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Effect of an external electric field on the smectic-C^*_{tilted} phases

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We are interested in the chiral tilted smectic subphases, smectic-C^*_{tilted}, including commensurate structures as well as the incommensurate smectic-C^*_α one. The continuum theory defines two scalar order parameters I and J, which can be coupled with an external electric field through a linear term $I \times E$ and a dielectric term $J \times E^2$. We have calculated the changes due to these terms in the phase diagrams in the (α,η) plane, where α comes from the spontaneous twist and η measures the strength of the biaxial order. The coupling with an external electric field induces the expansion of the areas with spontaneous polarization and the appearance of an electroazimuthal effect.

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I. INTRODUCTION

Liquid crystals exhibit many features that have been studied for years [1]. In particular, chiral smectic phases, in which rod-shaped chiral molecules are arranged in layers, may show interesting ferroelectric properties through a variety of subphases [1,2]. These subphases are designed simply with smectic-C^*_{tilted} because a specific molecular tilting arrangement with respect to the layer normal, which we take as the z axis, occurs in each one. Several techniques (calorimetry [3,4], ellipsometry [5,6], depolarized reflected light microscopy [7], dielectric spectroscopy [8], conoscopy [9], and resonant x-ray scattering [10,11]) allow us to distinguish one subphase from another [2,3,12–23]. These subphases have great technological potential for applications in fast electro-optical switching, especially in flat panel displays [2,12].

Typically, as molecules are arranged in layers, we are concerned with the laboratory coordinate system (x,y,z) and the smectic layer coordinate system (x_l,y_l,z_l). In Fig. 1, we represent the average molecular orientation in the lth smectic layer, defining the director $\vec{n}$, which varies on a case-by-case basis, and we show the axes for both the main coordinate frame (x,y,z) and layer coordinate frame (x_l,y_l,z_l), which is defined by taking the orientation of the main coordinate frame and simply rotating it about the z axis by an angle ϕ_0. In the case of the smectic-C^* phase, the director $\vec{n}$ is tilted nearly by the same angle θ from the layer normal [3,14], and its azimuthal orientation in the lth smectic layer is defined by an angle ϕ_l between the c-director $\vec{C_l}$ and the x_l axis [2,12]. The molecules in the smectic-C^*_A subphase tilt in opposite directions in adjacent lth and $(l+1)$th layers; from one layer to the next, the azimuthal angle varies from ϕ_l to $\phi_l+\pi$ [7,10]. The smectic-C^*_{Fi1} and smectic-C^*_{Fi2} subphases [24] have, respectively, three and four tilts in different azimuthal orientations, as shown by the resonant satellite x ray [10,11]. Eventually, the smectic-C^*_α subphase is incommensurate; it has a short-pitch helical structure where the ratio period over layer thickness gives a noninteger value in a subset of the interval 2 to 8. In the following, we consider these smectic-C^*_{tilted} subphases where molecules of the lth smectic layer tilt by a constant angle θ with respect to layer normal and their azimuthal orientation can be characterized by the angle ϕ_l, as shown in Fig. 1. Among chiral smectic compounds, the typical order of appearance of subphases on heating up is the following: smectic-C^*_A (anticlinic) $\rightarrow$ smectic-C^*_{Fi1} (intermediate with spontaneous polarization) $\rightarrow$ smectic-C^*_{Fi2} (intermediate without spontaneous polarization)$\rightarrow$ smectic-C^* (ferroelectric) $\rightarrow$ smectic-C^*_α (incommensurate) [3,14]. As matter of fact, one or more of these subphases, whose projections onto the smectic layer plane are shown in Fig. 2, may be missing in some materials, but the succession generally follows the same sequence. We can also mention subphases having higher periodicity. For instance, some with five layer periodicity (smectic-C^*_5) and another with six layer periodicity (smectic-C^*_6) were predicted theoretically [25,26] and were reported from experiment only by Wang *et al.*, Chandani *et al.*, and Takanishi *et al.* and not as widely as other structures [3,27–29].

In this paper, we report the results of a study of the influence of a moderate external electric field on the smectic-C^*_{tilted} subphases. In fact, the continuum theory defines two scalar order parameters I and J, which can be coupled with an external electric field through a linear term $I \times E$ and a dielectric term $J \times E^2$. We show that the changes due to these terms in the phase diagrams induce the expansion of the areas corresponding to subphases with spontaneous polarization. The results of our calculations also show that the azimuthal orientation depends on the electric field. That is why we predict the appearance of an electroazimuthal effect.

