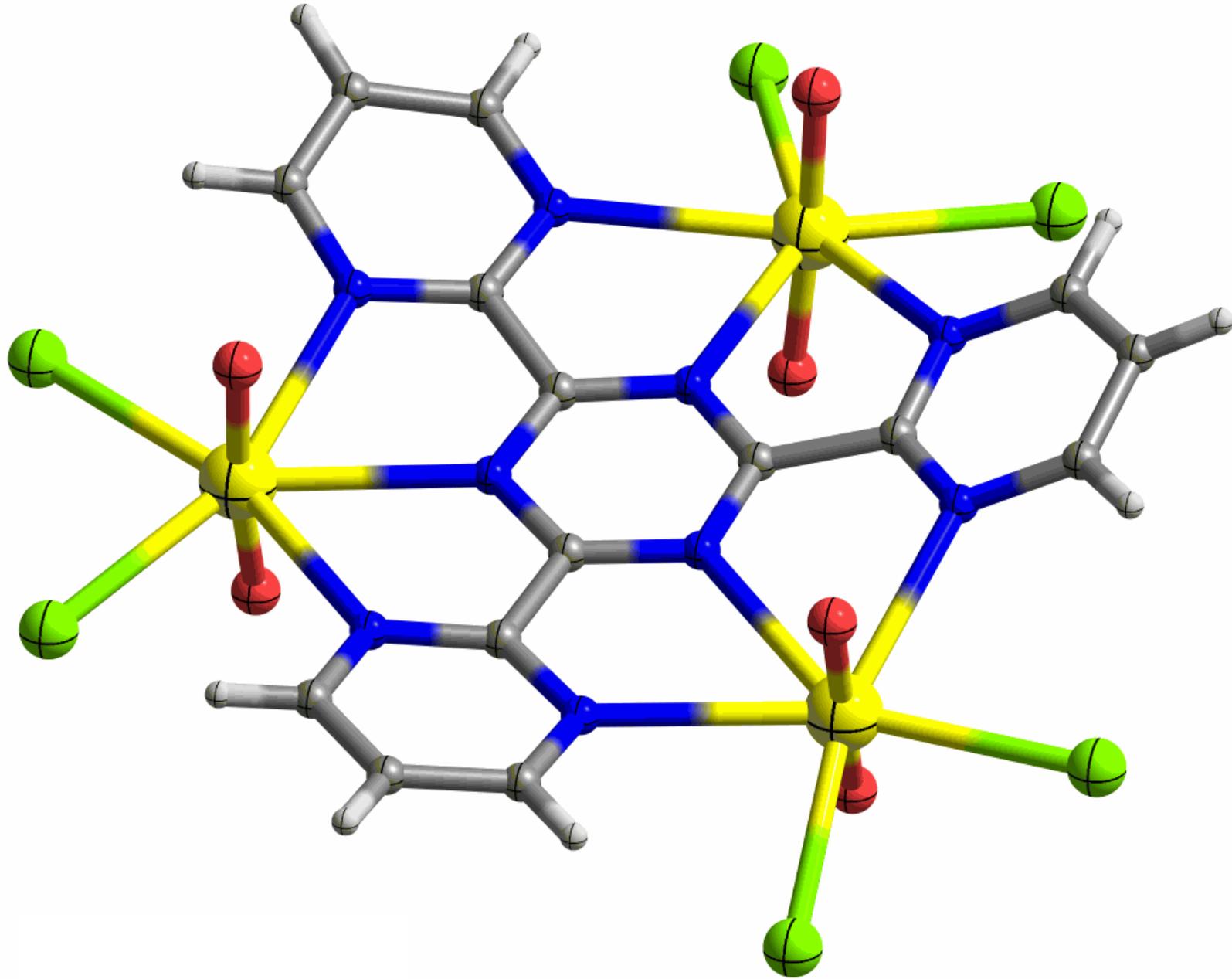
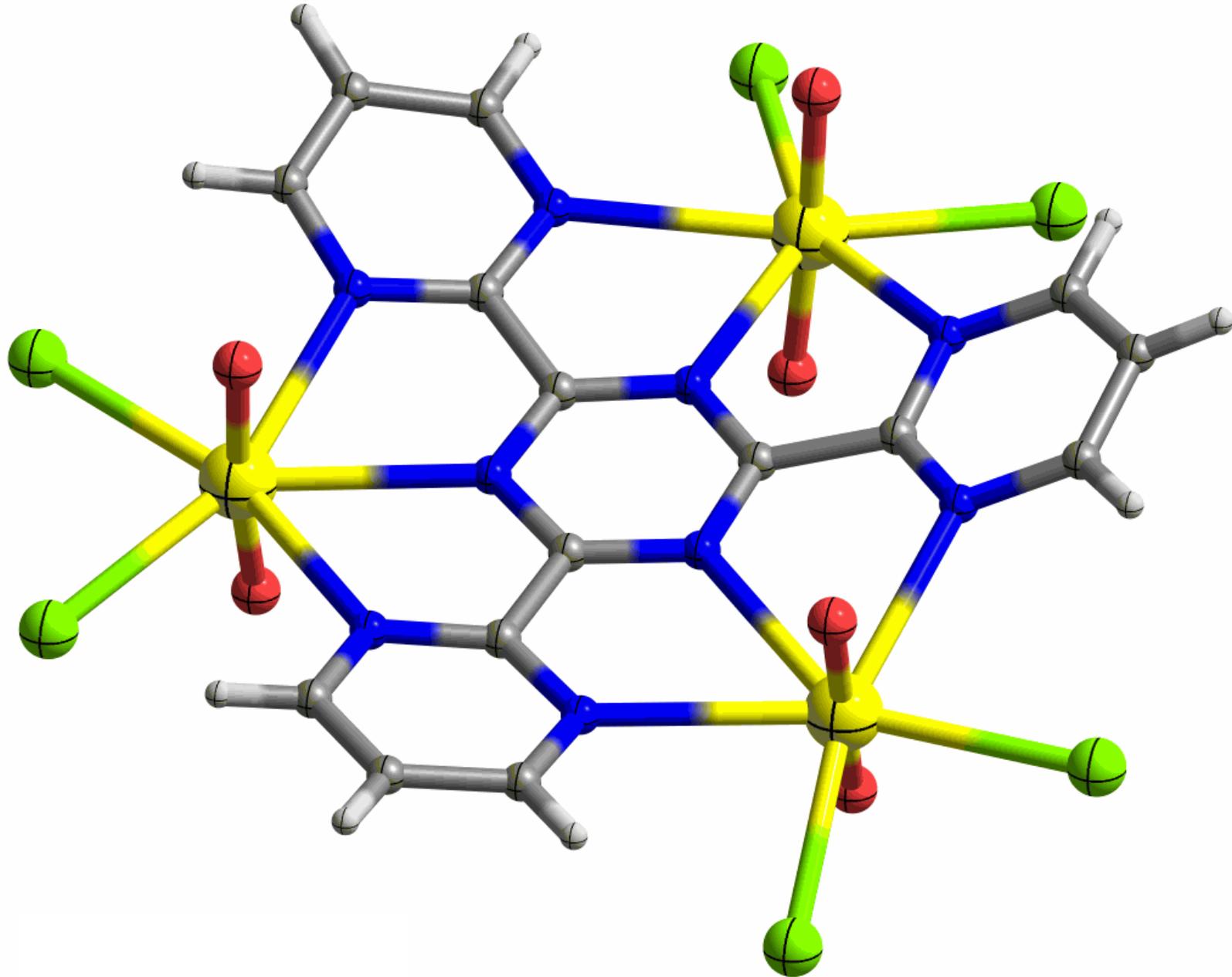


DFT computed {B3LYP/CRENBL (for U)/def2-TZVP (for Cl,O,N)/def2-SVP (for C,H)} IR and Raman active frequencies associated with uranyl group (between 800 cm^{-1} – 1000 cm^{-1}) and TPymT ligand (between 1000 cm^{-1} – 1200 cm^{-1}) performed on the $[(\text{UO}_2\text{Cl}_2)_3(\text{TPymT})]^{3-}$ moiety of the XRD structure of **1**. The $[\text{K}(18\text{-Crown-6})]^+$ group was removed from the model complex to minimise computational costs.

