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# Field-induced textures of polymer-dispersed chiral liquid crystal microdroplets

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# Field-induced textures of polymer-dispersed chiral liquid crystal microdroplets

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We have microscopically observed the textures of very large droplets of cholesteric liquid crystal in a polymer matrix under the influence of an electric field $E$. When $E = 0$, the droplets exhibit rings and often a disclination line extends from the centre to the periphery of the droplet. As $E$ increases, the droplet undergoes a progressive transition to a uniform-appearing texture. This uniform region first occurs near the centre of the droplet, then increases in radius as the field is increased. We propose that the field-off texture corresponds to the Frank–Pryce spherulite model while the uniform field-on texture is the planar texture.

## 1. Introduction

Recently, dispersions of liquid crystal droplets in a polymer matrix have been developed for their potential use in electrooptic applications [1–3]. Although motivation has been to construct useful display devices, there are also new and interesting scientific questions involved in the behaviour of a liquid crystal confined to very small (micron-sized) droplets [4].

The director configuration of liquid crystal droplets without the presence of an electric field has been studied by several authors [5–11]. In the simplest picture, the surrounding polymer medium imposes either a parallel or perpendicular orientation on the liquid crystal director **n**. The resulting bulk director field $\mathbf{n}(x, y, z)$ for each of these boundary conditions then depends on the liquid crystal's elastic constants and the droplet radius. For the case of nematic droplets, the simplest textures are the 'bipolar' texture for parallel boundary conditions (with two $s = 1$ disclinations at the north and south pole), and the 'hedgehog' texture for perpendicular boundary conditions (with a point disclination at the centre) [11].

When a field is applied, a competition arises between the surface forces and the bulk forces exerted by the field. There are three possible effects, which occur for increasing field. (1) The zero-field texture may simply be reoriented in the droplet. (2) The zero-field texture may itself be distorted but not topologically altered. (3) The texture may be replaced, partially or completely, by either a new, topologically different texture or a thermodynamically distinct phase. Thus, spherical *nematic* droplets in which the director orients *parallel* to the boundary take on a randomly oriented bipolar configuration. If the dielectric anisotropy is positive, the application of an electric field causes first an alignment of the bipolar axis along the common field direction, and then, for higher fields, an additional distortion of the texture as the director attempts to align itself more closely with the applied field [1]. For nematic

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droplets with *perpendicular* boundary conditions, however, the effect of an applied field is to replace the zero-field hedgehog texture with a new axial texture. These two textures have different disclination patterns; the conversion of the texture is discontinuous and analogous to a first-order phase transition [12].

If the previously described nematic droplets are made chiral, the situation should be dramatically different. Chiral nematics (cholesterics) possess an intrinsic helical structure [15] which is fundamentally incompatible with the alignment imposed by the boundary conditions of a spherical interface and of an applied field. In the present study we have made polymer-dispersed droplets of chiral liquid crystal in which both the droplet size and the pitch of the liquid crystal are much larger than the wavelength of light, so that the texture can be directly observed with an optical microscope. Our results show that without an electric field, the droplets take on a spherulite texture. When an electric field is applied, however, the spherulite texture reverts to a region of planar texture which starts at the centre of the droplet and increases progressively to larger radii as the field increases.

## 2. Experiment

The sample is a mixture consisting of 50 wt % thermoplastic polyvinyl butyral (PVB), 2 wt % chiral liquid crystal (R)-(−)-4″-(2-methylbutylphenyl) 4′-(2-methylbutyl)-4-biphenyl carboxylate (CE2), and 48 wt % nematic mixture ZLI-2806, which has negative dielectric anisotropy ($\varepsilon_a = -4{\cdot}8$). Both CE2 and ZLI-2806 were obtained from British Drug House. This mixture was first dissolved in chloroform, then pipetted on to an ITO-coated glass slide where the chloroform was allowed to partially evaporate at room temperature. The glass slide was then heated to 110°C to complete the evaporation. 72 $\mu$m mylar spacers were placed on the slide, a second ITO-coated slide placed on the spacers, and the whole sample raised to 140°C for 1/2 hour. When the sample cooled, phase separation of the liquid crystal from the polymer resulted in liquid crystal droplets, the size of which depended on the cooling rate. A cooling rate of 1°C/min yielded 10 $\mu$m diameter droplets; for the drops reported here, a cooling rate of 0·02°C/min resulted in drops 50 $\mu$m in diameter.

