



**Figure S1** (a) VUV-MATI spectrum of THT. Simulated spectra using the Franck-Condon factors and the vibrational frequencies calculated at the (b) B3LYP, (c) B98, (d) M06, (e) M062X, and (f) BMK levels with cc-pVTZ basis set for the C<sub>2</sub> conformer. All spectra are magnified by fourfold for detailed comparison.