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Domain growth dynamics and Fréedericksz transitions of liquid crystals: Brownian dynamics simulations on lattices

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A Brownian dynamics simulation using the Lebwohl–Lasher (LL) nematogen model is developed to investigate liquid crystals (LCs) on two-dimensional lattices. According to the examination of defect annihilation and domain growth, this methodology appears to be a successful approach in studying the LC dynamics, its main advantage being that the time evolution in the simulation mimics the physical time, in contrast to related modellings published recently. In addition, the Fréedericksz transition of an LC under static magnetic fields is reproduced. Further analysis of the simulation outputs and the associated continuum theory of LC elasticity reveals that the LL potential can, by analogy, be obtained from the Frank energy. As a result, the methodology of the combination of Brownian dynamics simulation with the LL model is proven valid and can be extended to study the director dynamics of LCs subjected to external fields.

1. Introduction

Liquid crystals (LCs) exhibit fruitful and characteristic microstructures [1, 2]. An appreciation of LC microstructures leads to an understanding of the properties of LCs and the possibility of tailoring them for specific requirements. LC microstructures can be dealt with by the Frank elastic theory, whilst restructuring dynamics is treated by the Ericksen–Leslie (EL) theory [1]. Both the Frank and EL theories describe LCs on the continuum level. On the other hand, molecular theories have been developed by Maier–Saupe, Onsager and Doi, etc. [2]. Computer simulation provides a third theoretical method, its main advantage being the easy inclusion of complicated static and dynamic interactions [3–11]. For instance, Zannoni and co-workers [3] and Yang *et al.* [4] performed Monte Carlo (MC) simulations of LCs, while Windle and co-workers [5–8] and Kimura and Gray [9, 10] developed an iterative algorithm to obtain the equilibrium state of an LC under a certain boundary condition. These modelling approaches are commonly based on the Lebwohl–Lasher (LL) nematogen potential model developed from the Maier–Saupe potential for simulation on lattices [11].

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The dynamics of defect annihilation and domain growth in thermodynamically unstable systems is an outstanding problem in statistical physics [12, 13]. One of the typical dynamic systems of interest is an LC undergoing a texture evolution [7]. Computational models on a supramolecular scale have enabled equilibrium structures to be predicted for given boundary conditions and have also facilitated the modelling of the process of microstructural relaxation of LCs [5–10, 14–17]. In two recent reports [8, 9], the LC textures initiated from a random isotropic state were calculated using a simple two-dimensional (2D) lattice model proposed originally by Lebwohl and Lasher [11] and then developed by Bedford *et al.* [5]. The computer simulations noted above [5–10] were carried out by iteration after minimizing an interaction energy between nearest-neighbour (NN) lattices. This work is of great help, with advantages as pointed out by the authors. On the other hand, the disadvantages are also obvious. The most serious problem is, in our opinion, that the number of iteration steps cannot be guaranteed to be linearly proportional to the physical relaxation time. The modellings are therefore not a vivid simulation of the evolution process and are unsuitable for quantitative investigation in the strict sense of dynamics.

Another important method which should be mentioned is MC simulation [3, 4]. The MC iteration step linearly relates to real time to some extent. In most MC simulations of LC systems [3, 4], whether the new orientation state is accepted or rejected depends only on the energy difference between them, but the details of the potential barrier between the two states cannot be taken into consideration. Therefore, the MC method cannot describe very fast relaxation. In order to resolve these problems, we have employed a Brownian dynamics simulation [14–18] to study the texture evolution in an LC system also using the LL model on lattices. With this method, the linearization of the simulation with real time is achieved, which is the main advantage over the previous modelling work on texture evolution [5–10]. We should further note that if the time step is small enough in the simulation, the evolution process can reflect the details of the local potential and, hence, the fast relaxation. This is the main advantage of the Brownian dynamics simulation over the MC method. In this paper, the evolution process of LC domains is described with the Brownian dynamics simulation, and the scaling law for domain coarsening with time is revealed plausibly.

