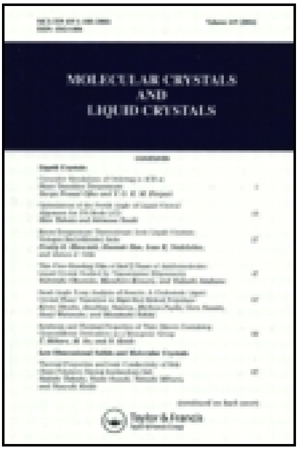

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Soft Modes in Confined Nematic Liquid Crystals

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Collective fluctuations of the liquid-crystalline order in two model geometries with a domain-like equilibrium configuration are analyzed within the Landau-de Gennes theory. The *wetting-induced heterophase structure* is characterized by a localized soft mode corresponding to fluctuations of the position of the interface between the two phases. In a very thin *hybrid cell*, the soft mode is associated with a structural transition from the configuration with a step-like profile of the tilt angle to the usual bent structure. This slowdown is exhibited by the lowest director mode. In both geometries the critical behavior is expected to be observable by the evanescent light wave scattering.

Keywords: confined liquid crystals; orientational fluctuations; light scattering

INTRODUCTION

In the past few years experimental results as well as theoretical studies have shown that the ordering and pretransitional dynamics of liquid crystals in restricted geometries differ from the equilibrium ordering and dynamics in the bulk.[1-8] Since the confined liquid crystals are very important both for present and future display technologies, considerable scientific efforts have been focused on determination of their properties and investigation of the influence of different types of confinement.

In our previous work,[6,7] we have intensively studied static and dynamic properties of a nematic liquid crystal film sandwiched between two identical parallel confining substrates. Here we present a summary of our previous work and preliminary results concerning pretransitional dynamics in a thin nematic hybrid cell with an emphasis on the soft modes in the vicinity of phase and structural transitions. A complete report on the study of the mean-field order and fluctuations in a hybrid cell will be presented and discussed elsewhere.[9]

In the following the model describing static and dynamic properties of the ordering within the Landau-de Gennes phenomenological approach is presented. In the second part of the paper we discuss the results and propose the setup for the evanescent light wave experiment that can confirm our predictions.

MODEL STRUCTURES AND DYNAMICS

As mentioned in the introduction, our model system consists of a nematic liquid crystal sandwiched between two parallel substrates. We consider three idealized cells with different confining substrates and, therefore, different equilibrium ordering and pretransitional dynamics (see Figure 1). The first two geometries are characterized by identical substrates with large disordering/ordering power: at temperatures just below/above the clearing point such substrates stabilize a heterophase structure consisting of an isotropic/nematic wetting layer and nematic/isotropic core. In the hybrid cell the substrates are different, i.e., they induce uniaxial nematic ordering in mutually perpendicular directions.

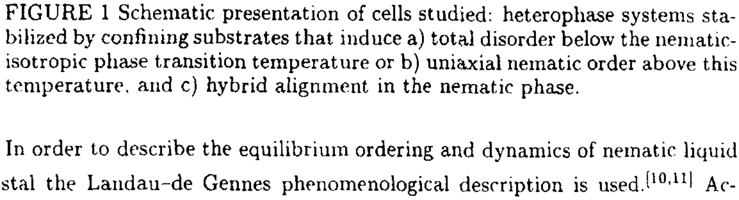

FIGURE 1 Schematic presentation of cells studied: heterophase systems stabilized by confining substrates that induce a) total disorder below the nematic-isotropic phase transition temperature or b) uniaxial nematic order above this temperature, and c) hybrid alignment in the nematic phase.

In order to describe the equilibrium ordering and dynamics of nematic liquid crystal the Landau-de Gennes phenomenological description is used.[10,11] According to this approach, in the vicinity of phase transition temperature the free energy density of the nematic phase can be expanded in terms of scalar invariants of the order parameter $\underline{Q}$, which is a symmetric, traceless, second-rank tensor,

$$
\begin{aligned}
f &= \frac{1}{2}A(T-T^*)\,\mathrm{tr}\,\underline{Q}^2 - \frac{1}{3}B\,\mathrm{tr}\,\underline{Q}^3 + \frac{1}{4}C(\,\mathrm{tr}\,\underline{Q}^2)^2 + \frac{1}{2}L\nabla\underline{Q}\vdots\nabla\underline{Q} + \\
&+ \frac{1}{2}G\,\mathrm{tr}\,(\underline{Q}-\underline{Q}_S)^2[\delta(z)+\delta(z-d)]\,, \qquad (1)
\end{aligned}
$$

