

Liquid Crystals – The ‘Fourth’ Phase of Matter

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The remarkable physical properties of liquid crystals have been exploited for many uses in the electronics industry. This article summarizes the physics of these beautiful and complex states of matter and explains the working of a liquid crystal display.

What are Liquid Crystals?

The term ‘liquid crystal’ is both intriguing and confusing; while it appears self-contradictory, the designation really is an attempt to describe a particular state of matter of great importance today, both scientifically and technologically. Thermodynamic phases of condensed matter with a degree of order intermediate between that of the crystalline solid and the simple liquid are called liquid crystals or mesophases. They occur as stable phases for many compounds; in fact one out of approximately two hundred synthesized organic compounds is a liquid crystalline material. The typical liquid crystal is highly anisotropic – in some cases simply an anisotropic liquid, in other cases solid-like in some directions.

Liquid-crystal physics, although a field in itself, is often included in the larger area called ‘soft matter’, including polymers, colloids, and surfactant solutions, all of which are highly deformable materials. This property leads to many unique and exciting phenomena not seen in ordinary condensed phases, and possibilities of novel technological applications.

Liquid crystalline materials have been observed for over a century but were not recognized as such until the 1880s. The most significant breakthrough came in 1888 when an Austrian botanist named Friedrich Reinitzer (credited for the first systematic report of the phenomenon) observed that a material known as cholesteryl benzoate had two distinct melting points. In his

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experiments, Reinitzer increased the temperature of a solid sample and watched the crystal change into a hazy liquid at 145.5°C. As he increased the temperature further, the material changed into a clear, transparent liquid at 178.5°C. The color of the turbid liquid also changed from red to bright blue-violet to pale blue, and the whole process was reversible. Reinitzer sent his early work to Otto Lehmann, a professor of natural philosophy (physics) in Germany. Lehmann had constructed a polarizing microscope with a stage to control precisely the temperature of his samples. He examined Reinitzer's substance with his microscope and noticed its similarity to other samples he was studying then. The term 'liquid crystal' was coined by him in 1900 (although at first he called them 'flowing crystals' (1889) and 'crystalline solids' (1890)).

Slowly the study of this phase spread to all the continents and there was an explosive growth during the 1970s and 1980s. Theories for the liquid crystal phases added a new dimension of study and the invention of liquid crystals displays gave the field a practical dimension. Scientific studies of liquid crystals involve both the chemistry and physics of this state, being concerned with liquid crystal synthesis and investigation of structure-property relationships. Technologically, liquid crystals have become a part of our lives, first showing up in wrist-watches and calculators, but now being used for all kinds of advanced instrumentation, including laptops and futuristic flat panel displays. Their advantage was first their low power consumption and small size; now they are competitive with other technologies for attractiveness, ease of viewing, cost and durability.

Classification of Liquid Crystals

The distinguishing characteristics of phases of condensed matter are

- Positional order
- Orientational order.

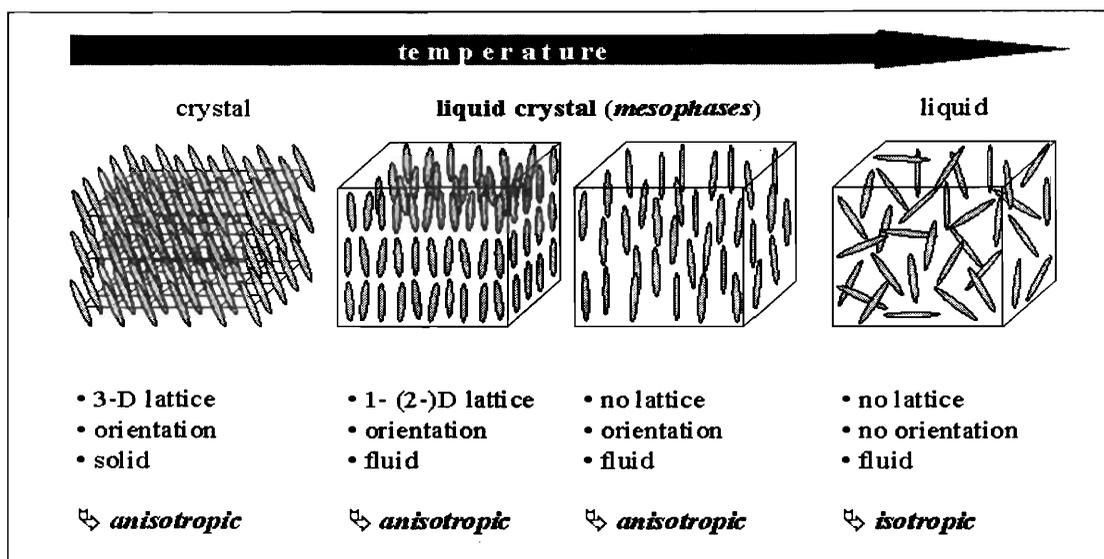


Figure 1. Solids, liquid crystals and liquids – the molecular picture.

