Defects in liquid crystals

Appearing under the polarizing microscope as ellipses, parabolas, hyperbolas, lines and points, colorful structural singularities are understood through topological and geometrical arguments.

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The most striking feature of liquid crystals is the wide variety of visual patterns they display. These patterns, such as those shown in figure 1 and on the cover, are due almost entirely to the defect structure that occurs in the long-range molecular order of the liquid. Indeed, historically, the underlying structure of the liquid-crystal phases known as nematic and smectic-A was discovered from a study of stable defects that characterize these phases. Such defects are easily visible in the optical microscope. By examining the thin and thick thread-like structures observed in nematic liquid crystals, Otto Lehman[1] and Georges Friedel[2] deduced that this phase involves long-range orientational ordering of the long axis of the rod-like molecules. (The direction of this orientational ordering is what we now denote by the unit vector $\hat{\mathbf{n}}$, called the director.)

More remarkably, Friedel's observation and elucidation of defects that look like confocal pairs of conic-section curves (focal conic defects, figure 1) in smectic-A liquid crystals—at that time often called the liquid with cones—led him to conclude that smectic A is a layered system. He futher found[2] that the planes of molecules are perpendicular to $\hat{\mathbf{n}}$. Nevertheless, generations of observers have been mystified by the focal conic domains and the extraordinarily constant layer spacing over macroscopic distances implied by their structure. Indeed, Friedel's fundamental geometrical arguments, which directly couple the existence of such domains to the smectic structure, seem frequently to become lost in the discussion.

Recent years have seen increased interest in liquid crystals and their defects. Research on the latter has focused on the detailed structure of individual defects and the ways in which groups of defects can arrange themselves.[3,4] Investigators did not realize until 1971 that line defects of unit strength in nematic liquid crystals could move out of a plane of orientational order, or "escape into the third dimension."[5] (Nematic liquid crystals are less ordered than smectic A. In nematics there is long-range orientational ordering of the long axes of the rod-like molecules, but they do not form layers.) The finding that line defects can escape, along with increasing interest in defects in gauge theories and in more complicated order parameters such as those occurring in superfluid helium-3, led researchers in France[6] and the Soviet Union[7] to discuss the singular defects in terms of the topological arguments of homotopic groups, which we will examine later. As we shall see, one can use such arguments to classify defects in nematic liquid crystals, while the classification of defects in smectics and cholesterics requires a combination of topological and geometrical arguments.

As researchers obtain a better understanding of the nature of defects in liquid crystal phases, they are gradually realizing that defects may be important in the statistical mechanics of phase changes. This is particularly true for the two-dimensional cases that Ronald Pindak and David Moncton discuss in their article on page 56, but it may also be so for the transition from the smectic-A phase to the nematic phase.[8] In addition, stable arrays of line defects have been proposed as a description of the so-called "blue phases" of cholesterics,[9] which we will discuss later.

Nematic phase defects

One characterizes the nematic phase by the direction of orientation of the long axes of the rod-like molecules. Because this direction may vary from place to place in a sample, we write the director as a function of position: $\hat{\mathbf{n}}(\mathbf{r})$. At defects in the liquid crystal, the director cannot be defined uniquely. There are two main classes of defects, or singularities: line defects, which are lines along which $\hat{\mathbf{n}}$ cannot be determined, and point defects, which are points where $\hat{\mathbf{n}}$ cannot be determined. Because the ends of the molecules are not distinguished, $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$ are equivalent. This equivalence essentially determines the only line defect that can exist.

Nematic line defects are referred to as disclinations[10] to indicate discontinuities in the "inclination" of the molecules. One commonly discusses these defects in terms of their behavior in the plane perpendicular to the line. A defect is said to be of strength S if on moving around a closed path in that plane the director rotates by S multiples of 2π. Because $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$ are equivalent, half-integer values of S are allowed. Sir Charles Frank has called such half-integer defects "Möbius defects" because they have the same topology as a Möbius strip. Figure 2 shows a disclination of strength $+\frac{1}{2}$ "frozen" into a liquid crystal by polymerization.[11] The central region, or core, of the disclination is the fine