Furthermore, we would like to point out that this algorithm can also be employed to treat external field effects on LCs by reproducing the Fréedericksz transition. This phenomenon refers to the transition of the orientational direction of the LC system sandwiched between two plates with the increase of the strength of the external magnetic fields. The cell configurations for splay and bend deformations are schematically presented in figure 1. The measurement of the associated transition points has conventionally been employed as a standard method to determine experimentally the principal Frank constants, K_{11} (splay), K_{22} (twist) and K_{33} (bend) [1]. The other purpose of this paper is to reveal the relationship between the LL potential used in the computer modelling and the Frank elastic energy used in the continuum theory. This aim will be achieved by (1) examining Frank constants implied in the LL potential according to the simulation of the corresponding Fréedericksz transition and (2) giving a theoretical analysis to the LL model by analogy to the Frank theory.

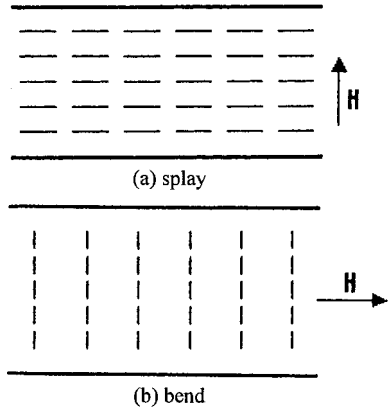

Figure 1. Schematic presentation of Fréedericksz transitions for (*a*) splay and (*b*) bend deformations. Corresponding transition points can be applied to measure the Frank elastic constants, K_{11} and K_{33}, respectively.

2. Simulation algorithm

For simplicity, the simulation is similar to those in [8, 9], performed on 2D square lattices. In fact, these modelling methods and our own can readily be extended into three dimensions. Our algorithm is unique in that the rotation of the local director at each lattice site is described by the Langevin equation written as

$$\xi \frac{\mathrm{d}\theta_i}{\mathrm{d}t} = f_{n,i} + f_{h,i} + f_{l,i} \tag{1}$$

where θ_i is the orientation angle of the local director at the ith lattice site with the zero angle defined along the plates or the horizontal direction; $f_{n,i}$, $f_{h,i}$, and $f_{l,i}$ denote the nematic torque due to anisotropic interaction between the NN lattices, the magnetic torque imposed on the local director and the Langevin stochastic torque, respectively; ξ is the friction coefficient and reads $\xi = kT/D$. Here, kT is the Boltzmann constant multiplied by the absolute temperature; D is the effective rotational diffusivity. Actually, the value of D depends, especially for a polymeric LC, upon the local orientation of the director and the local order parameter, due to the tube dilation effect [2]. In this paper, D is, for simplicity, assumed to be a constant. The back flow effect is also neglected, which, as usual, does not bring about serious consequences [1].

The 2D LL potential at position i, $E(\theta_i)$, can be written as

$$E(\theta_i) = \frac{1}{4} kT\, U \sum_{j \in NN} \sin^2(\theta_i - \theta_j) \tag{2}$$

where U designates the dimensionless strength of the nematic interaction reduced by kT. The summation is made over four NN sites. This potential is therefore free of the mean-field approximation. The corresponding elastic torque is readily expressed as $f_{n,i} = -\mathrm{d}E(\theta_i)/\mathrm{d}\theta_i$. Through this model, not only is the nematic interaction considered, but also the spatial correlation of the local directors is introduced.

The model system is divided into 50×50 lattices with the periodic boundary condition [3, 4, 14–17], unless otherwise indicated. When the Fréedericksz transition is dealt with, one dimension is restricted by the upper and lower plates. The surface-anchoring potential at position

i near the plates, $E_s(\theta_i)$, reads

$$E_s(\theta_i) = kT\, U_s \sin^2(\theta_i - \theta_a) \tag{3}$$

where U_s represents the dimensionless strength of the anchoring energy and is, tentatively, set equal to U in this paper; θ_a is the anchoring angle with $\theta_a = 0$ for the parallel anchoring associated with splay deformation and $\theta_a = \pi/2$ for the homeotropic anchoring associated with bend deformation.

