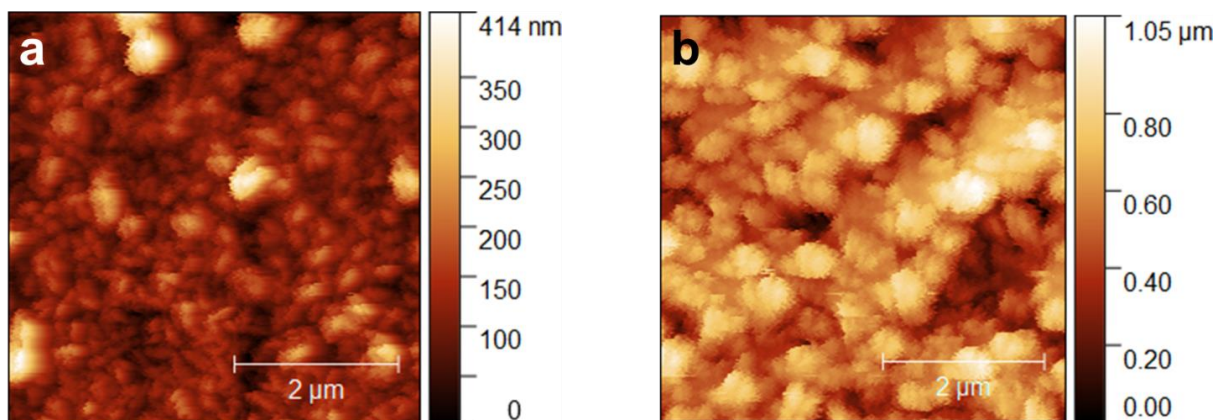


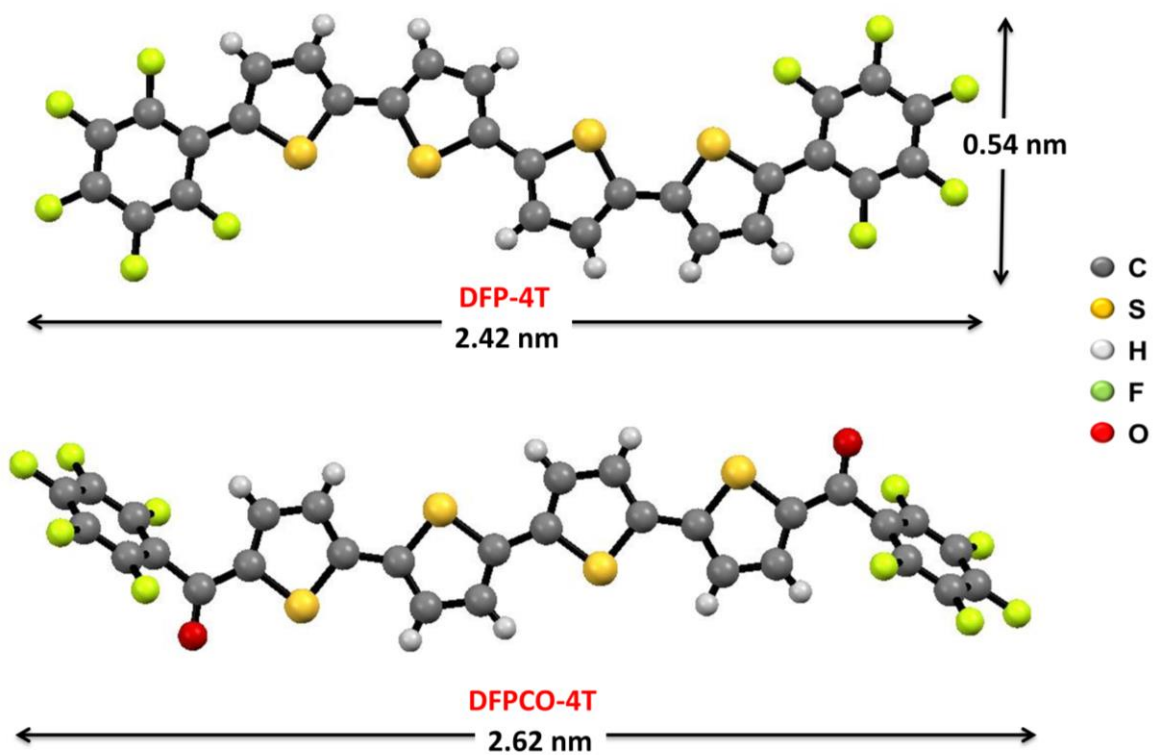
Supplementary Information

Molecular Engineering of Organic Semiconductors Enables Noble Metal-Comparable SERS Enhancement and Sensitivity

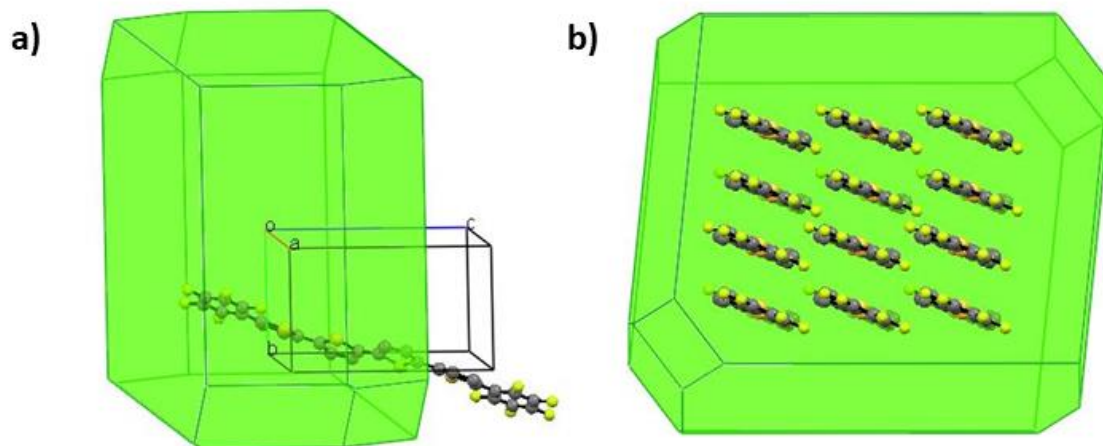
