

## Liquid Crystal Technologies for All Organic Displays

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### Abstract

*We discuss several key technologies of liquid crystal displays (LCDs) that share common features of all organic displays (AODs). An overview of the morphological effects associated with molecular ordering at interfaces on the crystallinity and the carrier mobility in organic thin film transistors (OTFTs) is given from the viewpoint of the alignment mechanism for LCDs. Recent progress of improving the carrier mobility in the OTFTs is also reviewed.*

### 1. Introduction

During the last three decades, polymeric and organic materials have extensively studied for the use as electrically active materials having the properties of conductivity and/or light-emission. Since a polymer-based transistor has been fabricated [1], organic material-based electronics have been suggested as an alternative to silicon-based integrated circuits (ICs) in applications of flexible and low-cost substrates [2]. The organic material-based ICs can be generally deposited onto a large area by simple, inexpensive, and low-temperature methods unlike conventional silicon-based technologies. In fact, the deposition over large area has significant impact on reel-to-reel manufacturing of thin, lightweight, and bendable flat panel displays [3].

Recently, all organic displays (AODs) where all the elements such as an active layer, substrates, ICs, and electrodes are made of the organic materials, have attracted much attention [2,4-6] since thin, lightweight, and bendable displays have several advantages of low-power consumption and simple fabrication processes. The technologies related to organic material-based ICs including organic thin film transistors (OTFTs) should play a key role in the area of the AODs due to the possibility of low-temperature processing of the IC fabrication. Particularly, low-

temperature processing is inevitably involved in fabricating ICs on a plastic substrate such as polyethyleneterephthalate (PET) offering a flexible medium [7].

The performance of the OTFTs has gradually improved and some OTFTs using pentacene as a semi-conducting layer now superior to that of amorphous silicon (a-Si) TFTs and comparable to that of polycrystalline silicon (p-Si) TFTs [8,9]. The improved performance of the OTFTs results mainly from the crystallinity and the interfacial ordering of the molecules in thin films [10]. From the viewpoint of the crystal growth, it is very important to minimize the role of grain boundaries and to maintain the uniform orientation of the molecules through a controlled growth mechanism. The charge carrier mobility, one of the critical factors governing the performance of the OTFTs, is highly dependent on the molecular orientation and the crystal grain size [11,12]. In order to obtain well-aligned organic films, several methods applied to liquid crystal (LC) devices, such as mechanical stretching [13], rubbing [14], Langmuir-Blodgett deposition [15], and liquid crystalline self-organization [16], have been recently proposed.

In this paper, we describe key technologies of LC displays sharing common features of AODs and discuss how the carrier mobility in the organic material can be improved for practical applications in view of the crystallinity and the molecular ordering. In section 2, a brief description of the OTFT and the carrier mobility is given. Section 3 deals with the morphological effects and the crystallinity on the performances of the OTFT. The essential features of surface interactions are discussed to how one can control the grain size and the anisotropic ordering of the molecules on various treated substrates. We then describe the relationship between the molecular ordering and carrier mobility in several different organic films in section 4. Some concluding remarks are made in the remaining section.

## 2. Organic Thin Film Transistor (OTFT)

Figure 1 shows a schematic diagram of the cross sectional view of an OTFT which is identical to a conventional a-Si TFT except for using organic materials. This structure is one of typical structures for low conductive materials and is widely used in flat panel displays [17]. As shown in Fig. 1, the source and the drain electrodes form ohmic contacts directly to the conducting channel (active layer) and there is no depletion region to isolate the device from the substrate. Thus, low off currents are only guaranteed by the low conductivity of the active layer. Using the pentacene as the active layer, it is often desirable to pattern the organic active layer using polyvinyl alcohol (PVA) to reduce current leakage and crosstalk between transistors.

In organic-based electronic devices, the electrical properties are governed by the electronic states of the semi-conducting molecules and the morphology of the thin film structure [10]. In general, it is believed that charge transport in organic materials occurs by hopping of charges between localized electronic states through  $\pi$ - $\pi$  interactions between neighboring molecules. This is different from the conventional inorganic semiconductor case, i.e., the band-like transport in delocalized electronic states. In conjugated organic materials behaving as a class of semiconductors, localization of electronic states occurs via the formation of polarons. Under the action of the charge, the deformation of the conjugated chain forms a polaron, meaning that the charge is self-trapped by the deformation of the conjugated chains. As a consequence, the transport of the polaron formed by self-trapping produces the charge transport in the organic materials [18,19].