where A, B, and C are temperature-independent material constants, T is the temperature, and T^* is the supercooling limit. The first three terms in the expansion represent the homogeneous part of the free energy density. The fourth term is the elastic part in the one-elastic-constant approximation and L is the elastic constant. The last term corresponds to the coupling between the liquid crystal molecules and the substrate; G is the strength of the coupling, $\underline{Q}_S$ is the preferred value of the tensor order parameter at the substrate, and d is the cell thickness. The numerical results reported in the next Section correspond to a 800 nm liquid crystalline film in a heterophase state and to a 100 nm thick hybrid cell. The material constants correspond to 5CB and read $A = 0.13 \cdot 10^6$ J/m^{3}K, $B = 3.89 \cdot 10^6$ J/m^3, $C = 3.92 \cdot 10^6$ J/m^3, $L = 9 \cdot 10^{-12}$ N, and $T^* = 307.1$ K.[12,13]

The mean-field ordering of a liquid crystal system is determined by the minimum of the free energy, while its dynamics can be described by an effective relaxation equation, i.e., time-dependent Ginzburg-Landau equation,[14,15]

$$
-\Gamma\frac{\partial \underline{Q}}{\partial t} = \frac{\delta f}{\delta \underline{Q}}\,, \qquad (2)
$$

where Γ is the effective rotational viscosity. In general, the changes of orientational order are coupled to the translational motion of molecules. Since the characteristic times for hydrodynamic processes are by two orders of magnitude shorter than the reorienatational ones, one can eliminate fast variables and describe the dynamics of the slow ones by the effective equation.[11]

In order to study harmonic excitations of the mean-field equilibrium the tensor order parameter $\underline{Q}$ is split into

$$
\underline{Q}(\mathbf{r},t) = \underline{A}(\mathbf{r}) + \underline{B}(\mathbf{r},t)\,, \qquad (3)
$$

i.e., into the mean-field equilibrium part which minimizes the free energy and the fluctuating part, which is governed by a linearized dynamic equation. The two tensorial equations, one for the mean-field part of the tensor order parameter $[\underline{A}(\mathbf{r})]$ and the other for the temporal evolution of its fluctuating part $[\underline{B}(\mathbf{r},t)]$, can each be split into five scalar equations for the five independent degrees of freedom.[6,7] For the uniaxial nematic liquid crystal the appropriate base consists of five traceless second-rank tensors, $\underline{T}_0 = (3\mathbf{n}\otimes\mathbf{n} - \underline{I})/\sqrt{6}$, $\underline{T}_1 = (\mathbf{e}_1\otimes\mathbf{e}_1 - \mathbf{e}_2\otimes\mathbf{e}_2)/\sqrt{2}$, $\underline{T}_{-1} = (\mathbf{e}_1\otimes\mathbf{e}_2 + \mathbf{e}_2\otimes\mathbf{e}_1)/\sqrt{2}$, $\underline{T}_2 = (\mathbf{e}_1\otimes\mathbf{n} + \mathbf{n}\otimes\mathbf{e}_1)/\sqrt{2}$, and $\underline{T}_{-2} = (\mathbf{e}_2\otimes\mathbf{n} + \mathbf{n}\otimes\mathbf{e}_2)/\sqrt{2}$, where $\mathbf{n}$-nematic director, $\mathbf{e}_1$, and $\mathbf{e}_2$ form the orthonormal triad, and $\underline{I}$ is the unit second-rank tensor.[16] Each base tensor corresponds to a particular degree of freedom of the nematic order parameter: $\underline{T}_0$ is related to the degree of order, $\underline{T}_{\pm1}$ to biaxiality of the ordering, and $\underline{T}_{\pm2}$ to the orientation of the director. The tensor order parameter can thus be rewritten as $\underline{Q} = \sum_{i=-2}^{2}[a_i(\mathbf{r}) + b_i(\mathbf{r},t)]\underline{T}_i$, where $a_i(\mathbf{r})$ and $b_i(\mathbf{r},t)$ $[i = 0, \pm1, \pm2]$ are the mean-field profiles and fluctuation modes respectively.

Once the mean-field profile is determined one can solve the equations for the fluctuation eigenmodes using the ansatz

$$b_i(\mathbf{r},t) = \exp\left[i(k_x x + k_y y)\right]\beta_i(z)\exp\left(-\mu_i t\right), \tag{4}$$

where k_x and k_y are the transverse components of the wave vector of the modulation, which are subject to periodic boundary conditions, β_i's are z-dependent amplitudes of the eigenmodes, and μ_i are the relaxation rates of fluctuation modes.

