

Figure S1. (a) ^{57}Fe PVDOS ($\circ\circ\circ$) for $[\text{N}(\text{n-Bu})_4]_2[^{57}\text{Fe}_4\text{S}_4(\text{SPh})_4]$ and UBFF simulation (—) for $^{57}\text{Fe}_4\text{S}_4\text{S}'_4$ model assuming T_d symmetry; sticks represent intensities of discrete normal modes. (b) UBFF simulation (—) for $^{57}\text{Fe}_4\text{S}_4(\text{SC}_6)_4$ model assuming C_2 symmetry; (c) UBFF simulation (—) for $^{57}\text{Fe}_4\text{S}_4(\text{SC}_6)_4$ model without symmetry constraints; (d) PED for $^{57}\text{Fe}_4\text{S}_4(\text{SC}_6)_4$ model without symmetry constraints. Curves represent contributions from Fe-S_t (—), Fe-S_{b1} (—), Fe-S_{b2} (—) and S-C (—) stretching.

