Electronic Supplementary Information (ESI) concerning the manuscript:

Variation of the ground spin state in homo- and heterooctanuclear copper(II) and nickel(II) double-star complexes with a *meso*-helicate-type metallacryptand core

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Table of contents

Preparation of the ligands

H_2Et_2mpba. Ethyl oxalyl chloride ester (14.0 mL, 120 mmol) was added to a solution of 1,3-phenylenediamine (6.5 g, 60 mmol) and triethylamine (16.8 mL, 120 mmol) in THF (250 mL) under vigorous stirring at 0 °C. The reaction mixture was brought up to reflux for 1 h and then it was filtered to remove the precipitate of triethylammonium chloride. Solvent elimination under vacuum of the filtered solution afforded a white solid, which was treated with water, collected by filtration, washed with diethyl ether, and dried under vacuum (16.6 g, yield 90%). *Anal*. calc. for $C_{15}H_{18}N_2O_6$ (M = 308): C, 54.55; H, 5.19; N, 9.09 %. Found: C, 54.57; H, 5.18; N, 9.04 %. ^{1}H NMR [$(CD_3)_2SO$, ppm]: δ 1.32 (t, 6 H; 2 CH_3 from ethyl groups), 4.31 (q, 4 H; 2 CH_2 from ethyl groups), 7.34 (dt, 1 H; 5–H from phenylene group), 7.50 (dd, 2 H; 4–H and 6–H from phenylene group), 8.22 (t, 1 H; 2–H from phenylene group), 10.83 (s, 2 H; 2 NH). IR (KBr, cm^{-1}): 3349 (NH), 1725, 1715, 1698 (CO).

$H_2Et_2Mempba$. Ethyl oxalyl chloride ester (14.0 mL, 120 mmol) was added to a solution of 2,4-toluenediamine (7.3 g, 60 mmol) and triethylamine (16.8 mL, 120 mmol) in THF (250 mL) under vigorous stirring at 0 °C. The reaction mixture was brought up to reflux for 1 h and then it was filtered to remove the precipitate of triethylammonium chloride. Solvent elimination under vacuum of the filtered solution afforded a white solid, which was treated with water, collected by filtration, washed with diethyl ether, and dried under vacuum (15.5 g, yield 80%). *Anal*. calc. for $C_{15}H_{18}N_2O_6$ (M = 322): C, 55.90; H, 5.59; N, 8.70 %. Found: C, 56.12; H, 5.63; N, 8.47 %. ^{1}H NMR [$(CD_3)_2SO$, ppm]: δ 1.80 (t, 3 H; CH_3 from ethyl group), 1.82 (t, 3 H; CH_3 from ethyl group), 3.24 (s, 3 H; CH_3 from phenylene group), 4.79 (q, 2 H; CH_2 from ethyl group), 4.82 (q, 2 H; CH_2 from ethyl group), 7.72 (d, 1 H; 5–H from phenylene group), 8.11 (d, 1 H; 6–H from

phenylene group), 8.62 (s, 1 H; 2–H from phenylene group), 9.85 (s, 1 H; NH), 10.37 (s, 1 H; NH). IR (KBr, cm^{-1}): 3460 (NH), 1735, 1700, 1610 (CO).

Preparation of the binuclear nickel(II) complexes

$Na_8[Ni_2(mpba)_3] \cdot 12H_2O$. An aqueous solution (15 mL) of NaOH (0.96 g, 24.0 mmol) was added to a suspension of the diethyl ester derivative of H_4mpba (1.84 g, 6.0 mmol) in water (35 mL) under stirring. $Ni(NO_3)_2 \cdot 6H_2O$ (1.16 g, 4.0 mmol) dissolved in water (5 mL) was then added dropwise to the clear solution under stirring. The resulting deep green solution was filtered on paper to eliminate the small amount of solid particles. Solvent reduction under vacuum of the filtered solution afforded a blue-greenish solid, which was collected by filtration, washed with ethanol and diethyl ether, and air-dried (2.15 g, yield 85%). *Anal*. calc. for $C_{30}H_{36}N_6Na_8Ni_2O_{30}$ (M = 1261): C, 28.53; H, 2.85; N, 6.66 %. Found: C, 29.11; H, 3.03; N, 6.59 %. IR (KBr, cm^{-1}): 1585 (CO).

