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E. F. Carr[a]
[a] Physics Department, University of Maine, Orono, Maine, U.S.A.

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Optical rotation at the free surface of a nematic liquid crystal

by E. F. CARR
Physics Department, University of Maine, Orono, Maine 04469, U.S.A.

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When a small droplet of a nematic liquid crystal is placed on a horizontal glass plate in the presence of a magnetic field, the plane of polarization of light transmitted upward through the liquid crystal can be rotated. A defect usually forms preferring a direction perpendicular to the magnetic field and forming a diagonal of the droplet. This defect divides the free surface into regions giving rise to optical rotations that are clockwise and counterclockwise. It is suggested that the defect may be similar to a Helfrich splay-bend wall and the optical rotation in the regions near the defect may be explained by surface effects at the free surface of a liquid crystal.

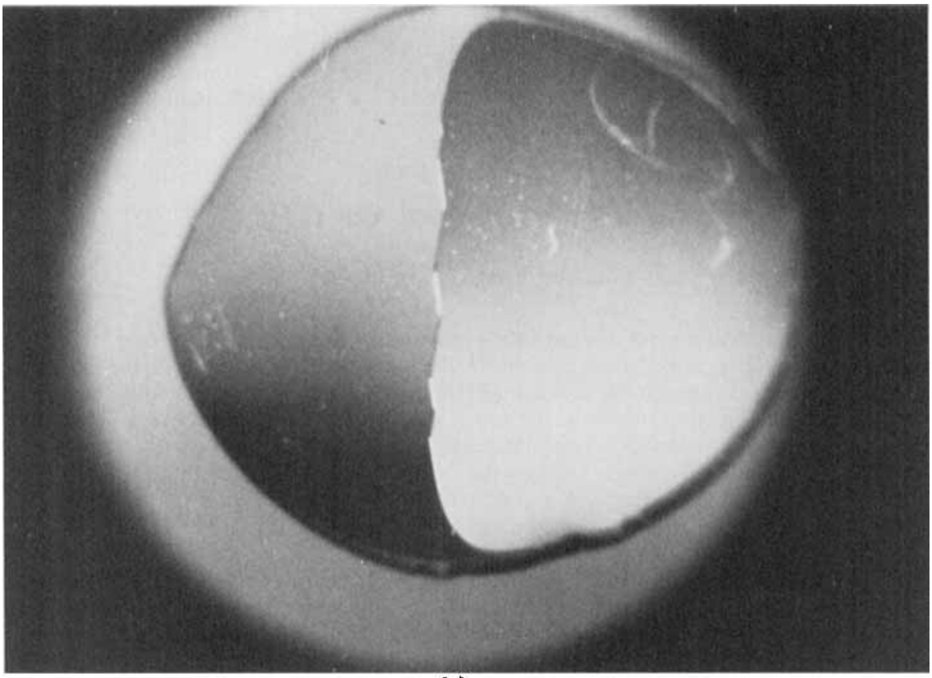
1. Introduction

Work by Williams [1, 2] showed a rotation of the plane of polarization of light when transmitted upward through the free surface of the nematic liquid crystal p-azoxyanisole (PAA). The effect was only observed in the presence of a magnetic field and there were two regions that exhibited optical rotary power of equal magnitude, but opposite sign. Williams suggested the rotation was a bulk property, but de Gennes [3] suggested it might be a surface effect. Meyerhofer, Sussman and Williams [4] made similar observations on *p* methoxy-benzylidene-*n*-butylaniline (MBBA). They proposed a model involving surface effects based on ideas of de Gennes [3].

The observations reported here are similar to those reported earlier [1, 2, 4] in that there are optical rotations of equal magnitudes but opposite sign. Although the experimental setup and patterns reported here are different from earlier work, some of the explanation of the results is similar to that discussed by Meyerhofer, Sussman and Williams [4].

2. Experimental

A small drop of nematic material MBBA was placed on a horizontal glass plate in the presence of a magnetic field. White light, polarized perpendicular to the field, was transmitted upward through the sample. Observations were made vertically above the sample (except figure 3) with a microscope using a sheet of polaroid as an analyser. The diameter of the droplet was approximately 3 mm. Since the free surface behaved much like a spherical surface, observations of points on the free surface, other than the centre, were made at an angle other than 90° with the surface. Before depositing the droplet on the glass the glass surface was rubbed in a direction parallel to the magnetic field. For the best results the sample was heated and cooled from the normal liquid phase in the presence of a magnetic field. All observations on MBBA were made at room temperature.



