Pentacene-Based Organic Thin-film Transistors

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Abstract—Organic thin-film transistors using the fused-ring polycyclic aromatic hydrocarbon pentacene as the active electronic material have shown mobility as large as 0.7 cm²/V-s and on/off current ratio larger than 10⁸; both values are comparable to hydrogenated amorphous silicon devices. On the other hand, these and most other organic TFT's have an undesirably large subthreshold slope. We show here that the large subthreshold slope typically observed is not an intrinsic property of the organic semiconducting material and that devices with subthreshold slope similar to amorphous silicon devices are possible.

I. Introduction

RGANIC electronic devices have made very rapid progress in the last several years. With field-effect mobility and current on/off ratio values comparable to amorphous silicon [1], it becomes increasingly likely that organic electronic devices will find use in broad area electronics applications. Displays are one particularly attractive application for organic devices. In active matrix liquid crystal displays (AMLCD's), for example, the use of organic thin-film transistors (TFT's) would allow polymeric substrates to replace the glass currently used for displays built using hydrogenated amorphous silicon (a-Si:H) devices. Polymeric substrates have important advantages in weight and ruggedness over conventional glass substrates. In addition, polymeric substrates would allow flexible form-factor displays and might also allow continuous web processing with dramatically lowered manufacturing cost.

Organic materials have been of interest for conductive and semiconductor applications for more than forty years. Since materials that allow bipolar conduction are not common, work on transistor structures has focused on field-effect devices, usually in thin film form. However, until fairly recently organic TFT field-effect mobilities have been disappointingly low, typically in the 10^{-5} – 10^{-6} cm²/V-s range, and far too low for most applications.

The playing field for organic TFT's changed significantly when research focused on small molecule organic semiconductors. This change of focus, pioneered by Garnier [2] and initially concentrating on α -sexithienyl as the active material, gave dramatically improved field-effect mobility, typically in the 10^{-2} – 10^{-1} cm²/V-s range. A possible explanation for the

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improved mobility is that materials like α -sexithienyl have a strong tendency to form molecular crystals and this allows improved intermolecular carrier transport.

Despite the improvements, the best reported mobilities for α -sexithienyl TFT's have been poor compared to a-Si:H devices and not adequate for wide application. For example, α -sexithienyl based TFT's have a typical field-effect mobility of 0.03 cm²/V-s [3] which, while encouraging for organic materials, is marginal at best for use in AMLCD's which currently use hydrogenated amorphous silicon with field-effect mobility greater than 0.5 cm²/V-s. Recently, Torsi *et al.* [4], [5] have presented a theoretical treatment of experimental measurements on α -sexithienyl organic TFT's using a formalism originally due to Holstein [6] and conclude that mobility may be limited to about 0.1 cm²/V-s in organic materials where electron-phonon interaction is strong.

It is useful to ask what mobilities should be possible in organic materials. Although the largest field-effect mobilities reported for α -sexithienyl-based TFT's are near $0.1~\rm cm^2/V$ -s, much larger mobilities have been observed in bulk organic materials. In anthracene, for example, there are many reports of time-of-flight mobilities in the 1– $10~\rm cm^2/V$ -s range [7]–[9]. Indeed, at low temperature, some organic mobilities are among the largest reported for any semiconducting materials (> $10^5~\rm cm^2/V$ -s at liquid helium temperature in tetramethyltetraselenafulvalene hexafluorophosphate, for example [10]). The problem is that, thus far, these mobilities have not been useful for building electronic devices (though they have certainly been useful in transport physics studies and may be useful in applications such as organic metal replacements).

II. PENTACENE TFT MOBILITY

We have fabricated organic TFT's using pentacene, a fusedring polycyclic aromatic hydrocarbon, as the active material. These devices have field-effect mobility as large as $0.7 \text{ cm}^2/\text{V}$ -s, comparable to hydrogenated amorphous silicon TFT's. While the bulk electrical conductivity of acenes such as pentacene ($\sim 10^{-15}$ S/cm) is well known [11], the field-effect properties of pentacene and many other conjugated materials remain relatively unexplored. Like α -sexithienyl, pentacene has a strong tendency to form molecular crystals and when deposited by evaporation will typically form well-ordered films even for low substrate temperatures. With pentacene, for example, ordered films are obtained when deposited by evaporation at temperatures as low as $0 \, ^{\circ}\text{C}$ [12].

