



## Short communication

## DNA adsorption measured with ultra-thin film organic field effect transistors

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## ABSTRACT

Organic ultra-thin film field effect transistors (FET) are operated as label-free sensors of deoxyribonucleic acid (DNA) adsorption. Linearized plasmid DNA molecules (4361 base pairs) are deposited from a solution on two monolayers thick pentacene FET. The amount of adsorbed DNA is measured by AFM and correlated to the concentration of the solution. Electrical characteristics on the dried DNA/pentacene FETs were studied as a function of DNA concentration in the solution. Shift of the pinch-off voltage across a wide range of DNA concentration, from very diluted to highly concentrated, is observed. It can be ascribed to additional positive charges in the semiconductor induced by DNA at a rate of one charge for every 200 base pairs. The sensitivity 74 ng/cm<sup>2</sup>, corresponding to 650 ng/ml, is limited by the distribution of FET parameters upon repeated cycles, and is subjected to substantial improvement upon standardization. Our work demonstrates the possibility to develop label-free transducers suitable to operate in regimes of high molecular entanglement.

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## 1. Introduction

A variety of biosensors have been developed for genomics and proteomics to monitor specific binding of biomolecules on solid-state substrates (Lyubchenko, 2004). Deoxyribonucleic acid (DNA) chips and DNA microarrays are used in molecular biology, pharmaceutical industry and clinical research to identify presence of specific biological targets. Fluorescence detection of labeled probes or targets is a widely used scheme for detecting specific recognition events (Hoheisel, 2006). Other methods include chemi-luminescence, surface plasmon resonance, quartz crystal microbalance and electrical conductivity (Drummond et al., 2003). Most of these methods lack a reliable quantitative response, which undermines their impact in early diagnosis or assessment of prognosis. Electronic transduction of the diagnostic event by low cost devices is desirable for in-field massive screening, without the need of sophisticated readout equipment or specialized personnel. For these applications, field effect transistors offer the advantage of a well-established scalable technology and ease of integration (Souteyrand et al., 1997).

Organic field effect transistors (OFET) are multi-parameter devices where charge carriers are generated and transported within the first few monolayers (ML) in contact with the gate dielectric

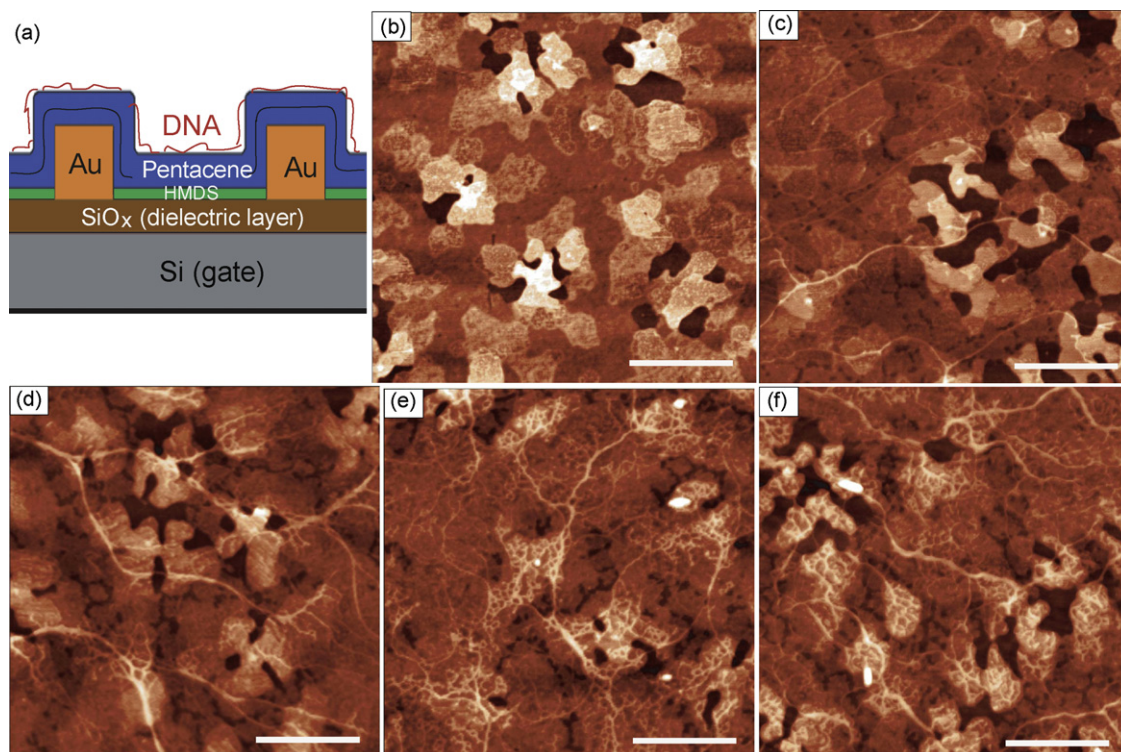
(Dinelli et al., 2004; Gomes et al., 2004). A schematic view of the device configuration is shown in Fig. 1a. In the case of ultra-thin film OFETs (1–5 ML), the exposure to the environment will affect the OFET response as molecules either adsorb on or approach the charge transport layer. Although this not desirable for the performance and stability of the device, it can become attractive for different applications. An example is the polyelectrolyte gate OFET (Said et al., 2006; Bernards et al., 2006), where the charge separation within a polyelectrolyte layer in contact with the organic semiconductor gates the charge carriers in the devices. As long as the faradic current within the polyelectrolyte is negligible, the response of the device reflects the change in the density of charge carriers induced by the double-layer. It can be envisioned that the capacitive coupling between large polyelectrolyte biomolecules adsorbed on the outer interface of an organic semiconductor will similarly modify the distribution of charge carriers. The electrostatic interaction of charge carriers and charged or polar species in the environment will be enhanced as their distance is small. Recently, OFET sensing DNA thick layers or highly concentrated solutions has been reported (Zhang and Subramanian, 2007; Stadler et al., 2007), although limited to the monitoring of the maximum on-current in the transfer characteristics.

