Supporting Information

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Fig. S1. Large-scale STM images of three chiral patterns of OPV3-CHO; each image contains two homochiral domains with opposite chirality. (a) Windmill structure: $V_{\text{bias}} = 683 \text{ mV}$; $I_t = 657 \text{ pA}$. (b) Chiral linear structure: $V_{\text{bias}} = 687 \text{ mV}$; $I_t = 482 \text{ pA}$. (c) Dense windmill structure: $V_{\text{bias}} = 634 \text{ mV}$; $I_t = 650 \text{ pA}$.



Fig. S2. Self-assembly of achiral linear structure. (a) Large-scale STM image: $V_{\text{bias}} = 650 \text{ mV}$; $I_t = 722 \text{ pA}$. (b) High-resolution STM image: $V_{\text{bias}} = 650 \text{ mV}$; $I_t = 750 \text{ pA}$. (c) Structural model for adlayer. The OPV backbones shift alternatively left or right along the direction "a" indicated by the arrow. Between these bright lines along the *a* direction, there are OPV backbones running roughly perpendicular to the *a* direction to form a network structure. Alkyl chains of OPV3-CHO extend and interdigitate along direction "b." The unit is outlined in the image with the parameters of $a = 4.0 \pm 0.1 \text{ nm}$, $b = 5.9 \pm 0.1 \text{ nm}$, $\alpha = 90 \pm 2^{\circ}$. For the H bondings here, one is 3.1 Å, 174°; another is 3.5 Å, 151°.



Fig. S3. High-resolution STM images and proposed models for OPV3-CHO/C₁₇H₃₅Br in (*a*) and (*b*) and OPV3-CHO/C₁₆H₃₃Br in (*c*) and (*d*). Imaging condition and unit cell parameter in (*a*): $V_{\text{bias}} = 650 \text{ mV}$, $I_t = 609 \text{ pA}$, $a = 1.5 \pm 0.1 \text{ nm}$, $b = 4.0 \pm 0.1 \text{ nm}$, $\alpha = 73 \pm 2^\circ$. Imaging condition and unit cell parameter in (*c*): $V_{\text{bias}} = 806 \text{ mV}$, $I_t = 579 \text{ pA}$, $a = 2.5 \pm 0.1 \text{ nm}$, $b = 3.2 \pm 0.1 \text{ nm}$, $\alpha = 90 \pm 2^\circ$.



Fig. S4. STM images of OPV3-CHO with alkyl bromides on HOPG. (a) OPV3-CHO/C₁₉H₃₉Br: $V_{\text{bias}} = 648 \text{ mV}$, $I_t = 660 \text{ pA}$. (b) OPV3-CHO/C₂₀H₄₁Br: $V_{\text{bias}} = 625 \text{ mV}$, $I_t = 652 \text{ pA}$. (c) OPV3-CHO/C₁₅H₃₁Br: $V_{\text{bias}} = 680 \text{ mV}$, $I_t = 635 \text{ pA}$.



Fig. S5. Self-assembly of OPV3-CHO with other alkyl derivatives. (a) OPV3-CHO/C₁₈H₃₈: $V_{\text{bias}} = 680 \text{ mV}$, $I_t = 600 \text{ pA}$. (b) OPV3-CHO/C₁₈H₃₇OH: $V_{\text{bias}} = 680 \text{ mV}$, $I_t = 630 \text{ pA}$.



Fig. S6. Calculated electrostatic potential of OPV3-CHO using the DFT method in Materials Studio 3.1.