OLED DISPLAY

FUNDAMENTALS AND APPLICATIONS

TAKATOSHI
TSUJIMURA

WILEY

SID Series in Display Technology

OLED Displays
Fundamentals and Applications

Wiley-SID Series in Display Technology

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Anthony C. Lowe

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Series Editor's Foreword

I have long aspired to publish a book on OLED technology in this series. From the beginning, the problem I encountered was that the technology was so new and was developing so rapidly that any book written would have been out of date before it was published. Although the technology is still developing at a prodigious rate, it has reached a level of maturity that at last makes possible the publication of a book covering all aspects of OLED technology from a fundamental description of light emission from OLED materials through TFT design to manufacturing techniques and applications not just to displays but also to lighting. Books previously published in the field have essentially been collections of research papers. Useful as such books are, they do not fulfill the objectives of the Wiley-SID series, so I hope that those who require a text covering all aspects of this important technology will consider this latest book in the series to have been well worth the wait.

To provide the reader with a précis of the scope of this book, after an introductory chapter which focuses on early OLED-based products, Chapter 2 contains a brief history of OLED development and then goes on to describe in detail and on the basis of actual device structures the electronic and thermodynamic fundamentals of light emission from small molecule and polymeric, singlet and triplet OLED materials, the mechanisms of charge injection, transfer and recombination, electron and optical efficiency, and degradation and lifetime issues. This chapter contains all of the science of the operation of a light-emitting OLED structure.

Chapter 3 describes the manufacturing processes for all of the layers in a working OLED device and covers purification and deposition processes, monitoring techniques for quality control, defects and degradation phenomena, and the methods for their detection and analysis. Chapter 4 describes

what is required to convert OLED devices into actual display modules which can use either active or passive matrix addressing. Chapter 5 discusses TFT addressing circuitry based on LTPS technology and describes in detail the specific requirements for the circuit designs required by current-driven OLED technology. Chapter 6 addresses the next generation of OLED technology, and covers patterning techniques, solution processed materials, new higher yield manufacturing processes, and improved TFT technologies. The book concludes with chapters on OLED lighting and on new display applications of OLEDs in large size and flexible displays.

Dr. Tsujimura has been actively involved in OLED display development since the early days in the 1990s when active matrix TFT technology reached a stage of development that made high pixel count OLED displays technologically feasible. His expertise in the technology is both deep and broad, as readers will appreciate when they delve deeper into this latest addition to the series.

Anthony C. Lowe
Series Editor
Braishfield, United Kingdom

Preface

Organic light-emitting diode (OLED) and liquid crystal display (LCD) technologies are often compared side by side; however, the LCD is the successful predecessor of the OLED display. According to the experience of this author, who was engaged in developing LCD displays from phase 1 thin-film transistor (TFT) LCD line manufacturing in the past, it is somewhat miraculous that the relatively new OLED and the well-established, mature LCD technologies are compared on an apples-to-apples basis or as parallel technologies. This comparison indicates the inherently high quality of OLED displays.

The success of the vast market for LCD displays can be attributed not only to the intensive research and development efforts of the display manufacturers proper (Samsung, Sharp, LG Display, etc.) but also to the contributions of various individuals involved in components and equipment testing, measurement, and manufacturing, and in both marketing and academic research. The author hopes that the OLED industry will benefit from equally vigorous and numerous contributions of specialists either directly or indirectly involved in OLED manufacturing, and that the OLED market will eventually become as wide and robust as the LCD market is today.

The LCD industry grew in tandem with developments in related fields (manufacturing equipment testing, etc.); thus there are many aspects applicable to OLED technology, which are discussed in detail in this book. Each topic is presented with a discussion of its basic theory and background, followed by description and examples of its practical application. The author has tried to present all of these technologies in as broad a scope as possible to ensure that readers involved in all aspects of OLED research and development will benefit from this book.

On receiving the SID Special Recognition award in 2007, the author expressed his desire to make significant contributions to, and thus fortify and expand the fledgling OLED industry. The author hopes that this book will help in achieving that objective, and that it will contribute to the overall industrywide growth of OLED technology.

