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Point Disclinations at a Nematic–Isotropic Liquid Interface†

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Abstract—The optical properties of a thin film of liquid crystal, with one of its surfaces bounded by the isotropic liquid of the same compound, reveal the existence of point defects in the liquid crystal. Particular sample geometries are examined in which the detailed structure of the optical patterns seen with polarized light can be analyzed to develop a qualitative picture of the structure of the liquid crystal layer near the defects. Changes in the optical pattern induced by an electric field are also analyzed. The observations indicate that the defects are point disclinations at the surface of the liquid crystal, of the kind recently described by deGennes, rather than line disclinations emerging from the liquid crystal.

Recently, P. G. deGennes has proposed the existence of regular arrays of point disclinations at the surface of a nematic liquid crystal in a magnetic field.[1] The idea of point disclinations in the center of nematic droplets, or at opposite points on their surface, is also a familiar one, and is supported by some observations by the author and others. However, point disclinations have not been studied nearly as much as the familiar line disclinations of nematic and smectic liquid crystals. In this paper, experiments are described in which point disclinations are observed in especially favorable circumstances which allow for detailed study of their properties. Although regular arrays have not been seen, the kinds of point defects described by deGennes have been confirmed to exist.

To obtain point defects of the kind described by deGennes, it is necessary to have certain conditions at the sample surfaces. Most important is a "free" surface, at which the optical axis of the nematic is free to orient itself in any direction parallel to the surface.

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In these experiments, the free surface was the interface between the nematic and isotropic phases, which exists at the nematic clearing temperature. Second, it is desirable that the nematic optical axis be normal to the other surfaces involved, for two reasons. First, this gives the sample some useful optical properties, and second, it eliminates line disclinations from the system, since they cannot easily attach to a surface with this boundary condition.

The normal boundary condition was achieved in two ways. The material studied was ethyl *p*-(*p*-methoxy bezylidene amino) cinnamate, which aligns normal to glass or tin oxide surfaces cleaned with hot chromic acid. This was determined by the observation that this material forms a homeotropic texture between glass slides. This material also aligns normal to its nematic–air interface, which was again determined by the appearance of a homeotropic texture in a thin droplet on a cleaned glass surface.

Using these boundary conditions, two sample configurations were studied. Most observations were made using a cell consisting of two tin oxide coated microscope slides separated by a five mil Teflon spacer. This cell was heated from the lower surface on a Kofler microscope hot stage, and the temperature was adjusted so that the nematic–isotropic liquid interface was maintained between and parallel to the two surfaces. Since the temperature difference between the slides was about one degree centigrade, it was fairly easy to maintain the interface during observation. As the temperature drifted higher, the interface would appear at the lower glass surface, and move up through the layer until it met the upper surface. This movement could be followed by refocusing the microscope to keep the interface in sharp focus. This and other qualitative observations, like the mobility of point defects, made it clear that an interface was being observed, and not merely a temperature dependent change of orientation at the glass surface. The second configuration studied was simply a droplet of material on a clean glass plate, again heated from below, so that a lens-shaped cap of nematic was obtained between isotropic liquid and air interfaces. In this case, one could clearly see the isotropic layer, the edge of which showed around the nematic cap.

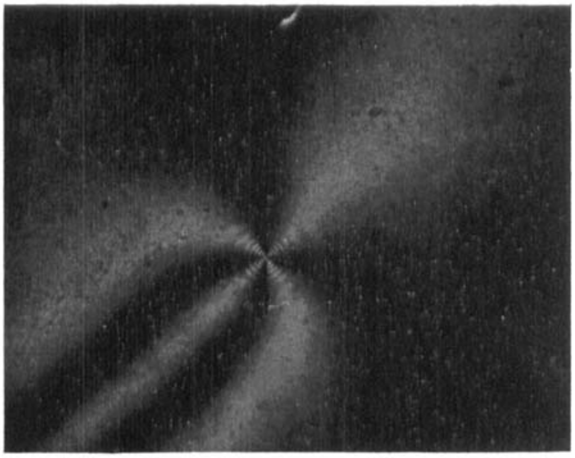
It was determined by the following observations that the nematic axis was locally parallel to the nematic–isotropic liquid interface.

