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Liquid Crystal Display Research: The First Fifteen Years

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This paper is a review of the more significant developments in the liquid crystal display field during the last fifteen years. Major effects and their applications are discussed, and the state-of-the-art is presented.

INTRODUCTION

The era of liquid crystal displays dawned in the mid-sixties with a series of publications emanating primarily from U.S. industrial research laboratories with RCA, Westinghouse, and Xerox particularly in evidence. While the early papers described a variety of new phenomena, materials and devices, the liquid crystal display field only began to expand in the seventies after the discovery of stable compounds and widespread adoption of the twisted nematic effect.[1]

This paper is an attempt to provide the reader with a capsule review of liquid crystal display R & D since its beginnings. Obviously, a thorough evenhanded review of the enormous amount of literature accumulated would have covered many pages and therefore gone far beyond what the editors had in mind when they solicited this paper. Two alternatives, each with its own shortcomings, presented themselves: to limit the material presented to only practically important display technologies, that is the ones which made or are about to make it to the market, or to cover as much ground as possible, even though necessarily in a superficial way. The latter approach, which includes the technologies which succeeded as well as the ones which led to a dead end, appealed more to this author who would like here to apologize for unavoidable omissions.

In tracing back the development of liquid crystal displays, the pioneering work of Fergason and co-workers[2] deserves special credit. While his investigations were primarily directed toward applications of cholesterics in thermography, many display concepts[3,4] were also proposed and demonstrated, which will be discussed later. The next milestones in the history of display development are the so-called William's domains and its follower, dynamic scattering.

WILLIAMS DOMAINS AND DYNAMIC SCATTERING

An early patent assigned to RCA by Williams[5] filed in 1962, and issued in 1967, described in effect all the elements, including scattering under the influence of electric fields, of the later widely publicized dynamic scattering devices. In the Williams patent, the electroptical device shown in Figure 1, consists of a nematic liquid crystal layer; 9, sandwiched between transparent tin oxide electrodes, 25 and 26, on a transparent substrate. Upon application of an electric field the device became scattering.

In later papers, Williams[6,7] discussed the domain formation and light scattering phenomena in more detail, but the use of nematics in displays remained dormant until 1968 when Heilmeier and co-workers began to publish a series of papers describing dynamic scattering.[8,9,10] In this electro-optic effect an initially clear and transparent nematic film layer of negative dielectric anisotropy and resistivity ranging between 10^8–10^9 Ω-cm becomes turbulent and scattering (milky white) upon passage of a dc or low frequency ac current which generates hydrodynamic instabilities resulting in the formation of optically inhomogeneous light scattering regions. After

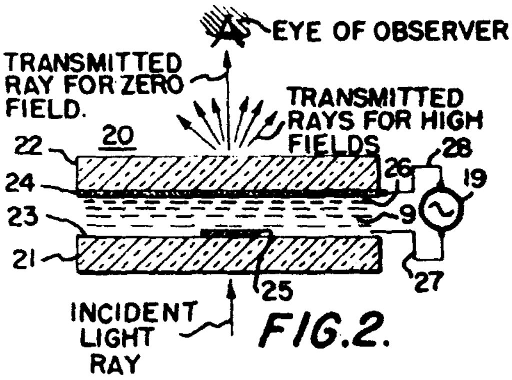


FIGURE 1 Schematic of Liquid Crystal Display Cell (Ref. 5).

field removal, the liquid crystals returns to its original state. Theoretical treatments of the phenomenon were developed by Felici,[11] Carr,[12] Helfrich[13] *et al.* Dynamic scattering occurs typically at fields of the order of 10^3–10^4 V/cm and cells normally require 15V. The milky appearance of dynamic scattering is quite pleasing, especially in cells having a back reflecting surface (Figure 2).

Technical applications of dynamic scattering at the time of its discovery were not practical because the materials used were either single compounds which had to be heated above room temperature to reach the mesomorphic range or mixtures which were not always stable. Dynamic scattering, however, became viable in 1969 with the synthesis of methoxybenzylidene-*p*′-*n*-butylaniline (MBBA) by Kelker and Scheurle,[14] and from then on materials ceased to be a problem because an ever increasing number of stable mixtures with wider temperature ranges began to become available.

Dynamic scattering has a place in any situation demanding large area, low cost displays with good angular viewing characteristics (no polarizers are necessary), and where multiplexing and power consumption are not issues. The need for such displays exists in advertising, where indeed dynamic scattering had some success. The applications proposed for dynamic scattering encompassed many of today's uses of liquid crystals ranging from timepieces and message displays to television screens. Yet,

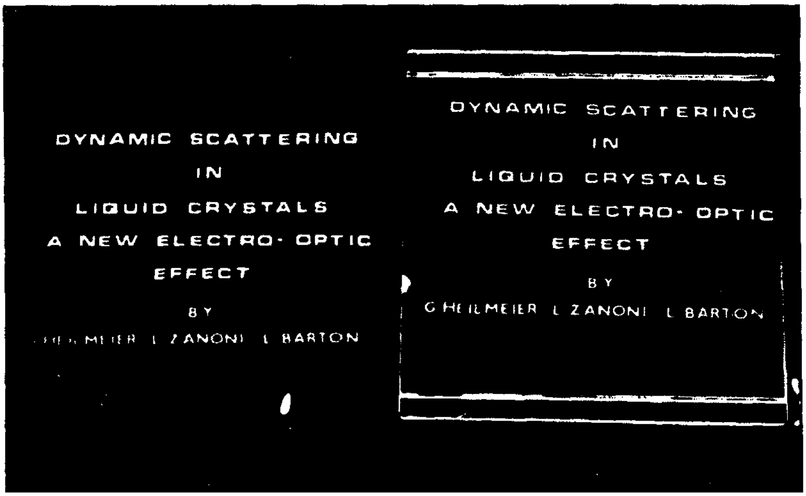


FIGURE 2 Left Lettered Card. Right Dynamic Scattering Display. Ref. 9., ©1968 IEEE. Reprinted, with permission, from Proceedings of the IEEE, Vol. 56, No. 7, p. 1171, July 1968.

due to the high voltage and power demands, and also because of the very limited life-times due to high current flow, dynamic scattering never became really successful and its predicted applications only became realized by the emerging of liquid crystal field effects. Dynamic scattering can be used in higher information content displays when the liquid crystal is in series with active electronic elements such as MOS devices,[15,16,17] or thin film transistors (TFT's).[18] Special addressing techniques can also be used to improve the multiplexing capacity.[19]

