Anchoring transitions in nematic-discotic lyotropic mesophase under magnetic field

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Magneto-morphologic and orientational properties of the lyotropic nematic-discotic N_D mesophase have been studied by the polarizing optical microscopy technique. Texture transformations from the schlieren texture to the homeotropic alignment and from the homeotropic alignment to the planar alignment have been investigated. Occurrence of these alignments has been studied by the optical and conoscopic/orthoscopic observations. The changes of the angle between the director and a normal to the surface of the sandwich-cell, and the optical birefringence under magnetic field vs. time have been determined.

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1. Introduction

Lyotropic liquid crystals are two and/or multicomponent systems, which are formed by mixtures of amphiphile, solvent (usually water) and other components (aliphatic alcohol, cosurfactant, inorganic salts, optical active material etc.). These systems under convenient concentration and temperature conditions display various types of the anisotropic mesophases. The structural units of these mesophases are the anisometric micelles. The rod-like micelles of quasi-indefinite length take place in the single hexagonal, two-dimensional hexagonal, rectangular and square mesophases; the plate-like micelles of quasi-indefinite diameter take place in the lamellar mesophases [1-4]. Other type of the anisometric micelles takes place in the lyotropic nematic mesophases. The nematic-calamitic N_C mesophase has the rod-like micelles of definite length; the nematic-discotic mesophase N_D has the disc-like micelles of definite diameter, and the biaxial nematic N_{bx} mesophase has micelles of definite sizes [5-9].

Lyotropic nematic mesophases are sufficiently sensitive to the external effects. These mesophases change the physical properties under influence of the magnetic field, temperatures, flows, surfaces, deformations, boundary conditions etc. [10-13]. Besides, in spite of the thermotropic nematic mesophases, in the lyotropic nematic mesophases the physical properties are connected not only with state of the anisotropic micelles, but also with their shapes and displacement of the amphiphile molecules in the micelles. Such peculiarities of the lyotropic nematic mesophases make these mesophases very interesting objects of the physical investigations and important materials for the technical and technological applications.

In this work the dynamics of the magneto-morphologic properties and processes of orientation and disorientation of the disc-like micelles in the nematic-discotic mesophase have been investigated. Peculiarities of the schlieren textures and the conoscopic images have been studies. Changes of the angle between the director $\vec{n}$ and a normal to the reference surface of the sandwich-cell vs. time during process of the homeotropic alignment, and also the dependence of the optical birefringence Δn vs. time during process of the planar alignment have been determined.

2. Experimental

2.1. Materials and samples

As the object of our investigations, the N_D mesophase, which is formed in the ternary n-hexadecyl-n,n,n-trimethylammonium bromide (HTAB) + water (H_2O) + decanol (DeOH) lyotropic system, has been used. HTAB was purchased from Sigma and DeOH was purchased from Merck. These materials had a high degree of purity and were used without further purification. H_2O, which was used as general solvent, was triple distilled and de-ionized.

For these investigations the sample compositions were as 22.85wt%HTAB + 73.86wt%H_2O + 3.29wt%DeOH and 23.00wt%HTAB + 72.75wt%H_2O + 4.25wt%DeOH. A sandwich-cell type glass plane capillary was used to prepare the samples under investigations. The thickness of liquid crystalline layer in the sandwich-cells was as 120.0 ± 1,0 μm. The sandwich-cells were hermetically closed at once after filled with the liquid crystalline system.

2.2. Methods

The polarizing optical microscopy method (POM) was used for the thermo-morphologic and magneto-morphologic investigations. The set-up consisted of trinoculer polarizing microscope with the conoscopic/orthoscopic observations, microphotographic system, λ plates (λ = 137 μm and λ = 530 μm) and optical filters from Olympus Optical Co., heater-thermostat with digital temperature control system, differential Cu-Co thermocouples, power supply and multimeters. The optical mapping method (OM) was employed to investigate the peculiarities of the typical and magnetically induced textures of the lyotropic nematic mesophase, and to determinate of the optical signs and the disclinations of strength for various singularities that take place in texture of the mesophase under investigations. The OM is presented in [14,15] and was widely used in [16-19] for detailed investigations of the peculiarities of liquid crystalline textures.

