

# The History of Liquid-Crystal Displays

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## Invited Paper

*The modern history of liquid crystals has been dominated by the development of electronic displays. These developments began in 1964, when Heilmeier of RCA Laboratories discovered the guest-host mode and the dynamic-scattering mode. He thought a wall-sized flat-panel color TV was just around the corner. From that point on, twisted-nematic (TN) mode, super TN mode, amorphous-Si field-effect transistor, and room-temperature liquid crystals were developed. In the beginning, liquid-crystal displays (LCDs) were limited to niche applications such as small-size displays for digital watches, pocket calculators, and small handheld devices. That all changed with the development of the notebook computer industry. In 1988, Washizuka et al. of Sharp Corporation demonstrated an active-matrix full-color full-motion 14-in display using a thin-film-transistor array. The electronics industries now recognized that Heilmeier's 25-year dream of a wall-hanging television had become reality. LCDs could be used to replace existing cathode ray tubes. Through the cooperation and competition of many electronics giants, the LCD industry was firmly established.*

**Keywords**—Active matrix, amorphous silicon, azoxy, birefringence, cholesteric, cyanobiphenyl, dielectric anisotropy, digital watch, DSM, DSTN, ester, Fergason, Gray, guest host, Heilmeier, Helfrich, history, LCD, liquid crystals, MBBA, Nehring, nematic, PCH, pocket calculator, poly silicon, Raynes, rubbing, Schadt, Scheffer, Seiko, Sharp, STN, TFT, TN mode, transmission minimum, Wada, wall-hanging television, Washizuka, Williams domain, Yamazaki.

## I. INTRODUCTION

The development of liquid-crystal displays (LCD) proceeded from early successes like the pocket calculator to the major milestone of a flat-panel television display you can hang on a wall. The history of that development spans the world's major industrial centers: the U.S., Japan, and Europe. I was fortunate to be a part of that history. When I joined RCA Laboratories at the David Sarnoff Research Center in April 1970, RCA was curtailing its efforts in liquid-crystal activities, but I had the opportunity to witness the developments there before the program's group head, George Heilmeier, left for Capitol Hill as a White House

Fellow. Then, in 1985, I joined the Sharp Corporation in Japan and met Tomio Wada, the man who developed the world's first liquid-crystal product, a pocket calculator, in 1973. At Sharp, I also witnessed major development efforts in LCDs at the Tenri Advanced Development Center. In 1990, I participated in the founding of the European Laboratories at Oxford, U.K., and in 1992, we welcomed Peter Raynes, known for his contributions to the applications of cyanobiphenyls, to the Laboratories. Through discussions with him, I learned about the achievements of British and European scientists.

The modern history of liquid crystals is predominantly the history of the development of electronic displays made of liquid crystals. The developments started when a dynamic-scattering mode (DSM) was discovered in 1964. Manufacturers of LCDs had been minor-league members of the electronic display industry and served a niche market, supplying small-size displays primarily to pocket calculators and digital watches. A major milestone was reached in 1988 when a 14-in active-matrix (AM) thin-film-transistor (TFT) display was demonstrated. The electronics industries then recognized that the dream of a wall-hanging television had become a reality, thus, promoting LCD manufacturers to the "major leagues" in the electronics industry. By 2000, the LCD industry had caught up to the giant cathode ray tube (CRT) industry. In this paper, I focus on the 25 years of LCD developments that gave birth to the present-day LCD industry. In writing this paper, I interviewed 37 scientists and engineers scattered throughout the world. Each section of the article concentrates on a key technical item that led to the ultimate goal—the flat-panel television.

What had been an obscure general and scientific curiosity for 80 years suddenly became the center of attention as the result of a new invention, spawning a new industry projected to reach 40 billion dollars by the year 2006. The history of LCDs is a story of the hard work, disappointments, and successes of worldwide competition and cooperation that encompassed the U.S., Europe, and Japan. Each industrial center contributed its particular strengths: in America, it was the quickness of forming new ideas and demonstrating their feasibility; in Europe, it was the fundamental science

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and synthesis of basic materials; and in Japan, it was the process of perfecting implementation and moving it to the production line.

