

## Supporting information

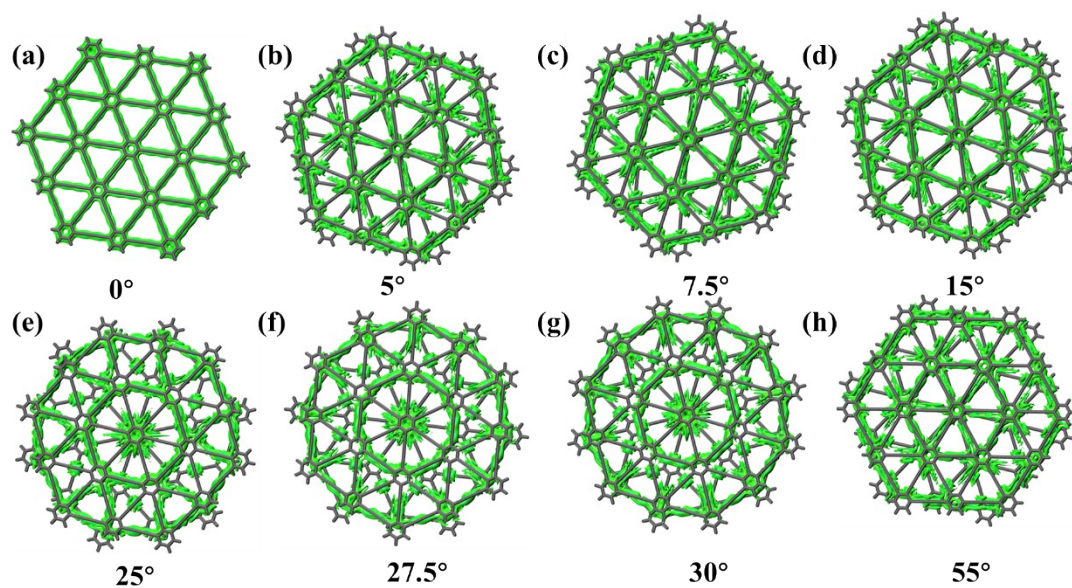


Figure S1: weak interlayer interaction in TwBLGDY when twist angle increases from 0° to 55°.

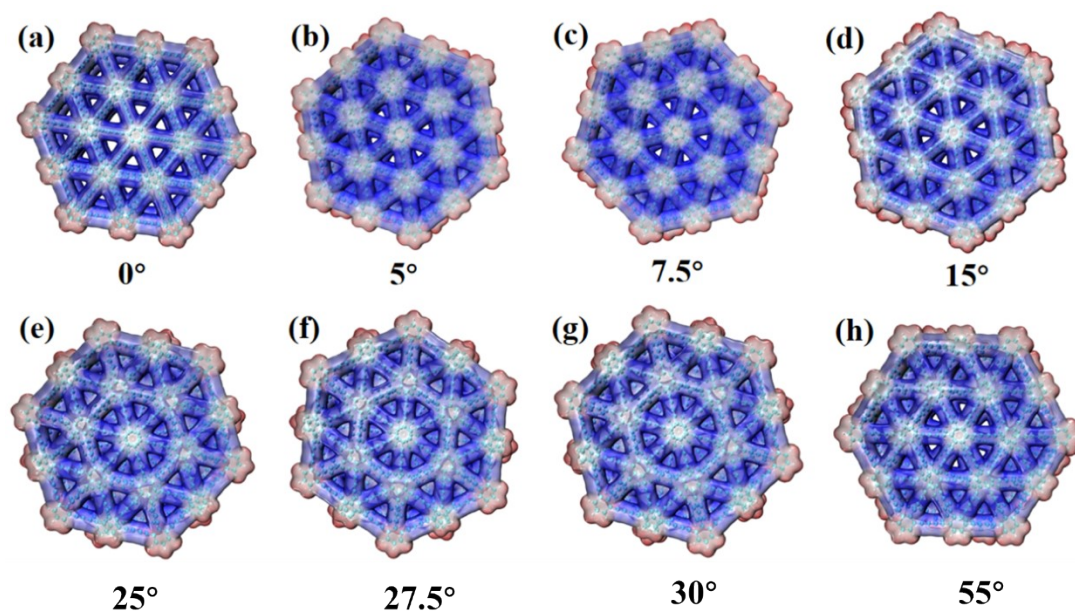


Figure S2: Molecular surface electrostatic potential of TwBLGDY as twist angle increases from 0° to 55°.

