Lyotropic nematic mesophases: peculiarities of singularities and inversion walls in specific and non-specific extures

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Peculiarities of various defects and disclinations in specific and non-specific textures of the optically uniaxial and biaxial lyotropic nematic mesophases were investigated. Sodium dodecylsulphate (SDS) [sodium lauryl sulphate (SLS)] /water/1-decanol lyotropic liquid crystalline system was used. The specific and magnetically induced textures were studied. The optical signs and the disclination of strength of various singularities were determined. The optical mappings and schematic configurations around the disclination lines, inversion walls and singular points, taking place in textures of lyotropic nematic mesophases, are presented in this work.

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1. Introduction

Liquid crystalline systems, based on amphiphiles, exhibit various types of physically anisotropic liquid crystalline mesophases. Lyotropic nematic mesophases have definite packing of structural units and are characterized by strong spatial symmetry. These mesophases are formed by mixtures of amphiphile and a solvent as water (or as water and aliphatic alcohol) under definite concentration and temperature conditions. Structural units in lyotropic nematic mesophases are the anisometric micelles of definite sizes [1-5]. Lyotropic liquid crystalline systems form two optically uniaxial nematic-calamitic N_C and nematic-discotic N_D mesophases and one optically biaxial N_{bx} mesophase. Structural units in N_C and N_D mesophases are the rod-like micelles with definite lengths and the disc-like micelles with definite diameter respectively. N_{bx} mesophase appears to be an intermediate mesophase appearing at the border between N_C and N_D mesophases [6-9]. This mesophase takes place in sufficiently small concentration and temperature intervals [10,11].

Unlike thermotropic nematic mesophase, lyotropic nematic mesophases exhibit large variety of specific and non-specific textures. In these mesophases various types of defects as the disclinations, inversion walls, singular points, point-like and linear defects can be arisen because of high sensitivity of lyotropic nematic mesophases to external effects, boundary conditions, additional lyotropic components etc. [12-15]. Additionally, lyotropic nematic mesophases are richer than thermotropic ones due to the freedom of choice of the parameters to obtain various defective formations. Such defective formations often arise spontaneously in ordered and non-ordered textures, but they can also be induced in controlled ways.

The physics of the defects in liquid crystals is important topic for the physics and application of the condensed matter. It is connected with important role of defects for various processes, namely liquid crystals identification and classification, for obtaining aligned textures, studying on phase transitions, elastic and inelastic deformations etc. [16-20]. Therefore, the aim of this work is investigation of character and peculiarities of various defects in lyotropic nematic mesophases for samples under external applied magnetic field.

In this work, the director distribution near optical positive and optical negative disclinations was studied. Optical mappings and schematic representations around defects and disclinations were presented for N_C, N_D and N_{bx} mesophases.

2. Experimental

In this work, lyotropic nematic mesophases of the ternary sodium dodecylsulphate (SDS) [sodium lauryl sulphate (SLS)])/water (H_2O)/1-decanol (DeOH) lyotropic system were investigated. SDS was purchased from Merck and was purified by recrystallization from ether/ethanol. This amphiphile is characterized by the melting point and the critical micelle concentration value. This value was determined as 0.00783 moles per liter. DeOH was also purchased from Merck, has high degree of purity and was used without further purification. Water, which was used as general solvent, was triple distilled and deionized. In Table 1, the compositions of N_C, N_{bx} and N_D mesophases are presented. These compositions were chosen in accordance with phase diagrams, which are given in [21].

The microslide samples as the sandwich-cells were used in this work. The thickness of liquid crystalline layer in the microslides was 120.0 ± 1.0 μm. The microslides were hermetically closed at once after filled with liquid crystalline system.

Table 1. Compositions of lyotropic nematic mesophases.

Type of mesophase	Compositions of lyotropic components, mass%		
	SDS (SLS)	H_2O	DeOH
N_C	28.00	67.50	4.50
N_C	28.10	67.08	4.82
N_{bx}	25.97	68.98	5.05
N_{bx}	26.11	68.90	4.99
N_D	23.95	70.90	5.15
N_D	24.21	70.17	5.62

