**Supplementary Material**

**Influence of Morphological Disorder on In- and Out-of-Plane Charge Transport in Conjugated Polymer Films**

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**Spano model for UV-vis absorption**

 The absorption spectrum of P3HT can be divided into contributions from two sources: a set of narrow absorption peaks at lower energies from π-stacked aggregates (interchain state), and a broad absorption at higher energies from the amorphous component (intrachain state). Within the limit of weak excitonic coupling, the cofacial interchain interactions of adjacent conjugated segments in P3HT leads to a formation of vibronic bands, which can be determined by applying the Frank-Condon principle according to the equation:

$$A\_{aggregate}∝\sum\_{m=0}^{}\left(\frac{S^{m}}{m!}\right)×\left[1-\frac{We^{-S}}{2E\_{p}}\sum\_{n\ne m}^{}\frac{S^{n}}{n!\left(n-m\right)}\right]^{2}×e^{\frac{\left(E-E\_{0}-mE\_{0}-\frac{1}{2}WS^{m}e^{-S}\right)^{2}}{2σ^{2}}}$$

*A* is the absorption of the aggregates as a function of photon energy *E*, *S* is Huang-Rhys factor, representing the overlap between vibrational states (assumed to be 1), *m* corresponds to different energy levels, and *E*p = 0.179 eV is the energy of the C=C symmetric stretch mode. In this equation, the two fitting parameters of interest are the exciton bandwidth *W* and the energetic disorder *σ* (Gaussian width of the absorption peaks). [References 17, 18 from main text]



**Figure S1** Overlay comparison of absorption spectra of spin-cast and MAPLE-deposited films. Plotted here are the absolute absorbance values of films of equal average thickness (55 nm); the close match of optical absorbances indicates that the films are of similar densities.



**Figure S2** Transfer and characteristics of transistors made from spin-cast and MAPLE-deposited P3HT on OTS-treated substrates.



**Figure S3** Plots of | *I* D|1/2 vs *V*gate for MAPLE and spin-cast P3HT transistors on bare substrates. The solid lines represent the linear regimes from which mobilities are calculated. The subthreshold regime of the MAPLE-deposited sample is significantly larger than that of the spin-cast sample, similar to devices fabricated on OTS-treated substrates.