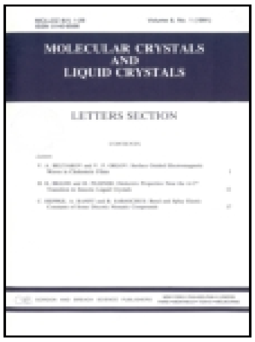
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Fredericks Transition in Pure and Nanodoped Surface Stabilized Ferroelectric Liquid Crystals

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Recently, ferroelectric liquid crystals (FLCs) doped with nanoparticles have attracted a wide interest not only from a scientific but also from a practical point of view and there is continuously growing interest in the effects caused by doping the ferroelectric nanoparticles. Furthermore, the presence of external fields exhibits a very interesting transition, reported by recent experiments. In this work, we investigate the response of a surface stabilized ferroelectric liquid crystal (SSFLC) doped with ferroelectric nanoparticles to an applied electric field. We assume that the smectic layers consist of uniform planes with a fixed orientation and the system is free from dislocation of constant layer thickness due to nanoparticles. We obtain the threshold field of orientational transition and the maximum deviation of the polarization vectors for a pure and a doped SSFLC medium. Then, we discuss the Fredericks transtion of the system and formation of inhomogeneous texture. It is found that the ferroelectric nanoparticles have significant influences in ordering behavior of a SSFLC medium and the threshold fields are critically changed by doping nanoparticles in the SSFLC, which is fundamental to operation of many liquid crystal devices.

Keywords Ferroelectric liquid crystal; nanoparticle; Fredericks transition

1. Introduction

It is well known that the ferroelectric liquid crystals (FLCs), exhibit smectic layer structures in which the director **n**(r), describing the time averaged direction of the long molecular axis, tilts with respect to the layer normal at an angle θ. The director precesses together with the local polarization about the layer normal axis. The polarization vector forms a helicoidal structure with the characteristic pitch. FLCs possess a permanent spontaneous polarization which is parallel to the layers [1]. Applying an external field has orientational effects on both director and polarization vectors. A Fredericks transition due to the presence of the spontaneous polarization in SmC* layers has been predicted by Zimmerman *et al.* [2]. These systems, recently, have attracted a wide interest not only from a scientific but also from a practical point of view [3–4]. Moreover, in recent years, there is continuously growing interest in the effects caused by doping the ferroelectric nanoparticles to FLCs [5]. Nanoparticle-doped liquid crystals have been studied broadly [6–8]. The theory of ferroelectric nanoparticles in nematic liquid crystals has been previously developed by Lopatina, et al [9]. Obviously, the presence of nanoparticles in FLC medium creates strong local fields that produce large alignment effects over the distribution of the nano dispersions [10–11]. Thus, the properties of these nanostructure materials can be significantly changed.

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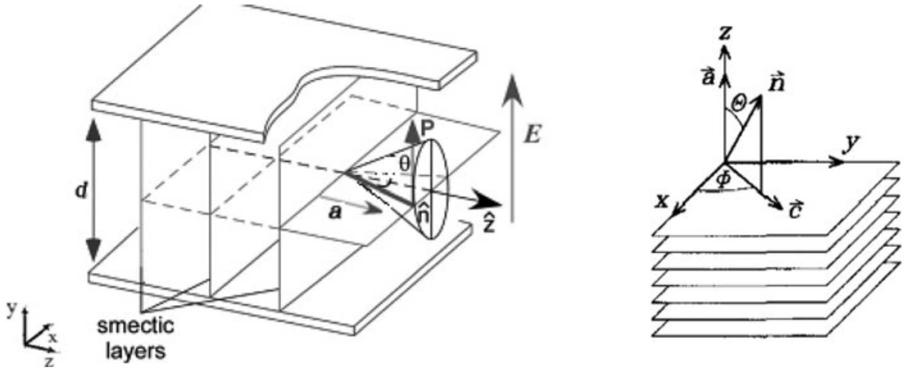

Figure 1. The geometry of the SSFLC system and the orientation of the directors with respect to the smectic layer normal.

In addition, the presence of external fields exhibits a very interesting transition, reported by recent experiments [12].