II. THEORETICAL APPROACH

The rich variety of structures that are observed in these smectic materials has initiated the development of several theoretical approaches to describe the tilted subphases [3,10,16–22,24,25,30–36]. The behavior of these subphases in an

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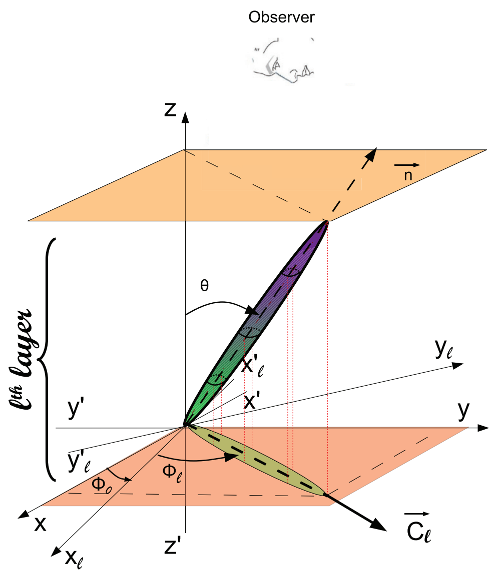


FIG. 1. (Color online) Schematic representation of the projection of a molecule onto the lth layer of a smectic-C^*_{tilted} subphase. θ and ϕ_l are the tilt and azimuthal angles.

external electric field attracts particular interest [25,36–39]. The majority of them consider the smectic layer as the basic element when taking into account the interactions between nearest-neighbor (NN) layers and even next-nearest-neighbor (NNN) layers [3,10,16,17,19–22,24,30–33].

In light of that, Dolganov *et al.* suggested a number of theoretical studies using the discrete phenomenological Landau model of the phase transition with a two-component order parameter. This approach is based on the vector $\overrightarrow{\xi_i}$, where i stands for the ith layer; it characterizes the tilt and the azimuthal orientation inside a smectic layer by the coordinates of the c director [30,39–41]. According to Dolganov *et al.* [39–41], one should drop the requirement of the constancy of the tilt angle to explain the formation of the smectic-C^*_6 subphase. In addition, for the sake of simplicity, it is possible to neglect the term related to chirality in the Landau expansion of the free energy.

Other approaches, disregarding the details of the structure on the smectic layer scale, are called continuum theories. For instance, one model was suggested by Hamaneh and Taylor (hereafter the HT model) [25,36]. That theoretical description links the appearance of the smectic-C^*_{tilted} subphases to a spontaneous microscopic twist, i.e., an angular increment α of the azimuthal angle from layer to layer. As developed by Dhaouadi *et al.* [35], an extension of the HT model introduced a new orientational order parameter that describes the contribution of the macroscopic polarization $\overrightarrow{P_S}$. Unfortunately, in this extension, the effect of electric fields is shown only with a linear term because the dielectric term is the same for all the subphases and does not depend on the azimuthal orientation. That can be considered an open question worthy of study. In fact, the behavior of the azimuthal orientation of a smectic-C^*_{tilted} subphase in an electric field constitutes the key idea of this work. Therefore, we suggest studying that effect using the continuum theory of tilted chiral smectic phases.

The basic idea of the continuum theory for smectics was put forward by de Gennes; an orientational order parameter $s_{ij} = n_i n_j - \frac{1}{3}\delta_{ij}$, where $\overrightarrow{n}$ is the director, was introduced to characterize uniaxial smectics [1]. The continuum theory of tilted chiral smectic phases introduces a macroscopic orientational order parameter (OOP) Q_{ij} [Eq. (1)], which is defined on a scale of at least ten layers and is valid in all smectic-C^*_{tilted} subphases taking into account the assumption of the constant angle tilt; Q_{ij} depends on two scalar order parameters, $J = \langle\cos(2\phi_l)\rangle$ and $I = \langle\cos(\phi_l)\rangle$, where the notation $\langle X\rangle$ represents the average of X taken on the variable inside the unit cell of the corresponding subphase. I and J describe, respectively, the uniaxial and biaxial orders [34]. Recently, we have reported the (α,η) phase diagrams of tilted chiral smectics, where α is a local angular parameter and η describes the variation of the temperature, without taking into account the effect of an external electric field [26]. In fact, we can see that the possible sequences are determined by the topology of a map in which the stable subphases appear as areas in a plot in which the free energy density is minimized with respect to the azimuthal angles. The effect of an external electric field on the smectic-C^*_{tilted} subphases may be readily taken into account through two terms: a linear term, $I \times E$, and a dielectric term, $J \times E^2$, due to the spontaneous polarization.