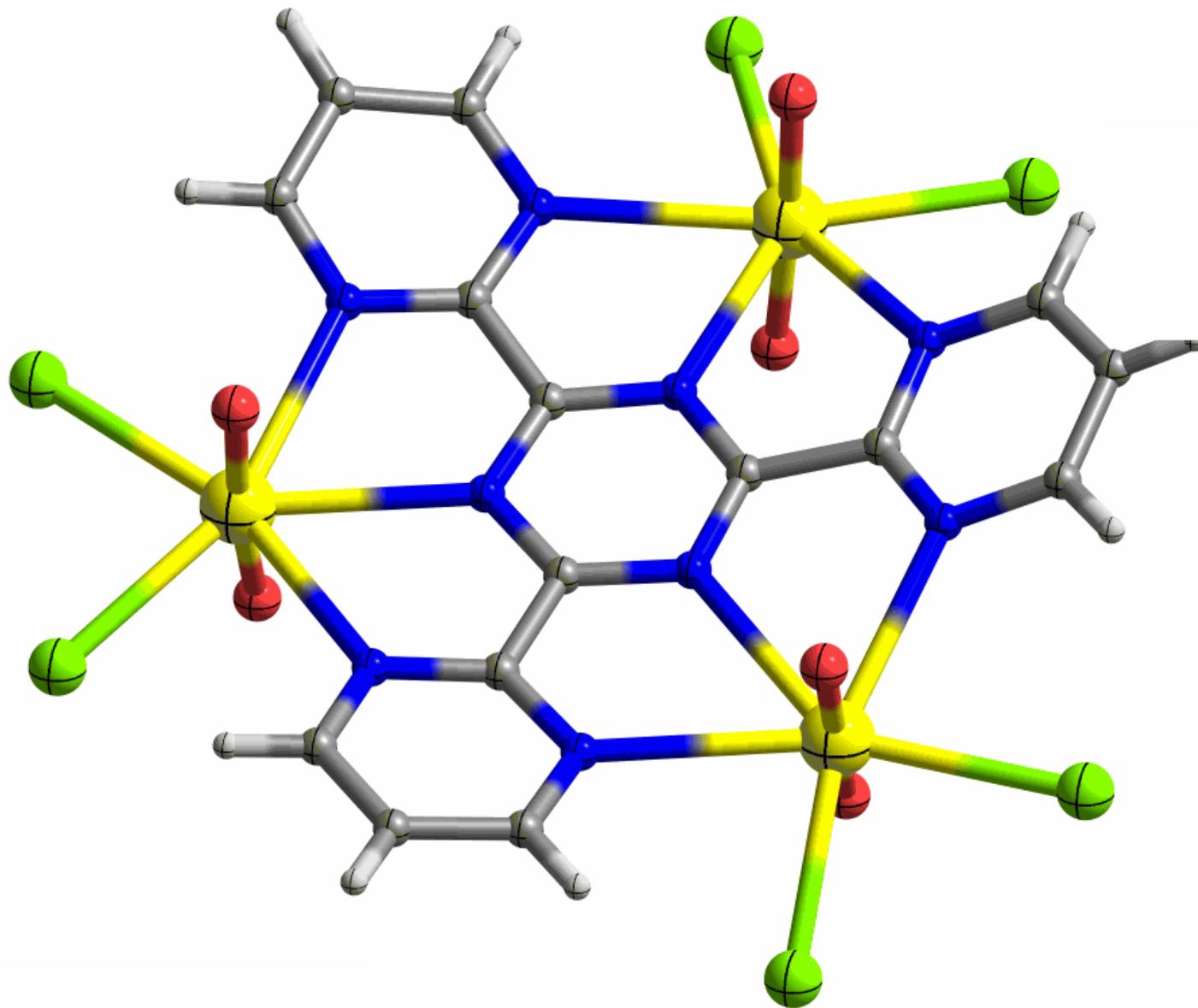
853.7 cm^{-1} - Symmetric U-O_{y1} Stretch (Raman Active)



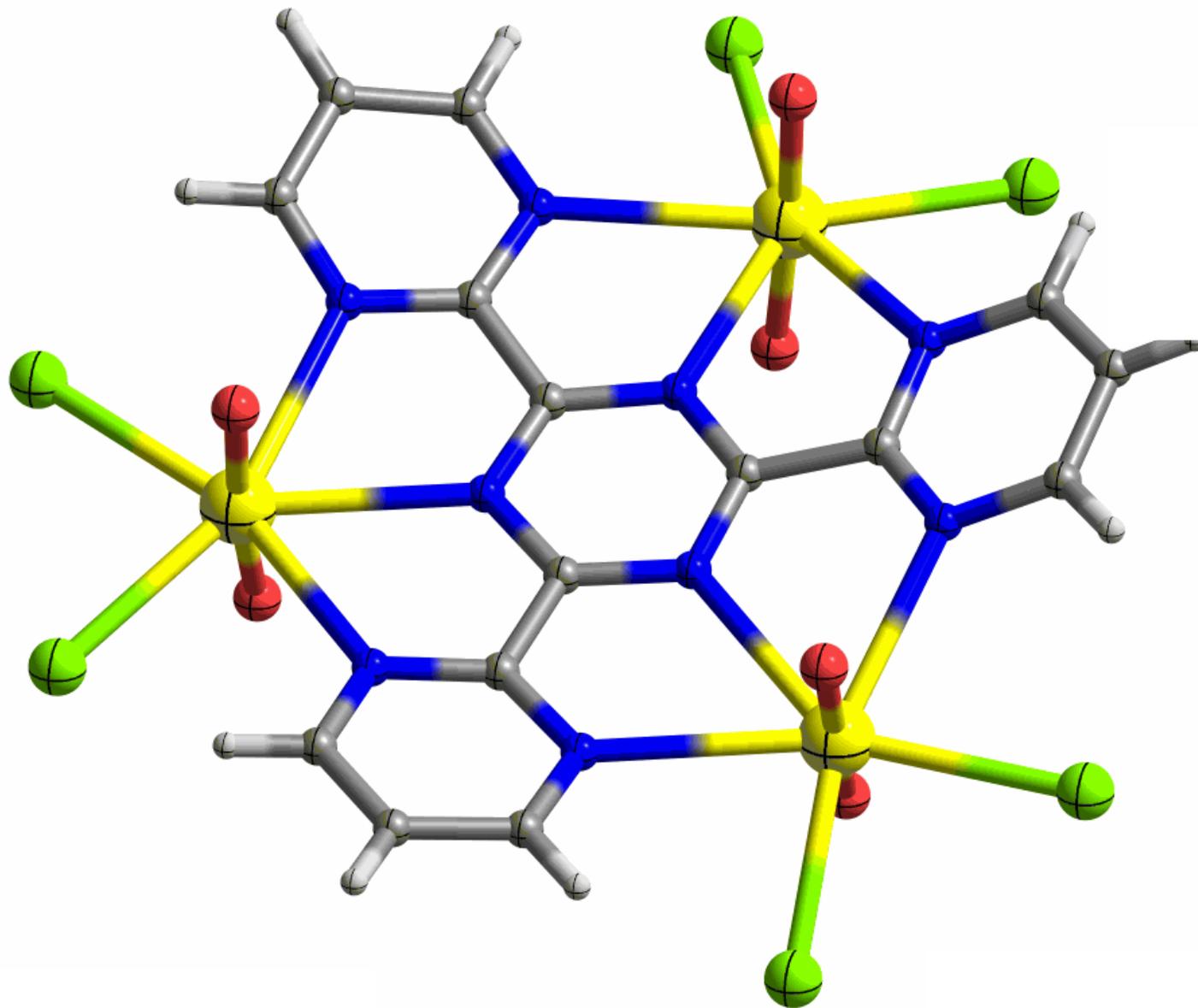
