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Calculation of the director configuration of nematic liquid crystals by the simulated-anneal method

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A new procedure for computing the equilibrium director pattern in a liquid-crystal-display cell subjected to an applied voltage is presented. It uses the simulated-anneal method which is based on the Metropolis Monte Carlo algorithm. The usefulness of the technique is illustrated by the simulation of three representative, but totally different kinds of liquid-crystal devices.

I. INTRODUCTION

Over the last few decades, the computation of electro-optical properties of twist-nematic liquid-crystal (LC) displays has been a matter of continued interest. These properties directly depend on the equilibrium director configuration of the LC material subjected to an externally applied field. This configuration is determined by the minimal total free energy of the LC system which depends on variations in the molecular tilt and twist angles, defining the local orientation of the director.

In most calculations, one determines the equilibrium director configuration by numerical integration of a set of differential equations, obtained analytically by applying the Euler-Lagrange equations on the Helmholtz free energy.[1-4] This technique, however, only finds an extremum of the free energy. Berreman[1] also mentions a second class of numerical methods, called relaxation methods, in which an initial director configuration is adjusted according to certain equations of motion that cause the free energy in the liquid crystal to relax towards a minimum. By including transverse motion of the fluid, these methods are also used to describe the dynamics.

This paper presents yet another method for determining the equilibrium director configuration. The Gibbs free energy given in detail in Sec. II is numerically minimized by the simulated-anneal (SA) technique. This algorithm, fully described in Sec. III, was first applied by Kirkpatrick *et al.*[5] to the optimization of computer design and recognized as a powerful minimization technique for multidimensional functions.[6] It has the advantage of not getting stuck in a local minimum by performing, in a controlled manner, uphill steps in the multidimensional parameter space. Moreover, it guarantees that a minimum instead of an extremum is found. In Sec. IV the usefulness of the method is illustrated by the simulation of three representative LC examples.

II. GIBBS FREE ENERGY

The equilibrium director configuration of a twist-nematic LC cell at a given voltage across the plates is found by minimizing the Gibbs free energy. Taking the z axis perpendicular to the cell surfaces with spacing d, this energy is expressed in terms of the angles $\theta\equiv\theta(z)$ and $\phi\equiv\phi(z)$. The tilt angle θ determines the orientation of the director measured from the xy plane, while the twist angle ϕ corresponds to the spherical ϕ coordinate. The Gibbs free energy G per unit area is then given by[7]

$$G\equiv G(\theta,\phi)=\int_0^d[G_E(z)-G_V(z)+G_S(z)]dz\ , \tag{2.1}$$

in which

$$G_E(z)=\frac{1}{2}\left[k_{33}(1-\gamma_1\cos^2\theta)\left[\frac{\partial\theta}{\partial z}\right]^2+k_{33}(1-\gamma_2\cos^2\theta)\cos^2\theta\left[\frac{\partial\phi}{\partial z}\right]^2-2k_{22}q_0\cos^2\theta\left[\frac{\partial\phi}{\partial z}\right]\right] \tag{2.2a}$$

is the elastic contribution,

$$G_V(z)=\frac{1}{2}\frac{D^2}{\epsilon_\parallel(1-\alpha\cos^2\theta)} \tag{2.2b}$$

denotes the electrostatic free-energy density, and the interaction with the surfaces of the cell is proposed to have the form[8]

$$G_S(z)=C_W[\delta(z)+\delta(z-d)]\sin^2(\theta-\theta_0)\ . \tag{2.2c}$$

Here $\gamma_1=(k_{33}-k_{11})/k_{33}$ and $\gamma_2=(k_{33}-k_{22})/k_{33}$, with k_{11}, k_{22}, and k_{33} being the elastic constants of the LC material for splay, twist, and bend, respectively. The dielectric constants of the material parallel and perpendicular to the director are denoted as $\epsilon_\parallel$ and $\epsilon_\perp$, respectively, and $\alpha=(\epsilon_{\parallel}-\epsilon_{\perp})/\epsilon_{\parallel}$. The natural pitch p of the material is incorporated in the formalism through the constant $q_0=2\pi/p$, and θ_0 denotes the pretilt angle. D stands for the dielectric displacement, C_W for the surface anchoring constant, and $\delta(z)$ for the Dirac δ function.

