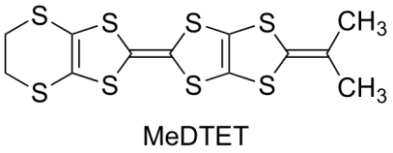
Supplementary Information

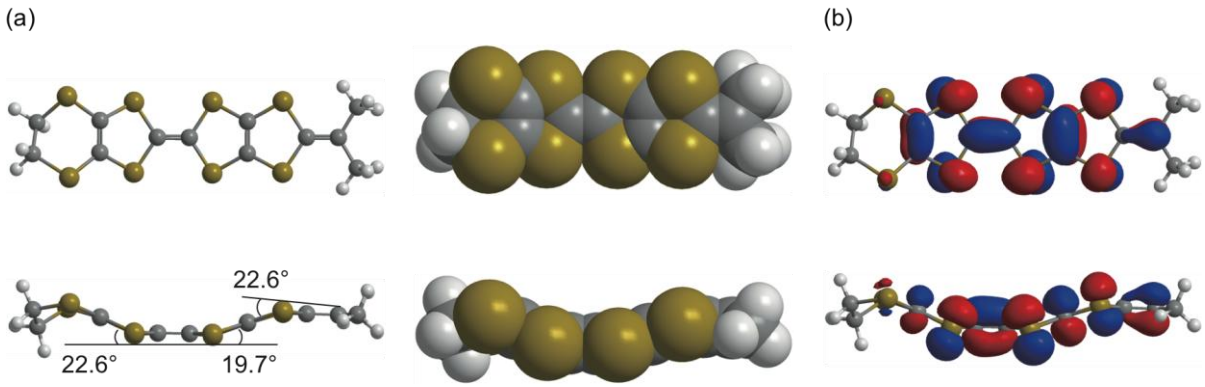
1. Theoretical Calculation of MeDTET

Density functional theory (DFT) calculations of MeDTET were carried out using a hybrid method of Hartree-Fock and B3LYP method [S1–S3]. They are performed with the Gaussian 09 program package [S4] using the 6-31G(d) basis set [S5].



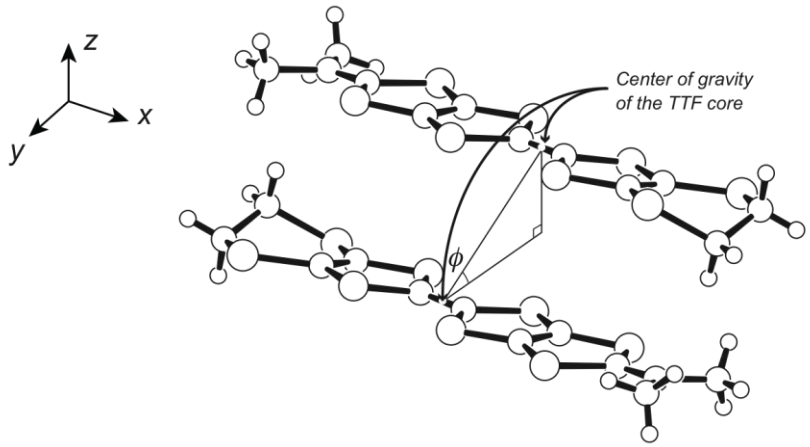
MeDTET

Figure S1. (**a**) Top view and side view of optimized structure for MeDTET are represented by ball and stick model (left) and space-filling model (right). (**b**) HOMO of MeDTET. The energy level is −4.74 eV.



