

## Electronic supplementary information (ESI)

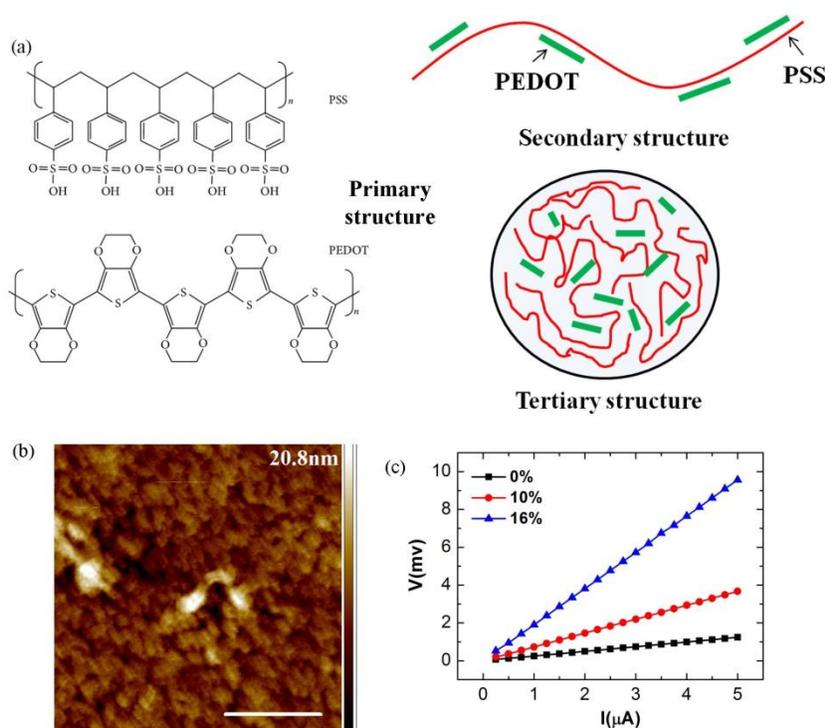
### Wrinkle and crack-dependent charge transport in a uniaxially strained conducting polymer film on a flexible substrate

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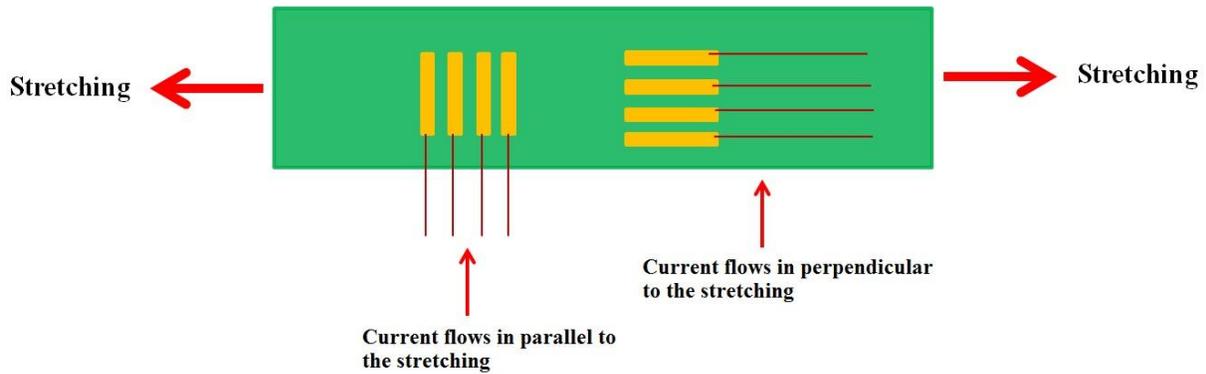
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### Structure, morphology and I-V characteristics of PEDOT:PSS film