A permanent magnet was used for the experiments to obtain magnetically induced and aligned textures of the mesophase under investigations. The field of H = 0.87 T was available. Magnetic field was applied perpendicularly and parallel to the reference surfaces of the sandwich-cells. During the magnetic field influence, the samples were kept at a stable temperature of 300.3 K.

Changes of the optical birefringence vs. the time for the samples under influence of the external magnetic field have been studied by the compensation method. For these studies the Berek compensator from Olympus Optical Co. has been used. The methods of the crystallo-physics and crystallo-optics also have been used for the optical investigations.

3. Results and discussion

Structural units of the N_D mesophase are the disc-like micelles with definite diameter. Thickness of these micelles is approximately equal to twice length of the amphiphile molecules [6,7,20-24]. The disc-like micelles in the N_D mesophase can be considered as the spherodiscs. Schematic representation of such micelles is given in Fig. 1. The director $\vec{n}$ of the N_D mesophase is perpendicular to the plane of the disc-like micelles. In this mesophase position of the director $\vec{n}$ is parallel to the amphiphile molecules. The arrangement of the director $\vec{n}$ and amphiphile molecules is cause of the positive optical birefringence $(\Delta n = n_{II} - n_{\perp} = n_e - n_o > 0)$, i.e. the positive optical anisotropy. Note that structural units of the lyotropic nematic-calamitic mesophase N_C is the rod-like micelles. In this mesophase the director $\vec{n}$ is in direction of the long axis of the rod-like micelle and perpendicularly to the amphiphile molecules. The arrangement of the director $\vec{n}$ and amphiphile molecules is cause of the negative optical birefringence $(\Delta n = n_{II} - n_{\perp} = n_e - n_o < 0)$, i.e. the negative optical anisotropy.

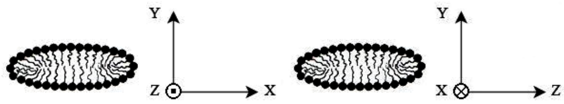
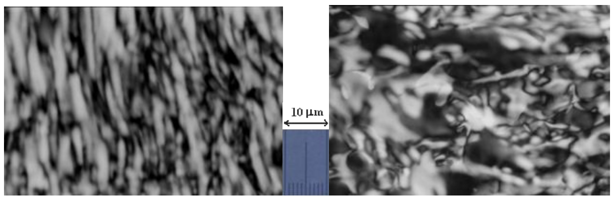


Fig. 1. Schematic representation of the disc-like micelles in the N_D mesophase.

The N_D mesophase in HTAB/H_2O/DeON lyotropic liquid crystalline system displays the schlieren textures (Fig. 2). These textures are classical for the N_D mesophase. These textures consist of several thread-like formations, singular points, and small uniform regions. Optical investigations showed that the N_D mesophase is optically uniaxial. The schlieren textures of the N_D mesophase are thermal sensitive and the mesomorphic properties of these textures change by a decrease of temperature. As is seen in Fig. 2, the schlieren texture at higher temperature (Fig. 2a) is unlike the schlieren texture at lower temperature (Fig. 2b). The texture, presented in Fig. 2b also consists of the loop-like formations. These formations are the inversion walls and boundaries between uniform aligned regions. Similar textures for the N_D mesophase have been also observed in [25-31].



a b

Fig. 2. Schlieren texture of N_D mesophase at temperature 300.3 K (a) and 307.0 K (b); Crossed polarizers; Magnification x100.

The investigations showed that the external magnetic field has sufficiently influence on the morphologic and orientational properties of the mesophase under investigations. By application of the magnetic field perpendicularly to the reference surfaces of the sandwich-cell, fluent texture transformations have been observed. After 25.0 min under magnetic field, the tilted texture of the N_D mesophase has been observed. Than, after 45.0 min, texture which is characterized by the specific conoscopic image, has been observed (Fig. 3a). As is known, such conoscopic image characterizes the tilted oriented texture [32-34]. Schematic representation of such conoscopic image in the spherical coordinates, and position of the angle between the normal and the optical axis of the tilted oriented liquid crystalline mesophase are presented in Fig. 4. The optical axis of the mesophase is placed in the XOZ plane; normal to the reference surfaces of the sandwich-cell is in the OZ direction. The θ angle is connected with the α, β and γ angles as $\cos\beta = \cos\theta \cdot \cos\alpha + \sin\theta \cdot \sin\alpha \cdot \cos\gamma$ [32].