The interaction energy between the magnetic fields and the local director can be described as

$$E_h(\theta_i) = -kT\, U_h \cos^2(\theta_i - \theta_h) \tag{4}$$

where U_h is the reduced dimensionless magnetic field strength and reads

$$U_h = \varepsilon_h/(kT), \qquad \varepsilon_h = 0{\cdot}5\chi_a H^2. \tag{5}$$

Here, H is the field strength; χ_a is the anisotropy of the magnetic susceptibility and is, without loss of generality, assumed to be positive; the term ε_h refers to the maximum magnetic energy under a given magnetic field strength. The magnetic torque can also be easily obtained by $f_{h,i} = -\mathrm{d}E_h(\theta_i)/\mathrm{d}\theta_i$.

The Langevin torque results from the Brownian thermal fluctuation. Its effect on the time evolution of the director rotation at position i can be realized by a white noise, W_i, which obeys the Einstein fluctuation–dissipation theorem

$$\langle W_i(t)\rangle = 0, \qquad \langle \mathrm{d}W_i(t)\,\mathrm{d}W_i(t')\rangle = \delta(t-t')\mathrm{d}t\mathrm{d}t'. \tag{6}$$

As a result, the Langevin equation (1) can be rewritten as

$$\mathrm{d}\theta_i = -\frac{1}{4}DU \sum_{j\in \mathrm{NN}} \sin[2(\theta_i - \theta_j)]\mathrm{d}t - DU_h \sin[2(\theta_i - \theta_h)]\mathrm{d}t + (2D)^{1/2}\mathrm{d}W_i. \tag{7}$$

In a computer simulation time must be discretized. The time step, $\Delta(Dt)$, is taken to be 0·01, which is reasonable [14]. A run-step is complete after all lattices are operated with equation (7) in sequence. (Operating randomly exhibits no essential difference for our simulation.) The number of run-steps or the evolution time in simulation is therefore directly proportional to the physical time with the coefficient of diffusivity, D. The LC dynamics can hence be described with a reasonable temporal scaling by the Brownian dynamics simulation.

3. Results and discussion

3.1. *Isotropic–anisotropic transition*

In an LC system, the calculation of the orientational order parameter is of central importance. In 2D the scaled order parameter, S, is defined as

$$S = \langle 2\cos^2(\theta_i - \Theta) - 1\rangle \tag{8}$$

where Θ represents the orientation of the whole system. S is a nonconserved order parameter in the evolution process. The equilibrium order parameter is obtained in the case without magnetic fields [$f_{h,i} = 0$ in equation (1)] and shown in figure 2 versus the dimensionless potential U. In order to reduce the scattering of the statistical value of S, the average is made over 5000 run-steps after the equilibrium state is reached or over 20 000 run-steps when U is near the transition point. The well-known statistical principle has been employed that the average over ensemble in the equilibrium state is equivalent to that over time. Our Brownian dynamics simulation gives reasonably a continuous isotropic–anisotropic transition in 2D (figure 2). The transition point, $U^* = 6{\cdot}7$, is higher than that predicted by mean-field theory ($U^* = 4$) where the thermal fluctuation is neglected [19]. (The value of U defined by Marrucci and Maffettone is only half of that by us, so $U^* = 2$ in their paper [19].)

3.2. *Domain growth dynamics*

Now we examine the dynamics of this system without magnetic fields and in the LC state ($U > U^*$). A typical trajectory relaxed from a random initial state is described by figure 3. This figure shows the texture evolution in a material that has been quenched from the athermal isotropic phase inside the nematic phase. Defects with different strengths and signs are produced and annihilated with time. But the defects with strength $|s| = 1/2$ predominantly remain because the energy of a defect is proportional to s^2 [1] and the defects with higher strength are therefore much less stable. This phenomenon is also reproduced in earlier computer modellings [8, 9]. The main advantage of our method