Demirel et al.



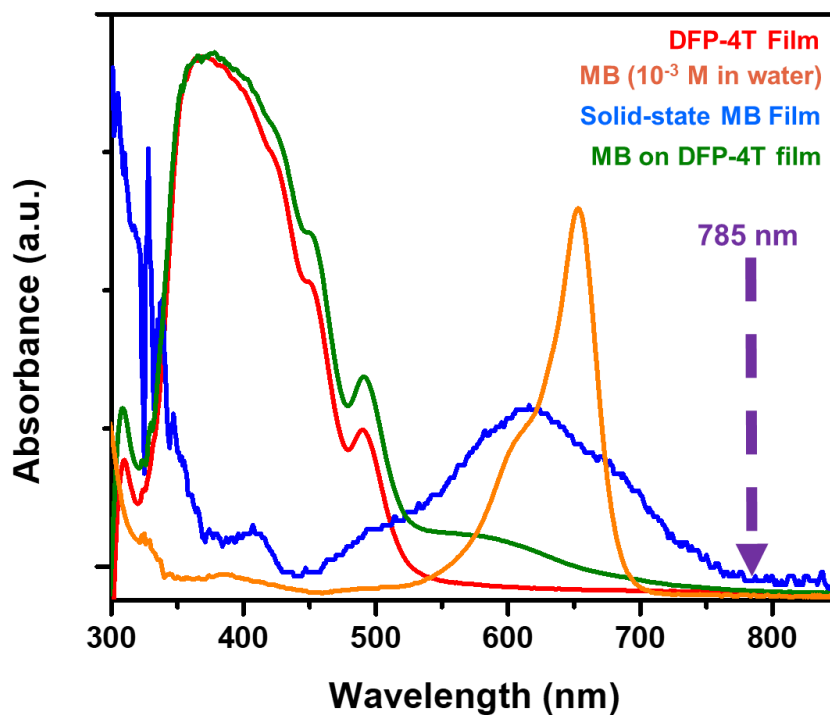
Supplementary Figure 1. AFM images of **DFP-4T** ($1.1 \pm 0.2 \mu\text{m}$ of thickness) (a) and **DFPCO-4T** ($1.7 \pm 0.4 \mu\text{m}$ of thickness) (b) films.



