

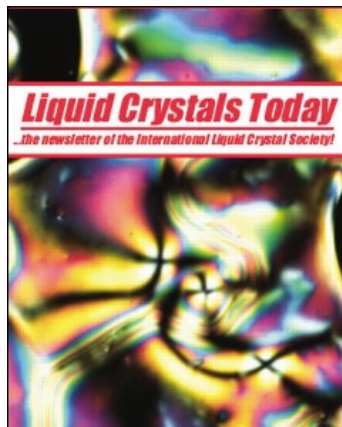
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Chiral Nematic Liquid Crystal Droplets

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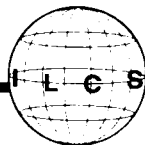
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Chiral Nematic Liquid Crystal Droplets

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Liquid crystal droplets, being relatively easy to produce, have been of interest since the early days of liquid crystals. This interest is both scientific and practical. Scientifically, we would like to know how confinement can alter the macroscopic properties of materials previously understood only in the bulk. In the case of liquid crystal droplets, the boundary conditions are fundamentally incompatible with parallel orientation in the bulk, which leads to topological defects in the structure. By applying an electric field or increasing the chirality, the situation is further complicated and new textures with new defects may evolve.

From a practical standpoint, an understanding of the above processes is necessary when using droplets for applications. Polymer-dispersed *nematic* liquid crystal

(PDLC) droplets have already been used in displays for a number of years. Polymer-dispersed *chiral-nematic* liquid crystal (PDCLC) droplets, with which this review is concerned, can also be used in displays, although they utilize a different principle than nematic PDLCs [1]. Nematic displays work in transmission and depend on refractive index matching; chiral nematic displays work in reflection and depend on the principle of selective reflection. Chiral nematic droplets have been studied for many years; however, studies of their behaviour under the influence of applied fields have been more recent. Experimentally, the situation is shown in figure 1 in which several large-pitch droplets of chiral nematic liquid crystal with negative dielectric anisotropy are viewed between crossed polarizers as an electric field is applied. The zero-field texture is called the Frank-Pryce texture, in which the helical axes are radial and a radial defect line can be seen when it is not aligned with the viewing direction. When an electric field is applied, the central region of the droplet takes on the more familiar planar texture in which the helical axis is aligned with the field. As the field increases, the radius of the central region increases correspondingly until at high field the whole droplet is planar.

What is happening to the droplet internally as it evolves from the Frank-Pryce to planar texture? Recent theory has given an explanation, as the rest of this article will show.

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Low fields

The director fields in chiral nematic droplets are determined by minimizing the total Frank free energy, which is a sum of elastic, surface, and field energies. The importance of these contributions may be understood in terms of four characteristic lengths. The radius R of the droplet and pitch P of the chiral nematic liquid crystal are obvious. The effectiveness of surface anchoring is given by the extrapolation length $b=K/W$, where W is the anchoring strength and K is an elastic constant. For a nematic drop, the anchoring is strong when $b \ll R$; however in a chiral nematic drop with $P < R$, the important length is the pitch. Consequently when $b \ll P$ the anchoring is strong; when $b \geq P$ the surface director does not completely obey the boundary conditions. It follows that the type of anchoring (weak or strong) in large chiral nematic droplets ($P < R$) is independent of the size of the droplet, unlike the case of nematics. Finally, the effect of the electric field is given by the coherence

(continued on page 2)

length $\xi = [K/(\epsilon_0 \epsilon_a |E^2|)]^{1/2}$ where E is the field and ϵ_a is the permittivity anisotropy of the liquid crystal. If $\xi \ll R$ the field is strong enough to overcome the elastic effects and create internal alignment.

In zero and low electric fields ($\xi \gg R$), stable structures are determined by the competition between elastic and surface effects only. For nematic droplets, a variety of structures has been predicted and/or observed, for example, radial and axial structures in the case of perpendicular anchoring, and bipolar in the case of parallel anchoring. For chiral nematic

droplets, the set of structures is even richer, but we shall restrict our attention to the case of chiral nematic droplets with parallel surface anchoring and negative dielectric anisotropy, due to their possible use in light-reflecting applications.