Briefly, the continuum theory of tilted chiral smectic phases explains the appearance of the tilted chiral smectics by the gain in electrostatic energy due to the lock-in of the unit cell to a number of layers that corresponds to the integer closest to the ratio of pitch over thickness of the subjacent smectic-C^*_α subphase [26,34].

For tilted chiral smectics, we can define a microscopic orientational order parameter S_{ij} obtained from $s_{ij} = n_i n_j - \frac{1}{3}\delta_{ij}$ through a rotation of angle θ around the x axis and another of angle ϕ around the z axis. The orientational order parameter

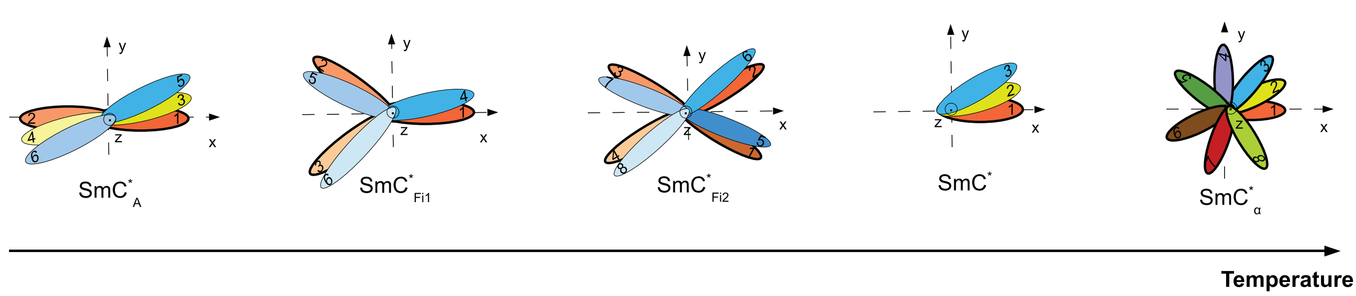


FIG. 2. (Color online) The five chiral smectic-C^*_{tilted} subphases drawn in the same order as usually observed upon heating. Ellipses, numbered by layer indices, represent the projections of the molecules onto the smectic layer plane.

Q_{ij} is the average of S_{ij} over the azimuthal angle ϕ inside the unit cell of the corresponding subphase [34]. Therefore, Q_{ij} reads as follows:

$$Q_{ij} = \left(1 - \frac{3}{2}\sin^2\theta\right)\begin{pmatrix} -1/3 & 0 & 0 \\ 0 & -1/3 & 0 \\ 0 & 0 & +2/3 \end{pmatrix} + \frac{J}{2}\sin^2\theta \begin{pmatrix} \cos 2\phi_0 & \sin 2\phi_0 & 0 \\ \sin 2\phi_0 & -\cos 2\phi_0 & 0 \\ 0 & 0 & 0 \end{pmatrix} - I\sin\theta\cos\theta \begin{pmatrix} 0 & 0 & \cos\phi_0 \\ 0 & 0 & \sin\phi_0 \\ \cos\phi_0 & \sin\phi_0 & 0 \end{pmatrix}. \quad (1)$$

As θ typically varies between 0 and $\frac{\pi}{8}$, we can use the approximation $\sin\theta \propto \theta$ and neglect terms with an order higher than 4 with respect to θ. Then, the Landau expansion for the free energy density describing the phase transition from the smectic-C_α^* to the smectic-C_{tilted}^* subphase can be written as [34]

$$\frac{F}{F_0} = \frac{1}{2}\langle(\Delta\phi - \alpha)^2\rangle - \eta J^2 - \gamma\sqrt{\eta}I^2, \quad (2)$$

where F_0 is the free energy density which is able to create a twist and is, in the smectic-C_α^* subphase, of the order of the 0.5 rad.

