Electronic Supplementary Information

Growth of 1D ClAlPc Molecular Chains Mediated by Graphene Moiré Pattern

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S1. ClAlPc and CuPc molecules on Gr/Rh(110) measured at RT.

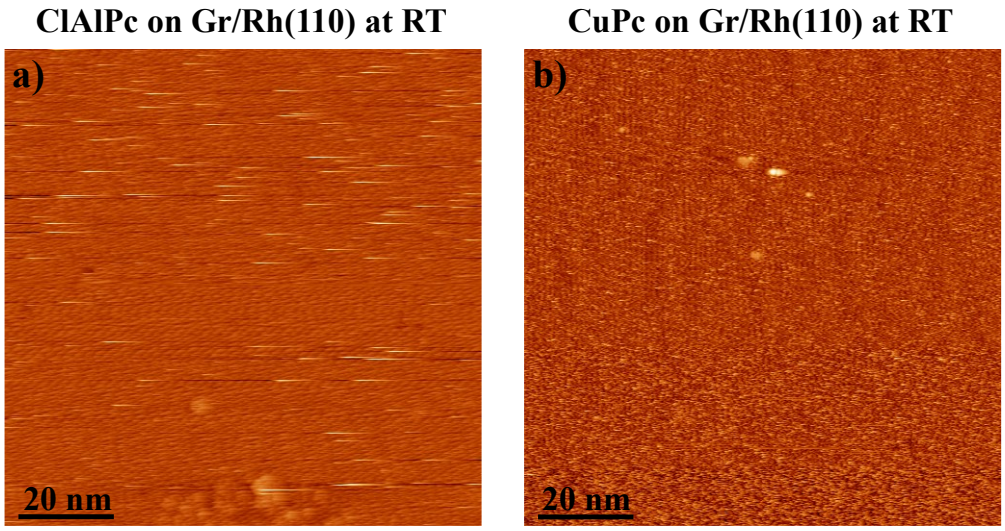

Fig. S1. STM data of a) ClAlPc and b) CuPc molecules on Gr/Rh(110) measured at RT. In both blurred images, ClAlPc and CuPc can not be imaged at RT with STM. Tunneling parameters: a) $V_s = 2.0$ V; $I_t = 20$ pA; 100×100 nm^2. b) $V_s = 2.0$ V; $I_t = 20$ pA; 100×100 nm^2.

S2. Square lattice structure for 1 ML of ClAlPc molecules on Gr/Rh(110) characterized at 40 K.

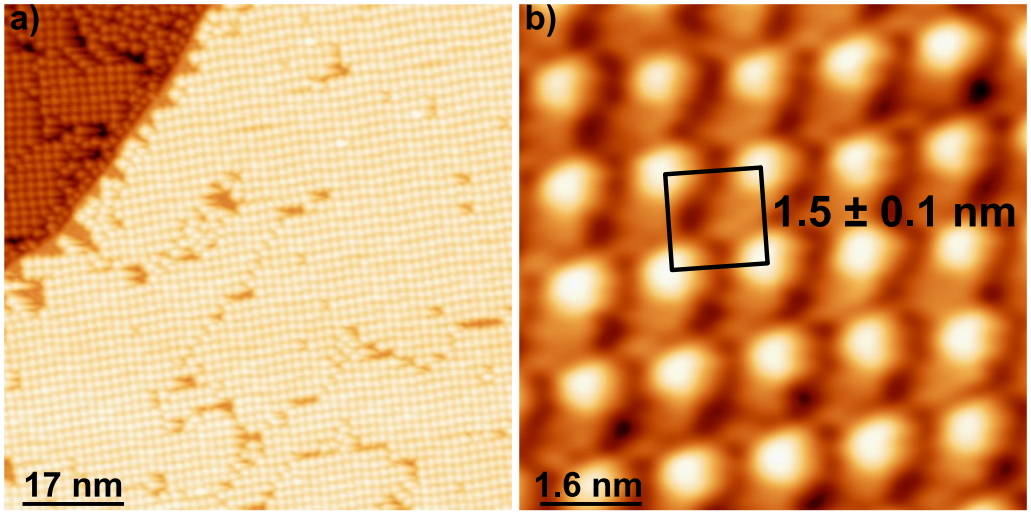

Figure S2. a) Large scale STM image after depositing a molecular coverage close to 1 ML. As can be seen, all the molecules form a close-packed square structure, with translational domains and vacancies. b) Close-up STM image on the square structure of ClAlPc molecules. The black square indicates the structure unit cell, and with a cell parameter of 1.5 ± 0.1 nm. Tunnelling parameters: a) V_s = 2.0 V; I_t = 20 pA; 85 × 85 nm^2. b) V_s = 2.0 V; I_t = 20 pA; 8 × 8 nm^2.

