Europhys. Lett., 12 (2), pp. 135-141 (1990)

Defects in Degenerate Hybrid Aligned Nematic Liquid Crystals (*).

O. D. LAVRENTOVICH and YU. A. NASTISHIN

Institute of Physics, Academy of Sciences of the Ukrainian SSR pr. Nauki 46, Kiev-28, Ukrainian SSR, USSR

(received 12 October 1989; accepted in final form 6 March 1990)

PACS. 61.30 – Liquid crystals.

Abstract. – High-strength and zero-strength defects have been observed for the first time in degenerate hybrid aligned nematic films. The defects possess complex asymmetrical structures, and their existence is the consequence of the splay cancelling principle. The relation between the strength of defects and the number of dark brushes in the textures is violated.

Introduction. – Topological defects in the distributions of the order parameter fields are extremely important objects of research in various branches of condensed-matter physics. Some clarity has now been brought by the homotopy classification of these defects (ref. [1], for example). In the experimental search for a fundamental understanding of topological defects there is considerable interest in the nonuniform states of nematic liquid crystals. A great deal is already known about defects in nematics, owing to the fact that experimental investigation of these objects can be made simply with the use of an optical polarizing microscope.

In thin flat nematic samples, taken between a slide and a coverslip, one usually observes textures with dark brushes. The brushes occur in areas in which director $\boldsymbol{n}$ is either parallel or perpendicular to the plane of polarization of the incident beam. As follows from numerous observations and from the simplest theoretical models, defects in nematics possess the following properties:

1) The distribution of $\boldsymbol{n}$ around the defect is symmetrical. For example, in the simplest model of the planar structure, in which $\boldsymbol{n}$ is confined to the horizontal (x, y)-plane and is not a function of z, the normal to the film $\boldsymbol{n}$ is distributed in such a way that $n_x \sim \cos m\varphi$ $n_y \sim \sin m\varphi$, where m is the strength (or topological charge) of the defect; $m = \pm 1/2, \pm 1, \ldots$. The strength m is defined as the number of revolutions of $\boldsymbol{n}$ by multiples of 2π in going once around the defect.

(*) This work was presented in part at the 8th Liquid Crystal Conference of Socialist Countries, Kraków, Poland, August 28-September 1, 1989.

2) There is a simple relation between the strength m of the defect and the number N of dark brushes in the corresponding texture [2]:

$$|m| = N/4 . \tag{1}$$

3) High-strength defects ($|m| > 1$) are prohibited in nematics. Really, a greater m implies a greater curvature of $\boldsymbol{n}$ and, as a result, a greater elastic energy F. For the planar model [1, 2] $F \sim m^2$. So, in textures one observes only points with either 2 or 4 brushes [2], which correspond to defects with $m = \pm 1/2$ and $m = \pm 1$, respectively. Rare cases of higher-strength defects have been observed in exotic systems: emulsions of lyotropic with thermotropic nematics [3] as well as in thermotropic nematics with special impurities [4, 5]. Unfortunately, up to now the question of the stability of these defects has not been clarified.

4) Any defect with $m \neq 0$ as well as any nonuniform state with $m = 0$ costs an elastic energy which is greater than the energy of the uniform state (for the latter $m = 0$ by definition).

With this background, we now report our observations of defects in a physically well-defined nematic system, for which all the above-mentioned statements are false. The system under investigation is a flat nematic film with different but degenerate (in the plane of the film) orientations of $\boldsymbol{n}$ on the lower and upper surfaces. We denote such degenerate hybrid aligned nematic films as HAND films. We describe the main structural properties of defects in these films, the optical peculiarities of their textures, and propose an interpretation of their stability as a consequence of the «splay cancelling» principle [6].

Experimental part. – To provide the above-mentioned degenerate hybrid boundary conditions we deposited the nematic film (pentylcyanobiphenyl, 5CB, or methoxybenzylidenebutylaniline, MBBA) on the surface of an isotropic liquid (glycerine or polyethylenglycole). The liquid surface imposed degenerated tangential boundary conditions for $\boldsymbol{n}$ on the lower surface of the nematic film. The upper boundary of the nematic layer was left free. Since we were studying 5CB and MBBA, the orientation of $\boldsymbol{n}$ at upper surface was nearly normal. As a result, the geometry of the HAND film was created.