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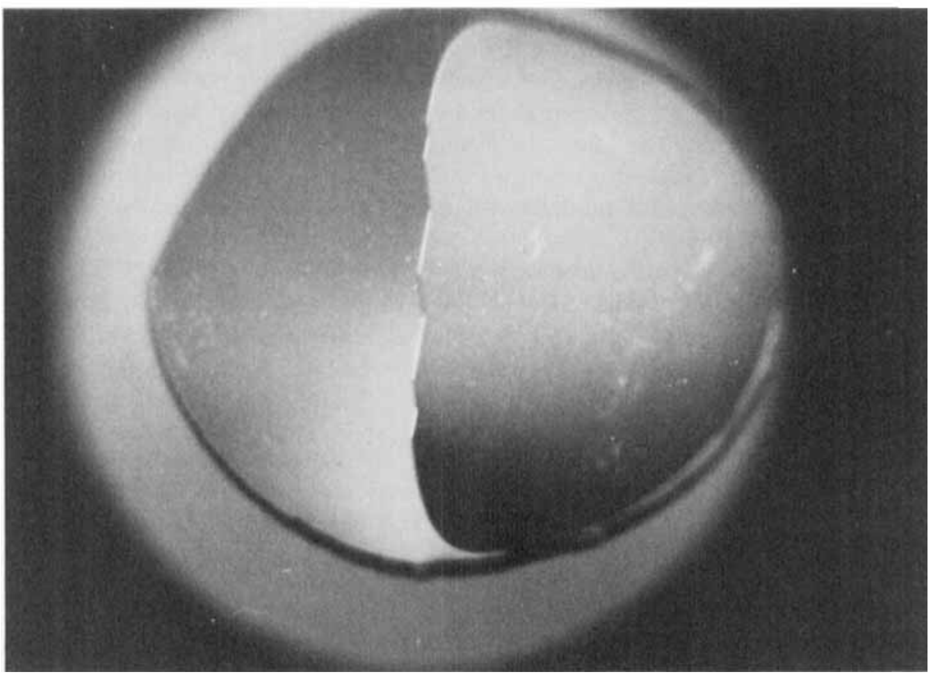


(*b*)

Figure 1. Vertical observation of a droplet of MBBA placed on a horizontal glass plate in the presence of a 0·4 T horizontal magnetic field. The diameter of the droplet was 3 mm and the light source, directed upward, was from an incandescent lamp. The polarizer was perpendicular to the magnetic field and the analyzer was oriented in (*a*) at an angle of +20° and in (*b*) at an angle of −20° relative to a crossed position of the polarizer and analyzer.

3. Results

A photograph of the free surface of a droplet of MBBA in the presence of a 0·4 T (4000 Gauss) magnetic field is shown in figure 1. A defect preferring a direction perpendicular to the magnetic field can be easily seen. The analyser was rotated $+20°$ in figure 1 (*a*) and $-20°$ in figure 1 (*b*) from a cross position of the polarizer and analyser. An examination of the photographs shows that the darker regions tend to become lighter and the lighter regions tend to become darker as the analyser is rotated from $+20°$ to $-20°$. For the analyser parallel to the magnetic field (0°) all regions were equally dark. This implies that for points on opposite sides but near the defect, the optical rotations are of equal magnitude but opposite sign. These photographs indicate optical rotations (clockwise and counterclockwise) that vary with position on the droplet and appear to have some dependence on the curvature of the free surface. Since the contrast between regions appears to vanish at the centre of the droplet which is the thickest part of the sample, we can conclude that this is not a bulk effect.

For an explanation of the results shown in figure 1 we refer to figure 2 which is similar to a model presented by Meyerhofer [4] *et al.* based on ideas of de Gennes [3]. For the present explanation we assume that there is only one defect forming a diagonal of the droplet and it prefers a direction perpendicular to the magnetic field. In figure 2 the lines indicate the direction of the director and the top of the diagram represents the free surface. If we assume [3] that the director at the free surface is perpendicular to the surface and below the surface it is parallel to the magnetic field, a defect can form as shown. Polarized light propagating perpendicular to the free surface will not experience a rotation, but if the light propagates perpendicular to the magnetic field at a small angle ϕ with respect to the plane of the diagram, a rotation should occur. At this angle a change of the director appears like a small twist in the alignment. One side of the defect should rotate the plane of polarization in one direction and the other in the opposite direction. If the angle the beam makes with the

