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Environmental stability and electronic transport of pentacene thin film transistors

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Abstract

The influence of environmental conditions on the electronic structure and the device stability of polycrystalline pentacene transistors were investigated. Electrical in situ measurements of pentacene TFTs were carried out to study the influence of dry oxygen on the device operation. The polycrystalline pentacene thin film transistors were fabricated by organic molecular beam deposition (OMBD) on thermal oxide dielectrics. Exposing the pentacene film to oxygen leads to the creation of acceptor-like states deep in the bandgap. The acceptor-like states cause a reduction of the subthreshold slope and a shift of the onset of the drain current to positive gate voltages. The threshold voltage and the mobility of the transistors are not affected by the dry oxygen. A simple model will be described which directly correlates the onset voltage of the transistors with the acceptor concentration in the pentacene film.

Keywords: Thin film transistors; Polymers and organics

1. Introduction

Polycrystalline pentacene has demonstrated the highest carrier mobilities of organic based thin film transistors (TFTs) [1,2]. Therefore, pentacene thin film transistors are promising candidates for large area electronic applications like sensor arrays or radio frequency identification tags (RFID). Despite the fabrication of pentacene transistors with high mobilities the electronic transport is not fully understood. The performance of pentacene TFTs strongly depends on the structural properties of the film, and the interfaces between the channel, the dielectric and the contacts [3–5]. Furthermore, the electronic transport is affected by environmental conditions like oxygen or moisture [6]. In order to gain insights in the electronic transport properties of polycrystalline pentacene TFTs electrical in situ measurements of pentacene TFTs were carried out. The influence of environmental conditions was studied by exposing the TFTs to dry oxygen. The influence of the conditions on the electronic transport will be discussed for polycrystalline pentacene TFTs prepared on thermal oxide.

2. Experimental

The cross section of a staggered pentacene thin film transistor with bottom drain and source contacts is shown in Fig. 1(b). A highly doped silicon wafer was used as a substrate. The gate dielectric of the transistor was formed by a 50 nm thick silicon oxide layer. The bottom drain and source contacts were defined by optical lithography. In order to improve the adhesion of the gold drain and source contacts on the substrate a 2–3 nm thick tungsten film was evaporated prior to the gold film. Before bringing down the pentacene molecules (Fig. 1(a)) the thermal oxide was treated by hexamethyldisilazane (HMDS). The pentacene molecules were deposited by organic molecular beam deposition (OMBD) using a deposition rate of 0.5 Å/s. The source material was two times sublimation purified before depositing the molecules at a base pressure of 5×10^{-6} Pa. The films were prepared by keeping the

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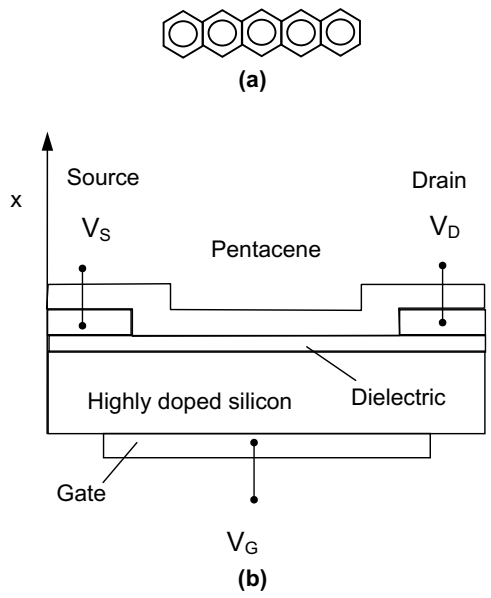

Fig. 1. Molecular structure (a) and schematic cross section of a polycrystalline pentacene thin film transistor (b).

substrate temperature constant at 70 °C. The source temperature was kept constant at 275 °C while depositing the pentacene films. The final pentacene layer has a thickness of 10 nm. Device structures were fabricated ranging from 1 μm to 50 μm channel length.

3. Experimental results and discussion

Understanding the electronic transport of organic semiconductors is essential to improve the device performance of thin film transistors. The transfer characteristic of a polycrystalline TFT fabricated on a thermal oxide is shown in Fig. 2. The transistor has a channel length of 10 μm and a width-to-length ratio (W/L ratio) of 10000. The transistor was measured under ambient conditions. The transistor exhibits a mobility of 0.4 cm^2/V s and an on/off ratio larger than seven orders of magnitude. Furthermore, the drain current starts to increase (onset of the drain current) for positive gate voltages, whereas the extracted threshold voltage of the TFT is negative. Such behavior is typical for polycrystalline pentacene thin film transistors characterized under ambient conditions [3,7]. Numerical simulations indicate that the shift of the onset of the drain current is caused by acceptor-like states deep in the bandgap [8,9]. Several publications reveal that the acceptor-like states are caused by oxygen and/or moisture, which is incorporated in the organic film [6]. However, the formation of acceptor-like states is unclear. The defects may be formed during the deposition as the source material is not pure enough. Alternatively, defects may be formed due to an exposure of the film to oxygen or moisture.