In this work, we investigate the response of a surface stabilized ferroelectric liquid crystal (SSFLC) doped with ferroelectric nanoparticle to an applied electric field. We assume that the smectic layers to consist of uniform planes with a fixed orientation and the system is free from dislocation of constant layer thickness due to nanoparticles. We obtain the threshold field of orientational transition and the maximum deviation of the polarization vectors for a SSFLC doped with nanoparticles. Then, we discuss the orientational order of the system and formation of inhomogeneous texture.

The article is organized as follows. In section 2, after formulation of the problem, the equation for threshold field is obtained. Then, the maximum deviation of the polarization vectors is calculated. In section 3, the calculations are performed for a nano-doped system and the transition field is obtained. Next, the variations of the director profile due to the presence of nanoparticles are determined. We, finally, make some concluding remarks, in the last section.

2. Fredericks Transition in a Pure SSFLC

We consider a stabilized ferroelectric liquid crystal cell of width d, placed in an electric field $\mathbf{E}$. In this work, we assume that the smectic layers consist of uniform planes with a fixed orientation parallel to the x-y plane (Fig. 1). The layer normal, thus being parallel to z axis, is denoted by a unit vector $\boldsymbol{a}$. Also, we assume the system is free from dislocation of layer thickness. Hence, the layer normal must fulfill the constraint [13]

$$\nabla \times \mathbf{a} = 0, \tag{1}$$

The average orientation of the liquid crystal molecular long axes is described by a unit vector called director $\mathbf{n}$, while the projection of the director into the smectic layers is denoted by a unit vector $\mathbf{c}$, called $\boldsymbol{c}$-director. In order to describe the orientation of the $\boldsymbol{c}$-director, an azimuthal angle φ, which is the angle between the $\boldsymbol{c}$-director and the x-axis, is introduced:

$$c = (\cos\varphi, \sin\varphi, 0)\,. \tag{2}$$

A unit vector **b** is introduced according to

$$\mathbf{b} = \mathbf{a} \times \mathbf{c}. \tag{3}$$

This unit vector will coincide with the spontaneous polarization vector ($\mathbf{P}\mathrm{s} = P_s\mathbf{b}$) [1].

Without applying an external field and in the absence of the nanoparticle, the ground state has a homogeneous structure. However, at a sufficiently large electric field$\mathbf{E} = E\mathbf{j}$, the Fredericks transition is induced. The presence of a spontaneous polarization leads to the torques on $\boldsymbol{P}$ and **c**-director. The polarization tends to be aligned with the field and on the other hand, it's coupling to the **c**-director leads to an additional torque on **c**, in an applied electric field. The director is strongly anchored at the boundaries and hence, the external field can cause an inhomogeneous texture. Also, the restoring orientational elastic torques, acts on the system as well. Then, the stable equilibrium state can be found by minimizing the total free energy with respect to variations in the **c**-director. In one constant approximation, the total density of free energy is written as

$$F = \frac{B}{2}\left(\frac{\partial\varphi}{\partial y}\right)^2 - \frac{1}{2}\varepsilon_a\varepsilon E^2\sin^2\theta\sin^2\varphi - P_sE\cos\varphi. \tag{4}$$

The first term comes from Frank free energy, in which B is the elastic constant of the ferroelectric liquid crystal. The second term is the contribution of the electric field to the density of energy, in which ε_a is the dielectric anisotropy and ε is the permittivity of the space. The last term describes the interactions of the electric field with the spontaneous polarization of the liquid crystal.

An applied electric field has an orientational effect on the director due to the dielectric anisotropy. It should be noted that if the dielectric anisotropy is positive, a parallel alignment of the director with the external field is energetically preferable and in the reverse case, if it is negative, a perpendicular alignment will be favorable.

