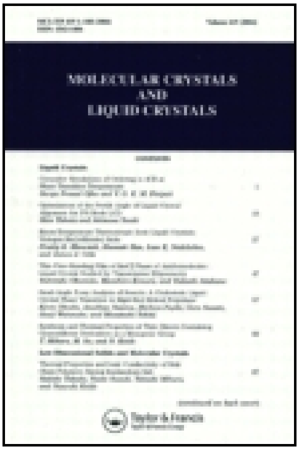

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A New Director Alignment for Droplets of Nematic Liquid Crystal with Low Bend-to-Splay Ratio

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The first observation of a new director orientation within nematic droplets is reported. This orientation consists of the nematic director field arranged concentrically around a central core running through the center of the droplet, with tangential alignment of the nematic at the droplet surface. It is shown that for tangential wall alignments, the observation whether a droplet adopts the axial alignment or the previously reported bipolar droplet alignment is correlated with the k_{33}/k_{11} ratio of the nematic, with a low ratio favoring the axial alignment. A qualitative argument for this preference is given. It is observed that distorting an axial droplet by placing it in a solid polymer host also distorts the director field and central core within the droplet. Droplets in the axial alignment in a polymer film can be forced into the bipolar alignment by the application of an electric field. The nature of the central core defect is discussed, with arguments made that it is a true line defect.

INTRODUCTION

Droplets of nematic liquid crystal suspended in an isotropic liquid have been studied intermittently in the past.[1–6] Recently, nematic droplets have reemerged as a topic of interest due to their application in a new generation of liquid crystal displays and light valves.[7–9] In this paper we described the first observation of a new alignment orientation of a nematic liquid crystal in a droplet, one with an axial line disclination rather than the previously observed bipolar point disclination configuration.[1,2,8] This axial[10] alignment is found in droplets possessing low values (<1.0) of the k_{33}/k_{11} ratio (bend-to-splay elastic constant ratio) and tangential wall alignment of the nematic director.

Two different alignments of nematic liquid crystal suspended in free droplets have previously been observed.[1-3,8] If the nematic director is aligned perpendicularly at the droplet surface boundary, a radial alignment is observed (Figure 1a). This alignment has spherical symmetry[3] with a center point disclination. For a tangential wall alignment, a bipolar arrangement is seen (Figure 1b). This orientation

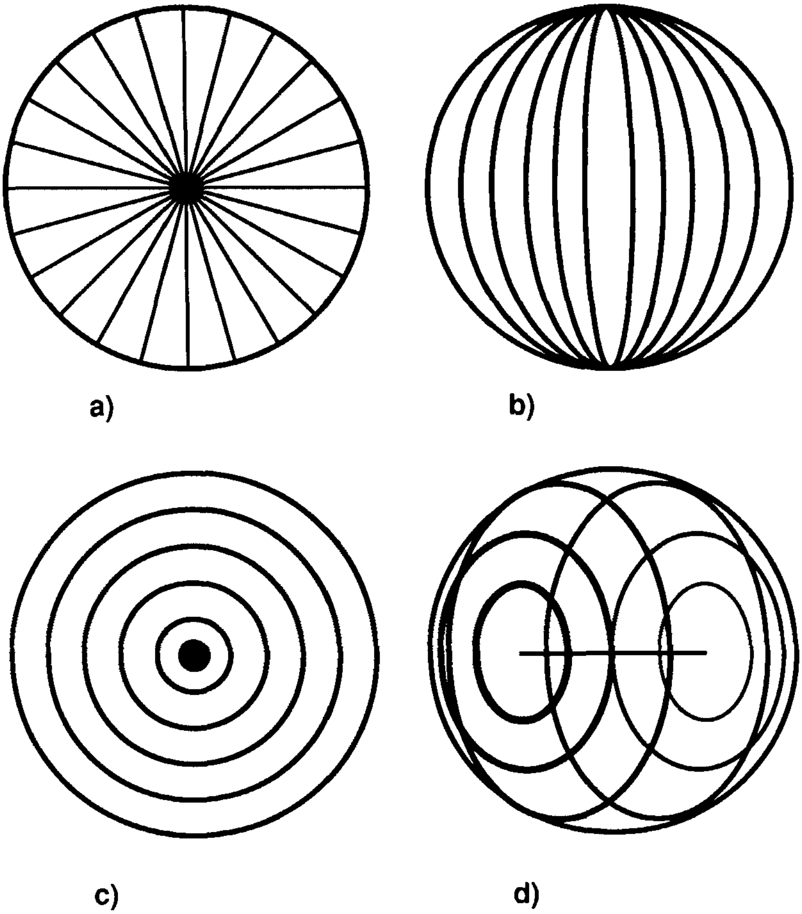


FIGURE 1 Idealized representations of different nematic director configurations within droplets. a) Cross section of radially aligned nematic droplet, possessing a center point defect. b) Cross section of bipolar nematic droplet, possessing two point defects. The view is perpendicular to the droplet symmetry axis. c) Idealized cross section of an axially aligned droplet. The view is parallel to the droplet symmetry axis. d) Three-dimensional representation of an axial alignment of nematic liquid crystal in a droplet. The circles represent the alignment of the nematic director field around the central core, shown as the line running through the center of the droplet.

