

## Supplementary Information

### Realizing Low-voltage Operating Crystalline Monolayer Organic Field-effect Transistors with Low Contact Resistance

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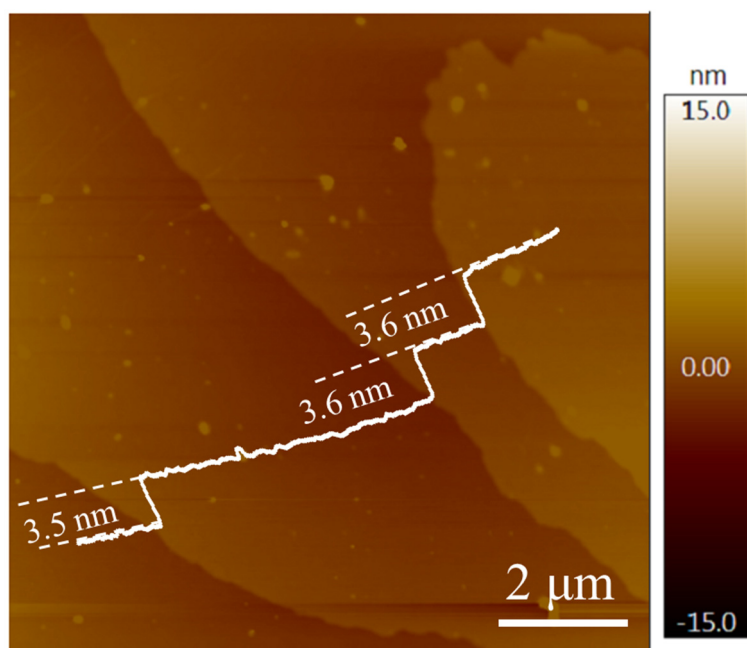
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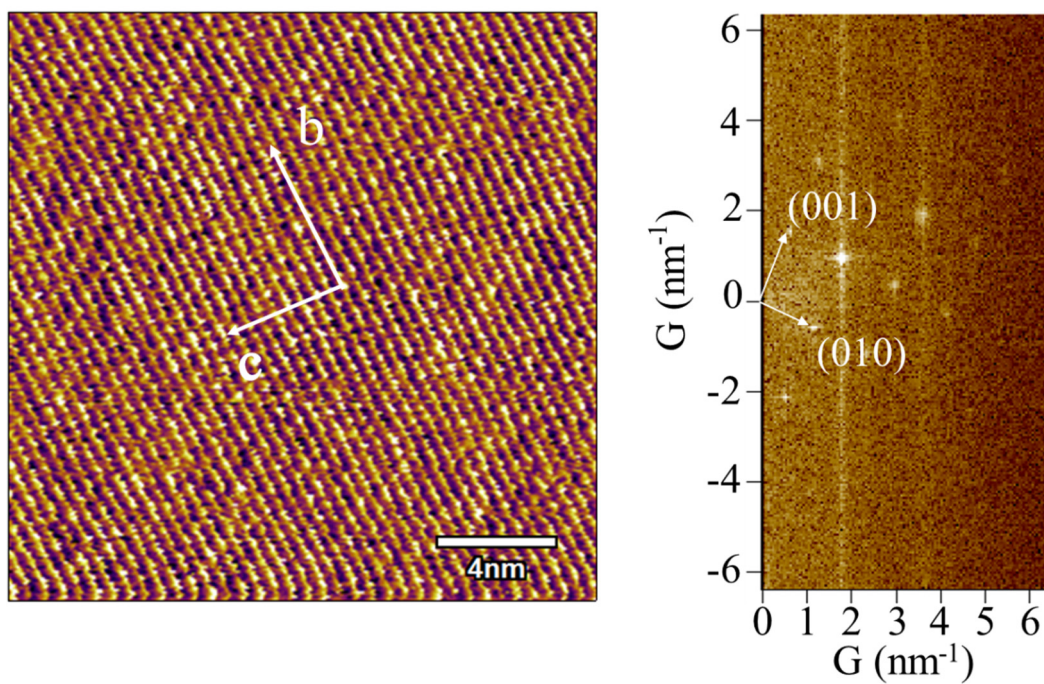
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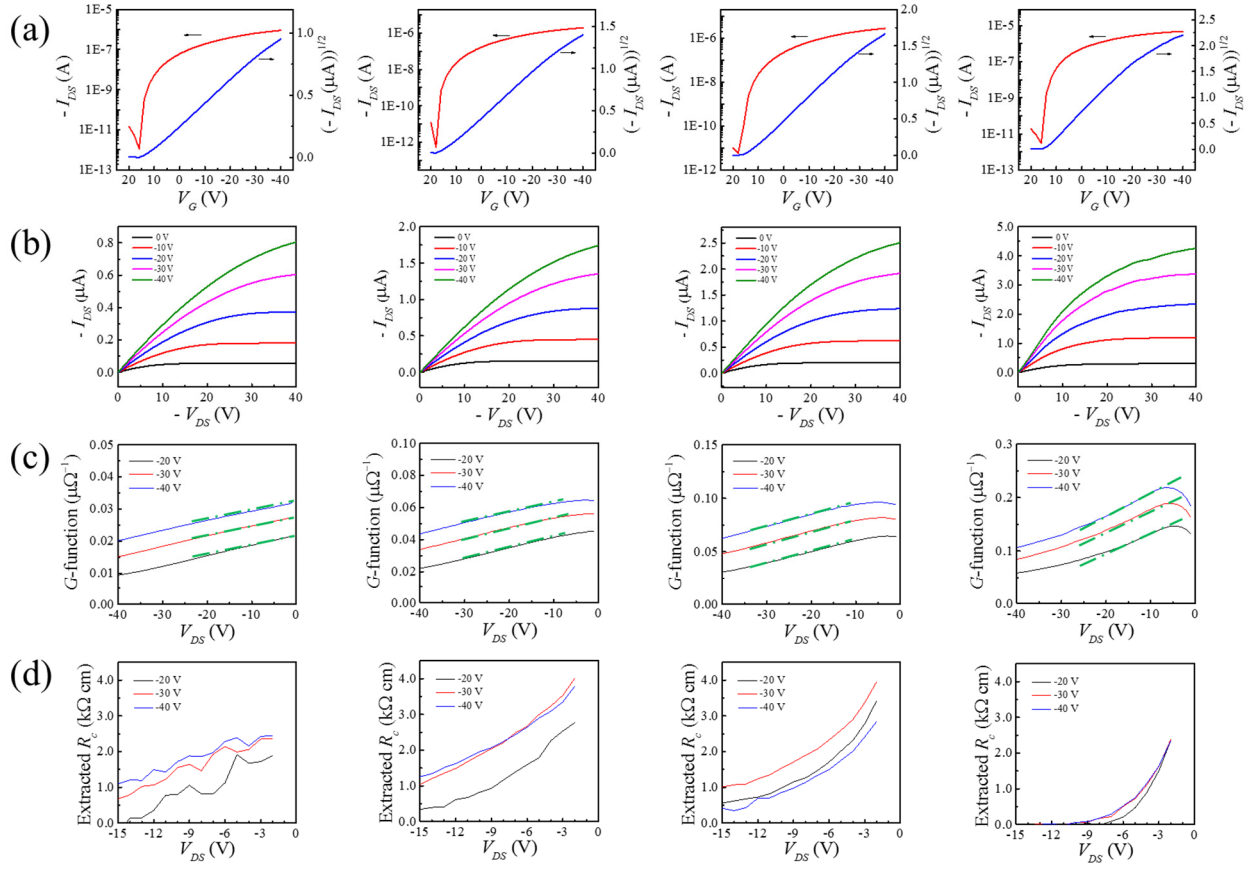
ljjiang@iccas.ac.cn.