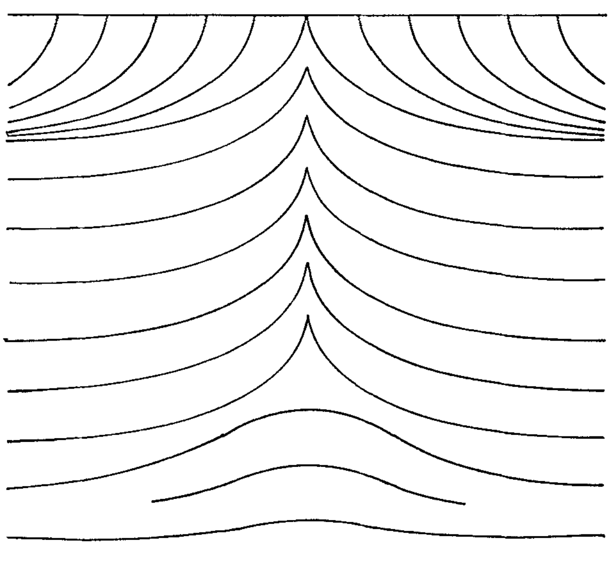


Figure 2. Schematic diagram for molecular alignment. The top horizontal line represents the free surface. Other lines indicate the direction of the director.

plane of the diagram is changed from $+\phi$ to $-\phi$ the direction of the twist should change resulting in a change in the sign of the optical rotation.

If we divide the droplet into four quadrants we notice the optical rotation changes from clockwise to counterclockwise as we pass from the upper right to the upper left quadrant. Also the rotation changes from counterclockwise to clockwise as we pass from the lower right to lower left quadrant. The changes in rotation can be explained by passing from the right to the left side of the defect as illustrated in figure 2.

The optical rotation changes gradually from the upper right to the lower right quadrant and also from the upper left to the lower left quadrant. These changes can be explained by changing the angle ϕ (angle the light beam makes with plane of diagram in figure 2) gradually. In figure 2 we assumed the direction of the light beam is changed, but in the work discussed here we are changing the direction of the free surface rather than the direction of the light beam.

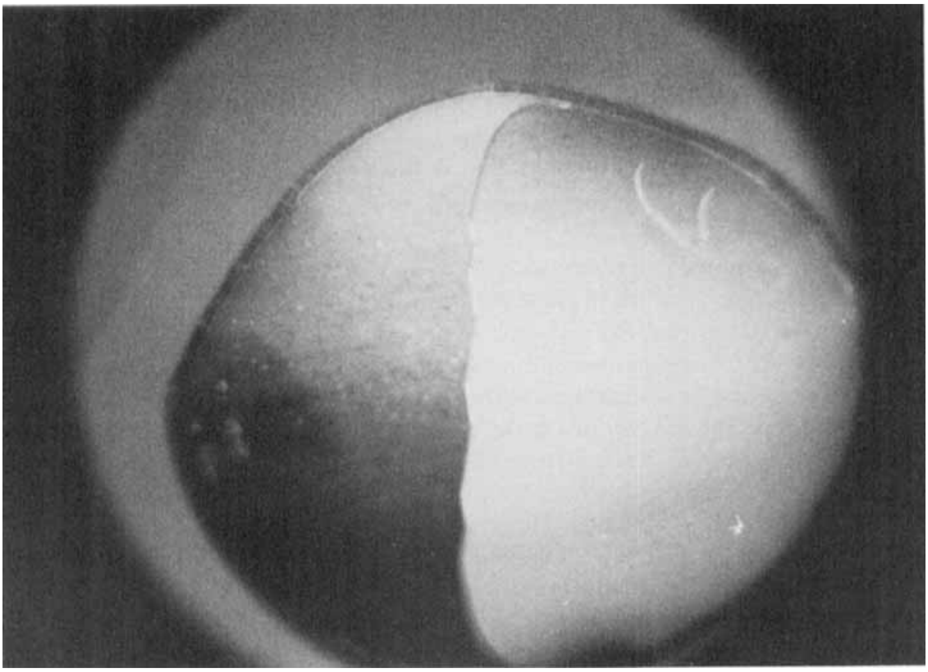
The area within the defect in figure 1 (*a*) also shows light and dark regions. The defect is made up of straight sections that alternate light and dark and form well defined angles. As the analyser is rotated (figure 1 (*b*)) from $+20°$ to $-20°$ the light sections become dark and the dark sections become light. To understand this change we may view the defect as similar to a Helfrich [5] splay-bend wall perpendicular to a magnetic field. As the magnetic field was increased we observed that the walls became thinner which is consistent with the behaviour of a Helfrich wall. One basic difference is that we assume the director within the wall to have a slight twist associated with it. This can give rise to an optical rotation at the wall that can be either clockwise or counterclockwise. When walls with clockwise and counterclockwise rotations join we have a change from dark to light. The angle formed by the walls is usually 160–165 degrees and does not show a field dependence as the field is varied from 0·15 T to 0·60 T.