SOFT MODES

Wetting-Driven Phase Transition

The equilibrium configurations in *cells with identical confining substrates* are characterized by a coexisting, spatially localized isotropic and nematic phase, the "metastable" phase being induced by the substrates. In the case of complete wetting of the substrates with the isotropic or nematic phase, the transition between the two phases is continuous. This results in the divergence of the thickness of the boundary layer at the clearing point if the sample is semi-infinite.[17] Such a growth of the wetting layer is induced by the lowest order parameter mode, whose relaxation rate is critical when approaching the bulk clearing point.[6,7] Slow, soft fluctuation mode is localized at the interface between the two coexisting phases (substrate-induced isotropic or nematic phase and central bulk-like nematic or isotropic phase) with its maximum coinciding with the maximum slope of the scalar order parameter profile (see Figure 2).

The complete wetting regime occurs if the anchoring at the substrates is strong

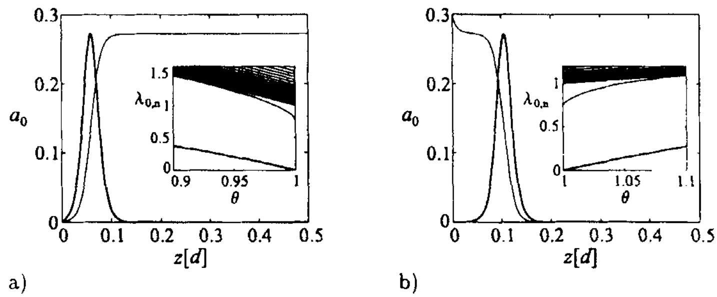

FIGURE 2 Scalar order parameter and soft order parameter mode (thick line) close to the nematic-isotropic phase transition temperature (T_{NI}), if the confining substrates a) decrease the nematic order or b) induce high uniaxial nematic order. In the inset, temperature dependence of the reduced relaxation rate [$\lambda_{0,n} = \mu_{0,n} - L(k_x^2 + k_y^2)/\Gamma$] measured in units of a rotational relaxation time $\tau_a = 27C\Gamma/B^2 \sim 10^{-8}$ s. The temperature is measured in units $\vartheta = (T - T^*)/(T_{NI} - T^*)$.

enough ($G > 10^{-4}$ J/m^2) and if the preferred degree of order at the substrate is either 0 (disordering substrate) or equal or larger than the degree of order in bulk nematic (ordering substrate).If the conditions for complete wetting are not fulfilled, the partial wetting regime occurs. In this case the nematic-isotropic phase transition is no longer continuous, and the excitation spectra are similar to the bulk one.

Structural Transition in a Confined Nematic Sample

In large *hybrid cell* the free energy is minimized by a bent configuration, where the nematic order is supposed to be uniaxial with director field bent with respect to the distance from one of the substrates. In thinner cells the director field cannot be bent continuously, but exhibits a step-like change resulting in a two domain structure, which is called director exchange structure or biaxial structure.[9,18] The latter name arises from the fact that the nematic order in the middle of the cell where the director exchange takes place is slightly biaxial [see Figure 3 a)]. The existence of the biaxial structure depends not only on the cell thickness but also on the strength of the surface anchoring, while the induced order at the substrates should be at least as high as in the bulk.

By changing the thickness of the system, a continuous structural transition

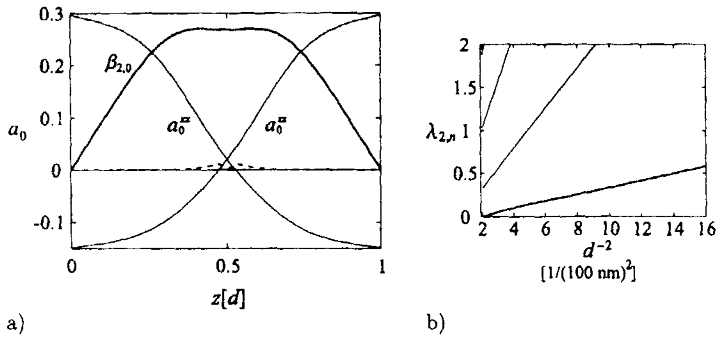

FIGURE 3 a) Scalar order parameters with respect to the director $\mathbf{e}_x$ and $\mathbf{e}_z$ are represented by a_0^{xx} and a_0^{zz}. Biaxiality of the nematic order is plotted with a dashed line. Thick line corresponds to the soft director fluctuation mode which bends the step-like profile of the tilt angle to the usual bent structure. b) Spectrum of director relaxation rate as a function of $1/d^2$. Thick line corresponds to the soft mode's relaxation rate.

can be induced, the corresponding soft mode being the lowest director mode which changes the step-like profile of the tilt angle to a continuously bent profile. The relaxation rate of the director fluctuations bending the director field decreases to 0 when the cell thickness is increased or the temperature is lowered toward the critical thickness or temperature, respectively [Figure 3 b)].