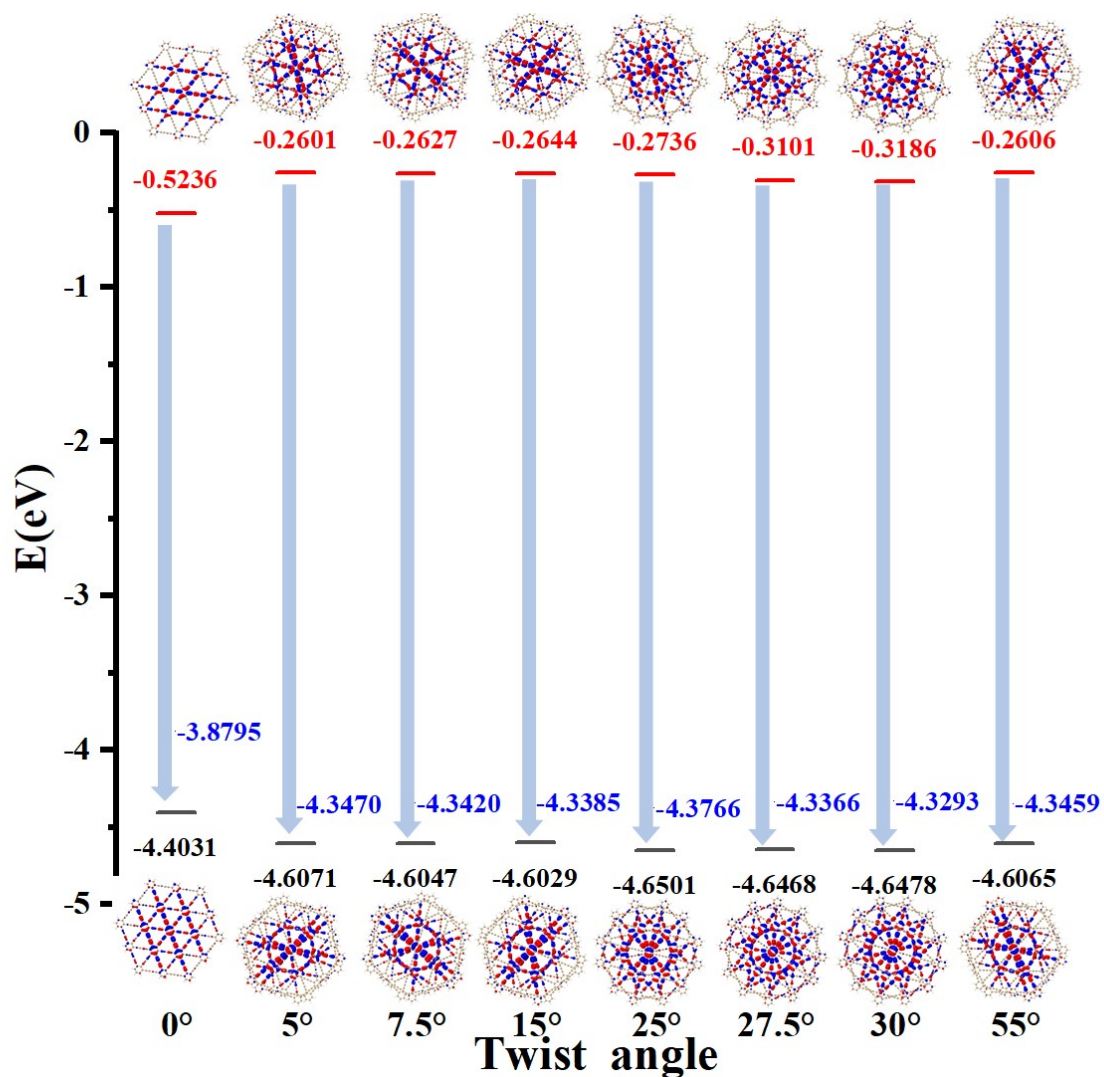


Figure S3: HOMO-LUMO molecular orbitals of TwBLGDY at different twist angles.

