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E. P. Raynes

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Electro-optic and thermo-optic effects in liquid crystals

By E. P. Raynes
Royal Signals and Radar Establishment, St Andrews Road, Great Malvern, Worcestershire WR14 3PS, U.K.

The twisted nematic electro-optic effect has become widely used as a low-voltage, low-power display in watches and calculators; however, it is only one of the many optical effects found in the various liquid crystal phases. Despite this wide variety, certain features of operation are general. Liquid crystals are birefringent, which, together with their ability to align on solid surfaces, allows the construction of thin layers with optical properties reminiscent of single solid crystals. The more common display devices use anisotropic electrical properties to produce electro-optic effects that are seen by using polarized light, scattering, or the absorption of light. Thermo-optic effects can be produced by varying the temperature of the liquid crystal in the vicinity of a phase transition, and are used in thermometry, thermography, and display devices.

1. Introduction

In this paper the major electro-optic and thermo-optic effects in liquid crystals will be reviewed. Selection of effects has been necessary, and some have been dealt with more briefly than their importance may warrant. The first part of the paper examines the building blocks of electro-optic and thermo-optic effects: the anisotropic properties of liquid crystals, ways of observing changes in orientation and construction of thin-layer devices. The second part looks at the various electro-optic and thermo-optic effects in the different liquid crystal phases.

2. Anisotropic properties

The anisotropic physical properties (de Jeu 1980) make liquid crystals both fascinating and useful. I shall now examine the anisotropic physical properties that contribute to display operation (Clark *et al.* 1980).

(*a*) *Refractive index*

The most obvious feature (invariably used by the organic chemist to identify a liquid crystal phase) is the large anisotropy of refractive index, or birefringence, given by

$$\Delta n = n_{\parallel} - n_{\perp},$$

where $n_{\parallel}$ and $n_{\perp}$ are the refractive indices measured parallel and perpendicular to the nematic director $\boldsymbol{n}$; typically $\Delta n \approx 0.25$.

(*b*) *Electric permittivity*

The analogous low-frequency property, the electric permittivity, is also anisotropic:

$$\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp}.$$

Liquid crystal molecules are free to rotate in an applied field, and permanent dipole moments within the molecule contribute to, and indeed usually dominate, the permittivity anisotropy. Cyano groups are frequently used in liquid crystal molecules, and their large dipole moment produces strong anisotropy of the permittivity, giving typically $\epsilon_{\perp} \approx 5$ and $\epsilon_{\parallel} \approx 20$.

The anisotropy of electric permittivity has become extremely important in the last few years because it is the 'driving force' in most electro-optic effects. The electric contribution to the free energy density contains a term that depends on the angle between the director $\boldsymbol{n}$ and the electric field $\boldsymbol{E}$, and is given approximately by

$$F_E = -\tfrac{1}{2}\epsilon_0 \Delta\epsilon\, (\boldsymbol{n}\cdot\boldsymbol{E})^2. \tag{1}$$

In the absence of any other constraints the director will rotate to minimize this contribution to the free energy. There are two possibilities: the director orients parallel to the field for positive anisotropy ($\epsilon_\parallel > \epsilon_\perp$), and perpendicular to the field for negative anisotropy ($\epsilon_\parallel < \epsilon_\perp$). An interesting variation, which can be useful in certain devices (Raynes & Shanks 1974), is the reversal in sign of the permittivity anisotropy (and hence field-induced orientation) with increasing frequency. In liquid crystals the normal Debye relaxation found in isotropic liquids in the region of 100 MHz is severely reduced by the nematic potential to 1 MHz or less (Maier & Meier 1961), and in extreme cases to 1 kHz. This influence of the nematic potential on the relaxation process only applies to the contribution of longitudinal dipoles to $\epsilon_\parallel$, so that $\epsilon_\parallel$ can relax below $\epsilon_\perp$ and the anisotropy change sign.