If a sample, which shows a pattern similar to that of figure 1, is rotated by an angle of less than 90 degrees about a vertical axis, a torque due to the presence of the magnetic field will rotate the defect back to a direction perpendicular to the field. If we assumed this to be a Helfrich type wall, the energy associated with the magnetic field should be a minimum with the wall perpendicular to the field.

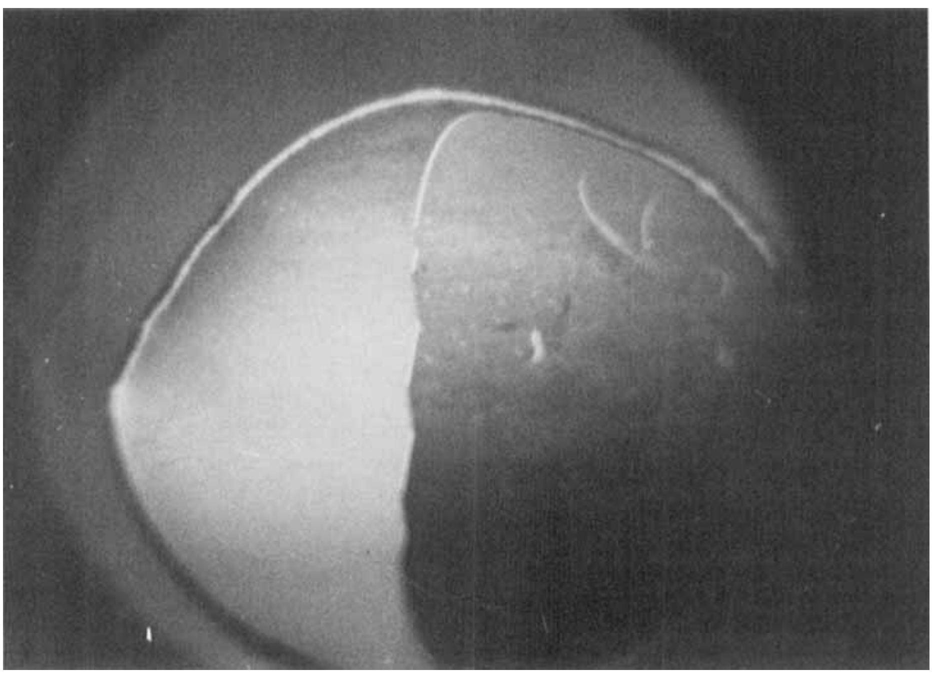
The defect shown in figure 1 does not extend to the bottom of the sample and may not extend [6] much more than a wavelength below the free surface. To support this statement a suspended film of MBBA was stretched over a small hole (3 mm diameter) which formed two free surfaces. Defects similar to those in figure 1 were observed on both top and bottom surfaces. In order to observe the effects due to the defect on the bottom surface, it was necessary to rotate the polarizer instead of the analyser. It was clear that one defect was not an extension of the other. Meyerhofer *et al.* made similar observations on suspended films. Though the appearance of their defects are different from that shown in figure 1, we cannot be sure the defects are of a different type. We also investigated defects on free surfaces facing downward by placing a sample on a glass plate and inverting the plate. Inverted samples produced defects like those shown in Figure 1 when cooled from the normal liquid in the presence of a magnetic field.

Since the defect in figure 1 does not appear to extend much below the surface we may wonder if it should be considered a wall or line defect. A defect with straight sections forming a well defined angle has been observed [7] for a somewhat different wall, but we are not aware of such an observation for any line defect.

