

## Supplementary Information

# Targeted Deposition of Organic Semiconductor Stripes onto Rigid, Flexible, and Three-Dimensional Substrates

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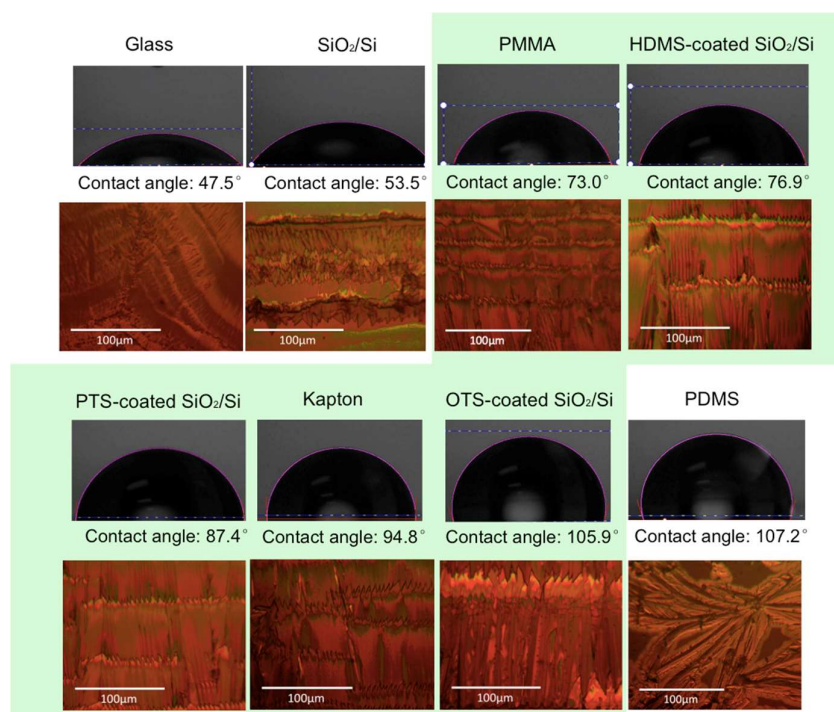
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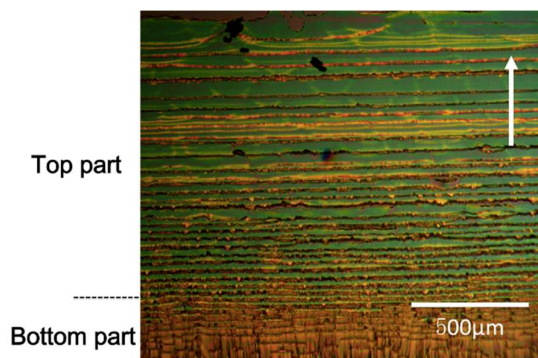
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## Water Contact Angle

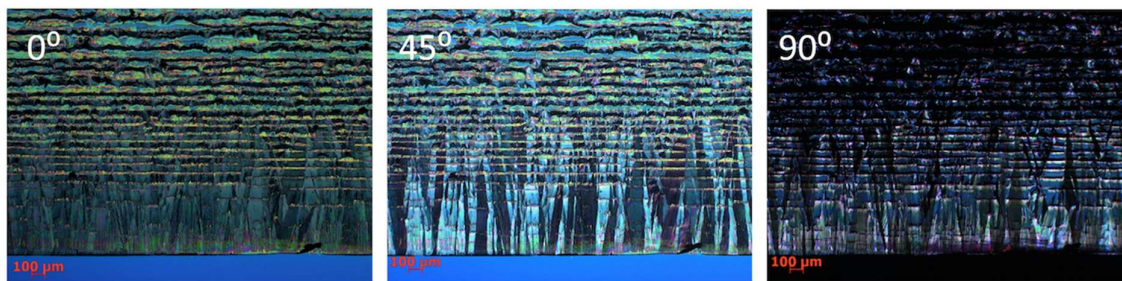


**Figure S1.** Photographs showing the water contact angle (CA) on various substrates and corresponding optical microscope images of TIPS-pentacene stripes grown on the same substrates. The growth of continuous stripes consisting of aligned crystallites occurs within the 70° – 105° CA range (highlighted in green).

