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Molecular Simulation of a Nematic Liquid Crystal Cell with Asymmetric Recurrent Boundary Conditions

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For several years now the physics and chemistry of condensed matter attracts a great deal of attention, while also occupying an extremely important position in the research activity worldwide. Liquid crystalline materials, in particular, present numerous applications in the fields of science and technology. Since the development of the liquid crystals display (LCD) technology, a significant concern was devoted to the development and characterization of these fascinating mesophases. In the present paper we perform several Monte Carlo simulations, by using the Lebwohl-Lasher model, for investigating the molecular director configuration in a nematic liquid crystal cell having varying boundary anchoring conditions in asymmetric circumstances. For this geometry, we analyze the molecular spatial behaviour, while mapping the local order parameter distribution for a nematic phase temperature. We also characterize the shape of the transition regions which strongly depend on the distance to the boundaries, and analyze the neighboring spins behaviour throughout the bulk. Furthermore, by using the Müeller matrix approach, we simulated the transmission of light through the nematic cell at normal incidence under crossed polarizers condition. These investigations seek to enrich our scientific knowledge about the fascinating research topics homeotropic in condensed matter physics by exploring interesting issues related to the orientational and optical properties of liquid crystals in confined geometries.

OCIS codes: (260.1440) Birefringence; (160.3710) Liquid crystals; (160.4760) Optical properties; (230.3720) Liquid-crystal devices; (350.2770) Gratings; (160.1190) Anisotropic optical materials

1. Introduction

Many molecular liquids consisting in anisotropic molecules have one or more liquid crystalline phases that, due to their particular properties of fluidity, transparency, optical anisotropy, represent a convenient testing ground for different complex phenomena [1, 2]. Because of the large number of potential applications in the fields of science and technology, liquid crystals represent a category of very interesting materials which have drawn the attention of physicist, chemist and engineers.

The orientational order the liquid crystals is determined by the anisotropic interaction between molecules consisting in both long-range attractive interactions and short-range repulsive ones. Moreover, the physical behavior of liquid crystals is also influenced by

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the surface properties and, particularly, the surface anchoring energies play an extremely important role in establishing the local orientation of the molecules [1, 2]. These competing phenomena complicate a theoretical description of such systems. Several theories based on the phenomenological treatment of Frank [3], or on the minimization of the free energy have been proposed [1]. Both approaches use rather complex calculus, involving more refined numerical methods, like using the finite element method with an adaptive [4], or a moving mesh [5].

Another way of describing the orientational order and the properties of liquid crystals is by using computer simulations. In the last decades, Monte Carlo and molecular dynamics simulations have become widely used tools in describing properties of liquid crystals, including optical properties of these classes of materials [6–11]. Much effort was also devoted to simulate the behaviour of liquid crystals displays [12–17] as well as liquid crystals in various confined geometries [18–22].

In this manuscript we suggest for investigation a special confinement geometry nematic cell presenting an interesting situation of periodic twist anchoring conditions with asymmetric circumstances at the two opposing boundary surfaces.

2. The Model and Simulation Method

We performed Monte Carlo simulations of a nematic liquid crystal cell using the well-known Lebwohl-Lasher model [23]. Here, the liquid crystal molecules are considered as unit vectors (versors or spins), which occupy fixed positions in the sites of a cubic crystalline lattice. The versors freely rotate in space and interact with each other through an orientation dependent energy:

$$U_{ij} = -\varepsilon_{ij} P_2(\cos\theta_{ij})$$

Here ε_{ij} is a positive constant for first order neighbor particles and zero otherwise. P_2 is the second rank Legendre polynomial and θ_{ij} is the angle between the versors, obtained from their dot product, $\cos\theta_{ij} = s_i \cdot s_j$.

Because the energy of interaction is invariant under a uniform rotation of all spins, the bend, splay and twist elastic constants of the liquid crystal are all considered to be equivalent [16–18].

The fact that the molecules' centers of mass are arranged in an ordered fashion does not contradict the fact that in a liquid crystal there is no positional order of the molecules, because, in a real liquid crystal, the molecules arrange themselves in ordered domains. In fact, each spin represents an ordered domain encompassing many molecules whose centers of mass are arbitrarily distributed [16, 17].