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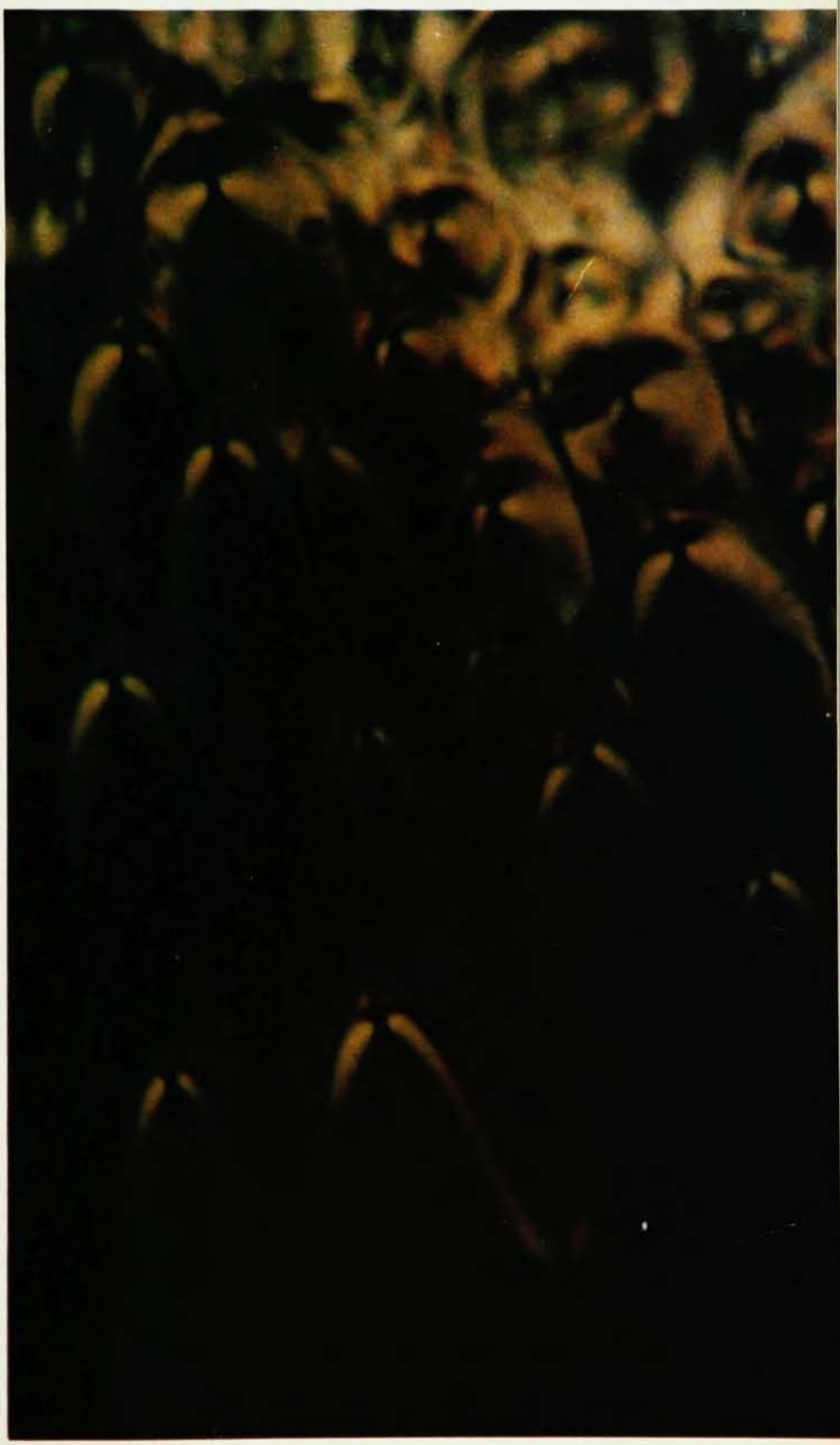
thread running through the material. Because the elastic energy density increases inversely with the square of the distance as one approaches the center of the defect, there is an isotropic core region at the center of the thread. This isotropic region is estimated to be on the order of 50–100 Å in radius. One observes defects of strength $\pm \frac{1}{2}$, but they are not as common as one might expect from the fact that the energy scales as S^2.