CHOLESTERIC-NEMATIC PHASE TRANSITION

In the same year dynamic scattering was first reported (1968), Wysocki *et al.*[20] reported another new important electro-optic effect, the cholesteric-nematic phase transition: In this effect the helical molecular arrangement of a cholesteric, having positive dielectric anisotropy, is broken-up by application of large external fields, and the cholesteric becomes a nematic with its molecules parallel to the field. In a conventional cell geometry this corresponds to a homeotropic uniaxial texture. If the field is removed, the nematic reverts again to a cholesteric and partial field removal delays the process. The magnetic analogue of the electric field transition was observed at Bell during a NMR experiment.[21] A theory for the magnetic effect was developed by de Gennes[22] and a similar treatment for electric fields was given by Meyer.[23] Starting out with the cholesteric planar texture, the sequence of events after application of a field is as follows: if the liquid crystal is planar (Grandjean), it first assumes the focal-conic texture, which is scattering, with an over-all aspect similar to dynamic scattering but without the violent turbulence associated with that effect. A further increase in field-strength leads to the clear homeotropic nematic texture which is dark between crossed polarizers. The optic sign of this texture is positive whereas the Grandjean texture is optically negative. Upon field removal a complex series of events occur which ultimately results in return to the focal-conic texture.[24] The effect can be very fast, milliseconds or less on-time, and has a very sharp threshold:

$$F_c = \frac{\pi^2}{2Z_0}\left(\frac{K_{22}}{\Delta\varepsilon}\right)^{1/2}$$

where F_c is the threshold field, Z_0 the pitch of the cholesteric helix, K_{22} an elastic constant, and $\Delta\varepsilon > 0$ the dielectric anisotropy. The threshold makes this effect ideally suited for multiplexing and, therefore, high information content displays. The moderate contrast between the clear homeotropic and scattering focal-conic textures is sufficient for acceptable viewing. The contrast can be enhanced further by the use of polarizers, which results in extinction in the homeotropic texture and light transmission in the focal-conic texture because the latter is birefringent like a polycrystalline substance.

Prototypes of displays based on the cholesteric-nematic phase transition were described by several authors, and the number of pixels in matrix addressed displays was gradually increased from thirty-five[25] to thirty-five hundred[26] and more recently to ten-thousand.[27]

Just as in 1969 MBBA made dynamic scattering viable, the advent of the cyanobiphenyls in 1973[28] made the phase transition a practical effect by lowering the required fields from 5×10^5 to 5×10^3 V/cm or less. Yet, the relatively low optical contrast of the phase transition prevented its wider use until in 1974[29] when White and Taylor circumvented this problem by the addition of pleochroic dyes to the mixtures. More about that effect later. Today's phase transition materials are generally complex mixtures of nematics doped with an optically active material to provide the cholesteric twist. They have very wide temperature ranges, (−20°C + 80°C) or better, are stable, colorless, and have dielectric anisotropies of $\Delta\varepsilon + 15.0$ and higher. Because the large market for twisted nematic materials demands it, improved mixtures reach the market in a steady flow benefiting at the same time, anybody interested in phase change materials which have practically the same characteristics.

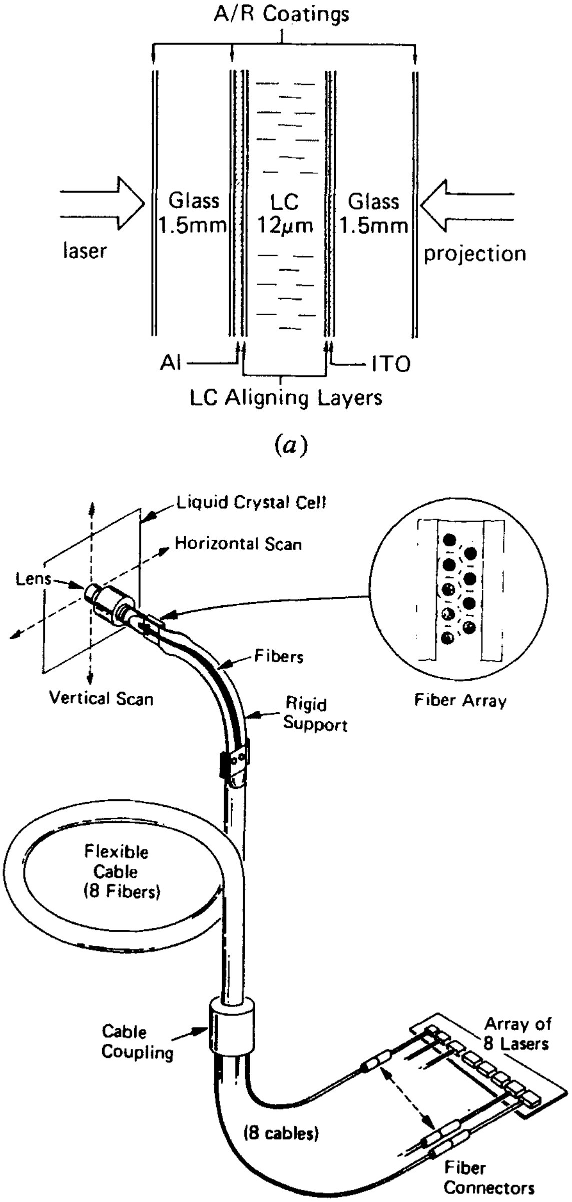
OPTICAL STORAGE IN LIQUID CRYSTALS

Reversible optical storage effects have been observed in all groups of liquid crystals, cholesterics, smectics, and, very recently, in nematics. Heilmeier *et al.*[30] reported that cholesteric-nematic mixtures become milky scattering upon application of electric fields. The effect was not unlike dynamic scattering; the difference is, however, that the mixtures remained scattering for long periods of time after field removal. Erasure, i.e. return to the initial clear state, could be accomplished, however, by application of ac fields above audio frequency. While the authors attributed the effect to an emulsification which could be reversed by an ac field, it was later shown that what occurred was, in fact, a reversible texture change from the scattering focal-conic to the clear Grandjean (planar) texture.[31,32,33] The turbulence associated with the ionic motion in dynamic scattering, plays a definite role in the formation of the scattering state in the storage mode. This effect is observable in many cholesteric-nematic mixtures, and erasure can always be accomplished by ac fields, provided that the overall dielectric anisotropy is negative; the frequency of the applied field sufficiently high to prevent ionic motion and the field-strength large enough. It is not necessary either that mixtures contain a cholesteric liquid crystal. Addition to nematics of soluble optically active materials which are not liquid crystalline, can result also in mixtures which exhibit reversible storage.[33] Thermally induced optical storage in cholesteric-nematic mixtures was described by Soref.[34] Initially clear mixtures were heated above the isotropic temperature and on cooling adopted in the focal-conic texture which could be erased by ac fields or shear. Thermo-optic storage was later used by Melchior *et al.*[35] to demonstrate the feasibility of scanned laser addressed storage display panels.

In another reversible optical storage mode, cholesteric-nematic mixtures with very large pitch (15–100 μ) are initially aligned homeotropically and upon application of an electric field acquire a stable bubble domain texture[36,37] which can also be erased with ac fields. The most recent storage effect reported in cholesterics is based on the bistability between two clear states with different polarizing characteristics.[38] One state is "untwisted nematic-like" with its optic axis oblique to the substrate and the second state, the addressed state, is helical with a twist of 360°. Electrical switching between the two states is possible.

Despite its promise, cholesteric storage effects have remained in the laboratories. Experimental display[39] and imaging devices[40,41] however, have been described by several authors, and recently Tani *et al.*,[42] discussed a matrix addressed storage display having 306 × 574 pixels and capable of displaying 320 Chinese characters on a 16 × 18 dot matrix and ≃64 lpi resolution.