Light Scattering

Fluctuations of the nematic tensor order parameter give rise to fluctuations of the dielectric constant tensor responsible for the scattering of light. The slow fluctuation modes, can be detected by choosing appropriate polarizations of the incoming (**i**) and scattered (**f**) light (see Figure 4). Should one detect the soft order parameter mode in either of the heterophase systems discussed, the polarizations should be parallel to the nematic director. The director fluctuations in the hybrid cell can be detected if the polarizations coincide with $\mathbf{e}_x$ and $\mathbf{e}_z$.

The correlation function observed in the evanescent light wave experiment is of form[19,20]

$$G(t)_{dyn} = \langle E_s(0)E_s^*(t)\rangle_{dyn} \propto \sum_n \frac{R_{m,n}^2}{\mu_{m,n;q}} \exp(-\mu_{m,n;q}t) \ , \tag{5}$$

where $R_{m,n} = \int \beta_{m,n}(z)\exp(-\kappa z)dz$, $q = |\mathbf{k}_i - \mathbf{k}_f|$ is the magnitude of the scatter-

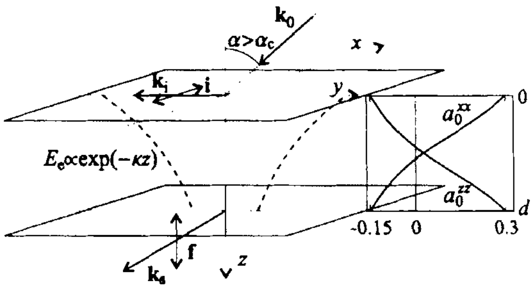

FIGURE 4 The setup of the evanescent light wave experiment in the hybrid cell. The incident light (wavevector $\mathbf{k}_0$) is totally reflected at the substrate-liquid crystal interface. In order to observe the director fluctuations the polarizations of the incoming (wavevector $\mathbf{k}_i$ and polarization $\mathbf{i}$) and scattered light ($\mathbf{k}_s$, $\mathbf{f}$) are mutually perpendicular.

ing wavevector, and $m = 0, 2$ for order parameter and director fluctuation modes, respectively. As shown by Figure 5, in the vicinity of a phase or structural transition the contribution of the soft mode dominates over the contribution of other modes. The correlation function can therefore be well described by a single exponential function corresponding to the relevant soft mode.

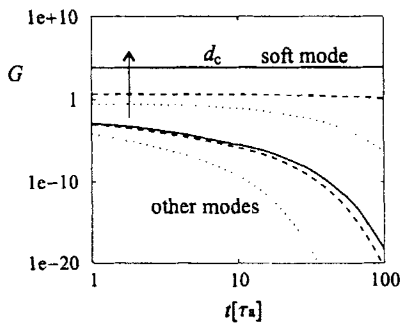

FIGURE 5 Contributions of the soft mode and other modes to the correlation function in a hybrid cell. The arrow denotes the direction of increasing cell thickness or decreasing temperature when approaching the structural transition towards bent configuration.

CONCLUSIONS

In a restricted geometry with high surface-to-volume ratio the properties of the nematic liquid crystal can differ considerably from those in bulk samples. For example, the confinement can make the nematic-isotropic phase transition continuous, and in a hybrid cell it can stabilize a domain-like ordering instead of a bent structure, the structural transition between these two configurations being continuous.

In the heterophase structure the soft mode corresponds to the phase transition and is associated to the nucleation of a (dis)ordered phase at the substrate. The soft fluctuations are localized in a small part of the cell, whereas the soft mode in a hybrid cell is spread over the whole sample. Being associated to the structural transition between two ordered phases, it changes the orientation of the molecules in the whole system.

The heterophase structure with a continuous nematic-isotropic phase transition can also be induced in a semi-infinite system, while the biaxial structure can only occur in a highly restricted geometry, i.e., in planar geometry with hybrid boundary conditions or in small cylindrical cavities with homeotropic anchoring. Although the modeled planar geometry is hardly a realistic one, the results obtained in our study can be applied to other host geometries as well, because they result from highly strained conditions and confinement.

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