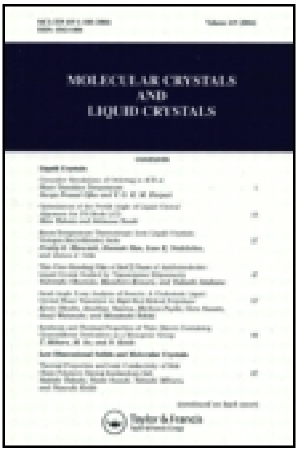

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Numerical Calculation of Nematic Director Fields with Stochastic Inner Boundaries

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NUMERICAL CALCULATION OF NEMATIC DIRECTOR FIELDS WITH STOCHASTIC INNER BOUNDARIES

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Abstract

The reorientation behaviour of a nematic liquid crystal restricted to thin planar cells in external electro-magnetic fields is well known. We discuss the effects of external fields on a bulk sample where a preferred director alignment is achieved by oriented small solid particles statistically dispersed in the nematic. The calculations may serve as a model for LC/polymer gels. They allow for the determination of physical properties, e.g. mesh sizes, from dielectric, optical or NMR measurements. A threshold behaviour for the director is found from the calculations. Critical fields, director distribution functions and characteristic relaxation times are derived.

INTRODUCTION

The director field orientation in a nematic liquid crystal has essential influence on the macroscopic optical, electrical and many other physical properties. In common applications, a macroscopically aligned director orientation is achieved by means of external electromagnetic fields or boundary conditions. The director alignment in thin display cells bases on the orienting surfaces of the cell planes. Recently, systems have become attractive to LC research where internal surfaces act on the director field and pose a definite alignment on the sample [1,2]. In liquid crystal dispersed polymers, a solid polymeric network inside the nematic pins the director field to a fixed orientation. By polymerization of appropriate monomers dissolved in the nematic phase, a complex inner surface is formed which aligns the director field in its orientational state maintained during polymerization. One can reorient the nematic director with sufficiently strong external fields. After switching the field off, the director relaxes back to the orientational state determined by the inner surface. Switching between transparent and opaque states can be achieved similar to PDLC systems. Electro-optic and NMR measurements [3,4,5] have shown that the director field deformation starts at some critical field strength which increases with the amount of polymer dispersed in the nematic. More systems are conceivable where the nematic alignment state is maintained not by external boundaries but by randomly embedded particles in the nematic bulk [6]. We introduce a simple model to investigate the properties of a nematic director field in such systems by means of numerical simulations. The director deformation in the sample is calculated as a function of field strength and mesh sizes (grain concentrations). From the calculated director fields, line shapes of NMR spectra and dielectric properties may be predicted. The computed director fields may also serve as the basis for simulations of optical properties.

THEORY

In view of the complexity of the system, we introduce some simplifications. We assume a nematic director field fixed with strong anchoring at statistically distributed fibers inside, and we restrict to a two-dimensional geometry, i.e. we assume that the inhomogeneity of the sample in the direction of the fibers y is much smaller than in the other directions and can therefore be neglected. The undisturbed director $\vec{n}_0$ be aligned planarly along y and we choose the coordinates such that the external field be in the yz-plane. Furthermore we assume that the director deformation stays planar in the plane of the field $\vec{B}$ and $\vec{n}_0$. This assumption might be violated at high fields (strong deformations) and large elastic anisotropy. Figure 1 gives a schematic view of the sample and some definitions.

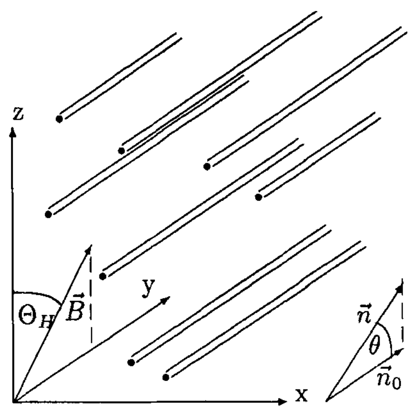

Fig. 1:
Geometry of the system. The magnetic field and the director are both in the y, z-plane. The director is fixed at the fibers along y.