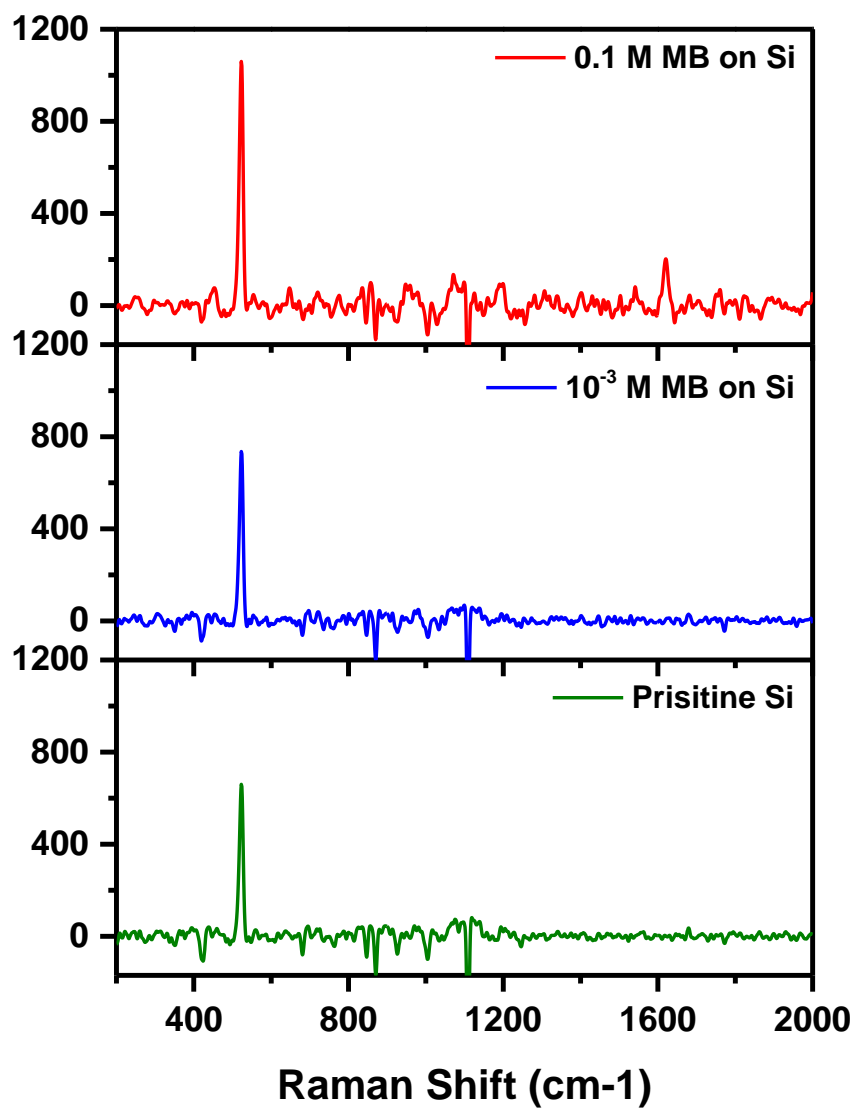
Supplementary Figure 2. Three dimensional molecular structures and lengths along the long molecular axis for **DFP-4T** and **DFPCO-4T** based on the single-crystal structure parameters.



