

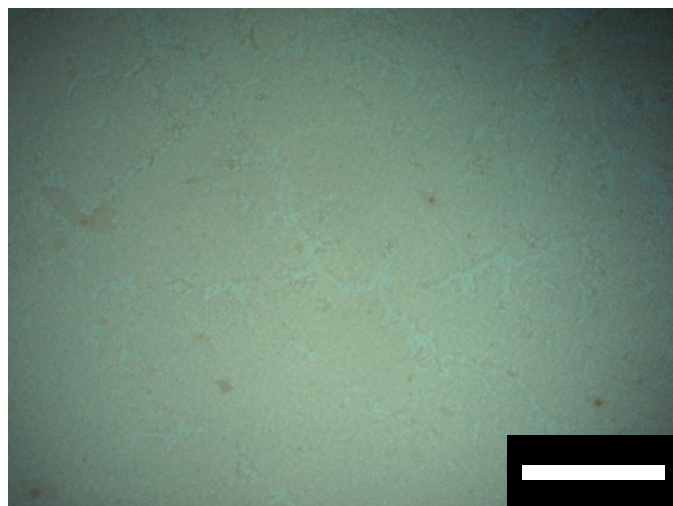
## Supporting Information

### **Directly Writing 2D Organic Semiconducting Crystals for High-Performance Field-Effect Transistors**

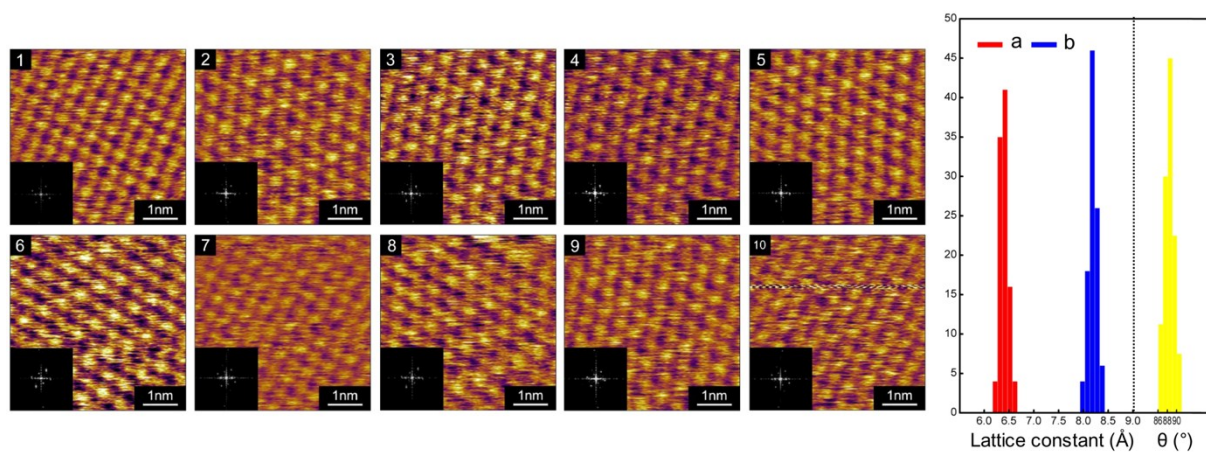
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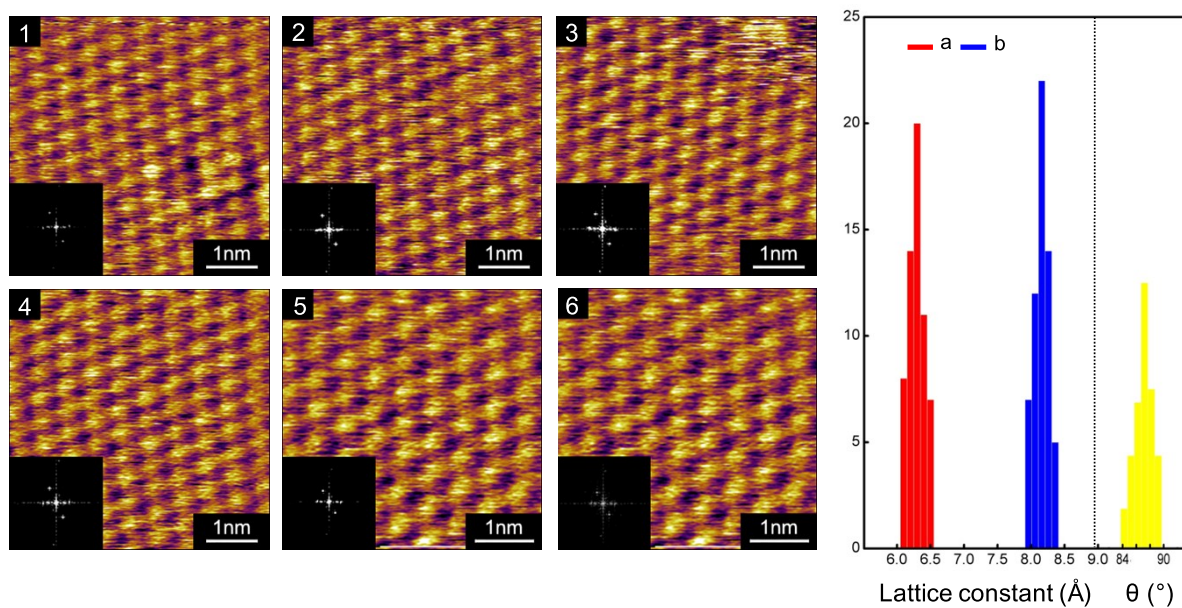
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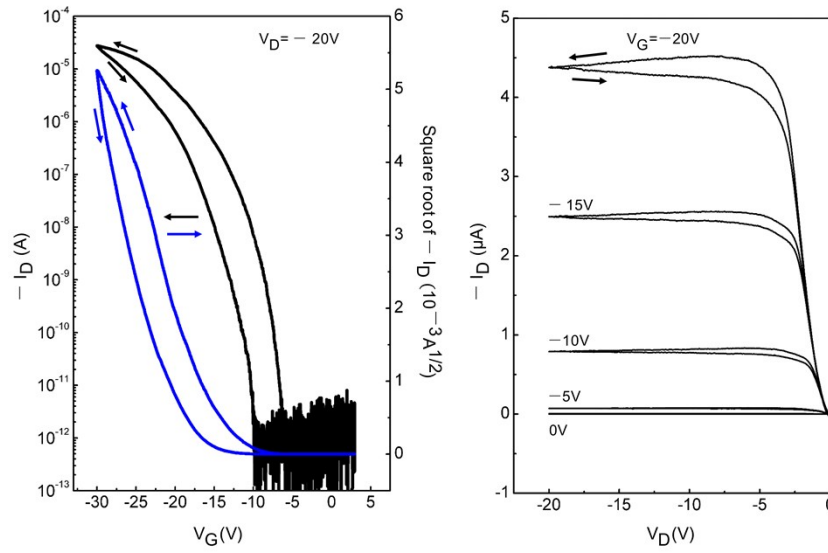
**Figure S1.** The silane of phenethyltrichlorosilane (PhTS) was used for the pretreatment on the  $\text{SiO}_2/\text{Si}$  substrate surface in order to improve its hydrophilicity. The deposited organic layer mainly consists of monolayer molecules. Because of the van der Waals interaction between the C8-BTBT molecules and the substrate, the molecules in the first layer are more horizontally oriented on the substrate. It thus leads to a weak  $\pi$ - $\pi$  interaction among the semiconducting molecules, which is not favourable for the effective charge transport. Scale bar is for 50  $\mu\text{m}$ .



**Figure S2.** Single-crystalline characterizations of 2L C<sub>8</sub>-BTBT molecules. High-resolution AFM image were taken from randomly chosen areas of a 2L C<sub>8</sub>-BTBT film. Histogram of lattice constants for 2L C<sub>8</sub>-BTBT molecules is shown in the right figure.



**Figure S3.** Single-crystalline characterizations of 3L C<sub>8</sub>-BTBT molecules. High-resolution AFM image were taken from randomly chosen areas of a 3L C<sub>8</sub>-BTBT film. Histogram of lattice constants for 3L C<sub>8</sub>-BTBT molecules is shown in the right figure.



**Figure S4.** Transistor characteristics for the 2D C<sub>8</sub>-BTBT crystal with the highest FET device performance. (a) Transfer characteristics at drain voltage of  $-20$  V. And a high carrier mobility of  $5.9 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  was obtained. (b) Output characteristics at various gate voltage.