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Vanishing Disclination Lines in Lyotropic Biaxial Nematics

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Faint disclination lines which vanish at the transition to the uniaxial nematic phase, are observed in the biaxial nematic phase of a ternary lyotropic mixture. Unlike the *m*-disclination lines, they do not exhibit observable breaks. They bear ghostlike point defects instead. The joint observation of both kinds of disclination lines could give a real proof of biaxiality, as well as the evidence of zigzagging disclination lines with temperature dependent breaking angles. Conversely, a quick analysis of the disclination lines observed in a uniaxial nematic phase may erroneously lead one to conclude to biaxiality.

Keywords: Lyotropic liquid crystals; uniaxial and biaxial nematic phases; zigzagging disclination lines; ghostlike point defects; real proof of biaxiality

PACS numbers: 61.30.Jf, 61.70.-r, 64.70.Md

The biaxial nematic phase, N_b , has been discovered in 1980 by Yu and Saupe in lyotropic mixtures of two different amphiphilic molecules in water [1]. Since this discovery, the N_b phase has been intensively studied with regards to the generalization of the physics of the uniaxial nematics that it yields [2]. In particular, the study of the disclination lines that the biaxial phase exhibits, is really fascinating. The lines in the N_b phase make zigzags under particular circumstances [3], and they change their nature when crossing themselves [4]. In this manner, they provided some proofs to the existence of the N_b phase in thermotropic liquid crystals [4, 5], where the other experimental means failed to give clear arguments for biaxiality, as for instance, conoscopy [2]. Unfortunately, the criterium of the zigzagging disclination lines was not so absolute as it was initially believed; it cannot be

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used so readily since the zigzagging lines can also be observed in the usual uniaxial nematics [6, 2].

In this paper, we report on the experimental observation of faint disclination lines in the N_b phase of a lyotropic liquid crystal composed of rubidium laurate, 1-decanol and H_2O , in proportions 31.0, 7.0 and 62.0 wt.%, respectively. These disclination lines which appear to be thin and weakly contrasted between crossed polarizers, make typical soft curls (Photo 1) close to the discotic nematic phase, N_d , one of the two uniaxial nematic phases available in this lyotropic system. They apparently do not break as the disclination lines reported earlier in the N_b phase [3]. They have the strength $1/2$ also since they are thin and weakly contrasted, but their nature is nevertheless different. Restricting our study to the vicinity of the N_b to N_d phase transition, we observe that the faint disclination lines in the N_b phase, vanish in a continuous manner, becoming fainter and fainter when heating up the sample towards the N_d phase, until they completely disappear at the biaxial to uniaxial discotic phase transition; for this reason, we also call them vanishing disclination lines. When the transition is crossed again backwards to the N_b phase, the faint, or vanishing, disclination lines reappear, but with different shapes and in different places. They also do not perceptibly shrink and collapse with time.

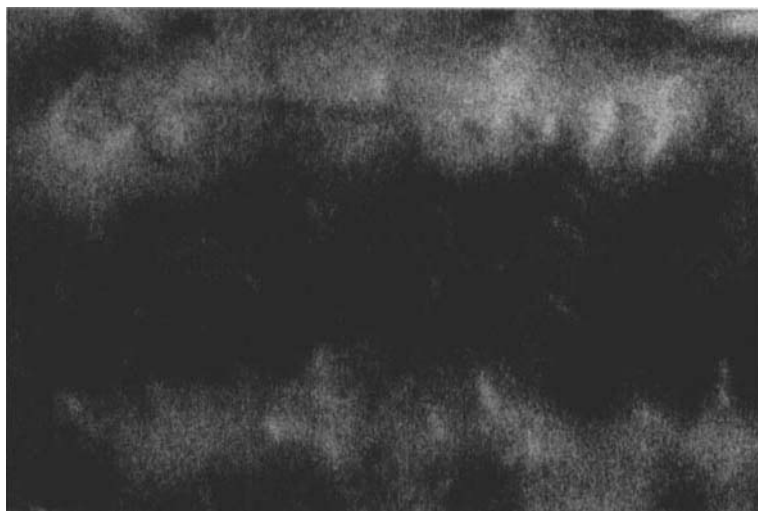


PHOTO 1 Vanishing disclination lines observed between crossed polarizers close to the uniaxial N_d phase. They generally draw entangled curls and loops. Here, the quasi uniform orientation of the director \mathbf{m} makes them undulate softly along the horizontal direction. (Courtesy of L. Liébert with whom these photographs have been taken, and who died four years ago.)