Most iterative calculations, however, start from the Helmholtz free energy,[1-4] which differs from the Gibbs free energy (2.1) only in that the sign of the electrostatic free energy is reversed. Hence the Helmholtz free energy takes the form

$$G_H=\int_0^d [G_E(z)+G_V(z)+G_S(z)]dz \ . \tag{2.3}$$

It has been demonstrated that both formulations lead to the same equilibrium director pattern.[9]

Since we want to know the equilibrium director configuration for a given voltage V, it is desirable to express the Gibbs free energy directly as a function of V by utilizing the relationship between the externally applied voltage and the dielectric displacement, the latter being constant throughout the cell. We have

$$V=\int_0^d \frac{D}{\epsilon_{\parallel}(1-\alpha\cos^2\theta)}dz \ , \tag{2.4}$$

from which it follows that

$$\int_0^d G_V(z)dz=\frac{V^2\epsilon_{\parallel}}{2}\left[\int_0^d \frac{1}{1-\alpha\cos^2\theta}dz\right]^{-1} . \tag{2.5}$$

For numerical work, the expression (2.1) has to be discretized with respect to z, i.e., the LC cell is divided into N layers, each layer having a thickness $h=d/N$. The derivative of the angles with respect to z is replaced by the simplest finite-difference approximation. The Gibbs free energy per unit area then becomes

$$\begin{aligned} G=\frac{1}{2h}\sum_{i=2}^{N} [(\theta_i-\theta_{i-1})^2 k_{33}(1-\gamma_1\cos^2\theta_i)+k_{33}(\phi_i-\phi_{i-1})^2(1-\gamma_2\cos^2\theta_i)\cos^2\theta_i] \\ -k_{22}q_0\sum_{i=2}^{N}(\phi_i-\phi_{i-1})\cos^2\theta_i-\frac{V^2\epsilon_{\parallel}}{2h}\left[\sum_{i=2}^{N}\frac{1}{1-\alpha\cos^2\theta_i}\right]^{-1}+C_W\sin^2(\theta_N-\theta_0) \ . \end{aligned} \tag{2.6}$$

It has to be stressed that in this formalism, the director configuration is simulated for the total thickness of the liquid-crystal-display (LCD) cell and no assumption concerning the symmetrical behavior of the tilt angle with respect to the middle of the cell is needed.[2,3,10] Only the tilt angle of the last layer is taken to be identical to that of the first layer, since the same boundary conditions are applied to both substrate plates. For practical purposes we have found it expedient to omit the contribution of the first layer [see (2.6)], but this is permitted as long as the layers have a sufficiently small thickness. Expression (2.6) will now be minimized with the help of the simulated-anneal method,[5,6] taking the angles θ_i and ϕ_i of the different layers as variables. It should be noted that it is not appropriate to replace the derivative of the angles by a finite-difference approximation of the form $(x_{i+1}-x_{i-1})/2h$. By doing so there is almost no coupling (except a small one due to the $\cos^2\theta_i$ dependence) between the angles of the odd- and the even-numbered layers, and consequently both sets of variables can be treated (almost) independently by the minimization procedure.