870.9 cm^{-1} - Symmetric U-O_y Stretch (Raman Active)



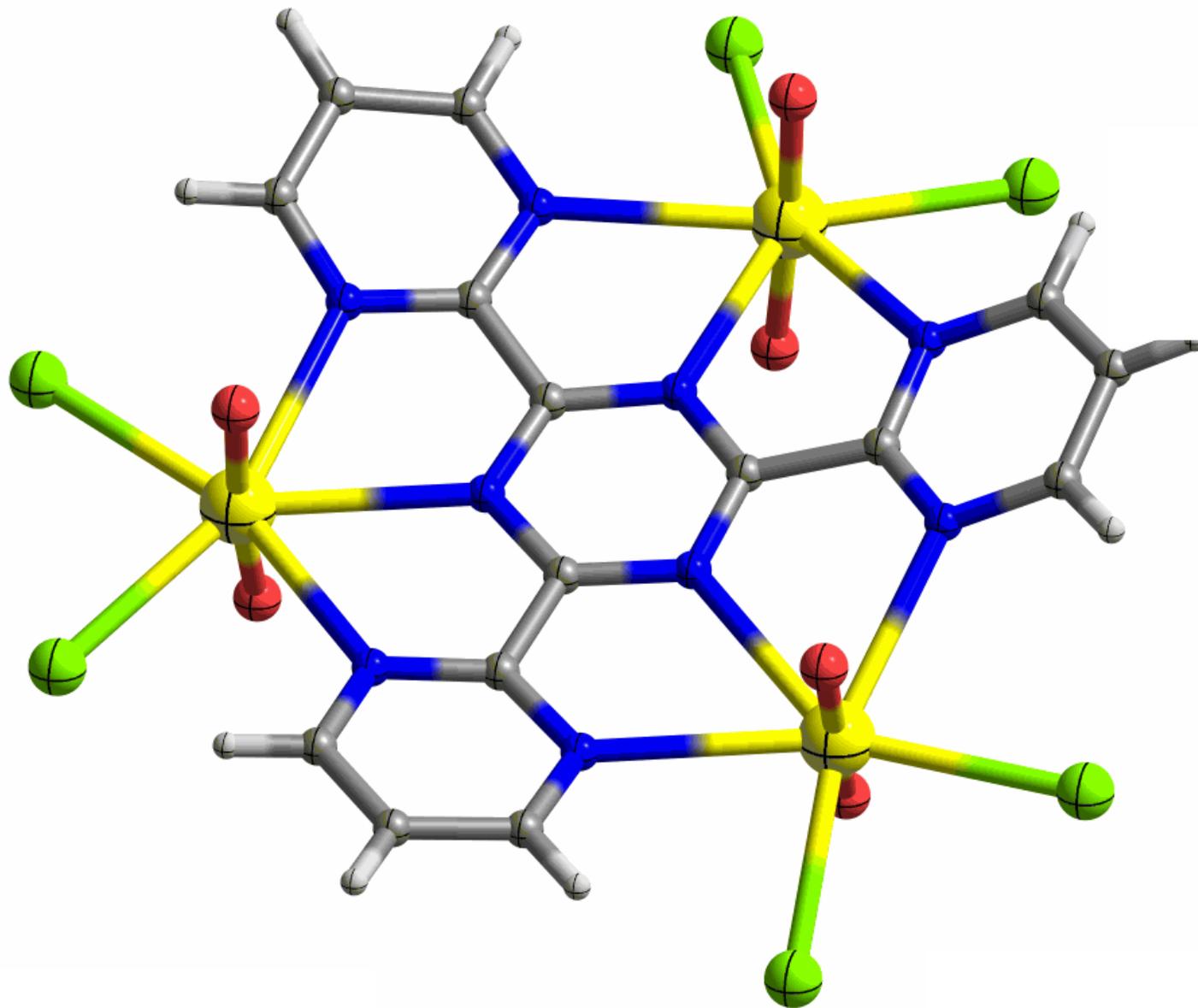