The major benefit of this simple model is the fact that, because spins' centers of mass are fixed, we can save computer time during the simulation, comparatively with models considering translational degrees of freedom. The system was deeply studied and gives a realistic representation of a nematic liquid crystal, with a first order phase transition at scaled temperature $T^*_{NI} = kT_{NI}/\varepsilon = 1.1232 \pm 0.0006$ [12, 15–17].

The nematic liquid crystal cell that was used in the simulation had a rectangular shape, having the dimensions $N_X \times N_Y \times N_Z$ in lattice spacings and electrodes parallel with XOY plane, situated at $z = 1$ and $z = N_Z$ as in Figure 1. The bottom electrode imposes planar boundary conditions, for the left half of the cell,$1 \le y \le \frac{N_y}{2}$, with spins oriented along OX axis, and for the right one, $\frac{N_y}{2} + 1 \le y \le N_y$, with spins oriented along OY axis. The

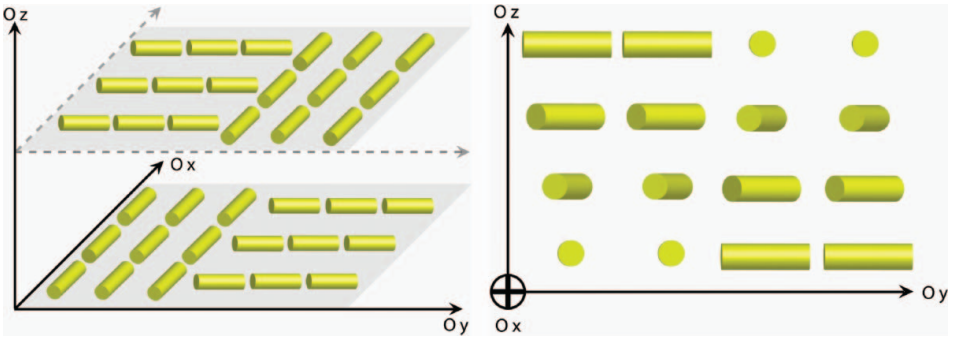

Figure 1. Geometry of the simulated cell. (left) Isometric view of the nematic cell and the boundary layers anchoring configuration. (right) Schematic representation of the expected molecular orientation in the YOZ plane view.

upper electrode also imposes planar conditions, but in opposite directions: the left side imposes the spins to orientate along the OY axis, and the right side along the OX. In all other directions, we consider periodic boundary conditions. For this kind of the geometry, each half of our cell has similarities with the geometry of the twisted nematic cell used in [11,12], but the presence of the stripes with different anchoring directions are rather analogous with the stripes we previously used in [17]. Because of the periodic conditions along the OY axis, each stripe of one kind is neighbor with two stripes of the other one, resulting in a complex interaction between the molecular spins.

For a system with such a rather complicated and competing boundary conditions, instead of using the bulk order parameter, it is preferably to employ the tensor parameter [6, 15–17, 20, 21]:

$$Q_{\alpha\beta} = \frac{1}{n}\sum_{k=1}^{n}\left(\frac{3}{2}\langle s_{k\alpha}s_{k\beta}\rangle - \frac{1}{2}\delta_{\alpha\beta}\right)$$

where $\alpha, \beta = x, y, z$, $\delta_{\alpha\beta}$ is the Kronecker delta, $< \ldots >$ is the ensemble average and n is the number of spins. For $n = 1$, the average is performed on Monte Carlo cycles only and $Q_{\alpha\beta}$ describes the local order [16, 17, 20–22].

We discussed elsewhere that, because the way in which the tensor order parameter is defined, it has two important properties [16, 17]: it is a symmetric tensor and it has a zero trace, meaning that only 5 out of 9 components are independent.

The diagonal components of the tensor order parameter represent the degree of order with respect the coordinate axis, and the off-diagonal ones represent the bending of the director field in the corresponding plane. The values of the tensor order parameter are between-0.5, when the director is perpendicular to the corresponding direction, and 1, when the director in perfectly parallel with the direction. Because the components of the tensor are obtained as statistical averages for each cell, their values are within these limits, with the value of 0 meaning the total disorder with respect a certain direction.

Following the discussion on the reference [4], the tensor order parameter approach may give a complete description of the nematic order: by diagonalization of $Q_{\alpha\beta}$, it's eigenvectors are the directions of the molecular orientations and the corresponding eigenvalues describe the degree of order. There are the following interesting situations: a) the largest eigenvalue corresponds to the classical order parameter and the related eigenvector is the

nematic director. The absolute difference between the remaining two eigenvalues can be also calculated. The deviation from zero of this value describes the biaxiality of the system; b) if all the eigenvalues are zero, we have an isotropic state.