possesses cylindrical symmetry, with two point disclinations at opposite ends of the droplet. These observed orientations are also the configurations calculated to possess the lowest energy for nematics in droplets with those respective wall orientations, under the assumption that all three elastic constants in the nematic were equal.[5,6] Droplet configurations other than the radial and bipolar types have been observed in nematics doped with cholesteric liquid crystal[2a] and in nematic droplets attached to treated glass slides.[4]

Recently, commercial mixtures of nematic liquid crystals possessing elastic constants with bend-to-splay ratios (k_{33}/k_{11}) less than 1 have become available. We will show that these mixtures, when suspended as droplets in a medium enforcing tangential director wall alignment, take on an axial type alignment. This configuration consists of the nematic director field arranged concentrically around a central line disclination which runs through the center of the droplet (Figures 1c, d). As will be discussed, this orientation possesses qualitatively very little splay deformation, at the cost of enhanced bending of the nematic director, qualitatively supporting the notion that the k_{33}/k_{11} ratio is the controlling factor in determining the nematic configuration. This configuration has been treated theoretically,[5,6] but has always found to be higher in energy than the bipolar configuration under the equal elastic constant approximation. Other topics that will be discussed are the nature of an axially aligned droplet in a solid polymer host, the effect of an electric field on an axially aligned droplet, and the nature of the core defect in an axial droplet.

EXPERIMENTAL

Nematic droplets were created by hand-stirring a mixture of glycerin and liquid crystal (20:1 ratio by weight, respectively). Such emulsions are ultimately unstable, but persist long enough (tens of minutes) to study. In order to unequivocally determine the director orientation within the various droplets of each nematic, each nematic was doped with 0.25% by weight of a blue pleochroic dye (B1, Hoffman-La Roche) prior to mixing with the glycerin. This is a high order parameter pleochroic dye, with its optical transition dipole along the long axis of the molecule.[11] When an individual droplet is viewed through the microscope using linearly polarized light, one can map the director field within the droplet by rotating the polarizer. When the absorption within a portion of the droplet is maximized, the director field is aligned in the same relative direction as the polarization axis of the polarizer. Using this technique, it is not difficult to differentiate the bipolar from the axial configurations.

The clearing point temperature of the dye-doped nematic (measured using a Mettler FP80/FP82 microscope hot stage) was not discernibly different from that of the non-dyed nematic, as expected for such a low concentration of dye dopant. Additionally, nematic eutectic mixtures that spent weeks in contact with glycerin showed no change in their clearing point behavior, indicating that differential solubility of the different constituents of the nematic mixtures in the glycerin is minimal. The lack of change in clearing point behavior gives us confidence that the elastic constants of dye-doped nematic in contact with the glycerin don't differ substantially from those of the neat nematic.

The observation of the bipolar alignment in nematics with high k_{33}/k_{11} ratios shows that glycerin enforces a tangential wall director alignment in those nematics. This tangential alignment is expected to be maintained for all the nematics, as the surface energy of the low k_{33}/k_{11} ratio eutectic mixtures should not be substantially different from the high k_{33}/k_{11} ratio mixtures. Radial alignment was not seen for any nematic dispersed in glycerin, although it could be seen for nematics of both high and low k_{33}/k_{11} ratio if the nematics were dispersed in a liquid of dramatically different surface energy, such as poly(dimethylsiloxane)(PS041.2, Petrarch Systems).

The various nematics were studied for orientation in a room held at 20 (± 1) C. This insured that the nematic mixtures both prior and subsequent to emulsification were at 20 C, the temperature where k_{33}/k_{11} data were available.[12] Additionally, ZLI 1957/5 was studied in a 25 C room since 25 C data was available for it.[12b] To study the nematic droplets, a drop of each stirred nematic/glycerin mixture was placed on a microscope slide, and covered with a glass slip raised with 125 micron spacers. Droplet sizes studied ranged from 5 to greater than 100 microns for each nematic. To insure that the droplet orientation was not being determined by interaction with the glass cell, alignments are reported only for droplets flowing slowly through the field of view of the microscope (unless otherwise noted in the text).