III. SIMULATED-ANNEAL METHOD

The simulated-anneal method[5,6] is a powerful minimization method based on thermodynamical concepts. In classical statistical physics, each state S with energy $E(S)$ occurs in a system in thermal equilibrium at a temperature T with a probability given by the Boltzmann distribution $e^{-\beta E(S)}$, in which $\beta=1/k_BT$ with k_B Boltzmann's constant. The mean value of a function f for such a system is then given by

$$\langle f\rangle=\frac{\int e^{-\beta E(S)}f(S)dS}{\int e^{-\beta E(S)}dS} \ . \tag{3.1}$$

From this, one can see that in the limit of $T\rightarrow 0$ $(\beta\rightarrow\infty)$, the mean value of f is determined by the function value of the ground state S_0 corresponding to the minimum energy $E(S_0)$. Replacing the energy $E(S)$ by a function to be minimized, e.g., $G(P)$, and the state S by the set of variables $P=\{p_i\}$, minimization of $G(P)$ is achieved by slowly increasing the value of β. If one wants to know the mean value of the set of variables P corresponding nearly to the minimum of $G(P)$, one has to calculate

$$\langle P\rangle=\frac{\int e^{-\beta G(P)}P\,dP}{\int e^{-\beta G(P)}dP} \ , \tag{3.2}$$

for a sufficiently large β, since in the limit of $T\rightarrow 0$ one finds

$$\lim_{\beta\rightarrow\infty}\langle P\rangle=P_0 \ , \tag{3.3}$$

where P_0 stands for the set of variables corresponding to the minimum of $G(P)$.

Now, for calculating expressions such as (3.1) or (3.2), one can use the Metropolis Monte Carlo (MMC) algorithm.[11,12] Defining the temperature-dependent probability density

$$\pi(S)=\frac{e^{-\beta E(S)}}{\int e^{-\beta E(S)}dS} \ , \tag{3.4}$$

the MMC procedure guarantees that the states S are generated according to this probability distribution by proceeding as follows.[11,12] Starting from a state S_i, a trial state S_j is constructed by application of a stochastic matrix M. Then the ratio $\pi(S_j)/\pi(S_i)=e^{-\beta[E(S_j)-E(S_i)]}$ is evaluated. If this ratio is greater than or equal to 1, the state S_j is accepted as a new state. Otherwise, this ratio is compared with a random number r uniformly distributed over the open interval (0,1) and the trial state S_j is only accepted if $\pi(S_j)/\pi(S_i)\geq r$. In case of rejection, the old state S_i is retained as the current state. Following this algorithm, each state S occurs with a probability $\pi(S)$ as long as the matrix elements of the matrix M fulfill the conditions[11]

$$(M)_{i,j}=(M)_{j,i}, \quad \forall i,j \ , \tag{3.5a}$$

$$\exists n \in \mathrm{N}:(M^n)_{i,j}\geq 0, \quad \forall i,j \ . \tag{3.5b}$$

The mean value of S can now be determined from

$$\langle S \rangle = \frac{1}{\mathrm{Card}(S)} \sum_{\{S\}} S \ , \tag{3.6}$$

where the sum covers those states generated by the MMC algorithm.

The power of this technique stems from the fact that states which do not necessarily possess a lower energy $E(S)$ are also accepted, and this with a probability density depending on the temperature. In terms of the minimization problem, this means that not only those sets of variables yielding lower function values are accepted. Consequently, this procedure allows for transitions out of a local minimum, the transition probability being regulated by the temperature.

For the problem at hand, expression (2.6) will be minimized by putting $G(P)=G=G(\theta,\phi)$ and regarding the angles $(\theta,\phi)=\{(\theta_i,\phi_i),\ i=1,2,\ldots,N\}$ as the set of variables. Of course the control parameter β is then to be considered as a fictitious "inverse temperature" and has no physical meaning. To implement the SA scheme we could, in principle, simply follow the prescribed procedure for setting up a MMC simulation algorithm, as given, for instance, in Ref. 12. However, due to the nature of our problem, an extension to the standard procedure is required. In practice, the MMC method usually consists of repeated attempts to change each of the variables one by one, i.e., by slowly moving through the "phase space." From (2.6) it can readily be seen that for problems of this kind such a scheme is bound to be highly inefficient. Indeed, as h is small relative to the other length scales of the LC model, changing one of the θ_i (or ϕ_i) by a significant amount implies a fairly large change in G. Hence, on average, such moves have a small likelihood of being accepted. Consequently, within a limited amount of computer time, the MMC will not sample the full phase space in an adequate manner. This is reflected in the simulation by excessively long annealing times. A way out of this problem is found by realizing that the structure of the integrals appearing in (3.2) is akin to that of the Feynman path integrals encountered in quantum-statistical physics. Therefore use can be made of the Monte Carlo techniques developed to compute such integrals.[13]