The investigations of the morphologic and crystallo-optical properties of lyotropic liquid crystalline mesophases were carried out by the polarizing optical microscopy method (POM). Our set-up consists of a trinoculer polarizing microscope, microphotographic system, optical filters, and λ-plates from Olympic Optical Co. and also an original heater-thermostat, differential Cu-Co thermocouples, a multimeter, a power supply, and a digital temperature controller. The experiments for obtaining magnetically induced and aligned textures of liquid crystalline mesophases were made by permanent magnet. Magnetic field was applied orthogonal to the reference surfaces of the microslides and accordingly to a liquid crystalline layer. Fields of 9.7 kG were available. The samples were kept at stable temperature as 297 K while the magnetic field was applied. The optical mapping (OM) method was used to determine of the optical signs and the disclinations of strength for various singularities, taking place in textures of lyotropic nematic mesophases. The OM method was given in [22,23] and was used in [24-27] for detailed investigations of peculiarities of liquid crystalline textures.

3. Results and discussion

In this work, peculiarities of defects in N_C, N_D and N_{bx} mesophases were investigated. N_C and N_D mesophases are optically uniaxial. Structural units of these mesophases are micelles which have the symmetry of the ellipsoids of rotation. Micelles of N_C mesophase are the rod-like supramolecular aggregated as spherocylinders (Fig. 1a); micelles of N_D mesophase are the disk-like supramolecular aggregated as spherodisks (Fig. 1b). In the rod-like micelle, the ellipsoid axis of rotation is the long axis of micelle; therefore the director $\vec{n}$ ($\vec{n} = -\vec{n}$) of N_C mesophase is directed parallel to the long axis of micelle. In the disk-like micelle, the ellipsoid axis of rotation is the short axis of micelle; therefore the director $\vec{n}$ ($\vec{n} = -\vec{n}$) of N_D mesophase is directed perpendicularly to the plane of micelle. Structural units of N_{bx} mesophase have the symmetry of the parallelepiped with the point group as D_2 (Fig. 1c). As is noted in [11,28-30], this mesophase is characterized by three orthogonal directors as the $\vec{l}$, $\vec{m}$ and $\vec{n}$, which are fixed in the micelle. These order parameters correspond to the orientational alignment of both the short and long axes of micelles. The $\vec{l}$, $\vec{m}$ and $\vec{n}$ are dependent. Therefore it is enough to fix two of them and the third one can be determined.

Investigations showed that the external magnetic field, which is applied in the orthogonal direction to the reference surfaces of microslides, leads to an appearance of various defective formations in textures of N_C, N_D and N_{bx} mesophases. Most of such defective formations were observed only in lyotropic nematic mesophases and were not observed in thermotropic nematic mesophase.

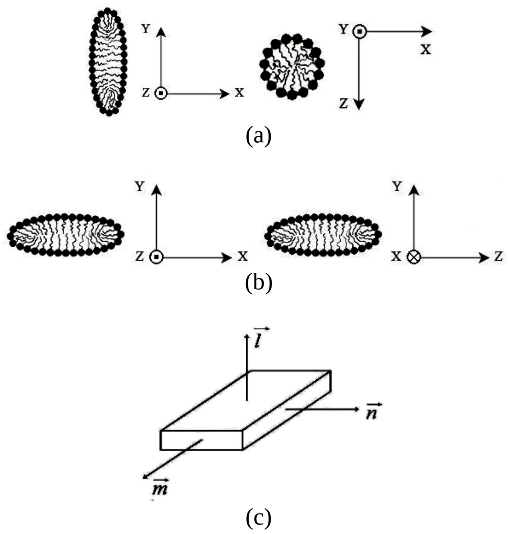


Fig. 1. Schematic representations of the spherocylinders (rod-like micelles) (a), spherodisks (disk-like micelles) (b) and structural units of N_{bx} mesophase (c).

N_C mesophase

When the external magnetic field was applied to the microslide with the schlieren texture of N_C mesophase, after persistent textural transformation, magnetically induced schlieren texture was observed (Fig. 2). This texture is characterized by the integer defects, which are randomly created. As it is seen in this figure, the texture consists of the system of brushes and the singular points. The brushes arise in regions where the optical axis of liquid crystalline layer is directed parallel or perpendicular to a polarization plane of the light entering the sample from the polarizer. The polarization of incident light is not changed by liquid crystalline layer and therefore, this light is not extinguished by the analyzer. In this case, when the polarizer and analyzer in cross position were rotated simultaneously in the same direction for stationary position of microslide on the microscope stage, the brushes were continuously moved. This fact indicates continuous change of the optical axis direction in the liquid crystalline layer of N_C mesophase. Similar type of the inversion walls was also observed for thermotropic nematic mesophase in [22,31-33]. However, unlike thermotropic nematic mesophase, textures with inversion walls were observed in lyotropic nematic mesophases only under the external magnetic field (Fig. 2).