Textures and structures of defects. – When observed under a polarizing microscope, HAND films with thickness $1\,\mu\text{m} \lesssim d \lesssim 20\,\mu\text{m}$ exhibit unusual Schlieren textures, which contain a rich array of point defects, located near the lower surface of the film. The first important feature of the textures is the existence of points with an enormously large number of dark brushes. Points with $N = 4, 6, 8, 10, 12, 16$ (and sometimes with $N = 5, 7$), etc., have been observed (fig. 1-4). Moreover, a large number of pairs of ± 1 defects can be seen appearing spontaneously from the uniform (in the horizontal plane) state (fig. 5). In thin HAND films ($d \sim 1\,\mu\text{m}$) defects form regular networks (fig. 6). The latter are similar to the networks of defect points observed at the nematic-isotropic interface [7].

The second important feature of these textures is the nonuniform distribution of the dark brushes around defect points. The angle ξ between two successive brushes is different for different sectors of the defect texture. There is a sector where $\xi = 90°$ and there are one or few sectors where ξ is much smaller ($\lesssim 10°$).

The existence of points with 6 or 10 brushes is surprising for hybrid aligned nematics. In accordance with (1), these textures must correspond to half-integer strength defects. However, half-integer defects are prohibited in the hybrid aligned thin films due to the polar character of the film structure. In order to solve this contradiction, we determined the real director distribution by making the observations under the polarizing microscope with a

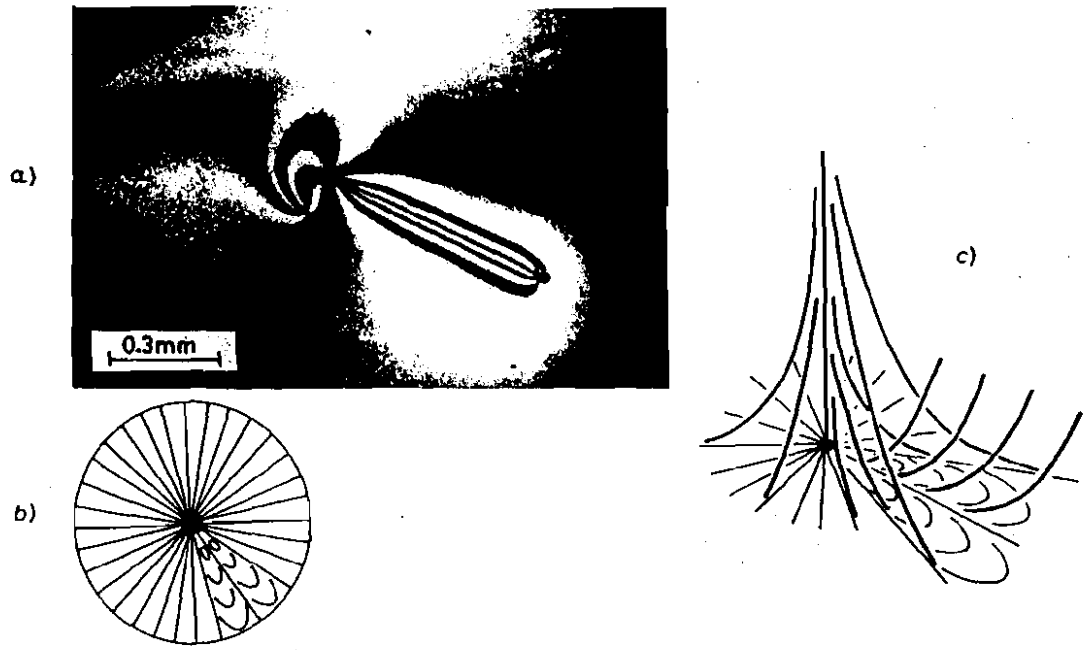

Fig. 1. – Defect with $m = 2$ and $N = 8$ in a HAND film of 5CB on glycerine: texture with 8 brushes (*a*)), structure in projection onto the film plane (*b*)), and general scheme of the structure (*c*)).

