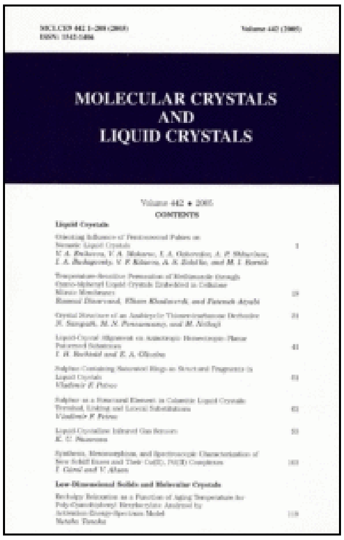

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On the Occurrence of Point and Ring Defects in the Nematic-Nematic Coexistence Range of a Binary Mixture of Rod-like and Disc-like Mesogens

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On the Occurrence of Point and Ring Defects in the Nematic-Nematic Coexistence Range of a Binary Mixture of Rod-like and Disc-like Mesogens

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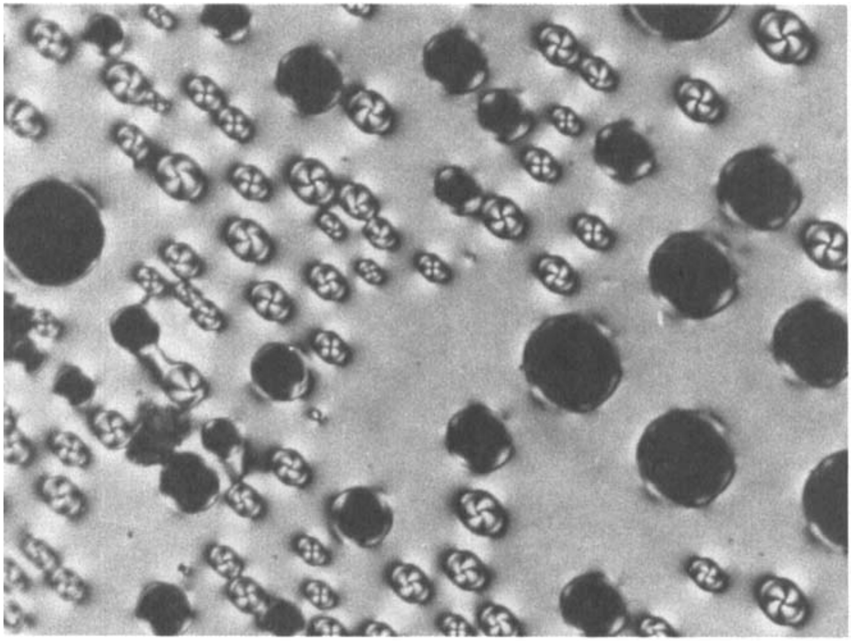
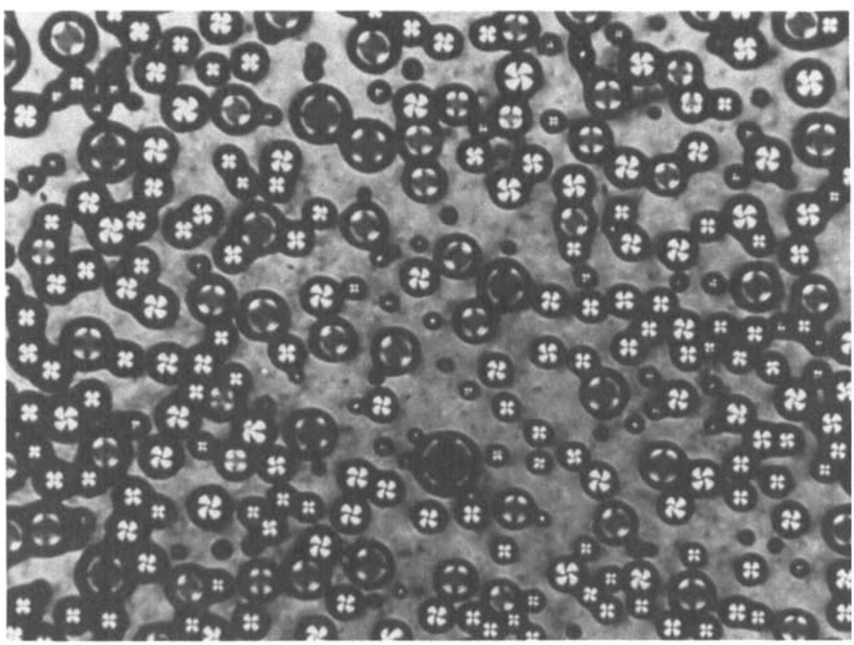
We report some observations on the director configurations in the coexistence range of two nematic phases, N_R and N_D in a binary mixture of rod-like and disc-like nematogens. The director alignment is continuous across the boundary of the N_R and N_D phases. Point defects of strength $+1$ in N_R droplets in an otherwise continuous N_D medium are found to be annulled by singular ring disclinations of strength $-\frac{1}{2}$. Further, the N_R drops which orient homeotropically at the glass plates have a point defect of strength $+1$ only if their size is less than about 7 μm. Larger drops have ring disclinations of strength $+\frac{1}{2}$ instead of $+1$ point defects. This changeover can be understood on the basis of simple energy considerations.