In order to understand these experimental observations, let us first briefly recall some physical features of the N_b phase. The N_b phase is characterized by three orthogonal directors, \mathbf{l} , \mathbf{m} , \mathbf{n} . The director \mathbf{n} is identical to the director of the N_d phase. \mathbf{n} therefore does not change at the discotic to biaxial phase transition. If we sketch the elementary objects of the N_b phase, which are micelles, as bricks, \mathbf{n} is the average direction of the shorter length of the bricks, \mathbf{m} is the average direction of their larger length, and \mathbf{l} is the average direction of their intermediate length. In general, a distortion in the N_b phase involves 12 elastic constants [7, 2], but close to the biaxial to discotic transition, the elasticity problem may be simplified. The directors \mathbf{m} and \mathbf{l} may be considered as biaxial directors. Their distortions involve elastic terms proportional to the square of the biaxial order parameter, i.e. to $(\delta n)^2$, where δn is the difference between the two biaxial optical indices. The biaxial to discotic transition being second order, and classical to a good approximation [8], these elastic terms continuously vanish at the transition. More exactly, the average biaxial elastic constant may be written as $K(\mathbf{m}) = \eta k_0 \Delta T$, where ΔT is the temperature distance to the transition, and η a scalar of the order of unity [3]. On the other hand, the \mathbf{n} -distortions which persist in both the biaxial and discotic phases, keep an about constant energy around the transition. As is general, the order along the defect lines is lower than in the bulk. The order parameter along the disclination lines in the N_b phase, is thus reduced to uniaxial values, exactly as in the classical uniaxial nematics the order parameter along the disclination lines vanishes, their core being isotropically disordered [9]. The disclination lines in the N_b phase thus bear a director, and we immediately see that three types of disclinations may exist depending on the nature of the director that they bear [2, 4]. In general, the director is not parallel to the lines. In this case, the uniaxial order is a little bit contradicted. This indicates that the director of the lines is preferentially anchored parallel to them, as is consistent with symmetry. In many cases, we may thus simplify the analysis on considering that the director is strictly parallel to the lines [3]. The energy per unit length of the disclination lines, also called the line tension, contains two types of terms. The first type of terms is the core energy which corresponds to the melting of the core of the lines into a uniaxial phase, and which is proportional to the average elastic constant associated to the destroyed directors, i. e. proportional to ΔT [9]. The other terms in the line tension expression, are relevant to the elastic energy of the distortion produced by the disclination lines all around themselves. In the N_b phase, these elastic terms arise from the distortion

of the three directors, and thus, close to the transition, they may be estimated according to the above approximations.

The faint disclination lines are places where, in the N_b phase, biaxiality is destroyed. They are visualized between crossed polarizers by means of their index difference with the bulk. This index difference, equal to the biaxial index difference δn , is small close to the N_b to N_d phase transition, so that the lines are weakly contrasted and often difficult to distinguish. They completely vanish at the N_b to N_d phase transition, growing up until they invade the whole sample, their core propagating the director \mathbf{n} everywhere. They may therefore be identified to the \mathbf{n} -type disclinations. As discussed above, the director \mathbf{n} that they bear is preferentially oriented along them, with an anchoring energy $1/2 k\alpha^2$ per unit length, α being the angle between \mathbf{n} and the lines, and k the anchoring constant of the director \mathbf{n} onto them. Their coupling to the bulk director \mathbf{n} vanishes at the transition. It thus directly depends on biaxiality. More precisely, the coupling of the vanishing \mathbf{n} -disclinations to the bulk director \mathbf{n} is proportional to $(\delta n)^2$, as the biaxial