The first smectic storage displays began to appear in 1973[43] and had the advantages over cholesteric displays of better image quality (smaller scattering centers) and local erasure capability. The basic device functions as follows: a smectic layer is sandwiched between two transparent electrodes (tin oxide on glass) and made to adopt the homeotropic texture by an aligning layer. If the smectic layer is heated above the isotropic transition temperature and then allowed to cool quickly, it will adopt a disordered scattering texture; if the cooling, however, proceeds slowly or an electric field is applied during cooling, the smectic layer returns to the clear nonscattering state. Kahn[43] was able to construct displays with a resolution of 50 l/mm over a 3 × 3 cm area. Addressing was carried out with a scanned 20 mW, 1.06 μ laser at a rate of 10^4 elements/sec., and the display was read-out with projection optics. Since then much progress was made[44] and Dewey *et al.* described in '82 a smectic storage display with 4 million pixels[45] addressable by an array of eight GaAs lasers in 75 s (Figures 3a and 3b).



(*b*)

FIGURE 3(a) Laser Addressed Smectic Display Cell., Ref. 44., (b) Fiberoptic Delivery System, Ref. 45. By permission of A. Dewey and the SID.

An interesting alternative to laser addressed smectic storage displays was proposed by Hareng and co-workers[46,47] who used the rows of a matrix as resistor elements to heat the smectic (row at the time) and supplied the electronic information to the flat panels via the columns. In 1982, flat smectic storage panels with a capacity of 1000 characters, 25 lines of 40 characters, on 5 × 7 matrix were reported. Characters were black on white.

The use of pleochroic dyes in conjunction with smectics was described by Tani,[48] and some years later, Lu *et al.*[49] constructed a smectic storage display based on the addressing method proposed by Hareng but containing pleochroic dyes. This device with enhanced contrast could display up to 1224 characters on a 7 × 12 dot matrix. As a whole, smectic storage displays offer the advantages of high multiplexability no need for polarizers, and large viewing angles, but on the other hand, they demand fairly high driving powers.

Pure nematics were not known to exhibit any intrinsic storage effects, but electrically or magnetically switchable bistable storage mode cells were reported in the literature;[50,51] the bistability and storage in these cells was achieved by treating the surfaces in such a way as to produce two oppositely tilted directions resulting in bistability. Polarizers are required to distinguish the two clear states.

THE TWISTED NEMATIC EFFECT

This effect is by far still the most widely used of all liquid crystal electro-optic effects and was reported by Schadt and Helfrich[1] in 1971. A nematic crystal with positive dielectric anisotropy is sandwiched between two transparent electrodes rubbed to obtain molecular alignment parallel to the direction of rubbing and the substrate (homogeneous alignment). The two electrode plates are separated by a spacer of about 1/2 mil thickness, and, prior to filling, are assembled with the directions of alignment at a 90° angle. The result of the procedure is a helical liquid crystal film with 90° twist across the cell (Figure 4).

The authors considered the helical texture of twisted nematic cells as corresponding to the planar cholesteric texture which, indeed, is true. Upon application of an electric field, the helical arrangement disappears and the texture becomes homeotropic with the optic axis parallel to the field and normal to the substrate which is the same situation as in the cholesteric-nematic phase transition.[20] After field removal, the liquid crystal returns to its initial planar state with a time constant which depends on cell thickness, viscosity, and the elastic constant.

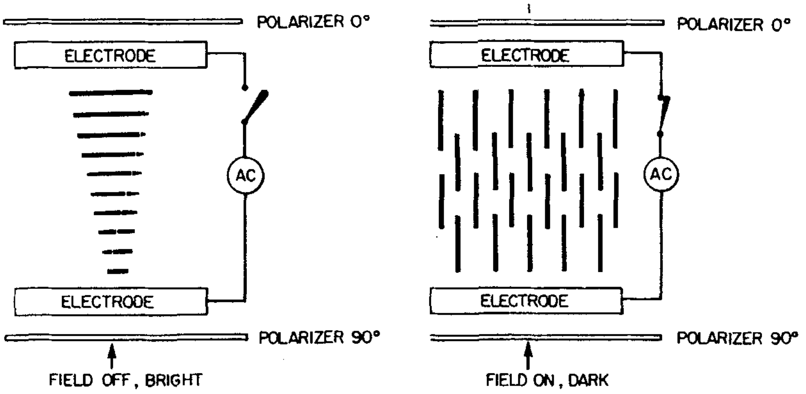


FIGURE 4 Schematic of Twisted Nematic Cell.

Twisted nematic cells are viewed between crossed or parallel polarizers depending on whether transmission is preferred in the field in "off" or in "on" condition, and the principal directions of the polarizer are generally parallel to the direction of alignment. In most applications, twisted nematics are viewed in reflection and, therefore, a diffuse reflector is placed behind one of the polarizing sheets. Prior to the application of an electric field, the cells transmit light when the polarizers are crossed because the plane of polarization of light is rotated by 90° in the cells. The propagation of light through helical media was treated by Mauguin,[52] deVries,[53] Chandrasekhar,[54] Baur,[55] Ward,[56] *et al.* The "Ward" reference, *"Optical Rotation of Helically Twisted Structures"*, unfortunately was never published, but copies can be obtained.

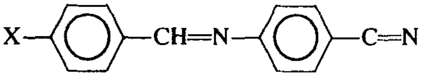
The advent of the twisted nematic cells spurred intense research in related fields, primarily liquid crystal materials, alignment methods, optical properties, fabrication, etc. The fruits of these efforts have now become part of our daily life.

a) Materials

In the materials realm, mixtures based on cyanobiphenyls[28] were rapidly augmented with the synthesis of benzoate esters,[57,58] Schiff bases with cyano groups,[59] phenylpyrimidenes,[60] phenylcyclohexanes[61] (PCH), cyanocyclohexylcyclohexanes[62] (CCH), just to mention some, which can be combined in innumerable ways to satisfy specific requirements of the display engineer. Excellent in-depth reviews of this very broad topic can be found in Refs. 63 and 64, and very complete tables of materials known up to 1972 were published by Demus *et al.*[65] To give the reader an idea of the problems associated with the formulation of suitable mixtures, let us rapidly review the most common requirements. First, it is necessary for the material to have positive dielectric anisotropy and be nematic at room temperature. Additional requirements are chemical, electrochemical and photochemical stability, high resistivity, preferentially low viscosity for enhanced response times, low birefringence to prevent the occurrence of undesirable optical effects, and absence of unwanted color. Doping with an optically active material is desirable to prevent problems arising from the so-called reverse twist. In matrix displays, where the columns of electrodes are addressed in parallel and the rows strobed one at a time, sharp threshold is yet another requirement. Not surprisingly, all these demands cannot be satisfied by a single liquid crystal component alone, but require the formulation of carefully balanced mixtures. In order to be useful in electro-optic applications, the material must be in the liquid crystalline, i.e. mesomorphic state, which only exists in a well defined temperature range. Above the upper limit of this temperature range, the liquid crystal becomes an isotropic liquid, and below is a crystalline solid. Within the mesomorphic range, the liquid crystal may undergo transitions also from one liquid crystal state to another, but this topic is beyond the scope of this paper.