Keywords: point defects, ring defects, nematic-nematic coexistence

We recently studied the phase diagram of two nematogens, one of them being the rod-like compound 4-(*trans*-4-*n*-propylcyclohexyl)phenyl-4-*n*-propylbenzoate (3CHP3B) and the other the disc-like compound hexa-*n*-heptyloxybenzoate of triphenylene (C_7OHBT).[1] In some composition ranges we found a coexistence of two nematic phases, one of them relatively rich in rod-like molecules (N_R) and the other relatively rich in disc-like molecules (N_D). When the glass plates are not specially treated, the N_R phase appears first on cooling from the isotropic phase in the form of droplets, each droplet having a point defect of strength $+1$ at the center. The N_D phase spreads around the N_R droplets and has a schlieren like texture. In this paper we report the director configuration in such mixtures taken between glass plates coated with SiO at an oblique angle. In such preparations, only the N_D phase which is spread in a continuous manner has a planar alignment (as has been observed previously[2] with pure C_7OHBT). N_R droplets which are isolated from one another have point defects of strength $+1$ at their centers only



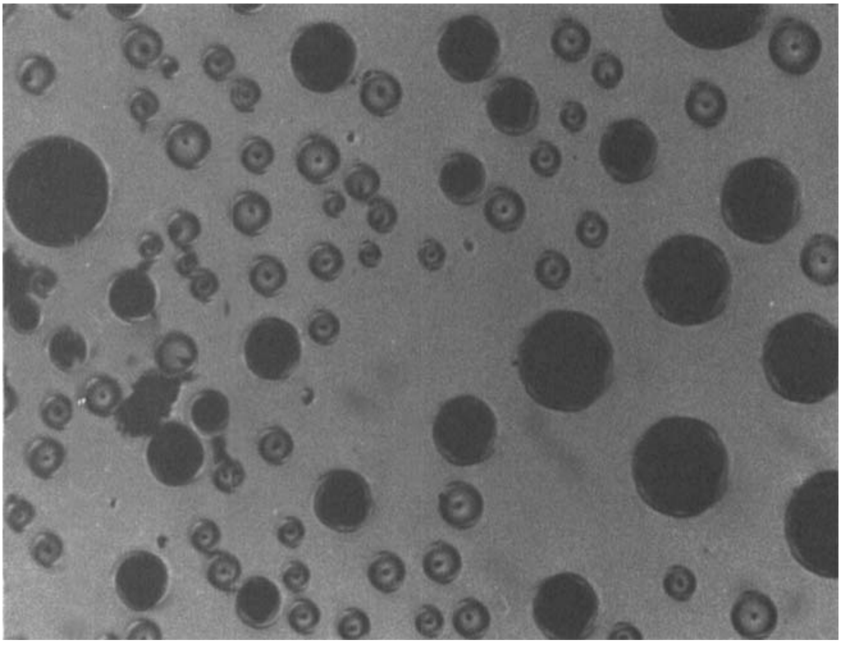
(a)



(b)

FIGURE 1a Coexistence of N_R and N_D phases in a mixture with 30 mol% of C_7OHBT at 88°C. Sample taken between SiO coated plates and crossed linear polarizers (× 675). The director in the N_D phase is aligned parallel to the lower edge of the photograph.

FIGURE 1b N_D phase going over to the I phase in a mixture with 30 mol% of C_7OHBT. Sample taken between SiO coated plates. Crossed linear polarizers (× 570).



(c)

FIGURE 1c Same sample area as in Figure 1a between crossed circular polarizers (× 675).

if the diameter is $\lesssim 7\ \mu m$ (Figure 1a). Observations on larger N_R drops clearly demonstrate that, surprisingly, the SiO coated glass plates favor a *homeotropic* alignment of the N_R phase.