sensitive colour plate and a quartz wedge [8]. It was found that the distortions of n are distributed nonuniformly in the film plane. Namely, inside one sector of the horizontal plane ($\Phi \leqslant \varphi \leqslant 2\pi$, sector I), the distribution is radial, as for the $m = 1$ defect. The scarcity of the director revolutions up to $m \neq 1$ is filled up in the remaining narrow sector ($0 \leqslant \varphi \leqslant \Phi$, sector II) (fig. 1-4). In other words, the defects in HAND films represent the combinations of two

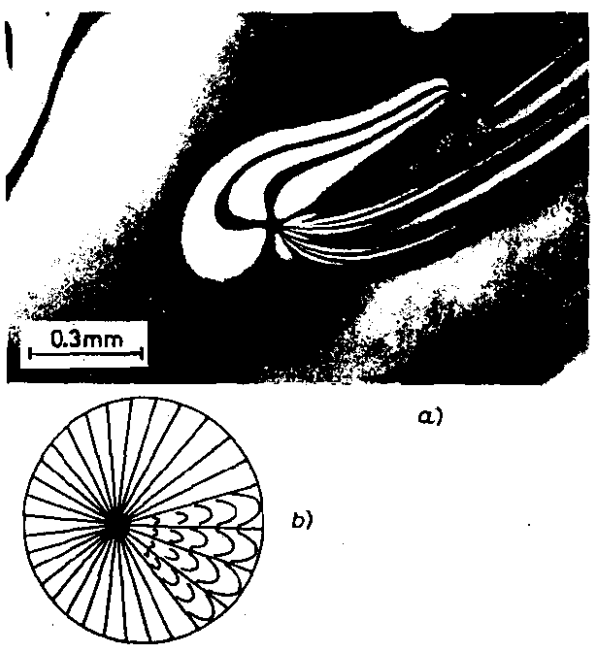

Fig. 2.

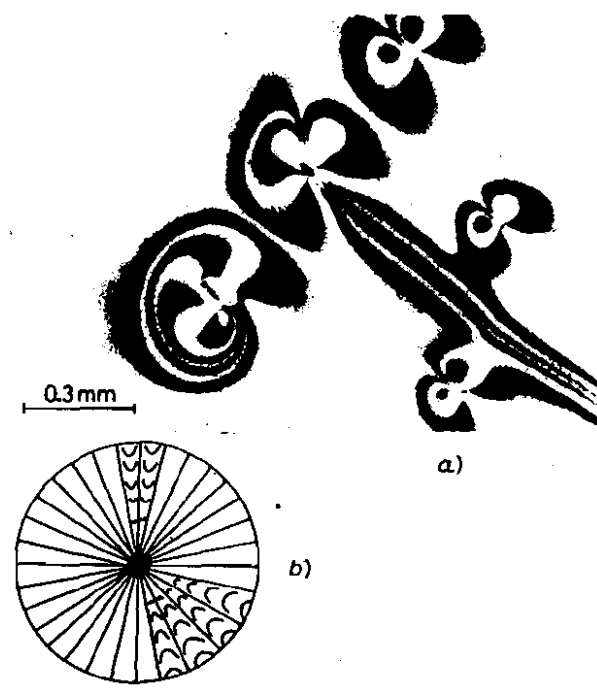

Fig. 3.

Fig. 2. – Defect with $m = 3$ and $N = 12$ in a HAND film of 5CB on polyethylenglycole: texture (*a*)) and structure (*b*)).

Fig. 3. – Defect with $m = -1$ and $N = 10$ in a HAND film of 5CB on glycerine: texture (*a*)) and structure (*b*)).

