Supplementary Information

Experimental and theoretical insights into magnetic exchange and anisotropy in multinuclear cobalt complexes displaying slow relaxation

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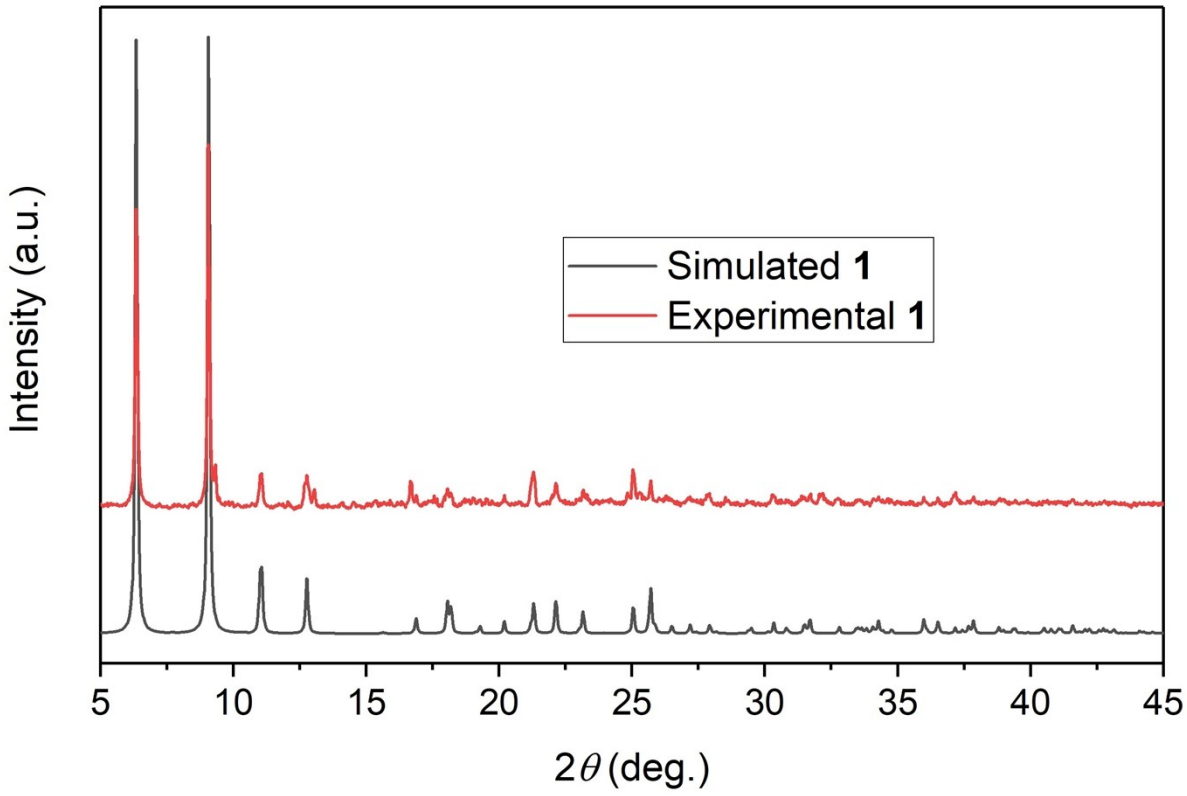


Fig. S1. PXRD data of complex **1**.