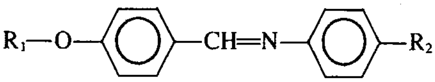
It is obvious that many display applications require fairly broad temperature ranges (the automotive industry offers a good example), and the mesomorphic range of single component liquid crystals is usually quite narrow. For example, MBBA[14] has a mesomorphic range of +22 to +47°C. Similarly, 4-cyano-4'-*n*-pentylbiphenyl,[28] an often used component in display mixtures, has only a liquid crystalline range of +22.5 to +35°C; however, mixed with 4-cyano-4'heptylbiphenyl in the ratio of 56 to 44 mol %, the liquid crystal range becomes +0.5 to +37°C. Numerous other examples exist.

As stated before, field induced alignment of liquid crystals with long axes parallel to (E) require a positive sign for the dielectric anisotropy which is the characteristic of numerous but not all liquid crystals. Strong positive $\Delta\varepsilon$ is generally associated with a CN end group. For example:



($X = C_nH_{2n+1}O$; C_nH_{2n+1}; $C_nH_{2n+1}CO$)

A large $\Delta\varepsilon$ is desirable whenever short turn-on times and low threshold voltages V_{th} are required. On the other hand, the use of mixtures containing only materials with positive $\Delta\varepsilon$ is undesirable in applications involving multiplexing.[66] Chemical, electrochemical and photochemical stability are, of course, a must to guarantee long life of the display devices. The first generation of liquid crystal displays, dynamic scattering devices, filled with Schiff bases:

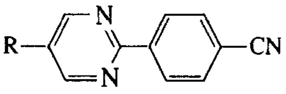


R_1 and R_2 are alkyl groups

were very sensitive to contaminations, especially water, and their use in displays required careful purification and hermetic sealing. The synthesis of cyanobiphenyls made liquid crystal displays much more viable products because these compounds are chemically, electrochemically, and photochemically stable. It should be noted that in order to minimize electrochemical interactions, liquid crystal mixtures for display applications have resistivities ranging from 10^{10}–10^{11} Ω-cm and are addressed using ac voltages. Resistivities of the order of 10^{10}–$10^{11}\Omega$-cm require high purity and judicious handling.

The viscosity of liquid crystal mixtures is one of the factors controlling the "turn-on" and "turn-off" times in liquid crystal devices.[67] Naturally, low viscosity corresponds to fast response, a desirable feature, which unfortunately runs against the demand for broad temperature ranges. Compounds of the CCH and PCH groups are photochemically very stable, which makes them valuable components in mixtures of broad mesomorphic ranges. An additional advantage of PCH and CCH compounds is the low birefringence, i.e. the small optical anisotropy. In many liquid crystals, the birefringence is about 0.2, but in PCH and CCH, it is only about 0.1. In practice, this means that the contrast ratio as a function of viewing angle is better for the same cell thickness.

Another group of liquid crystals used in compounding of display mixtures are pyrimidines such as for example:



R = alkyl group

whose $\Delta\varepsilon$ is large, +18.0. Almost all formulations of twisted nematic liquid crystal display mixtures include a chiral material, typically in concentrations of 0.1–3.0%. The function of this optically active additive is to impart a uniform sense of rotation, left or right-handed, to the 90° helix.

Without chiral additives regions of reverse twist may occur in the display giving it a patched appearance.[68] Optically active additives used frequently come from the cholesteric ester and biphenyl families, but the pool of candidates is very large indeed. It should be noted that the addition of any optically active substance to a nematic results in changing the nematic into a cholesteric material.

b) Alignment

The goal of all alignment techniques is to produce "single domain liquid crystal films", i.e., films resembling single crystals. Dynamic scattering, the cholesteric-nematic phase transition and several storage effects do not require special alignment even though the optical characteristics of the cells are generally improved by using aligned films. In nematic cells, however, alignment is a must.

Broadly speaking, two types of liquid crystal film alignment are known: homogeneous (molecular axes parallel to the substrate) and homeotropic (molecular axes normal to the substrate). In the case of homogeneous alignment, the long axes of the molecules can be oriented in any azimuth in the plane of the substrate and the designation homeotropic can be used also for films in which the molecular axes are somewhat oblique with regard to the substrate. Many techniques are known to produce uniform molecular alignment in liquid crystals. Homeotropic orientation has been achieved by judicious cleaning of the substrates,[69,70] addition of surfactants to the liquid crystal,[71] and deposition of surface coupling agents on the substrate.[72] Homogeneous alignment has been obtained by rubbing the substrate,[73] deposition of surface coupling layers,[71] and other techniques. Detailed discussions of the many existent alignment techniques are presented in Refs. 74 and 75.

A unique method to produce controlled alignment was reported by Janning[76] in 1972. This author used oblique vacuum depositions of very thin films of silicon monoxide and other materials onto the substrate to obtain very well-aligned liquid crystal films. Janning's technique found widespread use because of its reproducability and stability, which are crucial factors in the fabrication of liquid crystal displays. The techniques were later refined to produce special tilt angles[77] and alignment mechanisms, in general, became of interest to many authors.[78-84]

c) Optical Properties

The optical behavior of twisted nematic cells is complex and depends on the liquid crystal mixture used, cell thickness, the initial molecular tilt, and last but not least, on the conditions of viewing and illumination. As mentioned earlier, at zero voltage the twisted nematic acts as a 90° optical rotator for plane polarized light and, therefore, between crossed polarizers the cell appears bright in transmission; obviously, between parallel polarizers the situation is reversed. When a sufficiently high voltage is applied, the helical arrangement disappears and the molecules become aligned parallel to the field. The liquid crystal assumes the homeotropic texture and acts optically like a uniaxial crystal and does not rotate light. The result is that the cell, which in transmission between polarizers appeared bright, becomes dark. At smaller voltages, there is still transmission of light, but the details of the mechanism are beyond the scope of this review. The contrast between addressed and non-addressed regions in a twisted nematic display has a strong angular dependence, which is the Achilles heel of twisted nematic displays, making them the target of competing technologies. While the angular dependence of the contrast is tolerable in watches and displays, it becomes difficult to manage in highly multiplexed devices (>16:1). This topic is discussed in detail in Refs. 85 and 86.

d) Fabrication

The fabrication of liquid crystal displays involves basically the following steps: deposition of the electrode material onto the substrate followed by photofabrication of the electrode patterns; deposition of the alignment layer; spacing; materials selection, filling and sealing. "Many ways lead to Rome" and different manufacturers have achieved essentially the same final results following different paths. The electrode material most frequently used is sputtered indium-tin oxide which is transparent and has adequate conductivity. The pattern definition is carried out following conventional photoresist/etching techniques. The substrate is usually glass, but it was recently shown that plastic substrates can also be used.[87] Alignment of liquid crystals was discussed earlier and here also different approaches have been successfully used. It is important, however, to note that organic alignment layers are not compatible with the high temperatures required for glass frit sealing. In order to maintain uniform thickness of the liquid crystal layer, often spacers are required, and again a variety of approaches have been followed. What is demanded from the spacer material is easy availability in 10–15 μ range (the most typical cell thickness range), chemical inertness and transparency when placed in locations other than the perimeter of the cell.

