Supporting Information

for

Differentiation of Electric Response in Highly Oriented Regioregular Poly(3-hexylthiophene) under Anisotropic Strain

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1. Method of Calculating Orientation Function (f)

The orientation of P3HT orthorhombic crystals was investigated using mainly the strongest reflection at Bragg angles $2\theta = 5.3^{\circ}$. And Herman's orientation function *f* was calculated according to the following equations^[1,2]:

$$f = \frac{3\cos^2\varphi - 1}{2} \tag{1}$$

where φ is the angle between the normal of base plane and the normal of (100) crystal planes. Before calculating the orientation function, the azimuthal angles β on the two-dimensional detector were corrected to φ , using the following equation:

$$\cos\varphi = \cos\theta\sin\beta \tag{2}$$

Here,
$$\overline{\sin^2 \beta} = \frac{\int_{0}^{\pi/2} I(\beta) \sin^2 \beta \cos \beta d\beta}{\int_{0}^{\pi/2} I(\beta) \cos \beta d\beta}$$
 (3)

where $I(\beta)$ is the azimuthal intensity distribution for (100) reflection.

2. Supporting 2D-GIXRD data

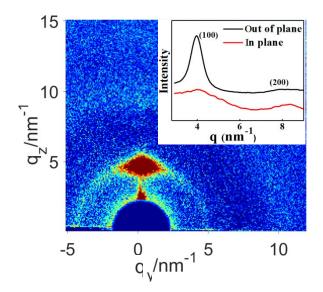


Figure S1. 2D-GIXRD pattern and its corresponding 1D diffraction profile (inset) of a dipcoated P3HT film measured with incident X-ray beam parallel to the dip-coating direction.

3. Supporting conductivity data

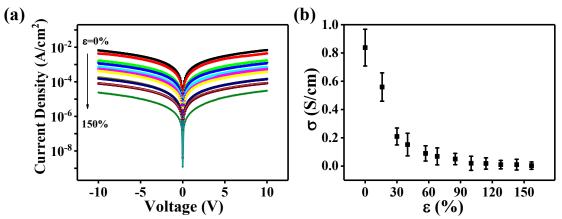


Figure S2. The (a) current density–voltage characteristics and (b) conductivity–strain relationship of non-oriented P3HT film by drop-casting and doped with F_4TCNQ .

4. Supporting POM and AFM data

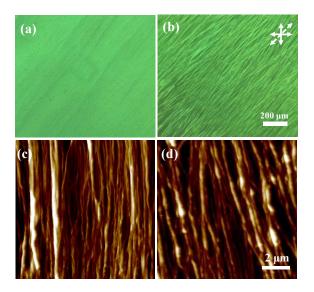


Figure S3. POM and AFM height images of the P3HT bundles on PDMS substrate with thicknesses of (a) & (c) 200 nm for conductivity measurement and (b) & (d) 80 nm for morphology characterization.

(1) Mueller, C. J.; Gann, E.; Singh, C. R.; Thelakkat, M.; McNeill, C. R. Control of Molecular Orientation in Polydiketopyrrolopyrrole Copolymers via Diffusive Noncovalent Interactions. *Chem. Mater.* **2016**, *28*, 7088–7097.

 (2) Zhang, Y.; Leblanc-Boily, V.; Zhao, Y.; Prud'homme, R. E. Wide Angle X-ray Diffraction Investigation of Crystal Orientation in Miscible Blend of Poly(ε-caprolactone)/Poly(vinyl chloride) Crystallized under Strain. *Polymer* 2005, *46*, 8141–8150.