Electronic Supplementary Information (ESI)

Electrochemistry-driven molecular tiling of a charged polycyclic aromatic compound

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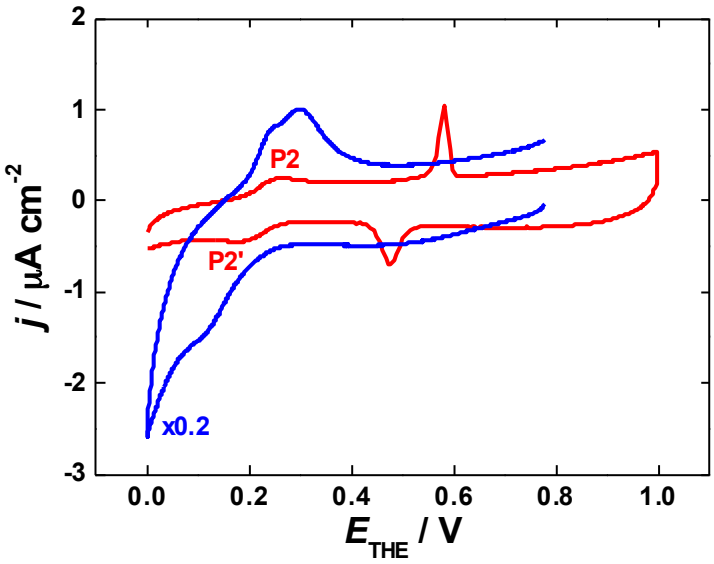

Figure S1. Cyclic voltammogram of $(PQPC_6)_2$AQDSA-modified Au(111) in 0.1 M $HClO_4$ (red trace, identical to Fig. 1 in main text; scan rate 10 mV s^{-1}), and of *ca.* 1 mM AQDSA + 0.1 M $HClO_4$ (polycrystalline Au, scan rate 50 mV s^{-1}, blue trace), identifying P2/P2' as related to anthraquinone electrochemistry. The larger peak separation and double peak in the second measurement may originate from contributions from both adsorbed and solution-based AQDSA, but centres around the same formal potential as P2/P2'.

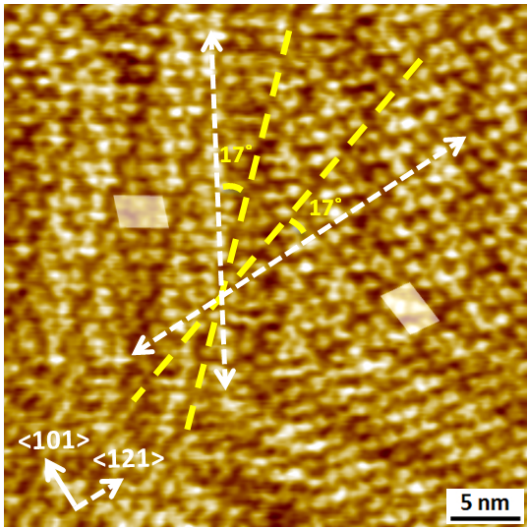

Figure S2. Mirror image domains within the hexameric adlayer. E_s = 0.3 V; U_{bias} = –0.65 V; I_t = 1 nA.

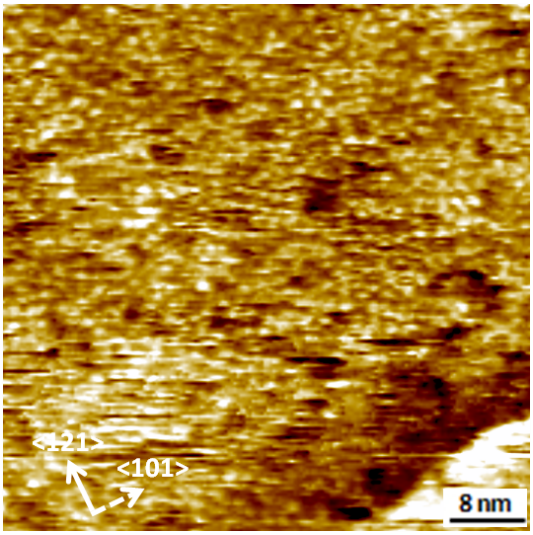

Figure S3. EC-STM image of amorphised $(PQPC_6)_2$AQDSA adlayer on Au(111) in 0.1 M $HClO_4$ at E_s = 0.8 V. U_{bias} = –0.65 V; I_t = 1 nA.

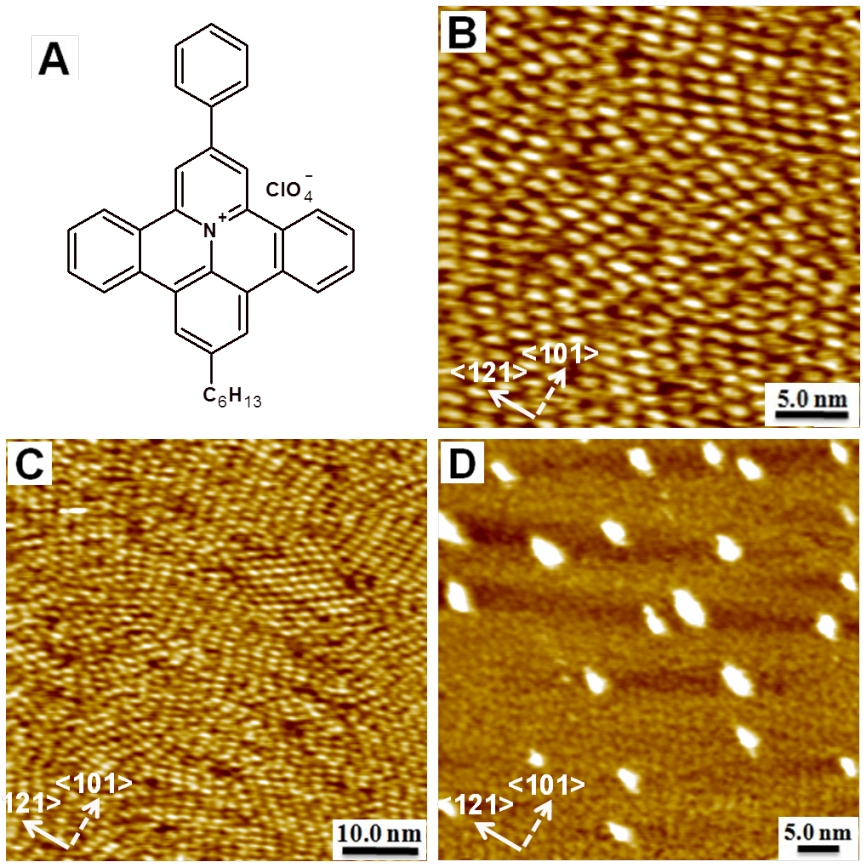

Figure S4. Preliminary results indicating anion effects in supramolecular ordering of PQP-based compounds: (A) Structural formula of $PQPC_6ClO_4$ (note that the cation is identical as discussed in the main text). (B, C) EC–STM images of $PQPC_6ClO_4$-modified Au(111) electrode in 0.1 M $HClO_4$, demonstrating that only short-range order is obtained. E_s = 0.45 V; U_{bias} = –0.6 V; I_t = 1.0 nA. (D) Idem, with E_s = 0.8 V.