Positional order refers to the extent to which molecules or groups of molecules, on average, show translational symmetry. Orientational order refers to the extent to which the molecules align along a specific direction on a long-range basis.

In a crystalline solid, molecules are ordered in both the above ways, i.e. they are constrained to occupy specific sites in a lattice *and* to point their molecular axes in specific directions. On the other hand, molecules in a liquid diffuse randomly throughout the sample without any orientational order. Thus, a crystal has orientational and three-dimensional positional order, whereas a liquid has none.

In contrast, liquid crystal phases have long range orientational order, but their long range positional order is generally at most two dimensional, although some exotic, very anisotropic three-dimensionally ordered structures such as the smectic B and the twist-grain boundary phase are included in this family.

Depending on the degree of positional order, different liquid crystal phases arise, e.g.

- no positional order, giving rise to the nematic phase;

- one dimensional positional order, giving rise to smectic phases;
- two dimensional positional order, giving rise to the columnar phase.

Additionally, liquid crystals are also classified into:

Thermotropics: These have small organic molecules, usually rod-like or disk-like, which show mesomorphic behaviour as a function of temperature.

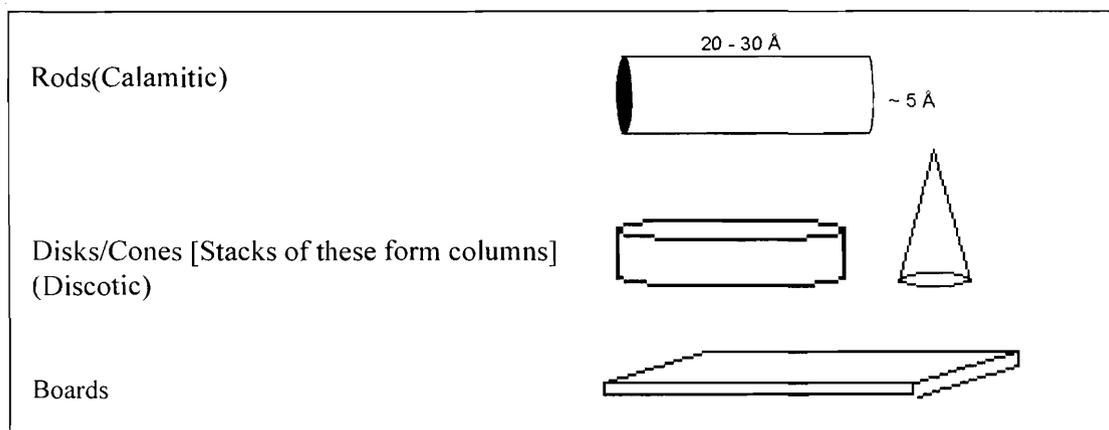
Lyotropics: These contain mixtures of organic molecules which show mesomorphic behaviour as a function of concentration of one or more of the molecular species in the mixture, as well as temperature.

Liquid crystalline phases occur most readily in systems whose molecules have shapes that favors parallel packing. A few representative shapes are shown in *Figure 2*.

Experimental Identification of Liquid Crystals

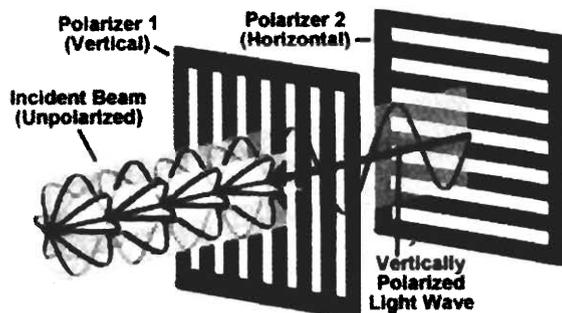
Liquid crystal phases can be identified by a variety of techniques like optical polarizing microscope, differential scanning calorimetry, X-ray analysis, miscibility studies, neutron scattering studies and nuclear magnetic resonance. A few of these are described here.

Figure 2 . Shape anisotropies of liquid crystalline molecules.



Box 1. Polarization

Light Passing Through Crossed Polarizers



Normal (unpolarised) optical radiation has light waves whose electric field vectors vibrate in all perpendicular planes with respect to the direction of propagation. When the electric field vectors are restricted to vibrate in a single plane by polarizing filters, then the light is said to be plane *polarized* with respect to the direction of propagation.

Differential Scanning Calorimetry (DSC)

Heat is needed to melt a crystalline solid to a liquid crystalline phase. The heat is measured using a DSC instrument. Although DSC cannot identify the type of phase, it provides valuable information like the exact transition temperatures and the enthalpies of the different phases.

Polarizing Microscope

In a polarising microscope, the light is polarized (*Box 1*) by passing it through a polarizing filter. It then passes through the sample, and then through a second polarizing filter called the analyzer. When a liquid crystal material is placed on a microscope slide with a cover slip and the slide is heated and viewed using a polarizing microscope, textures characteristic of each type of liquid crystal can be seen. Cooling the liquid can also yield these textures when liquid crystal phases are present.