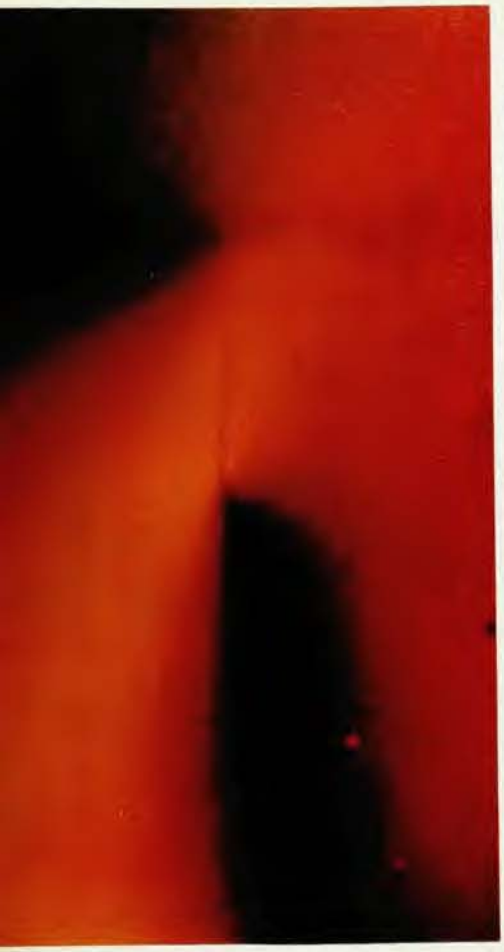
Figure 3 shows line defects of strength ± 1. One identifies these defects by the four dark brushes that become visible when the sample is held between crossed polarizers for observation. In the picture, the string of bubbles generated when the liquid crystal floats on diethylene glycol indicates the behavior of the director.[12] This photograph shows two important effects. The first is that the core region of the defects is quite large. The reason for this is that defects with integral values of S can "escape into the third dimension," that is, the director can simply rotate to lie along the line in the central region, obviating the need for a core.[5] Thus, lines of integral S are not singular. The second important feature in Figure 3 is that the $+1$ and -1 lines cancel the long-range elastic effects of one another. Ignoring the region of "escape," this cancellation gives rise to an attractive interaction between them. The force per unit length between two lines varies as the reciprocal of their separation.[3] The strings are oriented by the elastic forces of the nematic.

A disclination line of unit strength can break up into two lines of half-order strength. Because the line of strength 1 reduces its elastic energy by escaping, it can be favored energetically over two half-order lines.

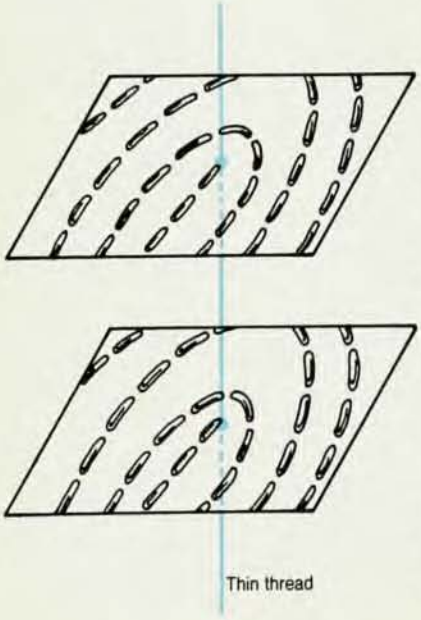
Point defects, such as those shown in figure 4, are another class of defects in nematic liquid crystals. One sees these defects in capillary tubes where the



Focal conic defect texture of liquid crystals in the smectic-A phase. The domains, which are on the order of tens of microns in size, become visible when the sample is placed between crossed polarizers. Each domain is a smectic-A layer suspended between a pair of conic-section curves that have a common focus. The crosses visible in the photograph occur where pairs of curves cross. The cholesteric phase also features focal conic defects, as the cover and figure 6 show. Figure 1



