$F=\sum_{j=1}^{N}\left[\frac{A}{2}\left(\xi_{j,x}^2+\xi_{j,y}^2\right)+\frac{B}{4}\left(\xi_{j,x}^2+\xi_{j,y}^2\right)^2+\frac{J_1}{2}\left(\xi_{j,x}\xi_{j+1,x}+\xi_{j,y}\xi_{j+1,y}\right)\right.$$
$$\left.+\frac{J_2}{8}\left(\xi_{j,x}\xi_{j+2,x}+\xi_{j,y}\xi_{j+2,y}\right)+\sigma_x\xi_{j,y}\right] \quad (6)$$

The change in the tilt of each layer may be calculated from the equation of motion, (5), using the approximation $\Delta\xi_{j,x}\approx-\frac{\Delta t}{\gamma}\frac{\partial F}{\partial\xi_{j,x}}$ for the x component where Δt is the increment of time. The time increment is allowed to proceed until $\xi_{j,x}$ reaches stability, that is $\Delta\xi_{j,x}\big|_{n\Delta t}\rightarrow 0$. Therefore, the new equilibrium tilt may be computed using

$$\xi_{j,x}(t)\approx\xi_{j,x}(t=0,E=0)+\Delta\xi_{j,x}\big|_{\Delta t}+\Delta\xi_{j,x}\big|_{2\Delta t}+\cdots\cdots+\Delta\xi_{j,x}\big|_{n\Delta t} \quad (7)$$

A similar procedure is repeated for the y component. Once stability is reached, the phase angle may be calculated based on $\alpha=\tan^{-1}(\xi_{j,y}/\xi_{j,x})$. The simulation is stopped at some value of the critical field, where the structure of the SmC^*_α phase is unwound ($\alpha=0$).

4. RESULTS AND DISCUSSION

At equilibrium, we assume the tilt angle ξ_0 is approximately 0.1 radian. For a three-layer pitch structure, the phase angle between each layer must be taken to be $(2\pi/3)$ radians. Based on this and by assuming that the tilt is not affected much by the changing of the E field, the values of A, J_1 and J_2 are estimated. One possible set of parameters that may be chosen is $A=-\frac{400}{397}$, $J_1=\frac{4}{397}$, $J_2=2J_1$ and $B=100$. Another is $A=-4$, $J_1=4$, $J_2=8$, and $B=100$. We have chosen the earlier set.

Figure 2 shows the sequence of changes in the structure of the SmC^*_α phase. The first figure depicts the equilibrium structure, $\sigma=0\,\text{V/m}$, where the phase angle between the three layer is $(2\pi/3)$. The second shows the structure just after application of the applied electric field, at $\sigma=0.02\,\text{V/m}$. When the electric field reaches a critical field value of $\sigma=11.64\,\text{V/m}$, an unwound structure as shown in the last figure is obtained. As expected, all three layers have collapsed together at a phase angle of 270°, thus producing an induced polarization perpendicular to this angle, in the direction of the field.

Figure 3 illustrates a curve of phase angle, α, versus applied electric field. Again, it is found that all layers converge to the unwound state as the field reaches the critical value, $\sim 11.64\,\text{V/m}$. It is also observed that the phase angle of each layer changes steeply at low field values

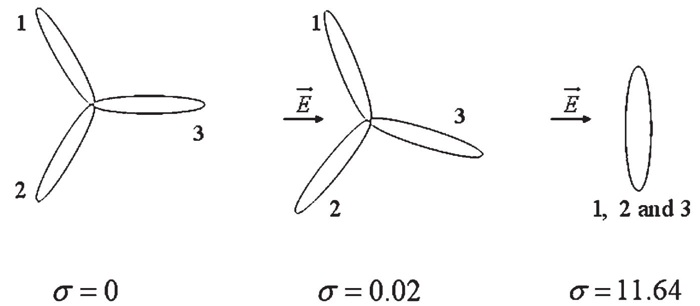

FIGURE 2 The structure of the SmC^*_α phase showing the position of the tilt of each layer in term of phase angle, α, measured in degrees for a sequence of electric field values: $\sigma = 0\,V/m$, $\sigma = 0.02\,V/m$, and $\sigma = 11.64\,V/m$.

before making its way gradually towards the unwound state as the field strength increases.

The behaviour of the SmC^*_α phase structure obtained may be due to the limitation of our model. In this work, we have neglected chirality

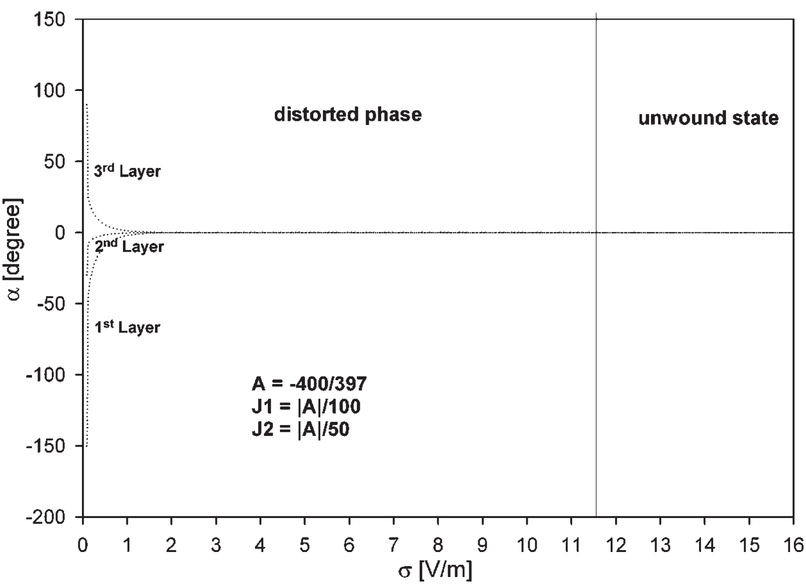

FIGURE 3 Variation of the phase angle, α, with applied electric field, σ, in unit of V/m for each layer. The values of parameters used in the computation are $A = -\frac{400}{397}$, $J_1 = \frac{|A|}{100}$, $J_2 = \frac{|A|}{50}$, and $B = 100$.

contribution in the free energy so that we can concentrate on the role of the NN and the NNN terms towards changes in the SmC$^*_\alpha$ phase with varying electric filed. The contribution of chirality may be important in pulling the tilts to the complete unwound state. We also have not included the role of quadrapolar interaction [13]. It would be interesting to extend this calculation to include these two terms one at a time to see its contribution towards the stable structure of the SmC$^*_\alpha$ phase. This work is currently in progress and will be reported elsewhere.

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