Zigzag Disclinations in Biaxial Nematic Liquid Crystals

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Disclination lines are observed in the biaxial nematic phase of the lyotropic system comprised of rubidium laurate, 1-decanol, and H_2O. They are shown to have a uniaxial nematic core which couples to the anisotropic elastic field produced by the surrounding medium. This interaction makes the disclination lines tilt and break into zigzags. The tilt angle is measured in the vicinity of the uniaxial to biaxial nematic phase transition, and found to vary linearly with temperature, which is consistent with the classical behavior of this phase transition.

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Lyotropic mesophases, which are formed by amphiphilic molecules dissolved in water, are known to exhibit different nematic phases depending on the temperature and concentration conditions[1]: a calamitic nematic phase N_c composed of (supposedly[2]) cylindrical micelles, a discotic nematic phase N_d made of discotic micelles, and a biaxial nematic phase N_{bx}, discovered by Yu and Saupe,[3] which is intermediate between the two former uniaxial nematic phases. A lot of experimental and theoretical studies have been stimulated by this new nematic phase.

It has been shown that the N_{bx} phase is composed of flat micellar aggregates of statistically elliptical shape[2] which are oriented, except for small fluctuations, with their symmetry axes parallel to three directors, **n** **m**, and $\mathbf{l}=\mathbf{n}\times\mathbf{m}$: **n** is the director perpendicular to the plane of the micelles (**n** identifies to the uniaxial director in the N_d phase where the micelles are statistically circular), and **m** and **l** are the directors along the long and short axes of the elliptic micelles, respectively. Continuous distortions in the orientation of these directors cost an elastic energy which has been shown to involve twelve elastic constants in general.[4,5] In fact, the problem is simplified in the vicinity of the N_d (or N_c) phase because the elastic constants vary continuously with temperature through the second-order phase transitions (e.g., the N_d-N_{bx} transformation[6]). Three out of twelve elastic constants are involved in distortions of the quasiuniaxial director **n**. They are of the Frank type, and, in the one-constant approximation, they can be taken equal to the same value K. The nine other elastic constants are involved in distortions of the biaxial directors **m** and **l**. They therefore vanish at the N_d-N_{bx} phase transition as $K' \sim KQ^2$, where Q is the scalar order parameter of the N_d-N_{bx} transition. Since the N_d-N_{bx} phase transition is of dau type,[6] Q is given by the relation $Q^2 \sim \alpha\Delta T$, where α is a proportionality coefficient, and ΔT is the temperature distance to the N_d-N_{bx} phase transition. Finally, the typically biaxial elastic constants scale as

$$K' \sim K\alpha\Delta T. \tag{1}$$

Discontinuities in the orientation of the directors, i.e., disclinations, corresponding to a local break of symmetry may also occur. In this Letter, we report the first observation of disclination lines in the N_{bx} phase, near the N_d-N_{bx} phase transition. Under particular conditions, the disclination lines make zigzags. This is actually a surprising behavior; the same disclinations in classical uniaxial nematics have a flexible shape (nema in Greek means hair).

The sample is a mixture of rubidium laurate (synthesized in the laboratory), 1-decanol (from Fluka), and H_2O, in proportions 31.0, 7.0, and 62.0 wt.%, respectively. A small amount of a ferrofluid ($<10^{-4}$ by weight) is added in order to help the alignment in the magnetic field.[7] As in similar lyotropic systems,[3] this sample presents the following nematic phases as a function of temperature: discotic uniaxial (N_d) below 13 °C, biaxial (N_{bx}) in a large temperature range, and discotic uniaxial again above 22.5 °C. After a complete mixing of the compounds, alternating shakings and centrifugations, the sample is placed in flat capillaries of 200 μm thickness (microslides from Vitro Dynamics, Inc.) and sealed at both ends with Parafilm and epoxy glue. These flat capillaries provide good optical conditions for observation under the polarizing microscope. They also have the great advantage of reproducibly[3] orienting the N_{bx} phase with the micellar aggregates parallel to the glass surfaces and their long axes along the shear velocity (that is, in the direction of the capillary) when the sample is introduced while in the N_{bx} phase, i.e., at room temperature for our mixture. Note also that this orientation is favored by the inequality $K > K' \sim KQ^2$, which makes the curvature of the director **n** more energetic than that of **m** or **l**. After this surface orientation is achieved, a magnetic field **H** is applied horizontally, perpendicular to the capillary and parallel to its plane windows. The micelles remain parallel to the plane windows and orient their long axes parallel to **H** in the bulk of the sample (right part of Fig. 1), which introduces a competition with the surface conditions previously defined on the edge of the capillary (left part of Fig. 1).