## II. DISCOVERY OF LIQUID CRYSTALS AND THEIR FUNDAMENTALS—THE NAME “LIQUID CRYSTALS” WAS BORN IN GERMANY

As a material's temperature is raised, it generally changes state from solid to liquid to gaseous. It is generally believed that an Austrian botanist, Friedrich Reinitzer [see, e.g., Fig. 1(a)], first observed liquid crystals in 1888. He discovered a strange material that exhibited a mesophase between solid state and liquid state [1]. At a temperature of 145 °C, it melted, becoming cloudy white and viscous. At a temperature of 179 °C, it became isotropic and clear. The material he discovered was cholesteryl benzoate. On March 14, 1889, he wrote a letter to Otto Lehmann [see, e.g., Fig. 1(b)], Professor of Physics at the Technical University Karlsruhe of Germany, telling him about the two melting points. Lehmann studied the material and discovered that the liquid at the mesophase exhibited a double refraction effect, characteristic of a crystal. Because it shared characteristics of both liquid and crystal, he named it “fliessende krystalle” and the name “liquid crystal” was born [2].

Liquid crystals were not popular among scientists in the early 20th century and the material remained a scientific curiosity for 80 years. It should be noted that E. Merck of Darmstadt, Germany, sold liquid crystals for analytical purposes as far back as 1907 (see Fig. 2). In the early 1960s, only a few institutions and corporations were known to have been carrying out research on liquid crystals. The prerequisites for designing liquid crystals with specific physical properties hardly existed, not to mention the lack of motivation to use it in a commercial product. Liquid crystals were unknown to the man on the street.

### A. Thermotropic and Lyotropic Liquid Crystals

The liquid crystals Reinitzer discovered by varying their temperatures are called thermotropic liquid crystals (see Fig. 3). As the temperature is raised, their state changes from crystal to liquid crystal at temperature  $T_1$ . Raising the temperature further changes the state from liquid crystal to isotropic fluid at temperature  $T_2$ . Generally speaking, the process is reversible by lowering the temperature, though there may be a small temperature hysteresis; the  $T_1$  value when reducing temperature may be slightly less than the  $T_1$  value when increasing temperature. The  $T_1$  is sometimes referred to as the “freezing” temperature and  $T_2$  as the “clearing” temperature. For the case where a liquid crystal is of nematic type as will be discussed later, the  $T_1$  is designated by  $T_{CN}$  and  $T_2$  by  $T_{NI}$ .

Another liquid crystal exists. It is called the lyotropic liquid crystal. This exhibits liquid-crystal behavior when it reacts with water or a specific solvent [4]. For example, the wall of a biological cell is made of lyotropic liquid crystals. Many biological structures such as the brain, nerve system, muscle, and blood contain lyotropic liquid crystals.



(a)



(b)

**Fig. 1.** Scientists who discovered liquid crystals. (a) Friedrich Reinitzer (1857–1927). (b) Otto Lehmann (1855–1922). (Historical Exhibition, the 12th International Liquid Crystal Conference, Freiberg, Germany, August 15–19, 1988).

The lyotropic liquid crystals are primarily investigated in the fields of biochemistry, biophysics, and bionics. We will leave the discussion of lyotropic liquid crystals to other publications while we focus on thermotropic liquid crystals.



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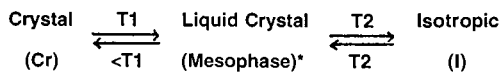
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Fig. 2. Liquid crystals sold as far back as 1907 for analytical works in research laboratories. Courtesy of Ludwig Pohl.



where  $T_2 > T_1$

\* A SINGLE PHASE OR A SEQUENCE OF PHASES

CRYSTAL----- PERFECT 3-D ORDER

LC STATES----- INTERMEDIATE DEGREES OF ORDER

ISOTROPIC -----TOTALLY DISORDERED

Fig. 3. Thermotropic liquid crystal [3].