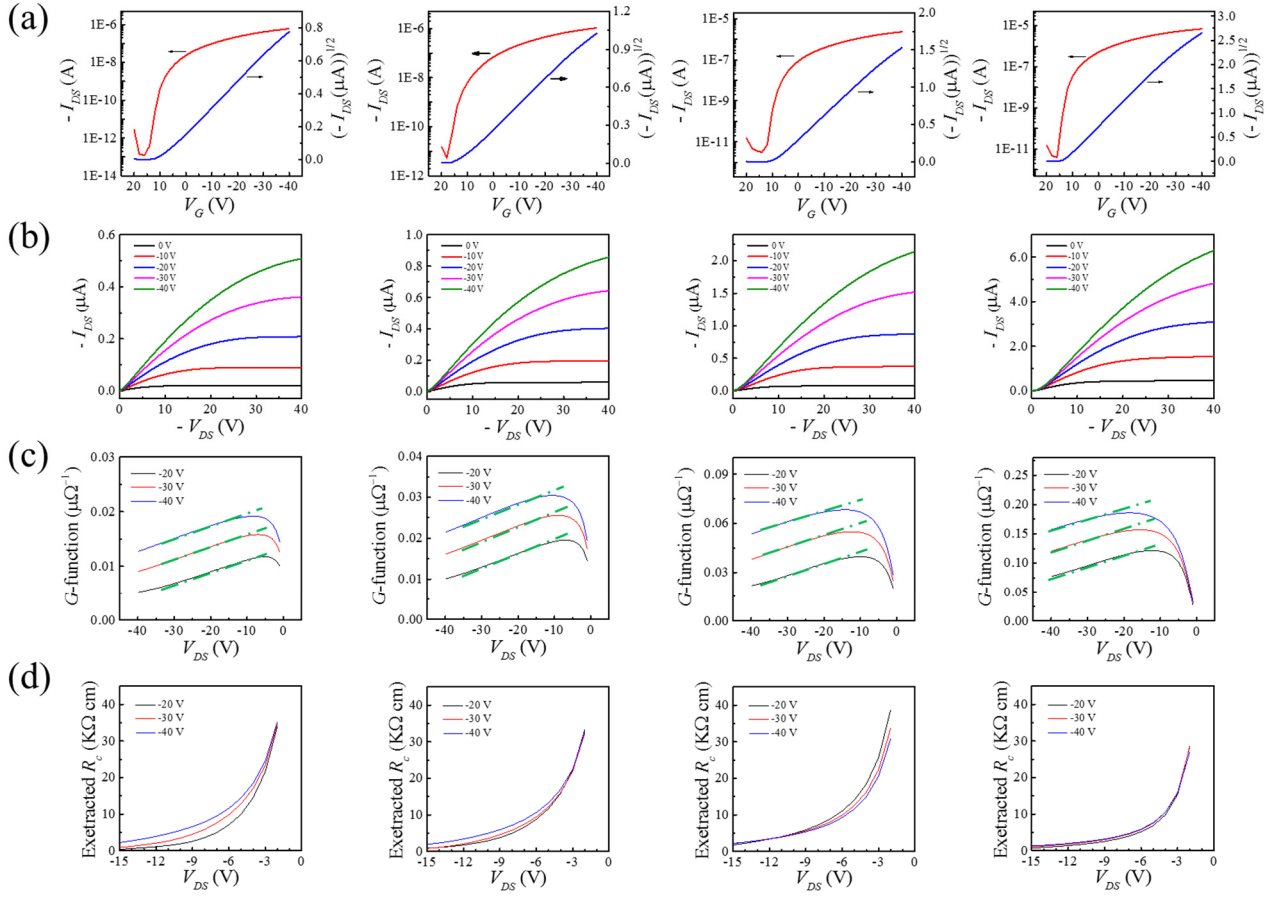
**Figure S1.** AFM image of HTEB 2D molecular crystals on Si/SiO<sub>2</sub> with layer-by-layer structure.