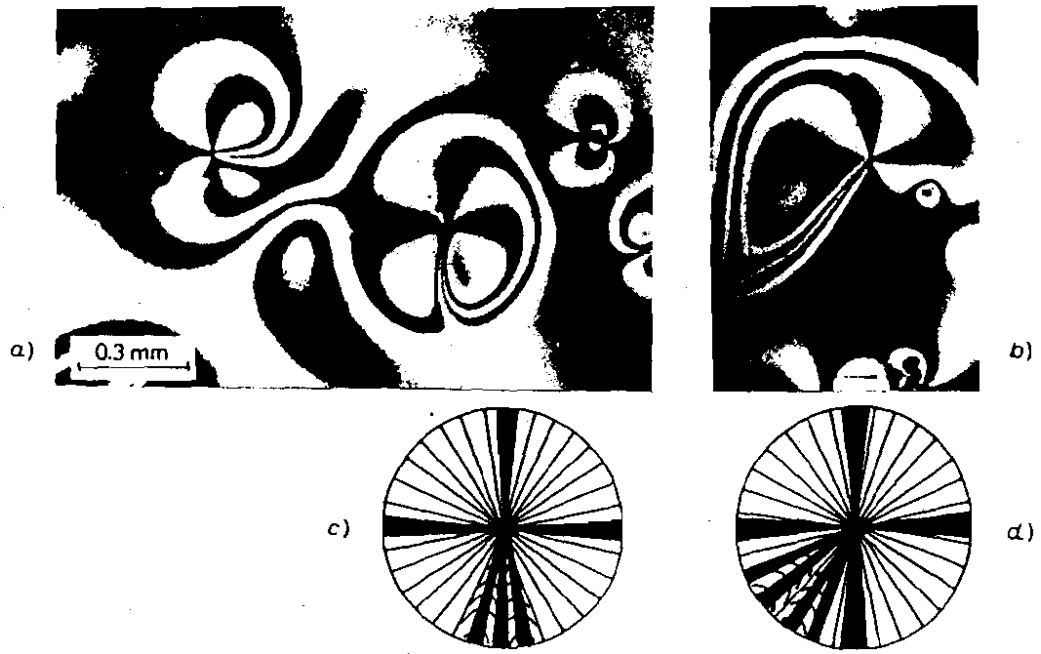

Fig. 4. – Defects with $m = 0$ in HAND films: textures with different numbers of dark brushes, $N = 6$ (a)) and $N = 8$ (b)); the corresponding structure provides $N = 6$ (c)) and $N = 8$ (d)) in two different sample orientations between crossed polarizers, which are oriented in the vertical and horizontal planes of the figure.

configurations with a common core but with different curvatures of $\boldsymbol{n}$ and different effective values of the strength M. As follows from experiment, these configurations may be written approximately as

$$n_x = \sin\theta(z)\cos M\varphi(x, y), \qquad n_y = \sin\theta(z)\sin M\varphi(x, y), \qquad n_z = -\cos\theta(z), \tag{2}$$

where $M = M_{\mathrm{I}} = 1$ for sector I and $M = M_{\mathrm{II}} = 1 + 2\pi(m-1)/\Phi$ for sector II. For the sake of simplicity, we suppose $\theta(z) = \theta_{\mathrm{L}} - \alpha z$, where $\alpha = (\theta_{\mathrm{L}} - \theta_{\mathrm{U}})/d$, $\alpha > 0$, θ_{L} and θ_{U} are the angles between $\boldsymbol{n}$ and the normals to the lower and upper surfaces, respectively. The last assumption is natural for large scales in the one-constant approximation, owing to the fact that $\theta(z) = \theta_{\mathrm{L}} - \alpha z$ exactly in the uniform state of hybrid aligned film [9].

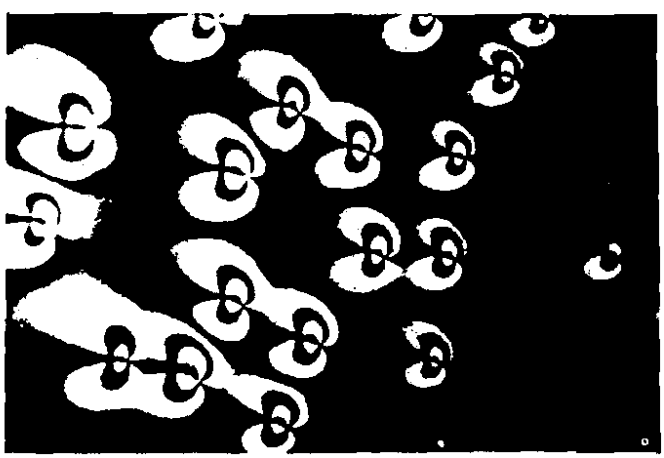

Fig. 5.

