

Figure S1. Atomic numbering scheme for the BEDT-TTF molecules **A-E** and the [Hg(SCN)₃]⁻ anion in the (BEDT-TTF)₂Hg(SCN)₃ structure at room temperature. Molecules **D** and **E** are located on inversion centers. Thermal ellipsoids are drawn at the 50% probability level.

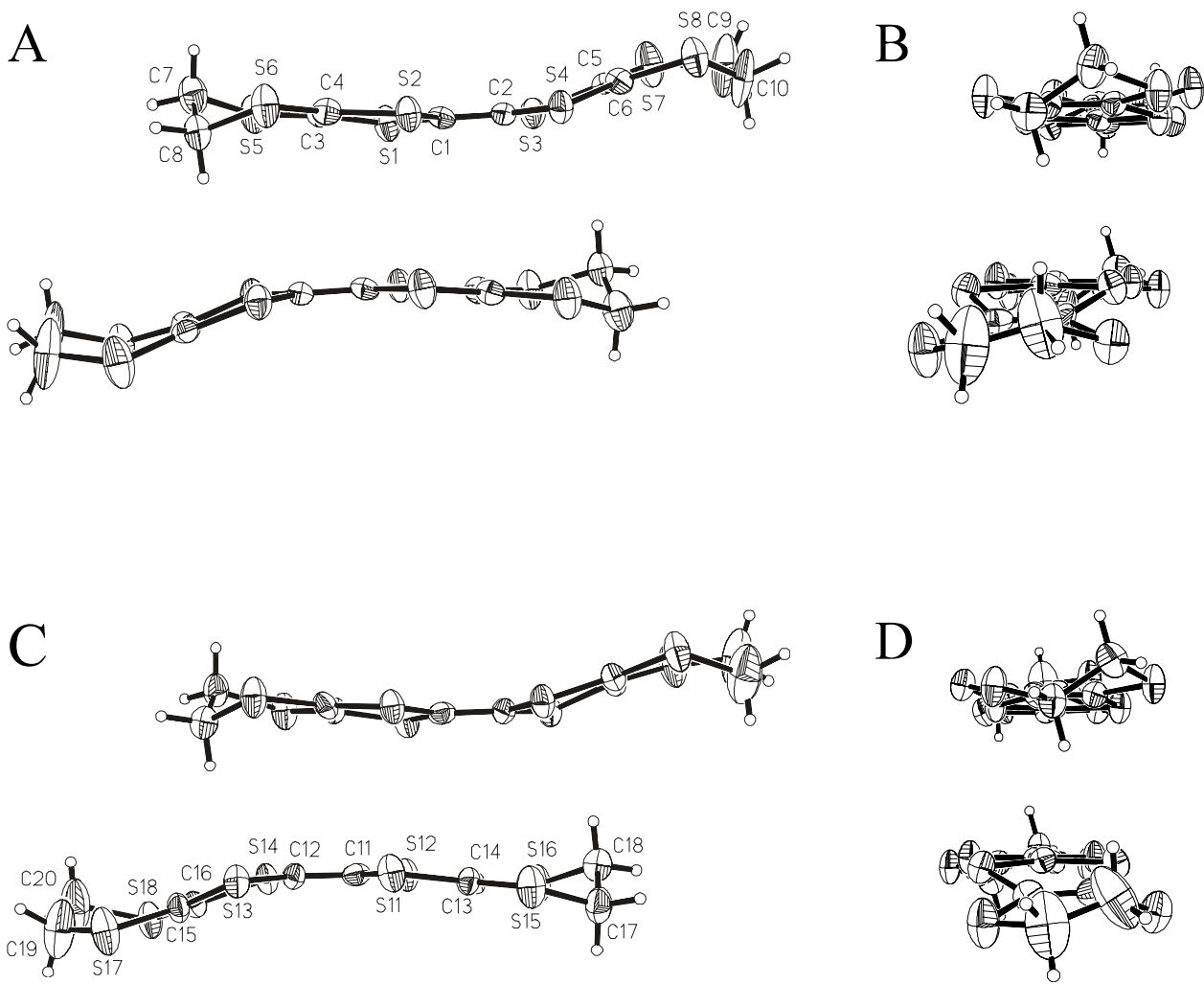


Figure S2. Dimerized BEDT-TTF molecules **A** and **B**. A and B illustrate the dimer of molecule **A** viewed along the short and long axis of the molecule, respectively. Similarly, C and D illustrate the dimer of molecule **B**. Thermal ellipsoids are drawn at the 50% probability level.