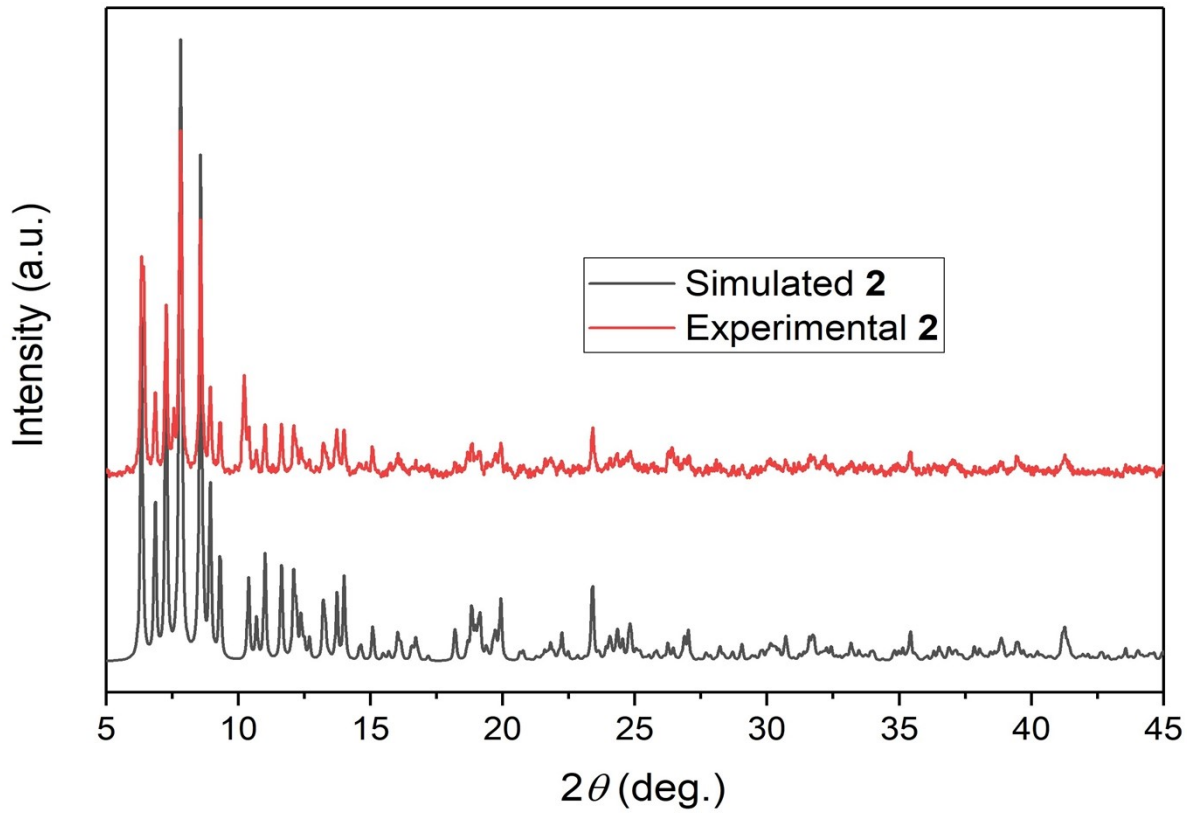


Fig. S2. PXRD data of complex **2**.

