## **Supporting Information**



**Figure S1:** POM images of TIPS-pentacene crystals prepared by zone-casting TIPS-pentacene/toluene solution. The concentrations were (a) 2 mg/ml and (b) 5 mg/ml, respectively. (b1) and (b2) were POM images taken from different zones of the substrate. The casting speed was 50µm/s. Discrete TIPS-pentacene crystals were observed when the concentrate was low (2mg/ml). The continuous crystals with poor orientation were obtained at 5 mg/ml.



**Figure S2:** POM images showing the transition from TIPS-pentacene spherulites to oriented crystalline array. The images were taken from the edge of the substrate, where the zone-casting procedure started. The right images indicated typical POM images of the spherulites and aligned crystals.



**Figure S3:** Typical characteristic transfer curve of OFET based on non-oriented TIPS-pentacene. The insert indicated the POM image of TIPS-pentacene crystals between the source and drain electrodes. The semiconductor layer was prepared by drop-casting TIPS-pentacene/CHCl<sub>3</sub> solution (5mg/ml) on the substrate. The calculated field effect mobility was 0.086 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>.