

Role of the substrate thickness for the structural properties of organic–organic heterostructures

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Abstract

F₁₆CuPc deposited on pentacene is characterized by the coexistence of two different configurations: F₁₆CuPc is found in the standing up phase (“*s*-configuration”) on top of pentacene terraces and in a lying down phase (“*l*-configuration”) at pentacene step edges. By combining AFM and grazing incidence X-ray diffraction we show that the ratio between F₁₆CuPc in *l*- and *s*-configurations increases with thickness of the pentacene substrate film, demonstrating the role of the pentacene steps as nucleation centers for the F₁₆CuPc *l*-configuration. Experiments performed with ultra-thin pentacene thicknesses disclose that the F₁₆CuPc *l*-configuration does not grow on top of the first and second pentacene layers, pointing to the action of long-range interactions with the substrate.

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1. Introduction

The high interest in organic semiconductor materials stems from the promise of the development of organic-based devices [1–4], and currently stimulates the vast amount of research in this field. Many of the electronic devices, such as ambipolar transistors, OLEDs or solar cells, rely on organic heterostructures based on organic materials with p- and n-type transport properties. The morphology and structure of the organic films as well as the structural properties of the organic interface are of paramount importance for the device performance. To date, very little is known about how the organic interface affects the structure, morphology and growth mode of the subsequently

deposited molecular film. Although numerous studies have been devoted to the performance of devices based on p–n organic heterojunctions, there are few structural studies addressing this important issue [5–12].

In a recent study of the growth of heterostructures of F₁₆CuPc (Fig. 1a) deposited onto pentacene (Fig. 1b), two oligomers used for n-type and p-type conduction, respectively, we have shown that the terrace steps of the underlying pentacene catalyze the growth of narrow and high crystallites of lying-down F₁₆CuPc molecules (“*l*-configuration”) [12]. Coexisting with this new F₁₆CuPc structure, the energetically preferred upright-standing configuration of F₁₆CuPc (“*s*-configuration”) forms on top of the pentacene terraces. This growth scenario is summarized schematically in Fig. 1c [12]. These results manifest a rather strong interaction of F₁₆CuPc with the pentacene molecules at the steps.

In this study we disclose that the structural characteristics of the F₁₆CuPc/pentacene bilayers is dependent on the pentacene film thickness and that this is in turn related to

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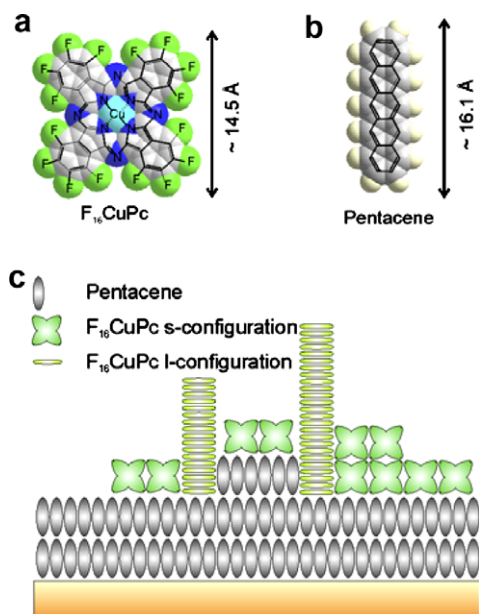


Fig. 1. Molecular structure of (a) pentacene and (b) $F_{16}\text{CuPc}$. (c) Schematic summary of the organization behaviour of $F_{16}\text{CuPc}$ onto pentacene.

the strong interaction of $F_{16}\text{CuPc}$ with the pentacene molecules at the steps. These important structural details have been found by combining atomic force microscopy (AFM) with in situ grazing incidence X-ray diffraction (GIXD).

2. Experimental

The organic heterostructures have been prepared by organic molecular beam deposition (OMBD), growing $F_{16}\text{CuPc}$ onto previously deposited pentacene films on Si(100) wafers covered by their native oxide. The substrates were cleaned ultrasonically with acetone and ethanol and then dried with nitrogen gas. Subsequently they were annealed under ultra high vacuum (UHV) above 500 °C and slowly cooled to the growth temperature. The molecules were purified twice by gradient sublimation before use. The film coverage and growth rate were controlled by means of a calibrated quartz crystal monitor (QCM). All heterostructures have been synthesized at room temperature. Those used for the AFM characterization have been grown under a base pressure of 10^{-10} mbar, those grown in the portable UHV chamber for the in situ X-ray experiments, at 10^{-9} mbar. The GIXD measurements, which enable the access to the in-plane film structure, have been performed at the beamline ID-3 in the ESRF (Grenoble) with a wavelength of $\lambda = 0.72316$ Å. To allow the in situ characterization, a specially designed portable UHV chamber, equipped with a beryllium window, a QCM, a Knudsen cell and a heatable sample holder has been used. The AFM measurements have been performed with a commercial Nanotec microscope in tapping-mode under ambient conditions.