### B. Cigar-Like and Disc-Like Molecules

There are two types thermotropic liquid crystals. One type has cigar-like molecules, while the other has disc-like molecules and is referred to as "discotic" (see Fig. 4). Both types are mostly organic, as seen from the formulae of the examples given. The liquid crystals used in electronic displays are primarily of the cigar type. In this article, therefore, we concentrate on thermotropic liquid crystals made of cigar-like molecules. Only in the late 1990s did the discotic molecules find an application in electronic displays. They are used to make a sheet of film that expands the viewing angle of a twisted-nematic (TN) display.

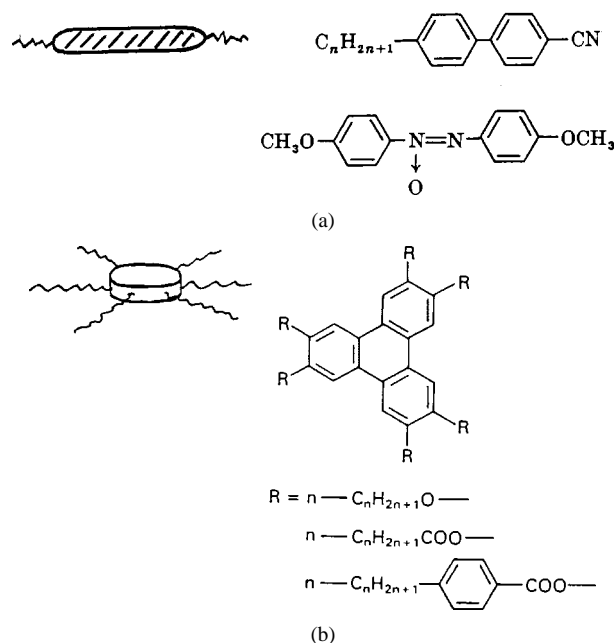


Fig. 4. (a) Cigar-like and (b) disc-like molecules [3].

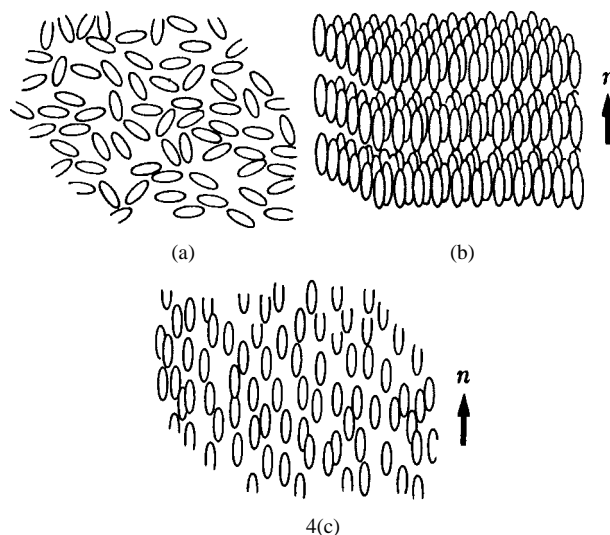


Fig. 5. Molecular alignments in liquid and liquid crystals. (a) Liquid. (b) Smectic. (c) Nematic.

### C. Thermotropic Liquid-Crystal Types

There are three types of thermotropic liquid crystals. These are based on a system proposed by G. Friedel in 1922 [5]. They are smectic, nematic, and cholesteric types.

1) *Smectic Liquid Crystals*: Smectic comes from a Greek word meaning grease or clay. In smectic type liquid crystals, the cigar-like molecules are arranged side by side in a series of layers as shown in Fig. 5(b). The long axes of all molecules in a given layer are parallel to one another and perpendicular to the plane of layers. The layers are free to slip and move over each other. The smectic state is viscous, but fluid and ordered.