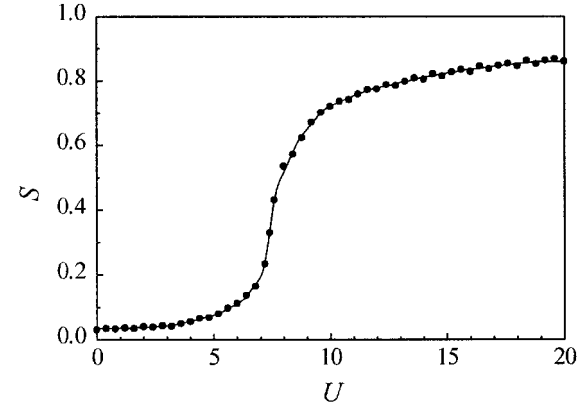

Figure 2. Scaled order parameter S as a function of reduced interaction potential U showing an isotropic–anisotropic transition.

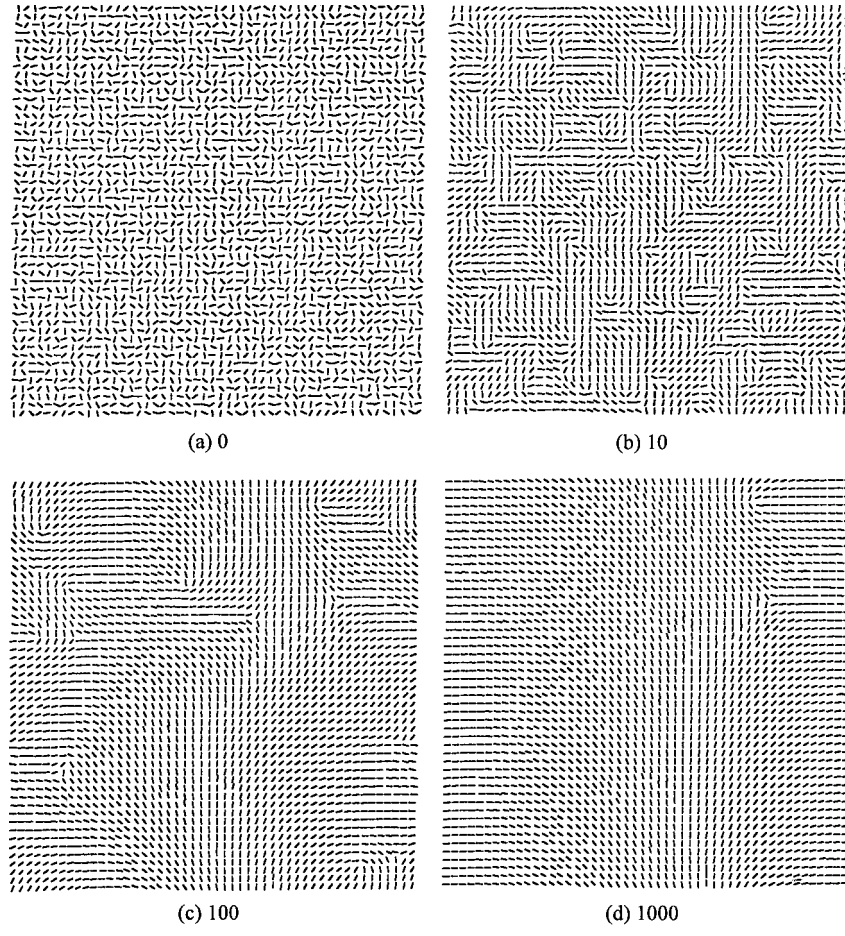

Figure 3. A typical trajectory for the texture evolution in an LC system initiated from a random isotropic state undergoing the indicated run-steps: $U=100$, $\Delta(Dt)=0{\cdot}01$ for each runstep.

is that the number of runsteps in the Brownian dynamics simulation is directly proportional to real time. Hence, domain growth dynamics can be studied quantitatively in the strict sense of dynamics.