Recently, most of the OTFTs with the high mobility and good on/off ratio have been fabricated using  $\alpha$ -conjugated oligothiophenes ( $\alpha$ - $n$ T, where usually  $3 \leq n \leq 8$ , especially  $\alpha$ -6T) [20,21], pentacene [8,9,22],

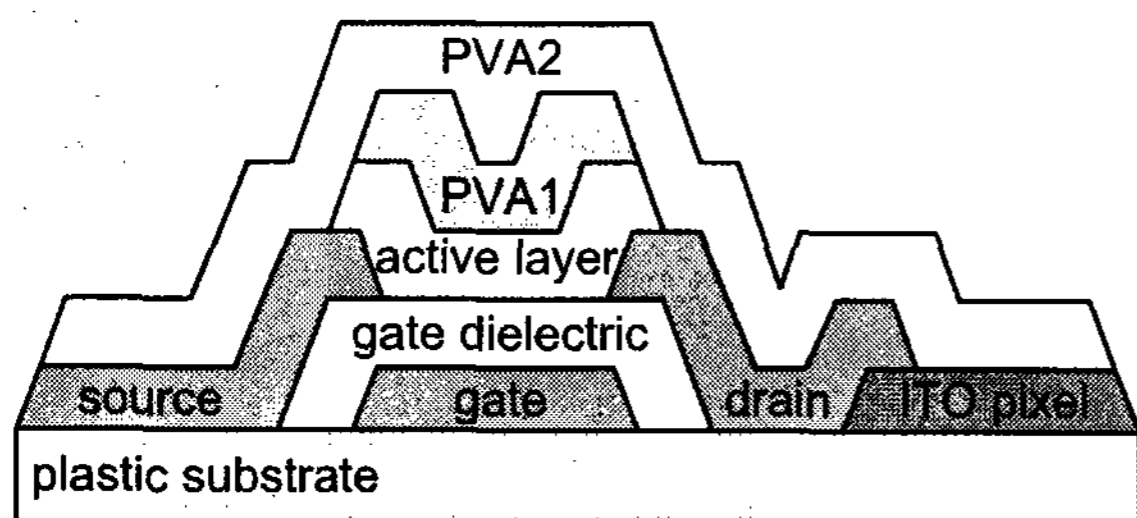


Fig. 1. Schematic cross section of OTFT

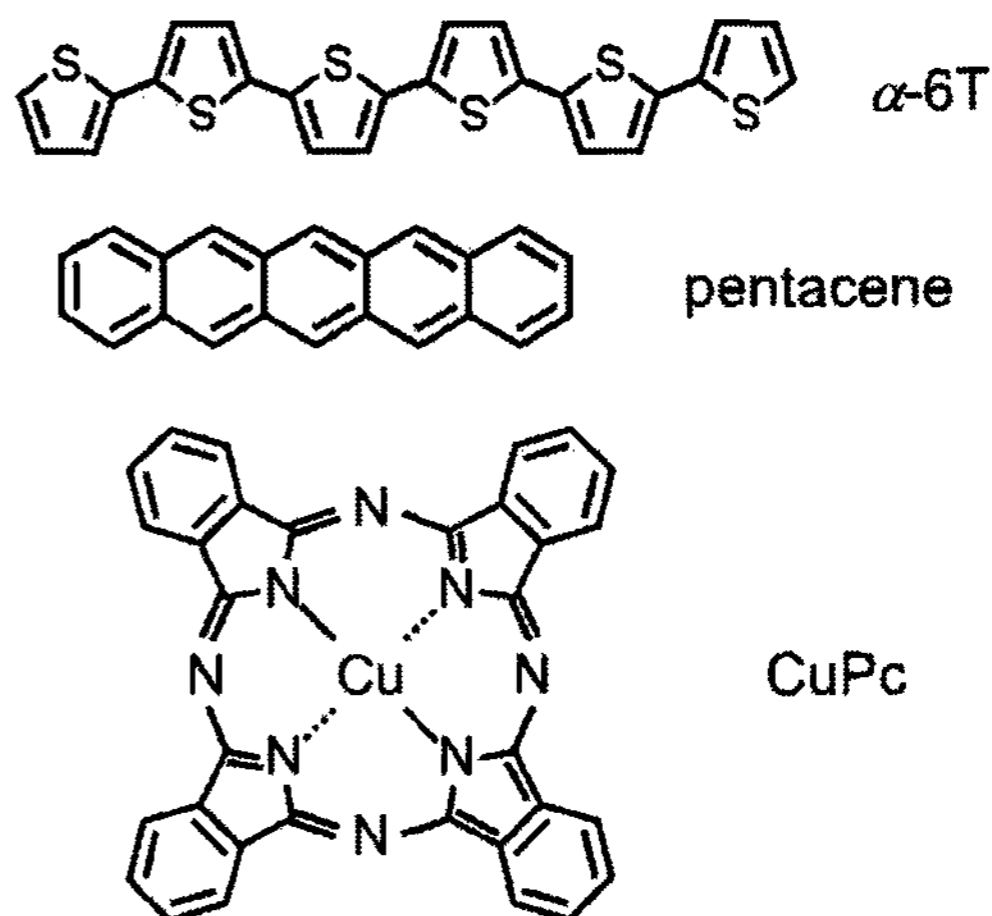


Fig. 2. Chemical structures of the organic semi-conducting molecules.

and copper phthalocyanine (CuPc) [12]. The molecules exhibit the semi-conducting behavior with strong anisotropy of the field-effect mobility. The chemical structures of some organic semi-conducting molecules are illustrated in Fig. 2.

It has been reported that some OTFTs possess the mobility exceeding  $1 \text{ cm}^2/\text{Vs}$  and the on/off ratio of  $10^6$  similar to a-Si TFTs [9,23]. However, a complete picture of the transport phenomenon in a single transistor device is not presented yet. Several models using a potential well [24], grain-boundary barriers [25,26], and trapping at localized gap states [23,27] have been proposed to describe the dependence of the field-effect mobility as a function of the gate voltage and temperature.

In order to fabricate fully patterned all organic TFTs, the conducting polymers for the gate, the source, and drain and the insulating polymer for the gate dielectric layer have been extensively studied [2,5,28,29]. For example, poly-3,4-ethylenedioxythiophene doped with polystyrene sulfonic acid (PEDOT:PSS) was used for the conducting polymer and poly-4-vinylphenol (PVP) for the gate dielectric layer [28,29]. Plastic materials such as polyethersulfone (PES), polyethyleneterephthalate, polycarbonate, and polyimide were used as flexible substrates [2,7,30].

## 3. Morphological Effects and Crystallinity

The charge carrier mobility is highly dependent on the molecular orientation and surface morphology, which in turn depend on the chemical structures of the