Thin-thread defect "frozen" by polymerization in the nematic phase and placed between crossed polarizers for observation. As the schematic diagram below shows, in the region of the thread each slice of the sample contains a point at which the direction of the long axes of the molecules varies discontinuously. The thin-thread defect is the locus of such singular points. Figure 2



boundary condition is that $\hat{\mathbf{n}}$ is radial at the surface, and where $\hat{\mathbf{n}}$ escapes in opposite directions in different parts of the capillary.[13] The two point defects in the figure are of opposite sign in the sense that they cancel each other's long range elastic fields. The total energy stored in the elastic field around a point defect grows linearly with the radius of the volume enclosed. Consequently, two defects have an interaction energy that grows linearly with separation. In this respect, they act like quarks interacting through a gluon field.

In addition to the three-dimensional nematics, one can consider a thin film of smectic-C liquid crystal (a layered structure in which $\hat{\mathbf{n}}$ is oriented at an angle with respect to the layer planes) to be a two-dimensional nematic in the plane of the film. In such films only disclinations of integral strength can exist. Some suggest that pairs of these disclinations dominate the statistical mechanics near the "nematic" to "isotropic" (really smectic-C to smectic-A) transition similar to the two-dimensional superfluid case.[14]

Topological analysis

Although we have learned much about nematic defects from optical studies, from solutions to elastic equations, and from simply drawing pictures, only in the last few years have researchers used topology to introduce[6,7] a general scheme for classifying defects into condensed-matter physics. The classification of defects in nematic liquid crystals represents a straightforward example of the applications of homotropic group theory. The applicability of homotropic theory becomes much less obvious for liquid-crystal phases with more complicated order parameters. We will discuss some of these phases in the following sections.

Here we give a very simplified discussion of the application of the theory to nematics. Those interested in greater detail might want to consult N. David Mermin's review of the subject.[15] One can quantify the "texture" of a nematic by specifying the orientation of the director at each point in the liquid. The set of all possible director orientations can be thought of as the set of points on a unit sphere. A configuration where the director is constant throughout a sample corresponds to a single point on the sphere and is called the uniform configuration. Because of the symmetry between $\hat{\mathbf{n}}(\mathbf{r})$ and $-\hat{\mathbf{n}}(\mathbf{r})$, the fundamental set of allowed values of $\hat{\mathbf{n}}$ (denoted by V) lie on a hemisphere as shown in figure 5. We can think of the definition of a texture in real space as a mapping of the points in real space ($\mathbf{r}$) onto the fundamental order-parameter space, V. The topological approach to the problem of classifying defects, then, is to study the classes of such mappings.

To determine the possible line defects, one considers all possible mappings of closed lines onto the order-parameter space, V. Two mappings are said to be in the same homotopic class if they can be continuously deformed into one another. Closed lines in real space that map into closed lines on the surface of the hemisphere of allowed values of $\hat{\mathbf{n}}$ (line 1 in figure 5) do not circumscribe a line defect because the gradual reduction of the loop size in real space ($\mathbf{r}$) will map into a vanishingly small loop in V space. The texture is thus homotopic to the uniform configuration. That is, the liquid crystal texture can be continuously deformed to the uniform configuration. The region between the two singularities in figure 4 is such a texture.

For a *vector* field such as occurs in ferromagnetic materials, V is the whole sphere. In such a case only closed loops exist and there are no line defects. For nematics, however, there is a second class of mappings, which corresponds to lines joining pairs of diametrically opposite but physically equivalent points on the hemisphere (line 2 in figure 5). Such mappings are continuous in real space because of the equivalence between $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$. This class of mappings represents the director configuration around a line defect of strength $\frac{1}{2}$, such as the thread in figure 2. It is easy to convince yourself that all maps fall into one of these two classes. Thus there is only one fundamental line defect in nematics.

These topological arguments regarding line defects in nematics do not reveal much that is new. For example, we have known for some time that all defects of strength $\frac{1}{2}$ are equivalent,[13] and the discussion of the absence of an $S = 1$ line in terms of the escape into the third dimension is not new. However, topological arguments do give a systematic approach and reassurance that higher-order lines do not exist.