Unconstrained chiral nematic liquid crystals organize in the planar texture, given by $\mathbf{n} = (\cos(qz), \sin(qz), 0)$, where the wave vector $q = 2\pi/P$ lies in the z -direction. Clearly the planar structure is not compatible with spherical confinement and parallel boundary conditions; this incompatibility leads in turn to a variety of

interesting director configurations. A simple description of such complex structures is based on the idea of a *chiral nematic surface*, which is defined as a surface to which the director field is tangent at every point or, equivalently, a surface to which the vector field $\mathbf{q}(\mathbf{r})$ is everywhere normal. For planar chiral nematics, these surfaces are equidistant planes (\mathbf{q} is a constant). In droplets, however, where the chiral surfaces are strongly curved, observations show that they are still nearly equidistant. Thus, for non-planar chiral structures, the assumption of constant chirality $|\mathbf{q}|$ and

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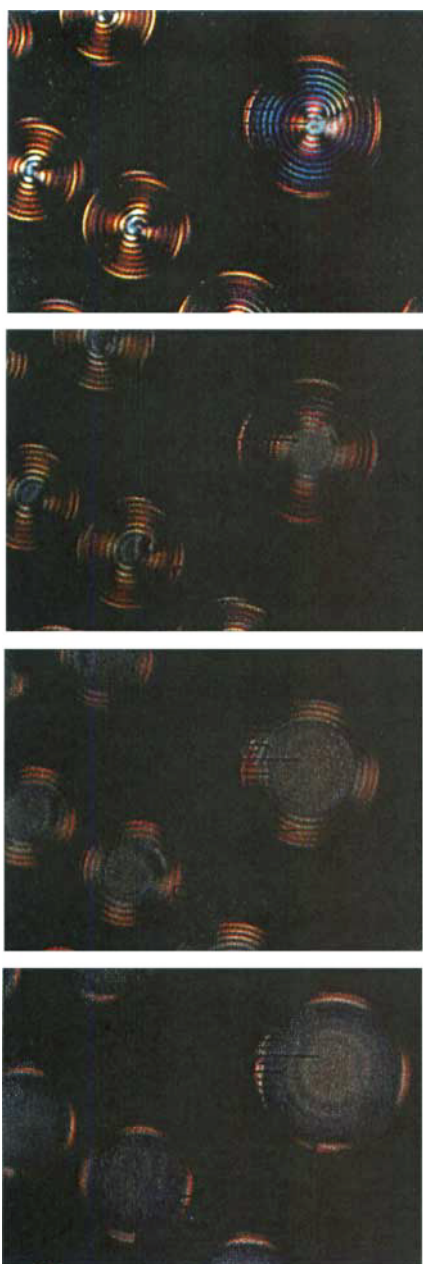


Figure 1. Photomicrograph of chiral nematic droplets subjected to increasing field. From top to bottom: 0 V, 15 V, 21 V, 63 V. The sample thickness is $\approx 72 \mu\text{m}$; the viewing direction is parallel to the field.