X-ray Crystallography

This can be used to study the extent of translational or positional order, and thus infer the type of liquid crystal phase.

The Thermotropic Mesophases

Friedel (1922) classified thermotropic liquid crystals broadly into nematic, cholesteric, and smectic. As time went on, more and more complex, exotic phases were discovered.

The nematic phase is the simplest liquid crystalline phase exhibited by long rod-like organic molecules. The word nematic stems from the Greek word *νημα* (nema) which means 'thread' referring to the thread-like structures seen in this phase when viewed through the polarizing microscope. This phase possesses orientational order of the long axes of the molecules about a particular direction referred to as the director \hat{n} . The director is apolar in nature (\hat{n} is physically indistinguishable from $-\hat{n}$) and hence nematics are not ferroelectric.

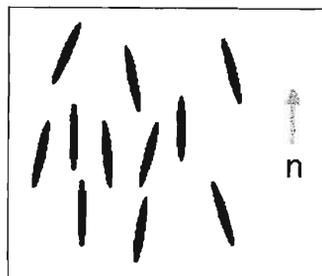


Figure 3. Nematic phase.

Cholesteric liquid crystals or chiral nematic liquid crystals are formed by optically active molecules. In this phase the director is no longer constant, but forms a helical structure causing the chirality of the phase. Due to the apolar nature of \hat{n} , the periodicity of the medium along the helical axis is half the pitch of the helix. (Chirality: A property in which the mirror image of the molecule cannot be superimposed on it.)

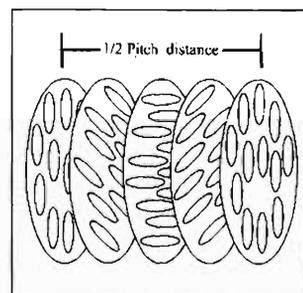


Figure 4. Cholesteric phase.

The word smectic means 'soap-like' in Greek. Smectic liquid crystals exhibit long range orientational order like the nematics and, in addition, layering perpendicular to some direction. There are three main types of smectic phases: A, C and C*.

Figure 5. Smectic phases.

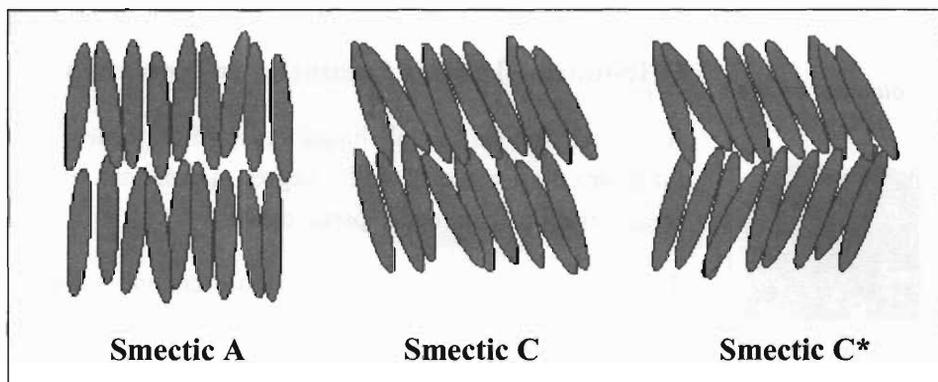
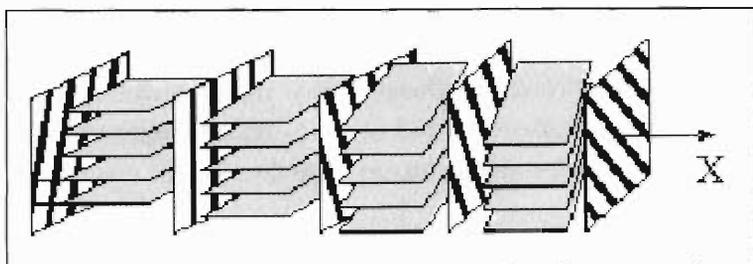


Figure 6. Twist grain boundary phase.



In smectic A, the director is normal to the plane of the layers. The phase has rotational symmetry about the director. Smectic C is similar to smectic A except that the director is tilted with respect to the layer normal. The (chiral) smectic C* phase is formed by optically active molecules or when smectic C is doped with a chiral substance.

The twist grain-boundary phase is formed when the layers of a smectic A phase are forced to twist by the presence of chiral molecules. The system accommodates the conflicting demands of constant layer spacing and a net twist by breaking up (see *Figure 6*) into a sequence of blocks, each with intact layers and no twist, but each rotated by a fixed angle relative to its neighbouring blocks. The interface between adjacent blocks is a twist grain boundary.