Droplets were observed using an Olympus BH-2 polarizing microscope. An important variable in the microscopic observations is the width of the field diaphragm. A stopped down field diaphragm showed the most structure within a droplet, including interference rings and the core defect (see Figure 2). A wider diaphragm made it somewhat

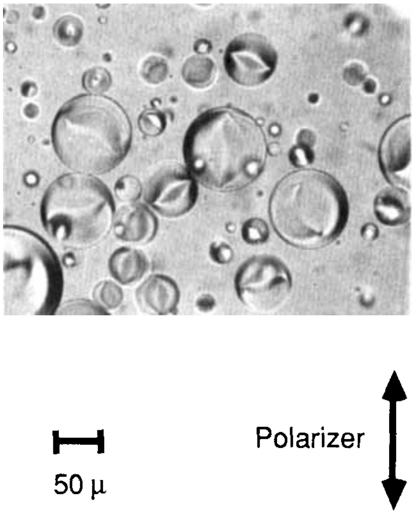


FIGURE 2 Droplets of ZLI 2620 in glycerin, photographed using a stepped down field diaphragm.

easier to observe and photograph other features of the droplets, such as dye absorption (Figure 3) and the behavior of the droplet core under the influence of an electric field (Figure 6). The lack of internal features in the droplets of Figures 3 and 6a compared to Figure 2, is more an indication of differing photographic conditions, rather than differences in internal droplet configuration.

For the study of nematic droplets in a solid film, the nematic was mixed with a 15% aqueous solution of polyvinylalcohol (PVA) (Vinol 205, Air Products). The PVA had previously been deionized by a Soxhlet extraction with methanol. A drop of this emulsion (20:1 weight ratio of PVA:nematic) was then smeared into a thin coating on a piece of indium-tin oxide (ITO) coated glass (Donnelly Mirrors). After drying, this cell was taped to a piece of 50 micron ITO coated glass (Deposition Technologies). This thin glass allowed for microscopic observations at high magnification while an electric field is applied across the nematic/polymer film. The nematic/polymer layer was generally 50 microns or greater in thickness; application of up to a 200 volt 1000 Hz square wave signal was usually sufficient to align all droplets greater than 5 microns in diameter.

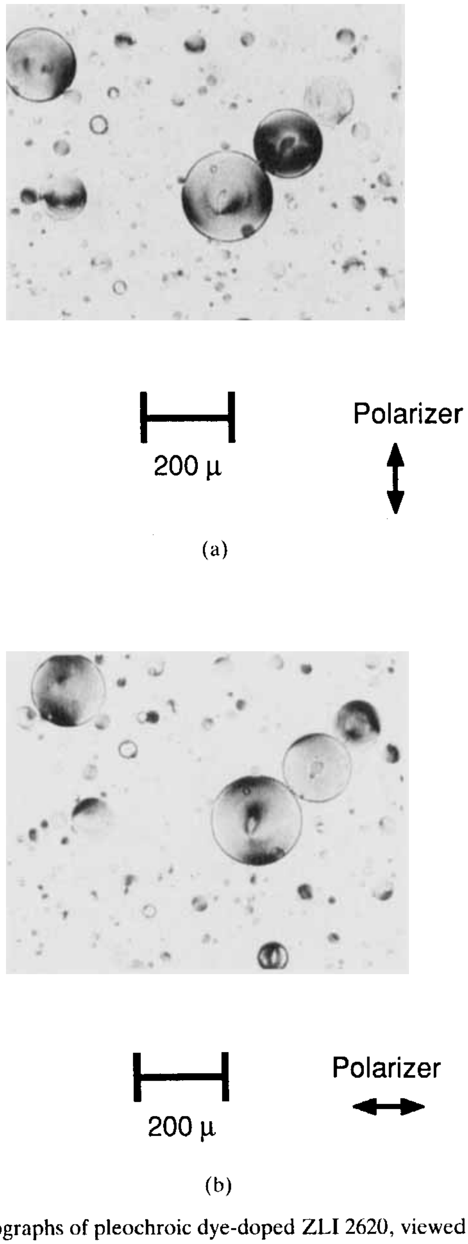


FIGURE 3 Photographs of pleochroic dye-doped ZLI 2620, viewed through a single (top) polarizer.