To study the influence of oxygen on the device characteristic electrical in situ measurements were carried out. Electrical in situ measurements of transfer curves for a drain voltage $V_D = -1.0$ V are shown in Fig. 3. The solid bold lines in Fig. 3 represent the transistor characteristic after the deposition of the pentacene film. The pentacene film has a thickness of 10 nm. The sample exhibits a saturation mobility of 0.5 cm^2/V s. The transistor exhibits a subthreshold slope of 0.2 V/decade, which is very low for pentacene TFT prepared on thermal oxide. The charge on the gate, which is required to modulate the drain current by one order of magnitude is typically 2 times higher (assuming the subthreshold slope is normalized by the gate capacitance) [3]. The onset of the drain current is observed for very small negative voltages. Therefore, the subthreshold behavior of the transistor is significantly different from the transistor characteristic in Fig. 2. The onset of the drain

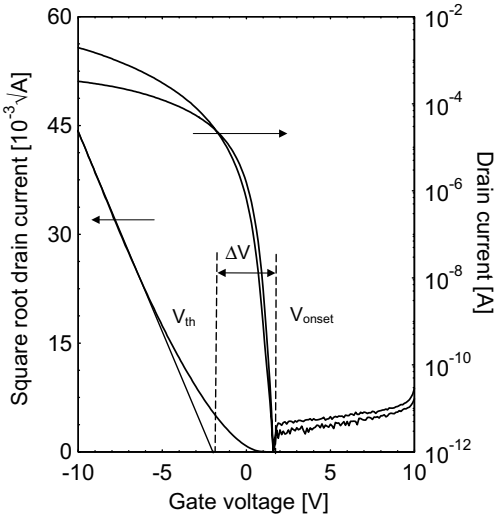

Fig. 2. Transfer characteristic of a pentacene TFT. The transistor was characterized under ambient conditions.

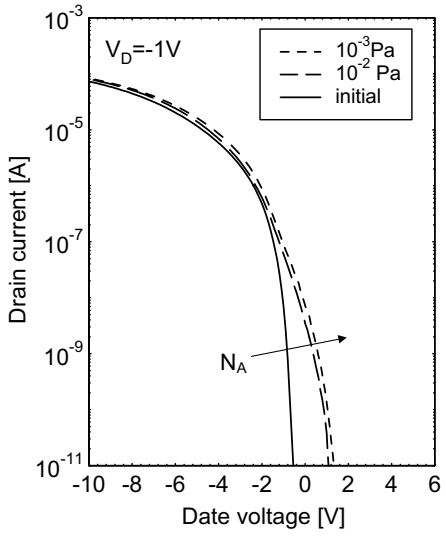

Fig. 3. Electrical in situ measurements of the transfer characteristic of a pentacene TFT for a drain voltage of $V_D = -1$ V. The transistors were exposed to dry oxygen leading to acceptor-like states, N_A, in the pentacene film.

current for small negative voltages and the low subthreshold slope are caused by the low concentration of acceptor-like states in the film.

In the following the sample was exposed to dry oxygen. The concentration of oxygen was controlled by the pressure in the high vacuum chamber. Due to oxygen exposure the onset of the drain shifted towards positive gate voltages. The mobility and the threshold voltage of the transistors were not affected by the oxygen contamination. Measurements of the device characteristic were carried out until no further change of the transfer characteristic could be observed. The transfer characteristic for a pressure of 10^{-3} Pa is shown in Fig. 3. We extracted an onset voltage of +1.0 V from the transfer curve. The onset voltage can be defined as the gate voltage for which the drain current starts to increase. The measurement of the onset voltage is limited by the electrical setup rather than the organic transistors. However, the error in determining the onset voltage is assumed to be small as the onset voltage can be extracted down to current levels of 10 pA. The oxygen exposure leads to an increase of the subthreshold slope from 0.2 V/decade to >0.5 V/decade.

In the following the oxygen concentration was further increased by lowering the pressure in the deposition chamber. However, increase the oxygen concentration leads to no further shift of the onset voltage.