This tendency to form ordered films at low temperature may be extremely important. Currently, hydrogenated amorphous silicon, the most widely used broad area electronic material, requires temperatures of 250–350 °C for high-quality

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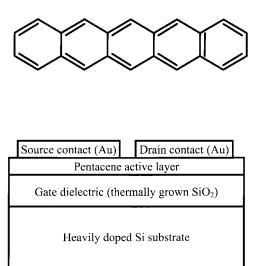


Fig. 1. Kekulé structure of a pentacene molecule and cross section of a pentacene TFT.

Gate electrode (Al)

material deposition and thus requires the use of glass or other high-temperature substrates. Pentacene, or other organic semiconductors, may allow device fabrication on flexible plastic substrates. For applications such as AMLCD's which currently use glass substrates, this would allow displays with decreased weight and improved ruggedness. The use of a flexible substrate could also dramatically lower fabrication cost by allowing continuous web processing. In addition, although the molecular bonds are often quite strong, individual molecules are held together by relatively weak van der Waals forces. Hence the electronic properties of the material are primarily determined by the molecular properties, potentially making organic devices more tolerant to at least some types of impurities than devices fabricated from covalently bonded inorganic materials.

The pentacene TFT's reported here use a staggered-inverted structure as shown in Fig. 1. For convenience, a heavily-doped silicon wafer is used as a substrate, and a 400-nm thick oxide layer is thermally grown for use as the gate dielectric. A 50-nm pentacene active layer is deposited by thermal evaporation at 7×10^{-5} Pascal after material purification by vacuum gradient sublimation [13]. The devices are completed by evaporating a 50-nm gold layer through a shadow mask to form source and drain contacts and a 100-nm aluminum layer onto the wafer back side to contact the gate.

Fig. 2 shows the I_D – V_{DS} characteristics for a typical pentacene TFT. This device has a channel length and width of 20 and 220 μ m, respectively, and a gate dielectric thickness of 400 nm. From the $I_D^{1/2}$ – V_{GS} characteristics for the same pentacene TFT (Fig. 3), we extract a field-effect mobility of 0.62 cm²/V-s in the saturation region at V_{DS} = –80 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.

Interestingly, previous reports of pentacene based TFT's exist, but with much lower field-effect mobility [14]–[17].

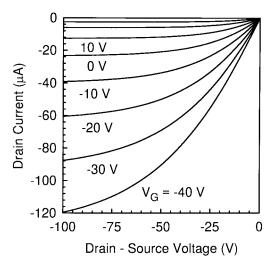


Fig. 2. I_D versus V_{DS} characteristics for a typical pentacene TFT with gate length and width of 20 and 220 μ m, respectively, and 400-nm gate dielectric thickness. The pentacene active layer has a nominal thickness of 50 nm.

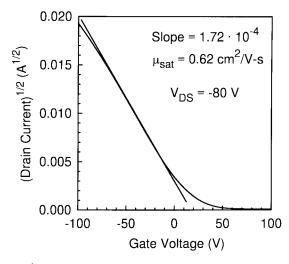


Fig. 3. $I_D^{1/2}$ versus V_{GS} characteristics for the pentacene TFT. The TFT field-effect mobility in the saturation region is greater than 0.62 cm²/V-s with V_{DS} biased at -80 V.

TFT's fabricated using a solution cast pentacene precursor had a field-effect mobility near 0.009 cm²/V-s [14], [15], while previously reported TFT's fabricated using evaporated pentacene had a field-effect mobility near 0.002 cm²/V-s [16], 0.03 cm²/V-s [17]. 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-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 because of disorder, defects, and chemical impurities which can form trapping states. Pentacene is a strong molecular crystal former and films deposited by evaporation onto substrates at room temperature or above are strongly three-dimensional (3-D) and show evidence of strong ordering effects. Consequently, we have optimized our pentacene deposition conditions (deposition rate and substrate temperature) to obtain high-quality pentacene films. For the

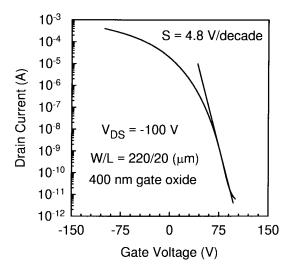


Fig. 4. $\log_{10}\left(I_D\right)$ versus V_{GS} characteristics for the pentacene TFT of Fig. 3. The TFT on/off current ratio is greater than 10^8 even with V_{DS} biased as high as -100 V.

transistors reported above we used a substrate temperature of about 120 °C and a deposition rate of 2–3 Å/s.