In the N_d phase, which is classically uniaxial, the sample solves this constrained situation by creating

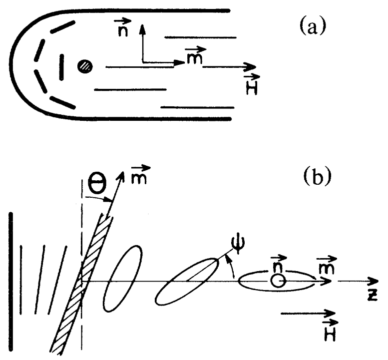

FIG. 1. (a) Transversal (vertical) and (b) longitudinal (horizontal) cuts of the sample in the N_{bx} phase. The biaxial micelles are sketched as ellipses, as short and thick dashes, or as long and thin dashes, in accordance to their being in-plane or cut along their short or long axes, respectively. The hatched part is the core of the disclination line. The broken line in (b) is the axis of the cylindrical edge of the capillary.

two $\frac{1}{2}$-disclination lines in the vicinity of the axes of the half-cylindrical capillary edges. In the core of these disclination lines, the orientation is not defined, i.e., the cores are melted in the isotropic phase. In the N_{bx} phase, because the three directors defined by the surface conditions on the capillary edges and those in the bulk are perpendicular to each other, the orientational configuration is also completely frustrated. A possibility for the sample to solve the problem could be to break completely the order along two straight disclination lines located about the axes of the cylindrical capillary edges, as in the N_d phase. The tensorial order parameter would then be zero in the core of the lines, meaning an isotropic order. In fact, such a solution is energetically disfavored if compared to a configuration where the disclination lines keep a uniaxial order parameter in their core (because the order parameter is not reduced to zero then). Figure 1 sketches such a disclination line in which the long axis of the micelles (director **m**) only undergoes continuous variations. This uniaxial disclination line bears a director [**m** in Fig. 1(b)] which is parallel to it, so that the uniaxial symmetries of the director and core of the line are consistent together. A direct consequence of the uniaxiality of the line is an elastic coupling with the anisotropic bulk, making the line tilt by an angle θ from its original position (parallel to the capillary).

The tilt angle θ of the disclination line in the N_{bx} phase may be derived from the θ variations of the free energy per unit length of the capillary. Schematically, three contributions already exist in the N_d phase and extrapolate to the N_{bx} phase, giving even θ functions in the expression of the free energy. A first contribution comes from the increase of the core energy due to the elongation of the line: $F_1 \sim K(1+\frac{1}{2}\theta^2)$, where the core energy per unit length of the line is estimated to be $\sim K$ in the N_{bx} phase as the uniaxial nematics.[8] The two other contributions of N_d-type to the free energy of the line are purely elastic. They correspond to the distortions sketched in the left part of Fig. 1. In Fig. 1(a) is shown the distortion that the line produces around it. At equilibrium (and the equilibrium position of the line is located at about the center of the cylindrical edge of the capillary), this energy per unit length of the capillary may be evaluated by[8] $F_2 \sim K(\pi/4)\ln(R/a)$, where a is the diameter of the core of the disclination line. If the line is shifted from its equilibrium position, F_2 increases as the square of the shift. F_2 depends therefore essentially on the amplitude of the shift, not on θ. Consequently, the contribution of F_2 may be neglected in the free energy of the lines which make zigzags of small enough amplitude (see below). The third term of N_d type expresses the elastic distortion directly produced by the tilting of the line. In the symmetry plane of Fig. 1(b) (left part), the distortion is a simple splay of the long axis of the micelles. Its elastic energy may be estimated by