Takatoshi Tsujimura

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1 Introduction

The basic structure of organic light-emitting diodes (OLEDs) was reported by Ching W. Tang and Steven Van Slyke at Eastman Kodak in 1987 [1]. This was a groundbreaking study and was later referred to as the "first OLED paper." Now, almost 25 years later, there is a large market place for OLED devices. After the first active-matrix-driven OLED (AMOLED) display was introduced by SK Display (a joint manufacturing venture by Eastman Kodak and Sanyo Electric), the first product using an AMOLED display was Kodak's LS633 digital camera (see Fig. 1.1). This was followed by the widescale development of many other OLED-based products, including cellular phones, audioplayers (Fig. 1.2), portable multimedia players (Fig. 1.3), and portable global positioning satellite (GPS) devices, which now provide high-resolution displays in brilliant, multitone colors.

Larger-display products have also been introduced on the market, such as those shown in Figs. 1.4 and 1.5. Much larger (e.g., 20–400-in.) prototypes have also been developed. Because of superior features such as slim flat-screen design and aesthetically pleasing screen image, and due to high-contrast image signal emission and very good response time, the current state of the art of OLED television technology that has debuted in the marketplace is indeed unprecedented. [2]

The main objective of this book is to explain the basics and application of this promising technology from various perspectives.

OLED Displays: Fundamentals and Applications, First Edition. Takatoshi Tsujimura.

Figure 1.1 The first active-matrix OLED display product on the market (Kodak LS633 digital camera).

Figure 1.2 Example of an audioplayer using active-matrix OLED (AMOLED) (Sony Walkman NW-X-1050).

Figure 1.3 Example of a personal multimedia player using AMOLED (Dynaconnective Dawin; original equipment manufacturer [OEM] product of the Neosol Cliod).

Figure 1.4 Digital photo frame obtained using 8-in. OLED screen (Kodak OLED wireless frame).

Figure 1.5 An 11-in.-screen television set (Sony XEL-1).

References

1. C. W. Tang and S. A. Van Slyke, Organic electroluminescent diodes, *Appl. Phys. Lett.* **51**(12):913–915 (1987).
2. T. Tsujimura, W. Zhu, S. Mizukoshi, N. Mori, M. Yamaguchi, K. Miwa, S. Ono, Y. Maekawa, K. Kawabe, M. Kohno, and K. Onomura, Advancements and outlook of high performance active-matrix OLED displays, *SID 2007 Digest*, 2007, p. 84.

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2 OLED Display Structure

2.1 OLED DEFINITION

Before any in-depth discussion of OLED display structure, let us consider the initial origins of OLED technology, which are based on early observations of electroluminescence. In the early 1950s, a group of investigators at Nancy University in France applied high-voltage alternating-current (AC) fields in air to acridine orange and quinacrine, which were dissolved in or deposited on thin-film cellulose or cellophane [1]. One mechanism identified in these processes involved excitation of electrons. Then in 1960 a team of investigators at New York University (NYU) made ohmic dark-injecting electrode contacts to organic crystals and described the necessary workfunctions (energy requirements) for hole and electron-injecting electrode contacts [2]. These contacts are the source of charge injection in all present-day OLED devices. The same NYU group also studied direct-current (DC) electroluminescence (EL) in vacuo on a single pure anthracene crystal and tetracene-doped anthracene crystals in the presence of a small-area silver electrode at 400 V [3]. The proposed mechanism for this reaction was termed *field-accelerated electron excitation of molecular fluorescence*. The NYU group later observed that in the absence of an external electric field, the EL in anthracene crystals results from recombination of electron and hole, and that the conducting-level energy of anthracene is higher than the exciton energy level [4].

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Because of the association between electroluminescence and later OLED development on the basis of these and other early EL studies, the term *organic EL* gradually emerged and is still used today. Electroluminescence includes two basic phenomena:

1. Light emission due to the presence of excited molecules caused by accelerated electrons (i.e., electrons that are accelerated to higher energy levels).
2. Light emission due to electron–hole recombination, as in all light-emitting diodes (LEDs).

Phenomenon 1 is the narrower definition. Current OLED devices, after Tang and Van Slyke's "first OLED paper," utilize LED-like emission mechanisms, that is, phenomenon 2.

Table 2.1 lists the differences between a liquid crystal display (LCD) and an OLED display. The OLED has a very short response time and is capable of using "punching" (an imaging technique for enhancing the local luminance to emphasize the highlighted region of an image). The punching technique is used in cathode ray tubes (CRTs), which can have much higher luminance of a dot than the screen luminance. An OLED can use a similar operation, while a normal LCD display cannot.

Table 2.2 outlines the chronological history of OLED technology development. This table was prepared by the Society for Information Display (SID), which holds the world's largest conference on this topic.

