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THE INFLUENCE OF ELECTRIC FIELD ON LIQUID CRYSTAL STRUCTURE IN THE $Sm\ C_{\alpha}^{*}$ PHASE USING THE DISCRETE MODEL

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ABSTRACT

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 based on a five-layer system is considered, which includes only the usual van der Waals, nearest-neighbour and next nearest-neighbour interactions. Using the finite-difference time domain technique, 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. Changes in the phase structure are presented.

INTRODUCTION

Chiral polar smectic phases of liquid crystal materials have attracted much attention in recent years, since these materials exhibits ferroelectric and antiferroelectric properties. In polar smectic phases, the molecules are formed in layered structures and tilted with respect to layer normal having non zero spontaneous polarization. Antiferroelectric smectic phase ($Sm\ C_{A}^{*}$) first reported in novel compound MHBOBC by Chandani *et al* [1]. Subsequently, discovery of various subphases of smectic in different liquid crystalline compounds follow. It is found upon cooling, the complete phase transition sequence below $Sm\ A$ is $Sm\ A\ -\ Sm\ C_{\alpha}^{*}\ -\ Sm\ C_{FI2}^{*}\ -\ Sm\ C_{FI1}^{*}\ -\ Sm\ C_{A}^{*}$ where the $*$ denotes a chiral phase and $Sm\ A$ is the isotropic high temperature phase [2, 3].
The $Sm\ C_{\alpha}^{*}$ is ferrielectric at low temperature and it is fundamentally an important phase because it is a rare example of ferrielectricity without long range positional order. It is also of technological importance for its potential as electro-optical switches [3]. The structure of SmC_{α}^{*} has been widely studied by many researchers but there are still some of its properties that are not well understood [4, 9, 10, 11]. Various experimental techniques prove that SmC_{α}^{*} phase is short pitch helical structure and the pitch varying from few layers to ten layers [12, 13].

In order to understand the $Sm\ C_\alpha^*$ structure, several models have been proposed. One of the most popular models is the phenomenological theory of liquid crystal based on the discrete model first developed by Cepic *et al* [4]. This model takes into account the configuration of each layer which was first introduced by Sun and Orihara [5]. 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 [4,9,10]. However, there is no systematic analysis performed to investigate the characteristic of each term in the free energy expansion in fluctuating and stabilizing the structure under an external applied field. We, therefore, are taking this challenge. In this paper we present 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 for the five-layer periodic structure with applied electric field. The three stable solutions corresponding to free energy model is presented. The effect of electric field on the structure is studied using finite-difference time domain method and changes of the phase structure are shown.

LANDAU FREE ENERGY EXPANSION OF DISCRETE MODEL IN AN EXTERNAL DC ELECTRIC FIELD

We consider a bulk sample of the $Sm\ C_\alpha^*$ phase, with five-layered periodical pitch. The molecular orientation within the j^{th} layer is described by the two-dimensional order parameter, shown in the Figure 1, as

$$\overrightarrow{\xi_j} = \xi_0\left(\cos j\alpha, \sin j\alpha\right) \tag{1}$$

where ξ_0 is the tilt magnitude and α is the tilt phase angle in each layer. Here, we assume Constant Amplitude Approximation (CAA), where the amplitude of tilt (ξ_0) is assumed constant in all layers and the phase difference of tilts between neighboring layers, $\alpha_{j+1} - \alpha_j$, is a constant.

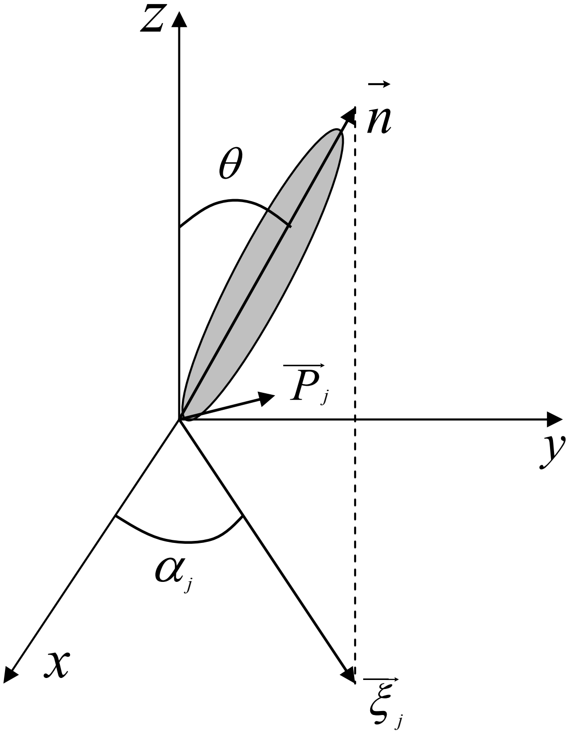


Figure 1: Presentation of the order parameters in the laboratory coordinate system.