$$F_3 = K(\nabla\theta)^2(\pi R^2/2) \sim K\theta^2,$$

where $\nabla\theta$ extending along R is approximated by θ/R (R is the radius of the capillary edge).

Only one elastic term is typically biaxial. It originates from the distortion in the flat part of the capillary (right part of Fig. 1), in which the micelles change from parallel to the magnetic field to parallel to the disclination line. Such a reorientation, if considered in the symmetry plane, can be measured by the angle $\psi(z)$ of the long axis of the micelles, referred to **H**; it is located in a range[9] ξ_H from the disclination line. (ξ_H is the magnetic correlation length. It measures the apparent thickness of the line. As seen in the photographs, it is much smaller than R for our H and ΔT values.[10]) If considered in a plane farther from the symmetry plane than ξ_H, the reorientation is more complicated: It is mixed to the distortion of energy F_2 considered above. It then relaxes over a larger distance ($\sim R$) than ξ_H, giving a much smaller, i.e., negligible, contribution to the elastic energy. The biaxial elastic interaction with the line is therefore essentially concentrated around the symmetry plane within ξ_H; it can be estimated by

$$F_4 = K'(\nabla\psi)^2\xi_H^2 \sim K'(\tfrac{1}{2}\pi-\theta)^2,$$

i.e.,

$$F_4 \sim -K'\pi\theta + \text{const}.$$

The total θ variations of the free energy per unit length of the capillary are therefore

$$F_1+F_3+F_4 = \tfrac{3}{2}K\theta^2 - K'\pi\theta + \text{const}.$$

With minimization of this free energy and use of Eq.

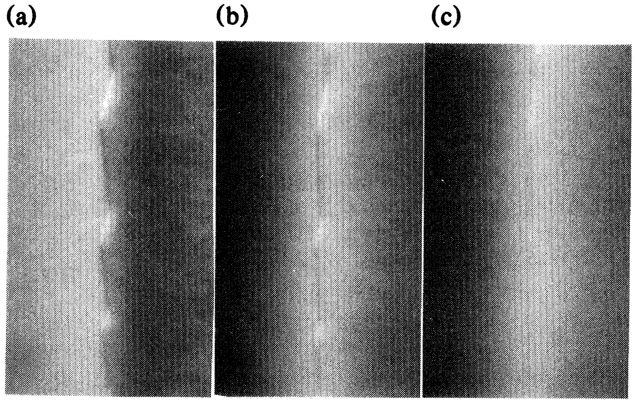

FIG. 2. Photographs of a disclination line near the edge of a flat capillary in the N_{bx} phase: (a) $T = 18.2\,°\mathrm{C}$, (b) $T = 19.4\,°\mathrm{C}$, and (c) $T = 21.5\,°\mathrm{C}$. The orientations of the capillary, of the magnetic field, and of the N_{bx} are sketched in Fig. 1(b). The sample is observed between nearly crossed polarizers in order to make lights and shadows on the zigs and zags. The asymmetric lengths of the zigs and the zags are due to a slight original tilt of the line in the N_d phase.

(1), we get

$$\theta = k\Delta T, \quad \text{with } k = \tfrac{1}{3}\alpha\pi, \tag{2}$$

which expresses that the angle of the zigzag line with the axis of the capillary should vary linearly with the temperature distance to the N_d-N_{bx} transition.

