

# Pentacene Organic Thin-Film Transistors—Molecular Ordering and Mobility

D. J. Gundlach, Y. Y. Lin, T. N. Jackson, S. F. Nelson, and D. G. Schlom

**Abstract**— Pentacene-based organic thin-film transistors (TFT's) with field-effect mobility as large as  $0.7 \text{ cm}^2/\text{V}\cdot\text{s}$  and on/off current ratio larger than  $10^8$  have been fabricated. Pentacene films deposited by evaporation at elevated temperature at low-to-moderate deposition rates have a high degree of molecular ordering with micrometer-sized and larger dendritic grains. Such films yield TFT's with large mobility. Films deposited at low temperature or by flash evaporation have small grains and poor molecular ordering and yield TFT's with low mobility.

## I. INTRODUCTION

ORGANIC thin-film transistors (TFT's) are of interest for use in broad area electronic applications. In active matrix liquid crystal displays (AMLCD's), for example, organic TFT's would allow the use of inexpensive, lightweight, flexible, and mechanically rugged plastic substrates as an alternative to the glass substrates needed for commonly used hydrogenated amorphous silicon (a-Si:H) devices. To date, the most extensively studied organic semiconducting material has been  $\alpha$ -sexithienyl [1]–[3].  $\alpha$ -sexithienyl based TFT's have a typical field-effect mobility of  $0.03 \text{ cm}^2/\text{V}\cdot\text{s}$  [4] and a typical on/off current ratio of  $10^6$  [5]. These values, while encouraging for organic materials, are marginal at best for use in AMLCD's which currently use hydrogenated amorphous silicon with field-effect mobility greater than  $0.5 \text{ cm}^2/\text{V}\cdot\text{s}$  and on/off current ratio greater than  $10^8$ . Recently, we have fabricated organic TFT's using pentacene, a fused-ring polycyclic aromatic hydrocarbon, as the active material [6]. These new devices have field-effect mobility greater than  $0.6 \text{ cm}^2/\text{V}\cdot\text{s}$  and on/off current ratio greater than  $10^8$ ; both results are comparable to hydrogenated amorphous silicon TFT's and are records for organic devices. In this letter we correlate the high mobilities observed to strong molecular ordering in pentacene films.

## II. FIELD EFFECT-MOBILITY

The pentacene TFT's reported here use a staggered-inverted structure. For convenience, the gate consists of a heavily-doped silicon wafer (which is also used as the substrate) on which a 400-nm thick thermal oxide is grown for use as the

gate dielectric. The pentacene active layer is deposited by thermal evaporation at low to moderate deposition rate ( $1\text{--}10 \text{ \AA/s}$ ) onto substrates held at room or elevated temperature after material purification by vacuum gradient sublimation [7]. The devices are completed by evaporating gold through a shadow mask to form source and drain contacts and aluminum onto the wafer back side to contact the gate.

Fig. 1(a) shows the linear  $I_D\text{--}V_{DS}$  characteristics and Fig. 1(b) shows the  $\sqrt{I_D}\text{--}V_{GS}$  and  $\log(I_D)\text{--}V_{GS}$  characteristics for a typical pentacene TFT. This device has a channel length and width of  $20 \text{ }\mu\text{m}$  and  $220 \text{ }\mu\text{m}$ , respectively, and a gate dielectric thickness of  $400 \text{ nm}$ . From the  $\sqrt{I_D}\text{--}V_{GS}$  characteristics, we extract a field-effect mobility of  $0.62 \text{ cm}^2/\text{V}\cdot\text{s}$  in the saturation region for  $V_{DS} = -80 \text{ V}$ . We note also that the pentacene TFT is a well-behaved square-law device and the fitting line used for mobility extraction fits the data over a wide range of gate voltage. The  $\log(I_D)\text{--}V_{GS}$  characteristics show that the on/off current ratio for this device is larger than  $10^8$  even at  $V_{DS} = -100 \text{ V}$ . Both the mobility and on/off current ratio for these pentacene TFT's are comparable to results typically obtained from hydrogenated amorphous silicon devices.

It is notable that this large on/off current ratio is obtained even at a drain-source bias of  $-100 \text{ V}$ . At lower drain-source bias the on/off current ratio is even larger and a large ratio can be obtained with a smaller gate voltage swing. The characteristics shown in Fig. 1 also indicate that this device has a positive threshold voltage and a positive gate voltage is required to turn the device fully off. We find that neither the threshold voltage nor the zero gate bias conductivity scale with active layer thickness. Thus, the positive threshold voltage does not arise from depletion of carriers in the bulk but rather from an initial accumulation layer at the pentacene/dielectric interface.