Topological arguments[3] show that the director distortion due to a $+1$ point defect can be annulled in the far field by a disclination loop or singular ring defect of strength $-\frac{1}{2}$, as the director is apolar (Figure 2). It has also been argued that point and ring defects can be transformed from one to another.[4,5] Usually in nematics point defects with strength $+1$ occur in the bulk when the sample is either taken in a capillary tube treated for homeotropic alignment[6] or when it is in the form of spherical droplets with normal boundary conditions.[7,8,9] To our knowledge, the compensation of the point defect by a singular ring defect in the sense mentioned above has not been observed in nematic samples. On the other hand, because of the layer structure, such observations are easier in the smectic A phase. Trebin[3] has interpreted some experimental observations of Perez *et al.*[10] on the smectic A phase to mean that a hyperbolic point defect is annulled by a singular ring. In this paper we present the first observations of such an annulment in a nematic sample which is in the coexistence range mentioned earlier. Further, while N_R drops with diameter $\lesssim 7\ \mu m$ have point defects of strength $+1$, in larger drops ring defects with strength $+\frac{1}{2}$ are favored demonstrating the equivalence of these two types of defects.

Elastic anisotropy plays an important role in determining the director configuration around the point defects. For example though one expects a radial distribution of the director in spherical droplets in the one elastic constant approximation,

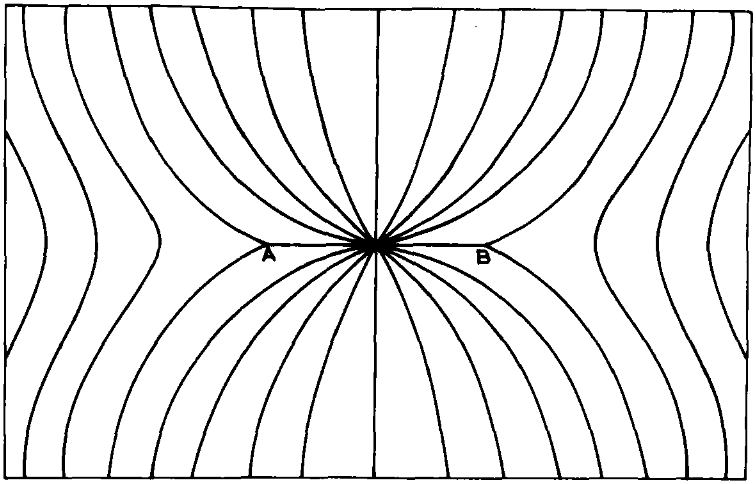


FIGURE 2 Schematic diagram of the director distortion around a point defect of strength +1 being annulled by a singular ring of strength $-\frac{1}{2}$. The singular ring intersects the paper at points A and B.

a more complicated configuration with a twist in the central region is quite often seen.[8,11] Numerical calculations using a cylindrical geometry show that such a twist distortion near the center of the point defect is possible when $k_{22} < k_{11}$. Very recently the director configuration in spherical nematic droplets of butoxyphenyl ester of nonyloxybenzoic acid which exhibits a smectic A phase below the nematic phase has been studied. Interestingly a transition from a radial point defect to a hyperbolic point defect accompanied by a non-singular ring of strength +1 occurs with increase in temperature[12] even though the boundary condition remains normal. This transition from the radial to the hyperbolic point defect is again a result of the elastic anisotropy. A radial structure is characterized by splay distortion and is favored when $k_{33} > k_{11}$. On the other hand a hyperbolic structure involves a lot of bend distortion and can be expected to occur when k_{33}/k_{11} has a favorably low value.[12] In droplets with tangential boundary conditions it has been observed[13] that depending on the ratio of k_{33}/k_{11} a bipolar or an axial alignment can be obtained. However, in the system studied by us, elastic anisotropy does not lead to any new qualitative features.

EXPERIMENTAL OBSERVATIONS

When the sample taken between SiO coated plates without any spacers is viewed between crossed polarizers, the following features are observed:

1. When the N_R droplets first appear in the I (isotropic) phase, the dark brushes that emanate from the centers of the point defects are parallel or perpendicular to the polarizers. With the onset of the N_D phase in the surrounding I region the brushes develop a curvature which can be either positive or negative (see Figure 1b). This indicates that the director makes a small angle with the normal to the

interface in the N_R–N_D coexistence region. By noting the position of the dark brushes with respect to the orientation of the polarizers the angle at the interface was found to be approximately 10°. The continuity of dark brushes across the N_R–N_D interface (Figure 1a) shows that the director is also continuous across the boundary.

2. Further, when viewed between crossed circular polarizers, we can see a dark spot at the center and two slightly elongated dark spots close to the periphery of each N_R droplet (Figure 1c). The central spot corresponds to the point defect. The other two spots are diametrically opposite to each other. The line joining these spots makes an angle of about 10° with the normal to the direction in which the N_D phase is aligned.

3. If the N_R droplets have a diameter >7 μm they look dark between crossed polarizers, implying that the director is homeotropically aligned, i.e., within these droplets the director is normal to the glass surfaces. The periphery of such a drop is bright, except in regions parallel or perpendicular to the polarizers (see Figure 1a).