DETERMINATION OF THE AXIAL ALIGNMENT

Small droplets (<20 microns) possessing the bipolar orientation are readily identified as such under crossed polarizers. Bipolar droplets show an overall two-fold symmetry upon rotation of the microscope stage, with a dark cross pattern appearing every 90° of rotation.[8] However, droplets of nematics possessing a low k_{33}/k_{11} ratio do not show this pattern. No cross pattern is seen, but rather interference rings and colors due to varying birefringence within the droplet are observed. The rings appear to be symmetric about the center of the droplet. Insertion of a full wave (red) plate into the microscope fails to produce sharp, readily identifiable yellow and blue colors within the droplet, as observed in most bipolar droplets.[8] These results indicate that light passing through the droplet does not experience the quasi-uniform director alignment that the bipolar droplets possess, but some orientation with a substantial curvature of the director field, giving the droplet a much greater apparent birefringence.

Figure 2 shows a microphotograph of droplets of ZLI 2620 (EM Industries, $k_{33}/k_{11} = 0.73$) in glycerin, using a single linear polarizer. The patterns seen within the droplets are not observed in bipolar droplets. A central core defect is evident in many of the droplets, along with other features indicative of optical interference effects. While many of the features within these droplets are consistent with an axial alignment, the use of a pleochroic dye in the droplets presents clearer evidence for an axial alignment.

Doping these low k_{33}/k_{11} ratio nematics with a small amount of pleochroic dye shows that these droplets possess a two-fold symmetry axis. Using a single polarizer to map the director field in the dye-doped droplets (*vide supra*), it appears that the nematic director field lies perpendicular to the droplet symmetry axis. In some droplets the symmetry axis was aligned parallel to the field of view; in these droplets the dye absorption indicates that director field is concentrically arranged around the droplet symmetry axis. By observing droplets flowing and rotating within the fluid, it was possible to determine that a given droplet shows both of the above descriptions, depending on the relative angle of the droplet symmetry axis to the viewer.

Figures 3a and 3b show photographs of nematic droplets of ZLI 2620 doped with a pleochroic dye, suspended in glycerin, and viewed using a single (top) linear polarizer. Figure 4 shows a schematic for the proposed axial orientation of the five major droplets in Figure 3. Note that the droplets in Figure 3 are darkest when the polarizer is aligned with the proposed director arrangement in Figure 4, and lightest when the directions are opposed, consistent with the proposed axial alignment. Other arrangements of the nematic within the droplet (including the bipolar and radial orientations) give distinctive extinction patterns consistent with these director patterns, but not consistent with the behavior shown in Figure 3.

This axial orientation was observed in ZLI 2620 droplets over a particle size range of 5 to greater than 100 microns. All nematic droplets studied possessed a readily discernable two-fold symmetry axis, although in droplets greater than 20 microns in diameter it is often difficult to differentiate between bipolar and axial droplets due to birefringence patterns and colors. Addition of a small amount of pleochroic dye resolves the problem: bipolar droplets show maximum dye extinction when a single polarizer is aligned with the symmetry axis, indicating the director field is predominantly parallel to the droplet symmetry axis. Axial droplets show the opposite effect, with maximum extinction perpendicular to the symmetry axis. Droplets oriented so that the viewing direction is parallel to the symmetry axis are also readily identified using a dye: axial droplets appear to have the nematic circularly arranged around the droplet center, while bipolar droplets appear to have the nematic radially arranged around the center. These effects are apparent in the axial droplets in Figure 3.

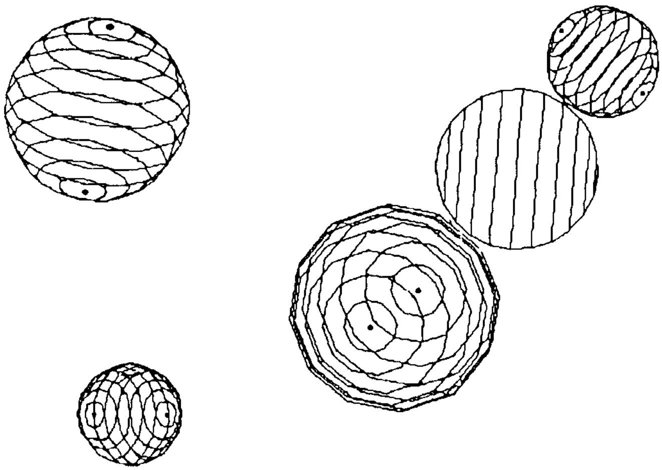


FIGURE 4 Schematic for the five major droplets seen in Figure 4, assuming axial alignment. The droplets photographed in Figure 3 are darkest when a linear polarizer is aligned with the director field within the droplet, as indicated in the schematic.

Careful examination of many droplets show that the representation of the central core of the axial droplets as a straight line is an idealization. Most axial droplets do not possess perfect two-fold rotational symmetry, but instead show features indicative of curvature of the central core, as seen in many droplets in Figure 2. These features will be discussed in a later section of this paper.