To test the importance of deposition conditions on TFT performance we fabricated TFT's using material deposited by flash evaporation. 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 °C, only very low mobility TFT's were obtained. For example, a flash evaporated film onto a substrate held at 160 °C gave TFT's with field-effect mobility near 0.0001 cm²/V-s (again extracted using fits to well-behaved $I_D^{1/2}$ - V_{GS} characteristics). Further, using rapid thermal annealing to rapidly heat the fabricated TFT's to 350 °C for a few seconds increased the TFT field-effect mobility to about $0.002 \text{ cm}^2/\text{V-s}$. 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.

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. We have obtained TFT's with field-effect mobility larger than 0.2 cm²/V-s for depositions onto substrates held at temperatures between 20 and 120 °C and with deposition rates between 2 and 8 Å/s. Maximum mobilities are obtained with substrate temperatures between 50 and 120 °C with deposition rate between 1 and 3 Å/s; field-effect mobility is typically greater than 0.4 cm²/V-s in this range, and mobilities as large as 0.7 cm²/V-s have been obtained.

III. OTHER ORGANIC TFT REQUIREMENTS

Although large mobility is a necessary condition for wide application of organic TFT's, it is not sufficient. For example, in AMLCD's TFT's are used to first place charge on the capacitor that controls pixel transmission and then to isolate the charge there. To be useful, they must have a sufficiently

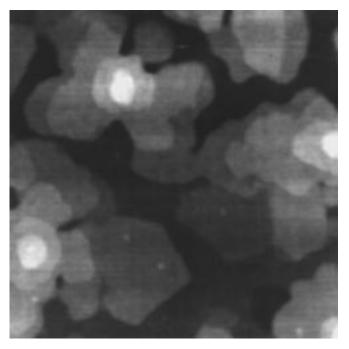


Fig. 5. A $1 \times 1 \mu m^2$ AFM image of a pentacene film with an average thickness of 7 nm deposited onto oxidized silicon.

large on current to charge the pixel capacitance during a line access time and a sufficiently low off current that the charge remains stable during the much longer frame time. a-Si:H TFT's have excellent on/off current ratio, typically near 10^8 , and this is one of the reasons they have found wide application in AMLCD's. α -sexithienyl based organic TFT's have shown on/off ratio as large as 10^6 [3]. Fig. 4 shows the $\log_{10}{(I_D)}$ - V_{DS} characteristics, for the pentacene TFT of Figs. 2 and 3; the on/off current ratio is larger than 10^8 even at $V_{DS} = -100$ V.

In contrast to the optimization of TFT mobility by control of film deposition conditions, we find that on/off current ratio is more strongly affected by film purity effects. Commercially available pentacene is typically 97–99% pure. Using as-received material we have been able to achieve TFT field-effect mobility greater than 0.5 cm²/V-s, however the on/off current ratio is typically only 10^3 – 10^4 . Using vacuum gradient sublimation [13] in two steps to further purify the pentacene before evaporation, the on/off current ratio is increased to greater than 10^8 . This large on/off ratio indicates that the large pentacene mobility is not achieved at the cost of large off conductivity.

Large field-effect mobility and on/off current ratio are important for organic TFT's, but other characteristics are also important for device application. For example, to take advantage of a large on/off current ratio it is necessary to swing the FET gate bias over some range. In saturation the FET drain current $I_D = K \cdot (V_{GS} - V_T)^2$ where V_T is the device threshold voltage and

$$K = \frac{1}{2} \cdot \mu \cdot C_{\text{ox}} \cdot \frac{W}{L}$$

where W and L are the device geometrical width and length, respectively, $C_{\rm ox}$ is the oxide (or other dielectric) capacitance

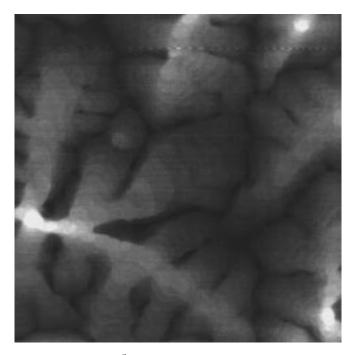


Fig. 6. A $2.5 \times 2.5 \,\mu\text{m}^2$ AFM image of a pentacene film with an average thickness of 30 nm deposited onto oxidized silicon.