The illustrated system was used to simulate the behaviour of a liquid crystal cell with planar boundary conditions imposed by the electrodes. The dimensions of the simulated cell were$N_x = 32$, $N_y = 32$ and $N_z = 18$ in lattice spacing, which means that we had 18,432 spins. Because spins located at electrodes are fixed and, because we have used periodic conditions along the OX and OY directions, the number of free spins is 16,384 and a Monte Carlo cycle consisted of 16,384 attempted moves.

The Monte Carlo procedure consisted in randomly choosing a spin and rotating it with a random angle [16, 17, 20, 21]. We calculated the energies in the old and in the new state and the move was accepted using the Metropolis acceptance criterion [6–10]. The mechanism was repeated for 160,000 Monte Carlo cycles with 60,000 of them used for equilibration.

The dimensions of the simulating cell were chosen in order to avoid finite geometrical effects and the big number of Monte Carlo cycles was set for having very good statistical averages.

The coupling interaction constant between free spins was $\varepsilon_B = 1$ and the interaction between a free spin and a fixed one was $\varepsilon_S = 1.5$. We have used this relatively large value for the anchoring constant, because we were interested only in the effects of the boundary conditions on the liquid crystal cell microscopic properties.

3. Results and Discussions

By means of Monte Carlo simulations we have obtained the components of the tensor order parameter in each point of our cell, at various temperatures. Due to the periodic boundary conditions along OX axes and because of the geometry of the cell the components of the tensor order parameter were averaged along the OX direction.

We performed a first set of Monte Carlo simulations at a reduced temperature, $T^* = 0.9$, which is very deep in the nematic phase. In Figure 2, we represent the maps of Q_{XX} and Q_{YY} in the YOZ plane.

From Figure 2, we notice that in the left side of the cell, Q_{XX} has values close to 0.79 near the bottom electrode and it decrease to approximately -0.39 near the upper electrode. On the contrary, in right side of the cell, near the bottom electrode, Q_{XX} is -0.39, increasing to 0.79 to the upper one.

In Figure 3, we plot the map of the Q_{YY}, which has a complementary and anti-symmetric behaviour regarding Q_{XX}: the smallest values are in the bottom left side and upper of the right side and vice versa.

This behaviour of the tensor order parameter is in perfect accordance with the directions imposed by the boundary conditions: in the bottom left side of the cell, the molecules are nearly parallel with the OX direction and, as z increases, the molecules rotate reaching a direction parallel with OY near the upper side of the cell. In the right side of the cell, the situation is different: in the bottom part the molecules are parallel with OY and rotate until became parallel with OZ in the upper one.

In order to have a more clear representation of the behaviour of the molecules in our nematic cell, we represented all the components of the tensor order parameter as a function of z, for $y = 8$, i.e. in the middle of the left region (Figure 4). We have chosen this value to illustrate the tensor order parameter, because near the boundaries of the region the influence

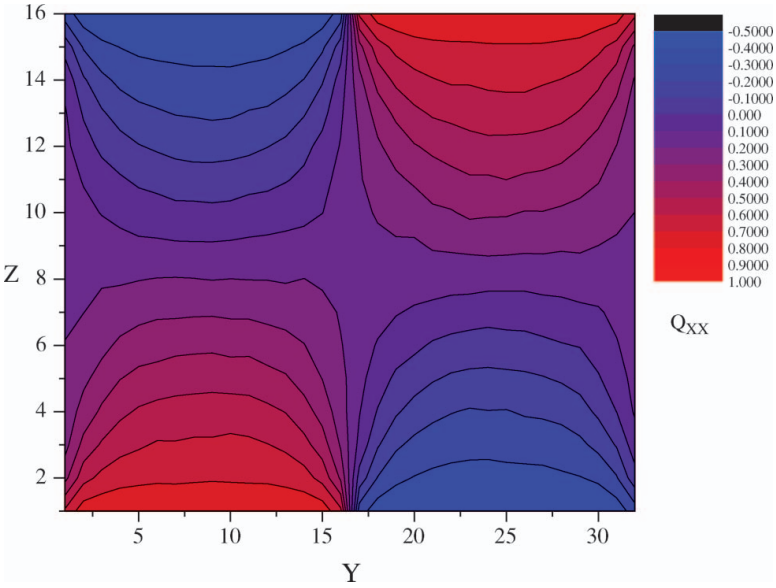

Figure 2. Map of Q_{XX} in the YOZ plane.

of the neighboring spins is very pronounced and the director behaviour is modified, as it can be observed in Figures 2 and 3.