All measurements reported here were made in normal room air with no precautions taken to prevent degradation of the pentacene film. Although pentacene is a fairly reactive material and oxidation in air might be expected, for our TFT's we see only small changes in device characteristics for time periods of days and weeks. We have not yet investigated long-term stability.

Interestingly, previous reports of pentacene based TFT's exist, but with much lower field-effect mobility [8]–[11]. TFT's fabricated using a solution-cast pentacene precursor had field-effect mobility near  $0.009 \text{ cm}^2/\text{V}\cdot\text{s}$  [8], [9], while previously reported TFT's fabricated using evaporated pentacene had a field-effect mobility near  $0.002$  [10] and  $0.03$  [11]  $\text{cm}^2/\text{V}\cdot\text{s}$ . We believe the most significant factor in obtaining the much higher mobility reported here is improved quality of the deposited pentacene layer. We expect that carrier transport in field-

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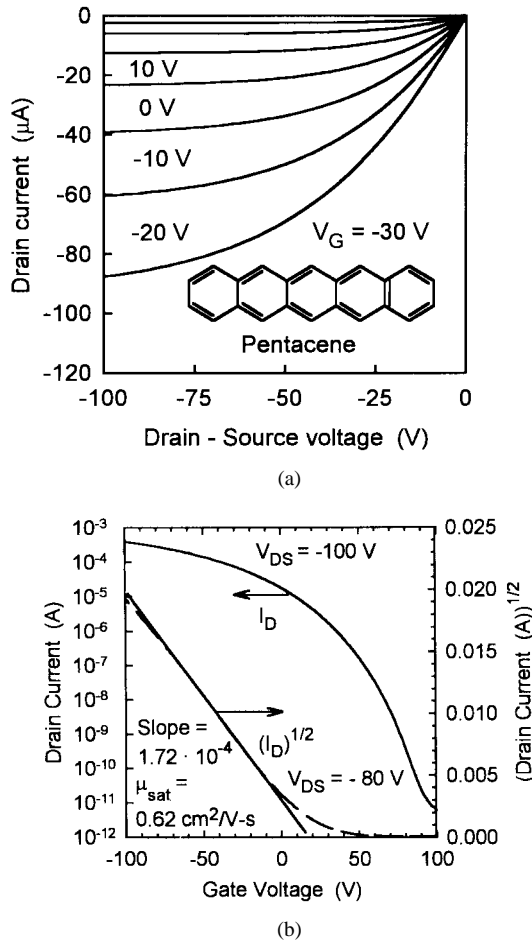


Fig. 1 (a)  $I_D$  versus  $V_{DS}$  characteristics for a typical pentacene TFT with gate length and width of 20  $\mu\text{m}$  and 220  $\mu\text{m}$ , respectively, and 400-nm gate dielectric thickness. The pentacene active layer has a average thickness of 50 nm. (b)  $I_D^{1/2}$  versus  $V_G$  and  $\text{Log}(I_D)$  versus  $V_G$  characteristics for the pentacene TFT of (a). The TFT field-effect mobility in the saturation region is greater than 0.62  $\text{cm}^2/\text{V}\cdot\text{s}$  with  $V_{DS}$  biased at  $-80$  V. The TFT on/off current ratio is greater than  $10^8$  even with  $V_{DS}$  biased as high as  $-100$  V.

induced channels in pentacene (and perhaps in most similar organic semiconductor systems) is dominated by the difficulty of moving carriers from one molecule to the next. Thus, a high degree of molecular ordering in the active organic material may be necessary for high-mobility devices, requiring careful control of film deposition conditions. For the TFT's reported above we used a substrate temperature of about 120  $^\circ\text{C}$  and a deposition rate of 2–3  $\text{\AA}/\text{s}$ .

### III. FILM GROWTH AND STRUCTURE

To test the effect of molecular ordering on TFT performance, we fabricated TFT's using material deposited by flash evaporation and also using material deposited onto substrates held at low temperature. Flash evaporation is often used for organic semiconductor depositions to avoid problems with thermal decomposition of the material being evaporated. However, using flash evaporation onto substrates held at temperatures between 20 and 160  $^\circ\text{C}$ , only very low mobility TFT's were obtained. For example, a flash evaporated film onto a substrate held at 160  $^\circ\text{C}$  gave TFT's with field-effect mobility near 0.0001  $\text{cm}^2/\text{V}\cdot\text{s}$  (again extracted using fits to well-behaved  $\sqrt{I_D}-V_{GS}$  characteristics). Further, using