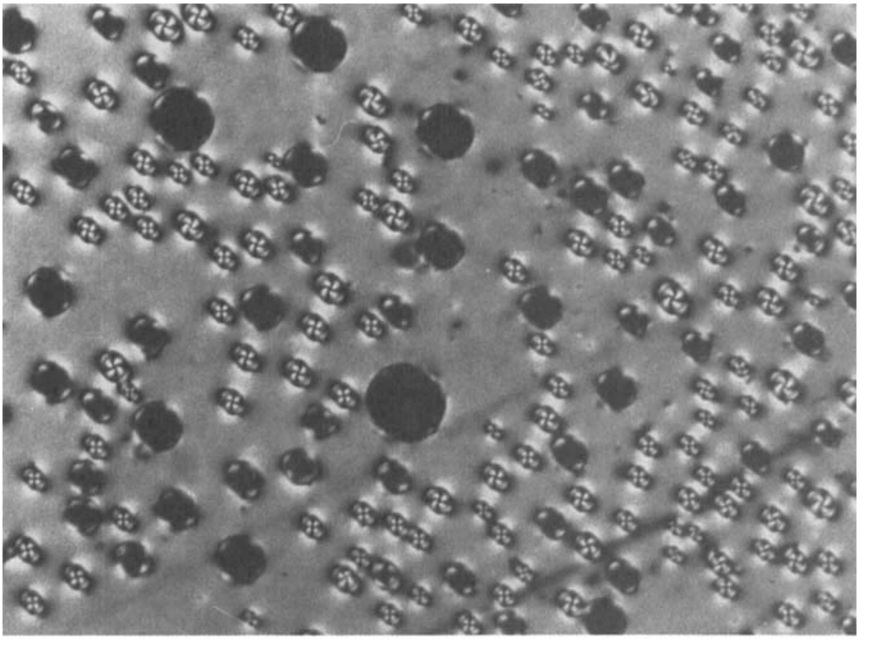
4. As the sample is cooled the $+1$ defects in small N_R droplets are found to move away from the centers of the droplets (Figures 3a and 3b). Further, in the larger N_R drops, the director field is no longer perfectly homeotropic.

DISCUSSION

As we noted earlier, the director alignment is continuous across the N_R–N_D interface. Considering a sample containing N_R drops with diameters $\lesssim 7$ μm, it is clear that we have a distribution of $+1$ point defects with ring defects of strength $-\frac{1}{2}$ surrounding them, in an otherwise uniformly aligned director configuration. As the sample is confined between two glass plates and the ring defect lies in a plane which is nearly perpendicular to the plates only two arcs of the singular ring are found. This provides the first example of a nematic specimen in which the ring defects of strength $-\frac{1}{2}$ are generated around a point defect of strength $+1$ to reduce the elastic energy. The two spots seen on the periphery of the droplet when viewed between circular polarizers (see Figure 1c) correspond to projections of the two arcs of the line defect. The director configuration within and outside the N_R droplet in the mid plane of the sample, which is parallel to the glass plates and contains the point defect is shown in Figure 4. The elongation of the outer spots in Figure 1c indicates that the plane containing the arcs of the ring defect is inclined to the vertical by a small angle.

This configuration can be checked by studying the pattern of dark brushes in relation to the orientation of crossed polarizers. Let the N_D phase have an overall planar alignment along the x-axis (see Figure 4) and let α be the angle between the radius vector at any point P outside the droplet and the x-axis. θ is the angle between the director and the radius vector, and ϕ that between the director and the x-axis and is equal to $\alpha \pm \theta$, the sign depending on the orientation of n with reference to the radius vector. Let ψ be the azimuthal angle made by the polarizer

(a)



(b)