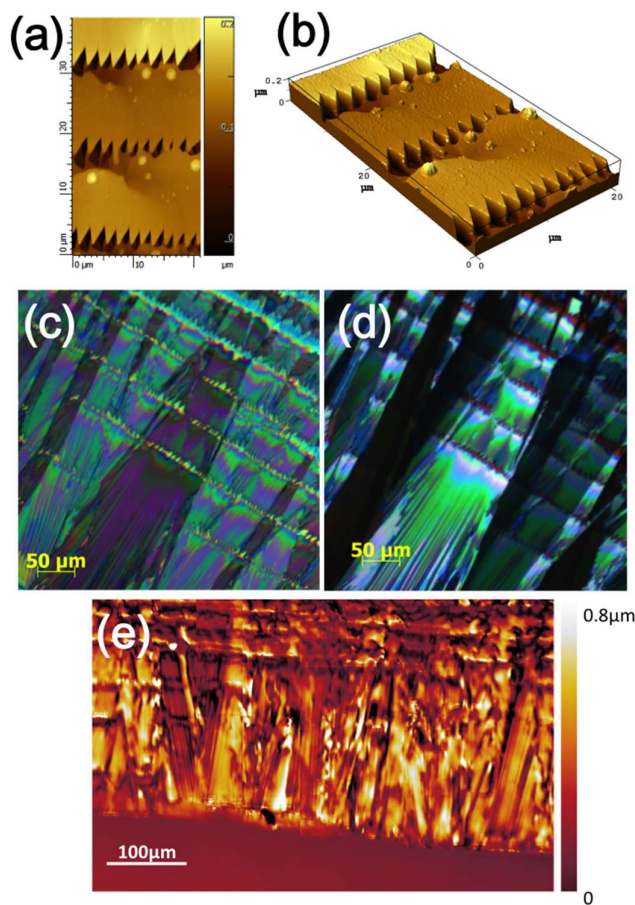
## TIPS-Pentacene Stripe Morphology



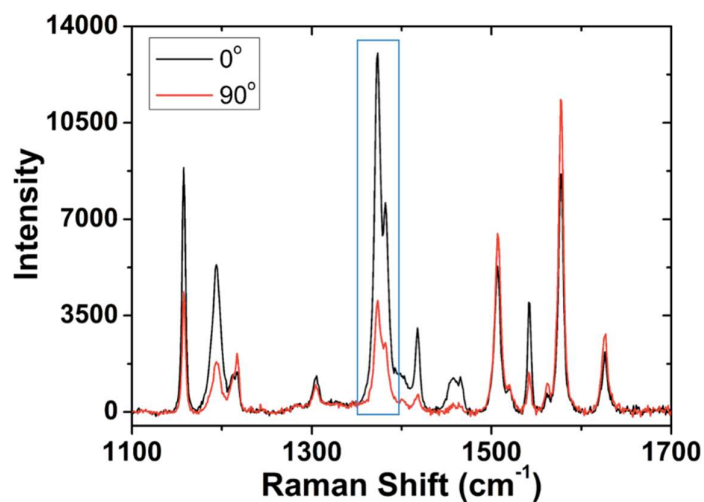
**Figure S2.** Optical microscope image showing the two types of TIPS-pentacene crystallite growth that occur within a deposited stripe, with a continuous morphology at the bottom and a sparser surface coverage at the top (sample prepared using 20 μL of solution).