The free elastic and magnetic energy per volume of the nematic are given by

$$F_{elast} = \frac{1}{2}K_1(div\vec{n})^2 + \frac{1}{2}K_3(\vec{n} \times rot\vec{n})^2 + \frac{1}{2}K_2(\vec{n}rot\vec{n})^2 \tag{1}$$

$$F_{mag} = -\frac{1}{2}\frac{\chi_a}{\mu_0}(\vec{B} \cdot \vec{n})^2. \tag{2}$$

The planar director orientation is described by the angle $\theta(x, z)$. Then the director field is $\vec{n} = (0, \cos\theta(x,z), \sin\theta(x,z))$, and Eqs.(1,2) take the form

$$F(x,z) = F_{elast} + F_{mag} = \frac{1}{2}(K_1\cos^2\theta + K_3\sin^2\theta)\theta_z^2 + \frac{1}{2}K_2\theta_x^2 - \frac{1}{2}\frac{\chi_a}{\mu_0}B^2\sin^2(\theta+\theta_H). \quad (3)$$

In a two-constant approximation, we set $K = K_1 = K_3$ and find

$$F(x,z) = \frac{1}{2}K\theta_z^2 + \frac{1}{2}K_2\theta_x^2 - \frac{1}{2}\frac{\chi_a}{\mu_0}B^2\sin^2(\theta+\theta_H). \quad (4)$$

The minimization of the free energy with appropriate boundary conditions gives the stationary director field. We use a relaxation technique to find the minimum of $\int F dV$. The balance equation of elastic, magnetic and viscous torques gives

$$\gamma_1\dot{\theta} = K\theta_{zz} + K_2\theta_{xx} + \frac{\chi_a}{\mu_0}B^2\sin(\theta+\theta_H)\cos(\theta+\theta_H). \quad (5)$$

This equation describes the relaxation of a nematic director field in absence of viscous flow. It reproduces sufficiently well the reorientation in low external fields and the response to slow changes of the field. At any given field B it will provide the correct stationary state with

$$\gamma_1\dot{\theta} \equiv 0 = K\theta_{zz} + K_2\theta_{xx} + \frac{\chi_a}{\mu_0}B^2\sin(\theta+\theta_H)\cos(\theta+\theta_H)$$

which corresponds to the Euler-Lagrange Equation

$$0 = \frac{d}{dx}\frac{\partial F}{\partial\theta_x} + \frac{d}{dz}\frac{\partial F}{\partial\theta_z} - \frac{\partial F}{\partial\theta}$$

for the equilibrium state of $F(x,z)$ in Eq.(4).
We introduce the coherence length $\xi_m = (K\mu_0/(\chi_a B^2))^{1/2}$, the relaxation time $\tau = \xi_m^2\gamma_1/K$ and the elastic ratio $\kappa = K/K_2$ and perform the coordinate transformations $\eta = (\kappa)^{1/2}x/\xi_m$ and $\zeta = z/\xi_m$. Then, Eq.(5) becomes

$$\tau\dot{\theta} = \theta_{\zeta\zeta} + \theta_{\eta\eta} + \sin(\theta+\theta_H)\cos(\theta+\theta_H). \quad (6)$$

This equation is solved by a finite differences method. The angle θ is kept fixed at M points $\Pi_m = (\eta_m, \zeta_m)$ $(m = 1..M)$ randomly distributed over a square of dimensions $l \times l$ corresponding to a region $l\xi_m \times l\xi_m/\kappa^{-1/2}$. The fixpoint density is $c = M/l^2$ corresponding to a fiber distribution density $C = c\kappa/\xi_m^2$ in the (x,z)-plane. A characteristic length of this distribution is $\xi_0 = C^{-1/2} = \xi_m(\kappa c)^{-1/2}$. The average distance from one Π_m to the next neighboured $\Pi_{m'}$ is $\xi_0/2$. In order to simulate a bulk sample, we have chosen the boundary conditions periodically with $\theta(\eta,\zeta) = \theta(\eta+l,\zeta) = \theta(\eta,\zeta+l)$.

In the first set of simulations, the fixpoints Π_m are assumed to be infinitely small in diameter, i.e. one grid point. In this situation, according to the scaling properties of Eq.(6), we find *one* universal behaviour of the director deformations. A scaling of the η or ζ coordinate does not change the properties of a random distribution in the η,ζ-plane except for the point density. Therefore, a set of calculated deformations $\theta(B)$ in η,ζ space completely describes the director fields for arbitrary K, K_2 and C if the magnetic field B is rescaled correspondingly with $(KK_2C)^{1/2}$ The director deformations $\theta(x,z)$ are obtained by correspondingly stretching the solutions in x and z. The angular distribution functions $n(\theta)$ describing the portion of the sample aligned in a certain orientation to $\vec{n}_0$ are independent of K, κ and C at the same reduced magnetic fields.