Since it is not very convenient to measure domain size directly from the micrographs we employ another, indirect, characterization of the domain size termed 'inverse perimeter density' by the calculation of energy. From [13] the domain size $R_E(t)$ is defined as

$$R_E(t)=\frac{E_0}{E(t)-E_{\mathrm{eq}}} \tag{9}$$

where $E(t)$ is the total energy of the system at time t [for each lattice, the energy can be obtained from equation (2)]; E_0 is the system energy in the random initial state; E_{eq} is that in equilibrium and equals zero if the thermal fluctuation is neglected. The 'inverse perimeter density' is associated with the number of domain boundaries and hence of domain sizes [12, 13]. The result under the same condition as that in figure 3 is given in figure 4 (*a*). The average is taken over 80 independent trajectories. The good linear relation reveals a convincing scaling law, $R_E(t)\sim t^{1/2}$. It should be noted that due to the square lattice and the NN interaction chosen in the present paper, the domain boundaries prefer to orient along the horizontal and vertical directions (figure 3), which will, in principle, stabilize domain walls and slow the coarsening. Evolution velocities are thus dependent upon the form of lattice and interaction considered. Nevertheless, we believe that the fundamental defect-evolution behaviours may not change with lattice as usual, and particularly the domain-coarsening exponent may not be altered.

Such a scaling law exists in many systems [12, 13, 20]. By the coupled maps method based on a mean-field approximation, Wang confirmed that the growth exponent for the nonconserved three-state Potts model is 1/2 [12]. Due to the limit of the mean-field approximation, Wang ignored thermal fluctuation. This effect was also neglected in [8, 9]. In contrast to these, thermal fluctuation has been intrinsically included in our Brownian dynamics simulation. The other advantage of our approach is that if we set $f_{l,i}=0$ in equation (1), the case without thermal fluctuations can also be dealt with. The simulated outputs with and without thermal fluctuations are shown in figure 4 (*b*). The scaling law

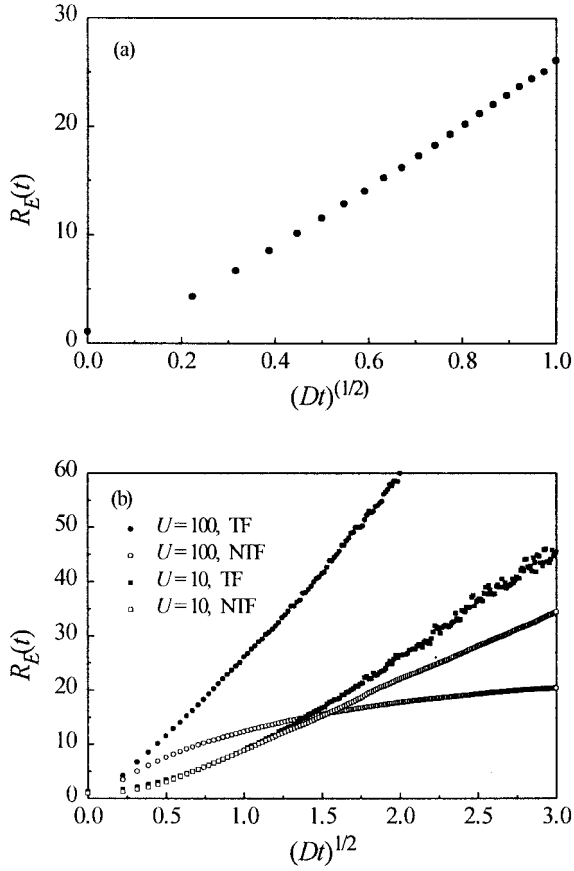

Figure 4. The domain size $R_E(t)$ vs $(Dt)^{1/2}$ for an LC system initiated from a random isotropic state. (*a*) Under the same conditions as those in figure 3; (*b*) comparison between domain growth dynamics with thermal fluctuation (TF) and without thermal fluctuation (NTF).

is not always obeyed without thermal fluctuations, especially for a high U, i.e. a low reduced temperature. It can be easily comprehended that because of the neglect of thermal fluctuations, the stationary state depends strongly on the initial conditions and the system is trapped in some local free energy minimum. Thermal fluctuations are, therefore, important and sometimes necessary to describe properly the dynamics of a non-equilibrium system. It is reasonable that the evolution is faster under a higher U than that under a lower U or with weaker nematic interactions, figure 4 (*b*). We can further see from figure 4 (*b*) that the neglect of thermal fluctuations does not enhance, but reduces the evolution velocity for the system studied in this paper.