In figure 1 we did not detect any optical rotation at the centre of the droplet. In figure 3 the direction of observation is at an angle of approximately 15° with the



(a)



(b)

Figure 3. Observations were the same as described in figure 1 except the microscope was oriented at an angle of 15 degrees with the vertical.

vertical. As the analyser is rotated from $+20°$ to $-20°$ (from crossed polarizer and analyser position) the centre of the droplet changes from light to dark and dark to light, indicating optical rotations of both signs. Because of the angle of observation in figure 3, the whole defect was not in focus but it appeared to be similar to that shown in figure 1. Since observations (see figure 1) along the vertical do not show optical rotation at the centre of the droplet, figure 3 provides additional evidence that the optical rotations depend on the angle formed by the light exiting the liquid crystal and the free surface.

The contrast shown in figure 1, as we move parallel to the defect, implies that the optical rotations increase as the angle between the light exiting the liquid crystal and the free surface decreases. We can not give a quantitative measure for the optical rotation because it varies with position and shows some variation with the magnetic field. The optical rotation decreases as the field is increased from 0·15 T to 0·6 T. We should point out that the contrast between dark and light sections of the defect can be improved by using an angle larger than 20° for the rotation of the analyser. The 20° angle was chosen because optical rotations both inside the walls and in the regions outside could be clearly indicated. The best contrast for the regions outside the defect was for a rotation of less than 20 degrees for the analyser.

In figure 2 the assumption was made that the molecules at the free surface were aligned normal to the surface. Bouchiat and Langevin-Cyuchon [8] have reported that for MBBA the director at the free surface makes an angle of about 70° with the plane of the surface. It may be that as a Helfrich type wall is forming, the director inside the wall may have to undergo a small twist in order to maintain the 70° degree angle at the free surface. This twist could be left or right handed which produces the dark and light walls of figure 1.

P-azoxyanisole (PAA) was also investigated but we were unable to observe defects comparable to that shown in figure 1. This is consistent with the observations of Williams [1, 2] in that he did not observe optical rotations that were independent of sample thickness. Meyerhofer *et al.* pointed out that the failure to observe surface defects in PAA may be due to the director alignment at the free surface which has been reported [8, 9] to be parallel to the free surface rather than perpendicular.

In order to produce the patterns of the four dark and light quadrants the droplet was cooled from the normal liquid in the presence of a magnetic field. After several minutes most of the patterns were similar to those of figure 1. Occasionally a defect formed that produced the pattern but the defect did not appear to be identical to the defect of figure 1. The apparent difference is not understood. Patterns similar to those shown here can be obtained by placing a droplet (in the nematic phase) in a magnetic field. The original pattern usually looks similar to that reported by Williams but gradually changes to that produced by a single defect perpendicular to the magnetic field. This defect is very stable and can remain for days (or probably much longer) if the field is left on. The shape will change but a single defect perpendicular to the field will remain.

A number of nematic mixtures have been investigated and the results are comparable to those of MBBA. This author previously published results on MBBA [10] with experimental techniques different from those discussed here and different from those reported previously [1, 2, 4]. At the time of the previous publication [10] this author was not aware of the earlier work, but the results are related to those of Meyerhofer *et al.* and the work reported here. It was reported in the article [10] that optical rotations in *p*-(anisalamino)-phenyl acetate were not obtained, but recent observations have shown that they can be observed for a curved surface.

4. Conclusions

When a small droplet of MBBA is placed on a horizontal glass plate in the presence of a magnetic field, a defect is formed preferring a direction perpendicular to the field and forming a diagonal of the droplet. This defect is very stable. Samples have been left in a magnetic field for several days and the defects have remained although they did undergo small changes in shape. Optical rotations which can be clockwise or counterclockwise can be accounted for by the presence of this defect. If we picture this defect to be similar to a Helfrich type inversion wall perpendicular to a magnetic field, we can use arguments similar to those used by Meyerhofer *et al.*, which are based on an idea of de Gennes, to explain the observations. They assumed that the director is normal to the free surface at the surface but parallel to the magnetic field in the bulk.

The defect shown in figure 1 does not extend to the bottom of the sample and may not extend much more than a wavelength below the surface. The defect appears to be made up of two types of defects connected in such a manner as to form a diagonal of the droplet. Since the director at the free surface has been reported as making an angle of 70 rather than 90 degrees with the surface, the director within the walls may have to undergo a twist when the walls are forming. With two possible directions for the twist the two types of walls can be accounted for.

Although the patterns Williams observed for PAA have some similarity to those observed in this work we believe that the mechanisms responsible are probably different. The defects responsible for changes in optical rotation that were reported by Meyerhofer *et al.* appear to be different from those shown here, but we are not certain.

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