

Instability of a Biaxial Nematic Liquid Crystal Formed by Homeotropic Anchoring on Surface Grooves *

ZHANG Zhi-Dong(张志东)^{1**}, XUAN Li(宣丽)²

¹Department of Applied Physics, Hebei University of Technology, Tianjin 300401

²State Key Lab of Applied Optics, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033

(Received 2 January 2011)

A method used to treat the elastic distortion of a uniaxial nematic liquid crystal induced by homogeneous anchoring on the surface grooves is generalized to biaxial nematic liquid crystals under the homeotropic anchoring condition. Employing some approximations for the elastic constants, we obtain an additional term in the elastic energy per unit area which depends on the angle between the minor director at infinity and the direction of the grooves, with a period of $\pi/2$. This leads to instability on the surface grooves so that two states with crossed minor directors are energetically indistinguishable. Our theoretical study explains why the homeotropic alignment method developed for uniaxial liquid crystals loses efficacy for biaxial nematics.

PACS: 61.30.Hn

DOI: 10.1088/0256-307X/28/10/106101

In most liquid crystal devices, the liquid crystals are sandwiched between two substrates coated with alignment layers. In the absence of externally applied fields, the orientation of the liquid crystal in the cell is determined by the anchoring condition of the alignment layer.^[1–3] One usually distinguishes three main types of liquid crystalline director alignment near solid walls: homeotropic, homogeneous (or planar) and tilted orientations. Here we study the first of these and consider the biaxial nematic phase, which was observed in lyotropic systems as early as 1980^[4] and has been confirmed by deuterium NMR spectroscopy. By 2004, firm evidence for this phase in thermotropics had been reported.^[5–8] For practical applications, new types of electro-optic devices with fast response are expected from this phase, due to the fact that the switching of the minor director \mathbf{m} might be much faster than that of the main director \mathbf{n} . Such devices require homeotropic alignment with in-plane switching.^[5,9]

In order to realize homeotropic alignment of biaxial nematic liquid crystals, different methods have been developed and used successfully for the uniaxial nematic phase. One is a special polymer coated on the substrate.^[9–13] A process of weak rubbing can be used to produce surface grooves which provide a preferred direction, so that a monodomain state with a uniformly tilted director field can be built up above the bend Freedericksz transition in a material with negative dielectric anisotropy. Recently, experimental realization of a submicron-scale surface grooved with sufficient geometrical precision has again aroused in-

terest in the notion of surface anchoring attributable to the geometry of the surface.^[14–17] In particular, Fukuda *et al.*^[14] re-examined the theory of homogeneous anchoring due to Berreman^[18] to discuss the possible effect of azimuthal distortions of a uniaxial nematic liquid crystal in contact with a sinusoidal wavy surface, and we have generalized this method to biaxial nematic liquid crystals.^[19] In the Berreman approximation,^[14,18,19] a surface groove was described by a sinusoidal wave with wave vector $q = 2\pi/\lambda$ and amplitude A , where λ is the spatial periodicity of the surface, and the condition that $qA \ll 1$ should be satisfied. In this Letter, we deal with homeotropic anchoring of biaxial nematic liquid crystals on surface grooves in the Berreman approximation.

It is well known that the molecular shape of a liquid crystal is a key factor in determining its properties. In particular, thermotropic biaxial nematics have been found mainly in bent core liquid crystals.^[6,7] However, even for a submicron-scale surface groove, the distances l ($l \sim \lambda$) over which significant variations of the tensor order parameter occur are much larger than the molecular dimensions (typically ~ 2 nm). Thus the deformations may be described by a continuum theory disregarding the details of the structure on the molecular scale.^[20] The molecular shape influences elastic properties according to the bulk elastic constants. For the present theoretical study, we assume that the particular shape of a liquid crystal molecule is not relevant, and that the mathematical description works for any biaxial nematic liquid crystal with the appropriate elastic constants. The elasticity of a biaxial ne-

*Supported by the National Natural Science Foundation of China under Grant Nos 60878047 and 60736042, the Hebei Provincial Natural Science Foundation under Grant No A2010000004, and the Key Subject Construction Project of Universities in Hebei Province.