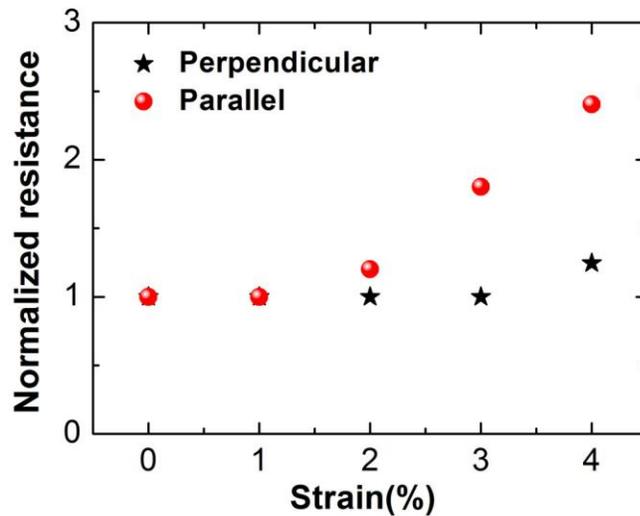
**Fig. S1.** (a) Schematic diagram of primary, secondary and tertiary structure of PEDOT:PSS. (b) Tapping-mode AFM image of pristine PEDOT:PSS film on O<sub>2</sub> plasma treated PDMS (scale bar: 1 μm with color bar: 20.8 nm). (c) I-V characteristics of PEDOT:PSS film on O<sub>2</sub> plasma treated PDMS under different applied strain up to 16%.

Schematic diagram illustrating the current flow for the two geometries considered



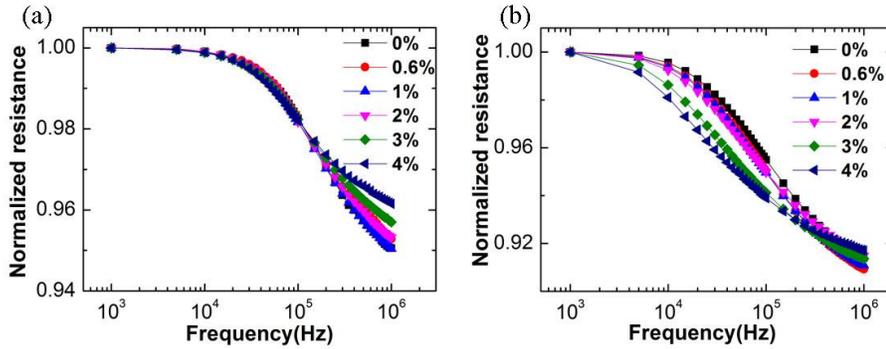
**Fig. S2.** Schematic diagram illustrating the current flow for the two geometries considered (yellow bars are contact electrodes).

**Zoom-in graph of normalized resistance versus strain up to 4%, measured for current being in parallel and perpendicular to the direction of stretching**



**Fig. S3.** Zoom-in graph of normalized resistance versus strain up to 4%, measured for current being in parallel and perpendicular direction to the stretching.

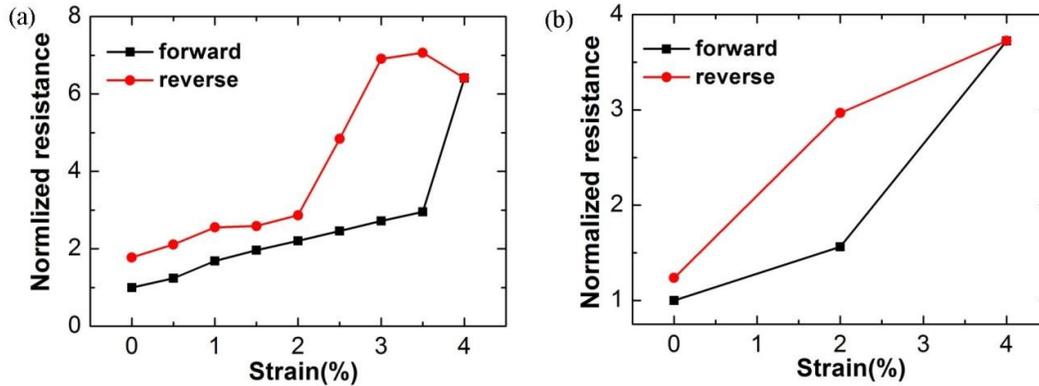
### Frequency-dependent-ac-response of PEDOT:PSS under strain (parallel geometry) on different interfaces



**Fig. S4.** Frequency-dependent ac-response of resistance in PEDOT:PSS film under varying applied strain (parallel geometry) on different interfaces. (a) SAM coated. (b)  $\text{H}_2\text{O}/\text{H}_2\text{O}_2/\text{HCl}$  (10:1:1 v/v ratio) treated PDMS.

