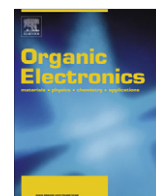




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Origin of mechanical strain sensitivity of pentacene thin-film transistors

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ABSTRACT

We report on bending strain-induced changes of the charge carrier mobility in pentacene organic thin-film transistors employing a combined investigation of morphological, structural, and electrical properties. The observed drain current variations are reversible if the deformation is below 2%. The morphology and structure of the active pentacene layer is investigated by scanning force microscopy and specular synchrotron X-ray diffraction, which show that bending-stress causes morphological rather than structural changes, modifying essentially the lateral spacing between individual pentacene crystallites. In addition, for deformations >2% the rupture of source and drain gold electrodes is observed. In contrast to the metal electrodes, the modification of the organic layer remains reversible for deformations up to 10%, which suggests the use of soft and flexible electrodes such as graphene or conducting polymers to be beneficial for future strain sensing devices.

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1. Introduction

Thin films of conjugated organic molecules are subject of intense research due to their applicability in novel (opto-)electronic devices, including organic thin-film transistors (OTFTs). Pentacene (PEN) is the prototypical hole-conducting material with notably high charge carrier mobilities of up to 5.5 cm²/Vs [1–3] in p-type OTFTs. One key advantage of organic electronic devices is the possibility to produce flexible all-organic OTFTs with the functional organic semiconductor films deposited on flexible plastic foils like Mylar® or polyethyleneterephthalate (PET) as substrate [4]. Through their intrinsic flexibility, OTFTs can be applied as mechanical strain-sensing devices [5,6] exhibiting significant advantages over conventional

types of sensors: they can be processed under ambient conditions and are generally inexpensive to fabricate [4–11]. For sensing strain, reversible changes in the electrical characteristics of OTFTs were employed, including drain current, charge carrier mobility, threshold voltage, and contact resistance for deformations up to 1–2% [5,6,9,12,13]. In particular, the reported changes in mobility were proposed to be due to morphological changes of the PEN layers under mechanical strain and/or the activation of trap states in the PEN/electrode interface region [5,6,9,14,15]. However, up to now there has been no direct experimental evidence to support these suggestions. In order to enable targeted research for improving current OTFTs for future reliable sensing applications, further experimental work is needed to complete the microscopic picture of strain-impact on OTFT performance. Here we report a comprehensive electrical characterization of flexible OTFT devices under applied bending-stress, relating the device characteristics to morphological and structural

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78 properties of the active layer, as determined by a combina-
79 tion of scanning force microscopy (SFM) and specular X-
80 ray diffraction (XRD). We identify the deterioration of the
81 metal electrodes to be responsible for irreversibilities in
82 the device characteristics, while the observed modifica-
83 tions of the active organic layer remain reversible for
84 deformations up to 10%, which suggests using soft organic
85 electrodes for future improved sensing applications.

86 2. Experimental

87 OTFTs were fabricated on 200 μm thin and flexible
88 polyethylenetherephthalate (PET) substrates. A gold layer
89 was vacuum-deposited on the substrates as gate electrode.
90 Polyvinylalcohol (PVA) with ammonium dichromate (AD)
91 salt (cross-linking agent) was deposited from water solu-
92 tion as gate dielectric by spin-coating and subsequent
93 UV-curing for cross-linking. Gold source and drain elec-
94 trodes (Fig. 1a) were patterned by thermal deposition
95 through a shadow mask. Pentacene (PEN) was used as or-
96 ganic semiconductor. PEN films were evaporated at a base
97 pressure $<10^{-7}$ mbar; the film thickness and the deposition
98 rate (0.5 nm/min) were monitored by a quartz crystal
99 microbalance placed next to the sample; the device is
100 sketched in Fig. 1a. A special apparatus for defined bending
101 of the OTFT (Fig. 1c) was developed to be suitable for use in
102 both SFM and specular XRD experiments. XRD measure-
103 ments were carried out at the beamline W1 at DESY-
104 HASYLAB (Hamburg, Germany) with a primary beam en-
105 ergy of 10.5 keV, using the bending apparatus (Fig. 1c) as
106 sample holder for *in situ* experiments, and a standard setup
107 (PVA samples fixed planar on a Si wafer) for reference