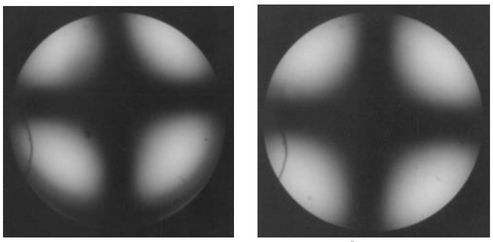


Fig. 3. Conoscopic images of orientational dynamics in the N_D mesophase; tilted oriented (a) and homeotropic oriented (b) textures.

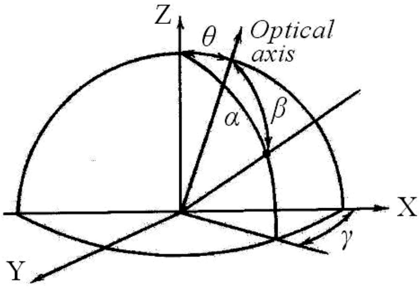


Fig. 4. Schematic representation of the conoscopic image in the spherical coordinates. θ is the angle between the normal and the optical axis of tilted oriented mesophase.

The conoscopic image, which is shown in Fig. 3a consists of the isogyres. These isogyres are not crossed in the centre of the field of vision. As is seen in Fig. 3a, the centre of the isogires is displaced from the centre of the field of vision. The isogyres in the conoscopic image are parallel to the direction of the oscillation, which are transmitted by the polarizer and analyzer. The angle θ between the director $\vec{n}$ and a normal to the reference surface of the sandwich-cell can be determined as $\theta = \arctan\frac{r}{L}$. (Here r is displaced the centre of the conoscopic image from the centre of the field of vision; L is the thickness of the liquid crystalline layer). Estimation showed that for this situation the angle θ between the director $\vec{n}$ and a normal to the reference surfaces of the sandwich-cell was $5^0 45'$. During the time, a fluent changes of the conoscopic image have been observed (Fig. 3). After 150.0 min the conoscopic image as the Maltese cross, presented in Fig. 3b, has been observed. This situation corresponds to the homeotropic alignment of the mesophase. Investigations showed that the character of orientational processes in the lyotropic systems under investigations is very similar. As an example, in Fig. 5 the dependence of the θ angle vs. time for 22.85wt%HTAB + 73.86wt%H_2O + 3.29wt%DeOH composion is presented. As is seen in this figure, the θ angle fluently decreases with time and full homeotropic alignment is characterized by the $\theta = 0$ and the conoscopic image as the symmetric cross. Schematic representation of the homeotropic alignment of the N_D mesophase under investigation, and position of the director and magnetic field are presented in Fig. 6a. Thus, the homeotropic alignment of the N_D mesophase has been obtained by application of the magnetic field perpendicularly to the reference surfaces of the sandwich-cell. In this case the optical axis of the mesophase under investigation is also directed perpendicularly to the reference surfaces of the sandwich-cell. Such behavior of the N_D mesophase shows that this mesophase in HTAB/H_2O/DeON lyotropic system has the positive diamagnetic anisotropy (i.e. $\Delta\chi > 0$).

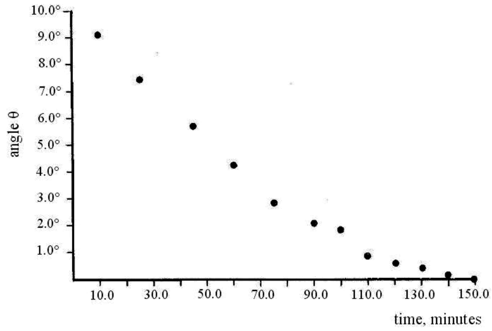


Fig. 5. Dependence of the angle θ between the director $\vec{n}$ and a normal to the surface of the sandwich-cell vs. time.