In general, the static director deformation that is finally reached by the relaxation method corresponds to the minimum of the free energy and should not be sensitive to the initial director field (except for the sign of θ). Nevertheless, we have used definite initial conditions. We take advantage of the fact that for small θ, Eq.(5) may be linearized and the solution can be separated by variables to $\theta(\eta,\zeta,t) = \vartheta(\eta,\zeta)\cdot T(t)$ with an exponentially growing or decaying general solution $\vartheta(\eta,\zeta)$. The shape of $\vartheta(\eta,\zeta)$ is found numerically. We have started the relaxation of the $\theta(\eta,\zeta)$ field from this universal solution scaled to a fixed mean amplitude $\bar{\theta}(t=0) = 0.1°$ Thus the relaxation of the director field was determined to positive angles θ, although the $-\theta$ reorientation is equivalent (as in ordinary Fréedericksz cells) for fields normal to $\vec{n}_0$.

RESULTS OF THE SIMULATIONS

In the first set of experiments, we have calculated the director deformations in dependence on the magnetic field B starting with different fixpoint distributions Π_k at each run. In Fig. 2, we show the calculated averaged $\bar{\theta} = \int \theta d\eta d\zeta / \int d\eta d\zeta$, crosses are for a 100×100 grid and diamonds for a 200×200 grid, resp., with the same density $c = 2.5 \times 10^{-3}$. One observes a pronounced critical behaviour for all distributions with a critical field $B_c \approx 1.09 B_0$ with $B_0 = 1/\xi_0(K\mu_0/\chi_a\kappa)^{1/2} \propto C^{1/2}$.

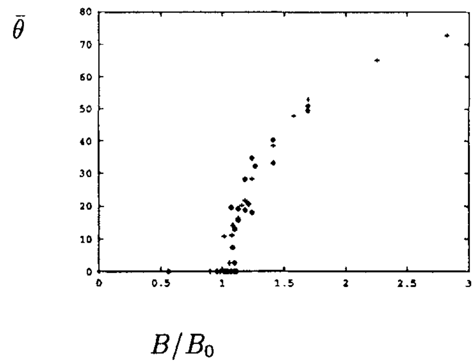

Fig. 2:
Average tilt angle $\bar{\theta}$ for random fixpoint distributions in the (x,z)-plane vs. reduced magnetic field B/B_0. Crosses and diamonds, corresponding to different grid sizes 100×100 and 200×200, resp., coincide.

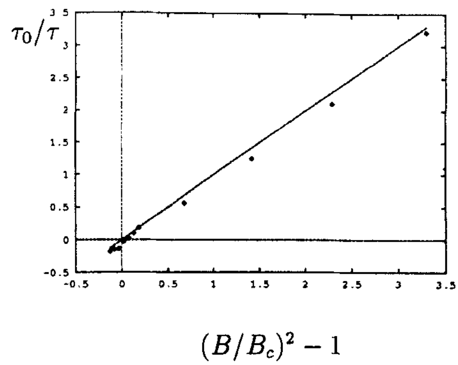

Fig. 3:
Relaxation time τ_0/τ vs. reduced magnetic field $(B/B_c)^2 - 1$ for random fixpoint distributions. The solid line is the fit to Eq.(7) with τ_0 calculated from corresponding using the average B_c for all runs.

Choosing realistic values of $\kappa = 1$, $\chi_a = 10^{-6}$, $K = 10^{-11} Nm$ and a density $C = 0.25 \mu m^{-2}$, one finds for example $B_0 = 1.772T$ and $B_c = 1.93mT$. The relaxation time $\tau(B)$ was determined from the increase/decay of $\bar{\theta}$ in the simulated director deformations after switching the field on/off. $\bar{\theta}$ scales with $(B/B_c)^2 - 1$ as shown in Fig. 3. The solid line corresponds to

$$\tau^{-1} = \tau_0^{-1}((B/B_c)^2 - 1) \tag{7}$$

with $\tau_0 = \gamma_1 \mu_0 / (\chi_a B_c^2) \approx 0.843 \gamma_1 \mu_0 / (\chi_a B_0^2)$
As expected, the time τ_0 which describes the relaxation to the off-state in absence of external fields decreases proportional to the inverse density C^{-1}, i.e. an increase of the concentration of fixpoints in the sample reduces the characteristic length and leads to faster elastic relaxation.