928.8 cm^{-1} - Asymmetric U-O_{yl} Stretch (IR Active)



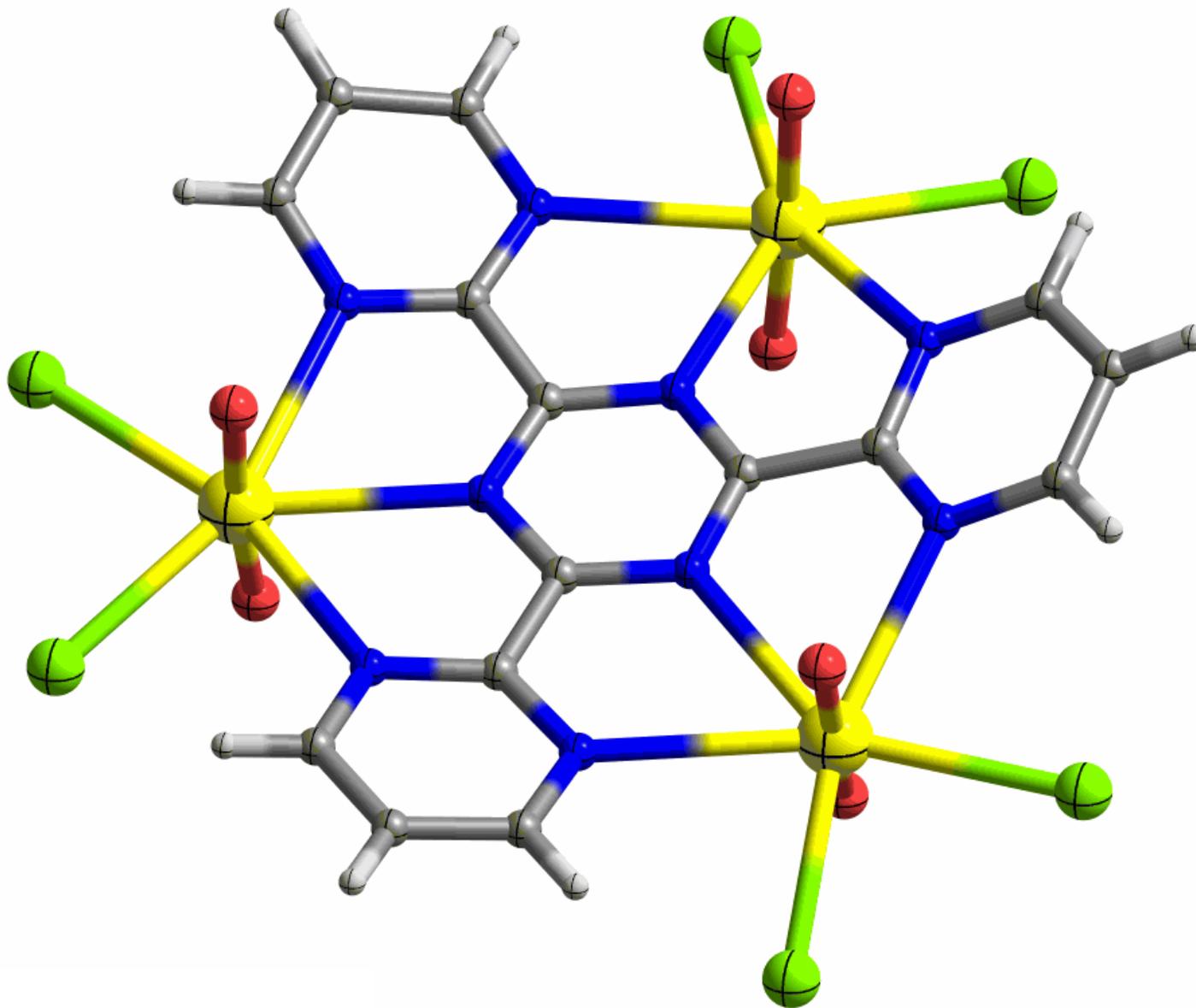
944.6 cm^{-1} - Asymmetric