Aim of the present work is to prove this concept using ultra-thin film transistors where the active layer consists of two pentacene monolayers only and the response is gated by a molecular polyelectrolyte, viz. DNA. We investigate the response of pentacene FETs upon the deposition of pBR322 DNA molecules from solutions of different concentration. We demonstrate the electrical

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**Fig. 1.** (a) Schematics of the OFET used in this work; (b) AFM morphology of pentacene film in the FET channel; each terrace is 1.5 nm high; (c–f) AFM images of DNA deposited on pentacene at different concentration: (c) 1  $\mu\text{g/ml}$ , (d) 5  $\mu\text{g/ml}$ , (e) 10  $\mu\text{g/ml}$  and (f) 20  $\mu\text{g/ml}$ . Scale bar for all AFM images is 500 nm.

transduction of DNA adsorption from low-coverage to highly entangled regime. DNA molecules induce an additional population of positive charges (holes) in the accumulation layer, contributing to the electrostatic field at the interface. The estimated sensitivity of the present devices is  $2.6 \times 10^{-14} \text{ mol/cm}^2$ , corresponding to about 160 pBR322 molecule/ $\mu\text{m}^2$ . Our result opens interesting perspectives for a new class of label-free transducers of DNA adsorption without requiring binding agents or immobilization.

## 2. Experimental part

Our device is a pentacene field effect transistor whose active layer is made of two stacked molecularly ordered monolayers (Dinelli et al., 2004, see Appendix A). Each monolayer is 1.5 nm high. DNA is a suitable prototype biomolecule for this study, since it is a polyanion with homogeneous charge density along the chain ( $2e^-/\text{bp}$ ), easy to visualize by scanning probe microscopy. As prototype, we use pBR322 plasmid (circular) DNA cleaved by a standard procedure at a precise position to yield DNA molecules of equal length (4361 bp) and sequence (see Appendix A). A drop of water solution (5  $\mu\text{l}$  volume) whose DNA concentration (DNA) varied in the range between 1 and 20  $\mu\text{g/ml}$ , and with constant buffer concentration of 10 mM tris-hydroxy-methyl-amino-methane (TRIS), is deposited on the transistors for 10 min. Then the drop is washed with UHQ water and dried with  $\text{N}_2$  prior to perform electrical transfer characteristic measurements.

Pentacene bottom contact field effect transistors were made by high vacuum sublimation. The transistor layout, the fabrication process and the electrical characterization were described earlier (Stoliar et al., 2007). Channel length  $L$  and width  $W$  range from 10 to 40  $\mu\text{m}$  and 1 to 20 mm, respectively. Pentacene coverage of 2.1 ML was chosen for all devices, so that the active channel is exposed to the environment (Gomes et al., 2004). Field effect response of these pristine transistors in air was measured as benchmark. For the devices with  $L = 30$  and 40  $\mu\text{m}$  the charge mobility in ambient

extracted from the transfer characteristics in the saturation regime is  $0.014 \pm 0.003 \text{ cm}^2/\text{V s}$ .

A differential measurement with respect to the bare pentacene and after contact with the buffer solution is carried out for every device. Each experiment involves three steps: (i) the OFET is fabricated and transfer characteristics acquired; (ii) a blank device is made by drop casting of the buffer solution (without DNA) on the device for 10 min, dried, and then characterized; (iii) DNA in the buffer solution is deposited and the device is measured again after drying it. The time lag between the drop deposition and the electrical measurement must be kept constant to 10 min because there is a small drift in time of the transistor response (i.e. the threshold voltage change with a rate of about 10 V/h).

## 3. Results and discussion

The morphology of DNA on pentacene is shown in Fig. 1. Pentacene stacked monolayers are clearly visible. Islands of the third and fourth monolayer are beginning to form above two almost complete monolayers, as commonly observed in several conjugated oligomer ultra-thin films. The effective coverage equals 2.1 ML, with an rms surface roughness of 1.1 nm. By comparison, surface roughness of the FET channel before sublimation of pentacene is 0.5 nm. Adsorption of linear DNA at low concentration (Fig. 1c) yields formation of bundles consisting of a few molecules. At higher concentration (Fig. 1d–f) a hierarchical DNA network is formed, where larger bundles are entangled with small DNA chains. The morphology does not seem to change substantially as strong entanglement is achieved at higher concentration (Fig. 1e and f). We monitor the morphological changes vs. [DNA] by measuring the length-scale saturated rms roughness  $\sigma$  (Palasantzas and Krim, 1995) from the AFM images acquired in the FET channels. The plot in Fig. 2a shows that  $\sigma$  first increases, as a result of increasing coverage and DNA bundling, then saturates at concentrations higher than 10  $\mu\text{g}/\mu\text{l}$ . Saturation may be ascribed to the adsorbed DNA chains repelling

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