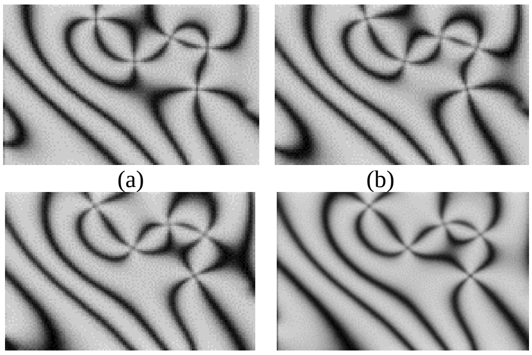


Fig. 2. Optical mapping of magnetically induced schlieren texture of N_C mesophase. Crossed polarizers; magnification x100; temperature 298.0 K.

As it is also seen in Fig. 2, when cross polarizers were rotated for stationary position of microslide, the singular points remained constant, while the dark brushes were moving in the plane of texture. If the brushes were rotated in the same direction as the rotation of the crossed polarizers, the optical sign of the singular point was positive; if the brushes were rotated in the opposite direction to the rotation of the crossed polarizers, the optical sign of the singular point was negative. Optical investigations showed that these singular points were the linear disclinations, which were placed perpendicularly to the reference surfaces of the microslides. The strength of disclination for singular points can be defined as $|S| = \frac{1}{4}N$, where N is the number of the brushes for the corresponding singular point [22,25]. As it is seen in Fig. 2, the singular points with the disclination of strength as both $S = +1$ and $S = -1$ were occurred.

In N_C mesophase, specific disclinations of integer strength were sometimes observed (Fig. 3). These individual defects were obtained as results of simultaneous effect of thermic and magnetic fields and were arisen on the planar alignment conditions. As it is seen in Fig. 3, these defects were the concentric line defects as closed curves. These defects were formed under planar alignment conditions. The diameter of the defects was microscopically determined as ~ 150 μm. The energy of an individual defect is proportional to S^2 [31,34]. In [35], it was shown that the concentric line defects with the disclination of strength as $S = 1$ were stable only if $K_{11} > K_{33}$ and $K_{22} > 2K_{33}$ (K_{11}, K_{22} and K_{33} are the elastic constants for the splay, bend and twist deformations, respectively). As it is known, in nematic mesophase, $K_{22} < K_{11} < K_{33}$ takes place [36-39]. Therefore it is expected that such defects rarely occurs in nematic mesophase because of small value of K_{22} elastic constant. We would like to note that the concentric line defects were observed for nematic mesophase of usual thermotropic liquid crystals in [22] and for polymer nematic mesophase in [31] only under homeotropic alignment conditions. In defect, presented in Fig. 3, the singular points were absent and this defect could be schematically presented as it is shown in Fig. 4.

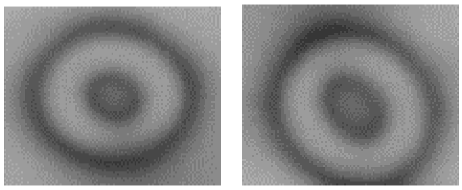


Fig. 3. Concentric line defects in N_C mesophase. Crossed polarizers.

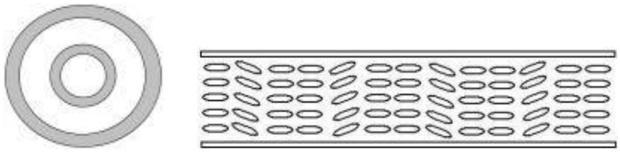


Fig. 4. Sketch of the integer defect structure. a- the top view; b- the side view.

N_D mesophase

After some heating-cooling processes and magnetic field application, texture, presented in Fig. 5, was obtained for N_D mesophase. This texture consists of particular loops on the planar background. We would like to note that planar texture of N_D mesophase was different from that of N_C mesophase. Namely, if the disc-like micelles of N_D mesophase were oriented perpendicularly to the reference surfaces, the optical axis of this mesophase will be parallel to the surfaces; if the rod-like micelles of N_C mesophase were oriented parallel to the reference surfaces, the optical axis of this mesophase will be also parallel to the surfaces.