U-O_{y1} Stretch (IR Active)



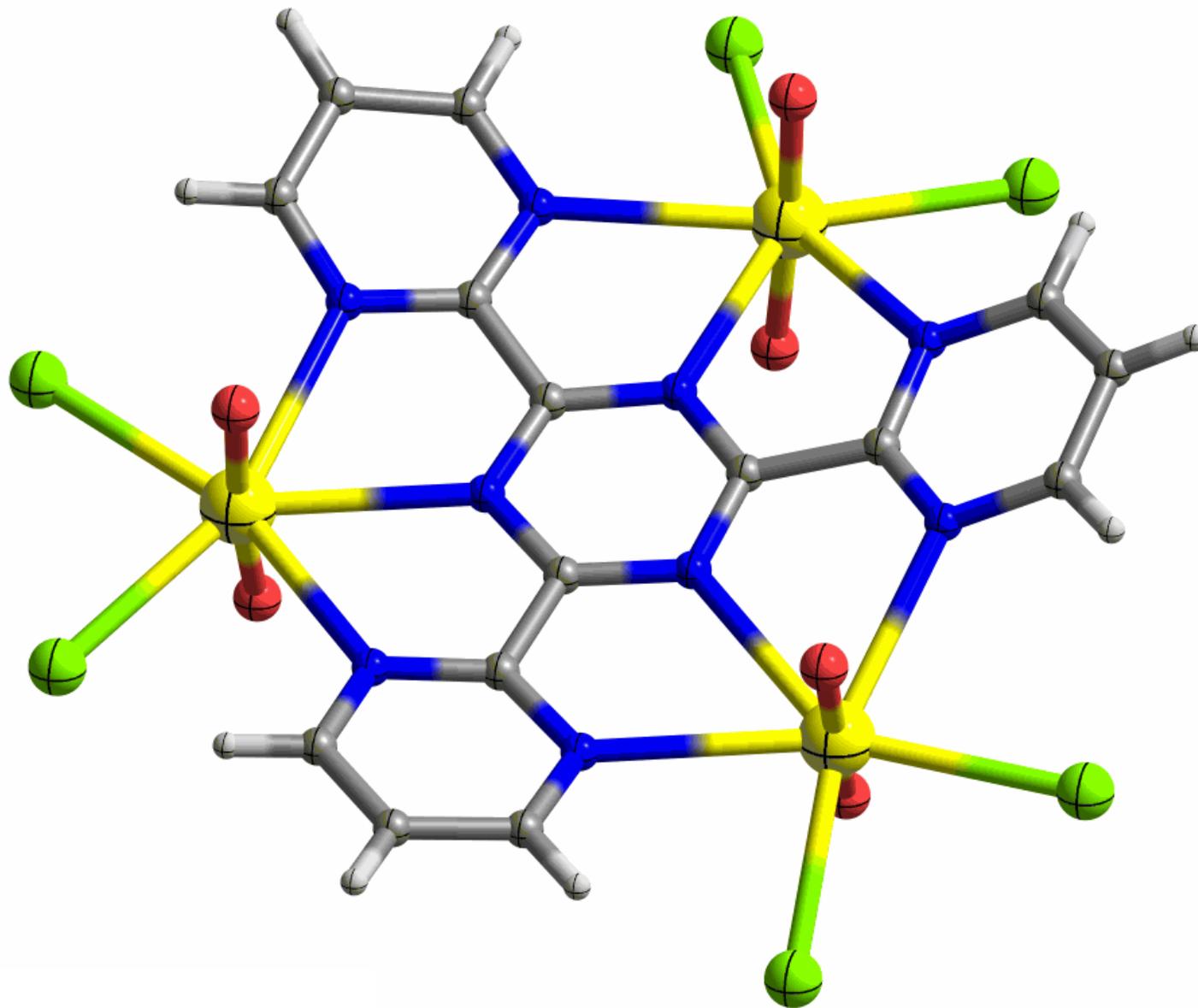
948.2 cm^{-1} - Asymmetric U-O_{yl} Stretch (IR Active)