First, with this interface present, the homeotropic texture disappears. Second, rank 1 disclination lines (around which there is a net rotation of π of the nematic axis) were observed to end at this interface without attaching to any surface disclination line. The first observation indicates that the optical axis is either parallel or at some oblique angle to the interface. The second observation eliminates the possibility of an oblique angle, on topological grounds. The topological argument is the following. Consider a rank 1 line emerging from the liquid crystal and ending at an interface to which the nematic axis is titled at an oblique angle. Following a path in the surface, around the end of the line, the direction of the tilt of the nematic axis changes so that it is exactly reversed at the end of one circuit, and cannot connect continuously with the nematic axis at the beginning of the circuit. This implies that there must be a surface disclination present, separating regions of opposite tilt. Since no surface disclination was seen in these samples, the nematic axis must be strictly parallel to the surface. This is the same argument that excludes rank one lines from smectic C phases.

The typical appearance of point disclinations in the two kinds of sample, viewed between crossed linear polarizers is shown in Figs. 1, 2, and 3. The two important elements of these pictures are the cross and concentric rings. The patterns bear some resemblance to the conoscopic figure obtained by looking down the axis of a uniaxial crystal. In fact, the origins of the patterns are similar, with the important difference that the point disclination pattern appears in the image of the liquid crystal interface, while the conoscopic figure appears in wave vector space, rather than in a real image.

Figure 2 is the easiest to analyze. A qualitative sketch of the internal structure of the sample, consistent with its boundary conditions and optical properties, is shown in Fig. 4. It contains a single point disclination at the center of the nematic–isotropic interface. This is always observed in samples of this kind. Topologically, the boundary conditions require the existence of some kind of disclination in the structure, and this appears to be the most stable one. In analyzing the optical properties of this and other structures, one important restriction is made; all light is assumed to be propagating normal to the plane of the sample. In practice, this means that collimated rather than highly convergent illumination was used.

(a)



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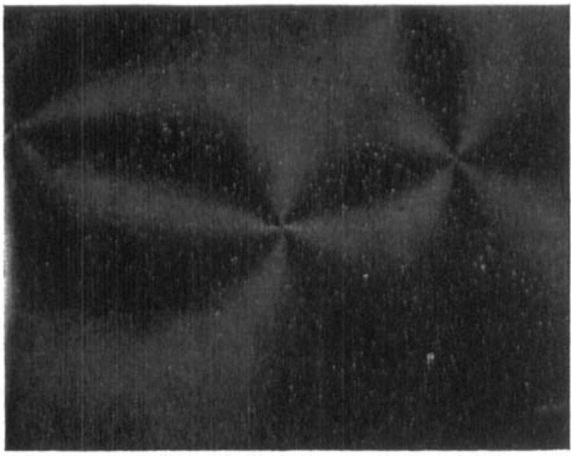


Figure 1. Point disclinations observed between crossed linear polarizers. (Mag. 66 ×)

a. A +2 point, using monochromatic light (5874 Å).

b. A +2 point (near the center) and a −2 point, in white light.