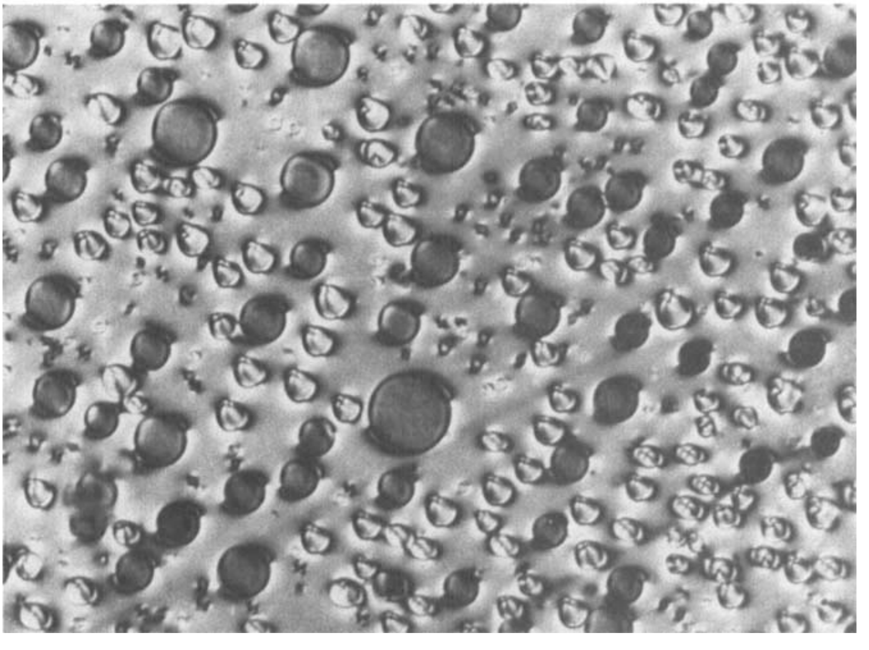


FIGURE 3a Coexistence of N_R and N_D phases in a mixture with 30 mol% of C_7OHBT at 88°C. Sample taken between SiO coated plates and crossed linear polarizers ($\times$ 560). Notice that the point defects are present at the centers of relatively small N_R droplets.

FIGURE 3b Same sample area as in Figure 3a at 80°C. The +1 point defects in most of the small N_R droplets have shifted to one end ($\times$ 560).

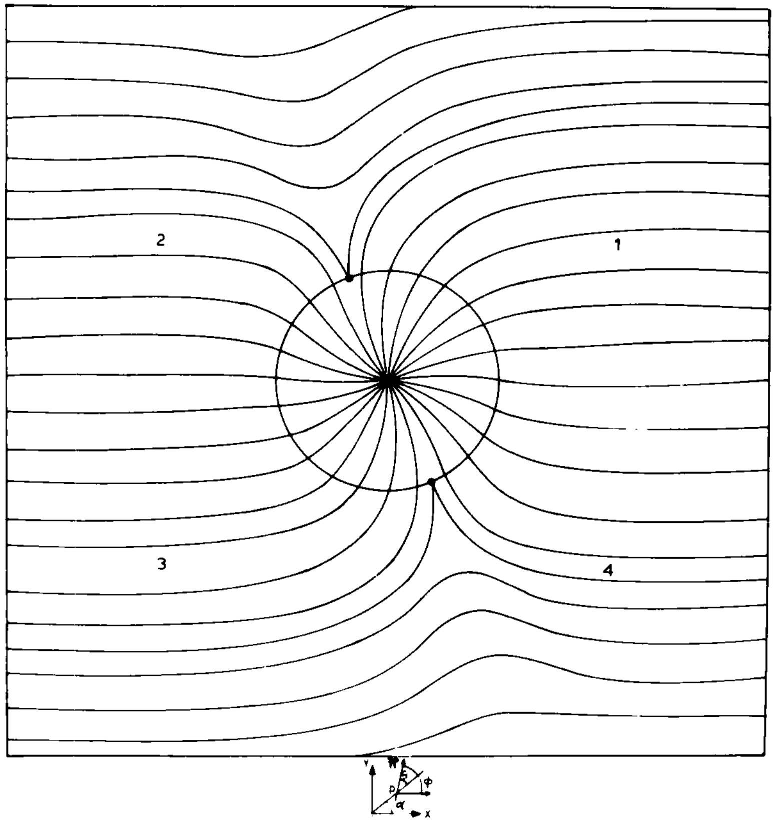


FIGURE 4 Schematic diagram of the director configuration within and outside an N_R droplet with a point defect in a plane parallel to the glass plates and containing the point defect, when the director makes a small angle with the N_R–N_D interface.

with the x-axis. The regions of the sample, where $\phi = \psi$ or $\psi + 90°$, are crossed out by the analyzer and appear dark.

When the polarizer is parallel to the x-axis, i.e., $\psi = 0$, regions with the director along or perpendicular to the x-axis are crossed out by the analyzer, i.e., there is an overall darkening of the sample except within and just outside the N_R droplets where the director makes an angle with the x-axis (Figure 5).

Figures 6a and 7a show photographs of the sample taken when $\psi \approx -10°$ and $-45°$ respectively. The expected patterns of dark brushes derived from Figure 4 in these two cases are shown in Figures 6b and 7b respectively.

The observation that N_R drops with diameters >7 μm have a predominantly homeotropic alignment clearly indicates that surprisingly the SiO coated plates favor a normal alignment of the director in the N_R phase. Taking this into account, we can expect the director configuration of N_R drops which have point defect of strength $+1$ to be as shown in Figure 8a. For relatively large drops, the director configuration changes over to that shown in Figure 8b which has a ring defect of