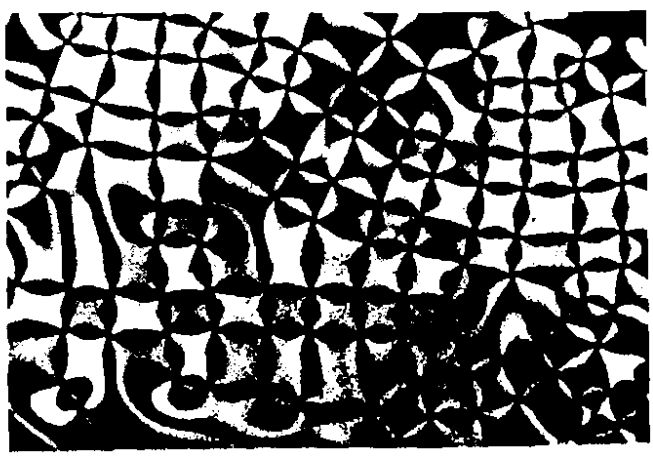

Fig. 6.

Fig. 5. – Texture of $m = \pm 1$ pairs which spontaneously arise in a HAND film of 5CB on polyethylenglycole.

Fig. 6. – Two-dimensional periodic network of $m = \pm 1$ defects in a HAND film of 5CB on glycerine.

The complex character of the defect structure can explain the above-mentioned textural paradox. Let us briefly consider the relation between the total strength m of the defect and the total number N of brushes in the observed textures.

As follows from the general optical properties of the nematics [2], the angle between two successive brushes is $\xi = \pi/2|M|$. It is clear that for sectors with different M this angle must hold different values: inside the sector I with $M_I = 1$, one finds $\xi = \pi/2$, and inside the sector II with $M_{II} = 1 + 2\pi(m-1)/\Phi$, one obtains $\xi = \pi\Phi/2|1 + 2\pi(m-1)|$. As a result, the number N of brushes for the complex distribution (2) may be estimated as $N_I = (4 - 2\Phi/\pi)$ inside the sector I and $N_{II} = 2\Phi|M_{II}|/\pi$ inside the sector II. Thus, for a defect with strength m in a HAND film, the total number of dark brushes is

$$\begin{cases} N = 4m, & \text{if } m \geqslant 1, \\ N = 4(|m| + 2 - \Phi/\pi), & \text{if } m < 1. \end{cases} \tag{3}$$

So, in the general case $N \neq 4|m|$. The model explains why one observes 6 or 10 brushes (even 5, 7, etc., brushes). These pictures correspond to integer m ($= 0, -1$), rather than to $|m| = 3/2$ or $5/2$, which are prohibited in HAND geometry (fig. 3, 4). It is important to note that for the same defect with $m = \text{const}$, N may be altered by changing Φ. Moreover, even for $\Phi = \text{const}$, N may be changed simply by sample rotation, as is shown in fig. 4*c*), *d*) for a defect with $m = 0$. For the sake of simplicity we did not take into account this fact in our calculations.

Elastic properties of defects. – The availability of two different sectors in the defect structure plays a crucial role in the defect stability. In order to comprehend the situation, let us consider the role of the large sector I with radial structure in the elastic energy saving. It is sufficient to compute the elastic energy F_1 of the defect with radial structure in the whole azimuthal plane ($m = 1$) and then compare this energy to the energy F_0 of the uniform (in the azimuthal plane) state of HAND films, see fig. 7.