The value of φ(y) at the critical field is calculated by minimizing the total free energy. Then, using the Euler-Lagrange equation and small values of φ(y) leads to

$$\left(\frac{d\varphi}{dy}\right)^2 = \left(\varphi_m^2 - \varphi^2\right)\left(\frac{\varepsilon_a\varepsilon}{B}\sin^2\theta E^2 - \frac{P_sE}{B}\right), \tag{5}$$

where φ_m is the maximum deviation of the director and can be determined variationally. Next, by performing the integrals of Eq. (5), from $y = 0\,\mathrm{to}\,y = \frac{d}{2}$ and $\varphi = 0\,\mathrm{to}\,\varphi = \varphi_m$, the threshold field is obtained as

$$E_{th} = \frac{P_s}{2\varepsilon_a\varepsilon\sin^2\theta}\left[1 \pm \sqrt{1 + \frac{4\varepsilon_a\varepsilon\sin^2\theta}{P_s^2}\frac{B\pi^2}{d^2}}\right]. \tag{6}$$

This formula contains both the conventional Fredericks transition behavior, [14–15], due to the dielectric torques, and the polarization Fredericks effect due to the ferroelectric torques. Obviously, at $E = E_{th}$ the Fredericks transition is induced by forcing the polarization and the director to reorient everywhere except at the slab boundaries. In the case of positive dielectric anisotropy with antiparallel alignment of the external field and polarization ($P_sE < 0$), the threshold field is reduced, or for the state of negative dielectric anisotropy with parallel alignment of the field and polarization ($P_sE > 0$), the threshold field is therefore increased. Indeed, due to the coupling of the spontaneous polarization and director, both orientational effects can enhance each other in the antiparallel case ($\varepsilon_a > 0$) and therefore, the threshold field is reduced. However, both effects contract each other in the parallel case ($\varepsilon_a < 0$) and hence increase the critical field. These obtained results show good agreement with the obtained solutions by Zimmerman *et al.* [2].

Moreover, to find the maximum deviations of the director, we first rewrite the total density of energy in an approximate form, keeping only the fourth order of φ,

$$F = \frac{B}{2}\left(\frac{\partial\varphi}{\partial y}\right)^2 - \frac{\varphi^2}{2}\left(\varepsilon_a\varepsilon\sin^2\theta E^2 - P_sE\right) + \frac{\varphi^4}{4}\left(\frac{2}{3}\varepsilon_a\varepsilon\sin^2\theta E^2 - \frac{P_sE}{6}\right) - P_sE. \quad (7)$$

Next, minimizing the total free energy with respect to φ, yields

$$\phi(y) = \varphi_m\cos\left(\tilde{k}y\right) + \phi_{\mathrm{m}}^3\frac{1}{\tilde{k}^2 - 9\tilde{k}^2}\frac{\varepsilon_a\varepsilon\sin^2\theta}{6B}\left(E^2 - \frac{P_sE}{4\varepsilon_a\varepsilon\sin^2\theta}\right)\left[\cos\left(3\tilde{k}y\right) - \cos\left(\tilde{k}y\right)\right], \quad (8)$$

where we have used the following definition

$$\tilde{k}^2 = k^2 + \Delta, \quad (9)$$

in which

$$k = \sqrt{\frac{\varepsilon_a\varepsilon\sin^2\theta}{B}\left(E^2 - \frac{P_sE}{\varepsilon_a\varepsilon\sin^2\theta}\right)}; \quad \Delta = -\frac{\varepsilon_a\varepsilon\sin^2\theta}{2B}\left(E^2 - \frac{P_sE}{4\varepsilon_a\varepsilon\sin^2\theta}\right)\phi_{\mathrm{m}}^2. \quad (10)$$

Clearly, φ_m indicates the maximum deviations of the director that can be determined by using the boundary condition at the walls of the slab$\varphi\left(\pm\frac{d}{2}\right) = 0$. Finally, we obtain

$$\varphi_m^2 = 2\left(E^2 - \frac{P_sE + (B\pi^2/d^2)}{\varepsilon_a\varepsilon\sin^2\theta}\right)\Big/\left(E^2 - \frac{P_sE}{4\varepsilon_a\varepsilon\sin^2\theta}\right). \quad (11)$$

It is evident that the solution $\varphi_m = 0$ is for$E < E_{th}$, when there is no inhomogeneous texture.

