Supporting Information

Tuning Intermolecular Charge Transfer in Donor-Acceptor Two-Dimensional Crystals on Metal Surfaces

Jonathan Rodríguez-Fernández,[†,¶,‡] *Maitreyi Robledo,*[§,‡] *Koen Lauwaet,*[⊥,∥,‡] *Alberto Martín-Jiménez,*[⊥] *Borja Cirera,*[⊥] *Fabián Calleja,*[⊥] *Sergio Díaz-Tendero,*[§] *Manuel Alcamí,*[§] *Luca Floreano,*[▼] *Marcos Domínguez-Rivera,*[▼] *Amadeo L. Vázquez de Parga,*[†] *David Écija,*[⊥] *José M. Gallego,*[Δ] *Rodolfo Miranda,*[†,⊥] *Fernando Martín,*[§,⊥] *and Roberto Otero**[†,⊥]

†Dep. de Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain

§Dep. de Química, Universidad Autónoma de Madrid, Madrid, Spain

⊥Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA-NANO), Madrid, Spain

▼CNR-IOM, Laboratorio TASC, Basovizza, Trieste, Italy

Δ Instituto de Ciencia de Materiales de Madrid (ICMM-CSIC), Madrid, Spain

Phase coexistence

As mentioned in the main text, the deposition of a given D:A coverage ratio on the solid surface leads to the coexistence of several phases. This coexistence is very remarkable when the D:A coverage ratio is relatively far from any of the D:A stoichiometric ratios of the stable phases. For example, Figure S1a shows the coexistence of the 1:1, 1:2 and 1:4 phases for a coverage ratio close to 0.35. On the other hand, when the coverage ratios are close to the stoichiometric ratios, the corresponding stoichiometric phase dominates, occupying above 75% of the surface and with domain sizes than can be as large as 1000 nm^2. This is shown in Figures S1b and c, where the morphology of samples with coverage ratios of 1.1 (b) and 0.27 (c) lead to samples dominated by the 1:1 or the 1:4 phases.

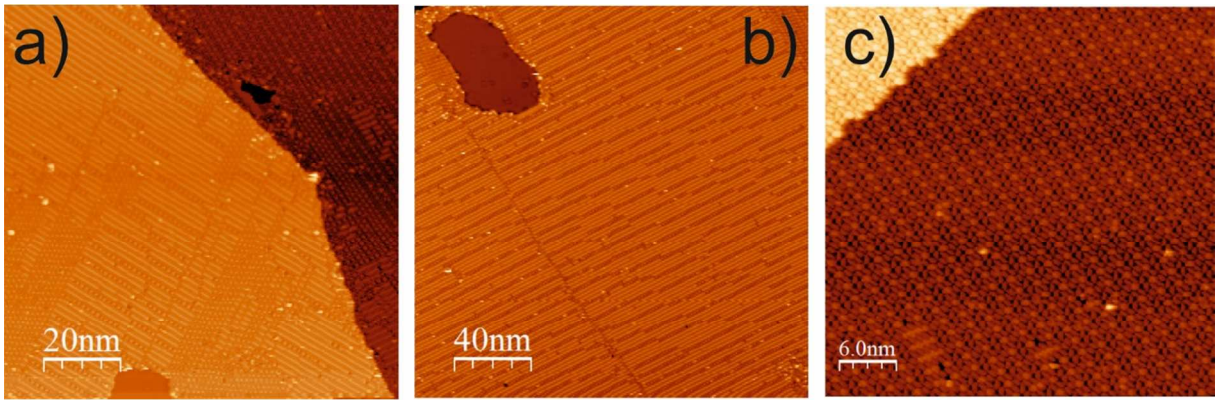

Figure S1. Large scale STM images showing the coexistence of phases for a coverage ratio of 0.35 (a) and the predominance of single phases for coverage ratios of 1.1 (b) and 0.27 (c).