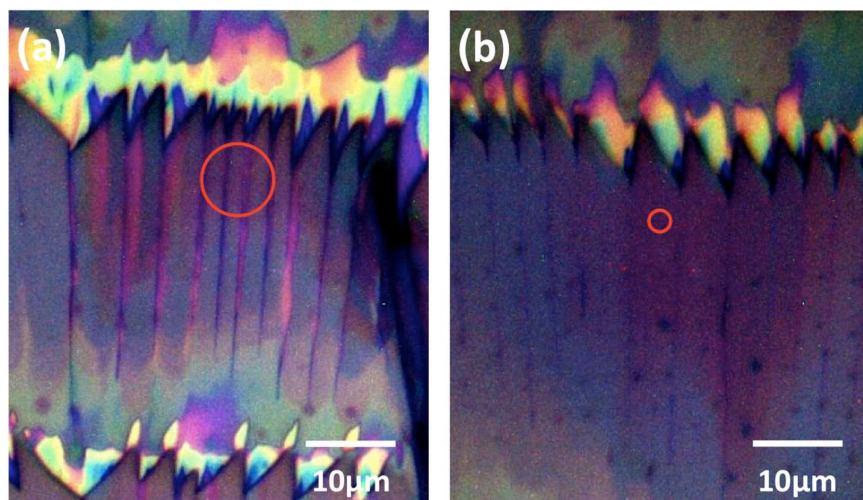
**Figure S3.** Polarized optical microscope images showing that the bottom region of a TIPS-pentacene stripe consists of large crystallites and a continuous morphology, while the top region is less continuous and exhibits smaller crystallites.



**Figure S4.** Atomic force microscope topography of TIPS-pentacene crystallites grown using WM-2PDC: (a) 2D-view and (b) 3D-view. Polarized optical microscope images of a continuous layer of crystallites (bottom part of the stripe) with the polarizer at (c)  $0^\circ$  and (d)  $90^\circ$  relative to the analyzer. (e) Optical profilometer image showing the continuity of the targeted stripe.



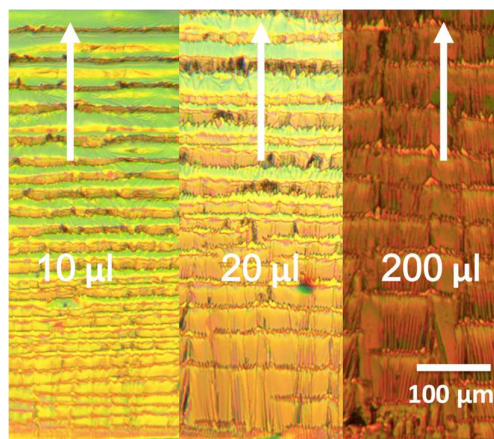
**Figure S5.** Raman spectra acquired at 0° and 90° polarization angles relative to the withdrawal direction for WM-2PDC TIPS-pentacene stripes. The vibrational mode associated with the C-C ring stretching mode (1374 cm<sup>-1</sup>, highlighted above) is used in the main text to track anisotropy.



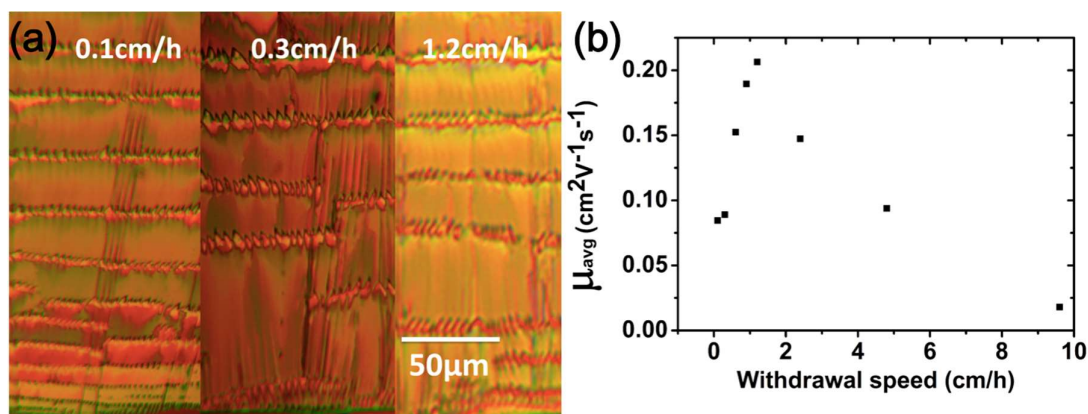
**Figure S6.** Optical microscope images of aligned TIPS-pentacene crystallites with the Raman laser spot size and location illustrated in red on (a) a small group of crystallites (8 μm laser spot diameter) and (b) a single crystallite (2.4 μm laser spot diameter).