EFFECT OF VARIATION OF k_{33}/k_{11} ON DROPLET ALIGNMENT

In previous studies of nematic droplets, the nematic alignment within a droplet has been determined primarily by the alignment of the nematic at the interface between the nematic and the isotropic surroundings.[1-4] In the case of the axial alignment studied here, the surface interaction and alignment is presumed to be the same as in bipolar orientation, but the elastic constants of the nematic enforce a different internal configuration. In order to gain support for the hypothesis that the k_{33}/k_{11} ratio is the force behind the new configuration, a number of nematics with a range of k_{33}/k_{11} ratios were studied as droplets in glycerin.

Table I shows the results of 25 different nematics, with k_{33}/k_{11} ratios[12] ranging from 0.73 to 2.05. For this study, each nematic was

TABLE I

Nematic droplet configuration as a function of k_{33}/k_{11} ratio

Nematic	k_{33}/k_{11} ratio	Droplet orientation	Nematic	k_{33}/k_{11} ratio	Droplet orientation
ZLI 1691	2.05	Bp	ZLI 2116-000	1.19	Bp
ZLI 3039	1.86	Bp	ZLI 2788-000	1.16	Bp
ZLI 3201-000	1.80	Bp	ZLI 1957/5 (20 C)	1.14	Bp
ZLI 1840	1.78	Bp	ZLI 2116-100	1.10	Bp
ZLI 3219	1.67	Bp	ZLI 1957/5 (25 C)	0.99	Bp
ZLI 1800-000	1.63	Bp	ZLI 2977	0.94	Ax
ZLI 3021-000	1.40	Bp	ROTN 3848	0.87	Ax
ZLI 2903	1.38	Bp	ZLI 2975	0.84	Ax
ZLI 2452	1.34	Bp	ZLI 2583-000	0.81	Ax
ZLI 1565	1.27	Bp	ZLI 2974-000	0.82	Ax
ZLI 3347-000	1.27	Bp	ZLI 2583-000	0.81	Ax
ZLI 2950	1.26	Bp	ZLI 2974-100	0.75	Ax
ZLI 3282	1.20	Bp	ZLI 2583-100	0.75	Ax
ZLI 2116-000	1.19	Bp	ZLI 2620	0.73	Ax

Bp = Bipolar, Ax = Axial configuration; ZLI prefix refers to nematic from E. Merck, ROTN prefix for nematic from Hoffman-La Roche. Data at 20 C, unless otherwise noted.

doped with B1, emulsified in glycerin, and observed at least two separate times using a single linear polarizer at 20 C (and at 25 C for ZLI 1957/5). There is a clear correlation relating the observation of bipolar and axial configurations for the nematics based on their k_{33}/k_{11} ratios. There was no correlation of the droplet configuration with the twist deformation (k_{22}) constant of the nematic, either in absolute terms or as a ratio with either the bend or splay deformation constants. Additionally, the absolute values of the bend and splay constants varied over a range of approximately 2x, but again no correlation with these absolute values and the droplet configuration was seen. The wall alignment of the director for each nematic is expected to be tangential, so it is unlikely that differences in surface energy between the nematics could account for the variations in internal alignment.

These data strongly suggest that the k_{33}/k_{11} ratio is the critical factor in determining whether a droplet adopts a bipolar or axial configuration. It appears that the switch from the bipolar to the axial orientation occurs somewhat in the k_{33}/k_{11} range of 0.94 to 0.99. To narrow and more precisely identify this critical range further will require a more detailed study with greater care paid to the elastic constants of a dye-doped material, as well as an indication of the variation of the k_{33}/k_{11} ratio with temperature.

One can qualitatively understand why a low k_{33}/k_{11} ratio would favor an axial alignment (Figure 5). The bipolar arrangement contains nematic director field distortions involving both bend and splay. Ideally, the axial alignment contains only bend distortions with no splay at all. If the splay deformation becomes costly enough in energy relative to the bend deformation (i.e., a low k_{33}/k_{11} ratio) the axial conformation can become favored. Recent calculations by Williams[6] indicate that the energy difference between axial and bipolar droplets for equal elastic constants is relatively modest, so it is not unreasonable that a small drop in the k_{33}/k_{11} ratio can lead to the axial alignment.