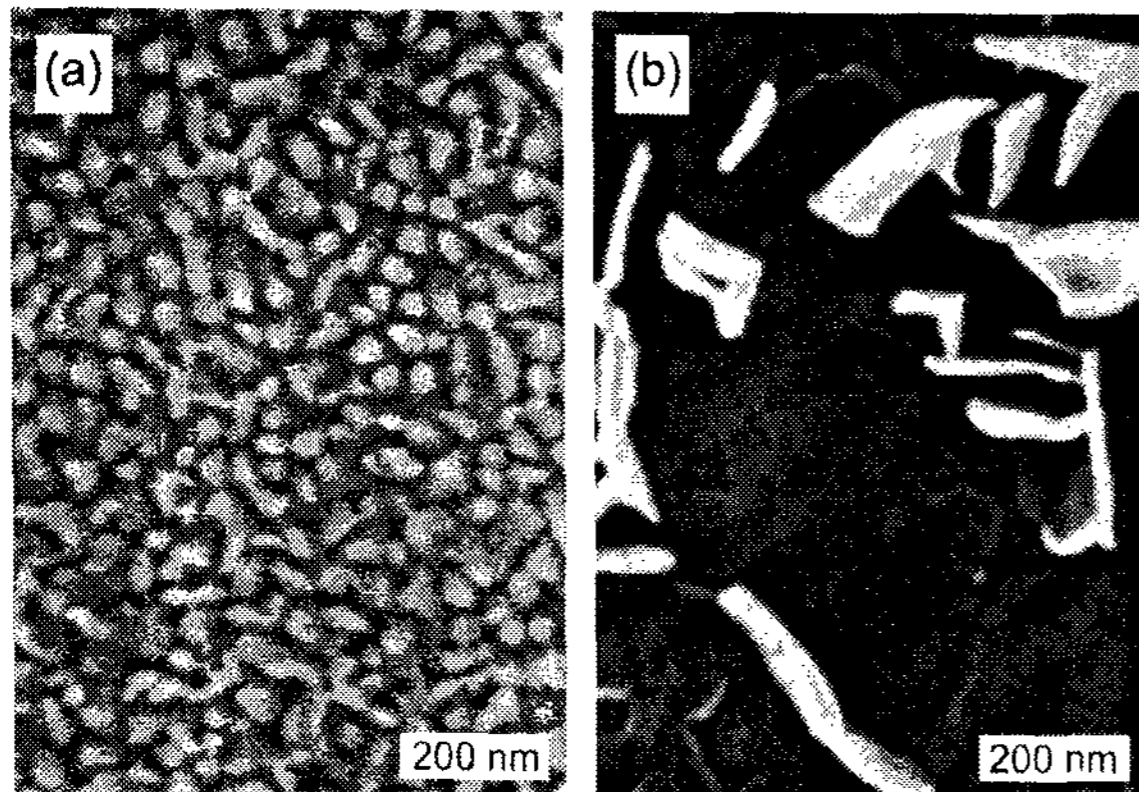


Fig. 3. SEM images of 8T films deposited on glass at (a) room temperature and (b) 175 °C [31].

constituent molecules and the deposition parameters such as the substrate temperature during deposition and the deposition rate [11,12]. In general, organic semiconductors including pentacene and  $\alpha$ -6T are strong molecular crystal formers that are believed to be very important to produce large field effect mobility [11,32]. The interface between the organic active layer and the gate dielectric layer such as silicon dioxide ( $\text{SiO}_2$ ) will play a critical role on the characteristics of the field-effect devices.

Figure 3 shows microscopic textures of scanning electron microscope (SEM) of octithiophene (8T) thin films deposited on a glass at various temperatures [31]. As shown in Fig. 3(a), the film deposited at room temperature consists of very small and homogeneous grains of the average diameter of about 60 nm. Increasing the substrate temperature, the grains remarkably increase in size and mostly lie flat on the surface. Note that the mobility increases with the grain size. It was reported that similar results were obtained in pentacene films grown on crystalline silicon substrates [33]. A progressive increase in the pentacene grain size on  $\text{SiO}_2$  was achieved by deposition at higher substrate temperatures, higher deposition pressures, and lower growth rates. Such deposition conditions favor the surface diffusion and growth on a few pentacene nucleation sites as shown in Fig. 4 [34].

Recently, it was found that the characteristics of pentacene TFTs were improved by using a self-assembled material like octadecyltrichlorosilane (OTS) as a buffer layer between the  $\text{SiO}_2$  gate dielectric and the pentacene active layer [27]. The SEM images of pentacene films grown on  $\text{SiO}_2$  and