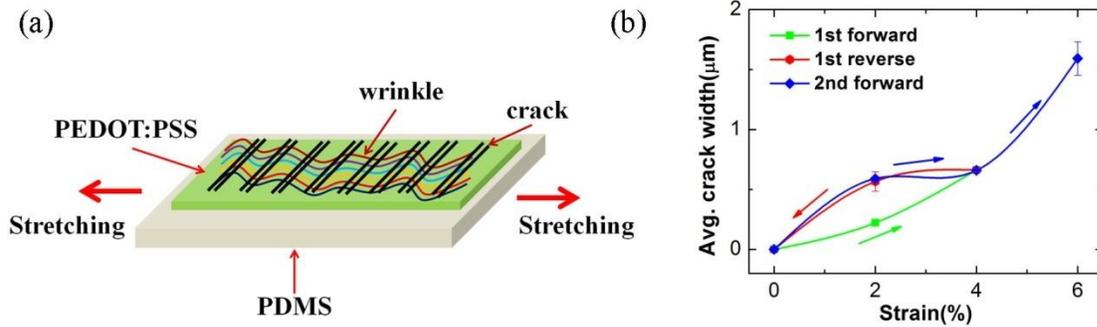
Frequency-dependent ac-resistance in PEDOT:PSS film on two different functionalized PDMS substrates, are plotted in Fig. S4 (a), (b). Characteristic features and key mechanisms of the frequency-dependent ac-transport under strain have already been discussed in the main text. Similar behavior has also been found for PEDOT:PSS film on SAM coated PDMS and also on  $\text{H}_2\text{O}/\text{H}_2\text{O}_2/\text{HCl}$  (10:1:1 v/v ratio) treated PDMS.

### Hysteresis in PEDOT:PSS film at very high ramp rates of applied strain in parallel geometry



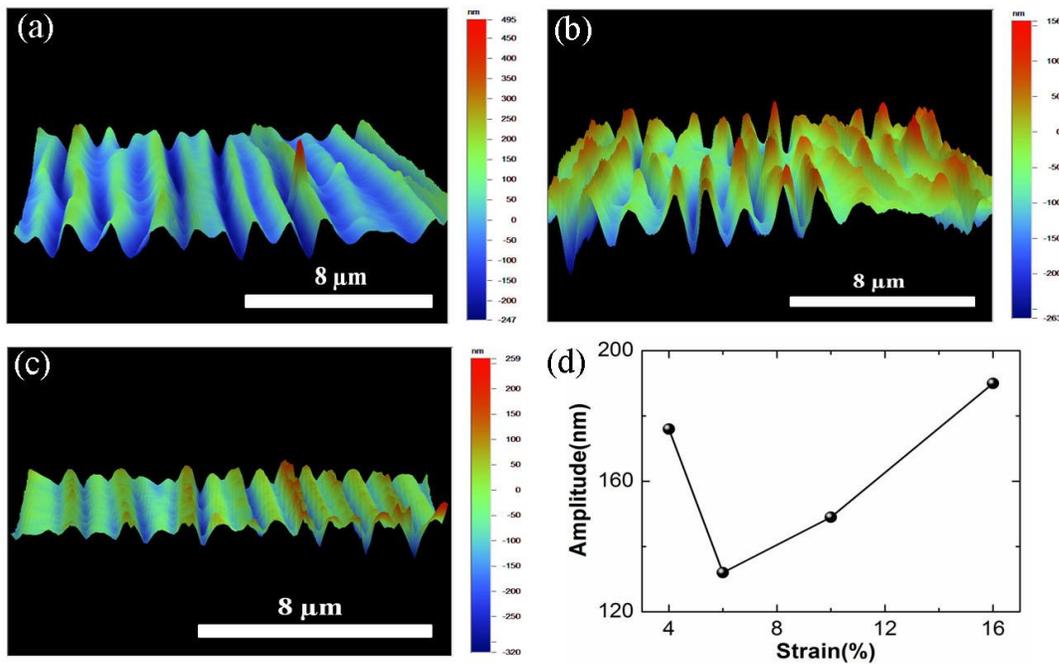
**Fig. S5.** Normalized resistance versus uniaxial strain (parallel geometry) in PEDOT:PSS film on  $\text{O}_2$  plasma treated PDMS substrate at very high ramp rate. (a)  $0.1\% \text{ sec}^{-1}$  (highest possible using linear translational manipulator). (b)  $0.4\% \text{ sec}^{-1}$  (manual).