$Na_8[Ni_2(Mempba)_3] \cdot 18H_2O$. An aqueous solution (15 mL) of NaOH (0.96 g, 24.0 mmol) was added to a suspension of the diethyl ester derivative of H_4Mempba (1.93 g, 6.0 mmol) in water (35 mL) under stirring. $Ni(NO_3)_2 \cdot 6H_2O$ (1.16 g, 4.0 mmol) dissolved in water (5 mL) was then added dropwise to the clear solution under stirring. The resulting deep green solution was filtered on paper to eliminate the small amount of solid particles. Solvent reduction under vacuum of the filtered solution afforded a blue-greenish solid, which was collected by filtration, washed with ethanol and diethyl ether, and air-dried (2.10 g, yield 75%). *Anal*. calc. for $C_{33}H_{54}N_6Na_8Ni_2O_{36}$ (M = 1411): C, 28.07; H, 3.83; N, 5.95 %. Found: C, 28.31; H, 3.80; N, 5.85 %. IR (KBr, cm^{-1}): 1590 (C=O).

Magnetic Properties

Fig. S1 Temperature dependence of $\chi_M T$ of $Na_8[Ni_2(mpba)_3] \cdot 12H_2O$ (○) and $Na_8[Ni_2(Mempba)_3] \cdot 18H_2O$ (□). Solid lines are the best-fit curves (see text, footnote ‡).

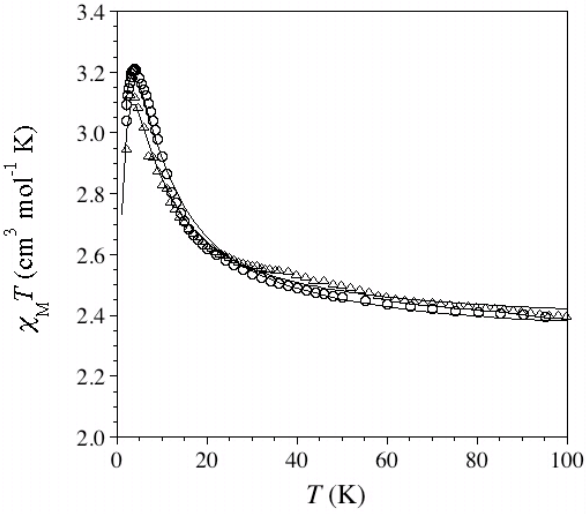

Table S1 Spin states and energies for an octanuclear copper(II) complex with a "dimer-of-tetramers" structure[a,b,c]

S^*_A	S_A	S^*_B	S_B	S	E	n
1/2	0	1/2	0	0	$-J$	4
1/2	0	1/2	1	1	$-2J$	4
1/2	0	3/2	1	1	$-1/2\,J$	2
1/2	0	3/2	2	2	$-5/2\,J$	2
1/2	1	1/2	0	1	$-2J$	4
1/2	1	1/2	1	0	$-3J$	4
1/2	1	1/2	1	1	$-3J$	4
1/2	1	1/2	1	2	$-3J$	4
1/2	1	3/2	1	0	$-3/2\,J$	2
1/2	1	3/2	1	1	$-3/2\,J$	2
1/2	1	3/2	1	2	$-3/2\,J$	2
1/2	1	3/2	2	1	$-7/2\,J$	2
1/2	1	3/2	2	2	$-7/2\,J$	2
1/2	1	3/2	2	3	$-7/2\,J$	2
3/2	1	1/2	0	1	$-1/2\,J$	2
3/2	1	1/2	1	0	$-3/2\,J$	2
3/2	1	1/2	1	1	$-3/2\,J$	2
3/2	1	1/2	1	2	$-3/2\,J$	2
3/2	1	3/2	1	0	$2\,j_{eff}$	1
3/2	1	3/2	1	1	j_{eff}	1
3/2	1	3/2	1	2	$-j_{eff}$	1
3/2	1	3/2	2	1	$-2J$	1
3/2	1	3/2	2	2	$-2J$	1
3/2	1	3/2	2	3	$-2J$	1
3/2	2	1/2	0	2	$-5/2\,J$	2
3/2	2	1/2	1	1	$-7/2\,J$	2
3/2	2	1/2	1	2	$-7/2\,J$	2
3/2	2	1/2	1	3	$-7/2\,J$	2
3/2	2	3/2	1	1	$-2J$	1
3/2	2	3/2	1	2	$-2J$	1
3/2	2	3/2	1	3	$-2J$	1
3/2	2	3/2	2	0	$-4J$	1
3/2	2	3/2	2	1	$-4J$	1
3/2	2	3/2	2	2	$-4J$	1
3/2	2	3/2	2	3	$-4J$	1
3/2	2	3/2	2	4	$-4J$	1