(c) *Orientational elasticity*

In nematic liquid crystals the preferred orientation is with the director everywhere parallel. All other orientations have a free energy density given by the Frank–Oseen continuum theory (Frank 1958)

$$F_k = \tfrac{1}{2}k_{11}(\operatorname{div}\boldsymbol{n})^2 + \tfrac{1}{2}k_{22}(\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{n})^2 + \tfrac{1}{2}k_{33}(\boldsymbol{n}\times\operatorname{curl}\boldsymbol{n})^2. \tag{2}$$

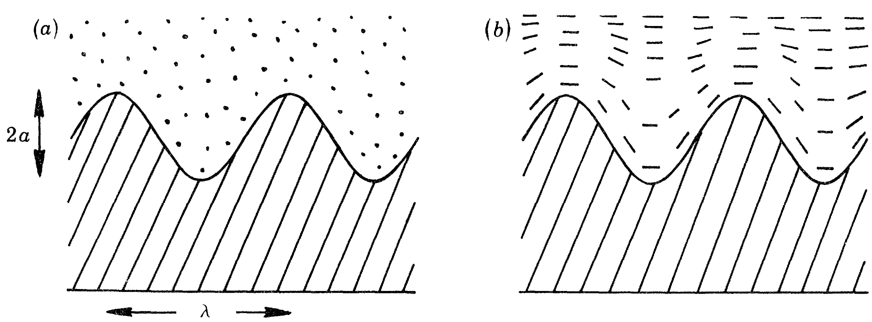

Figure 1. Alignment of a nematic liquid crystal on a microgrooved surface: (a) lowest-energy configuration with the director parallel to the microgrooves; (b) highest-energy configuration with the director perpendicular to the microgrooves.

The orientational elastic constants k_{11}, k_{22} and k_{33} describe the splay, twist and bend deformations respectively, and have a magnitude of 10^{-11} N. The orientation of liquid crystals on a microgrooved surface is an immediate consequence of (2). Surface alignment is usually a result of both the chemical nature and the topography of the interface of the liquid crystal and the surface. If the surface is microgrooved in one direction (figure 1) the difference in free energy between the two configurations is (Berreman 1972)

$$\Delta F_k = (\pi^3 k/2\lambda)\,(2a/\lambda)^2, \tag{3}$$

where k is an average elastic constant. With $k \approx 10^{-11}$ N, $2a/\lambda \approx 0.5$, and $\lambda \approx 0.05\,\mu$m, we find $\Delta F_k \approx 5\times10^{-4}\,\mathrm{J\,m^{-2}}$, the same order of magnitude as the energies associated with surface physico-chemical forces. A thin layer with two aligned surfaces adopts an orientation determined by the two surfaces and is the starting point for most optical effects in liquid crystals.

By adding the electric contribution F_E (equation (1)) to the orientation free energy F_k (equation (2)) it is possible to calculate many useful properties of electro-optic displays (Berreman, this symposium).

3. Visualization of orientational change

There are three main ways in which a change in the orientation of the liquid crystal can be seen by an observer. The changing refractive index can be observed by using two polarizers, one to polarize and one to analyse the light passing through the cell. Secondly, the change in anisotropic light absorption associated with the director reorientation can be observed directly or with a single polarizer. Most liquid crystals are transparent, so absorption is induced by adding pleochroic dye molecules (Heilmeier & Zanoni 1968). The last method of visualizing the director reorientation uses the scattering induced when the director becomes randomly oriented on a scale comparable with the wavelength of light. This scattering texture can be observed without polarizers against the clear aligned background areas.

4. Display construction

Display devices using the various electro-optic and thermo-optic effects have a fairly standard flat panel construction, shown in figure 2 (Clark *et al.* 1980). A thin layer of liquid crystal (*ca.* 10 µm) is sandwiched between two sheets of glass joined together by a thermoplastic or thermosetting seal around the perimeter. Layer spacing is usually controlled by small pieces of glass

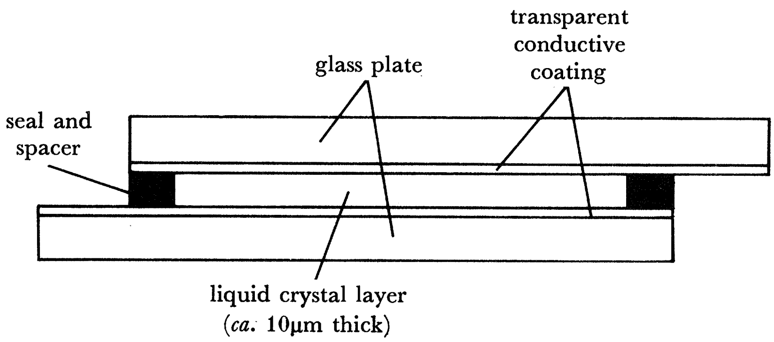

Figure 2. Construction of a typical liquid crystal cell (not to scale).

fibre of the appropriate diameter distributed across the display area. The liquid crystal is vacuum filled through a small gap left in the seal, which is subsequently closed by epoxy resin or solder. A field is applied across the liquid crystal by using inner glass surfaces coated with transparent conductors made of indium tin oxide and etched to provide the required pattern. Frequently a passivation layer is used to prevent damage by ion migration from the glass substrate and to act as a d.c. blocking layer to inhibit electrochemical degradation. Unidirectional homogeneous alignment of the liquid crystal is produced by using microgrooves produced either by a rubbing process or by obliquely evaporated dielectric layers (Janning 1972). Homeotropic alignment of the liquid crystal normal to the surface is achieved by chemical treatment of the surfaces with silane coupling agents (Kahn 1973*b*) or chrome complexes (Matsumoto *et al.* 1975).