Assuming the bulk sample is under the action of static external field, an expansion of the bulk free energy in the discrete order parameter ξ_j is

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

with $\vec{\sigma} = \varepsilon_0 c_p \vec{E}$ where ε_0 is the free space dielectric constant and c_p is the piezoelectric constant. The first two terms are the Van der Waal intra-layer interactions where the constants A and B are associated with the magnitude of the intra-layer interactions with $A = a_0\left(T - T_0\right)$. B is a material-dependent parameter. The third and the fourth term is the NN and the NNN interactions respectively. The last term describes the coupling of the spontaneous polarization of each layer with the external field.

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

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

Minimization of the free energy of the system with respect to phase angle without electric field gives the equilibrium structure. Three solutions are obtained, which are

$$\alpha = 0 \text{ and } \pi \tag{4}$$

$$\alpha = Cos^{-1}\left(-\frac{J_1}{J_2}\right) \tag{5}$$

The first solutions where $\alpha = 0$ and $\alpha = \pi$ correspond to the ferroelectric phase and antiferroelectric phase respectively. The second solution indicates the proposed structure of $Sm\ C_{\alpha}^{*}$ phase. This structure yields the global minimum and therefore our simulation is based on this solution.

NUMERICAL TECHNIQUE

In the bulk system of five-layered pitch with cyclic boundary condition, we assume the electric field $\vec{E}$ is applied in the x direction and is increased slowly in small increment. With each step, the system is fluctuated and is allowed to settle to its new equilibrium structure. The dynamic motion may be represented by the Landau-Khalatnikov equation of motion in the following forms,

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

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

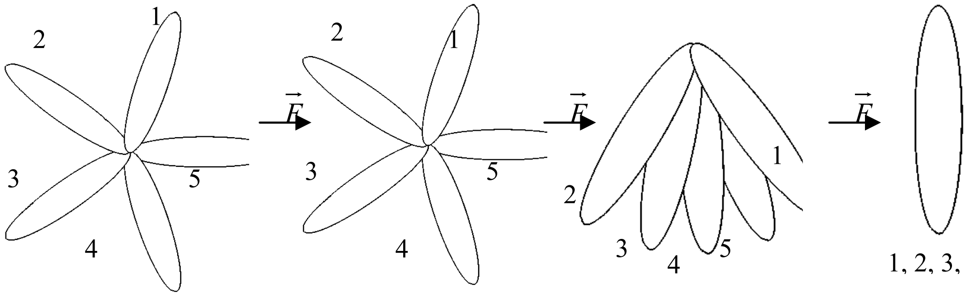
for the x and y component respectively. The change of the tilt can be calculated from these two equations using the free energy expression. Using the approximation $\Delta\xi_{j,x} \approx -\frac{\Delta t}{\gamma}\frac{\partial F}{\partial \xi_{j,x}}$ for the x component with increment of time Δt, the change of the tilt in each layer may be obtained. Similar approach is applied for the y component.

We begin our numerical procedure with predefined values of the tilt and phase angle. The initial approximation of the structure, $Sm\ C_{\alpha}^{*}$, is set to have some constant value of the tilt (CAA) and the symmetric orientation of the phase throughout the system. Then the change of the tilt under a small increment electric field is simulated. The calculation is repeated until the set of the tilt vectors stabilizes and satisfies a desired numerical precision. The simulation is stopped at some value of the critical field, where the structure of the $\mathrm{SmC}_{\alpha}^{*}$ phase is unwound, which corresponds to the tilt being perpendicular to the applied dc field.

RESULTS AND DISCUSSION

At equilibrium, we assume the tilt angle magnitude ξ_0 is approximately 0.1 radian. For a five-layer pitch structure, the initial equilibrium phase angle between each layer is taken to be $(2\pi/5)$ radian for an assumed symmetry orientation. We have chosen the following set of parameters: $A=-\left(1+\frac{\cos(2\pi/5)}{200}-\frac{1}{400\cos(2\pi/5)}\right)$, $J_1=\frac{A}{100}$, $J_2=-\frac{A}{100\cos(2\pi/5)}$ and $B=100$, for our simulation work.

Figure 2 shows the sequence of changes in the structure of the SmC_α^* phase. Figure 2(a) represents the equilibrium state of the structure without an external field. The structure changes slightly after we applying an electric field as shown in Figure 2(b). There is a continuous change when the field increases from $\sigma=0.02$ V/m to $\sigma=0.10$ V/m. At the field $\sigma=0.12$ V/m, a discontinuous change appears as shown in Figure 2(c). As the electric field reaches a critical value of $\sigma=11.58$ V/m, the fluctuated structure becomes unwound. The tilts of the five layers finally are aligned in the state where the tilts are nearly perpendicular to the direction of the applied electric field. This is as expected, in an unwound state all five layers collapse together at a phase angle of 270°, thus producing an induced polarization perpendicular to this angle, in the direction of the field.



(a) $\vec{\sigma}=0$ (b) $\vec{\sigma}=0.02$ (c) $\vec{\sigma}=0.12$ (d) $\vec{\sigma}=12.08$

Figure 2: (a), (b), (c) and (d) show the sequence of changes for $Sm\ C_\alpha^*$ phase under the influence of an external electric field.