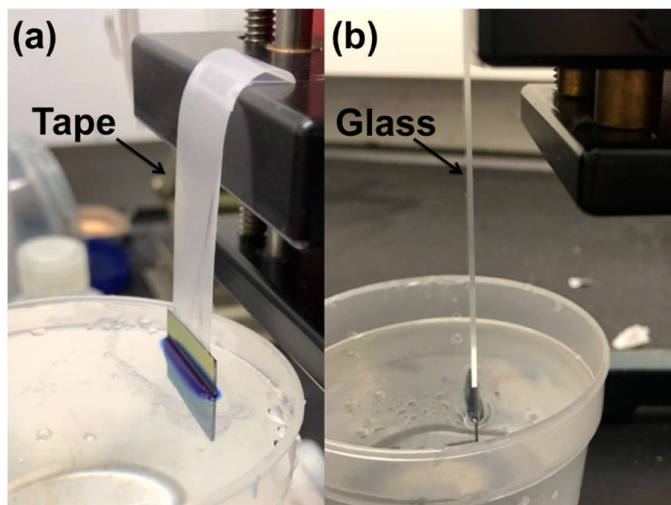
## WM-2PDC Robustness



**Figure S7.** Optical microscope image of TIPS-pentacene crystallites deposited by WM-2PDC using 10 μL, 20 μL and 200 μL of TIPS-pentacene solution. The crystallite morphology is highly similar in the two cases, with the primary difference being the width of the bottom, continuous region of the stripe. The continuous region is 170 μm wide for 10 μL of solution (0.50 μm in average thickness), 260 μm wide for 20 μL of solution (0.65 μm in average thickness) and 990 μm wide for 200 μL of solution (0.75 μm in average thickness).

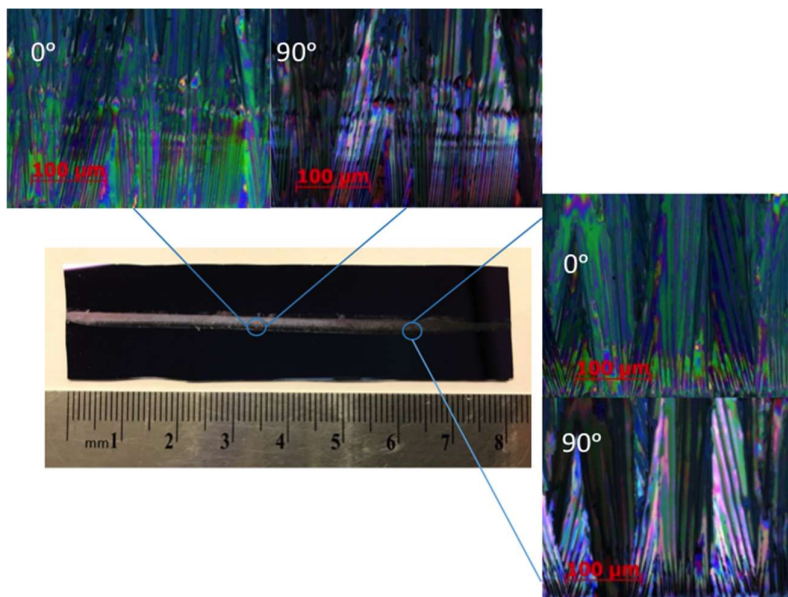


**Figure S8.** (a) TIPS-pentacene crystallites deposited by WM-2PDC using various substrate withdrawal speeds exhibit a consistent crystallite morphology. (b) Over a broad range of substrate withdrawal speeds (e.g. from 0.1 cm/h to 4.8 cm/h), the average hole mobility in TIPS-pentacene organic field effect transistors varies by less than a factor of three (between 0.08 and 0.21  $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ ).