**Schematic of wrinkles and cracks, crack width versus applied strain plot**



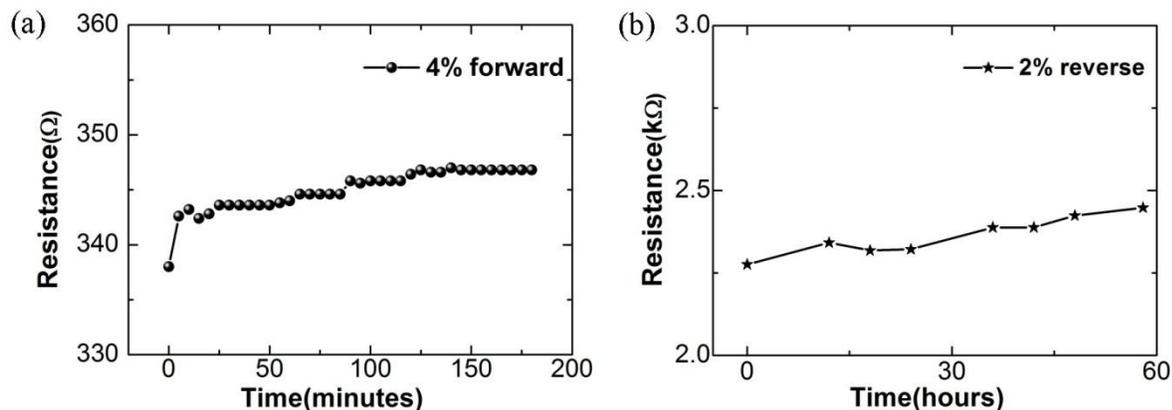
**Fig. S6.** (a) Schematic showing out-of-plane wrinkles and crack arrays in PEDOT:PSS film under applied strain. (b) Plot of average crack width versus applied strain (curves are spline function fits to the data points).

**3D optical profilometry images of PEDOT:PSS film under varying applied strain (forward) and wrinkle amplitude versus strain (forward) plot**



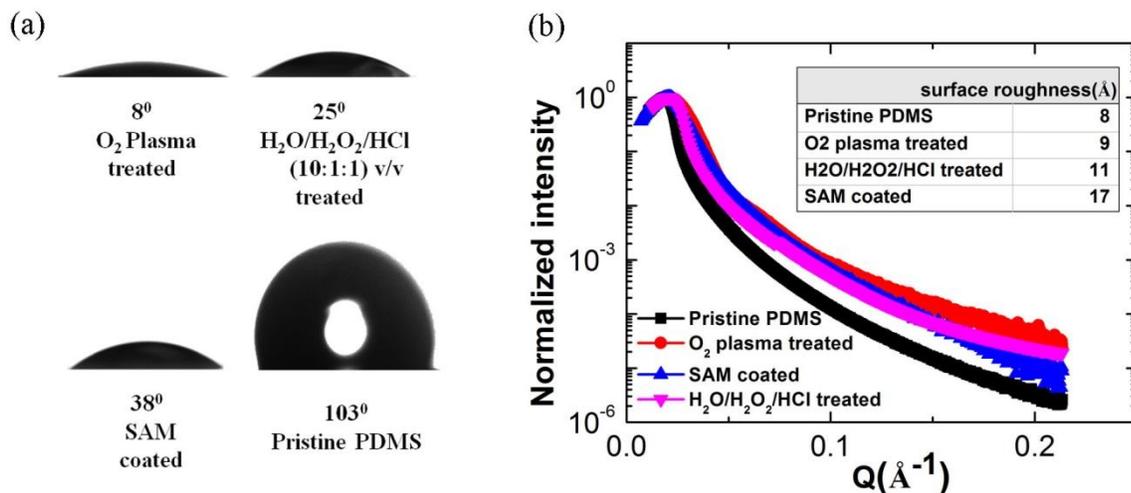
**Fig. S7.** 3D optical profilometry images of PEDOT:PSS films under varying applied strain (forward) (a) 4%. (b) 6% (c) 16%. (d) Plot of wrinkle amplitude versus applied strain.