Thus the standard procedure has to be supplemented with a different kind of Monte Carlo step in order for the MMC to be effective. Instead of changing only one of the angles θ_k or ϕ_k, the possibility of changing all the θ angles at once with the same amount $\delta\theta$ is also built in. For such a MMC step, the change of the elastic energy is small compared with the change of the electrostatic energy. The effect is that the pace at which the system evolves to equilibrium is much greater. This extension has proven to be essential for a successful application of the SA idea to the problem at hand. Note that for the ϕ_i no such procedure is necessary because from (2.6) it follows directly that there is no competition between contributions of different origins. Indeed, a change in ϕ_i only implies a change in the elastic contribution to the free energy. Hereafter we will call the first kind of MMC steps single-angle moves and the latter ones multiangle moves.

From the definition of the tilt and the twist angle, it is clear that they are related to the spherical coordinates $\theta'\equiv\theta-\pi/2$ and $\phi'\equiv\phi$, respectively. In a Monte Carlo simulation of a model described in terms of spherical coordinates, it is well known that one has to take into account the fact that the θ' coordinate is not distributed uniformly over the interval $[0,\pi]$. Instead, it has to be weighed by a factor $\sin\theta'$.[12] Accordingly, for the coordinate system used for the LC model, the probability density reads

$$\pi(\theta,\phi)=\frac{e^{-\beta G(\theta,\phi)}\cos\theta_1\cdots\cos\theta_N}{\int d\theta_1\cdots\int d\theta_N\int d\phi_1\cdots\int d\phi_N\, e^{-\beta G(\theta,\phi)}\cos\theta_1\cdots\cos\theta_N} \ . \tag{3.7}$$

Therefore the MMC strategy is applied as follows. Assuming the current configuration to be (θ,ϕ), a trial configuration $(\tilde{\theta},\tilde{\phi})$ is generated. The ratio

$$\frac{\pi(\tilde{\theta},\tilde{\phi})}{\pi(\theta,\phi)}=e^{-\beta[G(\tilde{\theta},\tilde{\phi})-G(\theta,\phi)]}\prod_{k=1}^{N}\frac{\cos\tilde{\theta}_k}{\cos\theta_k} \tag{3.8}$$

is evaluated and the trial configuration $(\tilde{\theta},\tilde{\phi})$ is rejected as the new state if the ratio (3.8) is less than a random number uniformly distributed between 0 and 1. Further details about the implementation of the MMC procedure will be discussed in Sec. IV on the basis of three examples of simulations.

IV. APPLICATION

The algorithm, as discussed in Sec. III, has been tested on three different systems and the results have been compared with data obtained earlier with the help of iterative schemes which solve the Euler-Lagrange equations.[3,4] Of particular interest are systems for which the transmission-voltage curve exhibits a discontinuity. In the analytical treatment of the Euler-Lagrange equations, this discontinuity manifests itself in the divergence of elliptical integrals, rendering the iterative solution unstable. This problem is circumvented by the nonanalytical and noniterative SA method, as shown by the results of our second and third example.