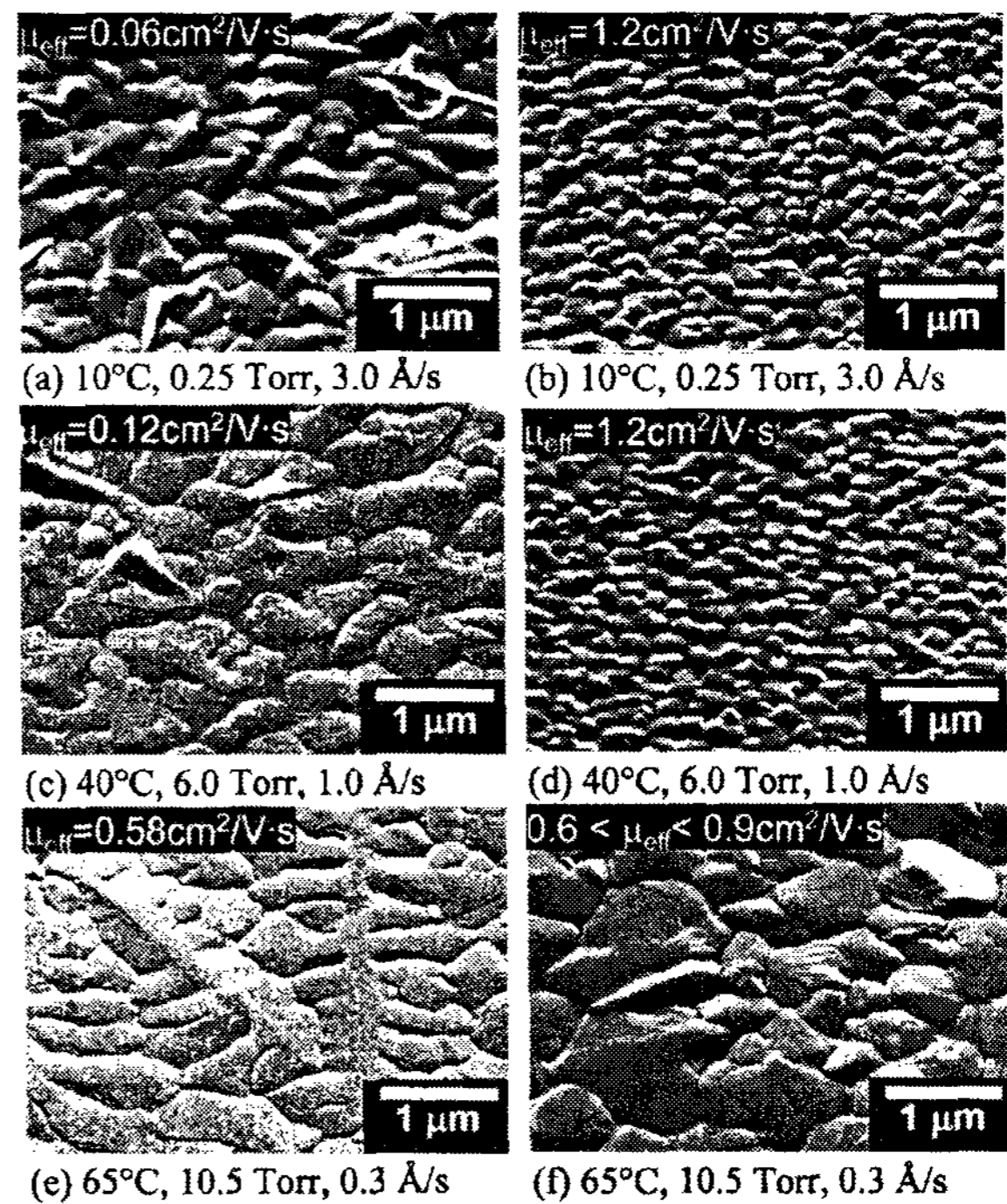


Fig. 4. SEM images of pentacene films deposited on  $\text{SiO}_2$  (left column) and on OTS-treated  $\text{SiO}_2$  (right column). Substrate temperature, deposition pressure, deposition rate, and field-effect hole mobility  $\mu_{\text{eff}}$  for the sample are given [34].

OTS-treated  $\text{SiO}_2$  are shown in Fig. 4 [34]. Here, the grain size on the OTS-treated  $\text{SiO}_2$  was remarkably smaller than the untreated  $\text{SiO}_2$  with improved contact between individual grains, and independent of the substrate temperature in the range 10-40 °C. This is possibly caused by the greater adhesion of pentacene to the OTS monolayer, as compared with that of pentacene onto the untreated  $\text{SiO}_2$ . As shown in Fig. 4(f), however, pentacene films deposited at high substrate temperature exhibit fractures and voids along grain boundaries due to the built-in strain and thermal contraction at room temperature after growth. The observed fractures and voids will significantly degrade the carrier mobility. Therefore, a stacked structure with a pentacene layer doubly deposited at high and low temperatures was proposed [11]. In this stacked structure, the first pentacene layer was deposited at high temperature to obtain high molecular ordering, and the second layer was deposited at low temperature to help fill-in voids and improve film continuity.