[a] S are the spin values of the Cu^{II}_8 entity, S_A and S_B are the spin values of each Cu^{II}_4 star unit, and S^*_A and S^*_B are the intermediate spin values corresponding to the coupling of the three peripheral Cu^{II} ions of each Cu^{II}_4 star unit. [b] E and n are the spin state energy and the number of degenerated spin states. [c] J and j_{eff} are the intratetramer and effective intertetramer coupling parameters as defined in eq S1 [with $S_{1A} = S_{3A} = S_{4A} = S_{5A} = S_{2B} = S_{6B} = S_{7B} = S_{8B} = S_{Cu} = 1/2$ and $S_A = S_B = 1$].

Table S2 Spin states and energies for an octanuclear nickel(II) copper(II) complex with a "dimer-of-tetramers" structure[a,b,c]

S^*_A	S_A	S^*_B	S_B	S	E	n
1/2	1/2	1/2	1/2	0	–3 J	4
1/2	1/2	1/2	1/2	1	–3 J	4
1/2	1/2	1/2	3/2	1	–9/2 J	4
1/2	1/2	1/2	3/2	2	–9/2 J	4
1/2	1/2	3/2	1/2	0	–3/2 J	2
1/2	1/2	3/2	1/2	1	–3/2 J	2
1/2	1/2	3/2	3/2	1	–3 J	2
1/2	1/2	3/2	3/2	2	–3 J	2
1/2	1/2	3/2	5/2	2	–11/2 J	2
1/2	1/2	3/2	5/2	3	–11/2 J	2
1/2	3/2	1/2	1/2	1	–9/2 J	4
1/2	3/2	1/2	1/2	2	–9/2 J	4
1/2	3/2	1/2	3/2	0	–6 J	4
1/2	3/2	1/2	3/2	1	–6 J	4
1/2	3/2	1/2	3/2	2	–6 J	4
1/2	3/2	1/2	3/2	3	–6 J	4
1/2	3/2	3/2	1/2	1	–3 J	2
1/2	3/2	3/2	1/2	2	–3 J	2
1/2	3/2	3/2	3/2	0	–9/2 J	2
1/2	3/2	3/2	3/2	1	–9/2 J	2
1/2	3/2	3/2	3/2	2	–9/2 J	2
1/2	3/2	3/2	3/2	3	–9/2 J	2
1/2	3/2	3/2	5/2	1	–7 J	2
1/2	3/2	3/2	5/2	2	–7 J	2
1/2	3/2	3/2	5/2	3	–7 J	2
1/2	3/2	3/2	5/2	4	–7 J	2
3/2	1/2	1/2	1/2	0	–3/2 J	2
3/2	1/2	1/2	1/2	1	–3/2 J	2
3/2	1/2	1/2	3/2	1	–3 J	2
3/2	1/2	1/2	3/2	2	–3 J	2
3/2	1/2	3/2	1/2	0	–3/4 j_{eff}	1
3/2	1/2	3/2	1/2	1	–1/4 j_{eff}	1
3/2	1/2	3/2	3/2	1	–3/2 J	1
3/2	1/2	3/2	3/2	2	–3/2 J	1
3/2	1/2	3/2	5/2	2	–4 J	1
3/2	1/2	3/2	5/2	3	–4 J	1
3/2	3/2	1/2	1/2	1	–3 J	2
3/2	3/2	1/2	1/2	2	–3 J	2
3/2	3/2	1/2	3/2	0	–9/2 J	2
3/2	3/2	1/2	3/2	1	–9/2 J	2
3/2	3/2	1/2	3/2	2	–9/2 J	2
3/2	3/2	1/2	3/2	3	–9/2 J	2