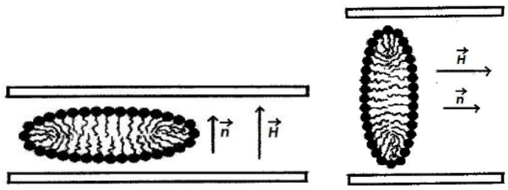


Fig. 6. Schematic representation of position of the director and magnetic field for the N_D mesophase under investigation. Homeotropic alignment (a) and planar alignment (b).

On the second stage of these investigations, magnetic field has been applied to a sample with the homeotropic alignment parallel to the reference surfaces of the sandwich-cell. Alignment process in the N_D mesophase has been investigated by texture transformations under magnetic field. Investigations showed that in this case sufficiently interesting orientational dynamics of the N_D mesophase takes place. On the initial stage, the homeotropic alignment has been gradually destroyed, and after 10 min texture presented in Fig. 7a has been observed. Then, after 30 min texture presented in Fig. 7b has been observed. Appearance of these textures is connected with the effect of the reverse flow because of the *homeotropic* $\rightarrow$ *planar* transition. This flow leads to an origination of formations as the cylindrical lenses [35,36]. As is seen in Figs.7a,b, these formations are nearly parallel to each other and form dense net. Appearance of such formations under influence of the magnetic field characterizes by three main conditions: availability of initial homeotropic alignment of the nematic micelles, strong coupling between these micelles and the reference surfaces of the sandwich-cell, and value of the magnetic field, which is three times more than the critical field for the Fredericksz effect [35-38]. As is noted in [6,35], the cylindrical lenses can be form only for samples, which have sufficiently well homeotropic orientation. For samples with the tilted oriented texture, which was characterized by degenerated conoscopic cross, we do not observed textures with the cylindrical lenses. In the case of the tilted oriented texture, during the *tilted orientation* $\rightarrow$ *planar* transition, texture with the inversion walls of the first order has been observed.

During the time, formations which are presented in Figs. 7a,b were destroyed and after 1.5 and 2.5 hours textures, presented in Figs. 7c,d, have been observed. As is seen in Figs. 7c,d, in these textures sufficiently large regions with the planar alignment take place. After 4.5 hours process of the texture transformations was finished and sufficiently uniform planar alignment was formed. In this case the optical axis of the mesophase under investigation is directed perpendicularly to the reference surfaces of the sandwich-cell. Schematic representation of the planar alignment of the N_D mesophase under investigation, and position of the director and magnetic field are presented in Fig. 6b. During process of the texture transformations, the dependences of the optical birefringence Δn vs. time have been determined. Investigations showed that these dependences for the lyotropic systems under investigations were very similar. As an example, in Fig. 8 the dependence of the Δn vs. time for 22.85wt%HTAB + 73.86wt%H_2O + 3.29wt%DeOH composion is presented. As is seen in this figure, fluent increase of the Δn with time takes place for the mesophase under investigation. Besides, as is seen in Fig. 8, the birefringence of the lyotropic nematic mesophases is usually ten times smaller than that in the thermotropic nematic mesophase [6,39-43]. We would like to note that, by application of the external magnetic field in OX and OZ directions, the same results for the Δn have been obtained.

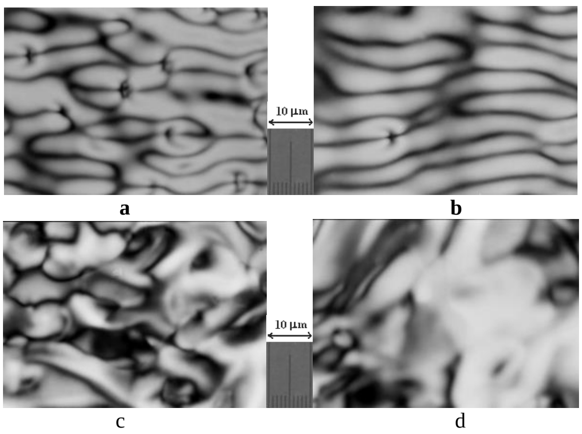


Fig. 7. Texture dynamics of for the N_D mesophase under investigation. Transformation from the homeotropic alignment to the planar alignment after 10 min (a), 30 min (b), 1.5 hours (c) and after 2.5 hours (d).