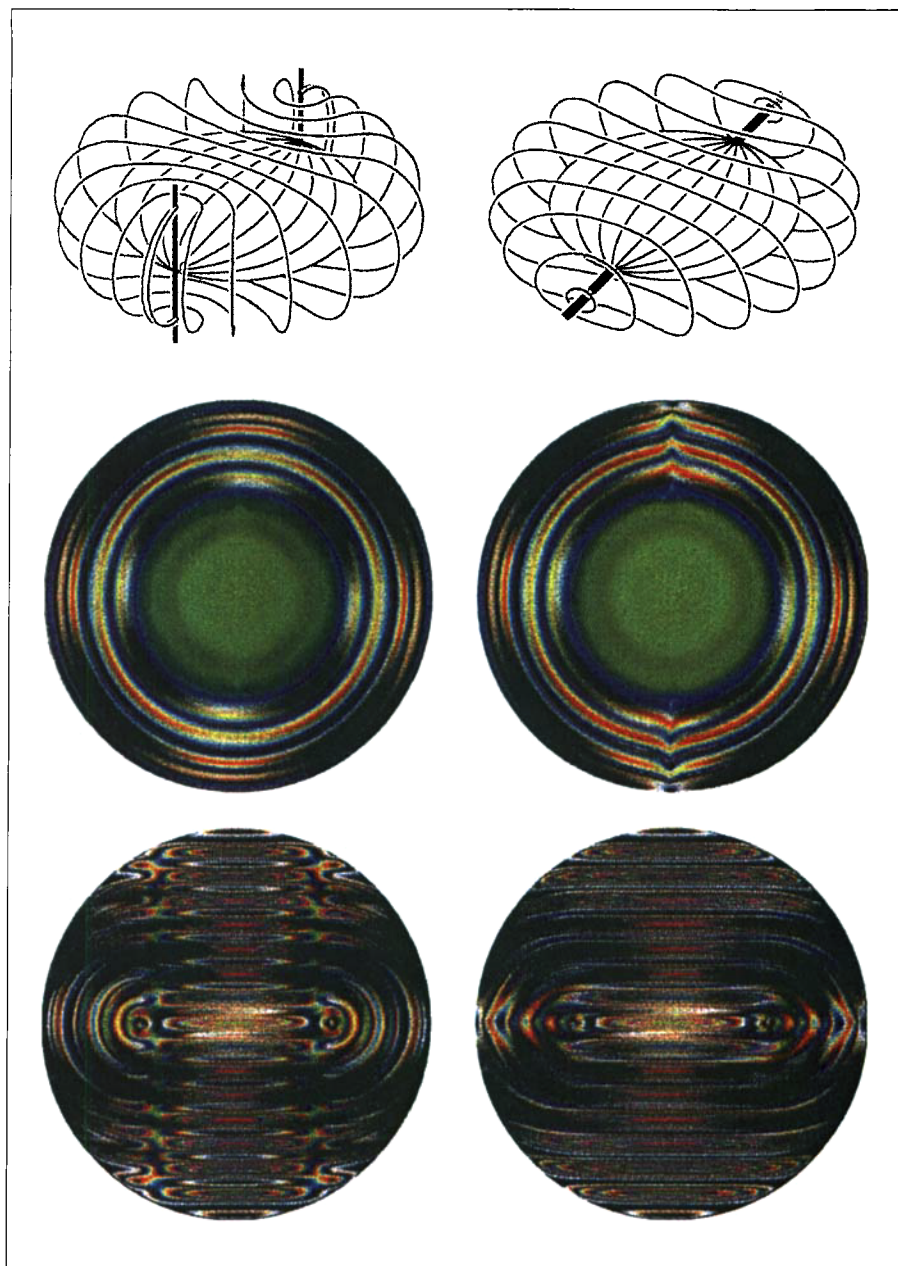


Figure 3. Theoretically predicted oblate droplets that have not been observed yet. Top: the director configurations of the structures; the thickness of a defect line corresponds to its strength. Centre: simulated textures of the upper structures when viewed along the electric field. Compare the right simulation with droplets in figure 1. The most obvious difference is due to different arrangement of the defect lines; the observed structure has one $s=2$ defect line and the diametrical oblate structure has two $s=1$ defect lines. Bottom: again simulated textures, but this time viewed perpendicular to the field.