Figure 1 shows the droplets for various electric fields as viewed by transmission microscopy between crossed polarizer and analyser. Common to all the droplets is a broad dark cross which is aligned with the polarizer and analyser directions. Each droplet also has a set of rings similar to the fingerprint texture seen in bulk chiral nematic samples. These are related to the helical twist structure and are spaced half a pitch apart.

In figure 1 (*a*) the field is zero. Similar droplet textures have been reported previously [5–12]. We observe two types of droplets. The first type consists of concentric circles with a disclination line extending from the centre to the edge. The second type exhibits no visible disclination line and the rings actually constitute a double spiral. As we shall see, these features are the signature of the spherulite texture.

The effect of applying 1 kHz AC voltages (reported as rms) to the sample is shown in the sequence figure 1 (*a*)–(*d*). For $V = 0$ (figure 1 (*a*)), the droplets are entirely in the spherulite texture. At 15 V (figure 1 (*b*)), the centre region of the droplet has transformed from the spherulite texture to a uniform-appearing texture. This uniform texture then increases in radius as the voltage is increased—to 21 V (figure 1 (*c*)), and finally to 63 V (figure 1 (*c*)) where the droplet has been transformed almost completely from the spherulite texture to the uniform texture.

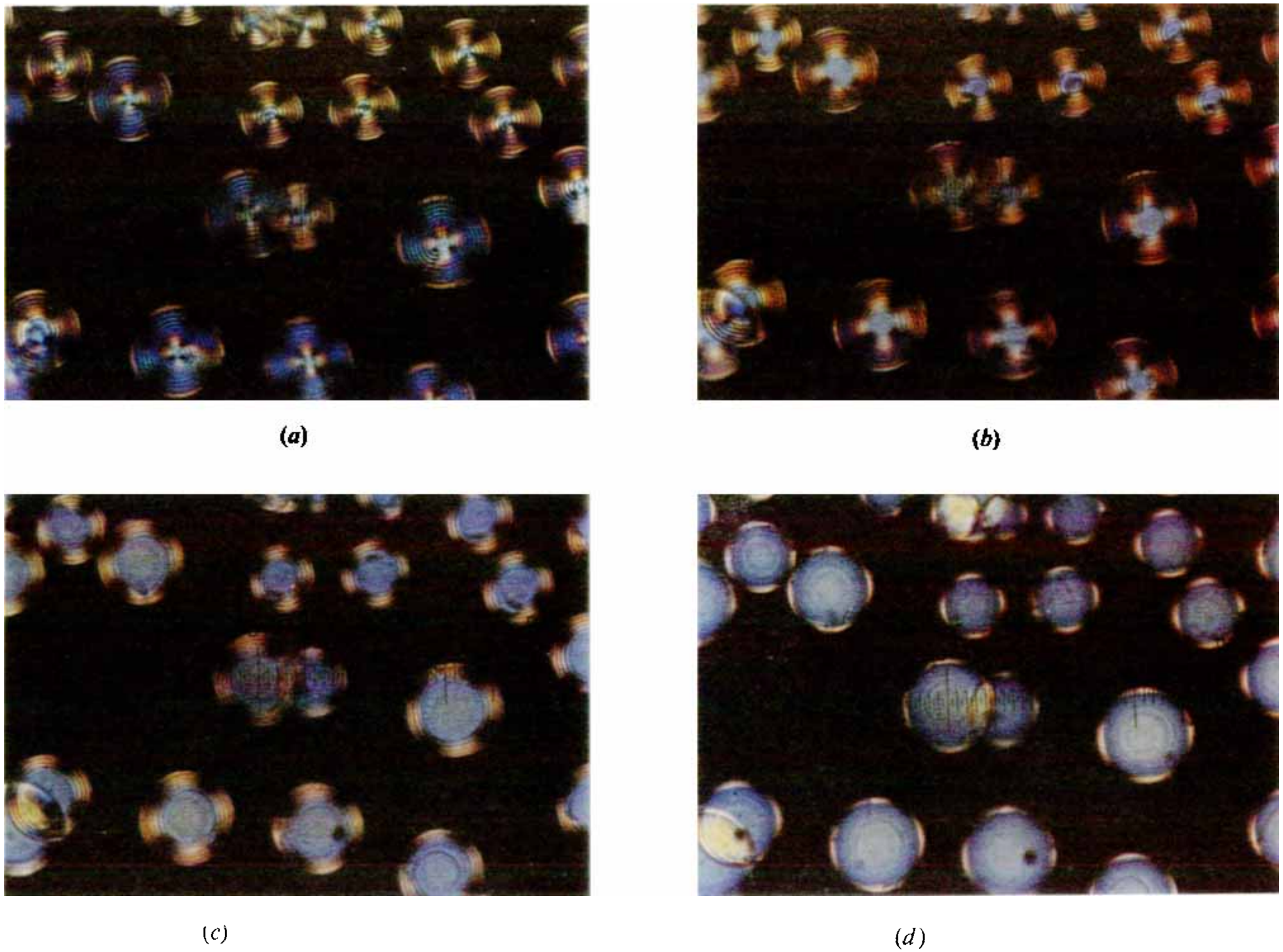

Figure 1. Microphotographs of droplets. Droplet diameter is $\approx 50\,\mu$m; sample thickness $\approx 72\,\mu$m. (*a*) $V = 0$ V; (*b*) 15 V; (*c*) 21 V; (*d*) 63 V.

As the voltage is increased, the radius of the central region increases in erratic steps as individual rings separately convert to the uniform texture. For those drops with a visible disclination line, a gradual increase of the field causes no immediate change until, at some field, one end of the innermost ring suddenly disconnects from the disclination line and annihilates 'around the circle', the annihilation process for that ring terminating when the ring has completely converted to the uniform texture.

To exhibit this process quantitatively, figure 2 shows the diameter of the central uniform region as a function of applied voltage for three different droplets. Note that there appears to be an initial threshold around 8 V, after which the diameter increases roughly linearly, levelling off at a maximum voltage of 30 V.

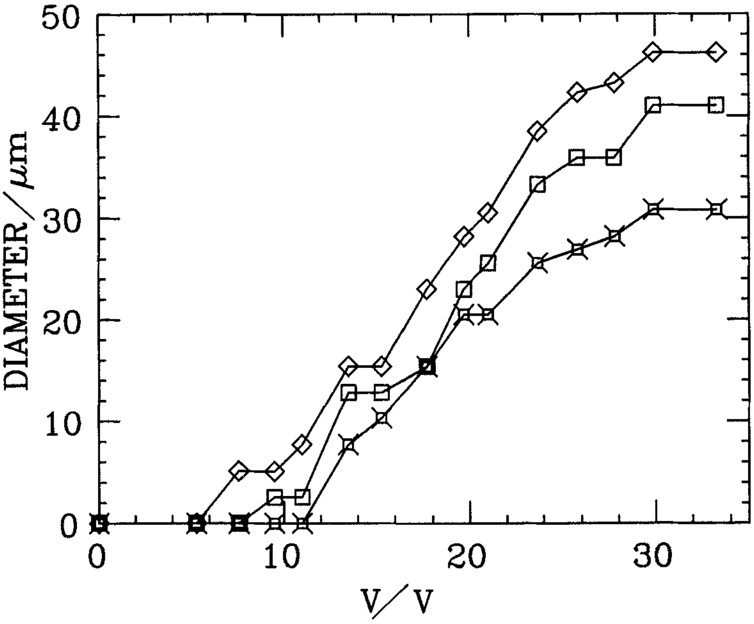

Figure 2. Radius of central uniform region as a function of applied voltage for three droplets.