3/2	3/2	3/2	1/2	1	–3/2 *J*	1
3/2	3/2	3/2	1/2	2	–3/2 *J*	1
3/2	3/2	3/2	3/2	0	–3 *J*	1
3/2	3/2	3/2	3/2	1	–3 *J*	1
3/2	3/2	3/2	3/2	2	–3 *J*	1
1/2	3/2	3/2	3/2	3	–3 *J*	1
1/2	3/2	3/2	5/2	1	–11/2 *J*	1
1/2	3/2	3/2	5/2	2	–11/2 *J*	1
1/2	3/2	3/2	5/2	3	–11/2 *J*	1
1/2	3/2	3/2	5/2	4	–11/2 *J*	1
3/2	5/2	1/2	1/2	2	–11/2 *J*	2
3/2	5/2	1/2	1/2	3	–11/2 *J*	2
3/2	5/2	1/2	3/2	1	–7 *J*	2
3/2	5/2	1/2	3/2	2	–7 *J*	2
3/2	5/2	1/2	3/2	3	–7 *J*	2
3/2	5/2	1/2	3/2	4	–7 *J*	2
3/2	5/2	3/2	1/2	2	–4 *J*	1
3/2	5/2	3/2	1/2	3	–4 *J*	1
3/2	5/2	3/2	3/2	1	–11/2 *J*	1
3/2	5/2	3/2	3/2	2	–11/2 *J*	1
3/2	5/2	3/2	3/2	3	–11/2 *J*	1
3/2	5/2	3/2	3/2	4	–11/2 *J*	1
3/2	5/2	3/2	5/2	0	–8 *J*	1
3/2	5/2	3/2	5/2	1	–8 *J*	1
3/2	5/2	3/2	5/2	2	–8 *J*	1
3/2	5/2	3/2	5/2	3	–8 *J*	1
3/2	5/2	3/2	5/2	4	–8 *J*	1
3/2	5/2	3/2	5/2	5	–8 *J*	1

[a] S are the spin values of the $Ni^{II}_2Cu^{II}_6$ entity, S_A and S_B are the spin values of each $Ni^{II}Cu^{II}_3$ star unit, and S^*_A and S^*_B are the intermediate spin values corresponding to the coupling of the three peripheral Cu^{II} ions of each $Ni^{II}Cu^{II}_3$ star unit. [b] E and n are the spin state energy and the number of degenerated spin states. [c] J and j_{eff} are the intratetramer and effective intertetramer coupling parameters as defined in eq S1 [with $S_{1A} = S_{2B} = S_{Ni} = 1$, $S_{3A} = S_{4A} = S_{5A} = S_{6B} = S_{7B} = S_{8B} = S_{Cu} = 1/2$, and $S_A = S_B = 1/2$].

Table S3 Spin states and energies for an octanuclear nickel(II) complex with a "dimer-of-tetramers" structure[a,b,c]