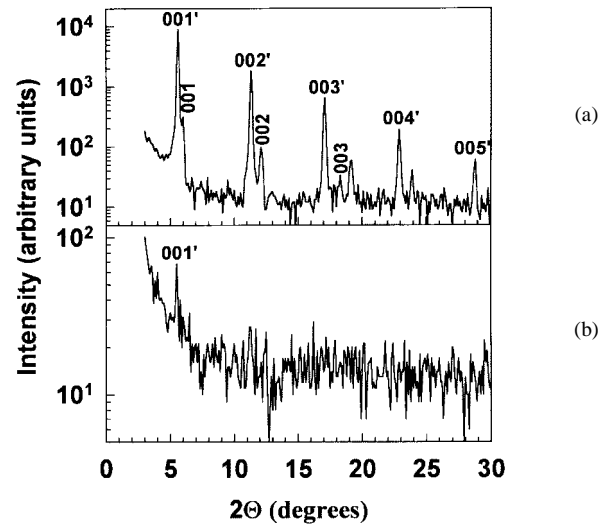


Fig. 2 (a) 4-circle X-ray diffraction pattern for a pentacene film with an average thickness of 150 nm deposited on  $\text{SiO}_2$  held at 60  $^\circ\text{C}$ . The diffracting planes are parallel to the surface of the substrate. (b) 4-circle X-ray diffraction pattern for a flash evaporated pentacene film with an average thickness of 84.5 nm deposited on  $\text{SiO}_2$  held at 135  $^\circ\text{C}$ . Again, the diffracting planes are parallel to the surface of the substrate.

rapid thermal annealing to rapidly heat the fabricated TFT's to 350  $^\circ\text{C}$  for a few seconds increased the TFT field-effect mobility to about 0.002  $\text{cm}^2/\text{V}\cdot\text{s}$ . We believe the initial poor mobility is due to poor molecular ordering in the flash evaporated film, and the mobility improvement comes from an improvement in ordering with the anneal. Similarly, pentacene films deposited at 4  $\text{\AA}/\text{s}$  onto substrates held at  $-20$   $^\circ\text{C}$  gave TFT's with relatively poor field-effect mobility, typically near 0.07  $\text{cm}^2/\text{V}\cdot\text{s}$  (extracted using fits to well-behaved  $\sqrt{I_D}-V_{GS}$  characteristics). Again, we believe the poor mobility is due to poor molecular ordering.

To further investigate the effect of molecular order in pentacene films on field-effect mobility, 4-circle X-ray diffraction (Cu  $K_\alpha$  radiation) was used to evaluate pentacene films deposited on oxidized silicon wafers for different substrate temperatures and deposition rates. Fig. 2(a) shows X-ray diffraction results for a pentacene film with 150 nm average thickness deposited at a rate of 2–3  $\text{\AA}/\text{s}$  onto  $\text{SiO}_2$  held at 60  $^\circ\text{C}$ . Single crystal pentacene is triclinic with unit cell parameters  $a = 7.90$   $\text{\AA}$ ,  $b = 6.06$   $\text{\AA}$ ,  $c = 16.01$   $\text{\AA}$ ,  $\alpha = 101.9^\circ$ ,  $\beta = 112.6^\circ$ , and  $\gamma = 85.8^\circ$  [12]. From these parameters the 001 plane spacing of single crystal pentacene is calculated to be 14.49  $\text{\AA}$ . The 00 $l$  peaks at positions expected for single crystal pentacene are labeled in Fig. 2(a). A family of peaks corresponding to a  $d = 15.5$   $\text{\AA}$  is also seen. Peaks corresponding to this  $d/n$  spacing are labeled for  $n = 1-5$  [00 $l'$  in Fig. 2(a)]. This spacing is longer than expected for the reported structure of single crystal pentacene. The nature of the distortion or rearrangement of the pentacene responsible for the elongated spacing is not known, however, this distortion or rearrangement has been reported elsewhere [11], [13]. These results indicate pentacene films deposited onto  $\text{SiO}_2$  at elevated temperature are highly ordered, with the long axis of the pentacene molecule perpendicular to the substrate interface.

Fig. 3(a) shows an atomic force microscopy (AFM) image for a similar, but thinner, pentacene film (30 nm average film

