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Monolayer coverage sets the mobility in self-assembled monolayer fieldeffect transistors

Simon G. J. Mathijssen, Edsger C. P. Smits, Paul A. van Hal, Harry J. Wondergem, Sergei A. Ponomarenko, Armin Moser, Roland Resel, Peter A. Bobbert, Martijn Kemerink, René A. J. Janssen, and Dago M. de Leeuw

Grazing incidence diffraction measurements

The internal microstructure of the chromophores was measured by grazing incidence diffraction measurements. The in-plane scattering vectors, q_{xy} , of the Bragg rods can be determined from the diffraction pattern, as shown in Fig. 2 of the original paper. The measurements are reproduced at higher magnification levels in Fig. S1 below. Bragg rods are found at following positions: $q_{xy} = 1.390 \text{ Å}^{-1}$, 1.607 Å⁻¹ and 1.967 Å⁻¹. Indexation yields a two-dimensional unit cell with lattice constants of a = 7.82 Å and b = 5.54 Å.

Since for quinquethiophene a full solution of the crystal structure does not exist, we compare the lattice constants with those derived from the sexithiophene crystal structure. Sexithiophene crystallizes in a herringbone packing with a lateral periodicity of a = 7.85 Å and b = 6.03 Å. The perpendicular distance between two neighboring sexithiophene molecules can be calculated from the lattice constants when taking into account the tilt angle of 23° in the direction of the *b*-axis. Perpendicular distances of 7.85 Å and 5.55 Å are obtained. These values have an excellent agreement with the lattice constants from the two-dimensional crystals within the SAM where the molecules are oriented perpendicular to the substrate. In contrast, packing of up-right standing aliphatic chains would result in a much smaller crystallographic unit cell of 7.4 Å and 4.93 Å obtained using values taken from polyethylene. Based on that comparison, we conclude that the lateral packing of the quinquethiophene units is responsible for the two-dimensional crystals and that the quinquethiophene units pack in a herringbone pattern.

The analysis presented above shows that the SAM is a densely packed ordered monolayer. The chromophores are highly ordered. An amorphous contribution, if any, is discussed below. To that end we look in more detail at lower intensity level around $q_{xy} = 1.5 \text{ Å}^{-1}$ in the grazing incidence diffraction pattern of Fig. S1. A background intensity is observed, that might be due to amorphous scattering of disordered carbon-hydrogen chains.



Fig. S1: Diffraction pattern from a partially covered SAM obtained with grazingincidence x-ray diffraction measurements. The diffracted intensity is presented as a function of the out-of-plane and in-plane scattering vector q_z and q_{xy} , respectively. The lower intensity levels are magnified.

However, amorphous silicon oxide has its halo exactly at the same scattering vector. A separate measurement of a reference substrate without SAM is presented in Fig S2. A background intensity is indeed observed at about 1.5 Å⁻¹. Therefore, we cannot unambiguously assign this intensity to amorphous chromophores. We note however that the SAM contains dominantly highly ordered chromophores, packed in a herringbone crystal structure.



Fig. S2: Grazing incidence diffraction of a thermally oxidized silicon substrate.

[1] Onclin S., Ravoo B.J. & Reinhoudt D. N., Engineering Silicon Oxide Surfaces Using Self-Assembled Monolayers, *Angew. Chem. Int. Ed.* 44, 6282 (2005).