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Supporting information

Potential-Selective Fate of 2D Chiral Crystallization on an Electrode Surface

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Figure S1. (a-b) Low- and high-resolution STM images collected with TTE – modified Au(111) electrode at 0.2 V in 0.1 M HClO<sub>4</sub>. Extended lines running in the <121> direction of the Au(111) substrate are ascribed to chains of TTE admolecules. Elongated protrusions are due to TTE admolecules standing on the Au(111) substrate with their S-ends of the dithiophene units, as revealed by the ball model shown in panel (c). This sample is prepared by immersion in 8  $\mu$ M TTE benzene dosing solution for 30 sec.



Figure S2. In situ STM image showing the hexagoanl array displacing the Au(111) -  $(7 \times 14)$  – TTE structure 3 min after the potential is changed from 0.5 to 0.2 V in 0.1 M HClO<sub>4</sub>. This structure is observed on the Au(111) –  $(1 \times 1)$  substrate. TTE admolecules are aligned in the <121> directions of the Au(111) substrate with a nearest neighbor spacing of ~12 Å.



Figure S3: (a-c) In situ STM images showing the Au(111) -  $(2\sqrt{3} \times \sqrt{13})$  – TTE structure observed at 0.2 V in 0.1 M HClO<sub>4</sub>. A molecular model is superimposed on the STM image to illustrate the TTE adsorption orientation. The brighter spot marked in dotted circle is assumed to stem from the thiophene unit tilting away from the Au(111) substrate. This corrugation pattern indicates an all S-TTEs domain. R and S –TTE domains are found on the same terrace, which have two opposite angles (30°) with respect to the main axis of Au(111) electrode, indicated by the dotted line (c).



Figure S4. (a) A high-resolution STM scan showing the zigzagged chains of the Au(111) -  $(7 \times 14)$  - TTE structure residing at the lower section of panel (b). As the intermediate structure (the upper half of (b)) gradually transforms into the  $(7 \times 14)$  - TTE structure (lower half), two patched  $(7 \times 14)$  - TTE structures (in the highlighted ovals) seen in the last STM scan (Fig. 4d) disappear. This STM scan is recorded 30 s after Fig. 4d. The white line marked in panel (b) denotes the initial boundary between two different TTE structures. These images are recorded at 0.5 V in 0.1 M HClO<sub>4</sub>. The close-packed atomic directions of the Au(111) substrate are indicated by arrows in panels a and b.