TABLE 2.1 Differences between Liquid Crystal and OLED Displays

Parameter	LCD	OLED
Response time	*Slow; hold-like (delayed)*	*Fast, impulse-like (rapid)*
Punching	*Difficult*	*Possible* 
Viewing angle	*Narrower high contrast angle region*	*Lambertian distribution**
Number of components	*More*	*Fewer*
Differential aging**	*Small*	*Larger*
Susceptibility to water and O_2	*Small*	*Larger*

*Outgoing light distribution whose luminace is proportional to cosθ.
**Luminance reduction in terms of use of a particular pixel and between colors.

TABLE 2.2 Timeline for OLED Technology Development

Year	Event[a]	Company/Institute
1960–mid–1970s	D OLED crystal molecule, anthracene, etc.	NRC (Canada), RCA
1987	P OLED diode structure paper in *Appl. Phys. Lett*.	Eastman Kodak
1990	P first PLED paper in *Nature*	Cambridge Univ.
1996	P first AMOLED demonstration (QVGA)	TDK
1998	D first phosphorescence OLED	Princeton Univ.
1999	D first passive OLED product	Pioneer
2001	D 0.72-in. headmount display by AMOLED on silicon	eMagin
2001	D 13-in. SVGA AMOLED prototype	Sony
2001	D 2.1-in. 130-ppi AMOLED prototype	Seiko Epson/CDT
2002	D 15-in. 1280 × 720 OLED prototype	Eastman Kodak/Sanyo
2003	D digital camera with 2.2-in. AMOLED display	Eastman Kodak
2003	D Tiled 24-in. AMOLED prototype with by 12-in. display	Sony
2003	D 20-in. phosphorescence AMOLED prototype by a-Si backplane	ChiMei/IDT/IBM

[a]*Abbreviations in this column*: a-si—amorphous silicon; AMOLED—active-matrix OLED; D—development of; P—publication or presentation/demonstration of; PLED—polymer (O)LED; ppi—pixels per inch; QVGA—quarter videographics array (320 × 240 pixels); SVGA—super videographics array (800 × 600 pixels).
Source: SID International Symposium (2003), 40 Years of SID Symposia—Nurturing Progress in EL/OLED Technology, Baltimore, MD.
http://www.sid.org/Archives.aspx

The chronological sequence of development listed in Table 2.2 reflects the emergence of some general form of classification of OLED technologies, including

- Small-molecule OLED (SMOLED) and polymer OLED (PLED)
- Passive-matrix OLED (PMOLED) and active-matrix OLED (AMOLED) displays
- Fluorescence emission and phosphorescence emission.

The developments listed here and in Table 2.2 indicate that the rapid advances in OLED technologies resulted from extensive experimental trial and error. Each technology is discussed in further detail later in the book.

2.2 OLED DEVICE PRINCIPLES AND MECHANISMS

2.2.1 Basic Device Structure

Emission from all OLED devices—whether of the small-molecular or polymer family—can be explained by the same principle. Through electron–hole recombination, a high-energy molecular state is formed. This state is called an *exciton*, as it behaves like a single molecule with high energy. This exciton generates light after an exciton lifetime period (Fig. 2.1). [Another type of emission, termed *photoluminescence* (PL) emission, is caused by light (e.g., UV)-induced molecular excitation.]

The wavelength of this light emission corresponds to the exciton energy, so it is possible to control the color of the emission by adjusting the molecular design of the color center. This feature is quite advantageous for OLED display applications.

In experimentation using tetracene-doped anthracene crystals and materials, OLED emission had been observed before the so-called first OLED paper in 1987 [5] (see Row 1 in Table 2.2 [6]). However, the voltage and efficiency levels were insufficient for actual application. The scenario depicted in Fig. 2.2 and described in the Tang–Van Slyke paper [5] represents advanced concepts that remain valid today:

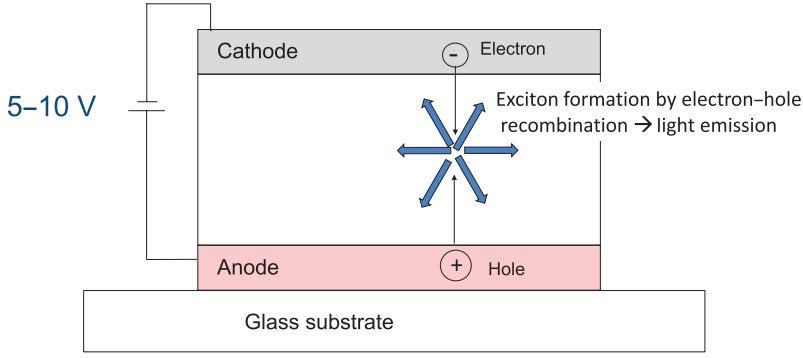

Figure 2.1 Diagram of the OLED emission mechanism.