In the one-constant approximation, the elastic energy of any state may be calculated as [1]

$$F = \frac{K}{2} \int_V [(\operatorname{div} \boldsymbol{n})^2 + (\operatorname{rot} \boldsymbol{n})^2] \, dV, \tag{4}$$

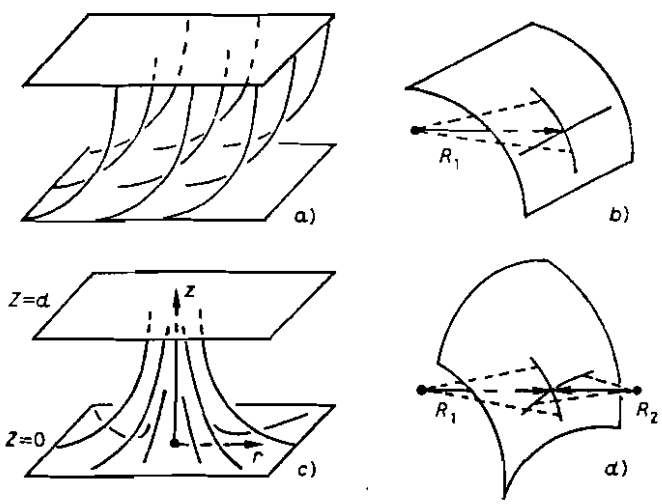

Fig. 7. – Distributions of n for a uniform HAND film (*a*)) and for a defect state with $m = 1$ (*c*)). The corresponding surfaces, which are perpendicular to n lines, correspond to $(R_1 R_2)^{-1} = 0$ in the uniform state (*b*)) and $(R_1 R_2)^{-1} < 0$ in the defect state (*d*)).

where K is the elastic constant and V is the volume of the sample. After substitution of the distribution (2) with $M=1$ in (4) and integration over $r_c \leqslant r \leqslant R$, $0 \leqslant \varphi < 2\pi$, $0 \leqslant z \leqslant d$ in cylindrical coordinates (r, φ, z), one obtains

$$F_1 = \frac{\pi K d}{2}[\alpha^2 R^2 + \ln(R/r_c) - 2\alpha R] + F_c, \tag{5}$$

where r_c is the radius of the defect core, $F_c \sim W r_c^2$ its energy, and W is the anchoring energy. On the other hand, the energy of the uniform HAND state for the film of the same size is

$$F_0 = \pi K d \alpha^2 R^2/2. \tag{6}$$

A comparison of (5) and (6) shows that the defect state may be energetically preferable to the uniform state, owing to the presence of the term $(-2\alpha R)$ in (5). For usual values $K = 10^{-11}$ N, $d \sim r_c \sim 10\,\mu$m, $W = 10^{-5}$ J/m^2, one obtains $F_1 < F_0$ if $R/d > 1$. This result is a consequence of the principle of splay cancelling [6]: if the boundary conditions force a variation of $\boldsymbol{n}$ in one direction, then a variation of $\boldsymbol{n}$ in another direction can lead to a cancellation of splay contribution. This principle may be illustrated by rewriting $(\mathrm{div}\,\boldsymbol{n})^2$ as $(1/R_1 + 1/R_2)^2$, where R_1 and R_2 are the principal radii of curvature of the surfaces, which are perpendicular to $\boldsymbol{n}$. As a consequence, the elastic energy can be reduced when $R_1 R_2 < 0$. For the uniform HAND film $R_1^{-1} \sim \alpha$, $R_2^{-1} = 0$, while for defect state with $m=1$, $R_1^{-1} \sim \alpha$, $R_2^{-1} \sim r^{-1} \neq 0$ and $R_1 R_2 < 0$, see fig. 7. The negative sign of the term $(-2\alpha R)$ in (5) is a direct consequence of the condition $R_1 R_2 < 0$.

Sector I with radial structure saves the elastic energy in other defect structures with $m \neq 1$. In fact, if this sector is absent, the energy of the trivial defect distribution with $m = M = M_I = M_{II} \neq 1$,

$$F_m = \frac{\pi K d}{2}[\alpha^2 R^2 + m^2 \ln(R/r_c)] + F_{cm}, \tag{7}$$

is greater than F_0 in all cases. However, F_m is reduced by the introduction of sector I with radial structure inside the defect distribution. After insertion of (2) with $M_I = 1$ and $M_{II} = 1 + 2\pi(m-1)/\Phi$ in (4), one obtains

$$F_m = \frac{\pi K d}{2}\{\alpha^2 R^2 + [2m - 1 + 2\pi(m-1)^2/\Phi] \ln(R/r_c) - 2\alpha R(1 - \Phi/2\pi)\} + F_c', \qquad F_c' \approx F_c. \tag{8}$$