Columnar/discotic phases arise when disk-like molecules are stacked together in columns. These columns, which are liquid-like, are packed together to form a two-dimensional crystalline array. The arrangement of the molecules within the columns and the arrangement of the columns themselves leads to new mesophases.

Figure 7. Columnar phase.



Orientalional Order Parameter in Nematics

As mentioned earlier, all liquid crystalline phases have long-range orientational order. This degree of order can be quantified by an 'orientational order parameter'.

The point is that even when the liquid crystal is oriented, e.g. vertically as in the nematic phase sketched in *Figure 1c*, all the molecules are not oriented vertically or completely parallel to

the z-axis. Rather, there is a distribution of tilt angles. If we define the tilt angle by θ as is sketched in *Figure 8*, we can define the order parameter for the nematic orientational order (for cylindrically symmetric molecules) in the following way:

$$S = (1/2) \langle 3 \cos^2\theta - 1 \rangle,$$

where $\langle \rangle$ denotes the average of the quantity enclosed inside. Since $\langle \cos^2\theta \rangle = 1/3$ in the isotropic state, $\langle \cos^2\theta - 1/3 \rangle$ is a good measure of the anisotropy. The order parameter S can take values in the range $-0.5 \leq S \leq 1$. We note that in general the average of a variable A that depends on the orientation of the molecules can be expressed as:

$$\langle A \rangle = \int_0^{2\pi} d\varphi \int_0^\pi \sin\theta d\theta A \cdot f(\theta, \varphi),$$

where $f(\theta, \varphi)$ is the distribution function, specifying the probability that a molecule is oriented with polar angles (θ, φ) . It is assumed to be normalized according to

$$\int_0^{2\pi} d\varphi \int_0^\pi \sin\theta d\theta f(\theta, \varphi) = 1.$$

In nematic liquid crystals, $f(\theta, \varphi)$ does not depend on the polar angle (φ) , and we can write $f(\theta, \varphi) = (1/2\pi) \tilde{f}(\theta)$ with $\int_0^\pi \tilde{f}(\theta) \sin\theta d\theta = 1$.

In the isotropic phase, with the molecules completely randomly oriented, $\tilde{f} = 1/2$ and $\langle \cos^2\theta \rangle = 1/3$; hence $S = 0$. In the fully aligned state with all molecules oriented parallel to the z-axis, $\langle \cos^2\theta \rangle = 1$ and $S = 1$. If, however, the orientation of the molecules is only 'on average' parallel to the z-axis with some mean square deviation, the order parameter will be $S < 1$.

Note that the nematic order parameter defined here can be expressed also as the expectation value or 'moment' of the second Legendre polynomial as $S_2 = \langle P_2(\cos\theta) \rangle$. The reason for this is that we can expand $\tilde{f}(\theta)$ in terms of the Legendre polynomials, $P_m(\cos\theta)$. The coefficients of the expansion are $S_m \equiv \langle P_m(\cos\theta) \rangle$. S_0 is 1 by the normalisation condition, and

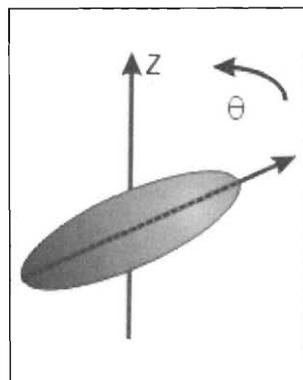
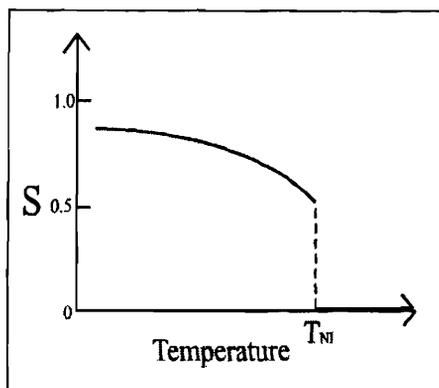


Figure 8. Molecule orientation.

$\langle \cos^2\theta - 1/3 \rangle$ is a good measure of the anisotropy.

Figure 9. Variation of 'S' with 'T'.



$S_1 = 0$ because only the molecular *axes* are oriented, not the *vectors* from tail to head. Hence $S_2 = S$ is the leading coefficient characterising $\tilde{f}(\theta)$.

The order parameter S is a function of temperature. As the temperature increases, due to increased kinetic motion of molecules, the amount of disorder in the system increases. As a result, the order parameter decreases. T_{NI} denotes the nematic-isotropic transition temperature. As soon as the liquid crystal enters the isotropic phase, $S = 0$. At the isotropic-nematic transition, which is a first order transition, the order parameter jumps abruptly to values ~ 0.3 to 0.4 and it can go up to 0.8 on cooling further.