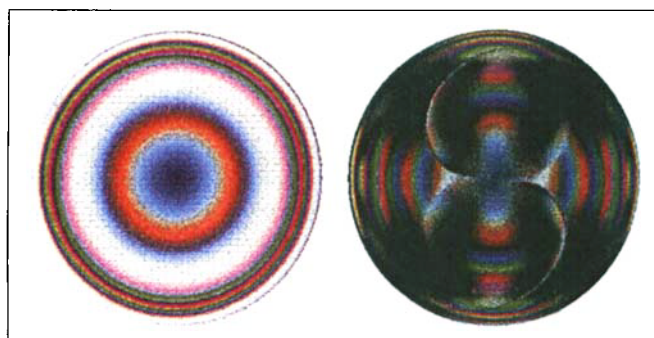


Figure 5. Simulated textures of two limiting planar structures when viewed along the applied electric field: very weak (left) and infinitely strong anchoring (right). Note that left texture is similar to the central texture in figure 1, whereas the right texture clearly shows the surface defect line. In both cases the chirality is the same $qR=\pi$.

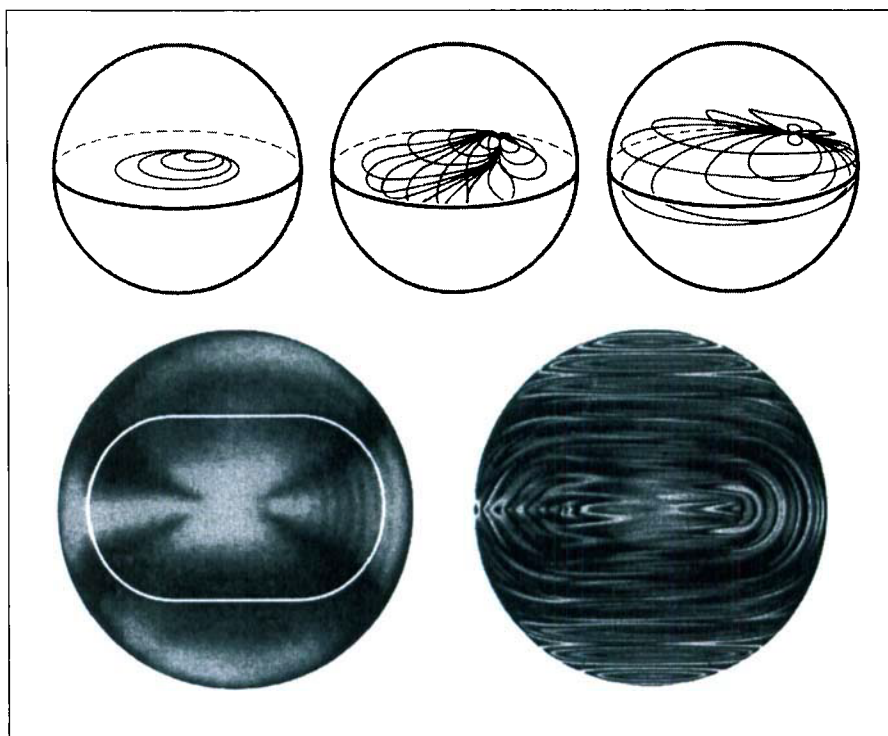


Figure 2. Observed and simulated droplet figures. Top: modelled director configuration of the observed oblate structures — director fields on several chiral nematic surfaces are shown separately. Bottom left: actual droplet observed by polarization microscopy; viewing direction is perpendicular to the defect line and to the electric field. One chiral nematic surface is drawn in white to guide the eye. Bottom right: simulated droplet in the same viewing direction; note that defect line is on the left side of the droplet.

equidistant chiral nematic surfaces is a good one.

In the absence of an electric field, the natural choice for the shape of chiral nematic surfaces in a spherical droplet with parallel surface anchoring is a sphere. Topology tells us, however, that a director field constrained to a closed three-dimensional surface must contain point defects, the sum of whose strengths must equal 2. (The strength s of a point defect on a surface is defined as the number of rotations made by the director along a loop encircling the defect.) On successive chiral nematic surfaces, these two-dimensional point defects combine to form defect lines. The resulting director field in spherical coordinates reads

$$\mathbf{n} = \cos\Omega \mathbf{e}_\theta + \sin\Omega \mathbf{e}_\phi, \quad \Omega = (s_0 - 1)\phi + \Omega_0 + qr, \quad (1)$$

where Ω_0 is an arbitrary constant and s_0 is the strength of the defect line along the $+z$ axis. The defect line along the $-z$ axis has the strength $2 - s_0$, so spherical structures can be labelled with a pair of numbers $(s_0; 2 - s_0)$. Of course $(s_0; 2 - s_0)$ and $(2 - s_0; s_0)$ represent the same spherical structure and a 'defect' line of strength $s = 0$ represents a

non-singular director field along this line.