AXIAL DROPLETS IN A POLYMER HOST

Emulsifying droplets of ZLI 2620 in an aqueous PVA solution results in droplets which clearly possess the axial orientation, as in the glycerin studies. As the water evaporates and a PVA film forms around the droplets, however, the director field becomes irregular and distorted, as determined by dye extinction. In some droplets in the solid host the central core becomes shows substantial curvature. In other

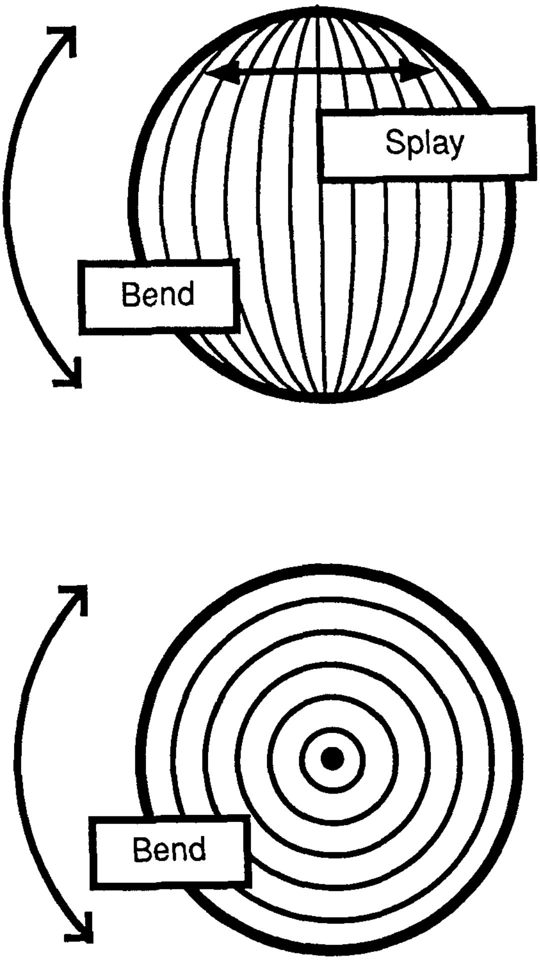


FIGURE 5 Schematic showing qualitatively the splay and bend deformations in cross sections of bipolar and axial droplets.

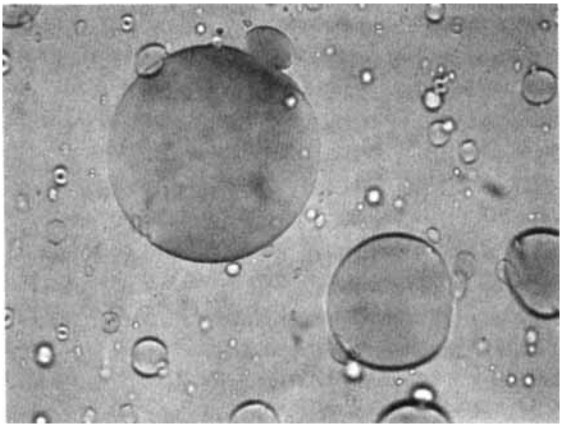
droplets the ends of the core no longer appear at opposite ends of the droplet, but somewhat closer together. This last effect is most apparent in droplets which are in contact with another droplet or a dust particle.

The most likely cause of this director distortion is distortion of the droplet cavity from a spherical shape (in the liquid binder) to an elliptical shape (in the polymer). Electron micrographs,[13] show that the nematic cavity in PVA films is highly distorted, forming an oblate cavity. Observations with an optical microscope show that the more distorted the cavity, the more distorted the director pattern and the core within the droplet.

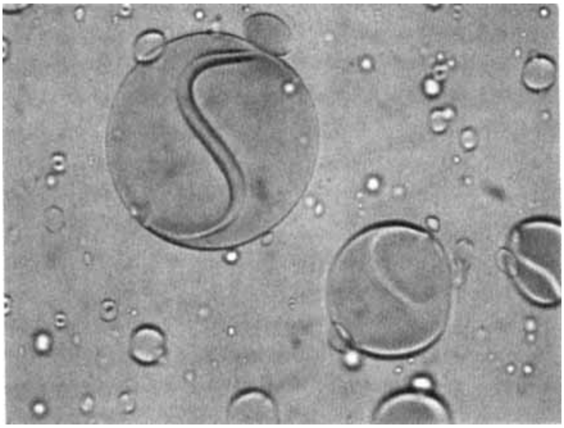
It is possible to transform an axially aligned droplet into a bipolar configuration by application of an electric field. Figure 6 shows a series a microphotographs of ZLI 2620 in a PVA host, with an electric field (on the order of 10^6 V/m) placed along the viewing axis. Initially, there is a small but noticeable movement of the bulk of the nematic within the droplet, concurrent with the very striking appearance of a twisted line running through the center of the droplet (see the largest droplets in 6a and 6b). The best interpretation of this observation is not the sudden growth of a central line defect, but instead a reduction of the apparent birefringence of the surrounding droplet. As most of the nematic aligns with the field, the distortion of light passing through the droplet will diminish. The line defect, present but difficult to observe in the unpowered droplet because of the high birefringence of the unpowered droplet, becomes visible due to a lensing effect; the core has a refractive index greater than the surrounding, well-aligned nematic.