For the first two examples, the material constants of ZLI-1132 (see Table I) are used and the thickness of the cell is taken to be 6.3 μm. The first example consists of a twisted-nematic (TN) LC cell ($\phi_T = 90°$) with a pretilt angle θ_0 of 3°. The natural pitch was chosen as 30 μm. As the second example, an SBE (supertwisted birefringence effect) display[14] with a total twist angle ϕ_T of 270°, a pretilt angle θ_0 of 30°, and a pitch p of 8.4 μm is considered. For both systems a strong anchoring with the cell plates is assumed, i.e., $C_W = 100 \times 10^{-6}$ J/m^2. The SA results of these systems can be checked with the iterative method described in Ref. 3. The third problem is defined equally to one of the systems of Ref. 4. It also consists of a TN system, but with a pretilt angle θ_0 of 0°, a natural pitch p of 63 μm, and weak anchoring with the substrate plates, which means once $C_W = 10.0 \times 10^{-6}$ J/m^2 and once $C_W = 8.3 \times 10^{-6}$ J/m^2. The material parameters for this system were fictive (see Table I), while the cell parameters were chosen such that the total free energy renders a discontinuity.

Most of the parameters controlling the SA simulation itself do not depend critically on the particular example, in spite of the fact that the physical behavior of the three systems differs substantially. The director pattern is computed for a voltage interval, the beginning and ending value of which can be chosen arbitrarily. The choice of the voltage step, however, depends on how fast the director changes as a function of the voltage. The starting configuration for the first value of the voltage is such that in each layer $\theta = 0.8\theta_0$ and that ϕ_i increases linearly from 0 to ϕ_T.

TABLE I. Material parameters of the liquid crystal ZLI-1132 (Merck) and of the fictitious material (FM) used for the calculation in Fig. 3.

Parameter	ZLI-1132	FM
k_{11}	10.1×10^{-12} N	10×10^{-12} N
k_{22}	8.6×10^{-12} N	10×10^{-12} N
k_{33}	19.7×10^{-12} N	30×10^{-12} N
$\epsilon_\parallel$	16.7	16
$\epsilon_\perp$	4.7	4
n_e ($\lambda = 480$ nm)	1.649	
n_0 ($\lambda = 480$ nm)	1.501	
n_e ($\lambda = 589$ nm)	1.630	
n_0 ($\lambda = 589$ nm)	1.492	

Trial configurations are generated as follows. In the case of a single-angle move, a randomly chosen element of the set $\{(\theta_i, \phi_i), i = 1, 2, \ldots, N\}$ is changed by an amount δ, chosen randomly from the interval $[-\Delta, \Delta]$. For a multiangle move all the θ_i are simultaneously changed by a similar amount. In our applications Δ was 1°. The ratio of single-angle moves to multiangle moves was kept to 50% for all systems. In order to fulfil condition (3.5a), all θ_i are taken modulo $\pi/2$, whereas all ϕ_i are taken modulo ϕ_T. On the other hand, by choosing $\Delta = 1°$, condition (3.5b) is not fulfilled for $n = 1$ since it is not guaranteed that each possible set of variables is reached by a single MMC step. But, in the limit of a large number of steps (which means large n), the matrix M becomes completely filled and, as such, requirement (3.5b) is met.

According to (3.8), the likelihood of accepting the trial configuration is "temperature" dependent. Cooling during the first voltage step is performed by gradually increasing β from 100 to 1000 over a number of MMC steps (equal to N_{wait}), while in each MMC step $(2 \times N)$ steps are performed so that each of the $(2 \times N)$ variables gets a chance to be updated. The choice of the final value of β is a compromise between a too inefficient calculation due to the low acceptance of trial configurations if β is large and a too inaccurate determination of the minimum of G if β is small.

For the next voltage, the initial director configuration is taken to be the final configuration of the previous voltage. Another amount of N_{wait} MMC steps is performed at the largest β, in order to let the system relax to its new equilibrium configuration. Cooling for each voltage step is possible, but this is less efficient since information obtained from the calculation for the previous voltage is then lost. From our examples it seems to be necessary to choose N_{wait} as large as 12 000, especially for those voltages where the director starts to rotate. After those N_{wait} steps another amount of steps (characterized by N_{samp},