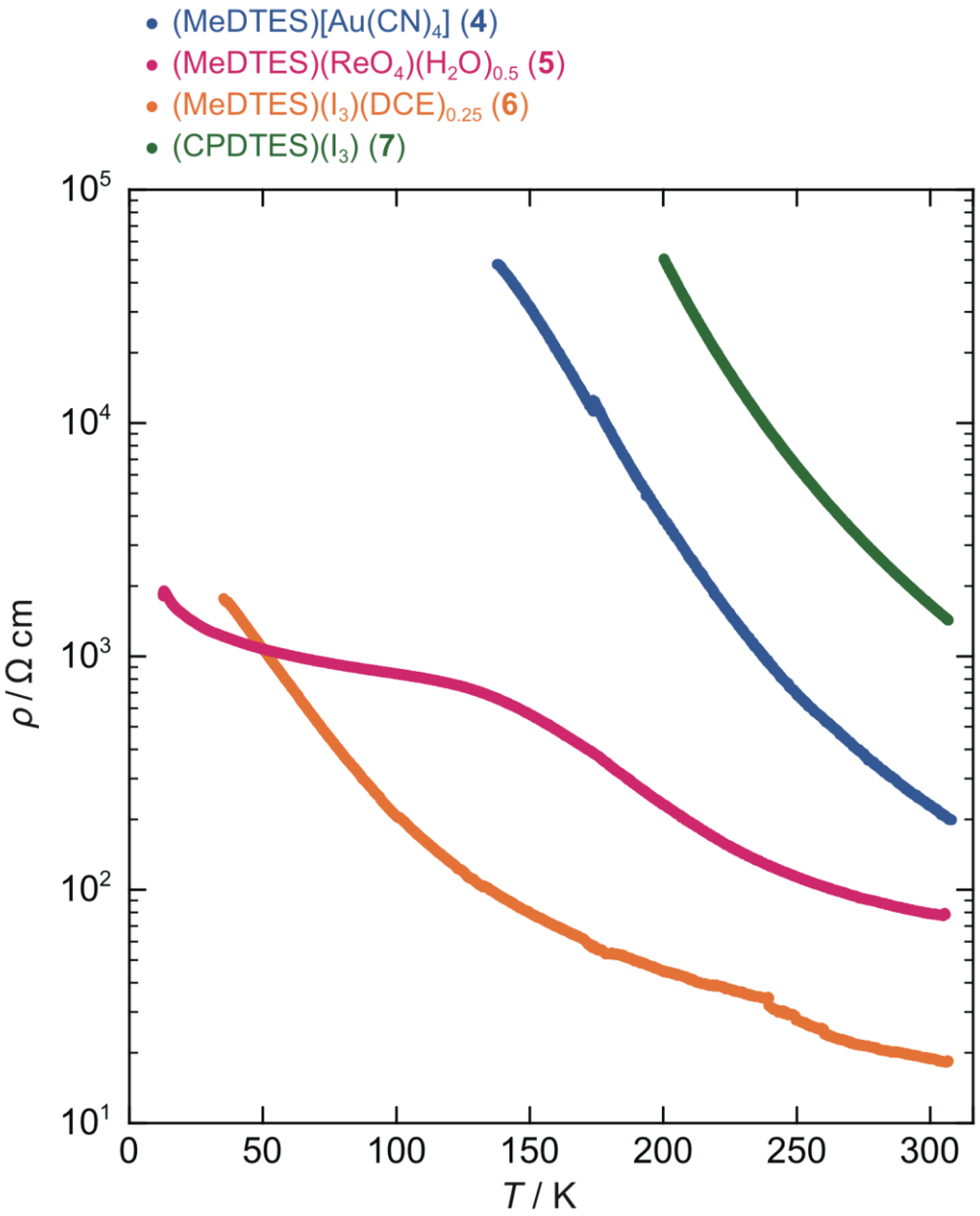
2. Definition of Geometrical Parameters

Figure S2. Definition of geometrical parameters: *x* and *y* are the slip distance along the molecular long and short axes, respectively; *z* is the interplaner distance between the molecular plane. The intermolecular vector is calculated based on the center of gravity of the TTF core of the unsymmetrical donor molecule.



3. Temperature Dependence of the Resistivity

Figure S3. Temperature dependence of the resistivity (ρ) of **4** (blue line), **5** (pink line), **6** (orange line), and **7** (green line).



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