We can examine point defects in a similar fashion by considering the mappings of spheres surrounding the defect into the order-parameter space V. It turns out that the number of times the sphere in V is "covered" by the mapping can serve as an index of point defects. Thus, there are an infinity of possible point defects. Energetically, it is unlikely that defects that have a covering greater than one exist, and indeed they are not observed.

We can make these topological arguments more rigorous and sophisticated. The classes of mappings of loops form a group known as the fundamental group of the mapping. The multiplication properties of the fundamental group determine the way in which defects combine.[15] The group for the nematic

ordering, for example, is a two-element Abelian group. As we shall discuss, non-Abelian fundamental groups occur for the cholesteric phase and for biaxial nematics.

Smectic-A defects

We can properly describe the defects of the smectic-A phase of liquid crystals using geometrical arguments rather than topological arguments. In this sense, smectic-A defects represent an opposing extreme to nematic defects, where topology gives the correct classification. Smectic-A phases are layered phases in which the director $\hat{\mathbf{n}}(\mathbf{r})$ of the molecules is perpendicular to the layers. The dominant parameter governing the deformation of material in the smectic phase is the energy required to change the layer spacing. In macroscopic defects, the spacing must be kept constant. This constancy tells us some geometrical characteristics of the possible defects because it implies that neither twist ($\hat{\mathbf{n}}\cdot\nabla\times\hat{\mathbf{n}}$) nor bend [$\hat{\mathbf{n}}\times(\nabla\times\hat{\mathbf{n}})$] distortions are allowed. Thus textures in the smectic-A phase consist of pure splay ($\nabla\cdot\hat{\mathbf{n}}\neq 0$). A defect, or singularity, with pure splay has the director pointing radially outward. We can imagine both a line disclination and a point defect with this property. In smectic-A liquid crystals, the disclination of strength 1 does not escape.[17] In general, the normal to a layer exhibits pure splay if the major radii of curvature of the layer surface do not vary along their respective major axes. Such considerations lead to a description of the possible singularities in terms of equally spaced surfaces known as "Cyclides of Dupin," which in their most general form are the focal-conic singularities that figure 1 shows.[2,16]

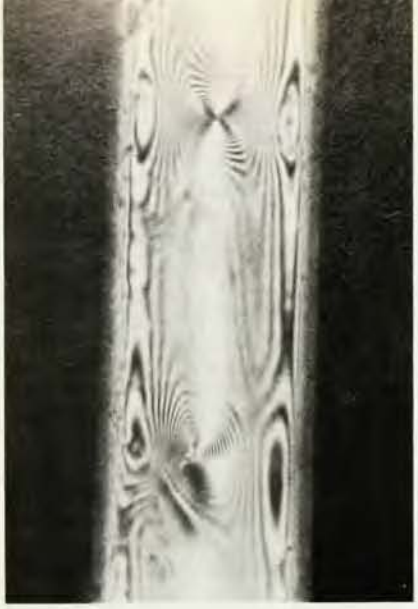
Focal conic defects, as we said earlier, appear as confocal pairs of conic-section curves. All focal-conic singularities are a generalization of the pair of defect lines that one gets by taking equally spaced smectic-A layers, wrapping them into concentric cylinders, bending the result into a torus and then adding more and more equally-spaced layers, going beyond the stage where the hole of the torus vanishes. (The result has a cross-section that looks like a topographical map of twin conical mountain peaks.) One of the resulting defect lines is the circular axis of the torus. The other one is the line normal to the plane of this circle, passing through its center; an infinity of directors meet along this line in a conical fashion.

In the focal-conic generalization of this texture, the circle becomes an ellipse and the straight line a hyperbola that shares a focus with the ellipse. The crosses in figure 1 occur at the intersections of the ellipses and the hyperbolas when they are projected onto a plane. Note that in the focal conic texture, such as shown in figure 1, only the conical volume whose base is the ellipse and whose peak lies somewhere on the hyperbola occurs. The extensive studies of focal conic defects have included the studies of parabolic focal conic arrays induced by dilation of the lyotropic layers discussed by Peter Pershan in this issue (page 34).