The above expression averaged over the unit cell of any tilted structure can be related to the expression introduced in the extension of the HT model as developed by Dhaouadi *et al.* [35]. The short-range term $F_{SR} = F_0\langle\cos(\Delta\phi - \alpha)\rangle$ was introduced empirically by HT [25,36]. On taking $\cos(\Delta\phi - \alpha) \backsimeq 1 - (\Delta\phi - \alpha)^2$, one gets $F_{SR} \backsimeq F_0(1 - \frac{1}{2}\langle(\Delta\phi - \alpha)^2\rangle)$ [34]. The long-range term represents the energy gain from the quadrupolar and dipolar ordering, which is given, respectively, by the J^2 and I^2 terms. The signs in front of these terms are opposite to the signs used in the extension of the HT model [35]. In fact, the J^2 term had been introduced for the first time by Hamaneh and Taylor in the long-range term [25,34,36]. Then, Dhaouadi *et al.* added a contribution due to the presence of the macroscopic polarization $\overrightarrow{P_S}$ in order to improve that approach [34,35]. Indeed, in the continuum theory of tilted chiral smectic phases, the long-range term comes from the difference of the quadrupolar density between the smectic-C_α^* subphase and the smectic-C_{tilted}^* ones, which is given by the full energy gain created due to the lock-in to commensurate subphases [34].

Expression (2) involves basically two parameters (α and η) and three variables ($\Delta\phi = \phi_{l+1} - \phi_l$, I, and J). α is the angular increment of $\Delta\phi$ in the smectic-C_α^* subphase; it is given by $\frac{2\pi}{n}$, where n is a noninteger number and is between 2 and 8. The terms ηJ^2 and $\sqrt{\eta}I^2$ come, respectively, from $\theta^4 J^2$ and $\theta^2 I^2$, which appear in the Landau expansion. η is an elastic coefficient which is dimensionless and has to be of the order of unity; according to the mean field approximation, its temperature dependence is due to $\theta^4 \sim (T - T_c)^2$, where T_c is the temperature where the tilt angle θ appears [26,34,35]. γ, which is also a dimensionless coefficient, depends only on the compound and not on the temperature, and it has to be of the order of 0.2 [26,35]. It must be noticed that the values of η and γ are chosen so that the terms $\eta\ J^2$ and $\gamma\sqrt{\eta}\ I^2$ are the same order of magnitude.

Continuing in the framework of the Landau–de Gennes theory, we can analyze the influence of an external electric field on the smectic-C_{tilted}^* subphases. Its contribution to the free energy density is given by

$$\Delta F_E = -P_i E_i - \tfrac{1}{2}\varepsilon_{ij}E_i E_j, \quad (3)$$

with ε being the dielectric permittivity tensor and P_i being the components of the polarization $\overrightarrow{P}$, which are given in the (x,y) plane by [1,34]

$$\begin{pmatrix} P_x \\ P_y \end{pmatrix} = C\begin{pmatrix} Q_{yz} \\ -Q_{xz} \end{pmatrix}, \quad (4)$$

where C is the flexoelectric constant.

In the uniaxial approximation, there are only two principal components of the local tensor ε_{ij}, $\varepsilon_\parallel$ and $\varepsilon_\perp$ [1]. To obtain the dielectric permittivity tensor of the tilted chiral smectic structure, we should imagine that this local tensor rotates in the laboratory coordinate system.

$$\varepsilon_{ij} = \left(\frac{\varepsilon_\parallel + 2\varepsilon_\perp}{3}\right)\delta_{ij} + (\varepsilon_\parallel - \varepsilon_\perp)Q_{ij}. \quad (5)$$

We assume that the electric field is applied in the y direction and that $J\ \sin^2\theta \sim \theta^2 J$ and $I\sin\theta\cos\theta \sim \theta I$. Taking into account all the expressions reported above, we can formulate the core of the question as consisting essentially of the addition of the following contribution to the free energy density:

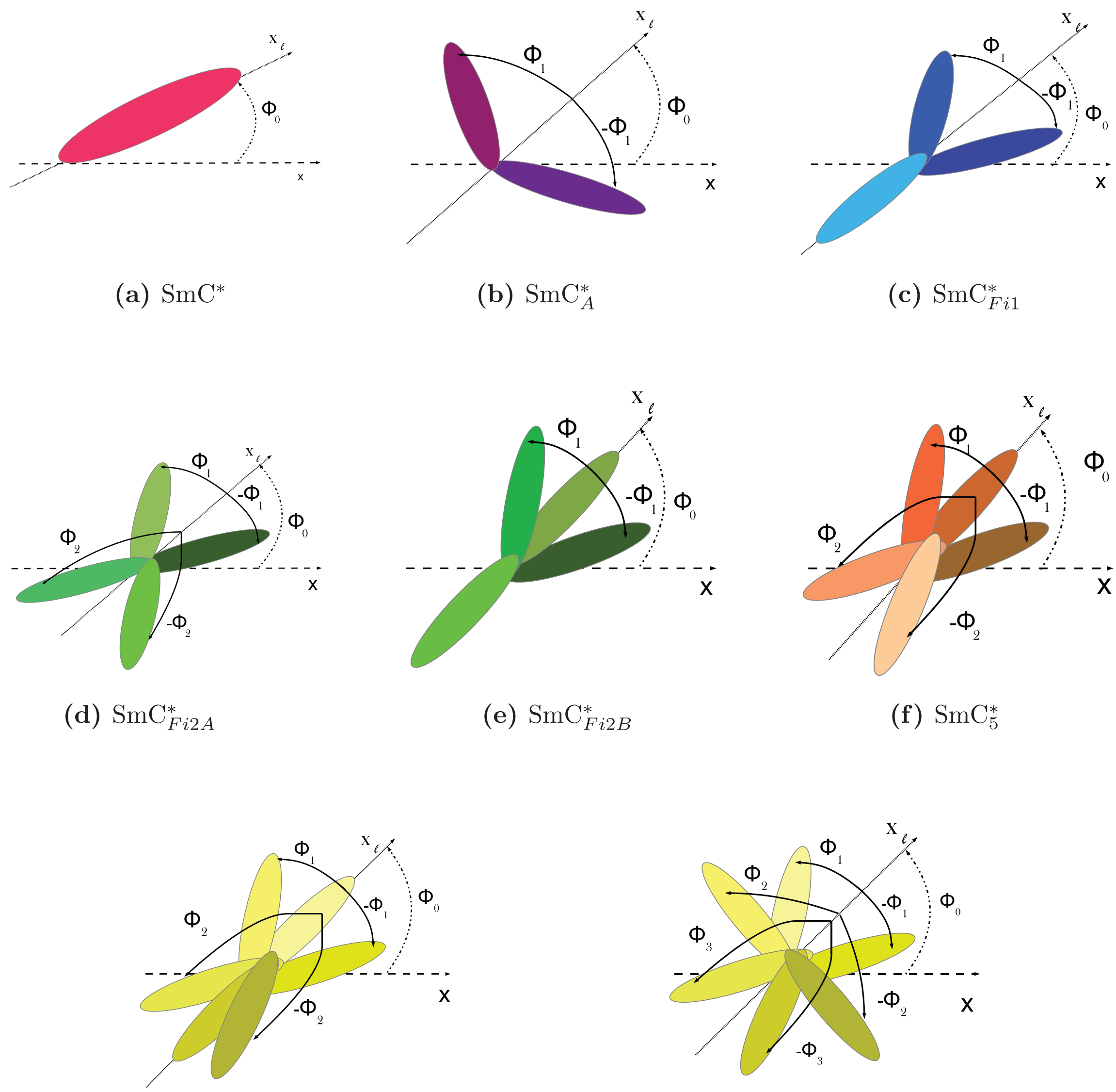
$$\frac{\Delta F_E}{F_0} = -\delta\sqrt[4]{\eta}\ I\cos(\phi_0) - k\delta^2\sqrt{\eta}J\cos(2\phi_0). \quad (6)$$

It must be noticed that the coefficient $\delta \sim \sqrt{\frac{C\varepsilon_0\chi}{F_0}}E$ is without dimension and depends only on the field electric magnitude, and neither the temperature nor the compound can disturb its value. The coefficient $k \sim \frac{10^8(\varepsilon_\parallel - \varepsilon_\perp)}{\sqrt{32\gamma}\varepsilon_0\chi}$ is a dimensionless constant depending on the compound and has to be of the order of unity.

In order to restrict the number of the estimated smectic-C_{tilted}^* subphases, we can assume that the symmetry is broken in such a way that the distribution of the ϕ_l is not symmetric about the x_l axis, so it constrains the terms $\langle\sin(2\phi_l)\rangle$ and $\langle\sin(\phi_l)\rangle$ to vanish [25,26]:

$$\langle\sin(2\phi_l)\rangle = 0; \quad \langle\sin\phi_l)\rangle = 0. \quad (7)$$

Figure 3 summarizes all the smectic-C_{tilted}^* structures which are physically possible from the subphase with one layer periodicity up to the subphase with six layer periodicity. In fact, it can be seen that the subphases mentioned above appear clearly through these schematic drawings of the projections of the molecules onto the layer plane in Fig. 3. We must clarify that the smectic-C_{Fi2}^* and the smectic-C_6^* subphases have two possible variants of molecular assembly indexed by the letters A and B. It should be mentioned that any gap or overlap between molecules is due to the handmade drawing and has no physical meaning.