The high degree of curvature in this line is likely due to distortion of the droplet cavity from sphericity. Droplets in glycerin do not show such a highly curved line structure. Figures 6c–e follow the behavior of the line defect in time. Over a period of many seconds, the ends of the lines move closer together. As the line shrinks, the ends of the line appear to leave behind two point defects, visible in both the two largest droplets in Figures 6c–e. When the ends of the lines meet, the line disappears, leaving a droplet which possesses the bipolar orientation (6e). The point defects left behind by the line become the poles of the bipolar droplet. Removing the field restores the axial alignment over a period of many seconds, although the high birefringence of the droplet makes it very difficult to discern the process by which the core line reforms. This general electric field-driven process is observed in most droplets within the film, although the exact shape of the line defect varies from droplet to droplet (presumably due to differences in droplet shape).

This type of behavior involving a line defect is observed in axial droplets formed in PVA films of both ZLI 2620 and ZLI 2975 (k_{33}/k_{11} ratio < 1.0). In contrast, nematics which take the bipolar configuration typically show only a rotation of the point defects to align the droplet with the applied field, as previously observed. Simple rotation of bipolar droplets was observed in PVA films of ZLI 1840 and ZLI 1957/5 (k_{33}/k_{11} ratio > 1.0). Tens of droplets of each nematic were

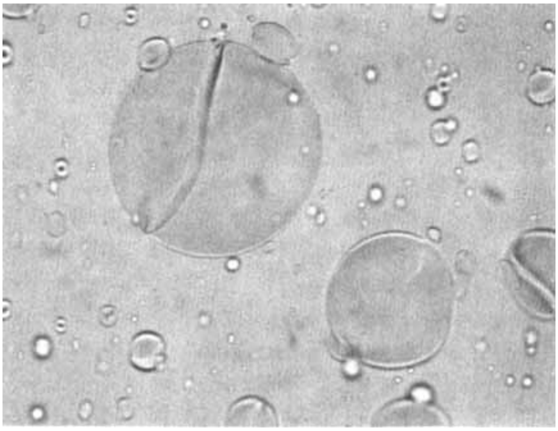


a)



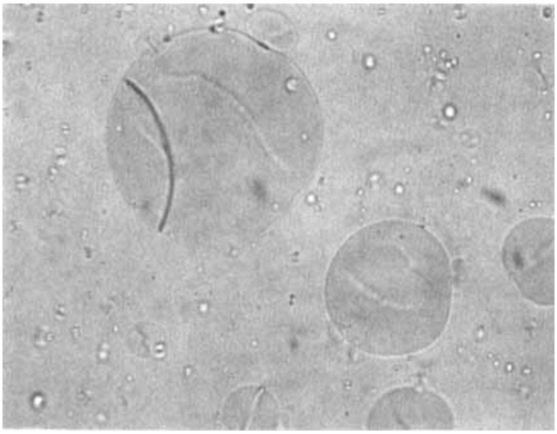
b)

FIGURE 6 Photographs of droplets of ZLI 2620 suspended in polyvinyl alcohol, viewed using a single polarizer. a) No electric field applied. b–e) Application of approximately 10^6 V/m electric field. The transformation observed in the photographs took several seconds to occur. The droplets in e) show the bipolar droplet alignment.

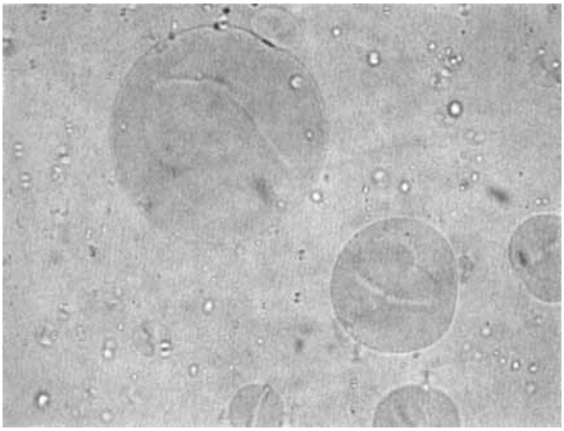


c)

25 μ



d)



e) 25 μ

seen to exhibit similar behavior, lending support to the generality of these processes.