3. Fredericks Transition in a Doped SSFLC with Ferroelectric Nanoparticle

In this section we determine the threshold field of the orientational transition for a system of ferroelectric liquid-crystal doped with ferroelectric nanoparticle and calculate the maximum deviation of the director in the system. The presence of ferroelectric nanoparticle affects the Fredericks transition threshold due to the strong local electric field around the ferroelectric particle and coupling between the director and the polarization of the particle. In order to obtain the threshold field, a further key assumption is to consider the case in which the ferroelectric nanoparticle is sufficiently small not to disturb the texture in its neighborhood. Near the nanoparticle, the electric field changes as a function of position but the liquid crystal order cannot follow these variations of the field, because it would cost too much energy. It is evident that coupling between the nanoparticles and liquid crystal orientations produces a small deviation of the polarization with respect to the y direction. Then, assuming that the tilt angle is constant [16], the total free energy density in one constant approximation, can be written as follows

$$F = \frac{B}{2}\left(\frac{\partial\varphi}{\partial y}\right)^2 - \frac{1}{2}\varepsilon_a\varepsilon\left(\mathbf{n}.\mathbf{E}\right)^2 - \mathbf{P}_s.\mathbf{E} - \mathbf{P}_s.\mathbf{E}_{np}(r) - \frac{1}{2}\varepsilon_a\varepsilon\left(\mathbf{n}.\mathbf{E}_{np}(r)\right)^2 - \mathbf{p}_{np}.\mathbf{E} \quad (12)$$

In this equation, the first three terms are the same terms defined for the pure system (Eq. (4)). The director **n,** as defined in previous section is given by $\mathbf{n} = \mathbf{a}\cos\theta + \mathbf{c}\sin\theta$. The remaining terms indicates the interaction energies of the particles with the surrounding SSFLC and the applied electric field. In these terms, $\mathbf{E_{np}}(r)$ is the electric field of ferroelectric nanoparticle and $\mathbf{p}_{np}$ is defined as the dipole moment of the ferroelectric nanoparticle. The field of nanoparticle, taken as a field of the element of electric dipole moment, is given by

$$\mathbf{E_{np}}(r) = \frac{1}{4\pi\varepsilon}\left[\frac{3r\left(r.p_{np}\right)}{r^5} - \frac{p_{np}}{r^3}\right], \tag{13}$$

neglecting the high order corrections caused by the electric anisotropy of the liquid crystal. Then, for sufficiently small nanoparticles and low concentrations, we can integrate the free energy of the interactions over whole volume of the system. We consider a low concentration of nanoparticles dispersed in liquid crystal and assume that the nanoparticles are spheres with radius R. Also, we make additional notations: $\emptyset_n$is the volume fraction of nanoparticles, $P_{np} = p_{np}/V_n$is the dipole moment of nanoparticles per unit volume and V_n is the nanoparticle's volume. Also, the strong orientational coupling between the particles and SSFLC is suggested and no interaction between the particles is assumed. Considering the small deviations of polarization vectors, as a result we obtain the total free energy as the following expression:

$$\begin{aligned} F = {} & \frac{B}{2}\left(\frac{\partial\varphi}{\partial y}\right)^2 - \frac{1}{2}\varepsilon_a\varepsilon E^2 sin^2\theta sin^2\varphi - P_s E\cos\varphi - \frac{4}{3}\pi\emptyset_n R^3 E P_{np}\sin\theta_1\cos\varphi_1 \\ & + \frac{1}{1728\varepsilon}\varepsilon_a^2\pi^2\left(r^{-3} - R^{-3}\right)P_{np}^2\emptyset_n R^6\left\{cos^2\theta\left[62 + 26Cos2\theta_1\right]\right. \\ & + sin^2\theta\left[36cos^2\theta_1 \; + \left(38 - 11Cos2\left(\varphi - \varphi_1\right)sin^2\theta_1\right)\right] \\ & \left. -4Sin2\theta Sin2\theta_1 Sin\left(\varphi - \varphi_1\right)\right\} - \frac{3}{8\varepsilon}\pi^2\emptyset_n \ln(\frac{r}{R})P_{np}P_s Cos\left(\varphi - \varphi_1\right), \end{aligned} \tag{14}$$

where r is defined as a radius of a cut sphere around the particles that gives the upper bounds beyond which the effect of particle becomes negligible, $\varphi_1$and $\theta_1$are respectively defined as the azimuthal and polar angles of the polarization vector of the nanoparticles. Next, we consider that φ_m is the value of φ in $y = 0$ and derive a stationary equation of the ***c***-director. Then, minimizing the total free energy and applying some approximations leads to