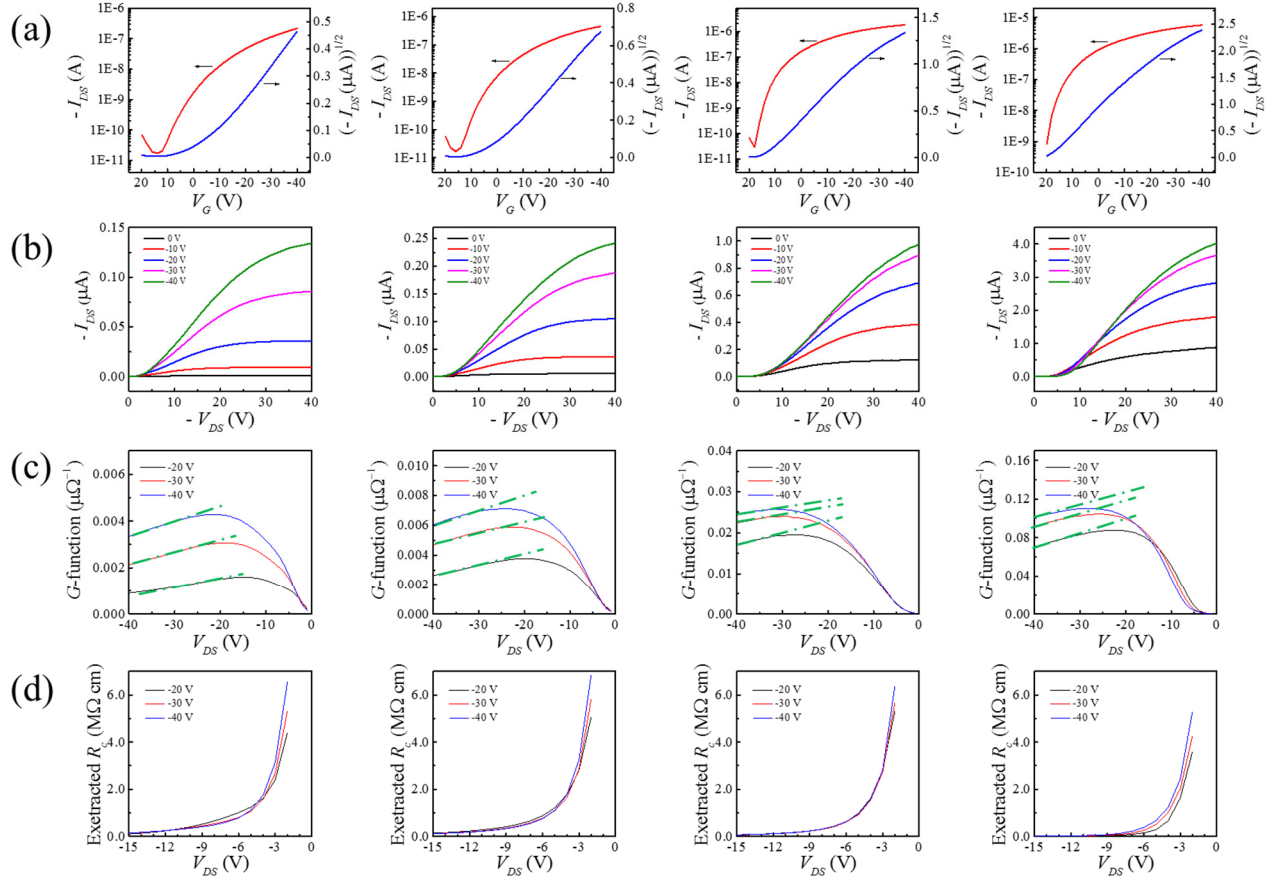
**Figure S2.** HR-AFM image and corresponding 2D FFT pattern of the HTEB multilayer crystal on Si/SiO<sub>2</sub>. Lattice constants were estimated to be  $b = 6.0 \text{ \AA}$ ,  $c = 7.8 \text{ \AA}$  and  $\theta = 85.9^\circ$ .