Two exceptions to the above trends must be noted. First, highly nonspherical droplets often showed behavior indicative of multiple defects within the droplet when an electric field was applied. This usually took the form of one or more defect lines forming within the droplets, and could be observed in all four of the nematics studied in PVA. Secondly, if a very strong field was rapidly pulsed across the film (rather than increased gradually), multiple defect lines were observed to appear. These tended to be non-reproducible in nature, and reflect the result of strong asymmetric flow patterns within the droplet.

NATURE OF THE CENTRAL DEFECT STRUCTURE

Previous theoretical and experimental work has cast doubt on the existence of nematic +1 defect lines, the type proposed for these droplets.[14-16] The theoretical work indicated that in a three dimensional structure, it is energetically favorable for the nematic director to twist as it nears the central core, rather than remaining in a plane (the cross sectional view in Figure 1c is a planar cut of an idealized line defect; the relaxation being discussed would twist the director field out of this plane as it approaches the core). The net result of this relaxation is the claim that a +1 defect line can always be replaced by one or more point defects, with a continuous director field between the point defects. Experiments[15,16] involving nematic placed in capillary tubes with perpendicular wall alignment (radial structure) support these calculations; line defects are not seen, but point defects connected by continuous nematic regions are. The theoretical results indicate that a circular +1 defect (of the sort proposed here) should also show similar relaxation effects, and a line defect will not exist. Experiments involving the circular +1 structure have not been performed to date.

The high birefringence of the nematic droplets studied here make it difficult to unequivocally determine the structure of the central core, but the bulk of the evidence indicates that a continuous line defect does propagate through the droplet, and the relaxation found for radial +1 defects in capillary tubes does not occur. The evidence for the existence of a line comes from a number of areas.

The electric field experiments in a PVA binder indicate a line defect exists in the unpowered film. The droplets, as exemplified in Figure 6, show a distinct (albeit highly curved) line structure when an electric field is formed. The most straightforward interpretation of the behavior of this droplets is that a line defect, already present within the droplet, becomes visible due to the reduction of the apparent birefringence of the surrounding nematic. If the core structure was a continuous nematic, it should have readily aligned with the electric field and not be visible.

The second piece of evidence comes from an observation technique performed by Williams *et al.* for nematics in a capillary tube.[16] It was pointed out that fluctuations of an aligned nematic give rise to Rayleigh scattering. With a stopped down diaphragm in the microscope, this is readily observed as a flickering within the droplet. This flickering is most intense if light linearly polarized along the director field is used in the observation. This light scattering technique is complementary to the dye-doping experiments described here, in that a rotating polarizer can be used to map the director field in both methods. In the capillary studies, this scattering technique clearly showed the presence of both a point defect and a nematic core with an alignment direction along the axis of the capillary, consistent with the relaxation mechanism proposed.

In the present study, obvious flickering can be seen within an axially aligned droplets of ZLI 2620 in glycerin when a linear polarizer is aligned perpendicularly to the droplet symmetry axis. A non-flickering core structure is also seen. Rotating the polarizer by $\pi/2$ damps the flickering within the droplet, but does not increase the fluctuations in the core region. Thus unless a flickering core is masked by the birefringence of the droplet, it appears that the core region is not anisotropic, consistent with a line defect.

A line defect is also consistent with the dye-doping experiments in assigning an axial orientation to these droplets. The dye doping experiments performed in this work do not show a well defined core structure with nematic alignment along the droplet's symmetry axis (Figure 3). An anisotropic core should be visible under some preferred polarization if it contains a pleochroic dye, but that effect is not visible in the present system.

Finally, microscopic observation show line-like structures within the axially aligned droplets. Figure 2 shows ZLI 2620 droplets in glycerin, photographed with a stepped down field diaphragm. Each droplet shows some sort of central line structure, although many of these droplets also show other structure within the droplet, presumably due to interference and lensing effects. The difficulty in determining the exact alignment within these droplets was the rationale for performing the dye-doping experiments of the previous section. Nonetheless, continuous line-like structures are seen; the weight of all of these combined observations supports a line structure for the central core, rather than a continuous nematic structure coupled to point defects.

SUMMARY

This paper reports the first observation of axially aligned nematic droplets, an orientation which has been treated theoretically but had not to date been observed experimentally. It is shown that for tangential wall alignments the k_{33}/k_{11} ratio of the nematic appears to be the controlling factor whether the droplet takes on a bipolar or axial alignment. A qualitative explanation for this effect is advanced. It is shown that an axially aligned droplet can be driven to a bipolar orientation through application of an electric field, and that the director field of axial droplets in a polymer host can be highly distorted. The evidence to date indicates that the core structure in these droplets is a true line defect.

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