$$\begin{aligned} \left(\frac{d\varphi}{dy}\right)^2 = \left(\varphi_m^2 - \varphi^2\right) & \left[\frac{\varepsilon_a\varepsilon E^2 sin^2\theta}{B} - \frac{P_s E}{B} + \frac{11}{864}\frac{\varepsilon_a^2\pi^2 sin^2\theta sin^2\theta_1}{\varepsilon B}R^3\emptyset_n\right. \\ & \left.\times\left(1 - \frac{R^3}{r^3}\right)P_{np}^2 - \frac{3}{8\varepsilon B}\pi^2\emptyset_n P_{np}P_s ln\left(\frac{r}{R}\right)\right]. \end{aligned} \tag{15}$$

Performing the integrals of Eq. (15) from $y = 0$ to $y = \frac{d}{2}$ and $\varphi = 0$ to $\varphi = \varphi_m$, and then taking the average value for orientation of the nanoparticles, yields the threshold field

$$E_{th} = \frac{P_s}{2\varepsilon\varepsilon_a}\left[1 \pm \sqrt{1 + 4\frac{\varepsilon_a\varepsilon^2}{P_s^2}\frac{B\pi^2}{d^2} + \frac{11d^2\pi^2\varepsilon_a\phi_n R^3 P_{np}^2 sin^2\theta}{432d^2P_s^2}\left(1 - \frac{R^3}{r^3}\right) + \frac{3}{2}\frac{\pi^2\phi_n ln(\frac{r}{R})P_{np}}{P_s}}\right]. \tag{16}$$

The maximum deviations of the ***c***-director can be calculated by a similar method, given in the previous section. Using the new introduced variables leads to the following expression

$$\begin{aligned}\varphi(y) = \varphi_m \cos\left(\tilde{k}y\right) + \varphi_m^3 \frac{1}{\tilde{k}^2 - 9\tilde{k}^2} \frac{\varepsilon_a \varepsilon sin^2\theta}{6B} \Bigg(E^2 - \frac{P_s E}{4\varepsilon_a \varepsilon sin^2\theta} \\ - \frac{11}{3456}\frac{\varepsilon_a}{\varepsilon}\pi^2 R^6 sin^2\theta_1 \emptyset_n P_{np}^2 - \frac{3\pi^2 ln\left(\frac{r}{R}\right)\emptyset_n}{32\varepsilon_a\varepsilon^2 sin^2\theta} P_{np} P_s \Bigg) \\ \left(\cos\left(3\tilde{k}y\right) - \cos\left(\tilde{k}y\right)\right). \end{aligned} \tag{17}$$

where

$$k = \sqrt{\frac{\varepsilon_a \varepsilon sin^2\theta}{B}\left(E^2 - \frac{PE_s}{\varepsilon_a\varepsilon sin^2\theta} + \frac{11}{864}\frac{\varepsilon_a}{\varepsilon}\pi^2\emptyset_n R^3 sin^2\theta_1\left(1 - \frac{R^3}{r^3}\right)P_{np}^2 - \frac{3}{8}\frac{\pi^2 ln\left(\frac{r}{R}\right)\emptyset_n}{\varepsilon_a\varepsilon^2 sin^2\theta_1}P_{np}P_s\right)}, \tag{18}$$

$$\Delta = \frac{-\varepsilon_a\varepsilon sin^2\theta}{2B}\left(E^2 - \frac{P_s E}{4\varepsilon_a\varepsilon sin^2\theta} - \frac{1}{4}\frac{11}{864}\frac{\varepsilon_a\pi^2}{\varepsilon}P_{np}^2\emptyset_n R^6 sin^2\theta_1 - \frac{3\pi^2 ln\left(\frac{r}{R}\right)\emptyset_n}{32\varepsilon^2\varepsilon_a sin^2\theta}P_{np}P_s\right)\phi_{\mathrm{m}}^2, \tag{19}$$