(a) SmC* (b) SmC_A^* (c) SmC_{Fi1}^*

(d) SmC_{Fi2A}^* (e) SmC_{Fi2B}^* (f) SmC_5^*

(g) SmC_{6A}^* (h) SmC_{6B}^*

FIG. 3. (Color online) The predicted smectic-C_{tilted}^* structures.

III. RESULTS AND DISCUSSION

We calculate the free energy density describing the phase transition from the smectic-C_α^* to smectic-C_{tilted}^* subphase using Eqs. (2) and (6). The minimization of this free energy is established through a combination of analytical and numerical methods in the hope of computing the complete phase diagrams, which are plotted in coordinates α versus η. To determine with precision the stable structures which correspond to the minimum of energy, the numerical transaction of minimization was repeated about 10^6 times.

Figure 4 illustrates the phase diagrams obtained for $\gamma = 0.2$ and some values of δ. These phase diagrams are a graphical representation which is able to tell us what subphase has the lower free energy density. This diagram is similar to that obtained without external electric field [Fig. 4(a)] [26]. In these phase diagrams, the increase in the value of δ does modify the domains corresponding to the smectic-C_{tilted}^* subphases where parameters I and J are not null.

Regarding the phase diagram without an external electric field obtained for $\gamma = 0.2$ and $\delta = 0$ and shown in Fig. 4(a), by following the vertical dashed line at constant η of about 0.8 from the bottom to the top, we begin at the smectic-C^* subphase and then, with an increasing angular increment α, cross the smectic-C_{Fi2}^* subphase and after that the smectic-C_{Fi1}^* subphase; by the end, we enter the smectic-C_A^* subphase. The smectic-C_α^*, smectic-C_{tilted}^* sequence is observed along the horizontal line at a constant angular increment α. For instance, the sequence smectic-C_α^* $\rightarrow$ smectic-C_{Fi2}^* $\rightarrow$ smectic-C^* is observed from right to left along the dashed line, which corresponds to $\alpha = \frac{3\pi}{7}$, as shown in Fig. 4(a).

With regard to the phase diagram in an electric field drawn in Fig. 4(b) for $\gamma = 0.2$ and $\delta = 0.1$, we note that the smectic-C_5^* and smectic-C_6^* subphases appear in a small area on the left-hand side of the diagram. By increasing δ [Figs. 4(c) and 4(d)], the smectic-C^*, smectic-C_{Fi1}^*, smectic-C_5^*, and smectic-C_6^* subphases clearly expand, to the detriment of the smectic-C_α^*