Figures 2(a)–2(c) show photomicrographs of the sample obtained when the temperature is increased from the N_{bx} to the N_d phase. They show that the disclination line breaks into zigzags at the N_d-N_{bx} transition, making an angle θ with the axis of the capillary. The angle θ does not depend on the magnetic field [this is consistent with Eq. (2)], but as soon as H is removed (or such that[10] $\xi_H > R$), the zigzags disappear, which means that the sample is not completely constrained any more.

In Fig. 3 are plotted two series of θ measurements taken in the same sample for increasing and decreasing temperatures. Except for a small hysteresis, probably due to the anchoring on the surfaces, these measurements show a linear variation of θ with temperature, which is consistent with the classical behavior of the N_d-N_{bx} transition. Moreover, they yield a determination of the coefficient $k \sim 4\times10^{-2}\,\mathrm{K}^{-1}$. This value may be compared to the estimate of the coefficient α. Taking the scalar order parameter of the N_{bx} phase $Q \sim \frac{1}{2}$ in the middle of the N_{bx} range, i.e., for $\Delta T \sim 5$ K, we find that $\alpha \sim 5\times10^{-2}\,\mathrm{K}^{-1}$, which is fairly consistent with Eq. (2) and with the model of the uniaxial disclination lines used to calculate it.

In summary, we have observed defect lines (disclinations) in the N_{bx} phase, with a core melted in the

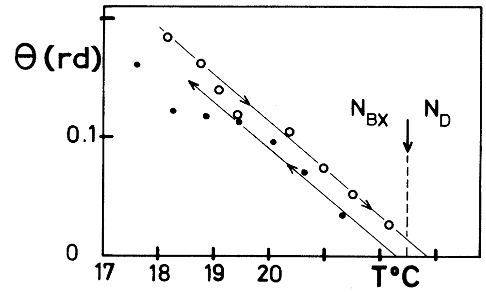

FIG. 3. Tilt angle of the disclination line vs temperature. The slight hysteresis ($\sim \pm\frac{1}{4}\,°\mathrm{C}$) observed in these measurements can be explained by anchorage of the biaxial orientation onto the glass surfaces. With a decrease in temperature far in the N_{bx} phase, new zigzags have to be created in order to reduce their amplitude. This makes the data lag a little more around $T = 17$–$19\,°\mathrm{C}$.

uniaxial nematic state. These lines bear a director parallel to them and are coupled to the anisotropic elastic field of the surrounding N_{bx} phase, which makes them tilted in a preferred direction ($\theta \neq 0$). But a consequence of this tilting is a shift of the lines from their equilibrium position. They finally find a compromise between preferred directions and preferred positions by breaking into zigzags, that is, by making point defects (melted into the isotropic state) at the peaks of the zigzags. The possibility of making such point defects on the uniaxial disclination lines in the N_{bx} phase gives them an escape from strong curvatures. They consequently adopt a stiff, solidlike shape, which at first sight is surprising in a liquid, even biaxial.

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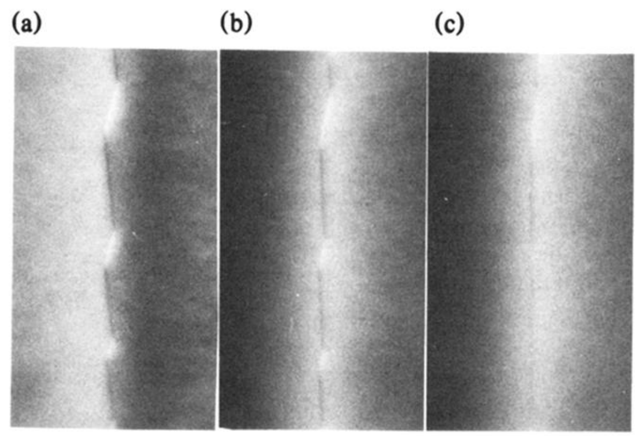

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