**Corresponding author. Email: zhidong_zhang@yahoo.cn

© 2011 Chinese Physical Society and IOP Publishing Ltd

matic is described by 15 elastic constants, 12 of which correspond to director distortions in the bulk, and 3 amount to surface-like elasticity.^[21] The elastic free energy density, as given in Ref. [21], is

$$F = \sum_{a,b,c} \frac{1}{2} [K_{aa}(\mathbf{a} \cdot \nabla \mathbf{b} \cdot \mathbf{c})^2 + K_{ab}(\mathbf{a} \cdot \nabla \mathbf{a} \cdot \mathbf{b})^2 + K_{ac}(\mathbf{a} \cdot \nabla \mathbf{a} \cdot \mathbf{c})^2] + C_{ab}(\mathbf{a} \cdot \nabla \mathbf{a}) \cdot (\mathbf{b} \cdot \nabla \mathbf{b}) + k_{0,a} \nabla \cdot (\mathbf{a} \cdot \nabla \mathbf{a} - \mathbf{a} \nabla \cdot \mathbf{a}), \quad (1)$$

where the summation is over a cyclic permutation of the three directors and indices. Hereafter, the director fields are denoted by \mathbf{l} , \mathbf{m} and \mathbf{n} for convenience. Let the orientation of the director triad in the uniform state be

$$\mathbf{l} = (1, 0, 0); \quad \mathbf{m} = (0, 1, 0); \quad \mathbf{n} = (0, 0, 1). \quad (2)$$

When the distortion of the biaxial nematics from the uniform state is small enough, we can write the director triad as

$$\mathbf{l} = (1, l_y, l_z); \quad \mathbf{m} = (m_x, 1, m_z); \quad \mathbf{n} = (n_x, n_y, 1). \quad (3)$$

As \mathbf{l} , \mathbf{m} , \mathbf{n} are orthonormal, one has

$$m_x = -l_y; \quad n_y = -m_z; \quad l_z = -n_x. \quad (4)$$

Thus, only three of the six perturbations in Eq. (3) are independent. Under the conditions given by Eqs. (3) and (4), the elastic free energy density is written as^[22]

$$g_b = \frac{1}{2} K_{LL} (m_{z,x})^2 + \frac{1}{2} K_{MM} (n_{x,y})^2 + \frac{1}{2} K_{NN} (l_{y,z})^2 + \frac{1}{2} K_{LM} (l_{y,x})^2 + \frac{1}{2} K_{MN} (m_{z,y})^2 + \frac{1}{2} K_{NL} (n_{x,z})^2 + \frac{1}{2} K_{ML} (l_{y,y})^2 + \frac{1}{2} K_{NM} (m_{z,z})^2 + \frac{1}{2} K_{LN} (n_{x,x})^2 - C_{LM} n_{x,x} m_{z,y} - C_{MN} l_{y,y} n_{x,z} - C_{NL} m_{z,z} l_{y,x} - 2k_{0,L} (l_{y,z} n_{x,y} - l_{y,y} n_{x,z}) - 2k_{0,M} (l_{y,z} m_{z,x} - l_{y,x} m_{z,z}) - 2k_{0,N} (n_{x,y} m_{z,x} - n_{x,x} m_{z,y}), \quad (5)$$

where the indices L , M and N are used instead of a , b and c . The equations of equilibrium are

$$-K_{LL} m_{z,xx} - K_{MN} m_{z,yy} - K_{NM} m_{z,zz} + C_{LM} n_{x,xy} + C_{NL} l_{y,xz} = 0, \quad (6a)$$

$$-K_{MM} n_{x,yy} - K_{NL} n_{x,zz} - K_{LN} n_{x,xx} + C_{LM} m_{z,xy} + C_{MN} l_{y,yz} = 0, \quad (6b)$$

$$-K_{NN} l_{y,zz} - K_{LM} l_{y,xx} - K_{ML} l_{y,yy} + C_{MN} n_{x,yz} + C_{NL} m_{z,xz} = 0, \quad (6c)$$