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5. Electro-optic effects

We have seen how a thin layer of liquid crystal can be aligned by the surfaces and reoriented by an electric field to produce a visual change in the optical properties. I shall now describe the various electro-optic effects that can be constructed out of these basic elements.

(a) *Nematic liquid crystals*

(i) *Dynamic scattering*

Dynamic scattering displays (Heilmeier *et al.* 1968) are worthy of a brief mention because although they are now rarely used, they were the first generation of liquid crystal displays. Their operation depends on a complex electrohydrodynamic effect (Helfrich 1969) not mentioned above; this disrupts the alignment to produce scattering of light, which is seen against a clear background of uniformly oriented liquid crystal. Dynamic scattering requires materials with $\Delta\epsilon < 0$, and in the early days such materials were unstable and contributed to a short lifetime for the displays. Although suitable stable materials now exist, the performance and use of dynamic scattering displays is limited by a number of factors:

short lifetime, resulting from the high electrical conductivity necessary for adequate scattering;
relatively high power consumption;
poor visual appearance (in reflexion a metallic or dielectric mirror was used to reflect the scattered light, resulting in specular reflexions in unenergized regions and a poor viewing angle and legibility).

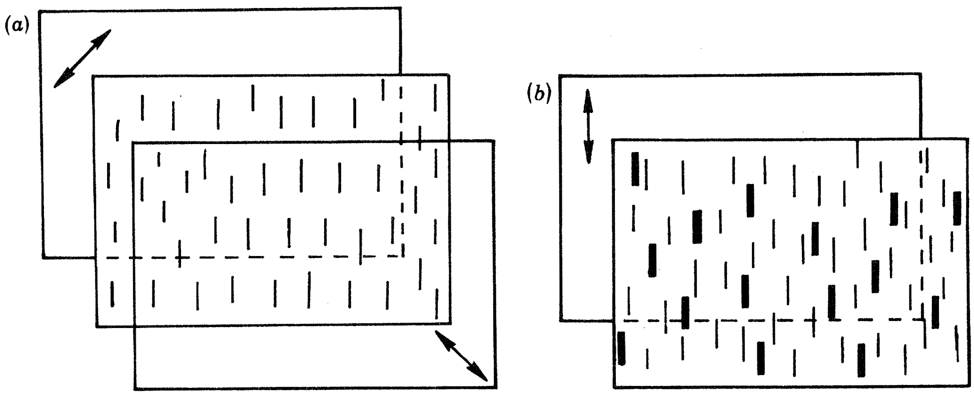

Figure 3. Two electro-optic effects in parallel aligned nematic layers: (*a*) variable birefringence with two polarizers; (*b*) guest–host effect with a single polarizer.

(ii) *Variable birefringence displays*

If a nematic liquid crystal with $\Delta\epsilon > 0$ is aligned unidirectionally in a thin layer by using two parallel aligned surfaces the change in birefringence when a field is applied can be observed by using two polarizers (figure 3*a*). The layer shows field-induced reorientation (Freedericksz & Zolina 1933) above a threshold voltage (*ca.* 1 V) given by

$$V_c = \pi\surd(k_{11}/\epsilon_0\Delta\epsilon). \tag{4}$$

It is also possible to use the opposite effect in a material with $\Delta\epsilon < 0$ in a homeotropically aligned layer. The variable birefringence effect (Schiekel & Fahrenschon 1971; Assouline 1971) has a performance limited by the strong dependence of contrast on wavelength, viewing angle, temperature and layer thickness. Its main use is a transmissive or projection display, particularly for multiplexed displays with a high information content.