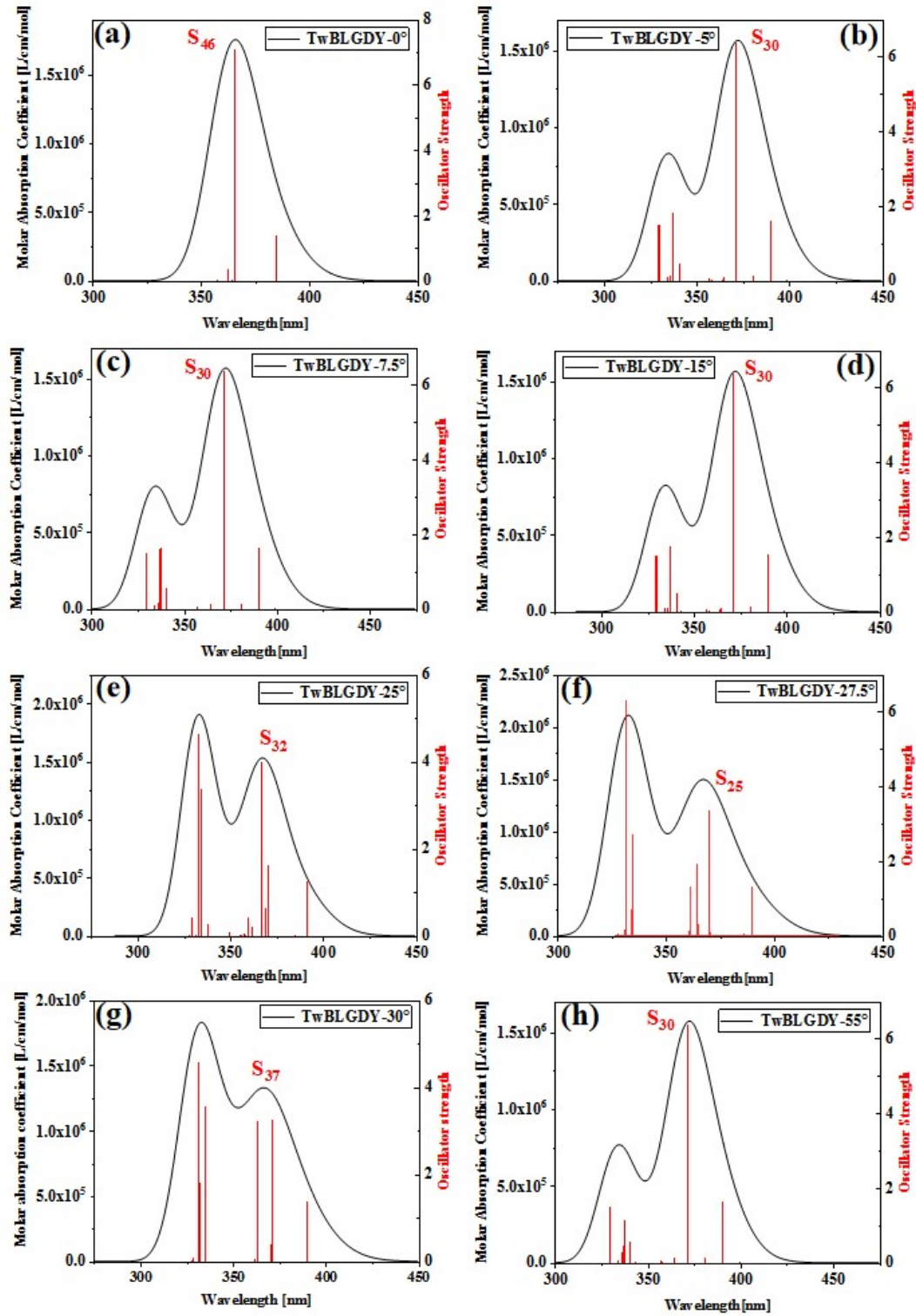


Figure S4: The OPA spectrum of TwBLGDY when the twist angle increases from 0° to 55°. The right vertical axis is the oscillator strength.