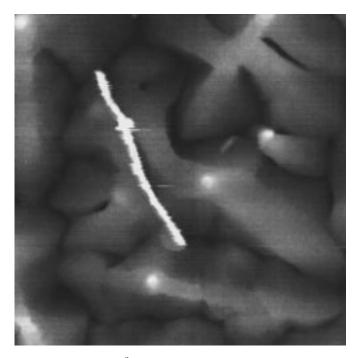


Fig. 7. A $2.5 \times 2.5 \ \mu\text{m}^2$ AFM image of a pentacene film with an average thickness of 150 nm deposited onto oxidized silicon.

per unit area and μ is the carrier field-effect mobility. Thus large field-effect mobility is the key to obtaining a large on current for a given device geometry and gate dielectric. Unfortunately, as Figs. 3 and 4 show, the TFT does not turn off abruptly at the threshold voltage V_T as the saturation current equation would suggest. Instead there is a subthreshold region where the drain current varies approximately exponentially with gate voltage. For single crystal silicon FET's the subthreshold region is well-behaved and ideally the subthreshold

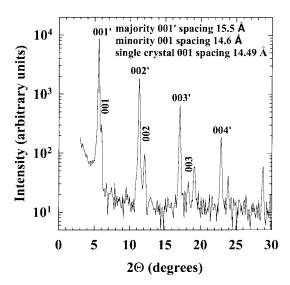


Fig. 8. X-ray diffraction results for a pentacene film with 150 nm average thickness deposited on SiO₂ held at 60 $^{\circ}$ C.

slope comes from the exponential activation of current with voltage compared to the thermal voltage; at room temperature this gives a subthreshold slope for drain current of about 60 mV/decade. Due to tail and midgap states, a-Si:H TFT's have a much larger subthreshold slope, typically 0.3–1.5 V/decade.

As shown in Fig. 4, the subthreshold slope for the pentacene TFT's discussed, is about 4–5 V/decade. This is a significant problem since, as Fig. 4 shows, to achieve a low off current it is necessary to swing the TFT gate voltage over a large range. The result shown for pentacene is typical of that commonly obtained for organic TFT's (our results using α -sexithienyl are similar) and it is worth asking if the large subthreshold slope is an intrinsic property of organic TFT's which might then limit their application.

As noted above, pentacene and similar organic semiconducting materials have a strong tendency to form molecular crystals. We believe the ease with which ordered films may be formed is an important factor in achieving high field-effect mobility devices and in considering organic TFT's in more detail it is worth looking more closely at film formation. Fig. 5 shows an atomic force microscopy image of a pentacene film with 7 nm average thickness deposited on SiO₂ held at 60 °C. The film is both strongly ordered and 3-D. The film forms a series of terraces, the minimum step height between terraces is about 15 Å which corresponds well with the pentacene molecular length of 14.5 Å [12], [17]. The islands shown typically begin with a two or three molecule high step followed by single molecule steps for succeeding terraces.

As the pentacene film thickness increases the initial terraced islands grow together and a dendritic structure results. Fig. 6 shows an atomic force microscopy (AFM) image of a pentacene film with 30 nm average thickness deposited on SiO₂ held at 60 °C. Individual molecular steps can still be seen and the 3-D film structure is notable. The film roughness does not increase significantly for films thicker than approximately 30–50 nm. Fig. 7 shows an AFM image of a pentacene film with 150 nm average thickness deposited on SiO₂ held at 60 °C. The surface roughness is similar to that of Fig. 6. In

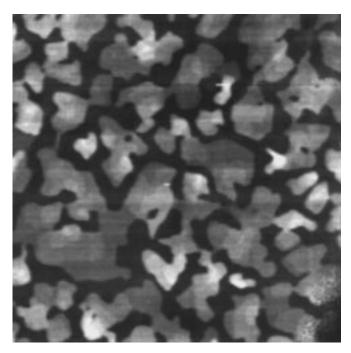


Fig. 9. A $2.5 \times 2.5 \ \mu\text{m}^2$ AFM image of a pentacene film with an average thickness of 7 nm deposited onto LiF at 60 °C.