S^*_A	S_A	S^*_B	S_B	S	E	n
0	1	0	1	0	–8 J	1
0	1	0	1	1	–8 J	1
0	1	0	1	2	–8 J	1
0	1	1	0	1	–6 J	3
0	1	1	1	0	–7 J	3
0	1	1	1	1	–7 J	3
0	1	1	1	2	–7 J	3
0	1	1	2	1	–9 J	3
0	1	1	2	2	–9 J	3
0	1	1	2	3	–9 J	3
0	1	2	1	0	–5 J	2
0	1	2	1	1	–5 J	2
0	1	2	1	2	–5 J	2
0	1	2	2	1	–7 J	2
0	1	2	2	2	–7 J	2
0	1	2	2	3	–7 J	2
0	1	2	3	2	–10 J	2
0	1	2	3	3	–10 J	2
0	1	2	3	4	–10 J	2
0	1	3	2	1	–4 J	1
0	1	3	2	2	–4 J	1
0	1	3	2	3	–4 J	1
0	1	3	3	2	–7 J	1
0	1	3	3	3	–7 J	1
0	1	3	3	4	–7 J	1
0	1	3	4	3	–11 J	1
0	1	3	4	4	–11 J	1
0	1	3	4	5	–11 J	1
1	0	0	1	1	–6 J	3
1	0	1	0	0	–4 J	9
1	0	1	1	1	–5 J	9
1	0	1	2	2	–7 J	9
1	0	2	1	1	–3 J	6
1	0	2	2	2	–5 J	6
1	0	2	3	3	–8 J	6
1	0	3	2	2	–2 J	3
1	0	3	3	3	–5 J	3
1	0	3	4	4	–9 J	3
1	1	0	1	0	–7 J	3
1	1	0	1	1	–7 J	3
1	1	0	1	2	–7 J	3
1	1	1	0	1	–5 J	9

1	1	1	1	0	–6 *J*	9
1	1	1	1	1	–6 *J*	9
1	1	1	1	2	–6 *J*	9
1	1	1	2	1	–8 *J*	9
1	1	1	2	2	–8 *J*	9
1	1	1	2	3	–8 *J*	9
1	1	2	1	0	–4 *J*	6
1	1	2	1	1	–4 *J*	6
1	1	2	1	2	–4 *J*	6
1	1	2	2	1	–6 *J*	6
1	1	2	2	2	–6 *J*	6
1	1	2	2	3	–6 *J*	6
1	1	2	3	2	–9 *J*	6
1	1	2	3	3	–9 *J*	6
1	1	2	3	4	–9 *J*	6
1	1	3	2	1	–3 *J*	3
1	1	3	2	2	–3 *J*	3
1	1	3	2	3	–3 *J*	3
1	1	3	3	2	–6 *J*	3
1	1	3	3	3	–6 *J*	3
1	1	3	3	4	–6 *J*	3
1	1	3	4	3	–10 *J*	3
1	1	3	4	4	–10 *J*	3
1	1	3	4	5	–10 *J*	3
1	2	0	1	1	–9 *J*	3
1	2	0	1	2	–9 *J*	3
1	2	0	1	3	–9 *J*	3
1	2	1	0	2	–7 *J*	9
1	2	1	1	1	–8 *J*	9
1	2	1	1	2	–8 *J*	9
1	2	1	1	3	–8 *J*	9
1	2	1	2	0	–10 *J*	9
1	2	1	2	1	–10 *J*	9
1	2	1	2	2	–10 *J*	9
1	2	1	2	3	–10 *J*	9
1	2	1	2	4	–10 *J*	9
1	2	2	1	1	–6 *J*	6
1	2	2	1	2	–6 *J*	6
1	2	2	1	3	–6 *J*	6
1	2	2	2	0	–8 *J*	6
1	2	2	2	1	–8 *J*	6
1	2	2	2	2	–8 *J*	6
1	2	2	2	3	–8 *J*	6
1	2	2	2	4	–8 *J*	6
1	2	2	3	1	–11 *J*	6
1	2	2	3	2	–11 *J*	6