In addition to focal-conic singularities, smectic liquid crystals exhibit edge dislocations in the layer spacing—a layer can be added that does not extend throughout the liquid.[18] Such edge dislocations are difficult to observe because the layers are typically 30 Å or less in thickness. The introduction of edge dislocations allows some bend in a texture if the boundary conditions require it.[3]

Topologically, one might expect to be able to describe smectic-A singularities in a fashion similar to those in nematics. Locally, the molecular order of the smectic A is described by the director $\hat{\mathbf{n}}$ and a phase that indicates the position in the layer. Thus, the smectic-A order-parameter space is a product space of the hemispherical shell of

A pair of disclinations, or discontinuities in the "inclination" of the molecules, show up in this nematic liquid-crystal sample that is held between crossed polarizers for observations. Strings of liquid beads, generated at the interface between the liquid crystal and another liquid, reveal the director field—the directions of the long axes of the molecules.[12] Figure 3



Point defects in nematic liquid-crystal material in a capillary tube. The tube is held between crossed polarizers and illuminated with monochromatic light. In the center of the cylinder, the long axes of the molecules are aligned with the axis of the cylinder; however, at the walls the director is radial. Point defects are places where the director "escapes" in opposite directions along the cylinder's axis. Figure 4

the nematic with another space that describes the layering. One can most easily think of this latter space as a ring; positions on the ring specify the phase of the layers. Possible smectic-A line defects separate into two classes. Defects involving $\hat{\mathbf{n}}$ are the same as in a nematic while those involving the phase are the edge dislocations discussed above. However, disclinations of the nematic type cause a change in the layer spacing that does not decay at large distances from the singular line. Topological arguments therefore predict singularities that are energetically not allowed and which do not occur. In addition, these arguments do not predict the dominant defect seen—the focal conic. These omissions occur because the requirement for constant layer spacing is a geometrical constraint, which is much more restrictive than the continuous deformations required by the classification of mappings in topology.[16] As we shall see, the cholesteric phase is intermediate between the smectic and nematic in this regard.

Cholesteric phase defects

In the uniform cholesteric the molecules twist about an axis perpendicular to the director with a definite pitch p that is typically on the order of a few thousand angstroms. (The pitch is the distance along the axis corresponding to a 360° rotation of the director, as the figure on page 40 shows.) Thus to specify the texture of a cholesteric one must specify at each point in space, the director $\hat{\mathbf{n}}$, the twist axis $\hat{\mathbf{t}}$ and the pitch. Both $\hat{\mathbf{n}}(\mathbf{r})$ and $\hat{\mathbf{t}}(\mathbf{r})$ are unit vectors.

If we consider the pitch to be a constant, then we can define $\hat{\mathbf{n}}(\mathbf{r})$ as:

$$\hat{\mathbf{n}}(\mathbf{r}) = \hat{\mathbf{n}}_0(\mathbf{r})\cos[(2\pi/p)\hat{\mathbf{t}}\cdot\mathbf{r}] + \hat{\mathbf{n}}_0(\mathbf{r})\times\hat{\mathbf{t}}(\mathbf{r})\sin[(2\pi/p)\hat{\mathbf{t}}\cdot\mathbf{r}]$$

where at each point in space we have specified an orthogonal set of coordinates $\hat{\mathbf{n}}_0$, $\hat{\mathbf{t}}$ and $\hat{\mathbf{n}}_0\times\mathbf{t}$. Then the functions $\hat{\mathbf{n}}_0(\mathbf{r})$ and $\hat{\mathbf{t}}(\mathbf{r})$ should vary slowly compared to the scale of the pitch. Note that because $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$ are equivalent, so are $\hat{\mathbf{n}}_0$ and $-\hat{\mathbf{n}}_0$, and $\hat{\mathbf{t}}$ and $-\hat{\mathbf{t}}$. This description reduces the cholesteric to a biaxial nematic[7,15,19] at least from a topological point of view.