3.3. *Fréedericksz transition*

Since the simulation is carried out on 2D lattices, only splay and bend deformations can be examined. When the Fréedericksz transition is dealt with, we let $U=15$, which guarantees that the system is in the nematic state. Corresponding Fréedericksz transitions are reproduced in a computer experiment by increasing the reduced magnetic field strength, U_h. A continuous transition takes place in 2D and the transition curves are shown in figure 5. In order to reduce the scattering of the data, the statistical values are obtained by averaging over 10 000 run-steps after the equilibrium state is reached at corresponding U_h. According to analysis of the Euler–Lagrange equation on the Frank energy [1], the critical transition point $U_h(\text{crit})$ ($\sim H^2$) must be inversely proportional to the square of the depth. The simulated outputs of $U_h(\text{crit})/U$ are 0·023, 0·012 and 0·006 under $d=10$, 14 and 20, respectively. The proportion is reasonably close to 4 : 2 : 1. Hence, Brownian dynamics simulation does seem to be an efficient approach in studying the external field effects on LC systems.

The more interesting result is that splay deformation and the corresponding bend deformation share the same Fréedericksz transition point, as shown in figure 6. This result reveals that the famous LL model implies the equal elastic-constant approximation, and the relationship between the 'molecular' model and the continuum model is thus elucidated by examination of the Fréedericksz transition with Brownian dynamics simulation.

3.4. *Relation between the LL potential and Frank energy*

At this point, we would like to ask how the LL potential is related to the Frank energy. In 2D and under the equal elastic-constant approximation, namely, $K_{11}=K_{33}=K$, the density of the Frank energy, g, can be easily expressed as

$$g=\frac{K}{2}\left[\left(\frac{\partial\theta}{\partial x}\right)^2+\left(\frac{\partial\theta}{\partial y}\right)^2\right] \tag{10}$$

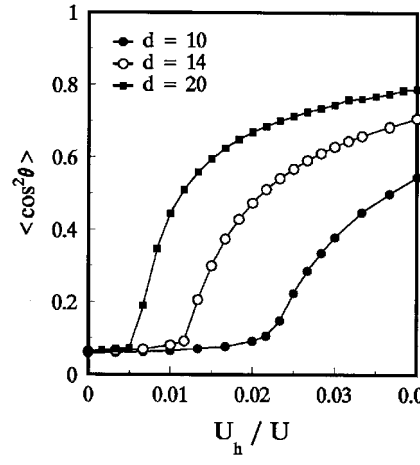

Figure 5. Fréedericksz transition curves with respect to the bend deformation and with different depths ($d=10$, 14, 20; $U=15$).

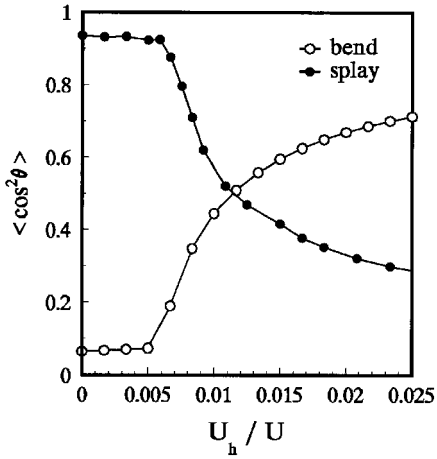

Figure 6. Fréedericksz transition curves with respect to the splay deformation and the bend deformation: $d=20$; $U=15$.

and the cross term such as $(\partial\theta/\partial x)(\partial\theta/\partial y)$ is fortunately eliminated under this approximation.