Optical Properties – Birefringence

Perhaps the most remarkable features of liquid crystals, crucial for their applications, are their anisotropic optical properties. A uniaxial liquid crystal is birefringent, meaning that it possesses two different indices of refraction. One index of refraction corresponds to light polarized along the director of the liquid crystal, and the other is for light polarized perpendicular to the director. Light propagating along a certain direction has electric and magnetic field vectors perpendicular to that direction. Supposing that the director lies perpendicular to the direction of propagation of the light, we can think of each field vector as being made up of two components, one parallel to the director of

At the isotropic-nematic transition the order parameter jumps abruptly.

the liquid crystal and one perpendicular. Essentially, what this birefringence property amounts to is that these two components of either the electric or the magnetic field will propagate through the liquid crystal at different speeds and, therefore, can be out of phase when they exit the crystal. As the temperature increases, the birefringence of the sample decreases and is zero at the nematic-isotropic transition.

Birefringence can be manipulated easily with the help of rather weak fields.

The orientational order and hence the birefringence can be manipulated easily, for example, with the help of rather weak magnetic, electric or optical fields, leading to huge magneto-optical, electro-optical and opto-optical effects. The most successful application of liquid crystals, namely displays in wrist watches, pocket calculators or flat screens of laptop computers, take advantage of electro-optical effects. More recently, it has been shown that the orientational order can be also affected by optical fields, leading to rather sensitive opto-optical effects and nonlinear optical properties which are important for optical switching and other photonic devices in future optical information technologies.

External Influences on Liquid Crystals

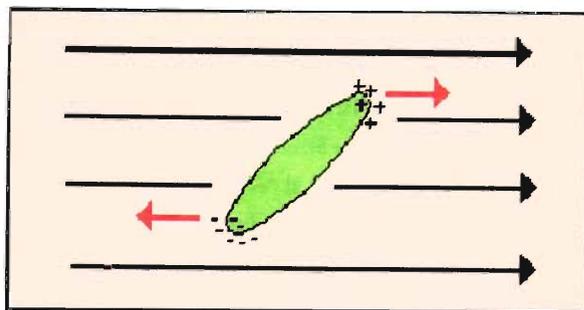
Scientists and engineers are able to use liquid crystals in a variety of applications because external perturbations can cause significant changes in the macroscopic properties of the liquid crystal system by forcing the director to align in specific directions. Both electric and magnetic fields can be used to induce these changes. The magnitude of the fields involved, as well as the speed at which the molecules re-align when the fields are changed, are important characteristics the industry deals with. Also, surface preparations can be used in liquid crystal devices to force specific orientations of the director.

Effects of Electric Field

The response of liquid crystal molecules to an electric field is the major characteristic utilized in industrial applications. The ability of the director to align along an electric field arises from



Figure 10. Influence of external electric field.



the dielectric nature of the molecules. An elongated molecule as in Figure 10 has direction-dependent polarizabilities, $\chi_{||}$ along and χ_{\perp} normal to its long axis. The components of the polarization in an electric field \mathbf{E} are simply $\chi_{||} E \cos \theta$ and $\chi_{\perp} E \sin \theta$ along and normal to the axis respectively, where θ is the angle between the axis and the field. Thus the electrostatic energy of such a molecule in an electric field is $-(1/2)(\chi_{||} E^2 \cos^2 \theta + \chi_{\perp} E^2 \sin^2 \theta) = -(1/2)E^2 [(\chi_{||} - \chi_{\perp}) \cos^2 \theta + \chi_{\perp}]$. Usually $\chi_{||} > \chi_{\perp}$, so that the lowest energy is at $\theta = 0$, that is, the molecule aligns with the field. In Figure 10, the black arrows represent the electric field vector and the red arrows show the electric force on the molecule.

Surface Preparations

When a thin polymer coating (usually a polyimide – a coating applied to the surface of the glass cells within which the sample is placed to predetermine surface alignments) is spread on a glass substrate and rubbed in a single direction with a cloth, it is observed that liquid crystal molecules in contact with that surface align with the rubbing direction. The currently accepted mechanism for this is believed to be an epitaxial growth (growth of one crystal on the surface of another crystal) of the liquid crystal layers on the partially aligned polymer chains in the near surface layers of the polyimide.

Fredericksz Transition in Nematic Liquid Crystals

An important example of the electro-optic effect, fundamental to the operation of many liquid crystal displays, is the Fredericksz transition. It is an electric-field induced transition in nem-



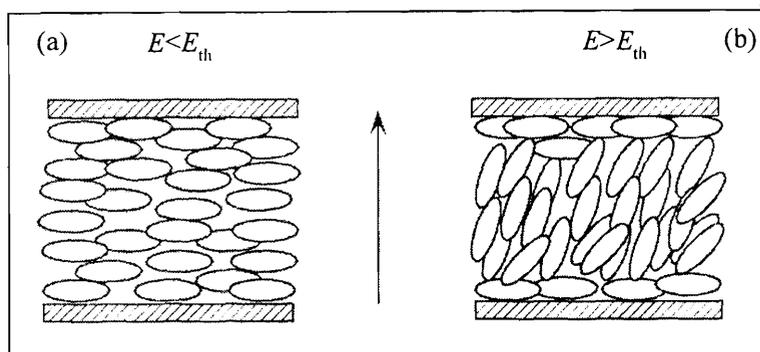


Figure 11. Director orientation (a) below the threshold voltage and (b) above.

atic liquid crystals oriented homogeneously between two glass plates, from an unperturbed to a non-uniformly deformed state.