S3. ClAlPc molecular transition from 1D to 2D network characterized at 40 K.

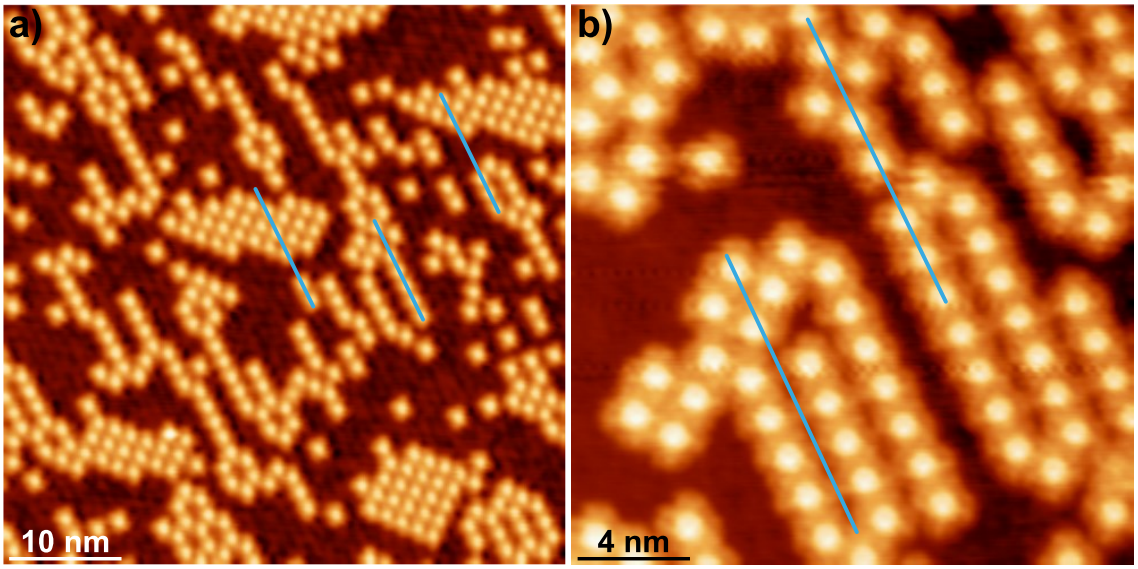

Figure S3. a-b) STM images showing the coexistence of molecular chains and square structures in a same region. As observed, the ClAlPc chains are roughly aligned with the diagonal direction of the square lattice (see the blue lines in the images). Thus, when the molecular coverage is high enough, the parallel molecular chains are joined to fold down into the square lattice. Tunnelling parameters: a) $V_s = 2.0$ V; $I_t = 40$ pA; 50×50 nm^2. b) $V_s = 2.0$ V; $I_t = 80$ pA; 20×20 nm^2.

Figure S4. ClAlPc molecular disordering on rotated domains of Gr/Rh(110) characterized at 40 K.