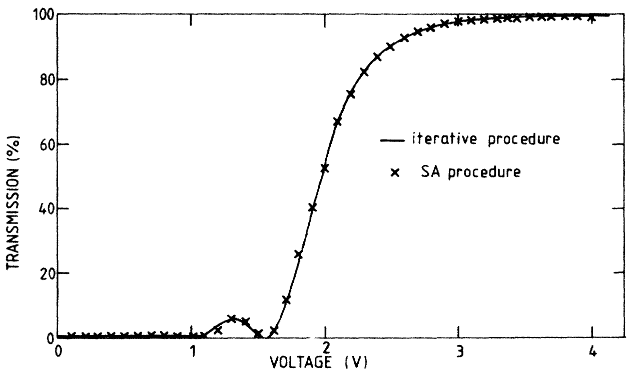

FIG. 1. Transmission-voltage curve for a twisted-nematic liquid-crystal cell ($\phi_T = 90°$, $\theta_0 = 3°$) of thickness $d = 6.3$ μm filled with ZLI-1132, having a natural pitch $p = 30$ μm, and a strong surface anchoring of $C_W = 100 \times 10^{-6}$ J/m^2 for light of wavelength $\lambda = 480$ nm incident on a cell with parallel polarizers. Crosses, SA calculation; continuous curve, iterative procedure (Ref. 3).

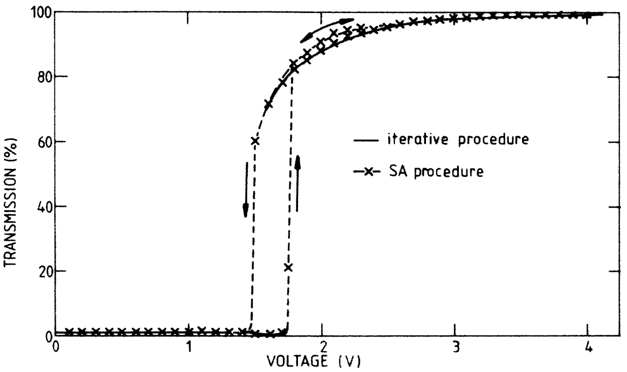

FIG. 2. Transmission-voltage curve for a nematic liquid-crystal cell ($\phi_T=270°$, $\theta_0=30°$) of thickness $d=6.3$ μm filled with ZLI-1132, having a natural pitch $p=8.4$ μm, and a strong surface anchoring of $C_W=100\times10^{-6}$ J/m^2, calculated for an increasing and decreasing sequence of voltages, for light of wavelength $\lambda=589$ nm incident on the cell with parallel polarizers. Dashed line through the crosses, SA calculation; continuous curve, iterative procedure (Ref. 3).

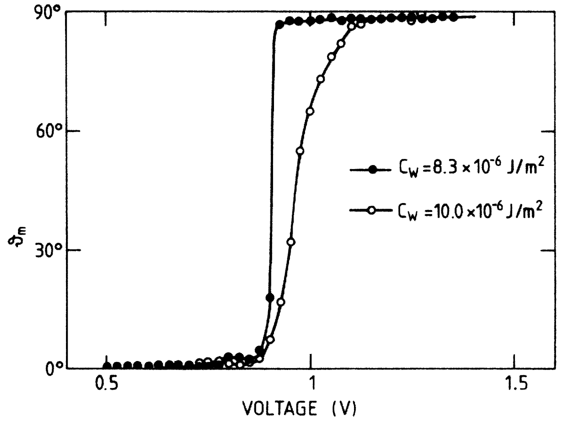

FIG. 3. Tilt angle in the middle (θ_m) of a twisted-nematic cell ($\phi_T=90°$, $\theta_0=0°$) as obtained from the SA procedure for two different values of the surface anchoring constant. The cell thickness $d=6.3$ μm, the natural pitch is infinite (in practice, $p=9000$ μm), and the material parameters, given in Table I, are fictitious.

which in our case equals 500) is performed to determine the averaged set of variables.