In the unperturbed state, which is maintained as long as the electric field E is below a threshold value E_{th} , the director (and the optic axis) is uniformly oriented parallel to the glass plate (see Figure 11a). If the sample is kept between two crossed polarizers and the optic axis is rotated by 45° with respect to one of the polarizers, below the threshold voltage the light will pass through the sample. When the dielectric anisotropy of the molecules is positive, if an electric field is applied perpendicular to the glass plate, the molecules try to align along the field direction. In the deformed state which results, many molecules become perpendicular to the glass plate and the intensity falls down. Thus, depending on the *off* and *on* state of the applied field we can get *on* and *off* states in the transmitted intensity.

The threshold field arises due to the competition between the elastic energy cost for the deformation of the director, and the dielectric energy gained by the alignment of the liquid crystal molecules. For a more detailed discussion see *Box 2*.

Textures and Defects in Liquid Crystals

The term ‘texture’ refers to the patterns in the orientation of liquid crystal molecules which arise because of their enforced orientation in the vicinity of a surface. Each liquid crystal mesophase can form its own characteristic textures, which are useful in their identification. We consider the nematic textures here.

Box 2. A Derivation of the Threshold Voltage

Let us assume a small deformation of the director under the applied field.

$$n = n_0 + \delta n(r), \quad (1)$$

where n_0 is the director when there is no deformation, and δn is the change in director orientation on applying the field. δn is normal to n_0 (since $n^2=1$) and is parallel to the applied electric field E . The *distortion energy* per unit volume can be written as (for one elastic constant approximation)

$$F_d = \frac{1}{2}K \left(\frac{\partial \delta n}{\partial z} \right)^2 \quad (\text{Analogous to Hooke's Law, } F = \frac{1}{2}K(\Delta x^2)), \quad (2)$$

where the z axis is the direction of the distortion and perpendicular to the glass plates, and K is the curvature elastic constant of the director. The *dielectric energy* is given by

$$F_e = -\frac{1}{2}\epsilon_a E^2 \delta n^2, \quad (3)$$

where $\epsilon_a(>0)$ is the anisotropy in the dielectric constant.

We assume that at the boundary ($z=0,d$) the molecules are so strongly anchored that δn vanishes. It is therefore convenient to express the distortion in terms of the Fourier series

$$\delta n = \sum_q \delta n_q \sin qz, \quad (4)$$

where $q = \nu \frac{\pi}{d}$ (ν is a positive integer). (5)

The total free energy density per unit area is given by

$$F = d (F_d + F_e) = \frac{d}{4} \sum_q \delta n_q^2 (Kq^2 - \epsilon_a E^2). \quad (6)$$

If the unperturbed state is to be stable, the increase in free energy F must be positive for all values of δn_q , i.e.

$$Kq^2 > \epsilon_a E^2 \quad \text{for all } q. \quad (7)$$

Thus only if $F_e > F_d$, we get a deformed state, otherwise not. At the threshold, which corresponds to an instability setting in at the smallest $q=\pi/d$, one has

Box 2. continued...



$$K \left(\frac{\pi}{d} \right)^2 = \epsilon_a E_{\text{th}}^2, \quad (8)$$

giving a threshold field of

$$E_{\text{th}} = \frac{\pi}{d} \sqrt{\frac{K}{\epsilon_a}}. \quad (9)$$

The corresponding threshold voltage is given by

$$V_{\text{th}} = E_{\text{th}} d = \pi \sqrt{\frac{K}{\epsilon_a}}. \quad (10)$$

If mesogenic materials are confined between closely spaced plates with rubbed surfaces (as described above) and oriented with rubbing directions parallel, the entire liquid crystal sample can be oriented in a planar texture. Mesogens can also be oriented normal to a surface with the use of appropriate polymer films, or in the presence of an electric field applied normal to the surface, giving rise to the homeotropic texture (See *Figure 12*).