(iii) *Single-polarizer guest–host effect*

The construction of this device is similar to the variable birefringence display (§5*a* (ii)) except that a pleochroic dye is dissolved in the liquid crystal and only one polarizer is used (figure 3*b*). The reorientation process is also identical with the threshold voltage given by (4), and the dye absorption decreases as the voltage is raised above V_c. The single-polarizer guest–host display (Heilmeier & Zanoni 1968) is well suited to transmissive or back-lit applications where its wide angle of view is an advantage. The effect just described shows negative contrast with bright characters on a dark background; the contrast is reversed if a material with $\Delta\epsilon < 0$ is used with homeotropic boundary conditions.

(iv) *Twisted nematic effect*

The vast majority of commercial liquid crystal displays use the twisted nematic electro-optic effect. The visualization of the twisted nematic effect uses a particularly subtle optical property not described in §3. The basic construction of the layer uses two aligned surfaces as in §§5*a* (ii) and (iii), but this time the grooving directions are orthogonal, so that the nematic director twists by 90° in going from one boundary surface to the other (figure 4). This twisted structure rotates

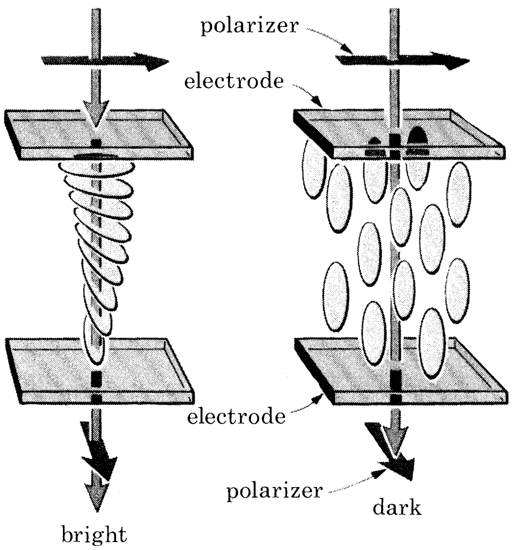

FIGURE 4. Operation of a conventional twisted nematic cell.

the plane of polarized light, which is guided by the twisted birefringent liquid crystal. This unusual property was first observed by Mauguin (1911) who also calculated that the guiding was effective only if

$$d\Delta n > 2\lambda, \tag{5}$$

where d is the thickness of the liquid crystal layer and λ the wavelength of the light. When an electric field is applied the director reorients above a threshold voltage (*ca.* 1 V) given by

$$V_c = \pi\sqrt{\{k_{11} + \tfrac{1}{4}(k_{33} - 2k_{22})/\epsilon_0\Delta\epsilon\}}, \tag{6}$$

and the guiding property is lost (Schadt & Helfrich 1971). This can be understood quite simply from (5) as the director realigns along the electric field, the effective Δn is lowered below the level for guiding. The twisted nematic layer can be used as an electro-optic shutter or more commonly as a display device.

Normally, twisted nematic devices show degeneracy of twist and tilt producing an unacceptably patchy contrast. These problems are overcome by using very long-pitch cholesteric materials and particular combinations of twist and surface tilt (Raynes 1975). The finite pitch P modifies the threshold voltage (Raynes 1975),

$$V_c = \pi \sqrt{\frac{k_{11} + \frac{1}{4}(k_{33} - 2k_{22}) + 2k_{22}\, d/P}{\epsilon_0 \Delta\epsilon}}, \tag{7}$$

and slightly degrades the optical performance (Raynes 1979), producing an increased transmission (T) between crossed polarizers calculated by the author to be given approximately (for $V \gg V_c$) by

$$T \approx (2\pi d V_c / PV)^2. \tag{8}$$

The guiding is independent of wavelength above the limit set by (5) and therefore the displays are not coloured and have a high contrast ratio (more than 10:1). Good operation is achieved at low voltages (*ca.* 2 V) with a lower power consumption ($< 1\,\mu\mathrm{W\,cm^{-2}}$). This performance, together with the long lifetimes now available, has contributed to making the twisted nematic display the leading low-voltage low-power display technology.

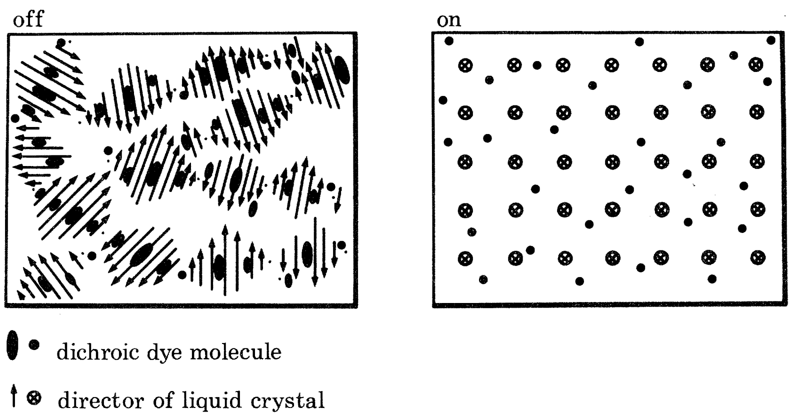

FIGURE 5. Operation of a cholesteric–nematic phase-change cell with dye: (*a*) no applied field; (*b*) electric field applied perpendicular to plane of figure.