## 3. Discussion

Structures of chiral droplets confined to spherical cavities have been studied by several authors [3, 5–11, 13]. In general, the helical structure will be distorted and disclination lines may be introduced because the undistorted helical structure is not topologically compatible with the boundary conditions imposed by the droplet interface. The resulting configuration will then depend on the boundary orientation, the elastic constants of the liquid crystal, and any applied fields. In this paper we consider tangential boundary conditions in which the molecules are constrained to lie parallel to the interface. Although perpendicular boundary conditions are also possible [8–11], parallel surface alignment has generally been found for polymer-dispersed liquid crystal droplets by other workers [1].

We consider two models. The *planar* model [9] shown in figure 3 satisfies both the tangential boundary conditions and perpendicularity to an applied field, the latter being appropriate for a liquid crystal with negative dielectric anisotropy. Let the droplet have a helix axis fixed in space, and consider a planar cross-section perpendicular to this axis (figure 3 (*b*)). The director lies in the plane and, except for distortions by the boundary, aligns itself parallel to a certain diameter. At the end of the diameter, however, point disclinations in the director field are required. These planes are then stacked perpendicular to and along the helix axis such that the averaged director of any plane rotates by angle $\delta\phi = 2\pi\delta z/P$, where $\delta z$ is the distance between two planes and $P$ is the pitch. The disclination points then trace out disclination lines on the surface of the droplet (figure 3 (*a*)).

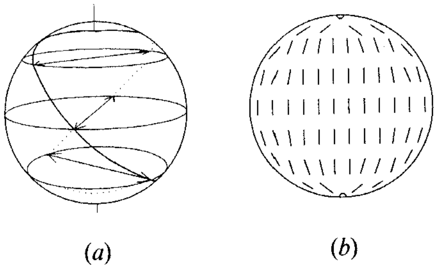

Figure 3. Planar configuration of a spherical droplet. (*a*) Locus of disclination line on droplet surface; (*b*) director field in a plane perpendicular to the helical axis.

If such a droplet is illuminated with white light along the pitch axis, the back-reflected light will consist of a circularly polarized selective reflection with wavelength $\lambda = nP$ ($n$ is an average refractive index of the liquid crystal). In transmission, the appearance will be uniform.

A second possibility is the *spherulite* configuration of Frank and Pryce [6, 11] shown in figure 4. Consider a sphere $S$ of radius $r$, as shown in figure 4 (*a*). The family of circles on $S$ corresponds to the intersections of the sphere $S$ with all planes passing through a line $d$ tangent to the sphere at point $D$. The director anywhere on the sphere $S$ is tangent to the circle passing through that point. $D$ is a defect point. Now consider other spheres $S'$ concentric to $S$ but with radii $r'$. The director configuration on

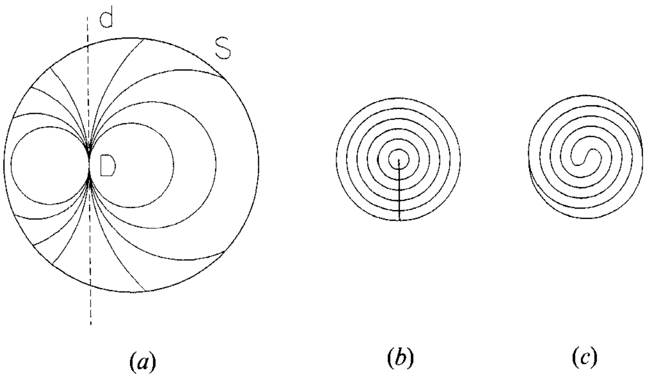

Figure 4. Spherulite director configuration. (*a*) The director is tangent to circles which are in turn intersections of the sphere with planes passing through line $d$ tangent to sphere at $D$; (*b*) Appearance of spherulite in transmitted light when the disclination line is perpendicular to the light and the pitch is much larger than the wavelength; (*c*) Appearance in transmitted light when disclination line is parallel to the light. See text for additional explanation.