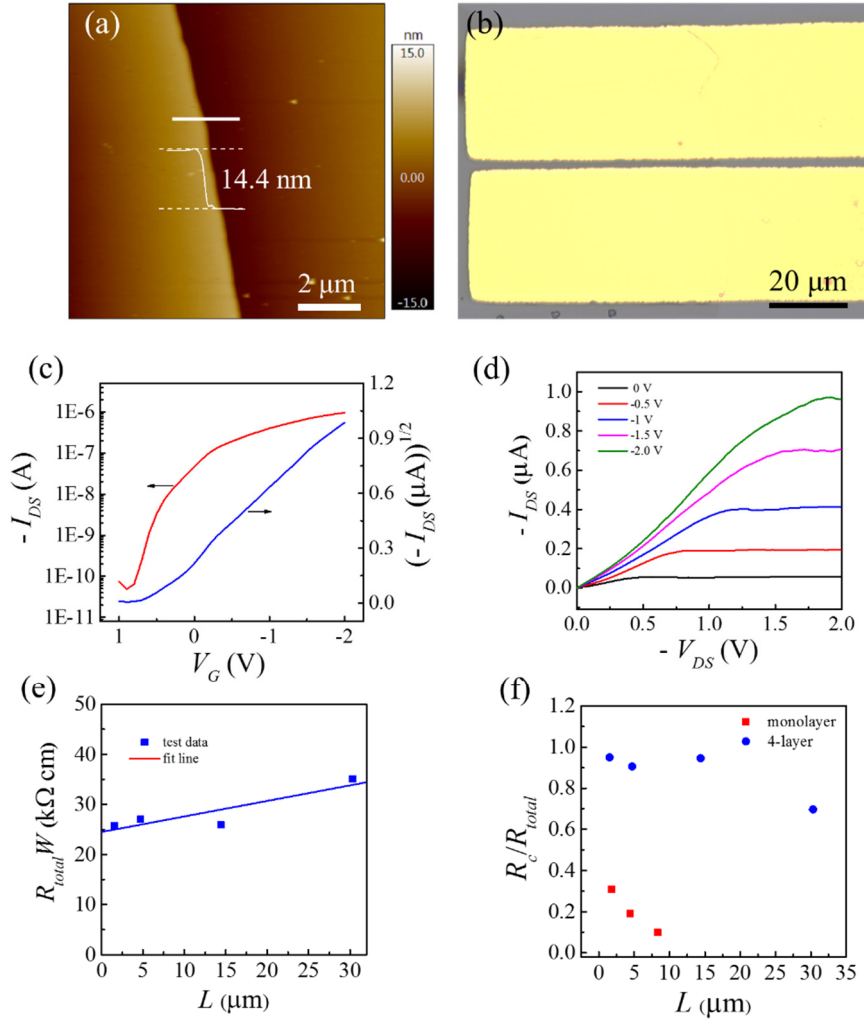
**Figure S3.** (a) Transfer curves, (b) output curves, (c)  $G$ -function curves and (d) extracted  $R_c$  curves of the four HTEB MMC devices fabricated on Si/SiO<sub>2</sub> with channel length of 85.3  $\mu\text{m}$ , 42.9  $\mu\text{m}$ , 28.0  $\mu\text{m}$  and 9.7  $\mu\text{m}$ , respectively.