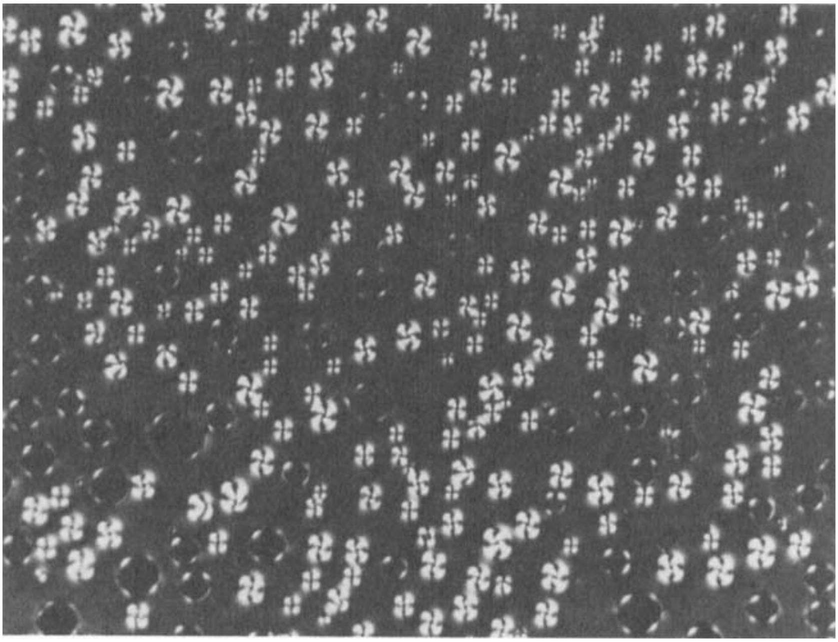
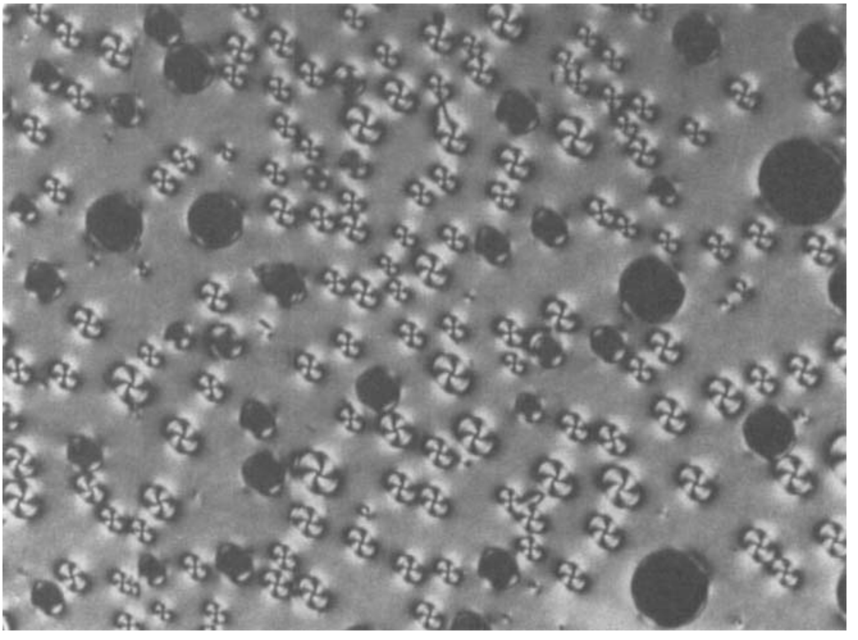


FIGURE 5 A coexistence region between N_R and N_D phases photographed between crossed linear polarizers with $\psi = 0°$ (× 570).

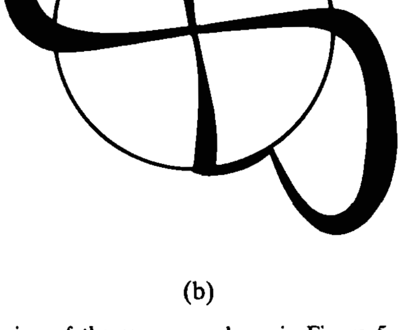
strength $+\frac{1}{2}$ near the periphery of the drop and contained in the mid plane of the sample. This again brings out the topological equivalence of point defects of strength $+1$ and ring defects of strength $+\frac{1}{2}$. Again in the far field a ring defect of strength $-\frac{1}{2}$ lying in an orthogonal plane can compensate the director distortion due to the $+\frac{1}{2}$ ring defect. The photograph between crossed circular polarizers (Figure 1c) shows the projections of the arcs of the ring defect of strength $-\frac{1}{2}$ outside the drops with homeotropic alignment in the center. They are better visible for drops with relatively small size, and appear to be more extended than the projection of the arcs around N_R drops with $+1$ point defects. This means that the arcs of ring defects of strength $-\frac{1}{2}$ make a considerable angle with the vertical plane.

We can understand the change in the director configuration when the diameter of N_R drop increases by energy considerations. For the sake of simplicity, we assume that the director is orthogonal to the N_R–N_D interface. Further, we assume that the interface has a cross section in the form of a circular arc of radius $d/2$ where d is the thickness of the sample, so that the director orientation can smoothly vary from the N_R-glass to the N_R–N_D interface (Figure 8).