The typical structure shown in figure 1 (top) is the Frank-Pryce or radial spherical structure, denoted by $(2;0)$. The diametrical spherical structure $(1;1)$ has also been observed, but only rarely. Given a proposed director structure, pictures like those in figure 1 can be simulated by computer. Comparison of simulated and observed polarizing microscope textures, viewed under different polarizations and droplet orientations, is an effective way to determine the structures of liquid crystal droplets (figures 2 and 3). The basic idea behind the simulation calculation is that the drop is made up of many slices of optical retarder whose retardance $\Delta\phi(\mathbf{r})$ can be calculated from $\mathbf{n}(\mathbf{r})$. Knowing the droplet thickness, the director configuration along the light path, the wavelength, and the orientation of the polarizers, the intensity of each ray can be calculated [2].

In all spherical structures the director rotates spatially along the radii and there is no macroscopically preferred direction for \mathbf{q} . As a result, the droplets weakly scatter light regardless of their orientation, but do not selectively reflect a particular colour.

Intermediate fields

At the lowest fields, no change takes place in the droplet texture, but the line defect aligns parallel or perpendicular to the field. Once a threshold field has been reached ($\xi \sim R$), however, the texture itself begins to deform. Because of the negative dielectric anisotropy, the helical axes begin to align with the electric field and the chiral nematic surfaces become *oblate* or flattened along the field direction. Initially the oblateness is small, with the flat portion of the surfaces appearing only near the axis of the drop. With increasing field the flat central part of the droplet grows in a step-like fashion (see figure 4).

An important consequence of the deformation of the now oblate chiral nematic surfaces is the change of the topology. Since the outer oblate surfaces no longer fit completely within the sphere, they are cut off by the droplet border. Two classes of chiral nematic surfaces appear. The closed ones in the central part of the droplet have already been discussed: they are topologically equivalent to a sphere and they have defects whose strengths sum to two. The internal parts of the cut off surfaces, however, are topologically equivalent to a disc with the director lying in the surface. The two dimensional defects contained on such a surface depend on whether the directors are tangent to the edge of the disc, and this, in turn, depends on the anchoring strength at the droplet surface.

If the anchoring strength is weak ($b \gg P$), the directors on the boundary of the cut off surfaces are undetermined; they are simply the continuation of the inner director field on the closed chiral nematic surfaces. For strong parallel anchoring ($b \ll P$), however, the cut off chiral nematic surfaces are equivalent to a disc with parallel boundary conditions at the border. From topology, we know that a two dimensional director field on a two dimensional closed surface must have point defects, the sum of whose strengths equals one. The resulting oblate surfaces can even have surface defect lines, which may become very long if the radius of the droplet is large compared to the pitch. Structures with long surface defect lines have not yet been observed, probably because of their high elastic free energy. Evidently it is cheaper for the liquid crystal to violate the boundary condition than to