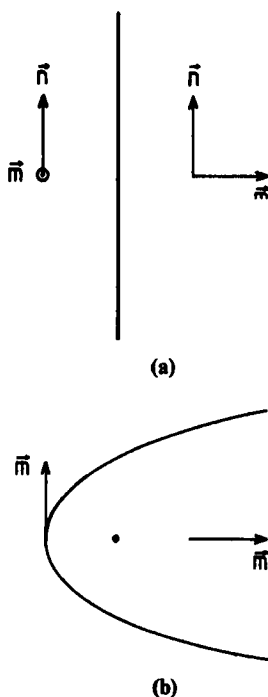


FIGURE 1 Sketch of a straight \mathbf{n} -disclination line parallel to a uniform \mathbf{n} -field. a - Cut parallel to the line. b - Cut perpendicular to the line.

elastic constants, i. e. $k = \eta k_0 \Delta T$, k_0 being a standard anchoring constant of the director \mathbf{n} onto the \mathbf{n} -disclination lines. The vanishing \mathbf{n} -disclination lines in the N_b phase have naturally different anchoring properties from the \mathbf{m} -disclinations in the N_b phase [3] or from the classical $1/2$ -disclinations in the uniaxial nematics [6] both reported to produce zigzags, and more evidently from the dislocations in the smectic O or smectic C_A phases [10]. Let us first consider a vanishing \mathbf{n} -disclination line roughly parallel to the bulk director \mathbf{n} (Fig. 1), the general direction of \mathbf{n} being perpendicular to the plates of the sample as is commonly observed in the lyotropic nematics. The disclination line arises from a π -rotation of the directors \mathbf{m} and \mathbf{l} around \mathbf{n} , i. e. around the line itself. The disclination line is thus a splay-bend disclination of strength $1/2$ for both the biaxial directors \mathbf{m} and \mathbf{l} . At the minimum of energy, the line keeps parallel to \mathbf{n} and does not need to break and to

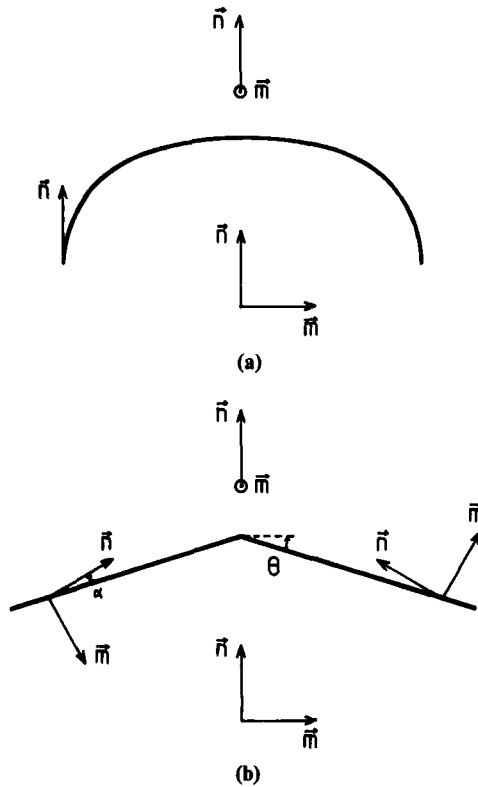


FIGURE 2 a – General view of a bent vanishing \mathbf{n} -disclination in a quasi uniform \mathbf{n} -field. b – Ghost point defect on the above \mathbf{n} -disclination line in a place where its average direction is perpendicular to \mathbf{n} . (Detail of Fig. 2a).

make zigzags. At little bit farther, while the director \mathbf{n} keeps always oriented about perpendicular to the plates, the line may rotate (Fig. 2a) and take an average direction perpendicular to \mathbf{n} , locally making the angle θ with its average direction (Fig. 2b). The energy per unit length of the line along its general direction, may then be written as:

$$W(\alpha, \theta) = \frac{1}{2} k \alpha^2 + \frac{1}{2} K(\mathbf{m}) / \cos \theta + K(\mathbf{n}) (\pm \pi/2 - \alpha - \theta)^2. \quad (1)$$