From Figure 4, we remark some other important conclusions. First of all, the value of Q_{ZZ} is close to -0.5, meaning that, as expected, the molecular director is always perpendicular on the OZ direction. On the other hand, with a very good accuracy, the

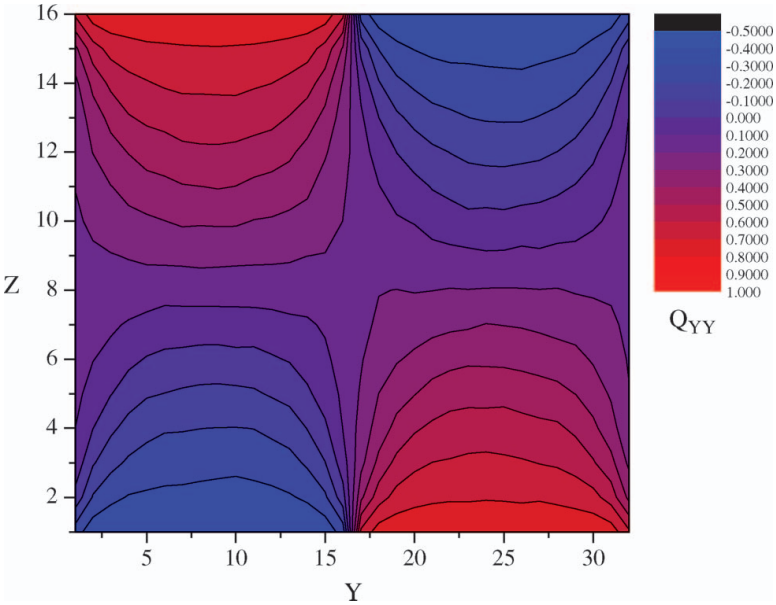

Figure 3. Map of Q_{YY} in the YOZ plane.

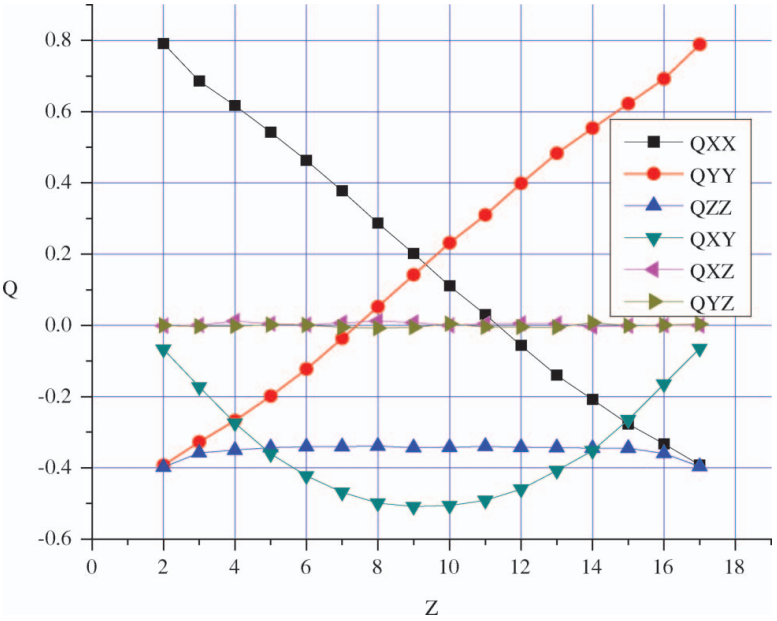

Figure 4. Components of the tensor order parameter for y = 8. Error bars sizes are of dimension of the symbols and were omitted. Lines are only guides to the eye.

values of Q_{XZ} and Q_{YZ} are zero, meaning no order, and all the rotations take place in the XOY plane. If we draw the same graphs, but this time for $y = 24$, we would find the same behaviour, except for the inverse role between Q_{XX} and Q_{YY}. This behaviour may be explained by the fact that both plots are for slices situated in the middle of the aligning regions, far enough from the boundaries.