1	2	2	3	3	–11 *J*	6
1	2	2	3	4	–11 *J*	6
1	2	2	3	5	–11 *J*	6
1	2	3	2	0	–5 *J*	3
1	2	3	2	1	–5 *J*	3
1	2	3	2	2	–5 *J*	3
1	2	3	2	3	–5 *J*	3
1	2	3	2	4	–5 *J*	3
1	2	3	3	1	–8 *J*	3
1	2	3	3	2	–8 *J*	3
1	2	3	3	3	–8 *J*	3
1	2	3	3	4	–8 *J*	3
1	2	3	3	5	–8 *J*	3
1	2	3	4	2	–12 *J*	3
1	2	3	4	3	–12 *J*	3
1	2	3	4	4	–12 *J*	3
1	2	3	4	5	–12 *J*	3
1	2	3	4	6	–12 *J*	3
2	1	0	1	0	–5 *J*	2
2	1	0	1	1	–5 *J*	2
2	1	0	1	2	–5 *J*	2
2	1	1	0	1	–3 *J*	6
2	1	1	1	0	–4 *J*	6
2	1	1	1	1	–4 *J*	6
2	1	1	1	2	–4 *J*	6
2	1	1	2	1	–6 *J*	6
2	1	1	2	2	–6 *J*	6
2	1	1	2	3	–6 *J*	6
2	1	2	1	0	–2 *J*	4
2	1	2	1	1	–2 *J*	4
2	1	2	1	2	–2 *J*	4
2	1	2	2	1	–4 *J*	4
2	1	2	2	2	–4 *J*	4
2	1	2	2	3	–4 *J*	4
2	1	2	3	2	–7 *J*	4
2	1	2	3	3	–7 *J*	4
2	1	2	3	4	–7 *J*	4
2	1	3	2	1	–*J*	2
2	1	3	2	2	–*J*	2
2	1	3	2	3	–*J*	2
2	1	3	3	2	–4 *J*	2
2	1	3	3	3	–4 *J*	2
2	1	3	3	4	–4 *J*	2
2	1	3	4	3	–8 *J*	2
2	1	3	4	4	–8 *J*	2
2	1	3	4	5	–8 *J*	2

2	2	0	1	1	–7 *J*	2
2	2	0	1	2	–7 *J*	2
2	2	0	1	3	–7 *J*	2
2	2	1	0	2	–5 *J*	6
2	2	1	1	1	–6 *J*	6
2	2	1	1	2	–6 *J*	6
2	2	1	1	3	–6 *J*	6
2	2	1	2	0	–8 *J*	6
2	2	1	2	1	–8 *J*	6
2	2	1	2	2	–8 *J*	6
2	2	1	2	3	–8 *J*	6
2	2	1	2	4	–8 *J*	6
2	2	2	1	1	–4 *J*	4
2	2	2	1	2	–4 *J*	4
2	2	2	1	3	–4 *J*	4
2	2	2	2	0	–6 *J*	4
2	2	2	2	1	–6 *J*	4
2	2	2	2	2	–6 *J*	4
2	2	2	2	3	–6 *J*	4
2	2	2	2	4	–6 *J*	4
2	2	2	3	1	–9 *J*	4
2	2	2	3	2	–9 *J*	4
2	2	2	3	3	–9 *J*	4
2	2	2	3	4	–9 *J*	4
2	2	2	3	5	–9 *J*	4
2	2	3	2	0	–3 *J*	2
2	2	3	2	1	–3 *J*	2
2	2	3	2	2	–3 *J*	2
2	2	3	2	3	–3 *J*	2
2	2	3	2	4	–3 *J*	2
2	2	3	3	1	–6 *J*	2
2	2	3	3	2	–6 *J*	2
2	2	3	3	3	–6 *J*	2
2	2	3	3	4	–6 *J*	2
2	2	3	3	5	–6 *J*	2
2	2	3	4	2	–10 *J*	2
2	2	3	4	3	–10 *J*	2
2	2	3	4	4	–10 *J*	2
2	2	3	4	5	–10 *J*	2
2	2	3	4	6	–10 *J*	2
2	3	0	1	2	–10 *J*	2
2	3	0	1	3	–10 *J*	2
2	3	0	1	4	–10 *J*	2
2	3	1	0	3	–8 *J*	6
2	3	1	1	2	–9 *J*	6
2	3	1	1	3	–9 *J*	6

