



Fig. S1 (a) Top view of the unit cell (b). The unit-cell vector  $\vec{c}$  (not shown) is along  $\hat{z}$  and extends from 0 to 80 Å. (c) is a projection of (b) along unit-cell vector  $\vec{b}$ . (e, d, f) show interfacial built-in electrostatic potential ( $V_{b-i}$ ) for the OPE atop junction (in eV) and is averaged along the  $\vec{b}$ ; the atoms pinpoint their corresponding positions in the  $\hat{x}-\hat{z}$  plane when projected along  $\vec{b}$ , as also shown in (c). All spatial coordinates in Ångstroms.