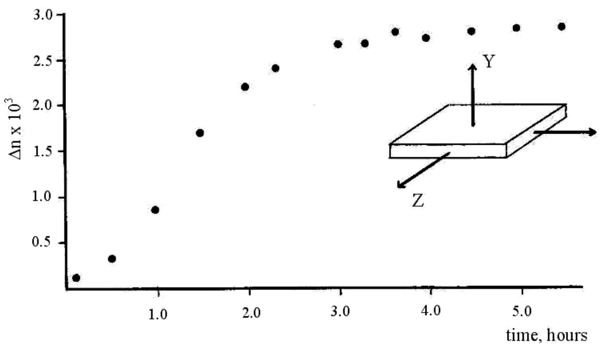


Fig. 8. Dependence of the optical birefringence vs. time in N_D mesophase by influence of magnetic field.

In conclusion we would like to note that, as is known, in the case of $\Delta\chi > 0$ the director of the thermotropic liquid crystalline mesophase is aligned parallel to the external magnetic field; in the case of $\Delta\chi < 0$ the director of the thermotropic liquid crystalline mesophase is aligned perpendicularly to the external magnetic field [37,44,45]. Analogous situation has been observed in different lyotropic liquid crystalline systems for the N_C and N_D mesophases by various scientists [29,36-39]. Besides, in the N_C and N_D mesophases both and $\Delta\chi > 0$ and $\Delta\chi < 0$, i.e. N_C^+, N_C^-, N_D^+ and N_D^- have been observed [6,29,50-58]. Obviously, in lyotropic liquid crystals the sign of the magnetic anisotropy in N_C and N_D mesophases is connected not only with the diamagnetic anisotropy of the micelle shapes but also with diamagnetic anisotropy of the amphiphile molecules [6,36,59,60].

4. Summary

Processes of the magneto-morphologic transformations, changes of the orientational and optical properties of the N_D mesophase in HTAB/H_2O/DeON lyotropic liquid crystalline system have been investigated. Investigations showed that in this mesophase process of the magneto-morphologic transformations is complicated and unremitting process of texture transformations.

Under influence of the external magnetic field, which was applied perpendicularly to the reference surfaces of the sandwich-cell, the homeotropic texture of the mesophase under investigations has been obtained. Such texture was characterized by the conoscopic image as the Maltese cross. Under influence of the external magnetic field, which was applied parallel to the reference surfaces of the sandwich-cell, the planar texture of the mesophase under investigations has been obtained. Such texture was characterized by the optical birefringence as $\Delta n = 0.0029$. Such behavior of the N_D mesophase in HTAB/H_2O/DeON lyotropic liquid crystalline system shows that this mesophase has the positive diamagnetic anisotropy (N_D^+ mesophase).

We would like to note that N_D^+ mesophase [46,59,61,62] and N_D^- mesophase [35,48,49,63,64] is known. In N_D^+ mesophase the director $\vec{n}$ is oriented parallel to the direction of the external magnetic field; in N_D^- mesophase the director $\vec{n}$ is oriented perpendicularly to the direction of the external magnetic field. Orientation of the diamagnetic micelles in the external magnetic field can be connected with both diamagnetic anisotropy of shapes of the anisometric micelles and diamagnetic anisotropy of the amphiphile molecules. Therefore the ordering in lyotropic nematic mesophases is characterized by the order parameter as $S = S_{mc} \cdot S_{ml}$ [6,59,65-67]. Here S_{mc} is the order parameter which describes the ordering of the anisotropic micelles relative to the director $\vec{n}$ of the mesophase; S_{ml} is the order parameter, which describes the ordering of amphiphile molecules relative to the chosen axis of the micelle.

Registry Nos.: n-hexadecyl-n,n,n-trimethylammonium bromide (cetiltrimethylammonium bromide, cetrimonium bromide, palmityl-trimethylammonium bromide) (HTAB) – Sigma-H9151; decanol (DEOH) – Merck-803463.

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