For higher molecular ordering of pentacene, thermal annealing was employed at high temperature [8]. It

was experimentally verified by the X-ray diffraction (XRD). However, more ordered structure was observed through the XRD patterns and atomic force microscopic textures but the field-effect mobility was found to decrease [35]. This seems quite intriguing and remains to be further explored.

#### 4. Molecular Ordering and Mobility

Since the charge transport in organic materials is attributed to the  $\pi$ - $\pi$  interactions between molecules through the hopping mechanism, the charges propagate preferentially along the stacking axis of molecules through their overlapping  $\pi$ -orbitals. Hence, highly aligned rod-like and disk-like molecules improve the mobility in the OTFTs. Considering that pentacene and oligothiophenes are rigid rod-like molecules similar to the core structure of the LC, it is expected that similar surface induced order can also be produced on pentacene and oligothiophenes. In order to align such rod-like and disk-like molecules, several alignment techniques employed in LC devices would be useful [13-16].

The highly anisotropic mobility was obtained in a bilayered film of friction-transferred polytetrafluoroethylene (PTEE) and regioregular poly-3-hexylthiophene (P3HT) on a  $\text{SiO}_2$  substrate [36]. However, since wetting problems at the inhomogeneous friction transferred PTEE surface, deposition of P3HT will not be uniform. Therefore, such films will yield poor device performances and non-uniform device characteristics [36].

In photovoltaic sandwich cells, the 8T film, horizontally aligned by the rubbing process, improves both light absorption and charge transport [37]. Note that 8T molecules are vertically aligned on an indium-tin-oxide glass with fast deposition at room temperature. From the absorption spectra of the rubbed 8T film under light polarized parallel and perpendicular to the rubbing direction, the absorption peak corresponding to the  $\pi$ - $\pi^*$  bonding in the 8T molecules becomes highly dichroic. In pentacene-based OTFTs on rubbed PVA and polyimide (PI) alignment layers such as Nissan SE150 and SE12710, large linear dichroism was observed [38].

Using photo-aligning techniques exposing linearly polarized ultra-violet (UV) light to photopolymers such as PVP, (fluoro- or nitro-) cinnamic acids and Nissan RN1332, the field-effect mobility of the OTFT was remarkably improved [38,39]. The mobility of the OTFT deposited on the UV treated PVP gradually