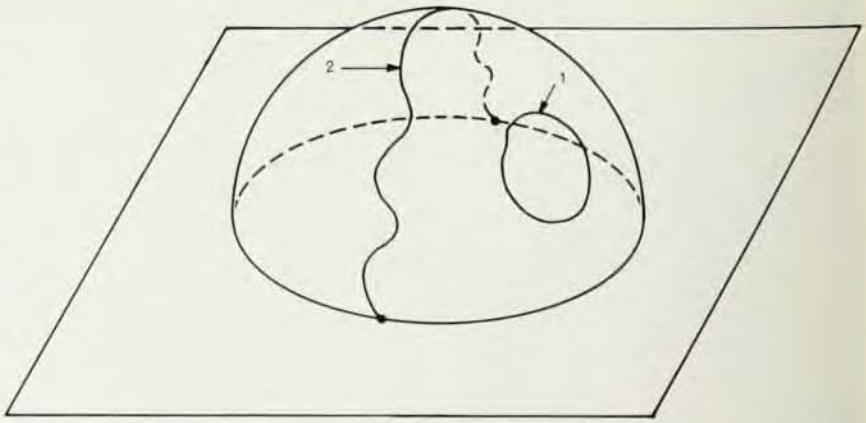
In a biaxial nematic our orthogonal set of coordinates would be the major axes of the dielectric tensor ϵ. The order-parameter space is then the set of all rotations in three dimensions, SO(3), reduced by the symmetry relations of the axes. The mappings of loops in order-parameter space give rise to a description of the line singularities in terms of the quaternion group, whose five elements we can represent by the three Pauli matrices, the identity matrix and its negative. Consequently, there are three line defects, τ, λ and χ, involving 180° rotations about the three inequivalent axes, and a class of line defects of equivalent 360° rotations.[4,7] The so-called $\tau(180°)$ line is a defect in which $\hat{\mathbf{n}}_0\times\hat{\mathbf{t}}$ is the axis about which the coordinate system rotates. The $\lambda(180°)$ line has $\hat{\mathbf{n}}_0$ as the axis of rotation. The third line, conventionally called χ, involves a rotation about $\hat{\mathbf{t}}$ and is simply a strength-½ nematic disclination (as shown in figure 2) in which the director twists about $\hat{\mathbf{t}}$. Because there is an orthogonal set of axes, one cannot rule out a strength 1 or 360° singular line as one could in the nematic phase. However, the continuous deformations of the director that are equivalent to "escape" into the third dimension make all 360° lines equivalent.

Cholesteric line defects are particularly interesting because the quaternion group is non-Abelian, a property that has many interesting consequences when one considers combining defects or entanglements of defects.[15] Unfortunately, this description suffers at least partially from the same difficulty as the topological description of smectic A.

Some of the possible arrangements of either the λ or τ defects have elastic energies that increase with volume because the pitch is modified out to very large distances from the core of the disclination. Although this energy is smaller here than in smectics, it nevertheless is clearly prohibitive as isolated λ and τ lines do not exist in nature, as far as we are aware.

A proper description of the cholesteric, therefore, is similar to that of the smectic as far as the constraint of constant layer spacing is concerned. Indeed, cholesterics show focal conic structure similar to smectics. (See the cover photo.) Because the pitch is often in the visible region of the spectrum, one can see the line singularity in more detail, as illustrated by figure 6, which shows a focal conic defect sliced close to one of the conic-section lines. The line along which the layers rapidly bend in this figure is very nearly the hyperbola characteristic of the focal conic.

In addition to focal conic defects, cholesteric liquid crystals show edge dislocations in which a twist with an extra multiple of π is introduced. Figure 6 shows one such edge dislocation near the lower left-hand corner where three lines merge into one. Such dislocations are among the most common,



Hemisphere of allowed directors. A nematic liquid crystal texture is a set of molecular orientations, which one can represent by a mapping onto a hemisphere. Each orientation corresponds to a point on the hemisphere. Only two kinds of topologically distinct closed curves can be drawn on this surface: loops (the line labelled 1) and lines that join a pair of diametrically opposite but physically equivalent points (the line labelled 2). Figure 5

Inside a focal conic defect in a cholesteric liquid-crystal polymer.[11] This slice through the defect is shown as it appears when held between crossed polarizing filters. The distance between two "stripes"—about 2 microns here—is a half pitch, as illustrated in the figure on page 40. This is the cholesteric analog of the smectic layer. As the cholesteric planes turn to line up with the polarizers, the contrast changes. A strong maximum in the twist axis occurs at A. The further the slice is from the focal line, the weaker this curvature. At I there is a dislocation composed of two λ defects, of strength $\frac{1}{2}$ and $-\frac{1}{2}$. Figure 6

probably because λ defects are not singular energetically. In analogy to the description of dislocations in crystals, we can describe edge dislocations in terms of bound pairs of disclinations.