2	3	1	1	4	–9 *J*	6
2	3	1	2	1	–11 *J*	6
2	3	1	2	2	–11 *J*	6
2	3	1	2	3	–11 *J*	6
2	3	1	2	4	–11 *J*	6
2	3	1	2	5	–11 *J*	6
2	3	2	1	2	–7 *J*	4
2	3	2	1	3	–7 *J*	4
2	3	2	1	4	–7 *J*	4
2	3	2	2	1	–9 *J*	4
2	3	2	2	2	–9 *J*	4
2	3	2	2	3	–9 *J*	4
2	3	2	2	4	–9 *J*	4
2	3	2	2	5	–9 *J*	4
2	3	2	3	0	–12 *J*	4
2	3	2	3	1	–12 *J*	4
2	3	2	3	2	–12 *J*	4
2	3	2	3	3	–12 *J*	4
2	3	2	3	4	–12 *J*	4
2	3	2	3	5	–12 *J*	4
2	3	2	3	6	–12 *J*	4
2	3	3	2	1	–6 *J*	2
2	3	3	2	2	–6 *J*	2
2	3	3	2	3	–6 *J*	2
2	3	3	2	4	–6 *J*	2
2	3	3	2	5	–6 *J*	2
2	3	3	3	0	–9 *J*	2
2	3	3	3	1	–9 *J*	2
2	3	3	3	2	–9 *J*	2
2	3	3	3	3	–9 *J*	2
2	3	3	3	4	–9 *J*	2
2	3	3	3	5	–9 *J*	2
2	3	3	3	6	–9 *J*	2
2	3	3	4	1	–13 *J*	2
2	3	3	4	2	–13 *J*	2
2	3	3	4	3	–13 *J*	2
2	3	3	4	4	–13 *J*	2
2	3	3	4	5	–13 *J*	2
2	3	3	4	6	–13 *J*	2
2	3	3	4	7	–13 *J*	2
3	2	0	1	1	–4 *J*	1
3	2	0	1	2	–4 *J*	1
3	2	0	1	3	–4 *J*	1
3	2	1	0	2	–2 *J*	3
3	2	1	1	1	–3 *J*	3
3	2	1	1	2	–3 *J*	3

3	2	1	1	3	$-3J$	3
3	2	1	2	0	$-5J$	3
3	2	1	2	1	$-5J$	3
3	2	1	2	2	$-5J$	3
3	2	1	2	3	$-5J$	3
3	2	1	2	4	$-5J$	3
3	2	2	1	1	$-J$	2
3	2	2	1	2	$-J$	2
3	2	2	1	3	$-J$	2
3	2	2	2	0	$-3J$	2
3	2	2	2	1	$-3J$	2
3	2	2	2	2	$-3J$	2
3	2	2	2	3	$-3J$	2
3	2	2	2	4	$-3J$	2
3	2	2	3	1	$-6J$	2
3	2	2	3	2	$-6J$	2
3	2	2	3	3	$-6J$	2
3	2	2	3	4	$-6J$	2
3	2	2	3	5	$-6J$	2
3	2	3	2	0	$6j_{\mathrm{eff}}$	1
3	2	3	2	1	$5j_{\mathrm{eff}}$	1
3	2	3	2	2	$3j_{\mathrm{eff}}$	1
3	2	3	2	3	0	1
3	2	3	2	4	$-4j_{\mathrm{eff}}$	1
3	2	3	3	1	$-3J$	1
3	2	3	3	2	$-3J$	1
3	2	3	3	3	$-3J$	1
3	2	3	3	4	$-3J$	1
3	2	3	3	5	$-3J$	1
3	2	3	4	2	$-7J$	1
3	2	3	4	3	$-7J$	1
3	2	3	4	4	$-7J$	1
3	2	3	4	5	$-7J$	1
3	2	3	4	6	$-7J$	1
3	3	0	1	2	$-7J$	1
3	3	0	1	3	$-7J$	1
3	3	0	1	4	$-7J$	1
3	3	1	0	3	$-5J$	3
3	3	1	1	2	$-6J$	3
3	3	1	1	3	$-6J$	3
3	3	1	1	4	$-6J$	3
3	3	1	2	1	$-8J$	3
3	3	1	2	2	$-8J$	3
3	3	1	2	3	$-8J$	3
3	3	1	2	4	$-8J$	3
3	3	1	2	5	$-8J$	3