Saupe pointed out that in the uniaxial phase, there

are^[21]

$$K_{LN} = K_{MN} = K_1, \quad (7a)$$

$$K_{MM} = K_{LL} = K_2, \quad (7b)$$

$$K_{NL} = K_{NM} = K_3, \quad (7c)$$

$$C_{LM} = K_1 - K_2, \quad (7d)$$

$$C_{MN} = C_{NL} = 0, \quad (7e)$$

$$2k_{0,N} = K_{24} - K_2, \quad (7f)$$

$$K_{NN} = K_{LM} = K_{ML} = 0. \quad (8)$$

Taking Eqs. (4) and (7) into account, Eqs. (6b) and (6a) lead to

$$K_1 n_{x,xx} + K_2 n_{x,yy} + K_3 n_{x,zz} + (K_1 - K_2) n_{y,xy} = 0, \quad (9a)$$

$$K_2 n_{y,xx} + K_1 n_{y,yy} + K_3 n_{y,zz} + (K_1 - K_2) n_{x,xy} = 0. \quad (9b)$$

Equations (9a) and (9b) completely correspond to Eqs. (7) and (8) in Ref. [14] (we assume that $\mathbf{n} = (n_x, n_y, 1)$ instead of $\mathbf{n} = (1, n_y, n_z)$ as in Ref. [14]).

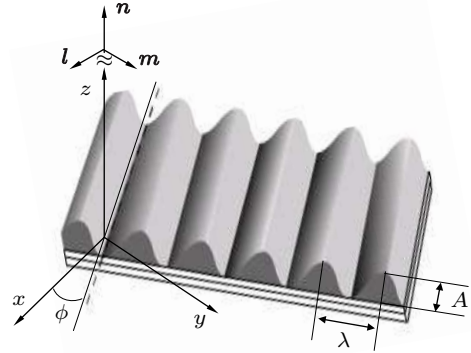


Fig. 1. Schematic representation of a sinusoidally shaped groove surface with amplitude A and spatial periodicity λ . At $z \rightarrow \infty$, there are $\mathbf{l} = (1, 0, 0)$, $\mathbf{m} = (0, 1, 0)$ and $\mathbf{n} = (0, 0, 1)$. Here ϕ is the angle between the x axis and the direction of the grooves, i.e. the angle between the minor director \mathbf{l} at infinity and the direction of the surface grooves.

Singh *et al.*^[22] predicted that in molecular theory, the seven elastic constants, namely, K_{LN} , K_{MN} , K_{MM} , K_{LL} , K_{NL} , K_{NM} and C_{LM} are of the order of the values found in the uniaxial nematic phase, and among the three C constants associated with mixed models of deformation, C_{LM} is about ten times larger than the other two. Similar results were obtained by Liu.^[23] Thus in the present preliminary work on biaxial nematics, we assume that Eqs. (9a) and (9b) can still be used approximately, and $C_{MN} = C_{NL} = 0$, i.e. the mixed elastic constants can be neglected except C_{LM} . This approximation means that the differences between splay and twist elastic constant are

neglected for both the \mathbf{l} director and the \mathbf{m} director. Consequently, Eq. (6c) becomes

$$K_{LM}l_{y,xx} + K_{ML}l_{y,yy} + K_{NN}l_{y,zz} = 0. \quad (10)$$

Following Fukuda *et al.*,^[14] we consider a surface groove whose shape can be described by

$$z = \zeta(x, y) = A \sin[q(x \sin \phi + y \cos \phi)], \quad (11)$$

where ϕ is the angle between the x axis (this axis coincides with the minor director \mathbf{l} at infinity) and the direction of the grooves, see Fig. 1. A biaxial nematic is filled in the infinite region $z > \zeta(x, y)$. We further assume that the \mathbf{n} director at the surface is perpendicular to it, so that one has

$$n_x = -Aq \sin \phi \cos[q(x \sin \phi + y \cos \phi)], \quad (12a)$$

$$n_y = -Aq \cos \phi \cos[q(x \sin \phi + y \cos \phi)]. \quad (12b)$$

Thus, Eqs. (9a) and (9b) can be solved under the boundary condition at the surface and under the boundary condition $n_x = n_y = 0$ at $z \rightarrow \infty$ (see Fig. 1). One easily obtains

$$n_x(x, y, z) = -Aq \sin \phi \cos[q(x \sin \phi + y \cos \phi)] \cdot \exp[-qz\sqrt{K_1/K_3}], \quad (13a)$$

$$n_y(x, y, z) = -Aq \cos \phi \cos[q(x \sin \phi + y \cos \phi)] \cdot \exp[-qz\sqrt{K_1/K_3}]. \quad (13b)$$

