Electronic Supplementary Information (ESI)

Tetranuclear Ni(II) and Co(II) Schiff-base complexes with an M₄O₆ defective dicubane-like core: zero-field SMM behavior in the cobalt analogue

Ivan Nemec, Radovan Herchel, Marek Machata and Zdeněk Trávníček

Fig. S1 Comparison of the calculated powder diffraction pattern for 1 (red line) with the experimental diffraction patterns collected for 1 (green) and 2 (black). Note: temperature of the single-crystal measurement was 150 K, while temperature of the powder diffraction experiments was 298 K.
**Fig. S2** The calculated the isodensity surfaces of the HS spin states using B3LYP/ZORA/def2-TZVP(-f) for 1. The spin density is represented by yellow surface with the cutoff values of 0.01 e·boh$^{-3}$. Hydrogen atoms were omitted for clarity.

**Fig. S3** The calculated the isodensity surfaces of the HS spin states using B3LYP/ZORA/def2-TZVP(-f) for 2. The spin density is represented by yellow surface with the cutoff values of 0.01 e·boh$^{-3}$. Hydrogen atoms were omitted for clarity.
Fig. S4 The DFT derived $J_{\text{ab}}$-parameters for compounds 1 and 2 vs. average $\text{M}_a$-$\text{O}$-$\text{M}_b$ angles. Full symbols calculated for X-ray derived molecular geometries, empty symbols for BP86-optimized geometries.

Fig. S5 The DFT derived $J_{\text{ab}}$-parameters for compounds 1 and 2 vs. interatomic distance $d$(M$_a$···M$_b$). Full symbols calculated for X-ray derived molecular geometries, empty symbols for BP86-optimized geometries.
Fig. S6 The CASSCF/NEVPT2 principal axes of ZFS $D$-tensors (DX - magenta, DY – dark green, DZ – cyan) visualized together with the molecular structure $[\text{NiZn}_3\text{L}_4\text{(CH}_3\text{OH)}_2]$ derived from X-ray data of compound 1. Hydrogen atoms were omitted for clarity.

Fig. S7 The CASSCF/NEVPT2 principal axes of ZFS $D$-tensors (DX - magenta, DY – dark green, DZ – cyan) visualized together with the molecular structure $[\text{CoZn}_3\text{L}_4\text{(CH}_3\text{OH)}_2]$ derived from X-ray data of compound 1. Hydrogen atoms were omitted for clarity.
Table S1. Basic crystallographic data for compound 1

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Table S2. Energy levels (cm⁻¹) of ligand field multiplets in zero magnetic field derived from CASSCF/NEVPT2/ZORA/def2-TZVP(-f) calculations for 1-2 using molecular geometries from X-ray data.

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Table S6. Energy levels (cm$^{-1}$) of ligand field multiplets in zero magnetic field derived from CASSCF/NEVPT2/ZORA/def2-TZVP(-f) calculations for optimized geometries of 1 and 2.

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