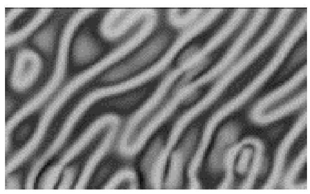


Fig. 5. Loop like defects in N_D mesophase. Crossed polarizers.

Optical investigations showed that the loops in Fig. 5 were the inversion walls. These walls pierce a liquid crystalline layer and are volume disclinations. Polarization optical study showed that the director $\vec{n}$ of the loops was approximately oriented under $\frac{\pi}{4}$ angle to the layer. Such geometry of the walls under effect of magnetic field, as it is indicated in [40], is comparable to that of the Néel walls in ferromagnetics. Such type of inversion walls was also observed for N_D mesophase in [6,41] and for N_C mesophase in [30,42,43].

Interesting and rare type of the defective formation was observed for N_D mesophase. In Fig. 6 this formation with $\sum_i S_i \neq 0$ is presented. Schematic representation of the director field for this formation is presented in Fig. 7. As it is seen in Fig. 6, the formation consists of five singular point and five pair of inversion walls on the planar background. Right singular point has $|S| = ½$, left singular point and singular points in the center of the formation have $|S| = 1$. This formation was stabilized by mutual attraction and repulsion of the singular points with different optical signs. Optical investigations showed that this defect was the volume disclination.

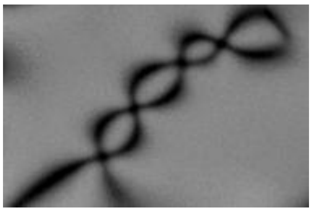


Fig. 6. Defective formation with $\sum_i S_i \neq 0$ in texture of N_D mesophase. Crossed polarizers; magnification x100; temperature 298.0 K.

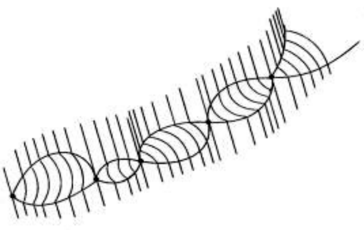


Fig. 7. Sketch of defective formations presented in Fig. 6.

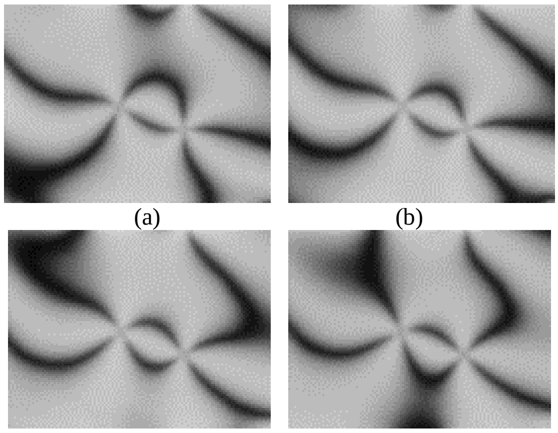
The formation, presented in Fig. 6, was sufficiently stable and only after 7.0 - 7.5 hours, it was transformed to other formation with $|S| = ½$ and low energy of deformation. We would like to note that, in [44] using the general expression for the energy density of elastic distortions of the director orientation,

$$F = \frac{1}{2}\left\{K_{11}(div\vec{n})^2 + K_{22}(\vec{n}\cdot rot\vec{n})^2 + K_{33}\left[\vec{n}xrot\vec{n}\right]^2\right\} \quad (1)$$

the steady disclinations were investigated ($\vec{n}$ is the director of liquid crystalline mesophase). And in above mentioned work, it was shown that volume disclinations were stable only if $2K_{22} < (K_{11} + K_{33})$. Additionally, for the case of $K_{11} \approx K_{22} \approx K_{33}$, the single disclinations with $|S| = ½$ and minimal energy were stable [27].