and $\widetilde{k}$ is defined as

$$\tilde{k}^2 = k^2 + \Delta. \tag{20}$$

Finally, applying the boundary conditions and taking the average values leads to

$$\varphi_m^2 = 2\frac{E^2 - \frac{P_s E + (B\pi^2/d^2)}{\varepsilon_a\varepsilon sin^2\theta} + \frac{11}{1624}\frac{\varepsilon_a}{\varepsilon}\pi^2 R^3\left(1 - \frac{R^3}{r^3}\right)P_{np}^2\emptyset_n - \frac{3}{8\varepsilon^2\varepsilon_a sin^2\theta}\pi^2 ln\left(\frac{r}{R}\right)\emptyset_n P_{np}P_s}{E^2 - \frac{P_s E}{4\varepsilon_a\varepsilon sin^2\theta} - \frac{11}{6912}\frac{\pi^2\varepsilon_a P_{np}^2\emptyset_n R^6}{\varepsilon} - \frac{3}{32}\frac{\pi^2 ln\left(\frac{r}{R}\right)\emptyset_n P_{np}P_s}{\varepsilon_a\varepsilon^2 sin^2\theta}}. \tag{21}$$

The angle φ_m shows the maximum deviation of the ***c***-director with respect to the x-axis that occurs at the middle of the cell. Comparing these results with the obtained threshold field and maximum deviation of the ***c***-director in the pure SSFLC system reveals the strong influence of nanoparticles even at low concentrations. Indeed, the presence of nanoparticles in SSFLC medium creates strong local fields that produce large alignment effects over the distribution of the nano suspensions. On the other hand, application of even a weak electric field breaks the central symmetry of the dipoles' orientation, generally due to the ferroelectric torques. The obtained results confirm that in these composite mediums, the critical field of Fredericks transition might be decreased or increased, depending on the external field's direction, as discussed for the case of a pure medium. The shift of transition field is obviously connected with the permanent polarization of the particles. The obtained results show that the threshold field of Fredericks transition depends on the polarization of the nanoparticles as well as both spontaneous polarization and dielectric anisotropy of the liquid crystal host.

4. Some Remarks and Conclusions

We have obtained the threshold field of orientational transition and maximum deviations of the **c**-director under a certain electric field, in a surface stabilized ferroelectric liquid crystal (SSFLC). In section 2, we considered a pure SSFLC system and in section 3, we studied a nano- doped surface stabilized ferrolectric liquid crystal. We are now in a position to make some remarks.

1. Under a given applied field, the polarization tends to be aligned with the field and on the other hand, it's coupling to the **c**-director leads to an additional torque on **c**. We assumed that the director is strongly anchored at the boundaries. Hence, the external field causes an inhomogeneous texture. We have determined the variations of the director profile due to this inhomogeneity of the texture.
2. The parallel and antiparallel alignment of the external field and polarization ($P_s E > 0$ and $P_s E < 0$) has been considered. Our obtained results confirm that due to the coupling of the spontaneous polarization and director, both orientational effects can enhance each other in the antiparallel case and therefore, the threshold field is reduced. However, both effects contract each other in the parallel case and hence increase the critical field.
3. For the nano doped SSFLC system we have considered ferroelectric nanoparticles with sphere shape. Doping the ferroelectric nanoparticle affects the orientational transition due to the strong local electric field around the particle and coupling between the director and the polarization of the particle. We introduced a new parameter for the range of aligning effects of the particles. Moreover, we supposed that the system is free from dislocation of layer thickness due to the nanoparticles and the tilt angle is constant. Then, the Fredericks transition field has been obtained for sufficiently small nanoparticles with a low concentration. The dependence of the critical field on volume fraction of the particles has been determined.
4. We have found that the threshold field can be decreased or increased depending on the dielectric anisotropies for doped SSFLC system, as in the pure system. In the other words, it is decreased for the state of positive dielectric anisotropy with antiparallel alignment of the external field and spontaneous polarization. But for the state of negative dielectric anisotropy with parallel alignment of the field and polarization, the threshold field is increased. However, the shift is closely related to the nanoparticles' concentrations and sizes, besides the polarizations and dielectric anisotropies.
5. The variations in the polarization vector show marked difference with the pure FLC medium and the ferroelectric nanoparticles have strongly influences on the critical fields, even at low concentrations. The significantly changes in threshold field and deviations of the director field, in nano-doped SSFLC, is fundamental to operation of many ferroelectric liquid crystal devices. Furthermore, such systems have been the focus of much research in recent years, because of the extreme sensitivity of the potential device properties to very low particle concentrations.

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