(*b*) *Cholesteric liquid crystals*

The major electro-optic effect in cholesteric liquid crystals uses materials with $\Delta\epsilon > 0$ that have pleochroic dyes dissolved in them. The natural pitch (P) of a cholesteric material is typically less than 10 µm, and the rate of twist of the director is therefore too rapid for guiding to occur (this can be seen by putting $d = \frac{1}{4}P$ in (5)), and consequently a dissolved pleochroic dye can absorb all polarizations of incident light (White & Taylor 1974). An electric field reorients the director, a process regarded as unwinding of the cholesteric helix in short-pitch materials (figure 5) and called the cholesteric–nematic phase-change effect (Wysocki *et al.* 1968). The threshold field for this unwinding (Meyer 1968; de Gennes 1968) is given by

$$E_c = (\pi^2/P)\sqrt{(k_{22}/\epsilon_0 \Delta\epsilon)}, \tag{9}$$

and for $P \approx 3\,\mu\mathrm{m}$, $\Delta\epsilon \approx 10$, and $k_{22} \approx 10^{-11}\,\mathrm{N}$ corresponds to a threshold voltage of 10 V in a 10 µm layer. In longer pitch systems the reorientation is more nematic-like in nature (Suzuki *et al.* 1981) with a threshold voltage given approximately by (7) and $V_c \approx 2$ V. In both cases adequate contrast can be achieved without polarizers and the brightness and viewing angle are significantly better than for twisted nematic displays.

The cholesteric displays just described show clear characters on a coloured background. The contrast can be reversed by a number of specialized display construction techniques or by using materials with $\Delta\epsilon < 0$.

(*c*) *Smectic liquid crystals*

Smectic liquid crystals are attracting increasing interest for electro-optic applications because they can be switched in quite short times and usually show long-term memory, making them useful in multiplexed displays of high complexity.

(i) *Smectic A scattering effects*

By applying a low-frequency voltage (*ca.* 100 V) to an aligned smectic A layer with high electrical conductivity, permanent scattering somewhat reminiscent of dynamic scattering in nematics can be induced in quite short times (10–100 ms) (Tani 1971; Steers & Mircea-Roussel 1976; Coates *et al.* 1978). With homeotropic boundary conditions and $\Delta\epsilon > 0$ this effect can be reversed by the application of a high-frequency signal (Coates *et al.* 1978). Figure 6 shows

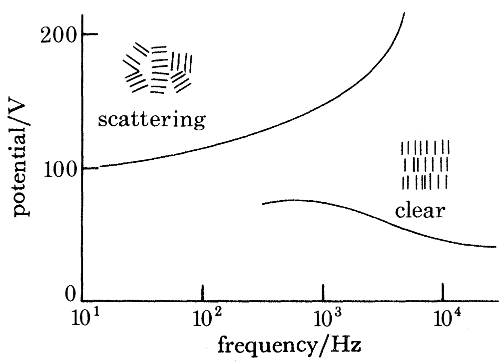

FIGURE 6. Threshold curves of a homeotropically aligned smectic A layer with $\Delta\epsilon > 0$.

typical threshold curves for both transitions. By applying a large voltage (*ca.* 100 V) and switching the frequency from 10^2 to 10^3 Hz a reversible transition from a clear to a scattering state is induced in times as short as 10 ms. Both states show long-term memory, making the effect useful in complex multiplexed displays (Crossland & Ayliffe 1982).