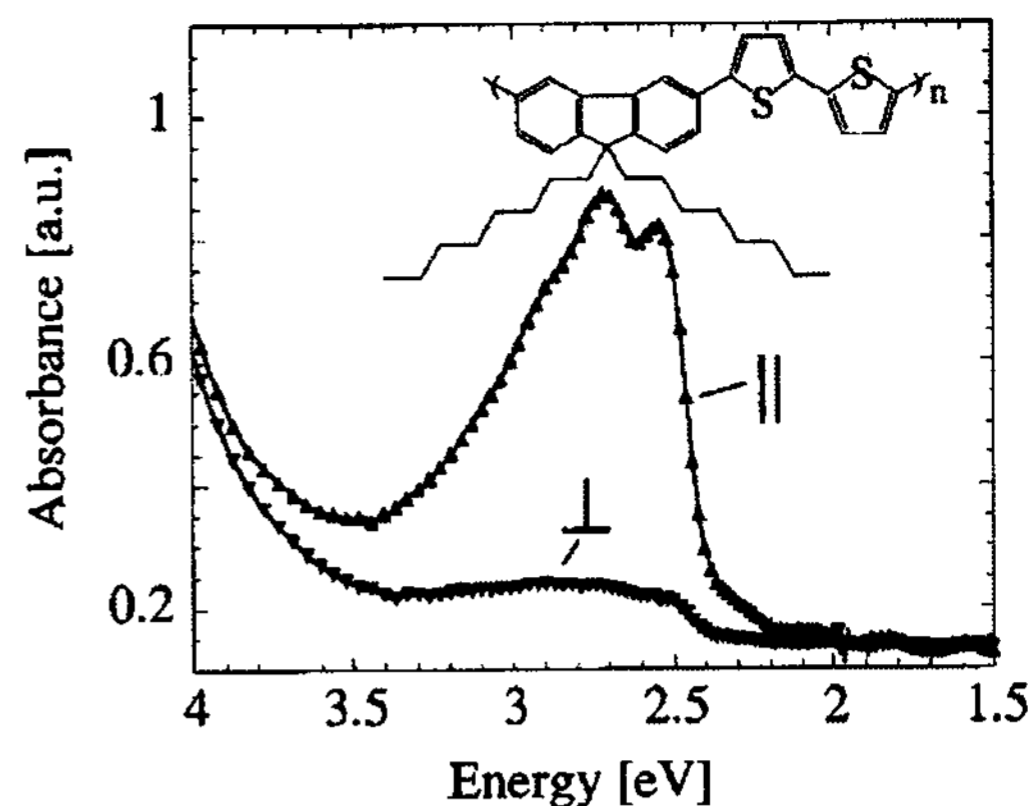


Fig. 5. Polarized optical absorption spectra of F8T2 TFT substrate. The incident light is polarized parallel (||) and perpendicular ( $\perp$ ) to the rubbing direction [40].

increases with increasing the exposure time of the UV light up to a certain threshold. Above the threshold, the mobility rapidly decreases due to the degradation of the PVP layer by the UV light [39].

More recently, the enhancement of the mobility in a semi-conducting polymer with the nematic LC phase has been reported [40]. Sirringhaus *et al.* [40] used poly-9,9' dioctylfluorencecobithiophene (F8T2) as the active material in the polymer TFT. The F8T2 molecules exhibit the nematic LC phase above 265 °C and are oriented to form a mono-domain on a mechanically rubbed PI alignment layer. The degree of alignment was determined by polarized optical absorption spectra as shown in Fig. 5. In the spectral region of the  $\pi$ - $\pi^*$  transition in the F8T2 molecules, the dichroic ratio of the absorption coefficients for the incident light polarized parallel (perpendicular) to the rubbing direction is 5-12 depending on the film thickness and details of the annealing and rubbing procedures. Here, the oscillations at energies below the fundamental  $\pi$ - $\pi^*$  absorption are caused by the interference effects in the gate insulating layer. Fig. 6 shows current-voltage (I-V) characteristics and the temperature dependence of the mobility of the aligned F8T2 TFTs with active channels parallel and perpendicular to the rubbing direction. The relevant dichroic ratio is always of the same order of the mobility-anisotropy. The isotropic device is prepared on the substrate with no alignment layer, and typically exhibits a certain value of the mobility between  $\mu_{\perp}$  and  $\mu_{\parallel}$  (see, the closed circles in Fig. 6(a)).