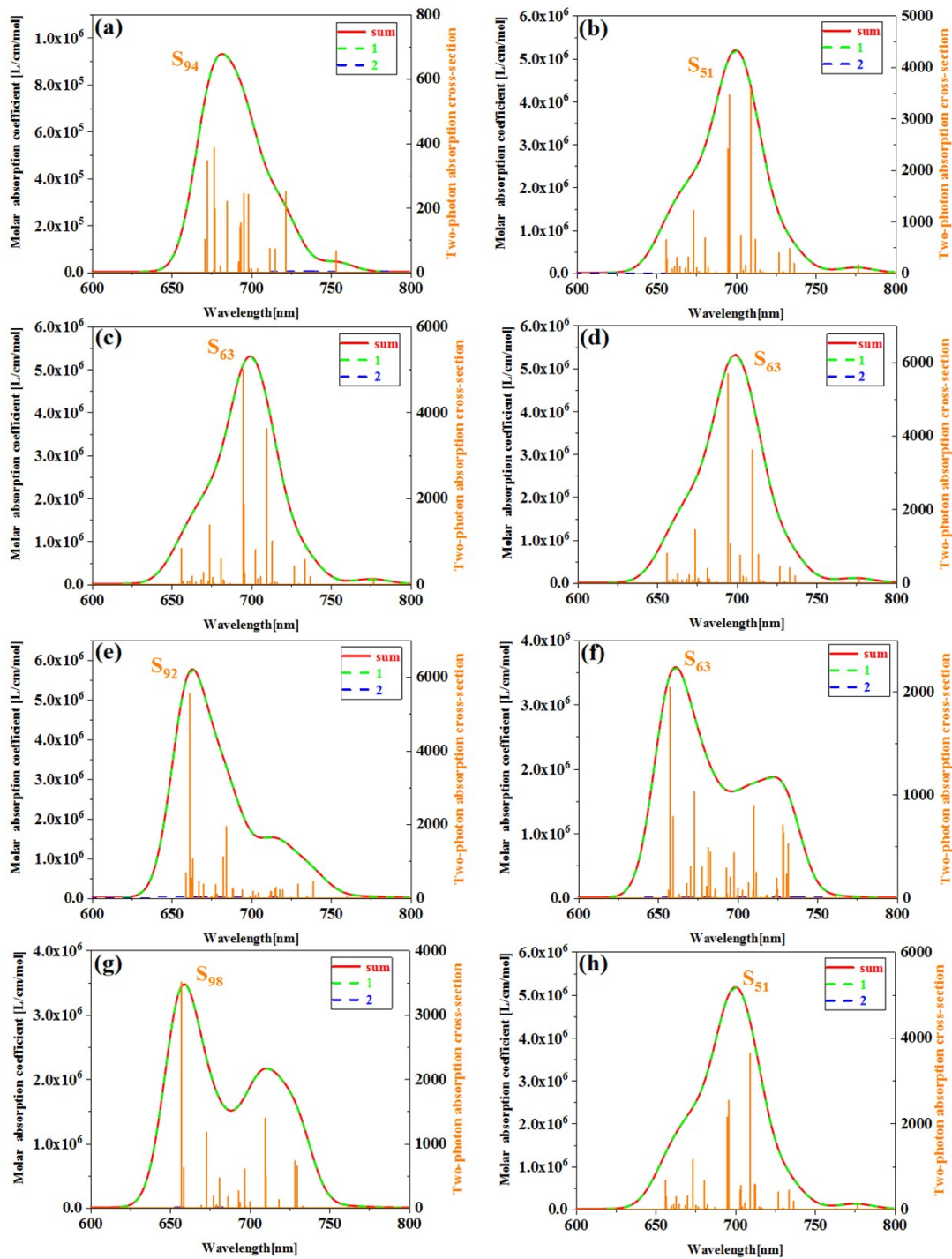


Figure S5: TPA spectrum of TwBLGDY as the twist angle increases from 0° to 55°. The right ordinate is the TPA cross-section.