It is difficult to obtain an analytical solution for the director configuration which depends on both r and z and contains the point defect. We estimate the distortion energy of the director field in the N_R drop in the two possible configurations in the one elastic constant ($k_{11} = k_{33} = k$) approximation using the following simple arguments.



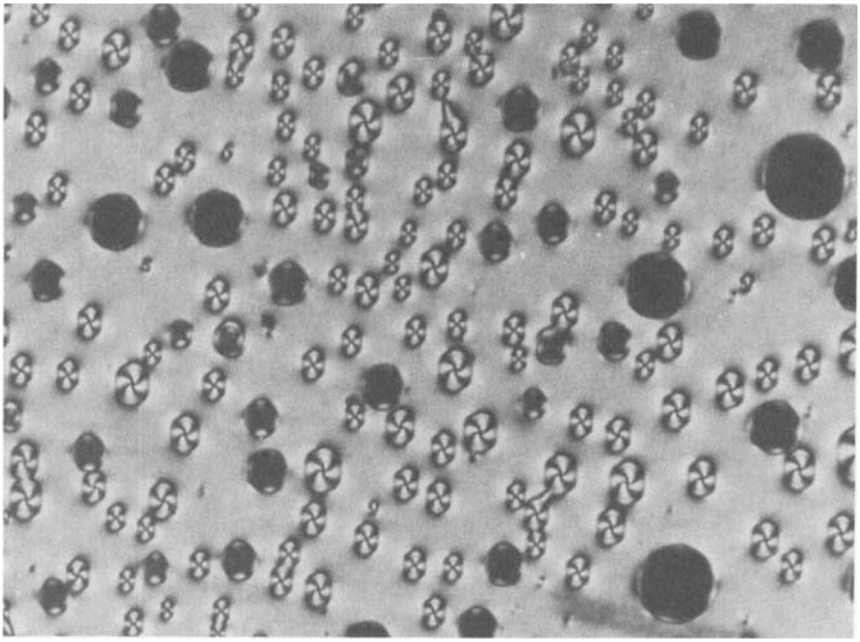
(a)



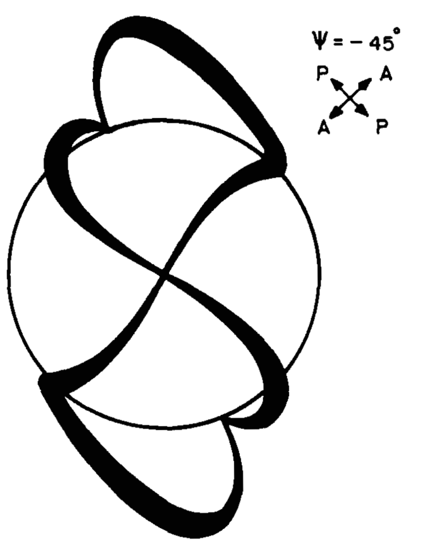
(b)

FIGURE 6a A different region of the same sample as in Figure 5, crossed linear polarizers with $\psi \simeq -10°$ (× 660).

FIGURE 6b Schematic pattern of dark brushes obtained from Figure 4 for $\psi = -10°$.



(a)



(b)

FIGURE 7a Same sample area as in Figure 6a, photographed between crossed linear polarizers with $\psi \approx -45°$ (× 660).

FIGURE 7b Schematic pattern of dark brushes obtained from Figure 4 for $\psi = -45°$.