**Time dependent measurement of resistance at different strain percentages (parallel geometry)**



**Fig. S8.** Resistance of PEDOT:PSS film in parallel geometry as a function of time. (a) 4% strain in the forward direction. (b) 2% strain in the reverse direction.

**Characterization of PDMS substrates with contact angle measurements and x-ray reflectivity (XRR) techniques**

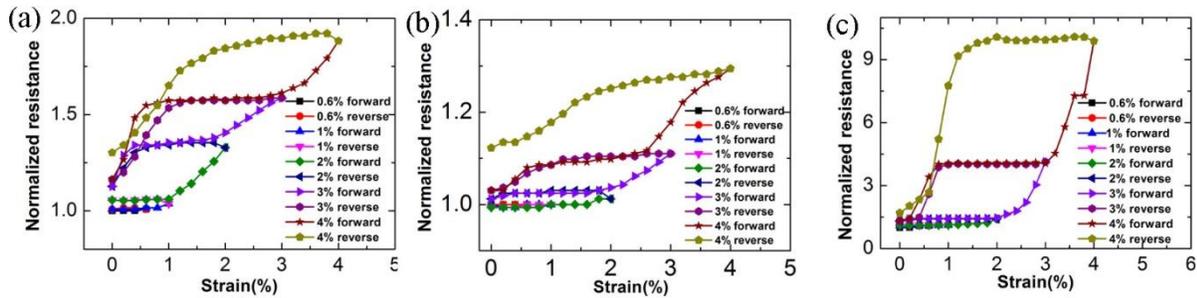


**Fig. S9.** (a) Contact angle measurement of DI water droplet (vol. 2.25  $\mu\text{L}$ ) on different functionalized PDMS substrates. (b) X-ray reflectivity (XRR) spectra of functionalized PDMS substrates.

A DI water droplet of volume 2.25  $\mu\text{L}$  was placed on different functionalized PDMS surfaces in order to obtain static contact angle using contact angle goniometer (Digidrop, GBX), equipped with a CCD

camera. Rigaku SmartLab x-ray reflectometer was used to perform out-of-plane specular XRR measurement in which the sample was illuminated with x-ray beam in theta-2theta geometry. To select a particular x-ray  $1.54\text{\AA}$ , germanium (200) 2-bounce crystals were used as the monochromator. X-ray reflectivity spectra were obtained by plotting intensity of the scattered X-ray against momentum transfer ( $Q = \frac{4\pi}{\lambda} \sin \theta$ ) along z-direction of the PDMS substrates. The reflectivity was scanned from  $0^0$  to  $3^0$ .  $Q$  corresponds to the maximum scattered intensity is called critical momentum transfer or critical wave vector and below which total external reflection took place. Above  $Q_C$ , scattered intensity falls off with some power of  $Q$ . Thickness oscillations or Keissig fringes are not observed in the XRR spectra as the PDMS substrates are micrometer thick. The value of surface roughness is estimated from the slope of the X-ray reflectivity spectra at reasonably high  $Q$ . The surface roughness values are similar for all functionalized PDMS substrates. The surface roughness versus surface modification of PDMS substrates are tabulated in inset of XRR plot.

**Observation of ‘Strain memory effect’ in PEDOT:PSS film on different interfaces, and in low conducting of PEDOT:PSS film (parallel geometry)**



**Fig. S10.** Cyclic measurement of normalized resistance in parallel geometry versus uniaxial strain for PEDOT:PSS film on different interfaces. (a) SAM coated PDMS (b) H<sub>2</sub>O/H<sub>2</sub>O<sub>2</sub>/HCl (10:1:1 v/v ratio) treated PDMS. (c) Cyclic measurement of normalized resistance in parallel geometry versus uniaxial strain for low conducting PEDOT:PSS film on O<sub>2</sub> plasma treated PDMS.