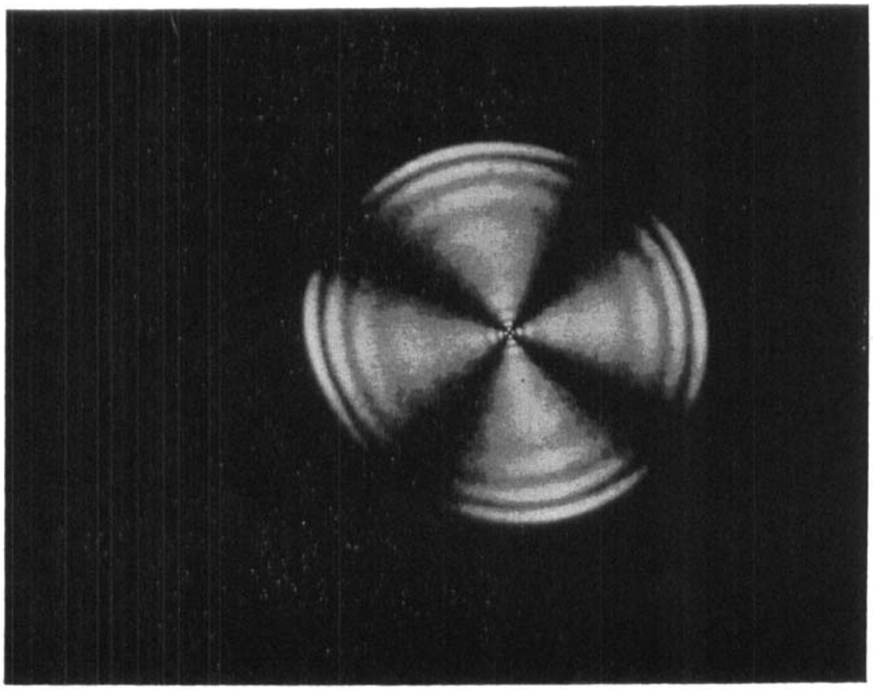


Figure 2. Point disclination in a droplet sample, observed with white light, between crossed linear polarizers. (Mag. 100 ×)

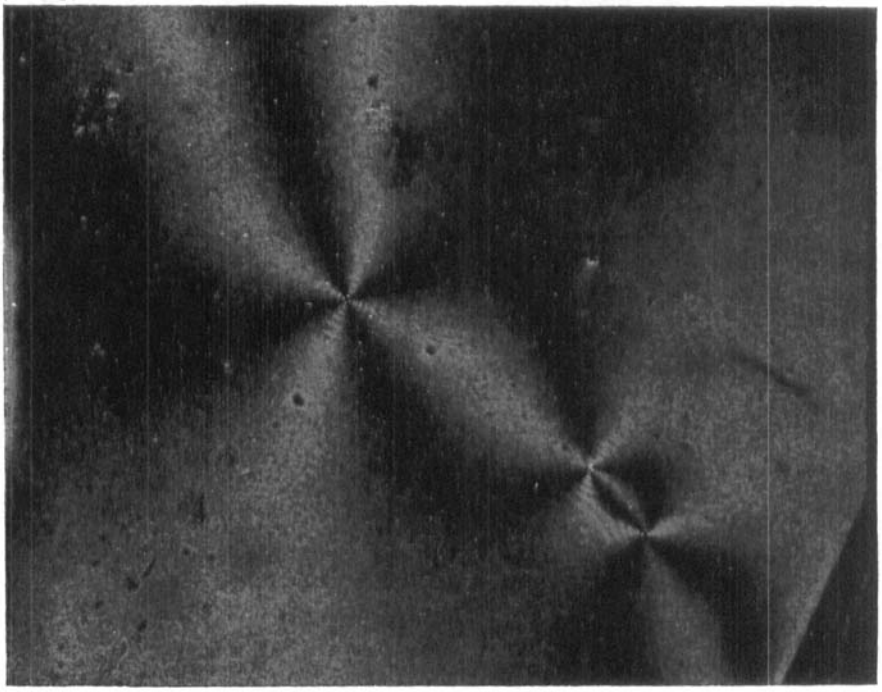


Figure 3. A pair about to be annihilated in the presence of another +2 point. (Mag. 100 ×)

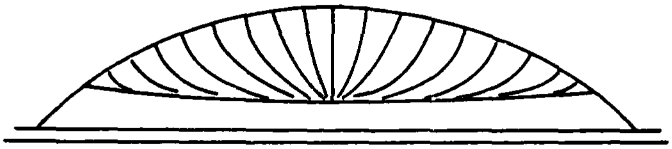


Figure 4. The internal structure of a droplet sample, which is symmetrical about a vertical line through the point disclination in the center of the sample. The curving lines indicate the direction of the optical axis in the nematic portion of the sample.

The extinction cross indicates where the light propagates as pure ordinary or pure extraordinary waves, that is, where the optical axis at the nematic–isotropic interface is parallel to either the polarizer or the analyzer. By observing the motion of the extinction arms while rotating the polarizer and analyzer together, one can map the optical axis over the whole interface.

In regions between the extinction arms, the concentric rings result from changes in the total phase difference between ordinary and extraordinary waves. Explicitly, at a point **r** in the pattern, this phase difference is given by

$$\Delta\phi(\mathbf{r}) = \frac{2\pi}{\lambda}\int (n_e - n_o)\,\mathrm{d}z$$

in which the integral is taken between the sample surfaces, along a path parallel to the optical wave vector and passing through point **r**. Ignoring some small refractive effects, this path is simply perpendicular to the glass plates. The ordinary index, n_o, is a constant, but the extraordinary index, n_e, is a function of the angle between the nematic axis and the wave vector, and this angle is a function of position in the sample, which in theory can be calculated.