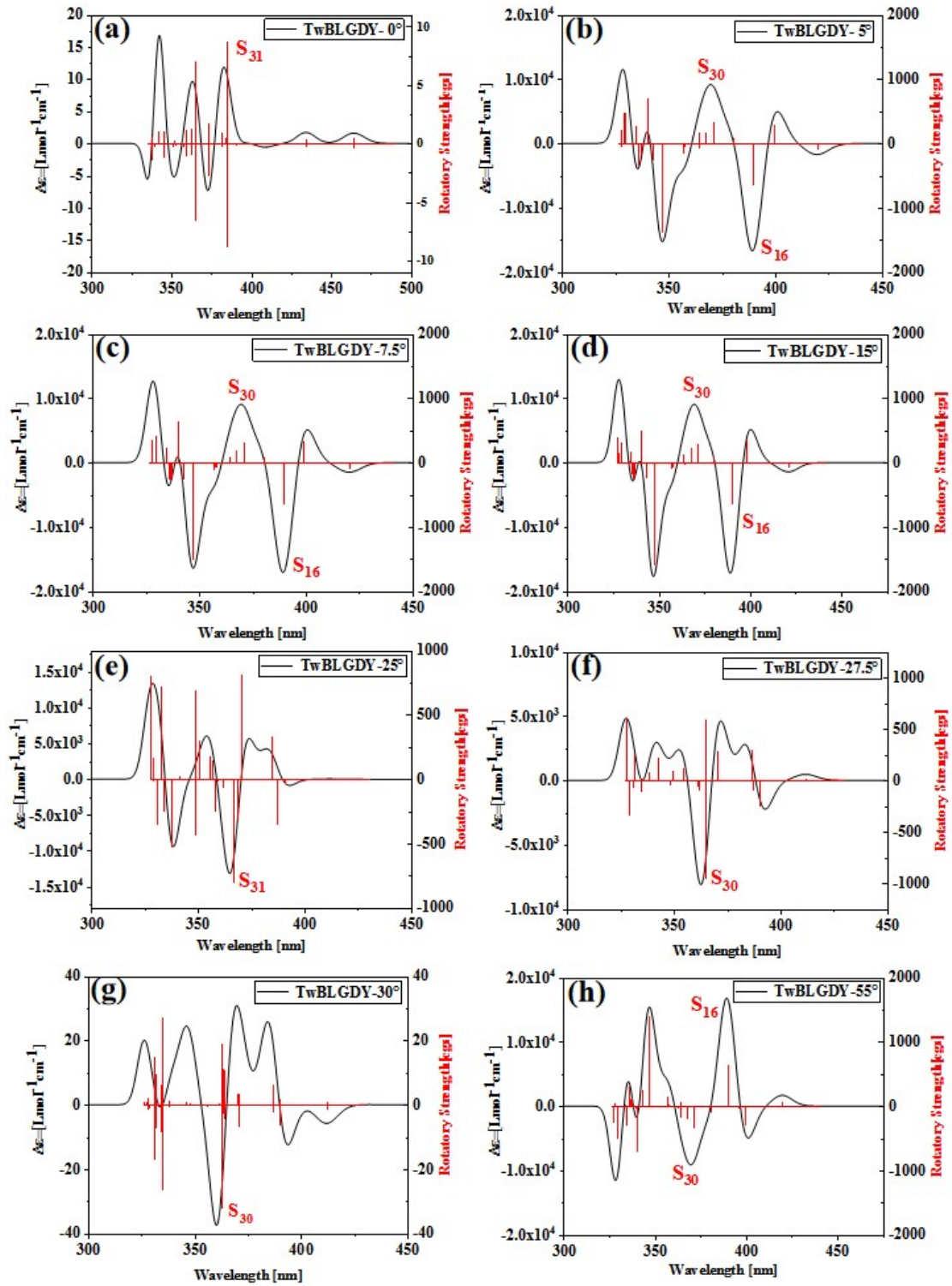


Figure. S6: ECD spectrum of TwBLGDY when twist angle increases from  $0^\circ$  to  $55^\circ$ , and the right ordinate is rotatory strength.