With regard to materials, many formulations and basic components are commercially available and, while some manufacturers synthesize their own chemicals, many others use ready mixtures or purchase the components and formulate their own proprietary compositions. The glass plates are held together by a seal which protects the liquid crystal from the environment, prevents leakage and also holds the plates together. Both glass frit and a variety of polymers have been successfully used as sealants; since modern compositions are much more inert than earlier materials used, such as Schiff bases, polymeric seals are becoming increasingly popular. Filling of the cells is usually done in a vacuum through a filling hole left open in the seal or on the cover glass plate. After filling, the hole is plugged with indium or a polymer. An alternative filling method is to place the liquid crystal directly between the glass plates and seal the cells afterwards. This process removes the need for filling holes. The last step in the cell fabrication cycle is the application of the polarizers. The large demand for liquid crystal displays using sheet polarizers was a strong incentive for the industry to improve the optical quality, polarizing efficiency, uniformity and resistance to heat and moisture of their products. Today, polarizing sheets are available in many colors and are usually laminates which may include diffuse reflectors, pressure sensitive adhesive and barriers against ultraviolet radiation.

ADDRESSING TECHNIQUES

The vast majority of today's liquid crystal displays contain many elements and, therefore, individual address, meaning that one driver per element is neither practical nor economical because of the large number of interconnections and drivers required. This difficulty can be circumvented by dynamic matrix addressing in which the display is subdivided into individual pixels in a chessboard-like arrangement. Generally, a matrix addressed display has one set of striped electrodes on one substrate and another set of striped electrodes (columns), at a 90° angle, on the other, the liquid crystal being sandwiched in between. During operation, the information is normally fed simultaneously to all columns (or rows) and the rows (or columns) are strobed. The underlying principle is that an element is "on" when two pulses coincide at a given location, otherwise the element is "off". The simplest form of matrix addressing is half-select which means that 1/2 of the voltage originates from the columns and the other half from the rows. It has been shown by Alt and Pleshko[88] that the maximum number of lines $N_{\max}$ which can be scanned in a display is given by

$$N_{\max} = \left[\frac{(1+P)^2+1}{(1+P)^2-1}\right]^2 \quad \text{where}$$

$$P = \frac{\Delta V}{V_{\text{threshold}}} = \frac{V_{\text{on}} - V_{\text{off}}}{V_{\text{off}}} = \frac{V_{90} - V_{10}}{V_{10}}$$

(rms voltages)

In the case of twisted nematic cells with parallel polarizers, V_{10} and V_{90} are the voltages at which 10% and 90% transmission, respectively, are measured; by convention, these are respectively the maximum acceptable "off" value and the minimum acceptable "on" value. What then is required for multiplexing of a large number of lines is a sharp threshold, i.e. a small $\Delta V/V_{\text{threshold}}$ ratio.

It has been shown that mixtures of positive and negative $\Delta\varepsilon$ can have sharper threshold characteristics,[89] than mixtures of only positive materials, and for this reason are preferred in applications involving multiplexing. The overall $\Delta\varepsilon$ of the mixture of course must remain positive for use in the twisted nematic mode. Another point which cannot be neglected is the temperature dependence of the threshold voltage, ideally, $dV_{\text{th}}/dT\ V_{\text{th}}$ should be as small as possible, which is true for azoxy compounds but not for biphenyls.[90] Again, design compromises will have to be made for each application.

Multiplexability reduces the number of drivers in a square matrix from N to $2(N)^{1/2}$, but the price to pay is a reduction in contrast and more stringent demands on the liquid crystal characteristics such as threshold voltage response curves and response times. Matrix addressing can also be used with segmented alpha-numeric displays, but appropriate connections must be used.[91]

In the last years, addressing techniques were substantially improved through the use of better liquid crystal mixtures,[92] special drive waveforms,[93] new matrix geometries and also improvements in the threshold characteristics by better anchoring of the liquid crystal molecules to the substrate. An example of a novel matrix geometry was demonstrated by Kawakami *et al.*,[94] who used horizontal electrodes controlling two rows of elements (instead of the conventional single row) and connected each element of the double row to a separate column (double matrix). This concept was later extended to four rows (quad-matrix) and demonstrated in a portable TV with 19200 pixels[95] (Figure 5).

An addressing method which deserves special attention is "two-frequency addressing"[96,97] which can provide or sharpen the threshold characteristics of liquid crystal cells, thus enhancing the multiplexability. This result is achieved by simultaneous application of electrical signals of different frequency and requires different orientational response to different frequencies. With dynamic scattering, "two frequency addressing" achieved only a limited success, but after Bucher *et al.*[98] synthesized materials in which the sign of the dielectric anisotropy changed with frequency, the concept became much more attractive. These materials, phenylbenzoyloxybenzoates mixtures, have typically values of $\Delta\varepsilon = 6.2$ at 2.5 KHz and $\Delta\varepsilon = -2.2$ at higher frequencies, i.e. the crossover is at 2.5 KHz. As previously stated, this characteristic not only increases multi-

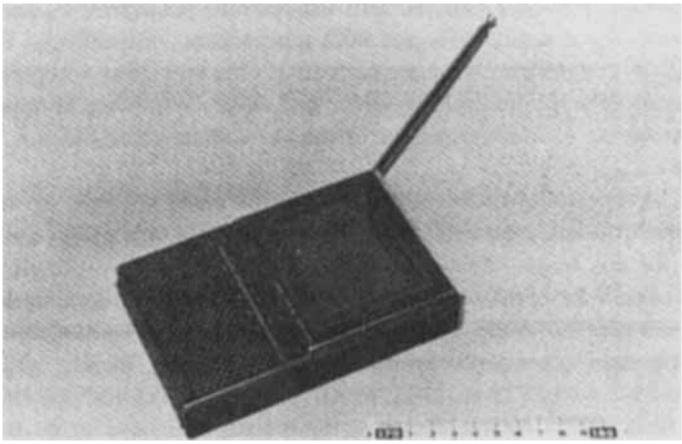


FIGURE 5 Portable Liquid Crystal TV, Screen: 3″ diagonal, Ref. 95. By permission of E. Kaneko and the SID.

plexability, but permits the construction of fast response devices because the molecules can be driven in both directions.

Dargent and Robert[99] demonstrated in 1977 a 2500 element twisted nematic display containing a Merck ZLI-518 mixture. Van Doorn and deKlerk[100] constructed a 100-line reflective display, and more recently investigators at Suwa Seikosha[101] described an 18 000 element device capable of displaying 8 rows of 64 characters each on 5 × 7 dot matrix. Frequencies used were 150Hz (low) and 50KHz (high). The parameters involved in the multiplexability of two frequencies addressed cells were studied by Clark and Harrison[102] for twisted nematic cells operated under varying conditions and with different liquid crystals.

Most recently new, two-frequency addressing materials, capable of multiplexing ratios up to 250:1 were reported.[103] Evidently, such materials could lead to fabrication of very high information content displays in the near future. Despite the continuing progress in direct matrix addressing, it is still widely believed that the need for displays with very high information content and good image quality will be best met by the incorporation of active electronic elements in the displays. This approach removes from the liquid crystal electro-optic effect the need for sharp threshold and the very special response characteristics required for real-time displays.