108 measurements. The electrical and morphological charac-
109 terization of the PEN layers under deformation was carried
110 out *in situ* under ambient conditions. For electrical charac-
111 terization a Keithley 2636A sourcemeter was used. The
112 surface topography was imaged with SFM (Nano Wizard,
113 JPK instruments). The PEN film strain was calculated with
114 the model of beam buckling (Fig. 1b), which allows to cal-
115 culate the film strain, ϵ , directly from the bending radius
116 [16], as demonstrated before [9,17]. The bending radius
117 was calculated taking into account the homogeneous
118 bending of the PET foil. This setup allowed to strain PEN
119 films up to 10%, with the upper limit defined by the small-
120 est accessible radius of curvature and the thickness of the
121 PET substrate. All electrical device characterization, XRD
122 and SFM measurements, were carried out under ambient
123 conditions.

124 3. Results

125 Transfer curves (i.e., drain current (I_D) versus gate volt-
126 age (V_G) characteristics) of the devices were recorded for
127 different degrees of strain. The bending-stress was released
128 after acquiring transfer characteristic for a particular strain
129 in order to verify the reversibility of the process (i.e., if the
130 current recovers to the initial value). We find, as expected
131 [9], a decrease in drain current as function of device defor-
132 mation (Fig. 2a). For deformations of up to 1.7%, straining
133 of the device causes reversible variations of the transfer
134 curves. Starting from 1.7% I_D does not recover to the initial
135 value of the pristine device upon bending-stress relief.
136 Transfer characteristics exhibit a pronounced hysteresis
137 with the hysteresis loop-area increasing with increasing

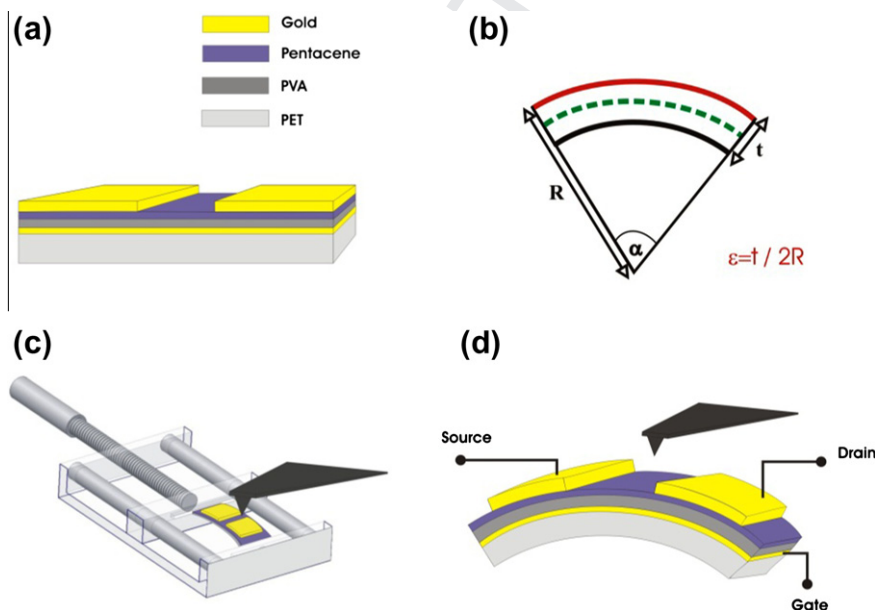


Fig. 1. (a) Schematic representation of the top-contact OTFT architecture. (b) Model used to calculate PEN film strain ϵ ; R denotes the radius of curvature, t the device thickness. Red and dashed green lines indicate the upper and neutral plane, respectively. Strain of PEN film is assumed to be equal to the strain of the beam surface. (c) The bending apparatus with the OTFT device in place. (d) Scheme of the device under bending stress with the SFM-tip; electrical contacts are indicated for the drain-current sensitivity measurements in the deformed OTFT and for the *in situ* morphological investigations on both electrodes and active layer by SFM. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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