There should be an additional boundary condition at the surface $z \sim 0$,^[14]

$$\frac{\partial g_b}{\partial n_{x,z}} \delta n_x + \frac{\partial g_b}{\partial n_{y,z}} \delta n_y + \frac{\partial g_b}{\partial l_{y,z}} \delta l_y = 0. \quad (14)$$

Because n_x and n_y are fixed at the surface and no condition is imposed on δl_y , Eq. (14) gives

$$\frac{\partial g_b}{\partial l_{y,z}} = 0, \quad (15)$$

so that one has

$$K_{NN}l_{y,z} + 2k_{0,M}n_{y,x} - 2k_{0,L}n_{x,y} = 0. \quad (16)$$

Now, one can find the analytical solution of Eq. (10) with the boundary conditions given by Eq. (16) at $z \sim 0$, and $l_y = 0$ at $z \rightarrow \infty$,

$$l_y = \frac{2Aq(k_{0,L} - k_{0,M}) \sin \phi \cos \phi}{K_{NN}\tilde{h}(\phi)} \cdot \sin[q(x \sin \phi + y \cos \phi)] \exp[-qz\tilde{h}(\phi)], \quad (17)$$

with $\tilde{h}(\phi) = \sqrt{(K_{LM} \sin^2 \phi + K_{ML} \cos^2 \phi)/K_{NN}}$.

From Eq. (6), distortion energy per unit area is written as

$$f_b(\phi) = f_a + \Delta f_b(\phi), \quad (18a)$$

$$f_a = \frac{1}{4}A^2q^3\sqrt{K_1K_3}, \quad (18b)$$

$$\Delta f_b(\phi) = -\frac{1}{4} \frac{A^2q^3(k_{0,M} - k_{0,L})^2 \sin^2 2\phi}{K_{NN}\tilde{h}(\phi)}. \quad (18c)$$

Firstly, for the uniaxial nematic phase, $f_b(\phi)$ reduces to f_a . Thus, homeotropic alignment on the surface is not destroyed by the surface grooves, at least in Berreman's approximation. Secondly, setting the approximation $K_{LM} \approx K_{ML}$, one has $\tilde{h}(\phi) \approx \sqrt{K_{LM}/K_{NN}}$ and Eq. (18c) gives the minimum at $\phi = \pi/4$ and $\phi = 3\pi/4$, respectively. In other words, it leads to instability on the surface grooves such that two states with crossed minor directors are energetically indistinguishable, i.e. $f_b(\phi + \pi/2) = f_b(\phi)$. However, in order to obtain a monodomain vertical alignment state, there should be $f_b(\phi + \pi) = f_b(\phi)$.

Lee *et al.*^[9] used a polyimide RN-1720 (Nissan for uniaxial liquid crystals, the pretilt angle 1° – 2°) to achieve homogenous alignment for a biaxial nematic liquid crystal (ODBP-Ph-C₇), but they used another polyimide AL-6010 (JSR for uniaxial liquid crystals, with pretilt angle 89°) to achieve only a midtilted alignment for the same biaxial nematic liquid crystal. Thus our theoretical study explains why the homeotropic alignment method developed for uniaxial liquid crystals loses efficacy for biaxial nematics.

In the case of homogeneous alignment, we have^[19]

$$\Delta f_b(\phi) = \frac{1}{4}A^2q^3K_{LM}h(\phi) \cos^2 \phi, \quad (19)$$

with $h(\phi) = \sqrt{(K_{ML} \cos^2 \phi + K_{NN} \sin^2 \phi)/K_{LM}}$. We should emphasize that Eq. (19) gives an additional distortion energy based upon the azimuthal anchoring energy,

$$F(\phi) = \frac{1}{2}W \sin^2 \phi, \quad (20)$$

where

$$W = \frac{1}{2}A^2qK_{24} \left(2 - \frac{1}{2}K_{24} \frac{K_1 + K_2}{K_1K_2} \right). \quad (21)$$

As a result, in the one-constant model for each director the stability condition is that the elastic constant of the main director is maximum.^[19] In fact, homogeneous alignment for biaxial nematics has been obtained over a wide range.^[9–13,24]