Active devices used for this purpose are diodes,[104,105] MOS's,[15,16,106-108] varistors,[109,110] and thin film transistors (TFT's)[111-114] with the latter receiving percently the bulk of attention. MOS driven liquid crystal displays appeared early in the history of liquid crystal displays, and have been developed over the years to the point where displays with ten thousands of pixels become possible. Workers at Hughes demonstrated in 1978[16] a video display of 30 000 pixels at 100 l/in., and Toshiba recently demonstrated portable TV sets with nearly 53 000 elements. The screen measured two inches in diagonal. Because of the opaque silicon substrate, displays of this type have to be readout in reflection and generally dynamic scattering has been used as the electro-optic affect. However, there is no basic obstacle to use several other liquid crystal effects with MOS arrays, and Crossland,[108] for example, recently discussed a guest-host display on a silicon wafer. Fundamental limitations of MOS addressed liquid crystal displays are the need for reflective read-out and the limited size of single crystal silicon substrates.

Varistors are a newcomer in the field of active electronic matrices and were first used by Castleberry[109] in 1979. An opaque substrate of pressed and fired ZnO with additives is the basic material which provides the strong non-linearity (sharp threshold) which permits extended multiplexing. As is the case with MOS displays, read-out must be in reflection. An advantage over MOS driven displays is that larger size displays can be made, but on the other hand, the resolution is somewhat limited. Best results to date are 36 lpi, a display size of 5″ × 7″ and a 180 × 252 pixel matrix.[110]

As mentioned before, a large fraction of the work presently ongoing worldwide in active matrices is dedicated to TFT's which can provide switching, gain and memory functions, thus giving the designer considerable freedom in the selection of electro-optic materials. Naturally, the existence of integrated electronic matrices not only benefits liquid crystal, but electroluminescent, electrochromic and other display technologies as well. A typical arrangement of a TFT in a liquid crystal display element is shown in Figure 6.[111]

The capacitor C_S is not required if the natural decay time is sufficiently long. Diligent efforts over the years have resulted in a continued increase in the number of pixels and line densities of TFT networks. Beginning with modest electroluminescent devices,[112] the technology had advanced in 1973 to 14 000 liquid crystal elements at 20 lpi[113] and to 62 500 elements at 50 lpi[114] in 1982 (Figure 7).

Until 1980, CdSe was generally the preferred semiconductor for TFT's, but recent progress in amorphous and crystalline silicon films has led several laboratories to investigate these alternatives. Progress in TFT liquid crystal displays based on polycrystalline and amorphous silicon has been rapid, and already devices with 57 600 silicon elements have been reported.[115] The interested reader is referred to a review of the state-of-the-art on silicon-based active matrices presented recently by Lakatos.[116]

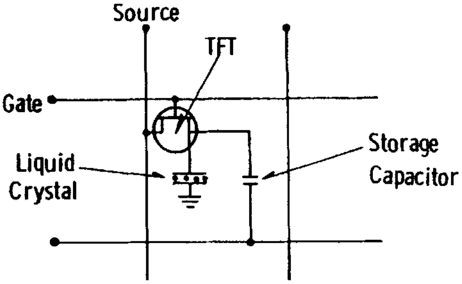


FIGURE 6 Matrix Circuit Element With Extra Capacitor, Ref. 111. By permission of Fang-Chen Luo and the SID.

GUEST-HOST EFFECT AND COLOR

Interest in liquid crystal color displays is as old as the display technology itself, and over the years four basic approaches have been pursued in the quest for color: a) Addition of pleochroic* dyes to nematics, cholesterics and smectics; b) Viewing of liquid crystal displays through transparent

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*In crystallography pleochroism means variation in the selective absorption of light with directions of vibrations. The term dichroism, a special case of pleochroism, applies when only two directions of absorption exist, but in the liquid crystal literature, pleochroism and dichroism have often been used interchangeably.

FIGURE 7 TFT-LC Panel with 2500 Elements, 1″ × 1″ size, Ref. 114. By permission of Fang-Chen Luo and the SID.

colored polarizers, filters, or systems of color stripes; c) Use of the natural colors exhibited by cholesterics with pitch in the visible; and d) Use of the large birefringence in nematics to generate interference colors.

a) Pleochroic dyes

The addition of pleochroic dyes to nematics to achieve color in displays was proposed by Heilmeier and Zanoni[117] who also coined the term "guest-host effect". In this effect nematic molecules (hosts) become oriented under the influence of an electric field and align pleochroic dye molecules (guests) dissolved in the host. Prior to the use of pleochroic dyes in liquid crystal displays, Dreyer,[118] in 1946, had already proposed the use of pleochroic dyes in nematics to make polarizing sheets.

In pleochroic dyes, the light absorption depends on the orientation of the molecular axis relative to incident polarized light, and for this reason optical differences are observable if the orientation of the dye molecules changes. Interest in the guest-host effect began to increase, after White and Taylor[29] used the cholesteric-nematic phase transition as the basic electro-optic effect. The phase transition provides superior alignment which, in turn, results in displays with high contrast.

Since then, guest-host displays in many configurations have been described in the literature. Pleochroic dyes were added to nematics of positive, as well as negative, dielectric anisotropy to combine the guest-host effect with the Freedericksz transition.[119] The twisted nematic effect was also optically enhanced by the addition of dichroic dyes.

Most recently, Uchida *et al.*[120] described a double layered guest-host cell which offers some improvements over former designs and does not require polarizers (Figure 8).

From a practical viewpoint, progress has been remarkable, and already a video display on a single crystal Si with 40000 pixels has been demonstrated.[121] With regard to the dyes used in guest-host cells, they should have:

1) Good solubility in the liquid crystal host
2) High dichroic ratio
3) Suitable color
4) Chemical, photochemical and electrochemical stability (non-ionic)

and, last but not least, a high order parameter (i.e., good alignment)

$$S = \frac{3\cos^2\theta - 1}{2}.$$

(θ is the time averaged angle between the axis of the dye molecule and the host director.)

