

## Supporting information

### Controlling Two-Phase Self Assembly of an Adenine Derivative on HOPG via kinetic effects

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#### Experimental section

##### Synthesis and Characterization of Adenine Derivative A-C22.

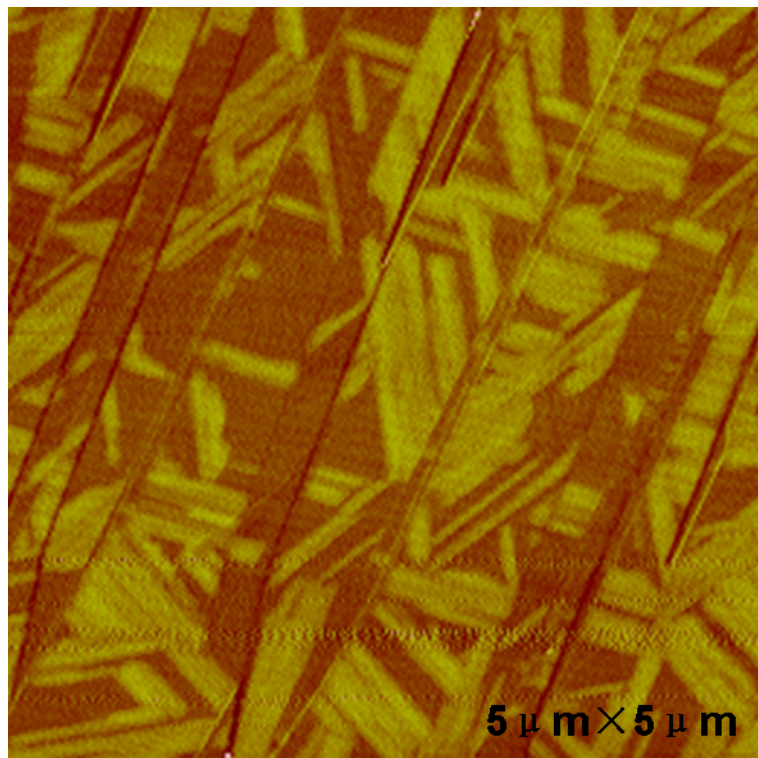
Commercially available reagents were used as received. NMR spectra were recorded with a Varian 500 MHz INOVA (1H, 500MHz; 13C, 126 MHz) and a Varian UNITYplus 600 (1H, 600 MHz; 13C, 151 MHz). For the determination of melting points, a DSC Q 20 (TA Instruments) and a DSC 2910 CE (TA Instruments) were employed. Mass spectra were recorded using a MAT8200 (Thermo-Finnigan-MAT) for ESI spectra or a LAZARUS IIIDE (Institute of Organic Chemistry, Muenster, Germany) for MALDI measurement. Elemental analyses were performed using a CHNO-Rapid (Foss-Heraeus).

##### Scanning Tunneling Microscopy (STM).

STM investigations were performed by using a commercial multimode Nanoscope III scanning tunneling microscope (Digital Instrument Co., Santa Barbara, CA) with mechanically cut Pt/Ir (90:10) tips at ambient temperature. The images shown were recorded in constant-current mode if not indicated otherwise. For measurements at the air–substrate interface, the adenine derivative deposited to a freshly cleaved surface of highly orientated pyrolytic graphite (HOPG, MaTeck GmbH). Experimental conditions are given in the corresponding figure captions. Different tips and samples were used to check for reproducibility and to ensure that there are no image artifacts caused by the tips or samples. Measurements obtained from STM images are corrected against the substrate lattice parameters obtained from HOPG images. Flattening of the images was carried out to compensate for the tilting of the substrate and scan line artifacts, and a low-pass-filtered transform was employed to remove scanning noise in the STM images.

**Figure S1**

This is the AFM phase image of A-C22 evaporated onto graphite surface. We could see that the molecules form very large-scaled films on the micron level. Under the experimental conditions, the almost-full single-layer molecular film could be prepared after evaporation of 20 minutes.



AFM phase-image of A-C22 physisorbed onto a graphite surface by evaporation,  $T_{\text{sub}} = 55^{\circ}\text{C}$ ,  $T_{\text{eva}} = 110^{\circ}\text{C}$ .

**Figure S2**

Large - scale STM constant-current image of A-C22 physisorbed onto a graphite surface by evaporation. The narrower stripes labeled with blue arrows correspond to the  $\alpha$  phase; the wider stripes labeled with white arrows to the  $\beta$  phase. 300 nm  $\times$  300 nm,  $V_{\text{bias}} = -1.12$  V,  $I_{\text{set}} = 0.100$  nA.