Figure 3 illustrates the curve of phase angle (α) versus applied electric field using the same set of values. It is found that all layers converge to the unwound state as the field reaches the critical value, ~11.58 V/m, with 5 layers having a final phase angle as expected. It is also observed that the phase angle of each layer changes steeply at low field values before making its way gradually towards the unwound state as the field strength increases. Figure 4 shows the curve of Figure 3 for low values of electric field, for the range $\sigma=0$ to $\sigma=0.7$, to clearly visualize the changes of phase angle at low fields.

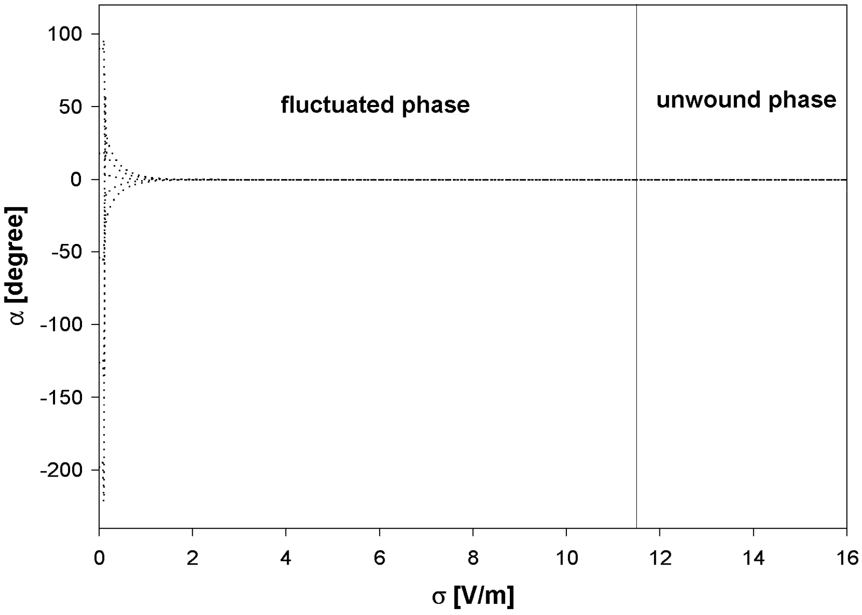


Figure 3: Phase angle (α) versus applied electric field

It would be interesting to compare the present results with our previous work on the electric field effects on a three-layer periodic structure of SmC_α^* [14]. Our present results are consistent with the results of the three layer pitch structure. In the three layer structure, the tilts also overlap at the angle of 270^0. Results for both systems reveals that the phase angle of the tilts at the unwound stage is perpendicular to the applied field.

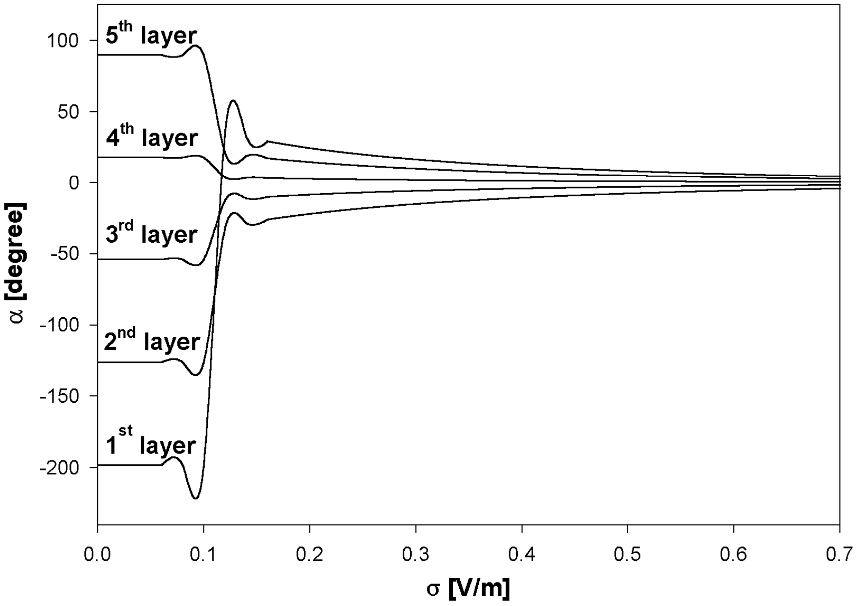


Figure 4: Zooming part of graph, phase angle (α) versus applied electric field, within the range from $\sigma = 0$ to $\sigma = 0.7$

CONCLUSION

In the unwound state, the tilts collapse to a single phase as we expected. In this work, we have neglected chirality contribution in the free energy since our aim is to concentrate on the role of the NN and the NNN terms towards changes in the $\mathrm{SmC}_{\alpha}^{*}$ phase with varying electric filed. The contribution of chirality may be important in pulling the third layer tilt 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 $\mathrm{SmC}_{\alpha}^{*}$ phase. This work is currently in progress and will be reported elsewhere.

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