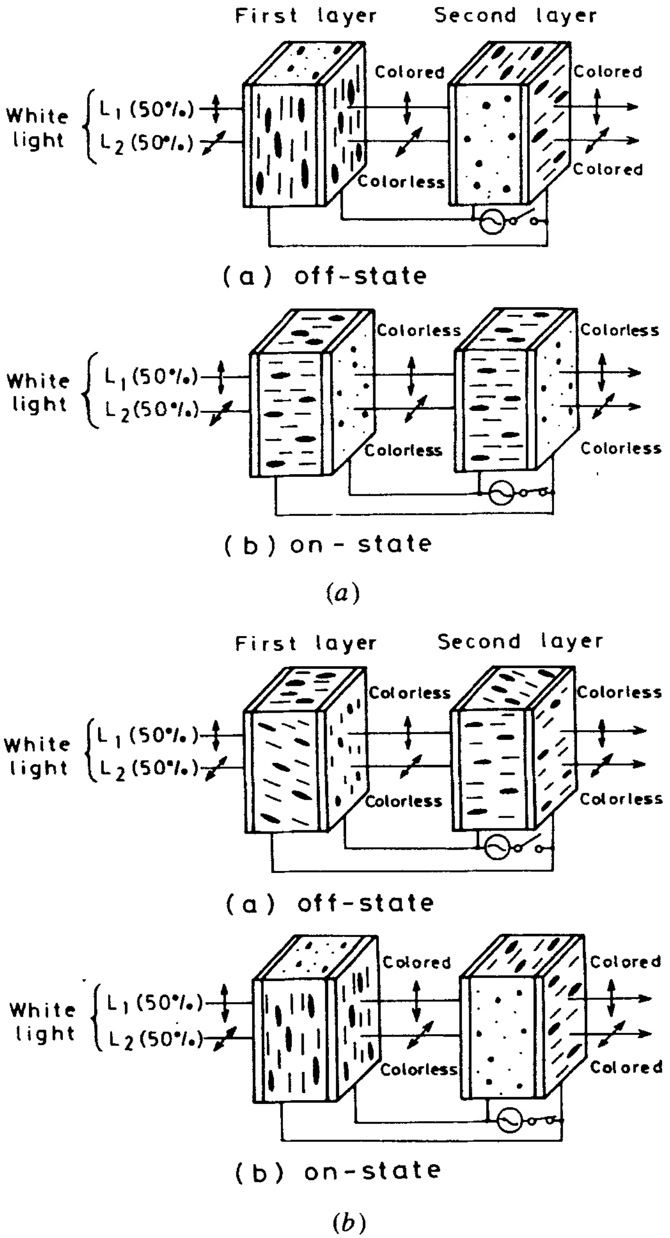

FIGURE 8(a) Negative Double Layer Guest-Host Cell, Ref. 120. Transparent Image on Colored Background; (b) Positive Double Layer Guest-Host Cell. Colored Image on Transparent Background. By permission of T. Uchida and the SID.

According to White and Taylor, $S = (A_\parallel - A_\perp)/(A_\parallel - 2A_\perp)$ where $A_\parallel$ and $A_\perp$ are the optical densities of incident polarized light vibrating, respectively, parallel and perpendicularly to the direction of alignment. Many dye molecules have been investigated for use in guest-host displays, and azo dyes with order parameters as high as 0.79,[122] and the more stable, anthraquinones, having S values as high as 0.70, are of special interest.[122]

The main advantages of dye displays in general are high brightness, good contrast over a large viewing angle and need for only one, or no polarizers at all. On the negative side is the inherent limited multiplexing capability which can be overcome with the help of thin film transistors or varistors. It has also been recognized that the combination of smectics and pleochroic dyes could provide improved contrast in thermally addressed displays[48] and also high multiplexability in electrically addressed displays.[49]

b) Filters

The use of filters to achieve multicolor capability has been discussed for years in the patent literature. Hanlon[123] in 1971 proposed a liquid crystal color modulator device based on adjacent narrow red, blue, and green color stripes. These stripes are located opposite to transparent electrode stripes, 800/inch which can activate a liquid crystal sandwiched between the stripes and a ground electrode. Without an electric field, the liquid crystal is transparent; hence all colors are transmitted and the device looks "white". Simultaneous application of a voltage to the electrodes in front of the green and blue filters, for example, results in blocking out of these colors and, therefore, the device solely transmits red light; alternatively application of a voltage to the electrodes corresponding to the green and red stripes results in transmission of blue light only. Other liquid crystal display concepts in which the color is first selected and then imagewise modulated by other means are disclosed in patents by Krueger[124] and Stolov.[125] The list is by no means exhaustive. Very recently, Uchida[126] discussed a multicolor display also based on filter stripes. The liquid crystal effects were the guest-host, and twisted nematic effects, and both additives and subtractive color systems were discussed. Color was generated by gelatin filters, interference filters, dyed polyvinyl alcohol films, and screen printed inks, all in striped geometry.

Simpler two-color systems have been achieved by sandwiching a twisted nematic cell between two crossed dichroic polarizers of different color as shown by Kobayashi *et al.*,[127,128] or a twisted nematic cell inserted between a polarizer and a birefringent plate and a second polarizer. The reader is referred to Ref. 129 for a detailed explanation of the optics involved.

Despite the relatively large number of technical paths explored to add color to liquid crystal displays, the introduction of commercial products has been cautious and focused on guest-host cells and displays using dichroic polarizers of different colors.

Big potential markets for multicolored liquid crystal displays exist in the automotive and television fields, but the former is traditionally conservative, and the latter will have to wait until all problems associated with the black and white liquid crystal TV are solved.

c) Cholesterics

Cholesteric liquid crystal films can exist in two states: planar and focal-conic, both of which can exhibit intense iridescent colors caused by Bragg-like reflections from optically anisotropic regions, stacked in a helical fashion.

The planar (or Grandjean) state is single-domain, uniaxial, and clear whereas the focal-conic state is multidomain scattering with its optic axes oriented predominantly in the plane of the substrate. The color of a cholesteric film for a given angle of light incidence and observation depends on the pitch (periodicity of the helical structure) and is sensitive to a variety of stimuli including temperature, organic vapors, UV irradiation, shear and electric fields. Much of the pioneering work in this area is due to Fergason[2–4] who also recognized the potential of electrically controlled color changes in cholesterics for displays, which can be effected directly or indirectly (via heating). Whereas the sensitivity of the cholesteric color (pitch) to temperature changes has found applications in many devices, including disposable clinical thermometers, cholesteric color displays have not become practical. Reasons for this can be found in the objectionable dependence of the cholesteric colors on viewing and illumination conditions, limited color saturation and difficulty in maintaining alignment under electric fields.

d) Electrically controlled interference colors

As every mineralogist knows, thin transparent plates viewed in white light between closed polarizers, in general, exhibit interference colors which depend on the thickness, the birefringence and the orientation of the plates studied. Aligned nematic films behave in much the same way, and since the alignment direction can be changed by application of moderate electric fields, it is possible to control the effective birefringence and, therefore, the interference color.

When an electric field is applied to a homeotropic cell, molecules with negative dielectric anisotropy become tilted toward the substrate and, in the case of homogeneous alignment and positive anisotropy, the opposite occurs. The molecules become aligned normally to the substrate that is parallel to the field. These reorientation effects are accompanied by color changes which can go through several orders.

Measurements of the changes in the effective birefringence of aligned nematics under the influence of electric fields were carried out by several authors[71, 130-134] and the significance of this effect for color displays was recognized early. While most of the work has been carried out on homeotropically aligned films (molecular axis normal to the substrate), the opposite configuration, homogeneous alignment, and positive dielectric anisotropy has also been studied. The birefringence of nematics can be quite high, up to 0.2, which permits the use of thin cells, 10.0μ, more or less. This is advantageous because it is possible to use small voltages, and also because it facilitates uniform alignment. As with other liquid crystal display cells, ac fields are used to minimize electrochemical reactions and undesirable electrohydrodynamic instabilities. One of the advantages of the homeotropic $\rightleftarrows$ homogenous transition (or Freedericks transition) as an electro-optic effect is the sharp threshold $V_c = \pi(K_{33}/\Delta\varepsilon)^{1/2}$, where K_{33} is an elastic contrast and $\Delta\varepsilon$ the anisotropy of the dielectric constant. This characteristic makes the effect well suited for matrix address and, in fact, Hareng, *et al.*[135] described a color 50 × 50 pixel device, and Schiekel and Fahrenschon[136] reported a 100 × 100 elements display. Despite the initial promise, the interest in homeotropic $\rightleftarrows$ homogenous displays diminished in time because of the optical sensitivity to viewing angles and the need for stringent thickness uniformity, which makes fabrication of larger displays difficult.