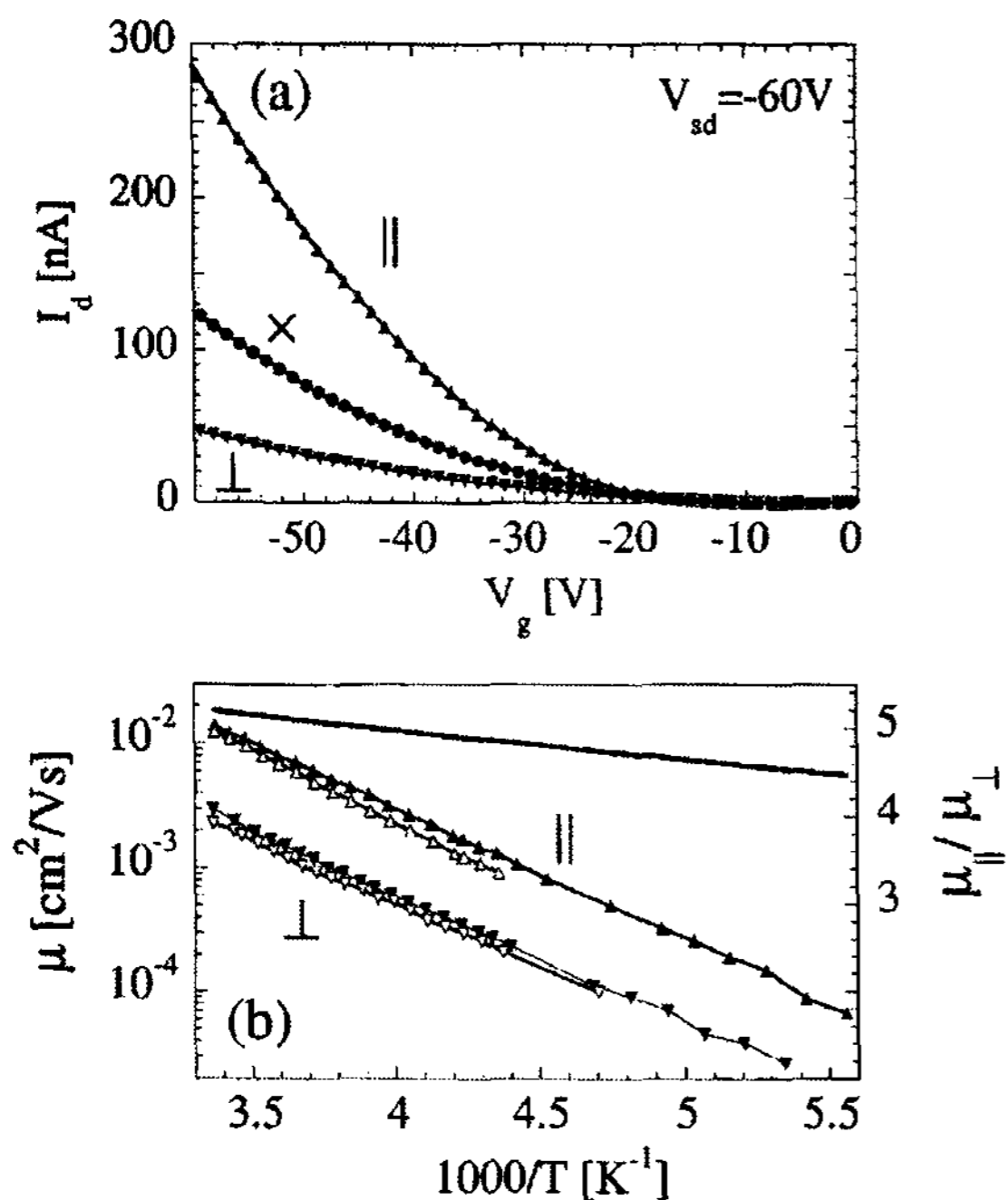


Fig. 6. (a) Saturated I-V characteristics and (b) temperature dependence of the linear (open symbols) and saturated (closed symbols) mobility of aligned F8T2 TFTs with active channels parallel (||) and perpendicular ( $\perp$ ) to the rubbing direction. The I-V characteristics ( $\times$ ) in an isotropic device without unidirectional alignment and the mobility-anisotropy  $\mu_{||}/\mu_{\perp}$  (right axis) are also shown in (a) and (b), respectively [40].

### 5. Concluding Remarks

After the pentacene film was first used as a semi-conducting layer, the performances of the OTFTs have been gradually improved. In general, the mobility is highly dependent on the molecular orientation, the degree of the crystallinity, and the grain size. Various efforts have been made to obtain highly ordered organic films using the alignment techniques employed in LC devices. For example, the surface order was produced using self-assembled monolayers (SAMs) and polymers that were applicable for aligning the LCs. The uniformly aligned LCs are obtained on rubbed or photo-irradiated polymer layers as well as SAMs. Especially, considering that pentacene is a rigid, rod-like molecule similar to the core structure of the LC, it is expected that similar surface order of pentacene can be induced on a treated substrate. In addition, since the electrical properties of

a multi-layered structure would be degraded by the contact resistance, the electrical contacts to organic semiconductors have been extensively studied in the context of organic light emission diodes [41,42]. The same principles should be applied to the OTFTs.

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## 18.2 / Plenary

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