2) *Nematic Liquid Crystals*: Nematic comes from a Greek word meaning thread-like. Under a microscope using polarized light, nematic liquid crystals appear as thread-like structures. In the nematic state, the molecules are not as

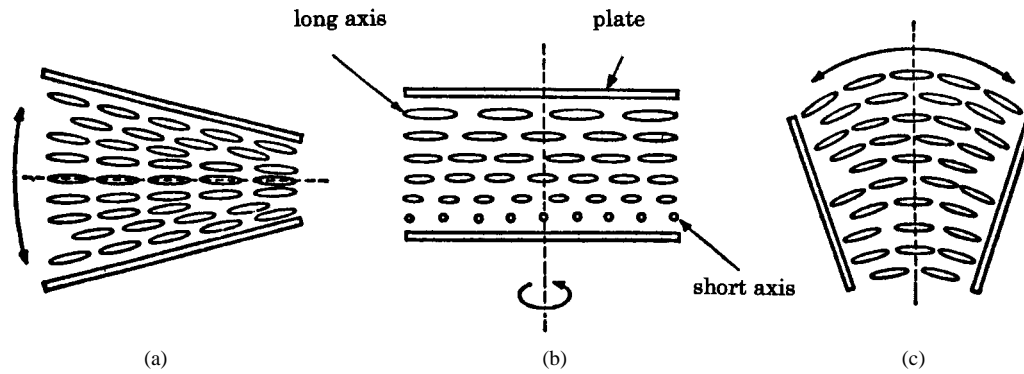


Fig. 6. Three molecular alignments of nematic liquid crystals [4]. (a) Splay. (b) Twist. (c) Bend.

highly ordered as in the smectic state, but they maintain their parallel order [see, e.g., Fig. 5(c)]. On average, the nematic liquid crystals are aligned in one direction. The direction is represented by a vector  $n$  called a director. Liquid crystals used in electronic displays are primarily of the nematic type.

Because of its specific molecular alignment, nematic liquid crystals exhibit anisotropic physical characteristics; their refractive index, dielectric constant, permeability, electrical conductivity, and viscosity measured in the direction of the long axis are different from those measured in the plane normal to the long axis. In nematic liquid crystals, the refractive index along the director axis is almost always larger than along the perpendicular axes. The electrical conductivity along the director axis is generally larger than along the perpendicular axes. The permeability is generally negative and its absolute value along the director axis is smaller than along the perpendicular axes.

3) *Cholesteric Liquid Crystals*: These materials are discussed later in this section.

#### D. Elasticity

Under mechanical stress, liquid crystals deform their molecular alignment. The deformation of nematic liquid crystals can be considered for three cases, shown in Fig. 6. The first is a “splay,” where molecules are spread by external stress, the second is a “twist” where molecules are twisted by an external stress, and the third is a “bend” where molecules are bent by an external stress. The relationship between the deformation and stress is expressed respectively by splay elasticity  $k_{11}$ , twist elasticity  $k_{22}$ , and bend elasticity  $k_{33}$ .

The elasticity of liquid crystals is of the order of  $10^{-6}$  to  $10^{-7}$  dyne and is much lower than that of ordinary elastic material. This makes alignment modification of liquid crystals by the application of electrical field, magnetic field and external stress much easier. The density of free elastic energy for nematic liquid crystals under deformation is expressed by [4]

$$f_d = \left(\frac{1}{2}\right) k_{11}(\text{div } n)^2 + \left(\frac{1}{2}\right) k_{22}(n \bullet \text{rot } n)^2 + \left(\frac{1}{2}\right) k_{33}(n \times \text{rot } n)^2. \quad (1)$$

#### E. Dielectric Anisotropy

Regarding the dielectric constant, there are two types of nematic liquid crystals. One is called positive dielectric anisotropy (p-type) and its dielectric constant along the director axis is larger than that along the axes perpendicular to the director [4]. The  $\Delta\epsilon$ , which is equal to  $\epsilon_{\text{parallel}} - \epsilon_{\text{perpendicular}}$ , is in the range of +10 to +20. The other type is called negative dielectric anisotropy (n-type) and its dielectric constant along the director axis is smaller than that along the axes perpendicular to the director. The  $\Delta\epsilon$  is in the range of -1 to -2.