In these experiments, because of the temperature gradient, among other things, a detailed comparison of theory with the observed patterns may not be worthwhile. However, a qualitative analysis is easily made from the above equation for $\Delta\phi$. At any point **r** for which

$$\Delta\phi = 2\pi m,\ m = \text{integer},$$

extinction is obtained between crossed polarizers. Referring to Fig. 4, it is clear that $\Delta\phi = 0$ at the point disclination, since the optical axis is vertical below that point, and $n_o = n_e$ along the whole path of integration. With increasing radius from the disclination,

$\Delta\phi$ increases continuously as the curvature of the optical axis increases. In the droplet case, $\Delta\phi$ decreases again near the edges of the sample as the thickness of the nematic layer decreases. These changes in $\Delta\phi$ produce two series of rings, one around the disclination, and one near the sample edges. The maximum value of m in Fig. 2 is about four.

The same kind of analysis accounts for the patterns in the other photographs. In the thin film sample, with a large surface area, there are usually a large number of point disclinations present simultaneously. Rotating the polarizers reveals two main types of points, designated by indices $+2$ and -2, in Frank's notation.[2] The extinction arms of the $+2$ points rotate with the polarizers, while those of the -2 points rotate the opposite way. The orientational structure of the nematic layer near the points is shown in Fig. 5. As

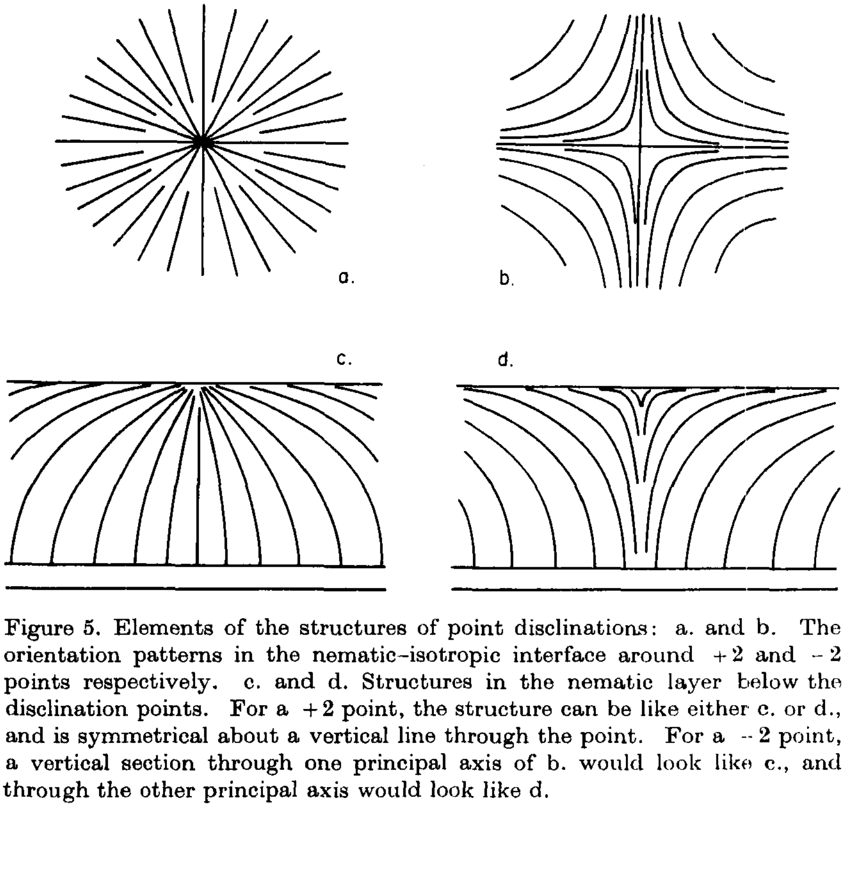


Figure 5. Elements of the structures of point disclinations: a. and b. The orientation patterns in the nematic-isotropic interface around $+2$ and -2 points respectively. c. and d. Structures in the nematic layer below the disclination points. For a $+2$ point, the structure can be like either c. or d., and is symmetrical about a vertical line through the point. For a -2 point, a vertical section through one principal axis of b. would look like c., and through the other principal axis would look like d.