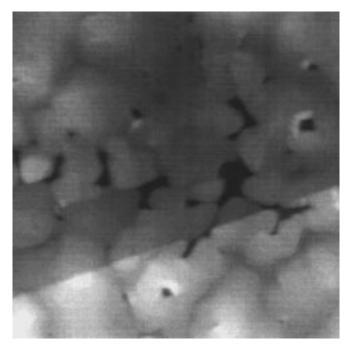


Fig. 10. A $2.5 \times 2.5 \ \mu m^2$ AFM image of a pentacene film with an average thickness of 50 nm deposited onto LiF at 120 °C.

addition, the spacing between peaks is similar for the films shown in Figs. 6 and 7. This suggests that peaks do not simply continue to grow with increasing average film thickness, rather, new peaks nucleate, most likely in the regions where previous peak slopes have met.

Fig. 8 shows X-ray diffraction results for a pentacene film with 150 nm average thickness deposited on SiO_2 held at 60 °C. The expected crystal structure of pentacene is triclinic [18], [19] with a=7.90 Å, b=6.06 Å, c=16.01 Å,

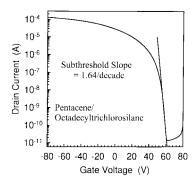


Fig. 11. $\log_{10}{(I_D)}$ versus V_{GS} characteristics for pentacene/octa-decyltrichlorosilane TFT.

 $\alpha=101.9^{\circ}, \beta=112.6^{\circ}$, and $\gamma=85.5^{\circ}$. This leads to an expected (001) plane spacing of 14.49 Å. In Fig. 8, we find this spacing present as a minority component with a majority component of 15.5 Å. Similar results have been reported [12], [17], and attributed to a "thin-film phase." We speculate that the initial phase is due to film substrate interactions and the pentacene is able to relax to the expected single crystal structure for thicker films. This points to the importance of the film substrate interface which may also be critical for device operation. The fact that we saw such strong peaks confirm that the deposited pentacene film is well ordered.

Fig. 9 shows an AFM image of a pentacene film with 7 nm average thickness deposited on cleaved crystalline LiF held at 60 °C. Comparison with Fig. 5 indicates that the growing film is strongly affected by the presence of a crystalline substrate. (note there is a change of scan scale between these figures) The islands shown in Fig. 9 are much flatter than those of Fig. 5. In each case the island begins with a multimolecule step (typically 2–4 molecules high) but the films on LiF show only single molecule additions (that is only a single terrace adds on the island surface). Fig. 10 shows an AFM image of a pentacene film with 50 nm average thickness deposited on LiF held at 120 °C. A step in the LiF substrate is included in this image and it is notable that despite the differences between growth on SiO₂ and LiF the film is not strongly affected by the presence of the step.

The atomic force images give some insight into pentacene film formation. In particular, a comparison of film growth on LiF and SiO₂ suggests that the film substrate interface can be strongly modified by the presence of an ordered substrate while the resulting bulk molecular crystal may be less strongly affected. Thus, it may be possible to use film substrate interface control as a means of improving subthreshold slope (assuming large slope is not an intrinsic property of the organic semiconducting material).

To test this, we looked at the effect of self-organizing monolayers on the properties of pentacene TFT's. Self-organizing materials are attractive because they allow the formation of an ordered template on an amorphous substrate such as ${\rm SiO_2}$ or perhaps a polymer. Fig. 11 shows the ${\rm log_{10}}\left(I_D\right)\!\!-\!\!V_{GS}$ characteristics for a pentacene TFT fabricated on ${\rm SiO_2}$ pretreated with octadecyltrichlorosilane. The subthreshold slope for this device is about 1.6 V/decade, a marked improvement over the 4–5 V/decade typically observed for pentacene on

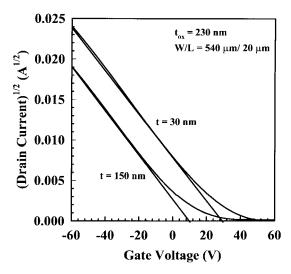


Fig. 12. $I_D^{1/2}$ versus V_{GS} characteristics for pentacene TFT's with active layer 30 and 150 nm, respectively.