From Figures 2 and 3, we also distinguish the presence of the transition region situated in the middle of the cell and to the margins, because of the periodic conditions. The width of the transition region depends on the distance to the electrodes: as the distance increases, the transition is smoother and the region is narrowed, as observed from Figure 5 were it is represented Q_{XX} as function of y for several distances from the bottom electrode.

Using the method described in [4], we tried to find regions of biaxiality in our simulation cell. The values for the biaxiality where very small, with a magnitude order of the error bars, concluding that our liquid crystal behaves uniaxial. We consider that this is consistent with the results of [15] where, for a system somehow analogous with ours, the calculated biaxiality was approximately 0.1 at temperature $T^* = 1.1$, which is very close to the nematic-isotropic transition temperature. These results may be generally explained by the simplification induced by the Lebwoh-Lasher model and, in our situation, especially by the very low temperature, $T^* = 0.9$, and strong anchoring regime, $\varepsilon_S = 1.5$, we have used.

We also compared our results with those in reference [11], where we also have a twisted nematic cell, but the simulation is done by using the off-lattice Gay-Berne molecular potential and a huge number of 10^6 particles. Although our system is a latticial one, we have used a more complicated geometry and a small number of particles comparative with [11], there are some similarities between these two approaches. Both of them predict the spontaneous manifestation of the helical torsion of the nematic, only under the boundary influences. Indeed, in our case this is true only in the middle of the left and right regions, the

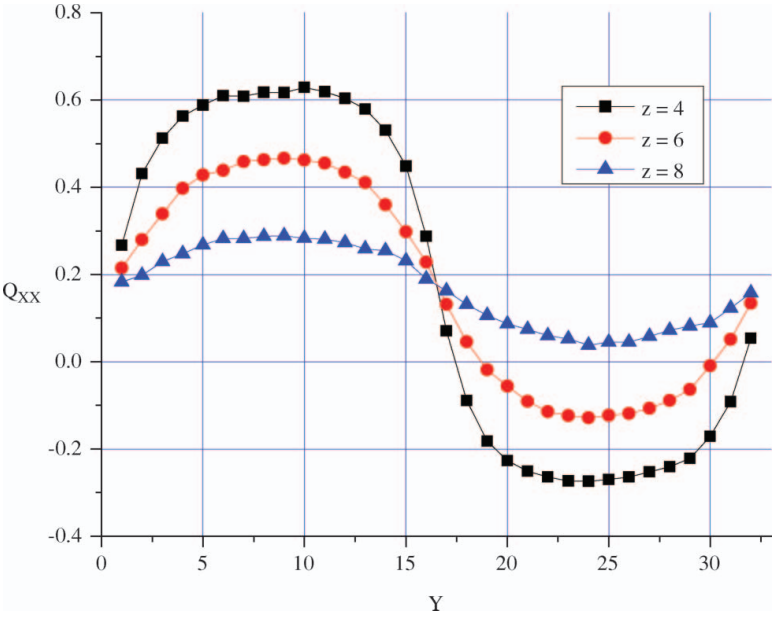

Figure 5. Behaviour of Q_{XX} as function of y for several distances from the bottom electrode. Error bars sizes are of dimension of the symbols and were omitted. Lines are only guides to the eye.

interfaces being, naturally, very disturbed, due to the competing influences. In the middle of the left and right regions of our cell, the dependence of Q_{XX} and Q_{YY} on z is virtually linear, very similar with the behaviour of the azimuthal angles calculated in [11].

It is well known that the main use of the liquid crystals is in optical devices, especially liquid crystal displays [1, 2, 9, 12–17]. We have also simulated the optical properties of our cell that were obtained via the Müeller matrix approach, also used in other Monte Carlo simulations of optical textures [12–15, 17, 19]. The key assumptions were that each lattice cell containing a liquid crystal molecule is a simple linear retarder described by a Müeller matrix, which is given by the orientation of the molecular director and optical properties of the molecule [17, 24, 25].

The light beam travelling in the liquid crystal display is retarded by a matrix resulting for a product of Müeller matrix corresponding to each crossed cell. In our case, we considered that the light is traveling in the OZ direction. The light modulated by the liquid crystal display is retarded and polarized and its intensity is represented by the Stokes vector [17, 24, 25] S_{OUT}:

$$S_{OUT} = P_{OUT} \cdot \prod_{i=2}^{N_z-1} M_i \cdot P_{IN} \cdot S_{IN}$$

where S_{IN} is the Stokes vector for the incoming unpolarized light, M_i is the Müeller matrix of the site i in a column of cell parallel with OZ. Note that on each side of the cell we have a polarizer (P_{IN} and P_{OUT}) in crossed conditions.