Topological Defects in Liquid Crystals

When conditions are not amenable for the formation of homogeneous orientation of the director, the textures obtained typically contain many topological defects. A topological defect in a medium with long-range order is a core region – a point, line or plane – where the order changes in a singular way that the order

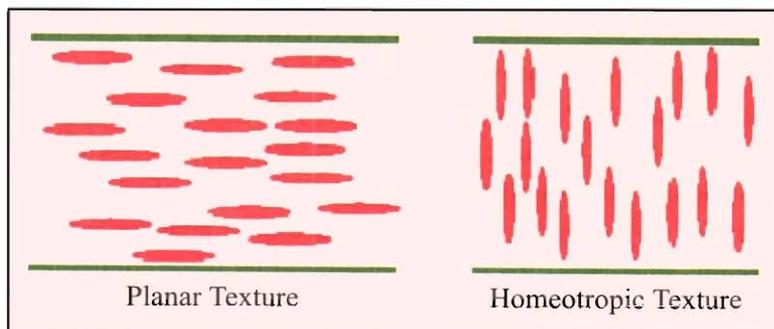
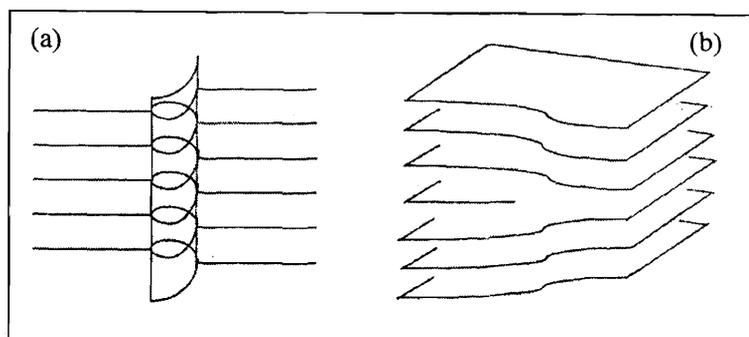


Figure 12. Planar and homeotropic textures.

Figure 13. Types of dislocations: (a) screw dislocations, (b) edge dislocations



parameter is destroyed at the core. A periodic crystal has line defects in its positional order called dislocations in which the phase of the mass density changes by an integral multiple of 2π in one circuit around the core. Liquid crystals like the nematic have line defects in their orientational order called disclinations. Disclinations are characterized by their strength s which is the number of multiples of 2π that the director rotates in a complete circuit around the core. Other liquid crystal phases with both positional and orientational order can have both types of defects.

Figure 13 shows some of the simple defect structures in smectic liquid crystals, called edge and screw dislocations.

Defects and Schlieren Textures

Defects in liquid crystal films can be imaged using transmission polarizing microscopy with the film placed between crossed polarizers (see *Box 1*), which lead to characteristic textures called Schlieren textures, as shown in *Figure 14*. The defect cores appear as black points which have either two or four dark regions, called ‘brushes’ emanating from them. The defect core regions are black because inside them the order parameter is zero, and the liquid in these regions is essentially isotropic. The brushes are black because the director field within them is either parallel or perpendicular to the polarizer or analyzer. Other regions where the director is so oriented that it has components along the polarizer and analyzer rotate the plane of polarization

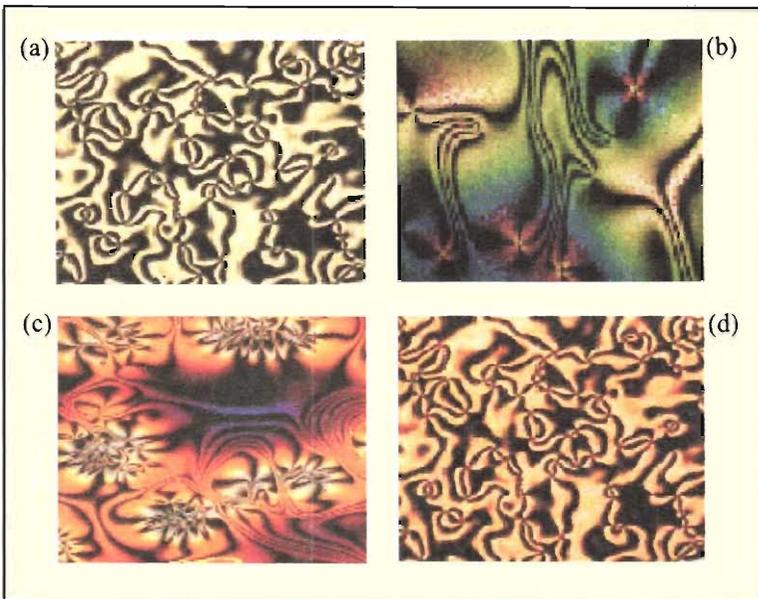


Figure 14. Schlieren textures for a nematic liquid crystal
(a),(d) : Two brush and four brush defects
(b): Four brush defects
(c): Unusual higher order defects

of some light from one to the other. Hence some light passes through and these regions appear bright.

The number of brushes is related to the strength s of a defect defined earlier, according to the relation:

$$\text{number of brushes} = 4s.$$

A defect of strength $s = \pm 1/2$ will have two brushes while one with $s = \pm 1$ will have four brushes. The motion of these brushes under rotating crossed polarizers will be determined by both the sign and magnitude of the defect strength. According to convention, clockwise rotation of the polarizer gives positive 's' values, and anticlockwise rotation negative values. Defects of positive strength will have brushes that rotate in the same sense as the crossed polarizers. Brushes associated with defects of negative strength will rotate in the opposite sense.