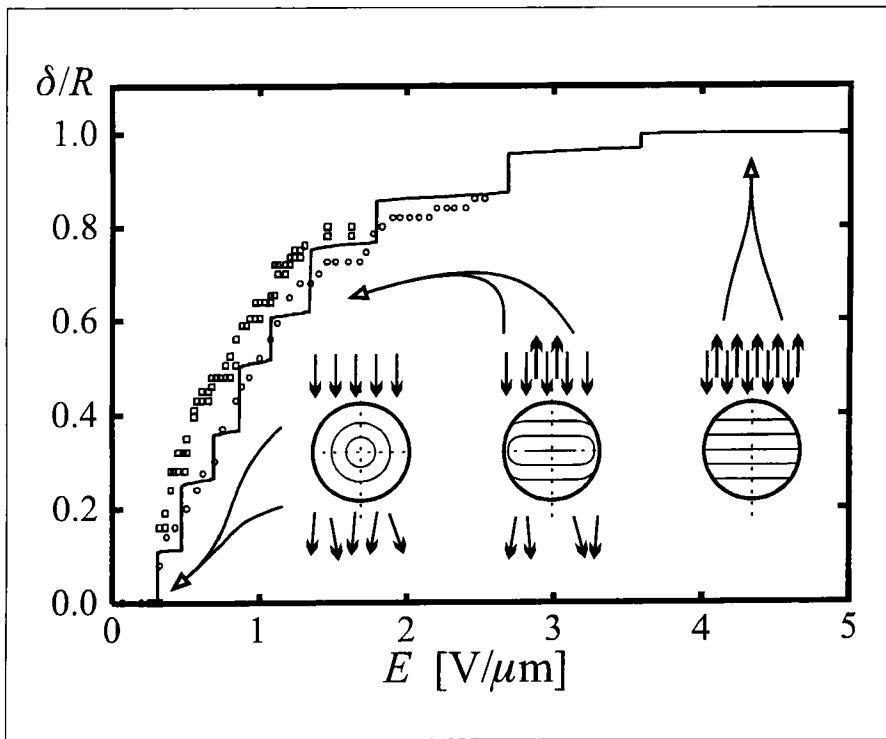
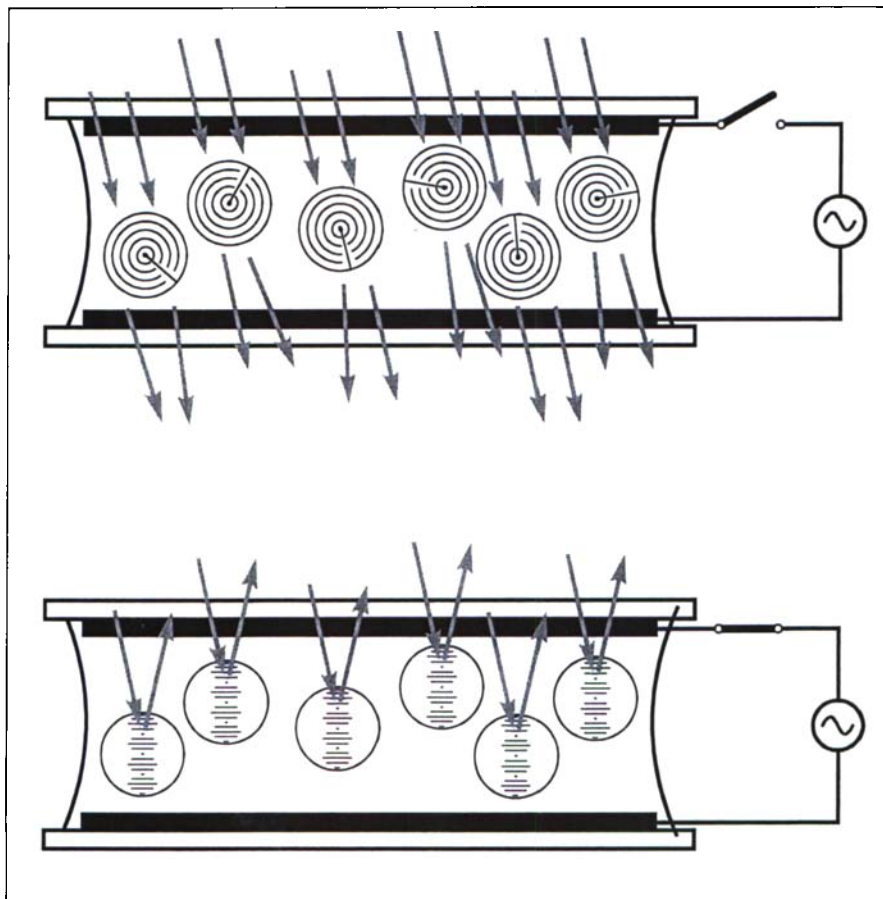