The equilibrium value of Φ is obtained by minimizing F_m:

$$\Phi_0 = [2\pi^2(m-1)^2 \ln(R/r_c)/\alpha R]^{1/2}. \tag{9}$$

An analysis of (8), (9) and (6) leads to the conclusion that for sufficiently large ratios R/d, defect states with $m \neq 1$ are also energetically preferable to the uniform state because of the negative term $(-2\alpha R)$ in (8). For example, for a $(m=2)$-defect with $\Phi_0 \approx \pi/2$ (fig. 1), one obtains $F_2 < F_0$ when $R/d > 6$. Moreover, defect configurations with $m=0$ (fig. 4) are also energetically preferred to the uniform HAND state. The latter can qualitatively explain the appearance of ± 1 pairs and arrays (fig. 5, 6) in HAND films.

Conclusions. – In summary, we find it interesting and surprising that a simple HAND film geometry leads to unexpected changes in the fundamental understanding of topological defects in nematics. Let us summarize the main properties of the defects in HAND films.

1) Defects possess asymmetrical structures, which are the combinations of sectors with different n curvatures. Each defect structure contains a large sector with radiallike distribution. The scarcity of the n revolutions is filled up in the remaining narrow sector.

2) The strength m of the defect and the number N of brushes in the corresponding texture do not fulfil the simple relation $|m| = N/4$ in the general case, see (3). Moreover, for the same defect with $m = \text{const}$, N may be changed by sample rotations as well as by modifications of the sector parameters.

3) High-strength defects as well as zero strength defects are systematically observed in HAND films. The stability of these defects is connected with the splay cancelling principle. The energy of the defect state may be smaller than the energy of the uniform (in the horizontal plane) HAND state.

We expect that the above-mentioned features cast light upon the earlier data on high-strength defects [3-5]. It seems likely that in emulsions of 5CB (or MBBA) with lyotropic nematics, which contained water and ethylenglycol [3], the HAND films of 5CB (or MBBA) with free upper boundary were in fact created on the surface of isotropic liquid. In impure nematics [4, 5] the action of the splay cancelling principle may be initiated by the spontaneous curvature induced by impurities.

The forthcoming aim in the investigation of HAND films is to connect the properties of isolated defects with properties of strings connecting pairs of defects [10] and domain structures, which have been recently observed in very thin HAND films [11].

* * *

We are grateful to V. M. Pergamenshchik for fruitful discussions.

REFERENCES

[1] KLÉMAN M., *Points, Lines, and Walls* (John Wiley and Sons, New York, N.Y.) 1983.
[2] CHANDRASEKHAR S., *Liquid Crystals* (Cambridge University Press, Cambridge) 1977.
[3] LEE H. and LABES M. M., *Mol. Cryst. Liq. Cryst., Lett.*, 82 (1982) 199.
[4] CHANDRASEKHAR S. and MADHUSUDANA N. V., unpublished work (1966).
[5] MADHUSUDANA N. V. and PRATIBHA R., *Curr. Sci.*, 51 (1982) 877; *Mol. Cryst. Liq. Cryst.*, 103 (1983) 31.
[6] PRESS M. J. and ARROTT A. S., *J. Phys. (Paris), Coll.*, 36 (1975) C1-177.
[7] MADHUSUDANA N. V. and SUMATHY K. R., *Mol. Cryst. Liq. Cryst.*, 129 (1985) 137.
[8] VOLOVIK G. E. and LAVRENTOVICH O. D., *Ž. Èksp. Teor. Fiz.*, 85 (1983) 1997 (*Sov. Phys. JETP*, 58 (1983) 1159).
[9] BARBERO G. and BARBERI R., *J. Phys. (Paris)*, 44 (1983) 609.
[10] LAVRENTOVICH O. D. and ROZHKOV S. S., *Pis'ma Ž. Èksp. Teor. Fiz.*, 47 (1988) 210 (*JETP Lett.*, 47 (1988) 254).
[11] LAVRENTOVICH O. D. and PERGAMENSHCHIK V. M., *Pis'ma Ž. Èksp. Teor. Fiz.*, 15 (1989) No. 5, 73; to appear in *Mol. Cryst. Liq. Cryst.*