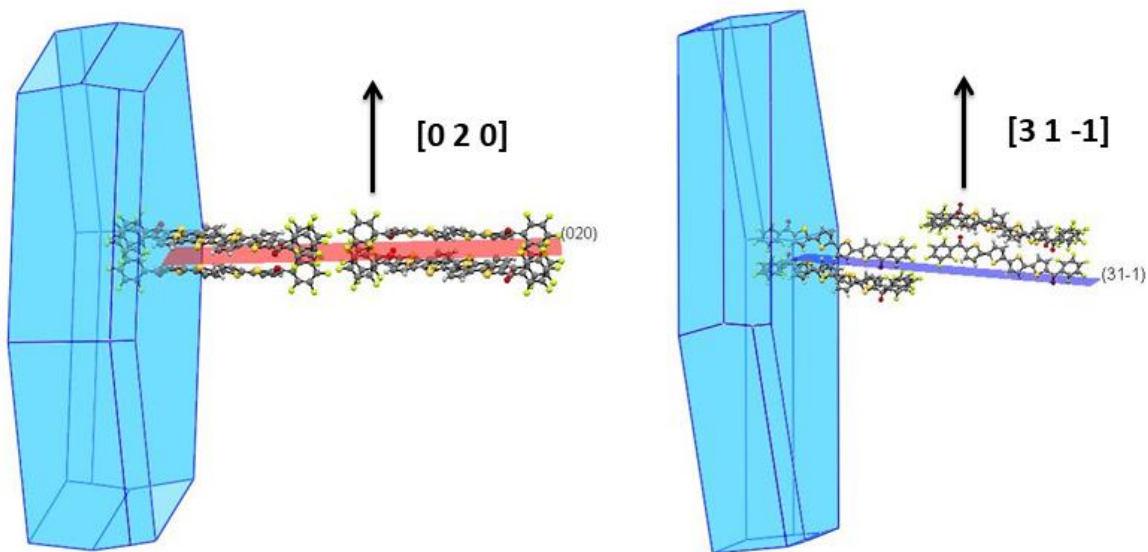
Supplementary Figure 3. BFDH (Bravais, Friedel, Donnay and Harker) theoretical crystal morphology simulated based on the single-crystal structure and view of the unit cell for **DFP-4T** (a), and molecular arrangement/packing in the 2-D crystal plane (b).



Supplementary Figure 4. UV-vis spectra of **MB** in aqueous solution (10^{-5} M) and solid state, pristine **DFP-4T** ($\sim 1.1 \mu\text{m}$) and 10^{-5} M of **MB** deposited **DFP-4T** films. (All samples were fabricated on cleaned glass slides).



Supplementary Figure 5. SERS spectra of MB on reference silicon substrates.



Supplementary Figure 6. BFDH (Bravais, Friedel, Donnay and Harker) theoretical crystal morphology simulated based on the single-crystal structure, and the views of molecular arrangements/packings along $[0\ 2\ 0]$ and $[3\ 1\ -1]$ crystallographic directions for **DFPCO-4T**.

Substrate	HOMO Energy (eV)	CT Excited State Energy (eV)	CT Excited State Oscillator Strength	SERS Enhancement Factor (CT resonance)	SERS Enhancement Factor (1.43 eV)
DFH-2T	-7.86	2.04	0.681	4.8×10^4	0.4
DFH-3T	-7.30	1.72	0.014	9.7×10^2	0.8
DFH-4T	-7.10	1.43	0.011	2.3×10^3	2.3×10^3
DFH-5T	-6.80	1.25	0.030	3.5×10^5	25.3

Supplementary Table 1. Highest occupied molecular orbital (HOMO) energies of isolated substrate molecules; CT excited state energies and oscillator strengths; and SERS enhancement factors for the substrate-**MB** complexes at the INDO/SCI level.