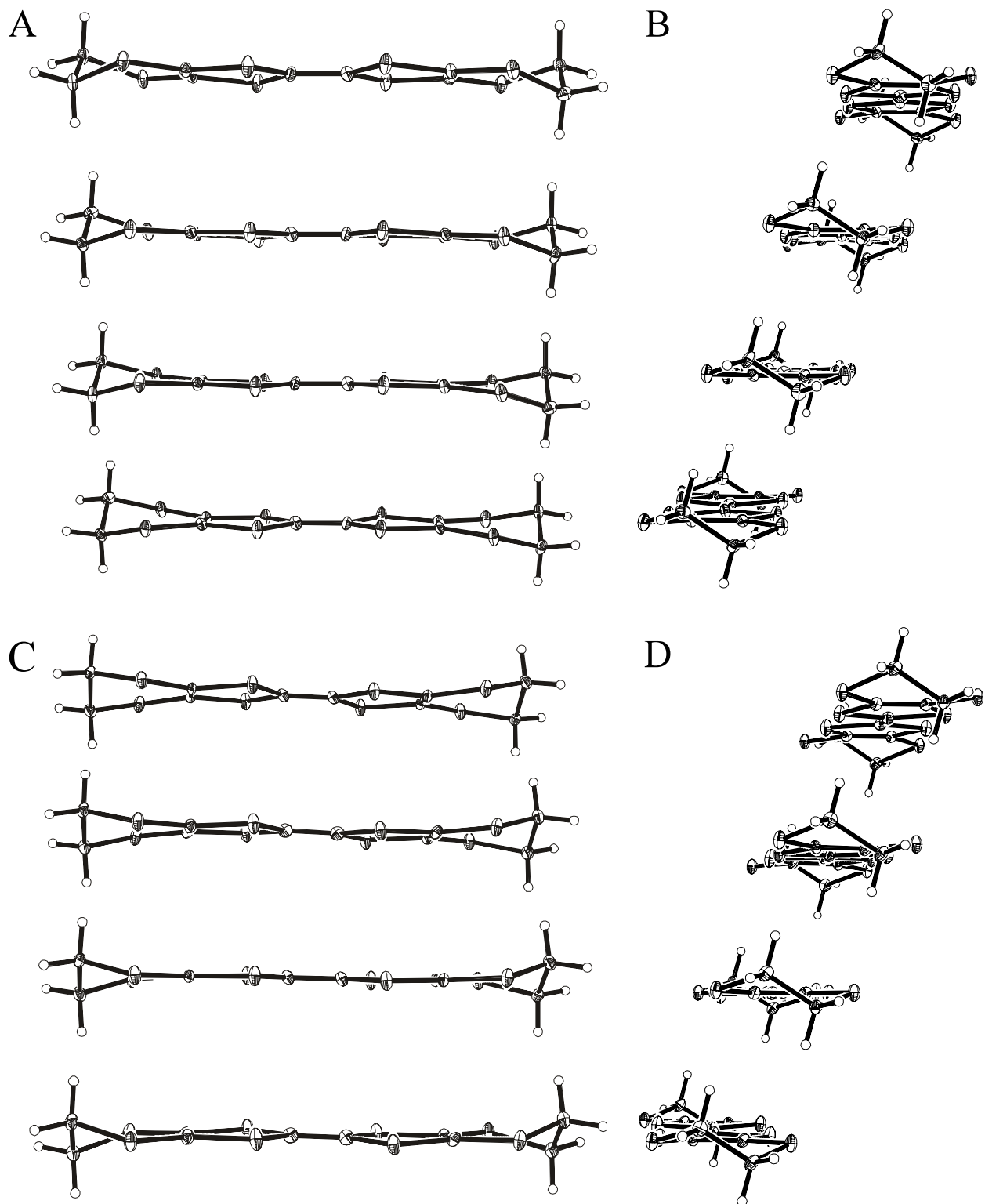


Figure S3. Stacks of BEDT-TTF molecules in the layer centered at $z = 0.5$. A) Stacks formed from molecule **C**. B) Projection of the stack formed from molecules **C** as viewed down the central C=C bond axis. C) Stacks formed by alternating molecules **D** and **E**. D) Projection of the stack formed from alternating molecules **D** and **E** as viewed down the central C=C bond axis. Thermal ellipsoids are drawn at the 20% probability level.