MORE REALISTIC SYSTEMS

The simulations performed in the previous section give insight in the general behaviour of systems with stochastic inner surfaces. Nevertheless, for a comparison with real systems, e.g. LC/polymer gels, some modifications have to be performed. At the fixpoints of radius zero, the free energy density diverges, i.e. the fixpoints behave like disclinations. A more realistic picture takes into account a finite size of the fibers corresponding to the correct volume ratio of LC and additive.

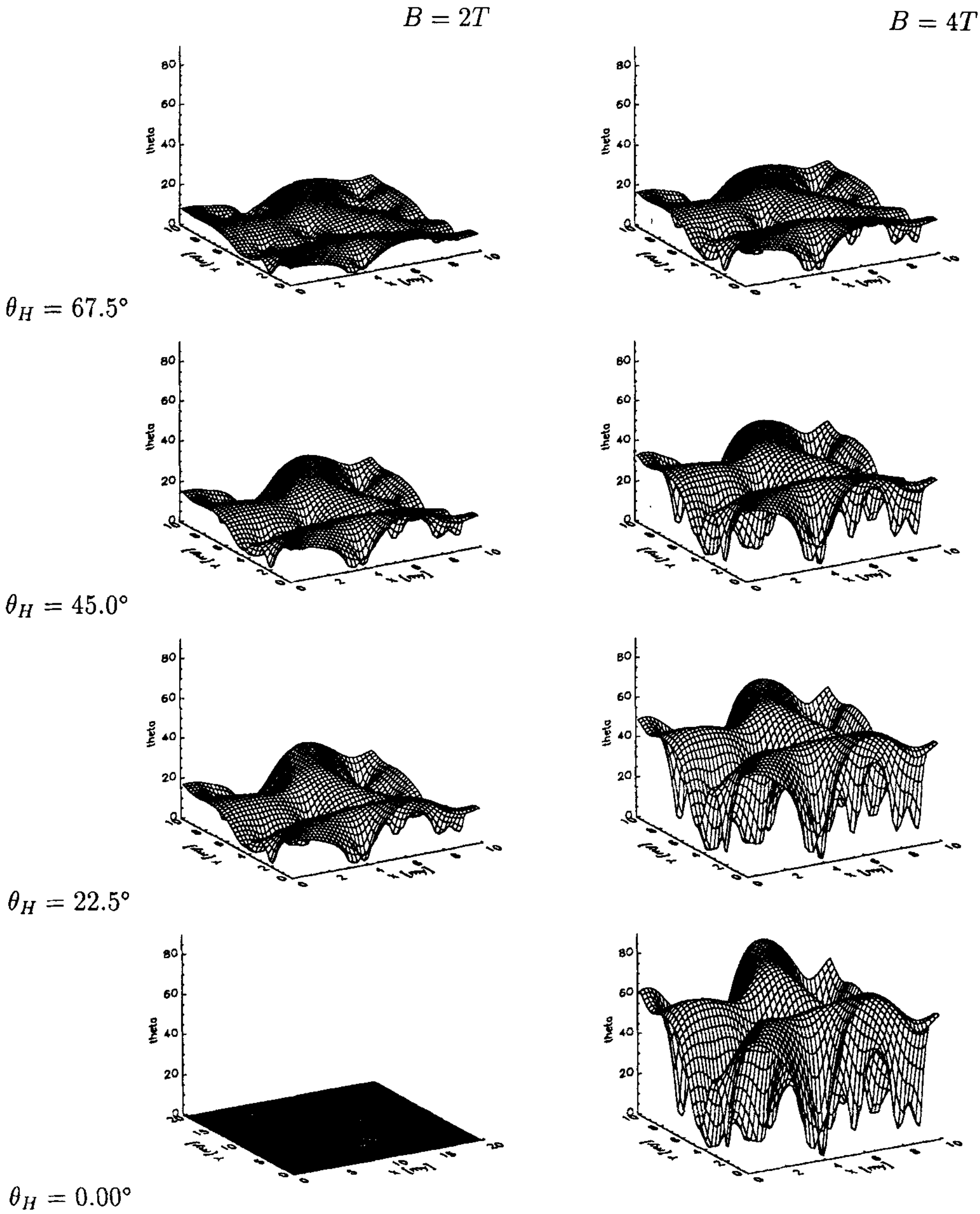

Figure 4: Influence of the magnetic field in different orientations to the z axis on the director deformation in a sample pinned with 3.35% additive with a cluster density of $C = 0.25\mu m^{-2}$, (distribution corresponding to Fig. 5). Parameters were $K = 10^{-11}, \chi_a = 10^{-6}, \kappa = 1$, yielding a critical field of $B_c = 2.495T$.