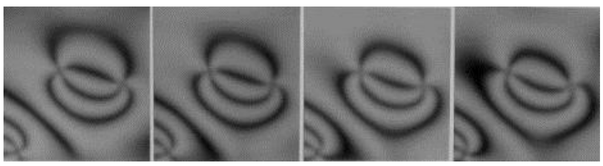
N_{bx} mesophase

Interesting types of the singular points with $S = \pm 1$ were observed in magnetically induced textures of biaxial nematic N_{bx} mesophase (Figs. 8 and 9). Figs. 8 and 9 present the singularities on the planar alignment conditions. As it is seen in Fig. 8, two brushes were closed by singular points and two other brushes were open. As it is also seen in this figure, the left singular point was optically positive whereas the right singular point was optically negative. Additionally, by even clockwise rotation of the polarizers, un-even rotation of the brushes was observed. Such rotation of the brushes took place for the case of $K_{11} \neq K_{22} \neq K_{33}$. In Fig. 10 the scheme of the director field for the singularities in Fig. 8, is presented. We would like to note that similar type of singularities was observed in [32] for thermotropic uniaxial nematic mesophase. Appearance of such defects in texture of biaxial N_{bx} mesophase is interesting peculiarity of magnetically induced textures in lyotropic nematic mesophases.



(c) (d)

Fig. 8. Optical mapping of singular points and open inversion walls in texture of N_{bx} mesophase. Crossed polarizers; magnification x100; temperature 296.8 K. Polarizers clockwise by 15^0 on going as a → b → c → d.



(a) (b) (c) (d)

Fig. 9. Optical mapping of singular points and closed inversion walls in texture of N_{bx} mesophase. Crossed polarizers; magnification x100; temperature 296.8 K. Polarizers clockwise by 15^0 on going as a → b → c → d.

In Fig. 9, the singular points, which unite the closed inversion walls, is presented. As it is seen in this figure, the left singular point was optically positive whereas the right singular point was optically negative. Optical investigations showed that the inversion walls in presented defective formation were placed perpendicular to liquid crystalline layer. As it is also seen in Fig. 9, by clockwise rotation of the crossed polarizers, the central inversion walls remained stationary. These central inversion walls stabilize the defective formation. Similar type of defective formation had been also observed for thermotropic nematic mesophase in [33,45], but on the planar background. In Fig. 11, schematic representation of this defective formation is given.

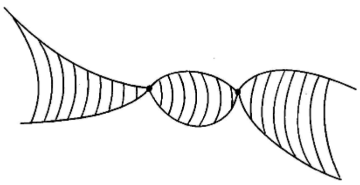


Fig. 10. Sketch of defective formations presented in Fig. 8.

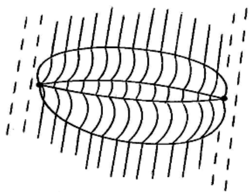


Fig. 11. Sketch of defective formations presented in Fig. 9.

In [30], it was shown that when one of the directors, $\vec{l}$ for example (Fig. 1c), was uniformly fixed in the entire sandwich-cell, the order parameter of the planar oriented N_{bx} mesophase reduced to the form of an order to the uniaxial nematic mesophase. In this case, under such configurations the physical properties of N_{bx} mesophase would present the properties similar to the uniaxial nematic mesophase. Such configuration was carried out for N_{bx} mesophase, investigated in this work.

The free energy density of liquid crystal near the disclination lines in spherical volume with radius R for single constant approximation can be determined by

$$\Phi = \Phi_1 + \Phi_2 + \Phi_{12} \tag{2}$$

Here Φ_1 and Φ_2 are the energy of separate disclinations per unit length between singularities; Φ_{12} is the energy of interaction between these disclinations. The energy of separate disclinations is proportional to S^2. Therefore, it is clear that disclination with larger disclination of strength has larger energy and is non-stable. Additionally, singular points with the strength of disclination as $|S| > 1$ are rarely observed. Such type of the singular points was observed in liquid crystalline polymers with high viscosity [46,47], in the hydrogen bond-induced nematic phases of p-alkoxycinnamic acids [48] and in nematic and smectic mesophases of 2,5-bis(4-alkoxybenzoyloxy)-p-benzoquinones [49].

Φ_{12} is determined by [36,37,50]

$$\Phi_{12} = 2\pi K S_1 S_2 \ln \frac{2R}{r} \tag{3}$$

Here K is the elastic constant for the single constant approximation ($K = K_{11} \approx K_{22} \approx K_{33}$); S_1 and S_2 are the disclinations of strength for connected singularities; r is the distance between singularities. In accordance with the theoretical approach, presented in [36,37,48], the interaction force between singularities can be determined by

$$F_{12} = -\frac{\partial \Phi_{12}}{\partial r} = \frac{2\pi K S_1 S_2}{r} \tag{4}$$

If the disclinations have the same sign, i.e. $S_1 \cdot S_2 > 0$, they will push each other; if the disclinations have the opposite sign, i.e. $S_1 \cdot S_2 < 0$, they will attract each other. In the case of singularities with the same disclination of strength the interaction force can be determined by

$$F_{12} = \frac{2\pi K S^2}{r} \tag{5}$$

Taking into consideration the elastic constant of lyotropic nematic mesophases as $K \sim 10^{-11}$ N [41,51,52] and distance between singularities, presented in Figs. 8 and 9, as ~ 23 μm, the mean interaction force was determined as $F_{12} \sim 2.730 \cdot 10^{-6}$ N·m^{-1}.