The cholesteric phase is different from the smectic phase in that it is strongly anisotropic in the planes of the layers because the true local anisotropy axis is the director. Thus, there can be χ defects[3] that in any given plane are simply nematic defects of strength $\frac{1}{2}$ and 1. The intralayer anisotropy also excludes the possibility of point singularities in which the layers form concentric spheres. This prediction is a result of the topological arguments given above. Thus, as far as defects are concerned, we can perhaps best think of the cholesteric liquid crystal as a smectic with an in-plane nematic behavior similar to that of the smectic-C phase, where the long axes of the molecules tilt at an angle with respect to the layer spacing.

Cholesteric blue phases

An interesting phenomenon in cholesterics is the appearance of the "blue phases" when the temperature is sufficiently close to that of the cholesteric–isotropic phase transition. Blue phases—there are three of them—were observed when liquid crystals were first discovered in the last century.[20] These phases exhibit a three-dimensional, cubic periodic variation on the scale of the cholesteric pitch,[21,22] when the pitch is shorter[23] than 7000 Å. Figure 7 shows a micrograph of a typical blue phase. Because of their three-dimensional structure, these phases exhibit many of the characteristics of ordinary crystals. Some authors think[9] that at least one of the blue phases involves cubic arrays of disclinations of strength $-\frac{1}{2}$.

The existence of the blue phases is related to the handedness, or chirality, of molecules in cholesterics.[24] For the present purposes, we can think of chiral molecules as screws that pack together more tightly if their long axes are tilted slightly relative to one another. In the cholesteric phase this tilting occurs in only one direction, but locally the molecules could lower their free energy if the tilt were in two or more directions. Thus, ideally, chiral molecules would like to twist doubly everywhere. However, it is topologically not possible for the director to twist doubly everywhere in space. Therefore the regions of double twist are compensated by singular lines.[9] Theoretical work on various cubic structures appears consistent with experimental observations.[9,22,23] The blue phases may be rare examples in nature of ordered arrays of defects that are thermodynamically stable.

Besides the four phases that we have discussed in this article—nematic, smectic A, cholesteric and cholesteric blue—there are a number of other phases—smectic C, discotic, and biaxial nematic—which exhibit different sets of defects. In addition, surfaces can induce new classes of defects depending on the boundary conditions that they impose on the order parameter. This allows us to study a rich variety of defects that have analogs in other fields. For example, the algebra of line defects of biaxial nematics is similar to that of three colored quarks, and, as we noted earlier, the force between point defects in nematics is independent of distance as are the forces between quarks induced by the gluon field. The defect structures of biaxial nematic liquid crystals are similar to, although not the same as, the superfluid-A phase of helium-3.

Studies of these diverse systems have enhanced our understanding of liquid crystals and vice versa. One of the important lessons is that even though topological theory gives a beautiful description, it must be confronted with geometrical constraints.[16] For example, the famous point-singularity, or "boojum" structure of $He^3(A)$ proposed for cholesterics, has not been observed experimentally. Lewis Carroll may have anticipated this when he wrote in *The Hunting of the Snark*,

> Then the ominous words "It's a Boo—"

The blue phase in a cholesteric liquid-crystal sample held between crossed polarizers. When the cholesteric pitch is less than 7000 Å, and when the sample is very close to the transition to the isotropic phase, the cholesteric transforms to this platelet texture, which is optically active but isotropic. The platelets are a cubic arrangement of cholesteric defects. The small pits at the platelet surface and grain boundaries are points where dislocations emerge. (Photography courtesy of Matthew Marcus.) Figure 7

Then, silence. Some fancied they
heard in the air
A weary and wandering sigh
That sounded like "—jum!" but the
others declare
It was only a breeze that went by.

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