(ii) *Chiral smectic C ferroelectric effects*

The only known liquid crystal phase that can possess ferroelectric properties is the chiral smectic C phase (Meyer 1977), in which every smectic layer possesses an electric dipole density perpendicular to the director $\boldsymbol{n}$ and parallel to the smectic layer plane (figure 7*a*). Normally the director and polarization spiral along the helix (figure 7*b*), resulting in a cancellation of the polarization and a reduction of the bulk polarization to zero. However, this can be overcome by unwinding the helix. In the device described by Clark & Lagerwall (1980) the spiralling is suppressed by using a thin cell and a surface alignment that allows the director to lie down within the plane of the surface without imposing any specific direction. There are now two possible orientations of the director corresponding to the intersection of the cone of angle θ

(where θ is the tilt angle of the smectic C phase) with the surface. They are at an angle of 2θ to each other and have dipole moments pointing up and down respectively (figure 8*a*, *b*). The device is usually sheared just after cooling from the isotropic phase to produce uniform alignment. The director can then be switched back and forth through an angle of 2θ by altering the polarity of the applied electric field. By using crossed polarizers with the incident polarization direction parallel to the director in one of the polarization states there is extinction of light by the second polarizer for that state (figure 8*c*). The other polarization state transmits an intensity

$$I = I_0\{\sin 4\theta \sin(\pi \Delta n d/\lambda)\}^2, \tag{10}$$

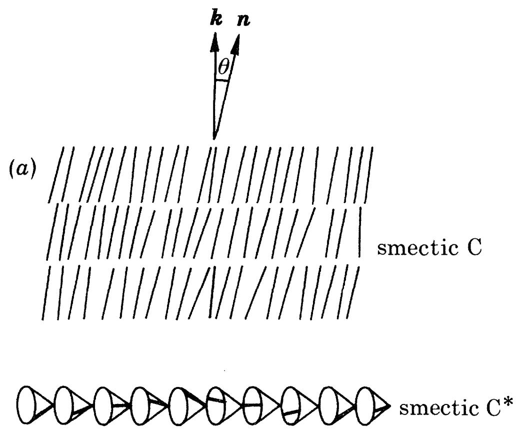

FIGURE 7. Smectic C configurations: (*a*) director tilted from layer normal $\boldsymbol{k}$ by θ; (*b*) spiralling of director in chiral smectic C.

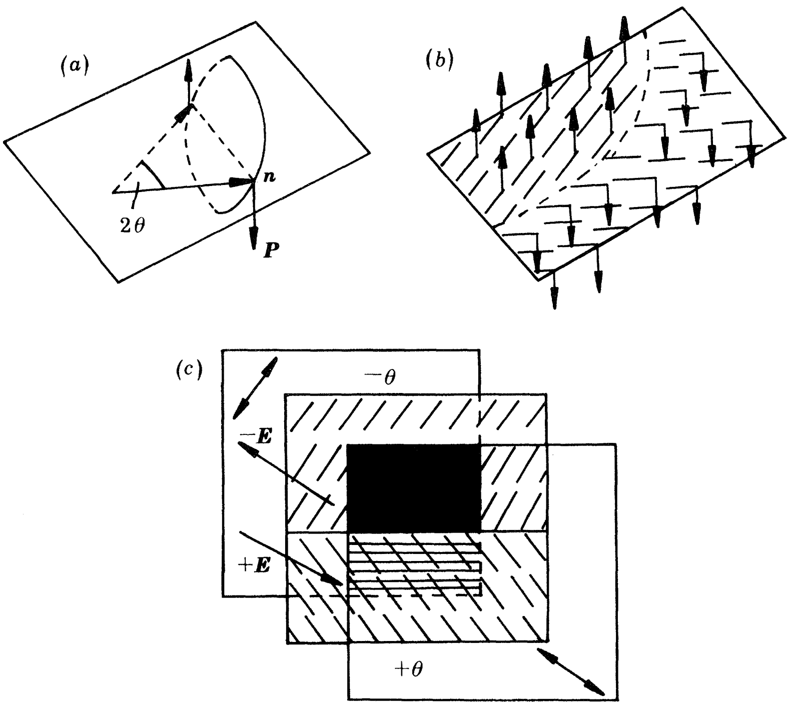

FIGURE 8. Operation of ferroelectric chiral smectic C* electro-optic devices: (*a*) two possible orientations on a surface imposing no specific direction; (*b*) dipole moments of the two possible orientations; (*c*) device operation with two crossed polarizers.

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where I_0 is the parallel polarizer transmission. The resulting electro-optic effect has several very attractive features (Clark & Lagerwall 1980):

high speed of switching even at low voltages – for example 5 μs at 10 V;
bistability – both states are stable with long-term memory;
threshold behaviour – the switching shows a pronounced threshold.

The ferroelectric chiral smectic C effect has many potential applications, from fast shutters to complex multiplexed displays. However, the technological problems that need to be solved before this effect can be used are quite severe. The liquid crystal layers must be much thinner than those currently fabricated (2 μm rather than the usual 10 μm), and the surface alignment required is not readily accessible. Finally, no suitable chiral smectic C material exists with a room-temperature phase, a large transverse dipole moment and a tilt angle close to the preferred value ($\theta = 22.5°$).