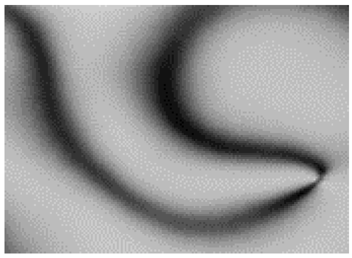


Fig. 12. Magnetically induced textures of N_{bx} mesophase after 24.0 hours. Crossed polarizers; magnification x150; temperature 296.8 K.

Additionally, investigations showed an interesting peculiarity of magnetically induced textures in N_{bx} mesophase. As process of unremitting texture transformations and after 24.0 hours the planar oriented texture and rare singular points with the disclination of strength as $S = \pm½$ was observed in N_{bx} mesophase (Fig. 12). In this texture, the singular points with the disclination of strength as S = ±1 were absent. We would like to note that in textures of N_C and N_D mesophases after 24.0 hours under magnetic field the singular points with the disclination of strength as $S = \pm½$ and $S = \pm1$ were observed. Because of such peculiarities of the magnetically induced textures, N_{bx} mesophase can be easily identified by behaviour of the typical textures in magnetic field.

4. Conclusions

External magnetic field is the cause of arising magnetically induced and oriented textures in lyotropic nematic N_C, N_D and N_{bx} mesophases in SDS/H_2O/DeOH lyotropic system. All magnetically induced textures of these mesophases show various types of defective formations and strong birefringence. Most of such defective formations were observed only in lyotropic nematic mesophases and were not observed in thermotropic nematic mesophase. Therefore, defect types and their differences in magnetically induced textures allow identifying the type of lyotropic nematic mesophase.

Orientation of diamagnetic configurations in lyotropic nematic mesophases is connected with the sign of the diamagnetic anisotropy. For the case of $\Delta\chi > 0$, the director of mesophase is oriented parallel to an external magnetic field; for the case of $\Delta\chi < 0$, the director of mesophase is oriented perpendicularly to an external magnetic field. Our investigations showed that N_C and N_D mesophases exhibit magnetically induced textures under planar conditions. A comparison between the peculiarities of the magnetically induced textures and character of the planar regions for both N_C and N_D mesophases, indicates that in SLS/H_2O/DeOH system N_C mesophase has $\Delta\chi > 0$ (N_C^+ mesophase) , while N_D mesophase has $\Delta\chi < 0$ (N_D^- mesophase).

We would like to note that lyotropic nematic mesophases are richer than thermotropic ones due to the freedom of choice of parameters to obtain the inversion walls and singularities. That is connected with differences in packing character of structural units in lyotropic nematic mesophases (micelles) and thermotropic nematic mesophase (molecules). As it is known, the ordering in lyotropic nematic mesophases is characterized by the order parameter S_{mc}, which describes the ordering of micelles relative to the director $\vec{n}$ of mesophase and by the order parameter S_{ml}, which describes the ordering of molecules relative to the chosen axis of micelle [6,53-56]. These parameters are connected with the order parameter S, describing the order of molecules relative to the director $\vec{n}$ as

$$S = S_{mc} S_{ml} \qquad (6)$$

Parameter S in lyotropic nematic mesophases is analogous to those in thermotropic nematic mesophase, but S_{mc} and S_{ml} parameters are absent in thermotropic nematic mesophase.

It was found that disclinations in lyotropic nematic mesophases could last longer than that in thermotropic nematic mesophase. It is obviously connected with the fact that viscosity of lyotropic nematic mesophases is higher than that of thermotropic nematic mesophases. Additionally, the mobility of N_C, N_D and N_{bx} mesophases with supermolecular structures (micelles) is lower than that in thermotropic nematic mesophase with molecular structure. These differences lead to stability and long life time of defective formations in N_C, N_D and N_{bx} mesophases.

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