3	3	2	1	2	–4 *J*	2
3	3	2	1	3	–4 *J*	2
3	3	2	1	4	–4 *J*	2
3	3	2	2	1	–6 *J*	2
3	3	2	2	2	–6 *J*	2
3	3	2	2	3	–6 *J*	2
3	3	2	2	4	–6 *J*	2
3	3	2	2	5	–6 *J*	2
3	3	2	3	0	–9 *J*	2
3	3	2	3	1	–9 *J*	2
3	3	2	3	2	–9 *J*	2
3	3	2	3	3	–9 *J*	2
3	3	2	3	4	–9 *J*	2
3	3	2	3	5	–9 *J*	2
3	3	2	3	6	–9 *J*	2
3	3	3	2	1	–3 *J*	1
3	3	3	2	2	–3 *J*	1
3	3	3	2	3	–3 *J*	1
3	3	3	2	4	–3 *J*	1
3	3	3	2	5	–3 *J*	1
3	3	3	3	0	–6 *J*	1
3	3	3	3	1	–6 *J*	1
3	3	3	3	2	–6 *J*	1
3	3	3	3	3	–6 *J*	1
3	3	3	3	4	–6 *J*	1
3	3	3	3	5	–6 *J*	1
3	3	3	3	6	–6 *J*	1
3	3	3	4	1	–10 *J*	1
3	3	3	4	2	–10 *J*	1
3	3	3	4	3	–10 *J*	1
3	3	3	4	4	–10 *J*	1
3	3	3	4	5	–10 *J*	1
3	3	3	4	6	–10 *J*	1
3	3	3	4	7	–10 *J*	1
3	4	0	1	3	–11 *J*	1
3	4	0	1	4	–11 *J*	1
3	4	0	1	5	–11 *J*	1
3	4	1	0	4	–9 *J*	3
3	4	1	1	3	–10 *J*	3
3	4	1	1	4	–10 *J*	3
3	4	1	1	5	–10 *J*	3
3	4	1	2	2	–12 *J*	3
3	4	1	2	3	–12 *J*	3
3	4	1	2	4	–12 *J*	3
3	4	1	2	5	–12 *J*	3
3	4	1	2	6	–12 *J*	3

3	4	2	1	3	–8 *J*	2
3	4	2	1	4	–8 *J*	2
3	4	2	1	5	–8 *J*	2
3	4	2	2	2	–10 *J*	2
3	4	2	2	3	–10 *J*	2
3	4	2	2	4	–10 *J*	2
3	4	2	2	5	–10 *J*	2
3	4	2	2	6	–10 *J*	2
3	4	2	3	1	–13 *J*	2
3	4	2	3	2	–13 *J*	2
3	4	2	3	3	–13 *J*	2
3	4	2	3	4	–13 *J*	2
3	4	2	3	5	–13 *J*	2
3	4	2	3	6	–13 *J*	2
3	4	2	3	7	–13 *J*	2
3	4	3	2	2	–7 *J*	1
3	4	3	2	3	–7 *J*	1
3	4	3	2	4	–7 *J*	1
3	4	3	2	5	–7 *J*	1
3	4	3	2	6	–7 *J*	1
3	4	3	3	1	–10 *J*	1
3	4	3	3	2	–10 *J*	1
3	4	3	3	3	–10 *J*	1
3	4	3	3	4	–10 *J*	1
3	4	3	3	5	–10 *J*	1
3	4	3	3	6	–10 *J*	1
3	4	3	3	7	–10 *J*	1
3	4	3	4	0	–14 *J*	1
3	4	3	4	1	–14 *J*	1
3	4	3	4	2	–14 *J*	1
3	4	3	4	3	–14 *J*	1
3	4	3	4	4	–14 *J*	1
3	4	3	4	5	–14 *J*	1
3	4	3	4	6	–14 *J*	1
3	4	3	4	7	–14 *J*	1
3	4	3	4	8	–14 *J*	1

[a] S are the spin values of the Ni^{II}_8 entity, S_A and S_B are the spin values of each Ni^{II}_4 star unit, and S^*_A and S^*_B are the intermediate spin values corresponding to the coupling of the three peripheral high-spin Ni^{II} ions of each Ni^{II}_4 star unit. [b] E and n are the spin state energy and the number of degenerated spin states. [c] J and j_{eff} are the intratetramer and effective intertetramer coupling parameters as defined in eq S1 [with $S_{1A} = S_{3A} = S_{4A} = S_{5A} = S_{2B} = S_{6B} = S_{7B} = S_{8B} = S_{Ni} = 1$, and $S_A = S_B = 2$].