From the voltage dependence of the director pattern, the transmission-voltage curve for normally incident light is calculated by means of a thin-slice method in which the LC layer is considered as a stack of birefringent slabs, each possessing a constant thickness and a uniform orientation of the optical axis. For the TN cell (example 1), characterized by a strong anchoring at the substrate plates, there is no discontinuity in the free energy as a function of V. Hence it is not difficult to calculate iteratively the director configuration from the Euler-Lagrange formalism. The transmission-voltage curve obtained by SA and the results of iterative calculations[3] are presented in Fig. 1 and clearly show excellent agreement. In contrast to the TN cell, the SBE cell (example 2) exhibits a discontinuity in the free energy, implying the typical hysteresis effect in the transmission-voltage curve. Therefore, in the iterative approach the upper part of the transmission-voltage curve has to be calculated starting from a different configuration than the one used to obtain the lower part.[3] Figure 2 shows the iterative results for the lower and upper part of the transmission-voltage curve in comparison with the results obtained with the SA technique. The width of the hysteresis found by SA is somewhat larger than that obtained iteratively but, apart from that, there is satisfactory agreement. In complete contrast to the iterative method, the SA technique yields the complete curve for increasing (decreasing) voltage V, starting from one configuration. Consequently, no unpredictable actions have to be programmed for the computation of the transition regime. For the third problem, the weak anchoring strength may cause a discontinuity in the transmission-voltage curve. Figure 3 shows SA data for the tilt angle in the middle of the cell [$\theta_m\equiv\theta_m(V)$], rather than the transmission-voltage curve, as a function of the voltage for two different values of the anchoring constant. It can immediately be verified that our SA results agree well with the ones given in Ref. 4, taking into account, however, that in Fig. 3 θ_m has been determined by searching for the global minimum of the free energy, whereas in Ref. 4, θ_m corresponds to an extremum in the free energy. For those parts of the curves of Ref. 4 where the slope of $\theta_m(V)$ is negative, the free energy is maximal rather than minimal.

To sum up, we have demonstrated the usefulness of the SA technique for the determination of the equilibrium director pattern within a LC cell with an external voltage applied to the substrate plates. The method searches directly for minima instead of extrema of the free energy. A disadvantage of the technique is that it uses about a factor of 10 more computer time compared with the procedure described in Ref. 3. On the other hand, it requires much less human intervention. Moreover, it has the advantage of being more flexible in the choice of the material and cell parameters and of being insensitive to the initial conditions. Finally, since no analytical manipulation precedes the numerical computation, it is straightforward to apply the technique to other forms of the free energy.

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[1] D. W. Berreman, Philos. Trans. R. Soc. London, Ser. A **309**, 203 (1983).

[2] P. A. Breddels and H. A. van Sprang, J. Appl. Phys. **58**, 2162 (1985).

[3] H. A. van Sprang and P. A. Breddels, J. Appl. Phys. **60**, 968 (1986).

[4] M. E. Becker, B. B. Kosmowski, and D. A. Mlynski, Proc. SID **26**, 109 (1985).

[5] S. Kirkpatrick, C. D. Gelatt, and M. P. Vecchi, Science **220**, 671 (1983).

[6] P. J. M. van Laarhoven and E. H. L. Aarts, *Simulated Annealing: Theory and Applications* (Reidel, Dordrecht, 1987).

[7] P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1974).

[8] A. Rapini and M. Papoular, J. Phys. (Paris) Colloq. **30**, C4-54 (1969).

[9] R. N. Thurston and D. W. Berreman, J. Appl. Phys. **52**, 508 (1981).

[10] F. C. Fraser, J. Phys. A **11**, 1439 (1978).

[11] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. **21**, 1087 (1953).

[12] K. Binder and D. Stauffer, in *Application of the Monte Carlo Method in Statistical Physics*, Vol. 36 of *Topics in Current Physics*, edited by K. Binder (Springer, Berlin, 1984).

[13] H. De Raedt and A. Lagendijk, Phys. Rep. **127**, 233 (1985).

[14] T. J. Scheffer and J. Nehring, Appl. Phys. Lett. **45**, 1021 (1984).