In summary, a method that treats the elastic distortion of uniaxial nematic liquid crystals induced by homogeneous anchoring on the surface grooves has been generalized to biaxial nematic liquid crystals under the homeotropic anchoring condition. With some

approximations for the elastic constants, we have obtained an additional term in the elastic energy per unit area which depends on the angle between the minor director at infinity and the direction of the grooves, with a period of $\pi/2$. This leads to instability on the surface grooves such that two states with crossed minor directors are energetically indistinguishable. We must point out that other reasons may give rise to the homeotropic alignment loss of efficacy for biaxial nematics. Taking the molecular shape into account, the alignment layers may only align one of the arms of bent core molecules, while the other arm has free orientation.^[25] Although a quantitative study in which much attention is paid to the molecular shape is beyond the scope of this Letter, we mention that an anchoring theory taking the molecular shape into account leads to the elastic constants being dependent on position in the liquid crystal-alignment layer interface.^[3]

References

- [1] Sonin A A 1995 *The Surface Physics of Liquid Crystals* (Luxembourg: Gordon and Breach)
- [2] Uchida U 1990 *Surface Alignment of Liquid Crystals* in *Liquid Crystals: Applications and Uses* ed Bahadur B vol 3 (Singapore: World Scientific)
- [3] Faetti F 1991 *Anchoring Effects in Nematic Liquid Crystals* in *Liquid Crystal Materials* ed Khoo I C (Amsterdam: Gordon and Breach)
- [4] Yu L J and Saupe A 1980 *Phys. Rev. Lett.* **45** 1000
- [5] Luckhurst G R 2004 *Nature* **430** 413
- [6] Madsen L A, Dingemans T J, Nakata M and Samulski E T 2004 *Phys. Rev. Lett.* **92** 145505
- [7] Acharya B R, Primak A and Kumar S 2004 *Phys. Rev. Lett.* **92** 145506
- [8] Merkel K, Kocot A, Vij J K, Korlacki R, Mehl G H and Meyer T 2004 *Phys. Rev. Lett.* **93** 237801
- [9] Lee G S, Cho J S, Kim J C, Yoon T H and Shin S T 2009 *J. Appl. Phys.* **105** 094509
- [10] Stannarius R, Eremin A, Tamba M-G, Pelzl G and Weissflog W 2007 *Phys. Rev. E* **76** 061704
- [11] Kumar P, Hiremath U S, Yelamagad C V, Rossberg A G and Krishnamurthy K S 2008 *J. Phys. Chem. B* **112** 9270
- [12] Le K V, Mathews M, Chambers M, Harden J, Li Q, Takezoe H and Jákli A 2009 *Phys. Rev. E* **79** 030701
- [13] Jang Y, Panov V P, Kocot A, Vij J K, Lehmann A and Tschierske C 2009 *Appl. Phys. Lett.* **95** 183304
- [14] Fukuda J I, Yoneya M and Yokoyama H 2007 *Phys. Rev. Lett.* **98** 187803; *ibid.* **99** 139902
- [15] Lamau L, Kondrat S and Poniewierski A 2007 *Phys. Rev. E* **76** 051701
- [16] Barbero G, Gliozzi A S, Scalerandi M and Evangelista L R 2008 *Phys. Rev. E* **77** 051703
- [17] Barbero G, Gliozzi A S and Scalerandi M 2008 *J. Appl. Phys.* **104** 094903
- [18] Berreman D W 1972 *Phys. Rev. Lett.* **28** 1683
- [19] Zhang Z D and Ye W J 2009 *Liq. Cryst.* **36** 885
- [20] de Gennes P G and Prost J 1993 *The Physics of Liquid Crystals* 2nd edn (Oxford: Oxford University) p 98
- [21] Saupe A 1981 *J. Chem. Phys.* **75** 5118
- [22] Singh Y, Rajesh K, Menon V J and Singh S 1994 *Phys. Rev. E* **49** 501
- [23] Liu H 2008 *Chin. Phys. B* **17** 1060
- [24] Xiang Y, Goodby J W, Görtz V and Gleeson H F 2009 *Appl. Phys. Lett.* **94** 193507
- [25] Le Khoa K V, Mathews M, Chambers M, Harden J, Li Q, Takezoe H and Jákli A 2009 *Phys. Rev. E* **79** 030701(R)