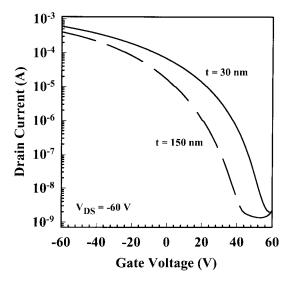


Fig. 13. $\log_{10}(I_D)$ versus V_{GS} characteristics for pentacene TFT's with active layer 30 and 150 nm, respectively.

untreated SiO₂ (Fig. 4). This device was fabricated as part of a series concentrating on film-substrate interface properties and no attempt was made to maximize device mobility or on/off current ratio (for this device the field-effect mobility was about $0.4~\rm cm^2/V$ -s and the on/off current ratio was about 10^7). The importance of this result is in showing that the large subthreshold slope typically observed is not an intrinsic property of the organic semiconductor. It is likely that other techniques for controlling film growth will also lead to improved device characteristics.

Fig. 14 shows the $\log_{10}\left(I_D\right)$ – V_{GS} characteristics for a pentacene TFT fabricated on SiO_2 pretreated with phenyltriethoxysilane. The subthreshold slope for this device is about 2.4 V/decade, but the threshold voltage has now been shifted to a negative value. In addition, although the maximum subthreshold slope is less than that for octadecyltrichlorosilane, the overall gate voltage swing required between large on current and small off current is reduced.

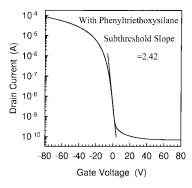


Fig. 14. $\log_{10}\left(I_D\right)$ versus V_{GS} characteristics for pentacene/phenyltriethoxysilane TFT.

The threshold voltage for our pentacene TFT's is typically positive and sometimes large and positive. This means that our devices are normally-on and a positive gate potential must be used to remove carriers present in the channel region at zero gate bias. This is somewhat surprising since bulk pentacene samples typically have extremely large resistivity ($\sim 10^{14}-10^{15}$ ohm-cm) which indicates that pentacene free surfaces normally do not accumulate carriers. Thus the threshold voltage must shift due to interface properties (since our devices are fabricated on high-quality thermally grown SiO₂ it is unlikely that charge in the dielectric is responsible, and work function effects are much too small).

It is important to note that the normally-on characteristics we measure for our pentacene TFT's do not come from bulk conductivity in the pentacene layer. Figs. 12 and 13 show $\log_{10}(I_D)$ versus V_{GS} and $I_D^{1/2}$ versus V_{GS} characteristics for pentacene TFT's with active layers 30 and 150 nm thick, respectively, (deposited at 60 °C). If the normally-on characteristics were due to depletion of bulk carriers in the pentacene films we would expect the threshold voltage for the thicker device to be much more positive than that of the thinner device. Instead, the device threshold voltages are similar (in fact, the threshold voltage for the thicker device is somewhat less positive). Bulk conductivity might also lead to a larger off current for thicker device if it is not possible to deplete all of the charge that is present. Again, this in not observed; the off currents for the two devices are similar. Since neither the threshold voltage, off-current, nor zero-gate-bias conductivity scale with active layer thickness we conclude that the positive threshold voltage does not arise from depletion of carriers in the bulk film but rather from an initial accumulation layer at the pentacene/dielectric interface.

IV. CONCLUSION

These results indicate that organic TFT's with large mobility, large current on/off ratio, and controlled threshold voltage and subthreshold slope are possible. This increases the likelihood that organic transistors will find application in AMLCD's, organic light emitting displays, and other broad area electronic applications. In flat panel displays alone, the current market is more than \$12 billion/year, and increasingly, display cost is a driving force for both current and future applications. The replacement of amorphous silicon TFT's

by high-performance organic devices may be a key factor in low-cost market expansion.

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