As in [17], in the above relation, the product is taken only for mobile spins having $z = 2$ to$z = N_z - 1$, the spins in the proximity of the electrode being fixed for simulating the anchoring conditions.

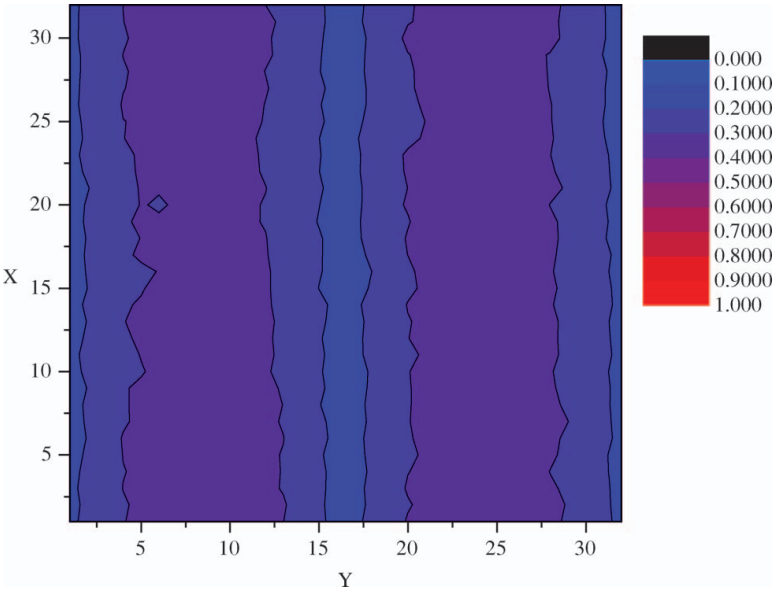

Figure 6. Map of the intensity of the transmitted light for $T^* = 0.9$ and $\lambda = 480$ nm.

For the simulated nematic liquid crystal we have chosen the ordinary refractive index $n_0 = 1.5$ and the extraordinary refractive index $n_e = 1.7$, similar of the 5CB liquid crystal [12, 17, 19], the wavelength of the light being $\lambda = 480$ nm and temperature $T^* = 0.9$.

We calculated the light intensity after the passage through the liquid crystal cell, obtaining the map of the intensity (Figure 6), where each pixel of the display corresponds to a lattice point in the XOY plane.

From Figure 6, we observe that the intensity map consists in a series of parallel stripes as it is dictated by the anchoring regime and boundary conditions. In the transition regions, the intensity of the transmitted light is smaller than in the bulk, due to the smaller order degree of the molecules. It is also important to notice that from the point of view of travelling light, the left and right regions are similar, both of them behaving like a twisted nematic cell. This observation is in close relation with our statement that components Q_{XX} and Q_{YY} of the tensor order parameter are mirroring each other.

Another set of Monte Carlo simulations was made for the same system, but at temperature $T^* = 1.3$, which is above the nematic isotropic transition temperature. We found that the intensity of the transmitted light was approximately zero. Indeed, by checking the values of the tensor order parameter components we obtained that, except for the vectors that are first order neighbors with the fixed spins, all the other components vanished.

4. Conclusions

By using the well known Lebwohl-Lasher model and Monte Carlo simulations we studied a special confinement geometry nematic cell. We suggest an interesting case of periodic twist conditions having asymmetric anchoring circumstances at the two opposing boundary surfaces.

From the simulations, we obtained the molecular director profile throughout the bulk, and we are able to observe the importance of the transition regions.

The consequent molecular organization of the nematic liquid crystal has also a high impact on the optical properties of the studied system. By means of Müeller matrix approach we analyzed a propagating 480 nm light wave inside the proposed nematic cell and the obtained transmission intensity mapping demonstrates the significant dependence on the imposed anchoring strength and spin stabilization after reaching a quasi equilibrium state. The simulation results were obtained deep in the nematic phase (for a temperature $T^* = 0.9$), while in the isotropic phase (for a temperature $T^* = 1.3$) the light intensity was close to zero.

We consider that the investigated nematic system has an interesting confinement imposed by the special boundary anchoring conditions and the resulting molecular distribution and optical behaviour prove worthy in understanding the control and operation of liquid crystal cells presenting complex geometries.

Acknowledgements

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