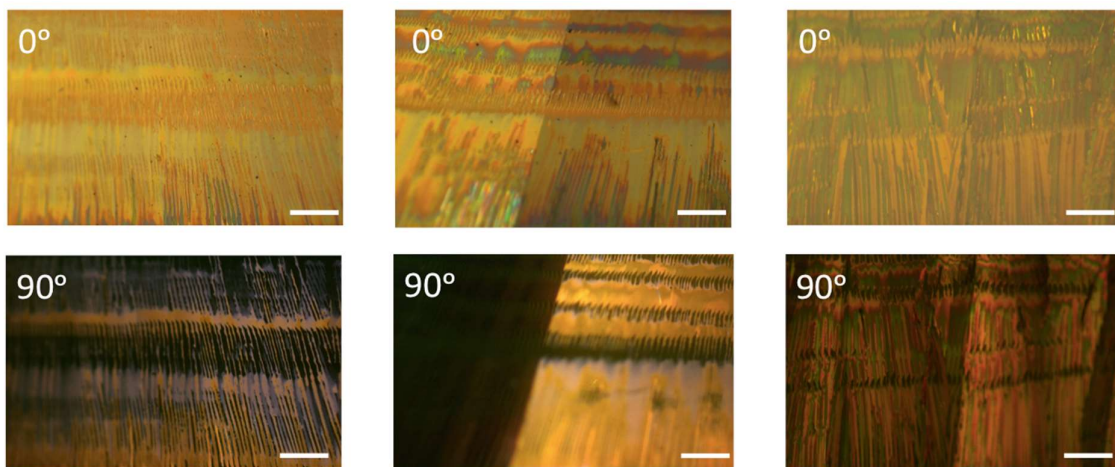


**Figure S9.** Mounting the substrate by (a) a flexible connector (tape) and (b) a rigid connector (glass) during deposition had no observed influence on the deposition process, despite visible motion of the flexible connector due to fume hood air flow.

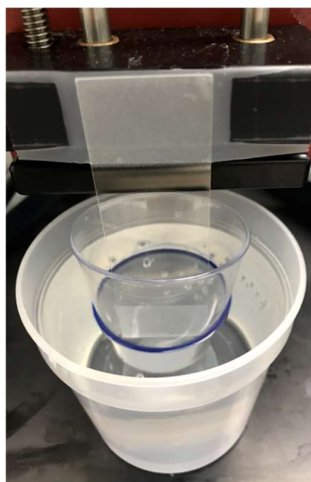
### **Large Areas, Three-Dimensional Substrates, and Multi-Stripe Patterning**



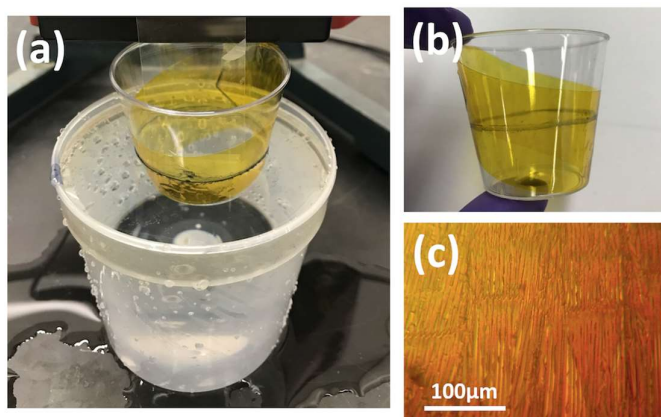
**Figure S10.** An example of WM-2PDC on a long rigid substrate (HDMS-coated  $\text{SiO}_2/\text{Si}$ ). The substrate size is only limited by the area of the reservoir. POM shows aligned crystallites across the 8 cm wide sample.