Applying an electrical field  $E$  to the liquid crystals, there appears an electrical energy  $f_e$ , where

$$f_e = -\left(\frac{1}{2}\right) \epsilon_{\text{perpendicular}} \epsilon_0 E^2 - \left(\frac{1}{2}\right) \Delta\epsilon \epsilon_0 (n \bullet E)^2. \quad (2)$$

Here, the first term is independent of the director  $n$ . The second term changes value depending on the direction of  $n$ . When  $\Delta\epsilon$  is positive (p-type liquid crystal), the application of an electrical field greater than some certain critical value ( $E > E_c$ ) aligns the long axis of the molecules parallel to the direction of the electrical field  $E$ . This happens because the electrical energy  $f_e$  is minimized when the director  $n$  is parallel with the electrical field  $E$ . On the other hand, when  $\Delta\epsilon$  is negative (n-type liquid crystals), the long axis of molecules aligns perpendicular to the electrical field because the electrical energy is minimized when  $n$  is at a right angle to  $E$ . Therefore, by applying an electrical field, we can control the direction of nematic molecules.

Consider the case when the alignment of molecules is such that the liquid crystals have been in their lowest energy state and an electrical field is then applied. The total free energy  $F$  of the liquid crystal is expressed by the summation of electrical energy  $f_e$  and elastic energy  $f_d$

$$F = \text{Integration} [f_e + f_d] d\tau. \quad (3)$$

An important case in applications occurs when nematic liquid crystals are sandwiched by two parallel plates and the electrical field is applied normally to the plates. A critical field  $E_c$  that causes the transition of the molecular alignment can be expressed by

$$E_c = \left(\frac{\pi}{d}\right) \left(\frac{k_{ii}}{|\Delta\epsilon \epsilon_0|}\right)^{1/2} \quad (4)$$

where  $d$  is the spacing of the plates. When the base state of the molecular alignment is homogeneous,  $k_{ii} = k_{11}$  and when the alignment is twisted,  $k_{ii} = k_{11} + (k_{33} - 2k_{22})/4$ . When the liquid crystals are positive and the alignment is twisted as shown in Fig. 6(b), the threshold voltage for transition is

$$V_C = \pi \left[ \frac{k_{11} + \frac{k_{33} - 2k_{22}}{4}}{|\Delta\epsilon\epsilon_0|} \right]^{1/2}. \quad (5)$$

#### F. Birefringence

Because of its double refraction or birefringence property, a liquid crystal exhibits the following optical characteristics [4].

- 1) It redirects the direction of incoming light along the long axis (director  $n$ ) of the liquid crystal.
- 2) It changes the state of polarization (from linear, ellipse, or circular polarization to one of linear, ellipse and circular polarizations) and/or changes the direction of polarization.

Liquid crystals are not as rigid as solids and are easily re-oriented, realigned, or deformed by applying electrical fields, magnetic fields, heat, and/or mechanical stresses. Accordingly, the optical characteristics based on the birefringence are easily affected. These make nematic liquid crystals attractive for use in electronic devices. The following sections trace the history of how scientists and engineers used such characteristics of nematic liquid crystals to construct electronic displays.