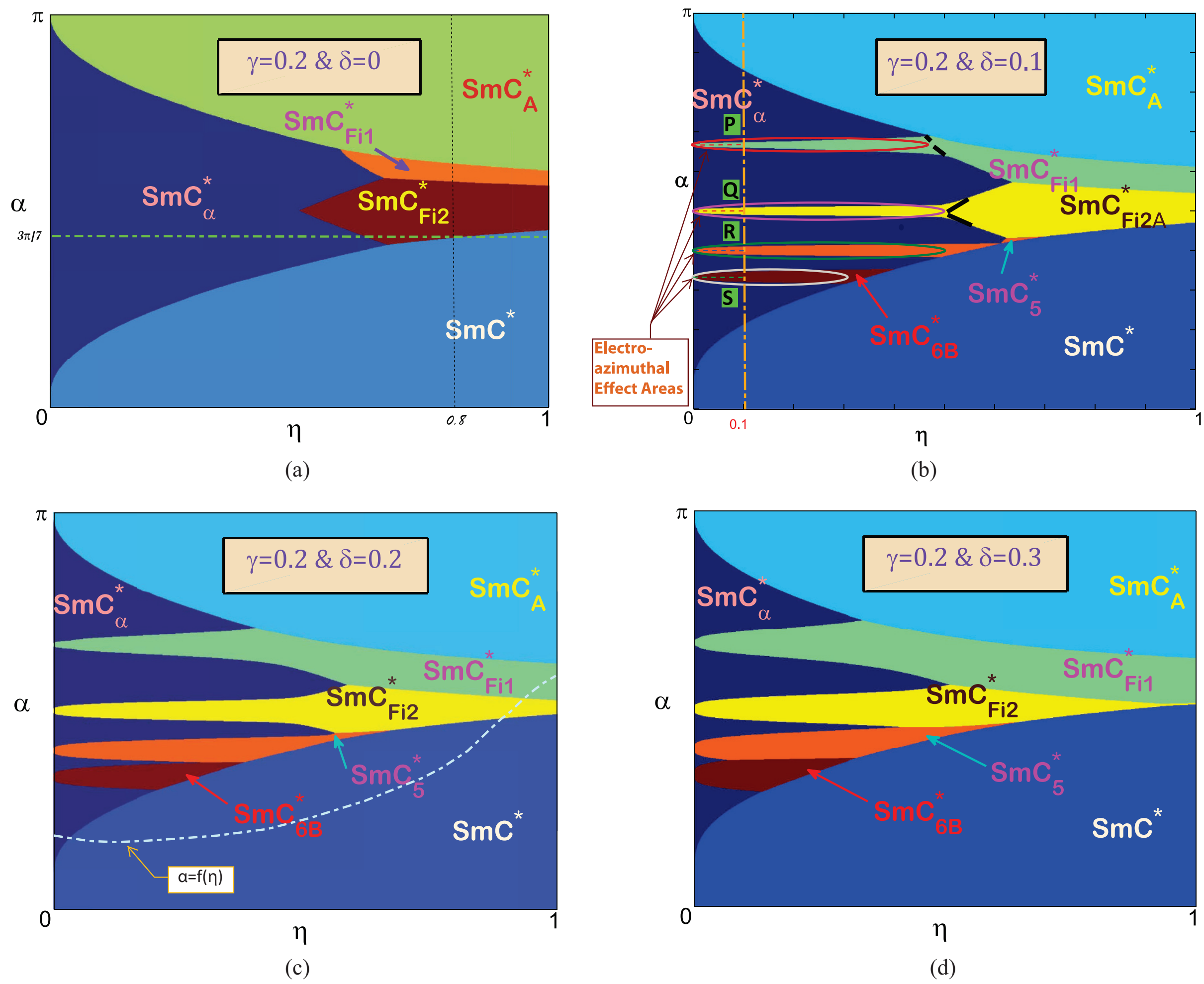


FIG. 4. (Color online) Phase diagrams. (a) The case $\gamma = 0.2$ and $\delta = 0$. The vertical dashed line corresponds to $\eta = 0.8$. The horizontal dashed line corresponds to $\alpha = \frac{3\pi}{7}$. (b) The case $\gamma = 0.2$ and $\delta = 0.1$. (c) The case $\gamma = 0.2$ and $\delta = 0.2$. The dashed line shows one possibility for the form of $\alpha = f(\eta)$. (d) The case $\gamma = 0.2$ and $\delta = 0.3$.

subphase. In addition, the domain of the smectic-C^*_{Fi2} narrows on the right-hand side of the diagram because it becomes hidden by the smectic-C^* subphase.

When we zoom in on the diagram obtained for $\gamma = 0.2$ and $\delta = 0.1$ [Fig. 4(b)], we can note the expansion of areas corresponding to the subphases with a spontaneous polarization (smectic-C^*, smectic-C^*_{Fi1}, smectic-C^*_5, and smectic-C^*_{6B}) by increasing the value of δ; this expansion is increasingly important for the subphases with lower periodicity. For the low values of η, the areas surrounded with ellipses take the form of tongues whose width grows with δ. The mentioned areas look like they correspond to the smectic-C^*_{Fi2A}, smectic-C^*_{Fi1}, the smectic-C^*_5, and smectic-C^*_{6B} subphases. Actually, they correspond to a distorted version of the smectic-C^*_α subphase.

Numerical calculations show that the parameter I is proportional to the external electric field. By analogy with the electroclinic effect [2], this phenomenon can be called the electroazimuthal effect. We grant this issue to the paraelectric property of the structure. For instance, for points P(0.1, $\frac{2\pi}{3}$), Q(0.1, $\frac{\pi}{2}$), R(0.1, $\frac{2\pi}{5}$), and S(0.1, $\frac{\pi}{3}$) shown in the diagram obtained for $\gamma = 0.2$ and $\delta = 0.1$ [Fig. 4(b)], we plot the variation of $I = \langle \cos(\phi) \rangle$ as a function of δ, and we get a linear curve for each point, as shown in Fig. 5.