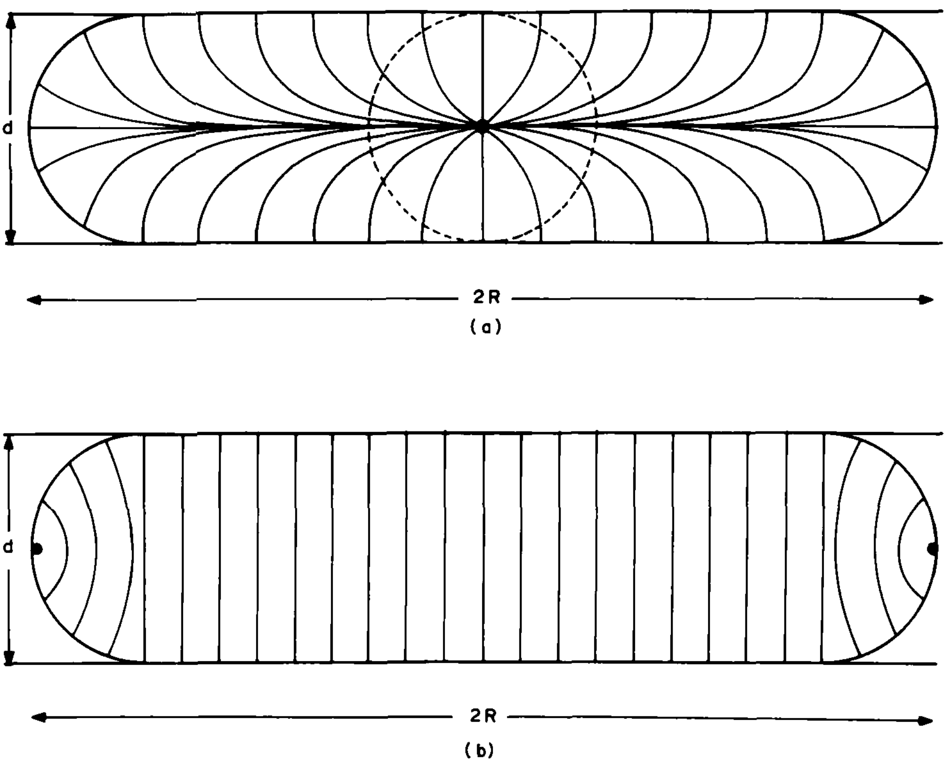


FIGURE 8a Vertical cross section of the director configuration of an N_R drop with a point defect of strength +1 in the center, with the director oriented homeotropically on the glass plates.

FIGURE 8b Vertical cross section of the director configuration of an N_R drop with a singular ring defect of strength $+\frac{1}{2}$ situated on the periphery of the drop.

For the drop with a point defect of strength +1 (Figure 8a) we can expect the region in the immediate vicinity of the point defect to have a simple radial configuration. Assuming that this extends over a radius or $\simeq d/2$, the energy of this part is $8\pi k\ d/2$ ignoring the core energy. Outside this region, the configuration should be predominantly determined by the requirement of matching both the distortion due to the point defect and to satisfy the boundary conditions. We can expect the director orientation to be approximately given by $\theta = \pi z/d$ where z varies from $-d/2$ to $+d/2$. The energy of this part is $\simeq k/2(\pi/d)^2\pi[R^2 - (d/2)^2]d$, where R is the radius of the N_R drop. The total distortion energy is then

$$E_{(\text{point})} \simeq 4\pi kd + \pi^3 k(R^2 - d^2/4)/(2d) \tag{1}$$

The contribution from the splay distortion in the region of $r = d/2$ to R is very small and we have ignored it.

For the configuration with the $+\frac{1}{2}$ ring defect (Figure 8b), the energy is approximately given by

$$E_{(\text{ring})} \simeq \pi k(\tfrac{1}{2})^2(2\pi R)\ln(d/2r_0) \tag{2}$$

again ignoring the core energy. The length of the line defect is $2\pi R$ and its strength $S = +\frac{1}{2}$. The distortion due to the defect extends over a distance $\simeq d/2$ and most of the distortion around the singular line is confined within the N_R drop. r_0 is the radius of the core of the defect.

Using the above crude approximations, it is clear that $E_{(\text{ring})} \propto R$ while $E_{(\text{point})}$ depends quadratically on R. When R is just larger than $d/2$, $E_{(\text{point})}$ can be smaller than $E_{(\text{ring})}$ for a sufficiently large value of d/r_0. For example, if $r_0 = 100$ Å and $d > 3$ μm, $E_{(\text{point})} < E_{(\text{ring})}$ for some range $R_1 > R > R_2$ (where $R_2 \simeq d/2$). For $R > R_1$, the ring defect carries the lower energy. (For $d = 3$ μm $R_1 = 3.215$ μm). This trend is in broad agreement with experimental observations.

The relative stability of the point and ring defects in a nematic sample has been discussed by Mori and Nakanishi.[5] Assuming the core to be in the isotropic phase, the core energy of the line defect is considerably higher than that of the point defect. Including the core energies it is clear that there is a reduction in the critical ratio (d/r_0) for which $E_{(\text{point})} < E_{(\text{ring})}$ for $R \simeq d/2$. Very recently it has been suggested that if the core region of a $+\frac{1}{2}$ line defect is characterized by biaxial symmetry, a ring defect with a diameter of a few 100 Å may be more stable than a point defect.[14]

In Figure 1a we see that on the right hand side there are relatively large drops with point defects of strength +1 but drops in the same region having the ring defects are much larger in size. On the other hand on the left hand side the drops with the latter structure are relatively small in size, but the drops with the point defects are still smaller. Based on the earlier discussion, this trend should arise from a decrease in the thickness d of the sample on going from the right hand to the left hand side of the field of view. As the temperature of the sample is reduced, the orientation of the director at the N_R-glass interface acquires a tilt. Simple energy considerations indicate that in such a case the point defects in small N_R droplets should be shifted towards one end, as has been observed (Figure 3b).

In conclusion, the observations reported in this paper demonstrate the close connection between point and ring defects in nematics.

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