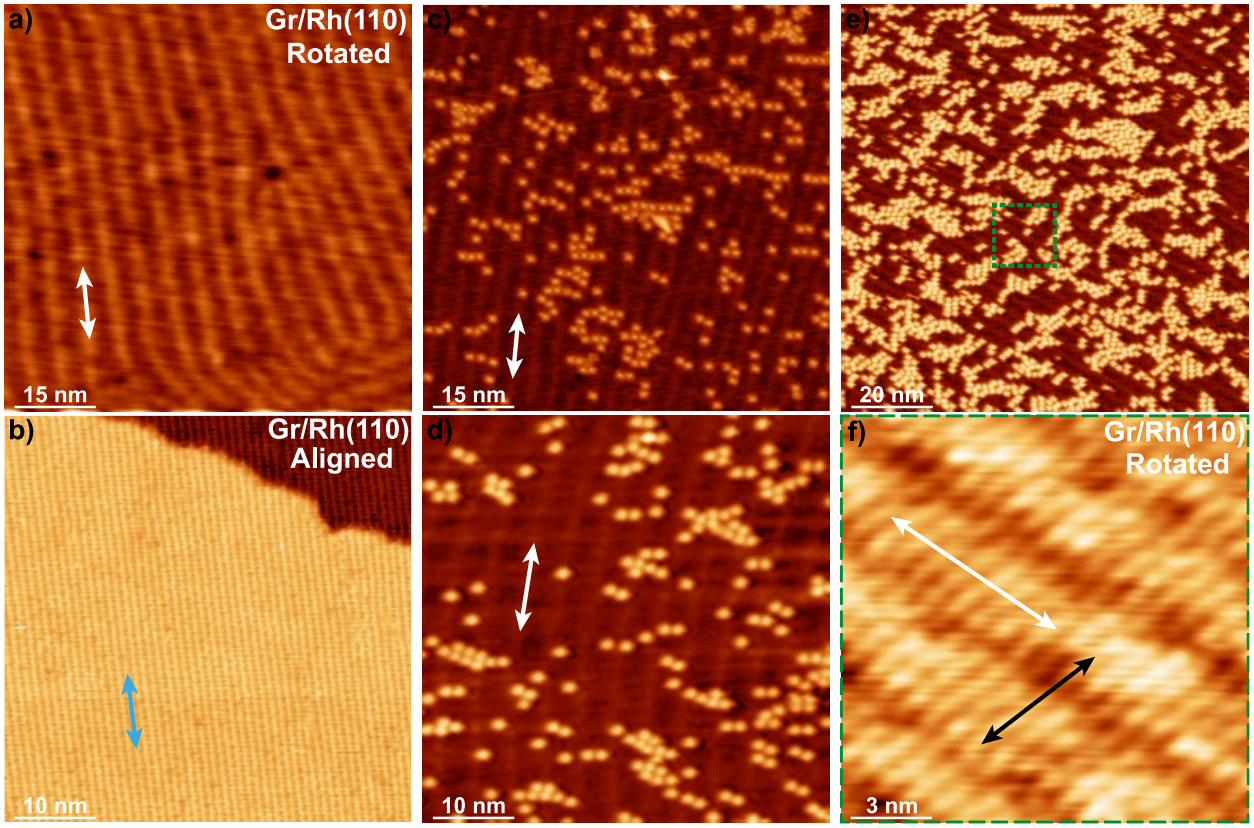

Figure S4. a-b) Representative STM images of pristine Gr/Rh(110) substrate before ClAlPc molecules deposition. The twisted lines in (a) represent a second set of moiré stripes (with a typical periodicity of ~ 4-6 nm) associated with domains, where graphene layer is rotated with respect to Rh(110) [1]. In contrast, the aligned graphene (panel b) features only one set of moiré stripes (periodicity ~ 1 nm). c-d) Set of STM images after depositing ClAlPc molecules on Gr/Rh(110). As can be observed, the molecules are distributed randomly on the surface, forming small clusters or short chains. In all the images, the second set of moiré stripes is observed, indicating that the molecules are placed on a rotated graphene layer. e) STM image acquired on a sample with higher ClAlPc coverage on Gr/Rh(110), compared to (c) and (d), where the molecules are still disordered. f) Underlying Gr/Rh(110) surface measured within the dashed blue square in (e). Two sets of moiré stripes can be observed (white and black lines indicate the two sets of moiré stripes), which is typical of a rotated domain of graphene on Rh(110). To access the substrate, the molecules were removed by scanning the region at high I_t and low V_s tunnelling values, following a similar procedure described previously [2]. Tunnelling parameters: a) V_s = 2.0 V; I_t = 0.24 nA; 75 × 75 nm^2. b) V_s = 0.84 V; I_t = 0.78 nA; 50 × 50 nm^2. c) V_s = 2.0 V; I_t = 20 pA; 75 × 75 nm^2. d) V_s = 2.0 V; I_t = 20 pA; 50 × 50 nm^2. e) V_s = 2.0 V; I_t = 20 pA; 100 × 100 nm^2. f) V_s = 0.64 V; I_t = 0.51 nA; 15 × 15 nm^2.

References

(1) A. J. Martínez-Galera, H. Guo, M. D. Jiménez-Sánchez, E. García Michel and J. M. Gómez-Rodríguez, *Carbon*, Submitted 2022, http://dx.doi.org/10.2139/ssrn.4146274.

(2) H. Guo, M. Izquierdo-Cid, M. D. Jiménez-Sánchez, A. J. Martínez-Galera and J. M. Gómez-Rodríguez, *Adv. Mater. Interfaces*, 2021, **8**, 2101389.