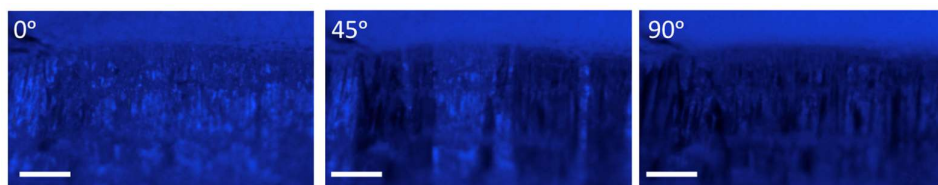
**Figure S11.** POM images of TIPS-pentacene crystallites deposited using WM-2PDC on a 0.8 m long Kapton substrate (scale bar = 100  $\mu\text{m}$ ). The three randomly selected sample locations (about 20 cm away from each other) show that aligned crystallites are obtained along the entire substrate.



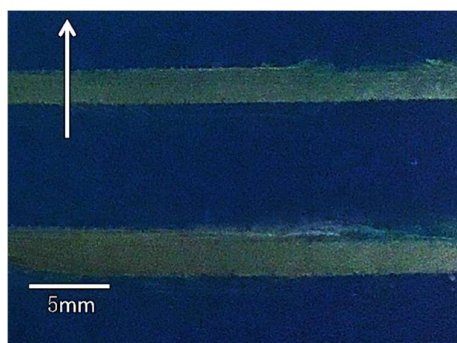
**Figure S12.** WM-2PDC of a TIPS-pentacene stripe onto the outside surface of a cup (corresponding to Figure 6c in the main text).



**Figure S13.** WM-2PDC of a TIPS-pentacene stripe onto a Kapton-covered cup. Microscope image (c), showing aligned TIPS-pentacene crystallites, was taken by peeling the Kapton tape off the cup and re-attaching it to a glass slide (scale bar = 100  $\mu\text{m}$ ).



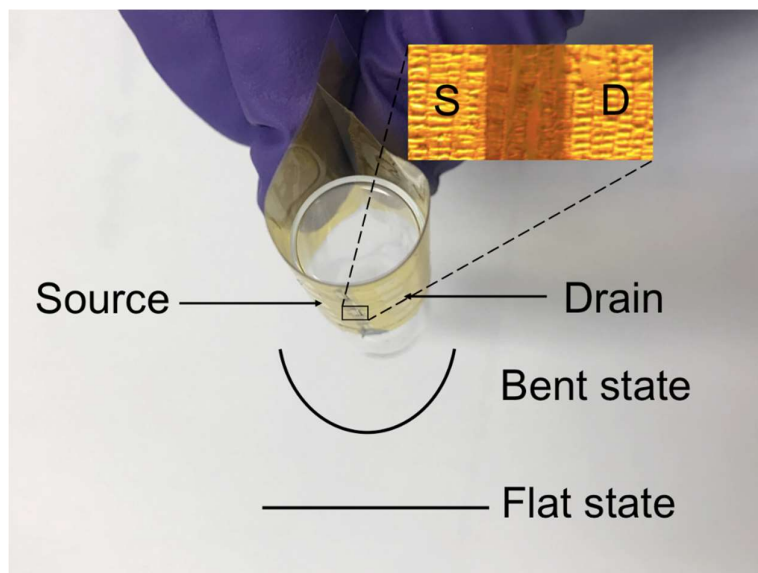
**Figure S14.** POM of TIPS-pentacene stripe on the outside surface of the toy car in Figure 6c (scale bar = 50  $\mu\text{m}$ ). Aligned crystallites can be observed, though the images are less sharp than others due to the non-planar surface of the car.



**Figure S15.** Multi-stripe deposition prepared by sequential solution deposition during substrate withdrawal. The white arrow indicates the withdrawal direction.



## Flexible OFETs



**Figure S16.** Photograph showing the geometry used for bend cycling.