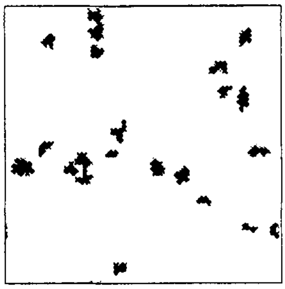

Figure 5: Example cluster pattern in the (η, ζ)-plane. The nematic fills the area between the clusters and is aligned along y at their surfaces. The filling factor C_0 is 0.0335.

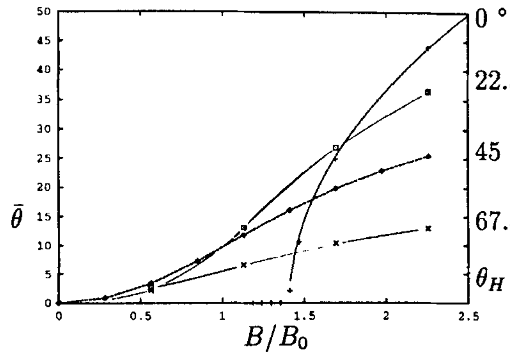

Figure 6: Average deformation angle vs. reduced magnetic field for different magnetic field orientations θ_H given at the right hand side. The lines are drawn to guide the eye.

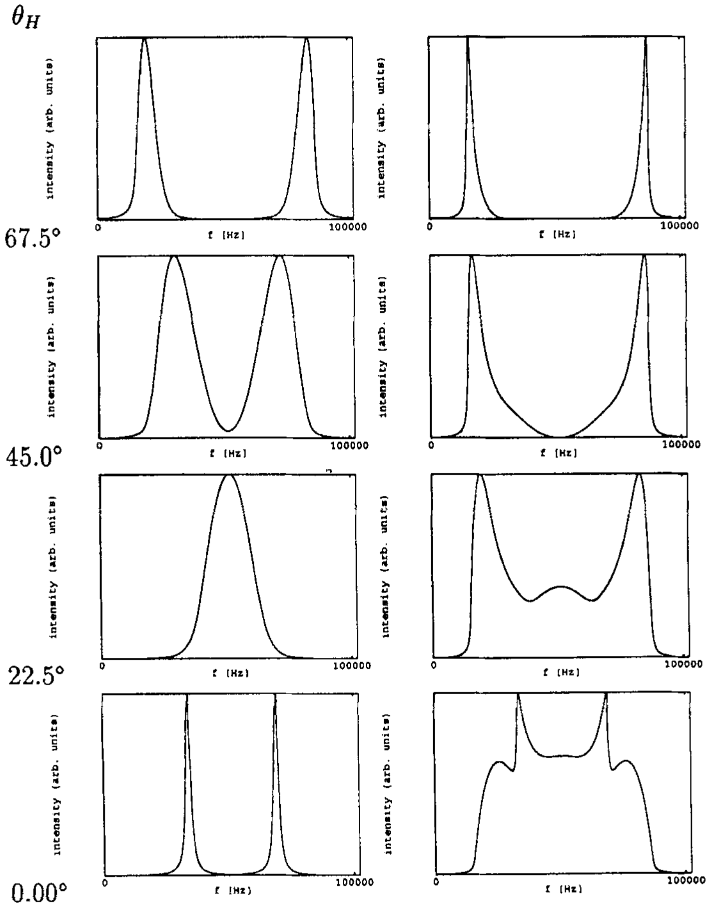

Figure 7: Angular dependence of simulated ^2H NMR spectra for a single line with $72kHz$ splitting, director deformations corresponding to $0.8B_c$ (left) and $1.6B_c$ (right), same parameters as in Fig. 4