LIQUID CRYSTALS AND PHOTOCONDUCTORS

The combination of photoconductors and liquid crystals offer many interesting possibilities for image storage, light amplification, wavelength conversion, and also conversion of incoherent to coherent light. In all liquid crystal-photoconductor devices, a biased, imagewise, exposed photoconductor controls the field (current) across a liquid crystal layer which responds by changing its optical properties. The result is a liquid image whose properties depend on the liquid crystal effect. Whether the image remains or not after exposure, depends on the liquid crystal electro-optic effect used. A light valve in the simplest form is shown in Figure 9.

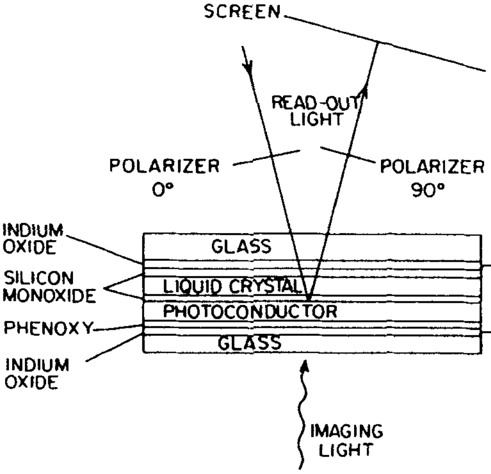

FIGURE 9 Schematic of Image Intensifier.

It is clear that such a device will function when storage effects are involved, i.e. read-out follows exposure, but it will not be effective in real-time situations because in this case the read-out light interferes with the imaging light. This problem, however, can be solved in several ways: for example, deposition of a mosaic of reflective metallic dots onto the photoconductor;[137] the use of read-out wavelengths to which the photoconductor is insensitive[138] insertion of an optical blocking layer and dielectric mirror with suitable conductivity between photoconductors;[139] and liquid crystal on the use of photoconductors with special absorption characteristics.[140]

Liquid crystal-photoconductor systems have been under investigation since 1968,[141] and a large number of combinations have been studied. Early devices used dynamic scattering[138,142] for real-time, and the planar ⇄ focalconic[138,141,143] texture change for storage displays. Other effects investigated in conjunction with photoconductors were the variable grating mode,[144] the cholesteric-nematic phase transition,[140] and the Freedericks transition.[145] Recently, the tilt appears to be more toward the twisted nematic and the "hybrid field effect", a combination between a pure optical birefringence and a 45° twisted nematic effect.[146]

Device operation is typically as follows: prior to exposure, the voltage drops essentially across the photoconductor and the voltage across the liquid crystal is below threshold. During exposure, the voltage across the liquid crystal increases to values above threshold, and the liquid crystal is activated. With regard to the photoconductor, ZnS,[138] ZnO,[142] CdS,[139] Se,[140,141,147] As_2Se_3,[148] and Si[149,150] have all been used with varying degrees of success. In general, the devices are read-out in reflection and may or may not have a dielectric mirror. Liquid crystal-photoconductor devices can have very good characteristics: sensitivity 5 ergs/cm^2 or better, resolution up to 90 l/mm, 9 shades of gray and high contrast. A photograph of an image projected onto a screen from a light valve as described in Ref. 148 is shown in Figure 10.

FLUORESCENT DISPLAYS

Fluorescence has been used in two fundamentally different ways to improve the optical characteristics of liquid crystal displays. In the first method, described by Baur and Greubel,[151] a twisted nematic liquid crystal cell is in close proximity to a plastic plate doped with fluorescent molecules. Ambient light collected by this plate is converted to fluorescent light and directed toward the addressed zones of display which become lit against a black background.

The second method which is in fact a guest-host effect,[152–154] in which fluorescent molecules of europium chelate are added to a cholesteric host capable of undergoing the cholesteric-nematic phase transition. Excited by

FIGURE 10 Photograph of Image Projected Onto Screen with Twisted Nematic Light Valve, Ref. 148.

an external source, the fluorescence is more intense in the scattering state (stronger absorption) than in the clear homeotropic state. Labes' group at Temple University recently reported another variety of fluorescent displays[154] based on the combination of a liquid crystal shutter cell with passive fluorescent elements.

STANDARDS

The desirability of liquid crystal display standards was recognized early by industry in the U.S. and Japan. Preliminary discussions initiated in 1972 at the Fourth International Liquid Conference in Kent, Ohio, led to the formation of the JC23.1 Committee under the aegis of the Joint Electron Devices Council (JEDEC) of the Electronics Industry Association. In Japan, in 1974, a LCD Committee under the Electronic Industries Association of Japan (EIA-J) was also formed to address the questions associated with standardization. Both committees performed valuable services and addressed terminology, test methods for electrical and electro-optical properties, glass and polarizer characteristics, etc. Yet, after many years of work, accepted international standards have not yet become a reality.

An issue which is becoming of increasing concern to users and manufacturers of displays in general is the human interaction with the display. As the information content of liquid crystal displays increases, viewability and legibility will receive increasing attention, particularly in the case of color displays which are coming of age, and whose evaluation is complex.[155]

CONCLUSION

The history of liquid crystal displays spans three continents, Asia, Europe, and North America, and is truly an example of international technical interactions. The initial impetus was provided by work in the U.S. which disclosed many of the basic electro-optic effects and demonstrated the possibilities inherent in liquid crystal displays. Major contributions, including enabling breakthroughs in liquid crystal materials, originated in Europe, but it was Japan's private industry which made liquid crystal displays a reality. By looking further ahead than others and by bringing its high technology manufacturing skills to bear on liquid crystals, reliable and affordable displays became available; and today, after piling up innovation upon innovation, Japan is the leading display manufacturer in the world.

Liquid crystal displays are continuously challenged by competing technologies such as vacuum fluorescence, electroluminescence, plasma, LED's, electrochromics, electrophoretics, etc., but rather than yielding ground, the use of liquid crystals is continuously expanding into new fields. In applications requiring low power consumption, and/or good viewability under high ambient light conditions, liquid crystals have become unbeatable, and beyond that they do better than simply holding their own. The ubiquitous watch and calculators are known to all of us, but liquid crystal displays are also used in many other applications such as instruments of all sorts, office equipment, graphic status panels, message panels, personal computers, printbars, audio equipment, games, gas pumps, etc. The list is very broad. Two new large fields are just on the horizon: automotive displays, and portable TV's. Beyond that lies the ultimate target of many liquid crystal technologists, the flat panel color display capable of competing head-on with full size color CRT's. Because of the steady and intense ongoing work on materials, new effects and electronically aided addressing, the probability is very high that viewability and information content of liquid crystal displays will be substantially augmented in the future. It is also likely that liquid crystal displays in coming years will find many new uses devised by clever innovators or will become practical for applications proposed earlier but buried in the literature for the lack of viable technologies.

In summary, the future looks bright for liquid crystals.

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