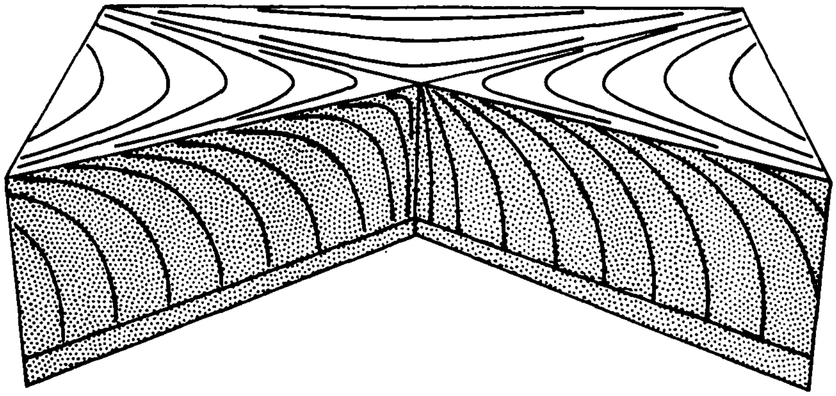
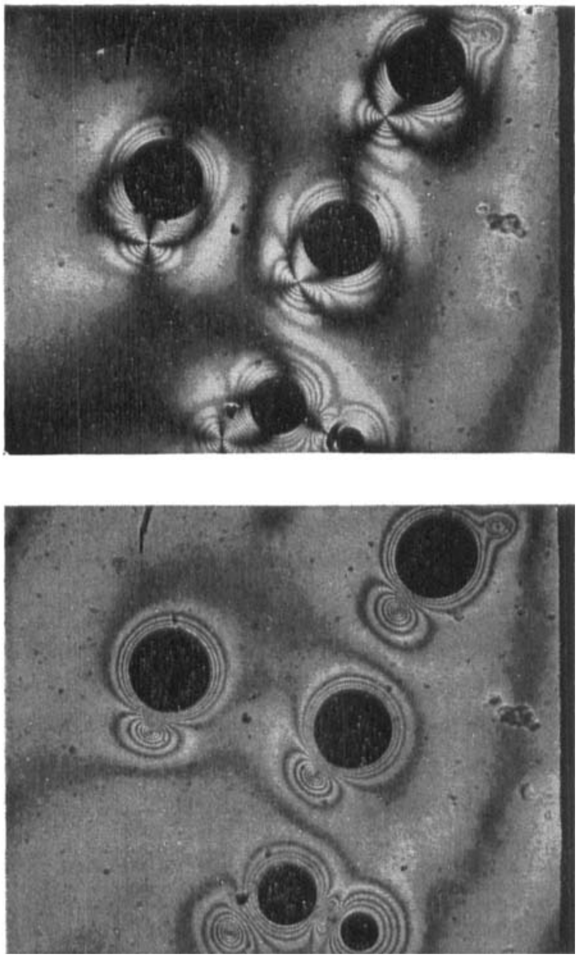


Figure 6. A three-dimensional view of the structure around a -2 point. The shaded areas are vertical sections.

deGennes indicated, there are two possible kinds of $+2$ points, and one kind of -2, containing a complex internal structure, as shown in Fig. 6. The points all produce qualitatively the same optical pattern, with cross and concentric rings. In each structure, $\Delta\phi = 0$ at the disclination, and increases with radius from it. The maximum value of $\Delta\phi$ depends on the thickness of the nematic layer, which changes as the temperature drifts. When the layer becomes very thin, so that $\Delta\phi$ is about 2π or less everywhere, some beautiful birefringence color patterns are seen, similar to those produced by an electric field, described later. For the -2 structure, one might expect a slight elliptical or rectangular distortion of the concentric rings, but none was evident in these observations. In these experiments, the two kinds of $+2$ points were not resolved, and the detailed structure of the -2 points was not confirmed. Qualitative checks of these properties of the disclinations could possibly be made by slight titling of the sample (avoiding any flow). The rings in the different $+2$ patterns would shift in opposite directions relative to the tilt, while the -2 pattern would undergo more complex distortions.