The amount (volume fraction) of an aligning additive in the nematic be C_0. For the simulations we assume it to form elongated clusters (fibers) along y with an average density $C = 1/\xi_0^2$ and average diameter $d_0 = (4C_0/\pi)^{1/2}\xi_0$ (see Fig. 5) where C has the same meaning as in the previous section. The nematic aligns in y direction at the cluster surfaces. We have deliberately chosen irregular cluster shapes generated statistically. The results presented are for $C_0 = 3.35\%$ additive. Figure 6 shows the average $\bar{\theta}$ as a function of the strength and direction of the external field. As might be expected, the critical B_c is slightly shifted to higher fields ($B_c \approx 1.41 B_0$) compared to the value 1.09 found above. The major influence on the director field comes from the size of the intermediate cavities which is primarily a function of the number of clusters per area, and not of the volume fraction C_0. The ratio 1.41/1.09 is close to the ratio of the average separation of the fiber surfaces $\xi_0/(\xi_0 - d_0)$ of both systems.

We have used the calculated director deformation to simulate deuteron NMR spectra which provide experimental access to the director field distribution. Results are shown in Fig. 7. One finds that a dramatic change occurs near B_c. From comparison with experimental data [3], one can estimate the mesh sizes in the liquid crystal dispersed polymer (LCDP) system. The 4.7T NMR magnetic field is of the order of the critical field $\approx B_c$ of a 2.16% polymer sample. With the relations given above, this B_c corresponds to a value $\xi_0 \approx 1\mu m$. This value agrees very well with the mesh size determined by Jakli [4] in electro-optical measurements. In first approximation the sample can be considered as a nematic bulk with solid polymer fibers of $\approx 0.2\mu$m average diameter and a distribution density C of $1/\mu m^2$.

CONCLUSIONS

We have shown by numerical simulations that in nematic LC with stochastically embedded inner boundaries, a critical behaviour of the director deformation in external fields is expected. For a dispersion of infinitely small fibers which align the nematic planarly along their surfaces, the director deformation starts at a value

$$B_c \approx \frac{1.09}{\xi_0}\sqrt{\frac{K\mu_0}{\chi_a \kappa}}$$

if the magnetic field $\vec{B}$ is perpendicular to the easy axis $\vec{n}_0$. The critical concentration that is sufficient to stabilize the director in an external magnetic field B is therefore

$$C_c = \xi_{0c}^2 \approx \frac{1.09^2}{\kappa \xi_m^2}. \tag{8}$$

For electric fields, one has to take into account the inner deformation of the electric field. Therefore the results for the magnetic field cannot be directly transferred. The threshold field for director deformations however is derived straightforward from Eq.(8) if the magnetic coherence length ξ_m is substituted by

$$\xi_{el} = \sqrt{\frac{K}{\Delta\epsilon\epsilon_0 E^2}}.$$

The relaxation time of the director field in the magnetic field is proportional to $(B/B_c)^2 - 1$. In absence of external fields, the relaxation time $\tau_0 = \gamma_1\mu_0/(\chi_a B_c^2)$ decreases linearly with increasing concentration C of fixpoints.

Calculations of the director field for a 3.35% dispersion of a solid polymer have been performed where a finite size of the aligning surfaces was taken into account. Comparison of simulated ^2H NMR line shapes with experimental spectra allow for the determination of the internal structure of LC/polymer dispersions. In the systems investigated in [3], a mixture of 5CB with 4,4'-disacryloyl-biphenyl (BAB),the fibers of the solid polymeric network are distributed with a density of approximately 1 μm^{-2}, their average displacement is 1μm. Relating this to the volume fraction of the polymer, one can conclude an average diameter of the agglomerates of $\approx 0.2\mu$m. The numerical formalism presented here may be applied to arbitrary shapes and arrangements of internal surfaces, however the two-dimensional simulations are restricted to oriented LCDP phases. If no macroscopic alignment is present ("powder" samples), calculations have to be performed in three-dimensional geometry. Such an extension of the calculations and inclusion of out-of-plane deformations could give more realistic quantitative simulations of LCDP but should not change qualitatively the results presented here.

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