$S'$ is identical to that of $S$, except that the tangent $d'$ is rotated by an angle $\delta\phi = 2\pi(r' - r)/P$. The defect point $D'$ is on the same radius as $D$; together all the defect points form a radial disclination line $R$.

The appearance of the spherulite in transmitted white light and crossed polars will be the fingerprint texture if the pitch is significantly longer than the wavelength [6, 11]. If the disclination line $R$ lies in the sample plane perpendicular to the incident light, the texture appears as concentric circles with the disclination line along one of the radii (figure 4 (*b*)). If, however, the disclination line is along the incident light direction, the texture consists of a double spiral and the disclination line is not observed (figure 4 (*c*)).

The stability of one configuration compared to another is found by calculating and comparing their free energies, which includes the elastic distortion energy, the energies of the respective disclinations, and the energy due to orientation of the director in applied fields. To our knowledge, a calculation of these effects for chiral liquid crystals has not been reported. Experiment indicates, however, that when the field is zero the observed spherulite texture evidently has the lower free energy. When, however, the field is non-zero and the material has negative dielectric anisotropy, the planar texture can lower the field energy significantly since the directors are turned to their lowest energy configuration. Whether this lowering of the energy offsets the increased elastic energy is not clear; again, the calculation is not available. Nevertheless, we are able to offer the following qualitative interpretation of our droplet textures.

When $V = 0$, the droplet appearance in figure 1 (*a*) corresponds to the spherulites of figure 4 (*b*)–(*c*). Droplets with concentric circles (with a defect line in the plane of the sample) and double spirals (with the defect line perpendicular to the plane of the sample) are observed. Interestingly, however, there is a lack of droplets with defect lines at some angle other than 0 or 90° to the light direction.

When $V > 0$, the uniform texture in the centre region of the droplet represents the planar texture. This assignment is based not only on the uniformity of the appearance in transmitted light, but also on the observation of coloured selective reflections in smaller droplets when $nP$ is in the visible wavelength region [3]. The novel aspect of this observation is that only the centre region of the droplet has changed texture, the size being dependent on the field. Since the deformation energy of the spherulite is highest near the centre, however, is seems likely that this region would be the first to revert to the planar texture under the influence of an applied field.

In this connection, we point out that several authors [8, 12] have reported subjecting spherulitic droplets to *magnetic* fields, with the field in the plane of the sample and perpendicular to the viewing direction. The application of the field caused the central region of a drop to transform to a uniform appearing texture of *oval* shape, the long axis of the oval lying along the field direction. Unlike our drops, however, the liquid crystal director tended to align *parallel* to the field. Therefore it is safe only to say that, for our drops, the three-dimensional shape of the central planar region may not be spherical.

These observations help to explain qualitatively the behaviour of the reflected light in the polymer-dispersed chiral liquid crystal (PDCLC) displays recently reported by Crooker and Yang [3]. In these dispersions, the droplets were $\approx 15\,\mu$m in diameter and $P \approx 300\,\mu$m, so that the fingerprint texture could not be observed. When the field was off, the PDCLC droplets exhibited a dark cross aligned with the polarizer and analyser and were weakly scattering, as might be expected from the spherulite texture when the light is near the selective reflection wavelength.

When an electric field was applied to the sample, the dark cross disappeared and the PDCLC droplets took on the coloured appearance characteristic of selective reflections from bulk cholesterics, the reflectivity increasing with increasing field. If, like the larger droplets reported here, the central core of the droplet had converted to the planar texture and the radius of the core increased with increasing field, the overall sample reflectivity would qualitatively increase as observed.

## 4. Conclusion

We have succeeded in forming polymer-dispersed chiral liquid crystal droplets with droplet size and pitch large enough to be observed by visual microscopy. When there is no applied field, the droplet is in the spherulite configuration. When a field is applied, the core of the droplet undergoes a transition from the spherulite to the planar configuration, the size of the core increasing with increasing electric field.

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