When several points are present simultaneously, they interact with one another, points of opposite sign attracting each other as would be expected from elasticity theory. When a pair of points of opposite sign meet, they annihilate one another. A pair about to be annihilated is seen in Fig. 3. Points can also be created in pairs by turbulence in the sample. An easy way to generate this turbulence



(a)

(b)

Figure 7. An array of bubble-disclination dipoles, observed with monochromatic light. (Mag. 66 ×)

a. between crossed linear polarizers, and
b. between crossed circular polarizers.

is to apply momentarily a large d.c. voltage ($\sim$100 V) across the tin oxide electrodes of a sample. This generates hundreds of disclinations per square millimeter, which rapidly recombine, leaving a few widely separated points. The fact that points appear and disappear in pairs is one of the topological conservation rules for arrays of points in the interface. Another example is the following: in a disc-like sample with an air boundary to which the nematic axis is perpendicular, the sum of the indices of all the points in the interface is $+2$. If there are any line disclinations ending at points in the interface, the indices of these points must be included in the sum. The simplest case of this topological conservation rule is shown in the droplet sample, containing a single $+2$ point.

In a multiply connected sample, i.e., one with some air bubbles making holes in the nematic–isotropic interface, an interesting situation develops. Each air bubble acts topologically as a $+2$ point, making it possible to have a sample with one air bubble and no disclinations, satisfying the above conservation rule. With more than one bubble present, a -2 point is needed to cancel each bubble. The point is attracted to the bubble, and sits at some equilibrium distance from it, forming a stable dipole pair. Figure 7 shows an array of these dipoles. A qualitative sketch of the structure around a typical dipole pair is shown in Fig. 8. At the surface of the air bubble, $\Delta\phi$ reaches a maximum, producing a series of concentric rings around the bubble. The curving extinction arms again map the nematic axis over the interface.

To study the ring pattern due to $\Delta\phi$ without interference from the extinction crosses, one can use circular polarizers, as shown in Fig. 7(b). The meaning of $\Delta\phi$ using circular polarizers is the same as that for linear polarizers. The circular polarization simply provides ordinary and extraordinary waves of equal amplitude and the same initial phase relationship at each point in the pattern, which eliminates the extinction crosses. Between " crossed " circular polarizers, a phase difference $\Delta\phi = 2\pi m$ again produces extinction rings for integral values of m.

In an a.c. electric field, the optical axis of this material tends to align parallel to the field direction. The effects observed were independent of frequency throughout the audio range. This alignment effect provided another means of studying the disclinations.

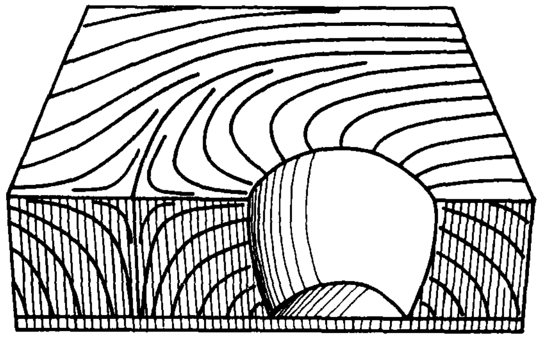
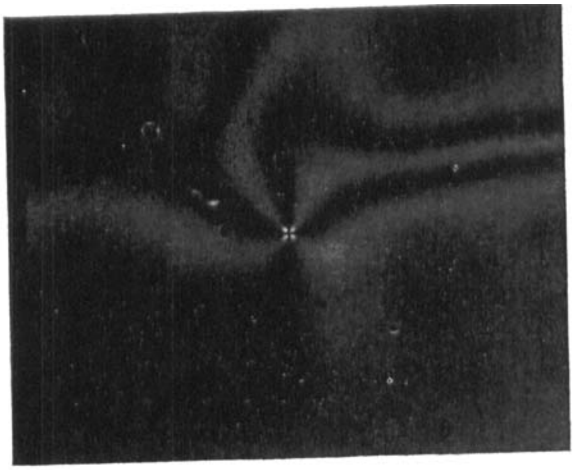


Figure 8. The structure around a typical bubble-disclination dipole, showing the orientation patterns in the nematic–isotropic interface, and in a vertical section through the disclination (shaded).

