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Microstructured surfaces have great potentials to improve the performances and efficiency of optoelectronic devices. In this work, a simple robust approach based on surface instabilities was presented to fabricate poly(3-hexylthiophene-2,5-diyl) (P3HT) films with ridge-like/wrinkled composite microstructures. Namely, the hierarchically patterned films were prepared by spin coating the P3HT/tetrahydrofuran (THF) solution on a polydimethylsiloxane (PDMS) substrate to form stable ridge-like structures, followed by solvent vapor swelling to create surface wrinkles with the orientation guided by the ridge-like structures. During spin coating of the P3HT/THF solution, the ridge-like structures were generated by the in-situ template of the THF swelling-induced creasing structures on the PDMS substrate. To our knowledge, it is the first report that the creasing structures are used as a recoverable template for patterning films. The crease-templated ridge-like structures were well modulated by the THF swelling time, the modulus of the PDMS substrate, the P3HT/THF solution concentration and the selective/blanket exposure of the PDMS substrate to O2 plasma. UV–vis and fluorescence spectrometry measurements indicated that the light absorption and fluorescent emission were improved on the hierarchically patterned P3HT films, which can be utilized to enhance the efficiencies of organic solar cells. Furthermore, this simple versatile method based on the solvent swelling-induced crease as the in-situ recoverable template has been extended to pattern other spin-coated films with different compositions.

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