6. Thermo-optic effects

I shall now consider the thermo-optic effects in liquid crystals that occur when the temperature of the liquid crystal is varied in the vicinity of a phase transition.

(a) *Cholesteric materials*

Another optical property of helically ordered liquid crystals is used in the thermo-optic (or thermochromic) effect in cholesteric materials (Elser & Ennulat 1976). In a well aligned planar sample, Bragg-like reflexions of one sense of circularly polarized light occur from the helical planes at a wavelength λ related at normal incidence to the average refractive index $\bar{n}$ and pitch P by

$$\lambda = \bar{n}P. \tag{11}$$

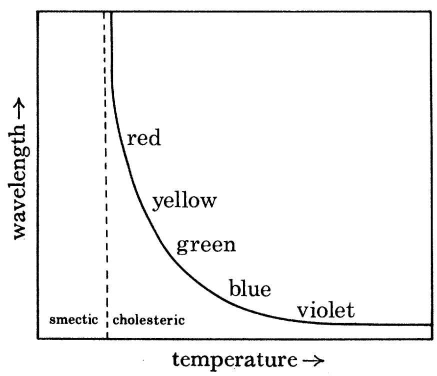

Figure 9. Temperature-dependence of selective reflexion from a cholesteric close to a smectic–cholesteric transition.

A strong thermochromic effect occurs just above the first-order phase transition from cholesteric to smectic phase, where P (and hence λ) diverges rapidly (figure 9). Thermochromic effects can be very sensitive, showing in some cases a colour shift through the visible spectrum for a change in temperature of only 0.5 K. Unlike all the other effects described in this paper, thermochromic devices are fabricated by printing an 'ink' containing the liquid crystal onto a substrate. The ink is produced by using one of two techniques. Droplets of the liquid crystal can be microencapsulated in polymer shells (diameters 5–50 μm) and the ink formed from a slurry of these capsules in water. Secondly, a dispersion of the liquid crystal droplets in a binder polymer film can be used directly without the need to form capsules. There is a wide range of temperature sensing applications of thermochromic devices, for example thermometry, medical thermography, non-destructive testing, radiation sensing, fashion and advertising.

(*b*) *Smectic A materials*

Smectic A liquid crystals can adopt two metastable textures in a thin layer. For simplicity, if we consider a layer with homeotropic boundary conditions, there is a clear transparent texture with the director normal to the plates, and secondly there is a scattering texture shown schematically in figure 10. If the material has $\Delta\epsilon > 0$, it can be changed from one texture to the other by using the overlying nematic and isotropic phases with the application of heat and an electric field, as shown in figure 10. Heating to the isotropic phase followed by rapid cooling through the

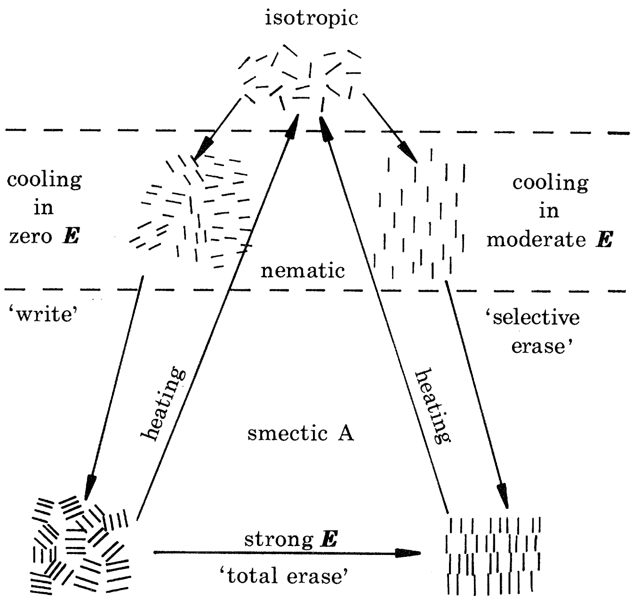

FIGURE 10. Transitions between the clear and scattering textures of a smectic A layer with the use of an overlying isotropic phase.

nematic phase produces the scattering texture. Applying a moderate voltage as well as the heat produces alignment in the nematic phase and results in the aligned smectic texture. The application of a higher voltage erases the whole display by transferring the whole layer to the transparent texture. There are two quite distinct methods of applying the heat pulse.