Voltage was applied between the tin oxide electrodes, producing a field parallel to the optical wave vector, and perpendicular to the sample surfaces. The alignment effect compresses all the curvature in the structures into a transition layer near the nematic–isotropic interface, therefore reducing $\Delta\phi$ everywhere. The thickness of the transition layer, and therefore the magnitude of $\Delta\phi$, varies inversely with voltage. As the field is increased, the rings around the disclinations expand, the outer ones disappearing, until $\Delta\phi$ is about 2π or less throughout the sample (except near air bubbles, around which rings contract into a narrow band). In this state (about 20 volts), the sample exhibits dramatic color effects, due to the dispersion of the birefringence. For instance, the sample may be generally bluegreen, except for bright yellow near the disclinations. At higher fields (about 40 volts), even these effects disappear as $\Delta\phi$ becomes much less that 2π, and only the extinction arms remain against a gray background, with their sharp crosses locating the disclinations. The typical appearance of samples in an electric field is shown in Fig. 9. Again, in a better defined system, one could calculate the details of these patterns from continuum elasticity and dielectric theory. Turbulant effects due to conductive instabilities do not occur at the voltages used here.

The electric field effects help to prove one essential feature of the structures described here. That is that $\Delta\phi = 0$ at the disclination, implying that the nematic axis is vertical below that point. This

(a)



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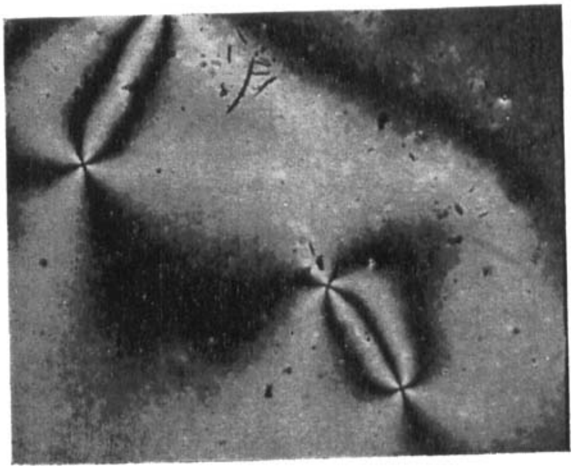

Figure 9. Disclinations in an electric field. (Mag. 66 ×)

a. A field in the range of maximum chromatic effects.

b. A slightly higher field at which chromatic effects have almost disappeared.

rules out a possible alternate explanation of the optical patterns, involving a line disclination running vertically from the glass to the isotropic liquid, as shown in Fig. 10. This would produce a maximum in $\Delta\phi$ at the center of a ring pattern. However, this ring pattern would shrink around the point as the electric field was increased, just as the rings shrink toward air bubbles. Another qualitative observation supporting the electric field effect evidence is the appearance of white light fringes around the disclinations. In these observations, the smallest ring around a point was very sharp and almost achromatic, while the larger rings showed increasing separation of hues, leading to a fading of contrast after about four orders. White light fringes would appear this way around a point at which $\Delta\phi = 0$, not around a maximum in $\Delta\phi$.

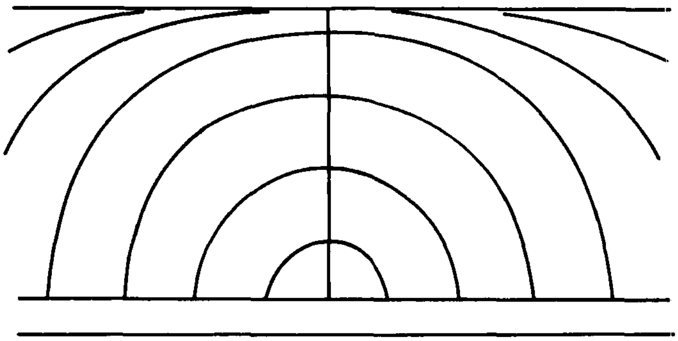

Figure 10. A possible structure, containing a vertical line disclination. The structure is symmetrical about the line. Although it would produce a pattern of rings around a +2 point, this structure is not consistent with the observation that $\Delta\phi = 0$ at the disclination point.

Perhaps it is possible to construct an explanation of these observations in terms of line disclinations. However, there were never any visible lines associated with the points described here, and it appears difficult to devise an explanation of the optical properties of the sample in terms of a vertical line at each point. On *a priori* grounds, point disclinations should be expected to exist under the proper conditions, such as those described here. There may be a historical bias toward explaining all observations similar to these in terms of line disclinations, but the author's reaction to that is to suggest that some of the past observations be reexamined to see if they could be equally well explained by point disclinations. In making the distinction between lines and points, the behavior of the birefringence pattern ($\Delta\phi$) near the singularities should be especially useful.

Acknowledgements

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