#### G. Cholesteric Liquid Crystals

The term cholesteric is used because the molecular structure of the liquid crystals characteristically has a large number of compounds containing cholesterol [6]. The molecules in cholesteric liquid crystals are arranged in layers (see Fig. 7). Within each layer, molecules are aligned in parallel, similar to those in nematic liquid crystals. The molecular layers in a cholesteric liquid crystal are very thin, with the long axes of the molecules parallel to the plane of the layers. A special aspect of the cholesteric structure is that the director  $n$  in each layer is displaced slightly from the corresponding director of the adjacent layer. The displacement is cumulative through successive layers, so that the overall displacement traces out a helical path. Because of the helical structure, it exhibits such interesting phenomena as optical rotation, selective reflection and two-color circular polarization.

The phenomenon of selective reflection was used in commercial applications before liquid crystals were used in display applications. The pitch  $p$  of the helical structure is of the order of the wavelengths of visible light. A beam of white light coming in the direction of the helical axis splits into two polarized beams: one beam with a right circular polarization and the other with a left circular polarization. Here, the light having its rotation of polarization in the same direction as the helical rotation is selectively reflected at the end surface; the light with the other rotation of polarization, transmits. Using the Bragg relation, we see that a film of the liquid crystal will

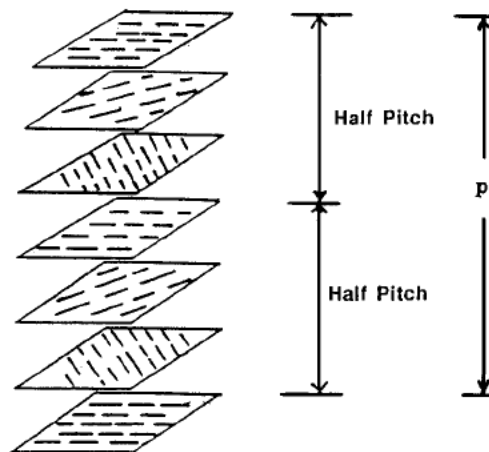


Fig. 7. Cholesteric liquid crystal [3].

reflect the light and appear colored. The wavelength  $\lambda$  of the reflected light is

$$\lambda = p \left( 2 \left( n_{\text{parallel}}^2 + n_{\text{perpendicular}}^2 \right) \right)^{1/2} \quad (6)$$

where  $n_{\text{parallel}}$  and  $n_{\text{perpendicular}}$  are refractive indices in parallel and rectangular respectively to the long axis of the molecule [4]. The pitch  $p$  is subject to change with temperature and so the color of the film also changes with temperature. Generally speaking, as temperature increases,  $p$  becomes small and the reflected light moves to a shorter wavelength. Ferguson and his group at Westinghouse Research Laboratories fabricated flexible films and tapes that can be applied to the surface of objects to record temperatures. They applied the film to the skin of a human body to locate veins and arteries and to electronic circuit boards to locate trouble spots [6].

One product incorporating cholesteric liquid crystals available in the market place is a stress testing card. This is a credit-card-sized plastic card on which cholesteric liquid crystals are painted. Ordinarily, the card is black. One presses the card between thumb and index finger for 15 seconds. If the color changes to blue, the card indicates that finger temperature is high and the subject is relaxed. If the color changes to red, the subject's finger temperature is low, indicating tension.

Cholesteric liquid crystals are also used as additives in a TN and super TN (STN) LCDs, which will be discussed later.

### III. DYNAMIC-SCATTERING MODE—AMERICA'S AGILITY IN TAKING A NEW IDEA AND DEMONSTRATING ITS FEASIBILITY

The development of LCDs started at RCA Laboratories in Princeton, NJ.

#### A. Williams Domain

In 1962, Richard Williams of RCA found that liquid crystals had some interesting electrooptic characteristics [7], [8]. He sandwiched a liquid crystal p-azoxyanisole (PAA), a negative nematic type liquid crystal, between two glass plates. The range over which it exhibited the liquid-crystal phase was from 117 °C to 134 °C. The liquid-crystal specimen between the glass plates was heated to a liquid-crystal tempera-

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