1. Significant enhancement of recombination efficiency by layer structure using multiple different materials (heterostructure)
2. Fabrication of low-voltage, high-quality devices through evaporation
3. Appropriate choice of electron–hole injection material and of work-function for cathode/anode electrode
4. High electric field obtained by ultra-thin-film formation

OLED devices could emit very dim light before these developments, but high-luminance operation was achieved only after the first OLED paper.

As mentioned earlier, the two basic families of OLED are small-molecule and polymer (SMOLED and PLED). In SMOLED devices, a small molecule is deposited by means of the evaporation technique, so the molecular size is small (however, the mass number of a small molecule can be relatively large). On the other hand, many of the PLED materials have structures containing substructures connected together, composed of a component suitable for dissolution in a solvent and a component suitable for light emission, so the molecule is designed with a larger mass. Figure 2.2 shows

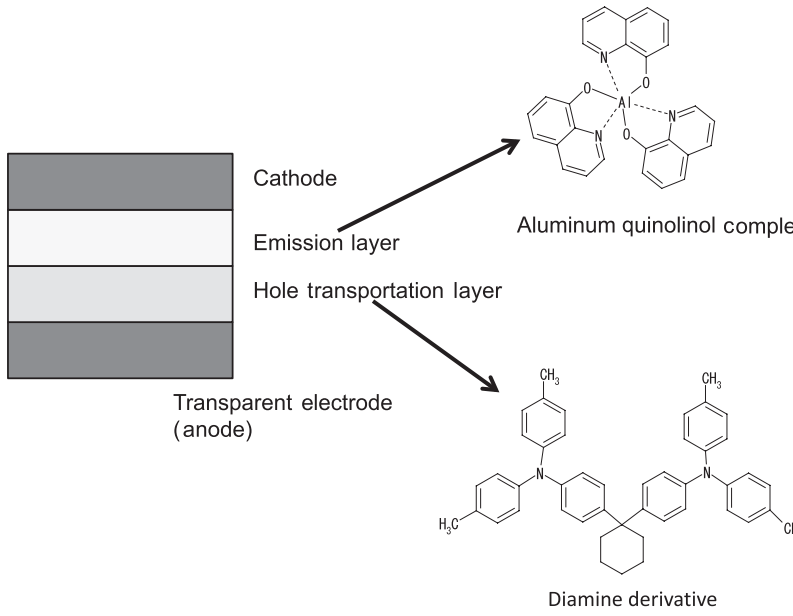

Figure 2.2 OLED device reported by Tang and Van Slyke in 1987 [5].

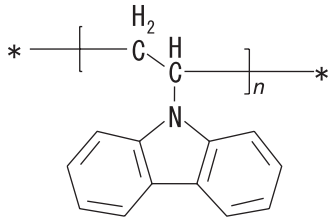

Figure 2.3 Example of a polymer OLED with polyvinylcarbazole molecular structure.

a typical example of small-molecule OLED materials: Alq_3 and diamine derivative. Figure 2.3 presents an example of a polymer OLED material: polyvinylcarbazole (PVK).

2.2.2 Light Emission Mechanism

The emission mechanism of OLED is discussed in this subsection.

2.2.2.1 Highest Occupied and Lowest Unoccupied Molecular Orbitals (HOMO and LUMO)

Of all the electron-filled orbitals, the orbital possessing the maximum electron energy is called the *highest occupied molecular orbital* (HOMO). Conversely, among all the unfilled electron orbitals, the orbital with the lowest electron energy is termed the *lowest unoccupied molecular orbital* (LUMO). The absolute values of HOMO and LUMO energies relate to the ionization potential and electron affinity (see Fig. 2.4). Ionization potential energy is the minimum energy required to extract one electron from the HOMO, and electron affinity is the energy required to add one electron to LUMO so that the system is stabilized [7].

2.2.2.2 Configuration of Two Electrons

Before considering the OLED light emission mechanism, it is important for readers to understand the electron configuration in both the ground state and the excited state.