Figure 4. Dependence of radius δ of the flat central region on the electric field. Experimental data (circles and squares) is from [3], calculations (solid line) are from [4]. Schematic presentation of the characteristic shape of chiral nematic surfaces and optical behaviour at certain fields is added; spherical structures are slightly scattering, oblate structures are partially selectively reflecting and partially scattering, and planar structures selectively reflect the incident light.



pay the cost of high deformation in the vicinity of the defect lines.

If the pitch is sufficiently short ($P \ll R$), the flat planar region of the oblate structure will selectively reflect light of a colour corresponding to the pitch. Electrical control of the size of this region is promising for use in continuous scale reflective colour displays.

High fields

In high electric fields ($\xi \ll P$), the twist axes in the entire droplet become aligned by the electric field, the structure becomes planar, and the director configuration is controlled by the competition between elasticity and surface anchoring. All chiral nematic surfaces are topologically equivalent to discs. If the anchoring is weak ($b \geq R$), the director field takes on the planar texture away from the boundary and is only modified by the droplet surface where the chiral nematic surface contacts the droplet boundary. Even if the anchoring is stronger ($b \ll R$), the situation does not change much as long as the pitch is short ($P \leq b$). Significant changes appear only when the pitch is much greater than the extrapolation length ($P \gg b$). In this case the parallel boundary conditions cause point defects to appear in each chiral nematic surface and these defects combine in turn to produce line defects. The latter have not been observed yet, probably because it has not been possible to make the anchoring strong enough. Were the anchoring strong enough, one of the most stable planar structures would be the *planar bipolar* structure. This structure is characterized by two $s=1/2$ surface defect spirals (figure 5).

Applications

Figure 6 shows a polymer-dispersed cholesteric liquid crystal (PDCLC) display [5]. When the field is off the droplets take on the Frank-Pryce texture and the display appears weakly scattering. When the field is on, the droplets take on the planar texture and selectively reflect incident light. The reflected light is reasonably

Figure 6. PDCLC display. Top: with the field off, the droplets are in the Frank-Pryce texture and the display is transmitting. Bottom: with the field on, the droplet structure is planar and the display reflects monochromatic light of one circular polarization.

monochromatic and circularly polarized, as determined by the type of liquid crystal used. Note that the display is viewed in ordinary ambient light—neither polarizers nor backlighting are required. Also, as with nematic PDLC displays, no surface alignment treatment is required.

One can think of several ways to utilize such a display. First it can be used as a black and colour display, much like currently-used black and white displays. For a multicolour reflective display, several colours may be stacked; red, green, and blue would additively mix to produce colours in a large fraction of the chromaticity diagram. If one wanted to reflect 100% of the light instead of the 50% associated with one circular polarization, one could use a sandwich of right and left handed cholesterics. On the other hand there may be an application for a switchable circular polarizing filter where it is only desired to switch, say, right circularly polarized

light, leaving the left circularly polarized light unchanged.

The PDCLC display can also be used in the transmission mode. When switched on, the display will transmit 50% of one monochromatic single circular polarization. Use of an additional circular polarizer will make the switching 100% and allow the device to be used as a switchable circularly polarizing filter. Note that the untransmitted light is scattered and not absorbed, which is important for heat-producing projection displays. For a multicoloured display, stacked colours will also work, but the colour mixing will be subtractive, not additive. A switchable 100% shutter for one colour can also be constructed by stacking left and right handed liquid crystals, with no additional polarizers required. In principle, by suitable stacking, a 100% light shutter can be constructed which would switch all light on and off, again with no additional polarizers.

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