Now, we discretize the LC system into lattices and, similarly to the LL model, consider only the interactions between the central lattice and every one of the four NN lattices. Equation (10) is thus rewritten as

$$g(x,y)=\frac{K}{4}\{[\theta(x,y)-\theta(x-1,y)]^2+[\theta(x+1,y)-\theta(x,y)]^2+[\theta(x,y)-\theta(x,y-1)]^2+[\theta(x,y+1)-\theta(x,y)]^2\}. \quad (11)$$

Obviously, the differentials in equation (10) are replaced by the simple differences in equation (11), which stands only if $\Delta\theta\to 0$. Under this limitation condition,

$$\Delta\theta\approx\sin(\Delta\theta). \quad (12)$$

The elastic potential at the i-th lattice site can be expressed as

$$g_i=\frac{K}{4}\sum_{j\in \mathrm{NN}}\sin^2(\theta_i-\theta_j). \quad (13)$$

By comparison between this relation and the LL potential, equation (2), we get

$$K=kT\,U. \quad (14)$$

Hence, the interaction strength in the LL potential is equivalent to the Frank constant, and the macroscopic elastic distortion arises, in physical essence, from the microscopic or mesoscopic nematic interaction.

It is necessary to point out that, in the strict sense, equation (13) cannot be derived from equation (10), because the difference approximation used in obtaining equation (11) cannot always be satisfied in computer modelling. The analysis given above is, therefore, only based on analogy. In our opinion, equation (12) is a key to making the LL model successful, because $\sin^2(\Delta\theta)=\sin^2(\Delta\theta+m\pi)$ with m denoting an integral. This property of the sinusoidal function eliminates the possible mistake of directly using $\Delta\theta$ and guarantees the equivalence between $\mathbf{n}$ and $-\mathbf{n}$ for a nematic LC, where $\mathbf{n}$ is a unit vector standing for the director field.

Such an analysis might shed light onto a reasonable mesoscopic potential embodying unequal Frank constants and in three dimensions. It should be mentioned that Windle and co-workers have revised the LL model by introducing the two different Frank constants, K_{11} and K_{33}, in 2D [6]. In another paper, the twist elastic constant, K_{22}, was further included in three dimensions [7]. These pioneering researches are very helpful. Nevertheless, the so-termed K_{11}, K_{22} and K_{33} in their papers seem to be merely the coefficients of the energy function and no convincing proof has been given, up to now, to verify that these coefficients are really the Frank constants. Hence, further effort to propose and confirm a simple and reasonable expression of the nematic interaction for computer modelling, including unequal Frank constants, is strongly desired.

4. Conclusions

We have combined the LL model with Brownian dynamics simulation to study LC systems on lattices. The algorithm is verified to be a successful approach to simulation of the dynamic process, its main advantage being that the evolution time in the simulation embodies the physical time in the strict sense of dynamics, in contrast to other recent publications [8, 9]. The texture evolution of an LC system involving multiple defects is observed and the scaling law concerning domain growth is reasonably revealed with the growth exponent $\sim 1/2$ in 2D. Thermal fluctuations are found to be important and beneficial to the computer modelling of the present problem. We would like to suggest that this simulation methodology might be helpful for studies on nonlinear dynamics and nonequilibrium statistics of LCs besides the texture evolution described in this paper. As a preliminary extension, the Brownian dynamics simulation has been applied to investigate the unusual director tumbling phenomenon in liquid crystalline polymers under simple shear flow [14–16].

The Fréedericksz transition of an LC under static magnetic fields is further reproduced using Brownian dynamics simulation. We confirm that the simulation approach developed by us is also suitable for dealing with external field effects on LCs. Using Brownian dynamics simulation and the LL nematogen model, we have made the first investigation of the external static

magnetic field effect on director dynamics and flow instability in LCs subjected to simple shear flow [17]. A comparison of the transition points associated with splay and bend deformations shows that the LL model implies equal Frank elastic constants. With this approximation, the Frank energy used in the continuum theory can, by analogy, be discretized to yield the LL potential used in computer modelling studies. This analysis relates the two theories and suggests possible ways to derive a reasonable potential including different Frank constants.

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