Let us assume that two electrons, 1 and 2, are allocated in different states. Also, let us define H_1^0 and H_2^0 as Hamiltonians when electrons 1 and 2 exist independently:

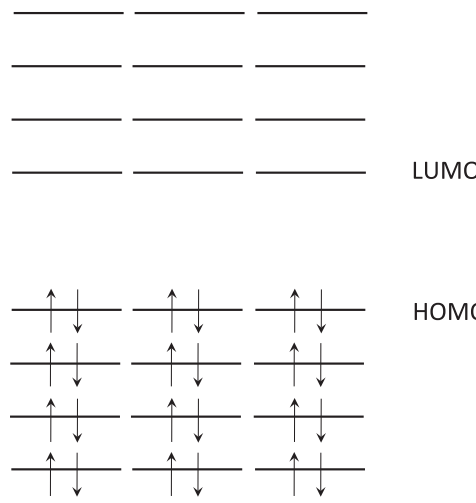

Figure 2.4 Diagrams showing orientation of highest occupied and lowest unoccupied molecular orbitals (HOMO and LUMO) of typical organic materials.

$$H_1^0 = -\frac{\hbar^2}{2\mu}\nabla_1^2 - \frac{Ze^2}{r_1} \tag{2.1}$$

$$H_2^0 = -\frac{\hbar^2}{2\mu}\nabla_2^2 - \frac{Ze^2}{r_2} \tag{2.2}$$

$$H' = \frac{e^2}{r_{12}} \tag{2.3}$$

Here, H' is perturbation due to electron repulsion.

When two electrons are allocated in different states, the Hamiltonian can be expressed as follows:

$$H = H_1^0 + H_2^0 + H' \tag{2.4}$$

If there is no perturbation, the solution of the wave equation can be expressed as follows.

$$\psi = C_1\chi_A(1)\chi_B(2) + C_2\chi_B(1)\chi_A(2) \tag{2.5}$$

Calculus of variation is useful here for solving the equation that accounts for perturbation, as follows:

$$E\psi = (E^0 + E')\psi = H\psi \tag{2.6}$$

Energy E should be the minimum in calculus variation:

$$E = \frac{\int \psi H \psi \, d\tau}{\int \psi^2 d\tau} \tag{2.7}$$

As ψ_A, ψ_B are orthogonalized and normalized with respect to each other, the equation can be expressed as

$$\begin{aligned} \int \psi H \psi \, d\tau = \iint \{C_1 \chi_A(1)\chi_B(2) + C_2 \chi_B(1)\chi_A(2)\}(H_1^0 + H_2^0 + H') \\ \cdot \{(C_1 \chi_A(1)\chi_B(2) + C_2 \chi(1)\chi_A(2)\} d\tau_1 \, d\tau_2 \end{aligned} \tag{2.8}$$

Here

$$\begin{aligned} &\iint \chi_A(1)\chi_B(2)(H_1^0 + H_2^0 + H')\chi_A(1)\chi_B(2) d\tau_1 \, d\tau_2 \\ &= \int \chi_A(1) H_1^0 \chi_A(1) d\tau_1 + \int \chi_B(2) H_2^0 \times \chi_B(2) d\tau_2 + J \\ &= E_A + E_B + J \\ &= E^0 + J \end{aligned} \tag{2.9}$$

and by applying the orthogonalization condition, we obtain

$$\begin{aligned} \int \chi_A(1)\chi_B(1) d\tau_1 = 0 \\ \int \chi_A(2)\chi_B(2) d\tau_2 = 0 \end{aligned} \tag{2.10}$$

Therefore, Eq. (2.8) can be expressed as follows:

$$\int \psi H \psi \, d\tau = (C_1^2 + C_2^2)(E^0 + J) + 2C_1 C_2 K \tag{2.11}$$

Here, J is the Coulomb integral, and K is the exchange integral, which can be expressed as follows:

$$\begin{aligned} J &= \iint \chi_A(1)\chi_B(2) H' \chi_A(1)\chi_B(2) d\tau_1 \, d\tau_2 \\ &= \iint \chi_B(1)\chi_A(2) H' \chi_B(1)\chi_A(2) d\tau_1 \, d\tau_2 \end{aligned} \tag{2.12}$$

$$K = \iint \chi_A(1)\chi_B(2) H' \chi_B(1)\chi_A(2) d\tau_1 \, d\tau_2 \tag{2.13}$$