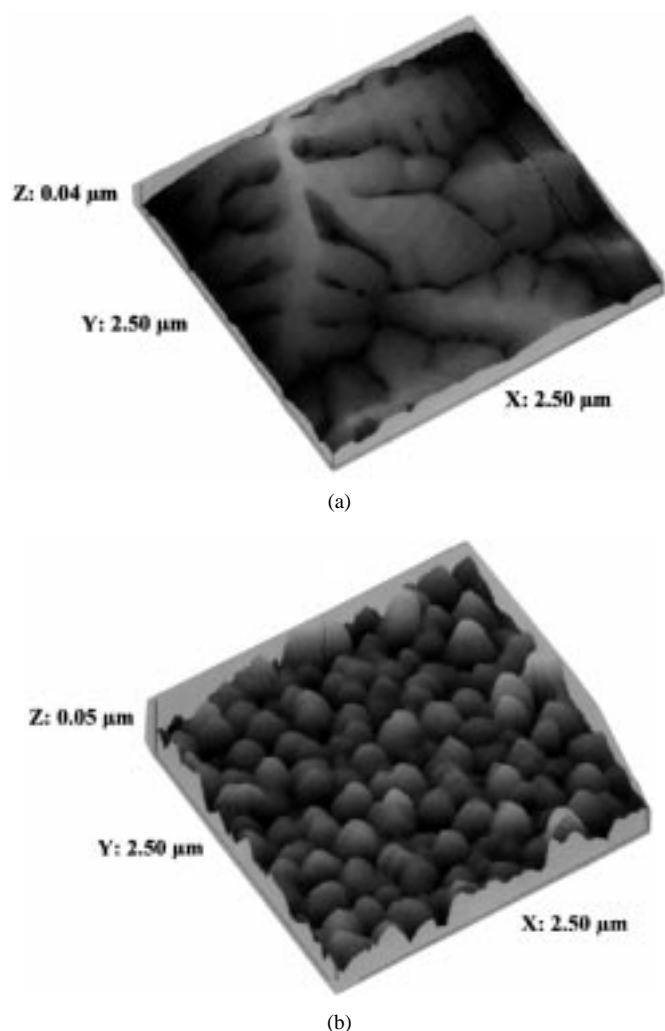


Fig. 3 (a) A  $2.5 \times 2.5 \mu\text{m}^2$  AFM image of a pentacene film with an average thickness of 30 nm deposited onto oxidized silicon held at 60 °C. (b) A  $2.5 \times 2.5 \mu\text{m}^2$  AFM image of a flash evaporated pentacene with average film thickness of 84.5 nm deposited onto oxidized silicon held at 135 °C.

thickness). The image confirms that the film is highly ordered, and reveals large (several micrometers in size) dendritic grains. The grain surface is terraced and AFM measurements indicate that the typical terrace is one pentacene molecule in height (that is, about 15 Å). It is notable that the grain structure extends over a distance of several micrometers even though the film is deposited on an amorphous substrate. There is also some indication that coordination may exist between grains so that an ordered structure may extend over even larger distances. These results are consistent with the high mobilities typically seen for TFT's fabricated using similar films. Ordered films are obtained for depositions at low to moderate rates (1–5 Å/s) onto substrates held at temperatures from 20 °C to 120 °C. Relatively high mobility devices are obtained for depositions over this entire range: typically  $>0.3 \text{ cm}^2/\text{V}\cdot\text{s}$ , the maximum we have observed thus far is  $0.7 \text{ cm}^2/\text{V}\cdot\text{s}$ .

Fig. 2(b) shows an X-ray diffraction pattern for an 84.5 nm pentacene film flash evaporated onto a substrate held at 135 °C. Only one very weak peak with a measured  $d$  spacing of  $16 \pm 0.4$  Å is observed which roughly corresponds to the expected  $d$  spacing of 14.49 Å. Compared to the deposition at lower rate, this film shows very poor molecular ordering.

Fig. 3(b) shows an AFM image for a similar film. The large dendritic grain structure observed for the pentacene film deposited onto a substrate at elevated temperature is absent and only small irregular grains are observed. These results are consistent with the low mobilities obtained for TFT's fabricated using flash evaporated films. Results for pentacene films deposited onto cooled substrates (–20 °C) are similar and we believe the absence of molecular ordering severely impacts and limits carrier transport in these films.

#### IV. CONCLUSION

The results presented above show a strong correlation between molecular ordering and pentacene TFT mobility. Despite the need for well-ordered films for large field-effect mobility, the range of acceptable deposition conditions for high mobility pentacene layers is quite large. Field-effect mobility larger than  $0.3 \text{ cm}^2/\text{V}\cdot\text{s}$  is obtained for depositions onto substrates held at temperatures between 20 and 130 °C and with deposition rates between 2 and 8 Å/s. For depositions in this range onto  $\text{SiO}_2$ , well-ordered films with large dendritic grains (often micrometer sized or larger) are observed. Maximum mobilities are obtained with substrate temperatures between 50 and 120 °C and with deposition rate between 1 and 3 Å/s; field-effect mobility is typically greater than  $0.4 \text{ cm}^2/\text{V}\cdot\text{s}$  in this range, and mobilities as large as  $0.7 \text{ cm}^2/\text{V}\cdot\text{s}$  have been obtained.

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