The electrical in situ measurements prove that oxygen is incorporated in the film by exposing the sample to atmosphere. The two times sublimation purified source material is not the source of the impurities as no shift of the onset current is observed for the initial measurement. Further measurements of different device geometries indicate that the morphology of the pentacene film and the contacts have a strong influence on the stability of the devices. The measurements reveal that with increasing size of the pentacene crystals, the influence of oxygen on the device operation is reduced.

In the following we tried to directly derive the dopant concentration from the experimental data. If the device operates in the linear region it can be assumed that the electric field in the channel is uniform, so that a uniform depletion layer is formed in the channel. Under such conditions the width of the depletion layer is given by

$$W(x) = d_{\mathrm{die}} \frac{\varepsilon_{\mathrm{pen}}}{\varepsilon_{\mathrm{die}}} \left[\sqrt{1 + \frac{2\varepsilon_{\mathrm{die}}(V_{\mathrm{G}} - V_{\mathrm{fb}} - V(x))}{qN_{\mathrm{A}} d_{\mathrm{die}} \varepsilon_{\mathrm{pen}}}} - 1 \right], \qquad (1)$$

where d_{die} and d_{pen} are the thicknesses of the dielectric and the pentacene film. $\varepsilon_{\mathrm{die}}$ and $\varepsilon_{\mathrm{pen}}$ are the relative permittivity of the dielectric and the pentacene film. q is the elementary charge, V_{G} and V_{fb} are the gate and the flat band voltage, and N_{A} is the acceptor concentration in the film [10]. We can substitute the voltage $V(x)$ by $V_{\mathrm{G}} - V_{\mathrm{onset}}$. Furthermore, we assumed that the complete pentacene film is depleted as the film is only 10 nm thick. Subsequently we can derive an expression for the acceptor concentration in the film.

$$N_{\mathrm{A}} = \frac{V_{\mathrm{onset}} - V_{\mathrm{fb}}}{q d_{\mathrm{pen}}} \cdot \frac{2\varepsilon_0 \varepsilon_{\mathrm{pen}} \varepsilon_{\mathrm{die}}}{\varepsilon_{\mathrm{die}} d_{\mathrm{pen}} + 2\varepsilon_{\mathrm{pen}} d_{\mathrm{die}}}. \qquad (2)$$

As $\varepsilon_{\mathrm{die}} \cdot d_{\mathrm{pen}}$ is much smaller than $2 \cdot \varepsilon_{\mathrm{pen}} \cdot d_{\mathrm{die}}$ Eq. (2) can be simplified, so that we get the following expression for the acceptor concentration:

$$N_{\mathrm{A}} \approx \frac{V_{\mathrm{onset}} - V_{\mathrm{fb}}}{q d_{\mathrm{pen}}} \cdot C_{\mathrm{G}}, \qquad (3)$$

where C_{G} is the gate capacitance. The voltage $V_{\mathrm{onset}} - V_{\mathrm{fb}}$ can be determined by comparing unexposed and exposed transistors. In this case, the unexposed sample is used as a reference. Such approach was already suggested by Meijer and coworkers for polymer TFTs [6]. Based on the experimental results a minimum detectable acceptor concentration of $5 \times 10^{16}\ \mathrm{cm}^{-3}$ can be estimated. The transistor in Fig. 3 exhibits an acceptor concentration of $5 \times 10^{17}\ \mathrm{cm}^{-3}$. It is important to mention that different device geometries exhibit different shifts of the onset voltage. The shift of the onset voltage depends on the morphology of the film and/or the contacts. Short channel transistors exhibit acceptor concentrations of up to $3 \times 10^{18}\ \mathrm{cm}^{-3}$. The results reveal that the pentacene films are not uniformly doped. The oxygen seems to be incorporated in the areas of low structural order.

The calculated acceptor concentrations are in good agreement with numerical simulations of TFTs assuming a density-of-states model, which accounts for localized states [9]. The simulations assume an exponential defect distribution, which extends deep in the bandgap.

4. Conclusion

The influence of environmental conditions on the electronic structure and the device operation of pentacene thin film transistors were investigated. Electrical in situ measurements of pentacene TFTs were carried out to study the influence of oxygen on the device operation. Staggered TFT structures with bottom drain and source contacts were used for the device fabrication. Oxygen exposure of the pentacene film leads to a significant change of the subthreshold characteristic of the transistors. Under vacuum conditions the onset of the drain current is observed for small negative gate voltages. The oxygen incorporated in the pentacene leads to a shift of the onset of the drain current towards positive gate voltages. As a consequence the subthreshold slope is increased from 0.2 V/decade for 50 nm thick dielectrics to >0.5 V/decade. Furthermore, the results indicate that the morphology of the film has a strong influence on the stability of the device structures.

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