**SUPPORTING INFORMATION**

**Supplementary Figure Legend**

**Fig. S1** Conformational changes by the nitrite binding to the resting state. The red and green sticks represent the geometrical structures before and after the nitrite binding, respectively. The amino residues included in the sensor loop, His244, substrates and two catalytic residues are shown in figure upper part. The geometrical parameters around T2 Cu site after the binding to the resting state are shown in figure lower part. The interaction distances are shown in Å.

**Fig. S2** The conformational changes by the proton addition to [T1Cu(II)-T2Cu(II)-NO2- His244Nε]. The red and green sticks represent the geometrical structures before and after the proton addition. The amino residues included in the sensor loop and His249, ligand molecule and two catalytic residues are shown in figure upper part. The geometrical parameters around T2 Cu site after the proton addition are shown in figure lower part. The interaction distances are shown in Å.
Figure S2.