(i) *Laser addressing*

A high-resolution image can be produced by scanning a focused laser beam across a smectic layer by using galvanometer mirrors and converting the laser energy into heat within the layer (Kahn 1973*a*) with the use of either absorptive surface layers (Kahn 1973*a*; Dewey 1980) or a dye dissolved in the liquid crystal (Hughes 1983). The system is shown schematically in figure 11, together with the projection optics for displaying the written image. The normal contrast is black written lines on a bright background, but this is reversed by the insertion of a Schlieren stop. Selective erasure is achieved by simultaneously scanning the laser and applying a voltage across the layer, and total erasure by the application of a higher voltage alone. The performance of laser-addressed systems is impressive with a possible resolution of up to several thousand lines in each axis. Writing speed depends upon the power of the laser used, and times as low as 3 s have been reported for writing over 10^6 picture elements with a high-power argon laser. There is almost indefinite storage, and capabilities exist for selective erasure, grey scale and multicolour display.

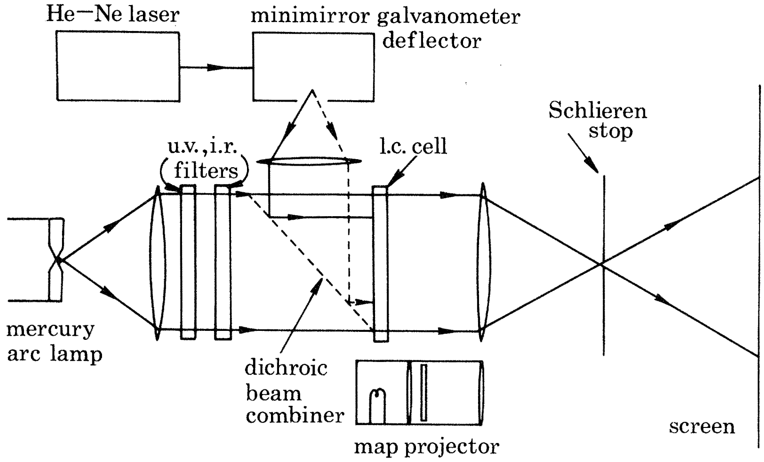

FIGURE 11. Construction of a transmissive laser-addressed smectic projection display.

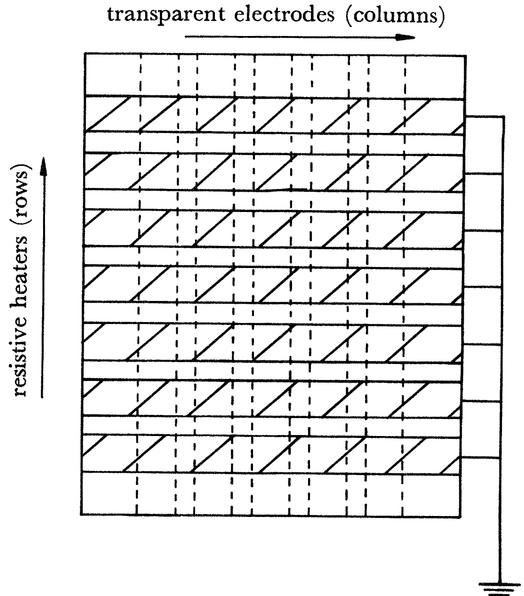

FIGURE 12. Electrode configuration of an electrically addressed smectic display.

(ii) *Electrical addressing*

The laser can be replaced as a source of heat by using an array of electrical heating elements (Hareng & Le Berre 1978). The heating elements (figure 12) are scanned sequentially and appropriate voltages applied to write or to erase the required information. The scattering can be observed directly (Hareng & Le Berre 1978) or a pleochroic dye can be dissolved to give an absorption contrast (Lu *et al.* 1982). Large displays, showing up to 100 alphanumeric characters by direct viewing, have been demonstrated (Lu *et al.* 1982; Le Berre *et al.* 1982) operating from

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0 to 40 °C. The whole display is written in 2–5 s with a power consumption of 25 W, although the permanent memory significantly reduces the power consumption if only part of the information is changed.

7. Conclusions

There is a wide variety of electro-optic and thermo-optic effects in the nematic, cholesteric and smectic phases of thermotropic liquid crystals. Despite this wide variety there are a number of common features that form the building blocks of the various effects. In this paper many of these effects have been described in outline only; later papers, however, will examine displays based on the twisted nematic and pleochroic dye effects in significantly more depth.

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