Technological Applications

Certain properties like birefringence and chirality make liquid crystals find a numerous range of applications. Typical applications include displays, temperature measurement (by changing



Box 3. Light and Liquid Crystals – A Panoply of Color

The optical properties of liquid crystals such as birefringence are not only interesting but also lead to vividly visible effects. The combination of anisotropy of the shape of the molecules and director configurations varying on a scale equal to wavelength produces a wide variety of marvellous color patterns which can be observed in a polarizing microscope, by placing the sample between crossed polarisers. Some of these are shown here. This property is utilized by all commercial liquid crystal devices.



Samples of liquid crystal films and their colours.

of colors in cholesterics), solvents for NMR, etc. Of these the most well-known application is the liquid crystal-display (LCD), used in watches, calculators, flat panel displays used recently even in TV sets, and so on.

The anisotropic properties of liquid crystals in refractive index and dielectric constant can be exploited for the purposes of creating flat-panel displays. Usually what is done is a thin film of liquid crystal is placed between two pieces of glass or transparent plastic treated for alignment of the medium. These plates are usually manufactured with transparent electrodes, typically made of indium tin oxide, that make it possible to apply an electric field across small areas of the film of liquid crystal. Polarizing filters are usually placed on one or both sides of the cell. Usually these polarizers are crossed, which means that, without the liquid crystal present, light entering from the top of the cell will not be transmitted through the cell due to the crossed polarizers, and no light will be able to pass through the display. The liquid crystal, however, can modify the polarization of the light in

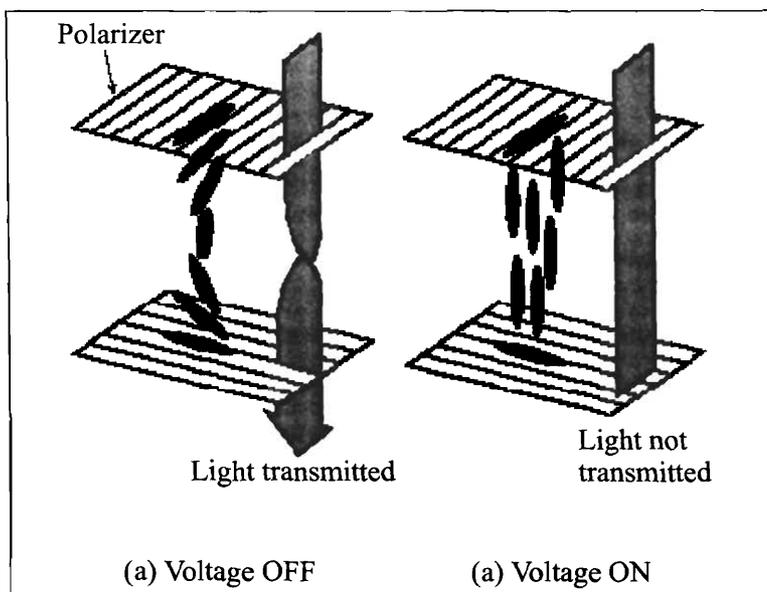


Figure 15. Liquid crystal response, on application of external electric field.

some way that is dependent on the electric field being applied to it. Therefore, it is possible to dynamically create spots where light will get through and spots where it will not.

Specifically, the director is twisted by 90° between the two plates of the cell, and the polarized light from the upper polarizer has its plane of polarization also rotated by 90° and is transmitted through the lower polarizer (analyzer). When a voltage is applied across the twisted nematic (TN) display, the molecules align with the electric field due to a dielectric interaction between the molecules and the electric field. This changes the earlier orientation of the director or the helical structure and the light cannot be transmitted. In a display the zero voltage, transmitting state is white and the non-zero voltage, non-transmitting state is black.

Simple liquid crystal displays use seven segments per numeric character and have one wire connection for each addressable segment. For example, the numeral 8, seen on a calculator screen is made up of 7 discrete segments. So, it requires 7 connections to these lines, and an additional one for the common (ground) electrode. As displays become larger, requiring more characters,

it becomes difficult and uneconomical to make a connection to each segment.

To reduce the number of connections, a matrix addressing scheme has evolved called time multiplexing. For example, in a full graphic display of $(N \times M)$ segments, multiplexing can reduce the number of connections from $(N \times M)$ to $(N + M)$. Thus when the number of characters are large, time multiplexing is an economical way to make a display.

Liquid crystal displays (LCDs) offer several advantages over traditional cathode-ray tube displays that make them ideal for many applications. LCDs are flat, and they use only a fraction of the power required by CRTs. They are easier to read and more pleasant to work with for long periods of time than most ordinary video monitors. Recent advances in technology have improved the quality of the display and reduced the cost of production to such an extent that flat panel TNLCDs are now used in computer monitors as well as commercial TV sets.

Suggested Reading

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