The first term in Eq. 1 is the anchoring or coupling term between the line and the field of the directors. The second term expresses the core energy as proposed by de Gennes [9]. It takes into account the increased length of the line due to its deviation angle θ . With $K(\mathbf{m}) = \eta K_0 \Delta T$, this second term is consistent with the idea that the core energy continuously vanishes at the transition together with the disclination line itself. The last term describes approximately the elastic energy of the \mathbf{n} -distortions around the line [9]. Its coefficient, the average elastic constant $K(\mathbf{n}) \sim K_0$, is roughly independent of temperature close to the N_b to N_d phase transition. Other elastic terms involving the biaxial directors should also be included in Eq. 1. They are of two kinds: Either they are proportional to ΔT , and thus they may be integrated to the second term of Eq.1, or they may be neglected in front of the third term. The minimization of Eq.1 yields:

$$\begin{cases} \alpha = \frac{K_0}{k_0} \theta \\ \theta = \frac{\pm \pi/2}{1 + \frac{K_0}{k_0}} \end{cases} \quad (2)$$

Eq. 2 shows that the vanishing \mathbf{n} -disclination lines break and make zigzags as the \mathbf{m} -disclinations, but with a breaking angle θ independent of temperature. The behavior of the vanishing \mathbf{n} -disclinations thus resembles formally that of the classical 1/2-disclinations in the uniaxial nematics, but with an unobservable breaking angle θ . We deduce that $k_0 < K_0$, and consequently, that Eq. 2 reduces to:

$$\begin{cases} \alpha \approx \pm \frac{\pi}{2} \\ \theta \approx \pm \frac{\pi k_0}{2 K_0} \end{cases} \quad (3)$$

The anchoring of the vanishing \mathbf{n} -disclination lines onto the director \mathbf{n} is therefore very weak. The vanishing \mathbf{n} -disclination lines are thus practically decoupled from the \mathbf{n} -director field. (Moreover, the \mathbf{n} -director field keeps almost uniformly oriented perpendicular to the large windows of the cell, which gives to \mathbf{n} a second reason not to perturb the \mathbf{n} -disclination lines). They apparently do not break. However, as can be shown in different manners, their shape is actually determined by the field of the biaxial directors \mathbf{m} and \mathbf{l} . For instance, they never appear if the \mathbf{m} -field, and consequently the \mathbf{l} -field, are uniformly oriented before entering into the N_b phase, as can be realized by applying a magnetic field parallel to the windows of the cell. On the opposite, they arise at the N_d to N_b phase transition when, the magnetic field being removed, the \mathbf{m} and \mathbf{l} -fields are disoriented, that is for instance, each time that the N_b phase is recrystallized, after a few minutes melting into the discotic phase. With this operation, new chaotic anchorings of \mathbf{m} and \mathbf{l} are defined at the glass surfaces. They determine new continuous \mathbf{m} and \mathbf{l} fields in the bulk of the sample, which produce in turn, new vanishing \mathbf{n} -disclination lines with different shapes.

Usually, the disclination lines shrink as soon as they are produced, and eventually collapse after a while. In this manner, they reduce their length and consequently, their core and elastic energies. Surprisingly here, the vanishing \mathbf{n} -disclination lines seem motionless after they have been created. In fact, they do move very slowly as observations at long intervals of time show. Such a slow behavior is a characteristic feature of these lines, though it may partly come from anchorings onto the glass surfaces. The time life of free disclinations is inversely proportional to the ratio of their tension to the bulk viscosity. The tension of the vanishing \mathbf{n} -disclination lines is approximately $\sim K(\mathbf{m})$ when they are oriented parallel to \mathbf{n} (Fig. 1), or $\sim k$ when they are perpendicular to \mathbf{n} (because then, the anchoring and bulk elasticities are associated in series as can be shown on minimizing Eq. 1). In both cases, the line tension is small and proportional to ΔT . On the other hand, the viscosities of the lyotropic nematics are known to be larger by about one order of magnitude than the typical viscosities of the thermotropic nematics [11]. So, the life time of the vanishing \mathbf{n} -disclination lines should be larger by two or three orders of magnitude than the life time of the classical disclinations in the thermotropic nematics. They therefore cannot be observed to move noticeably in the typical experimental duration of one hour.