3. Results and discussion

The stepwise $F_{16}\text{CuPc}$ deposition (up to 225 Å) onto 130 Å pentacene has been monitored in situ by GIXD (Fig. 2a) providing information of the in-plane structure of both films. The pentacene film exhibits its well known thin film structure [13–15] (black graphic). Upon subsequent deposition of $F_{16}\text{CuPc}$, the coexistent growth of $F_{16}\text{CuPc}$ *s*- and *l*-configurations is clearly observed. The in-plane Bragg reflections characteristic of the $F_{16}\text{CuPc}$ *s*-configuration [16,17] emerge and evolve with the same $F_{16}\text{CuPc}$ thickness-dependence as observed on SiO_2 : β bilayer-structure for the first layers followed by the development of the β -structure for increasing coverage (inset) [18]. Notice that the most intense $F_{16}\text{CuPc}$ reflection, the

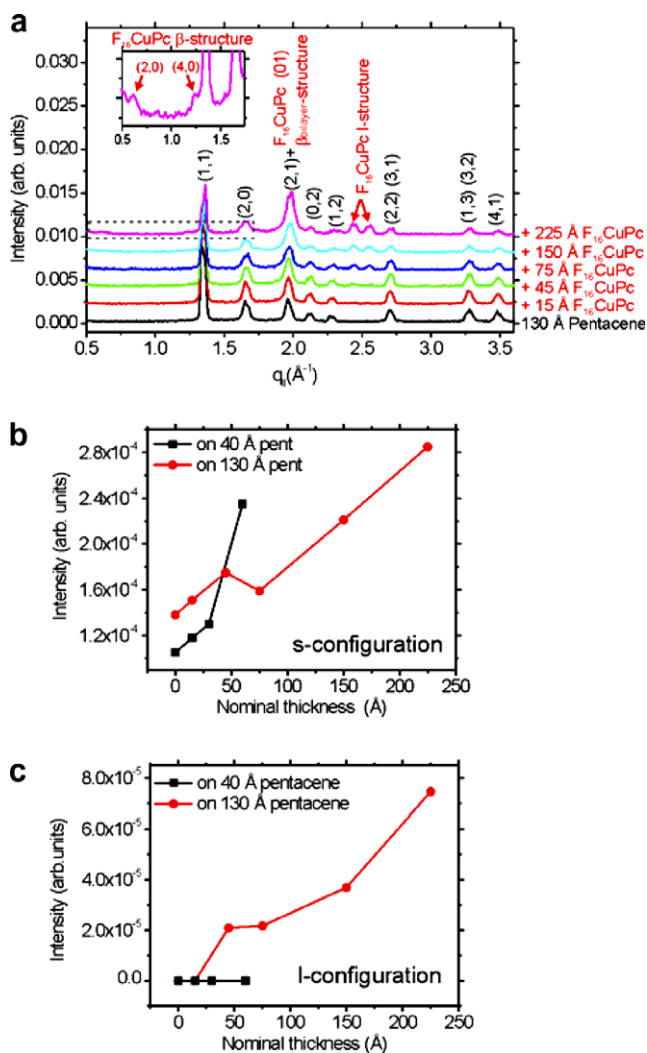


Fig. 2. (a) Evolution of the GIXD data of a 130 Å pentacene film upon stepwise deposition of $F_{16}\text{CuPc}$ on top. The inset shows a zoom of the area marked with the dashed line. (b) Evolution of the $F_{16}\text{CuPc}$ β bilayer-structure (01) reflection (*s*-configuration) intensity upon $F_{16}\text{CuPc}$ deposition on 40 and 130 Å thick pentacene films. (c) Evolution of the *l*-structure reflection at $q_{\parallel} = 2.4387$ Å $^{-1}$ upon $F_{16}\text{CuPc}$ deposition on 40 and 130 Å thick pentacene films.

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