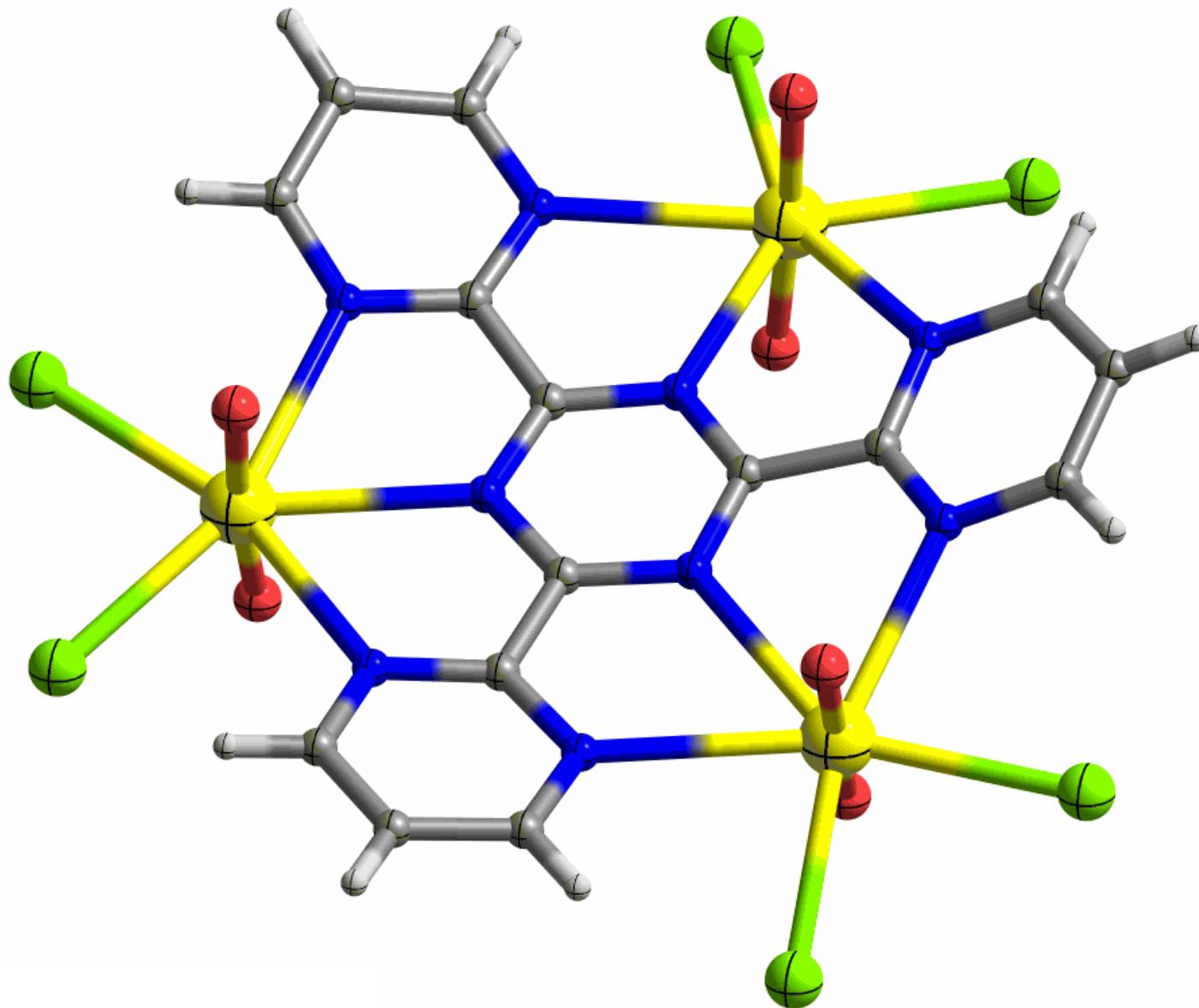
TPymT-radical based frequencies (1037.1 cm^{-1})



TPymT-radical based frequencies (1046.7 cm^{-1})



TPymT-radical based frequencies (1052.3 cm^{-1})



TPymT-radical based frequencies (1059.9 cm^{-1})