**Figure S4.** (a) Transfer curves, (b) output curves, (c) G-function curves and (d) extracted  $R_c$  curves of the four HTEB 4-layer single-crystal devices fabricated on Si/SiO<sub>2</sub> with channel length of 86.8  $\mu\text{m}$ , 54.1  $\mu\text{m}$ , 22.3  $\mu\text{m}$  and 9.2  $\mu\text{m}$ , respectively.



**Figure S5.** (a) Transfer curves, (b) output curves, (c)  $G$ -function curves and (d) extracted  $R_c$  curves of the four HTEB 15-layer single-crystal devices fabricated on Si/SiO<sub>2</sub> with channel length of 96.7  $\mu\text{m}$ , 54.8  $\mu\text{m}$ , 27.4  $\mu\text{m}$  and 8.8  $\mu\text{m}$ , respectively.



**Figure S6.** HTEB 4-layer crystal on HfO<sub>2</sub> insulator layer. (a) AFM image of HTEB 4-layer crystal, (b) OM image, (c) transfer curve and (d) output curve of the 4-layer single-crystal OFET with  $L = 1.5 \mu\text{m}$  and  $W = 83.5 \mu\text{m}$ . (e)  $R_{total}W$  as a function of channel length for 4-layer single-crystal OFETs at  $V_G = -2 \text{ V}$ . (f)  $R_c/R_{total}$  as the function of channel length for monolayer and 4-layer single-crystal OFETs.