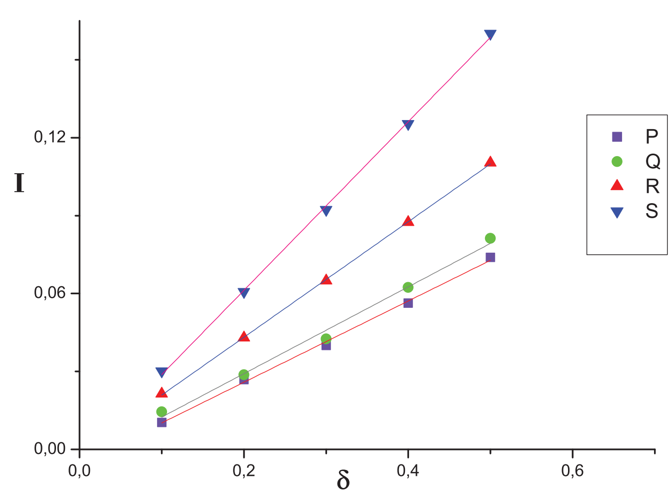


FIG. 5. (Color online) Points P(0.1, $\frac{2\pi}{3}$), Q(0.1, $\frac{\pi}{2}$), R(0.1, $\frac{2\pi}{5}$), and S(0.1, $\frac{\pi}{3}$).

In order to make a comparison with experiment, we have experimentally studied two compounds in our group: C7F2 and C10F3, the (E,T) phase diagrams of which are confirmed by Dhaouadi *et al.* through the extension of the HT model [35]. For each compound, a comparative study of the phase sequence observed under an electric field enables us to confirm a high level of concordance between results obtained numerically and those given by the experiment. Indeed, the dependence of α and η on the temperature T leads us to represent in Fig. 4(b) an estimated path $\alpha = f(\eta)$ to convert the (α,η) phase diagram into a prediction of the sequence in which the different subphases appear when only the temperature is raised or lowered and the other material parameters are kept constant. The most common form of this path would be a curve connecting the lower left to the upper right of the (α,η) phase diagram [25,35]. In fact, by following this path, we can determine the phase sequence obtained as the temperature is lowered. As we move up the curve, we pass from the smectic-A, represented by the vertical axis along which $\eta = 0$, to the incommensurate smectic-C_α^* subphase. Decreasing the temperature further takes us into the ferroelectric smectic-C^* subphase. We then pass into the intermediate smectic-C_{Fi2}^* subphase. Finally, we reach the smectic-C_{Fi1}^* subphase. The sequence smectic-$A \rightarrow$ smectic-$C_\alpha^* \rightarrow$ smectic-$C^* \rightarrow$ smectic-$C_{Fi2}^* \rightarrow$ smectic-C_{Fi1}^* has been observed under a relatively low external electric field ($E \sim 0.8$ V/μm) in C10F3 [35,42–44]. Similar lines can be drawn to trace the path of the phase sequence for each compound. In addition, we predict the appearance of some subdomains due to the electroazimuthal effect in the area corresponding to the smectic-C_α^* subphase. This result confirms many experimental indications which have always been neglected and assigned to disruptive factors, particularly for the compounds with a wide zone corresponding to the smectic-C_α^* subphase such as C7F2 [35,45].

IV. CONCLUSION

In conclusion, using the continuum theory of the tilted chiral smectics [34], we have studied the effect of a moderate external electric field on the smectic-C_{tilted}^* subphases. By adding two suitable terms to the free energy density expression, numerical and analytical methods of minimization lead us to show precisely the domain of stability physically possible for each structure. Consequently, this operation enables us to build the phase diagram for different values of δ in the (α,η) plane. By comparing them with those obtained without an external electric field [26], our results show that there is an expansion of the areas corresponding to the smectic-C^*, smectic-C_{Fi1}^*, and smectic-C_5^* subphases by increasing the value of δ. In addition, the analysis of the obtained phase diagrams reveals that an electroazimuthal effect can take place in some areas of the (α,η) plane and induced distorted versions of the smectic-C_α^* subphase. Finally, taking into account the electric field effect gives results that appear to be in good agreement with experiment and allows us to explain different phase sequences observed under the field. In the future, we plan to develop the experimental study of the influence of an external electric field on compounds in which the domain corresponding to the smectic-C_α^* subphase is large in order to reveal the behavior of the tilted chiral smectics under the electroazimuthal effect. In addition, we plan to study the effect of higher fields and to introduce the helicity and its sign evolution in the phase sequence.

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