In summary, the faint or vanishing disclination lines which are observed here in the N_b phase, are identified to be $1/2$ -disclinations of the \mathbf{n} -type. They formally break as all the $1/2$ -disclination lines do in the N_b phase, because of a competition between the elastic energy involved all around, the

core and the anchoring energies. Close to the N_b to N_d phase transition, the breaking angle of the **n**-disclination lines keeps so small that it can even not be noticed nor measured unlike on the **m**-disclination lines [3]. The breaks on the vanishing **n**-disclination lines nevertheless do exist. They are like ghost point defects. The lines are more and more easily observable the farther the sample is from the N_b to N_d phase transition, with a visibility proportional to $(\delta n)^2$, i. e. to ΔT .

Only the two **m** and **n** types of disclinations have been observed up to now in the N_b lyotropic phases. Basically, they should play reversed roles close to the biaxial to calamitic (N_c) nematic phase transition. By symmetry, the **m**-disclinations should there become vanishing lines with ghost point defects, while instead the **n**-disclinations should be well contrasted and should present finite breaking angles, though reaching linearly zero at the transition. About in the middle of the N_b phase, both the **m** and **n** disclination lines should resemble very much each other, with almost similar contrasts and breaking angles, and therefore clearly visible point defects (Photo 2). The **l**-disclination lines which constitute the third family of 1/2-disclinations in the biaxial nematic phase should have a uniaxial core of director **l**. They would therefore transform into 1/2-disclinations with an isotropic core, in both the uniaxial N_c and N_d nematic phases. Breaking the



PHOTO 2 Same sample as in Photo 1, but farther from the uniaxial N_d phase. After waiting relaxation for several hours, the **n**-disclination lines form well-contrasted zigzags as the **m**-disclination lines.

largest index difference, the **l**-disclination lines should always be well contrasted, and exhibit observable zigzag angles. Their zigzag angles should continuously vanish at the two uniaxial phase transitions, as the **m**-disclination lines do at the N_b to N_a phase transition. Though these features are clear cut and should be easily observable, we failed in finding any of these **l**-disclination lines. The explanation is probably that they are rare because, breaking the largest index difference, they also have the largest energy among the three possible types of biaxial disclination lines of strength $1/2$. They would therefore probably escape this difficulty by decomposing themselves immediately into disclination lines of lower energetic cost, of the **m** and **n** types [12, 13, 4]. However, such a decomposition could not be systematic. According to De' Neve *et al.*, the three types of disclination lines and their crossings can be observed in biaxial thermotropic polymers [4]. Indeed, this observation is questionable, because the reported disclination lines are not observed to bear any point defect and to break. It may be argued then that the biaxiality of the sample is weak. In this case, as discussed above, we should observe that one of the disclination lines is much fainter than the others. This line should then be of the **m** or **n**-type, and bear ghost point defects. The two other lines conversely should be easily observable with small, but measurable breaking angles. This is obviously not the case. We therefore have some doubts on the real biaxiality of the thermotropic polymer used for the experiment. The third line observed by De' Neve *et al.* [4] could then be due to light refraction across strongly distorted domains, all the more than this line exhibits typical variations of width and contrast which could arise from variations in the distortion. Similar mirages are commonly observed in nematic liquid crystals submitted to a constant voltage, small vortices sharply folding the director field. These mirages may easily be confused with disclination lines because they often take looplike shapes, if one does not pay enough attention to their anomalous variations of width and contrast.

As discussed earlier [2], biaxiality is interesting, but difficult to proof experimentally. The measurements integrated over a large volume of the sample (conoscopy, X-ray diffraction,...) are frequently biased with slight distortions, leading to apparent biaxiality. The local arguments, as deduced from a study of the defects, have also to be used with care. The simultaneous existence of two types of disclination lines, the vanishing and non-vanishing lines, could yield a good proof for the biaxiality of a sample, together with the observation of zigzagging disclination lines with temperature dependent breaking angles, as already suggested (see for instance, Ref. [2]).

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