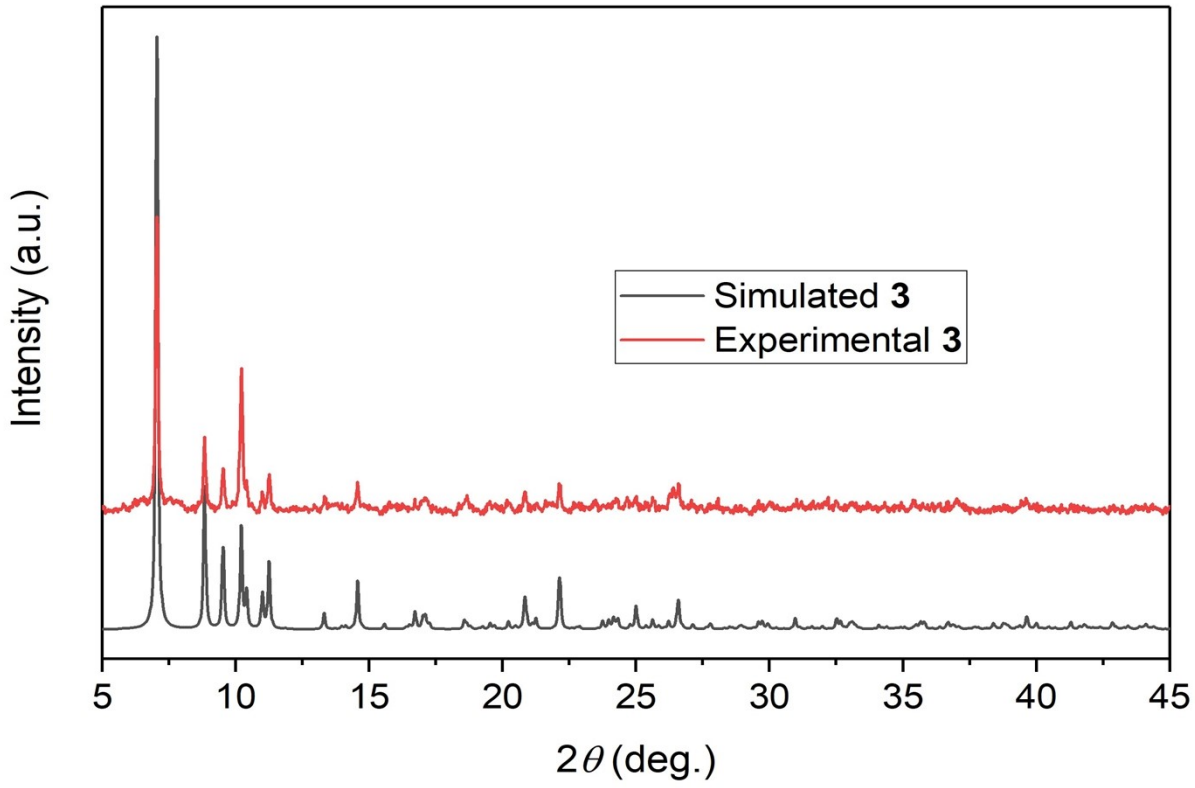


Fig. S3. PXRD data of complex **3**.

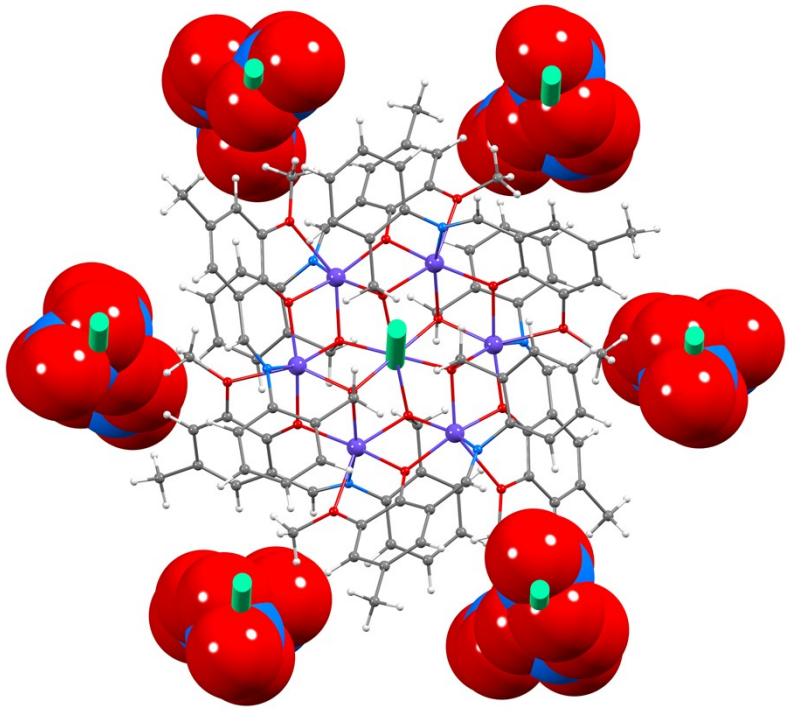


Fig. S4. A part of the crystal packing in **1** showing the complex cation surrounded by six helical pillars of nitrate counter ions.

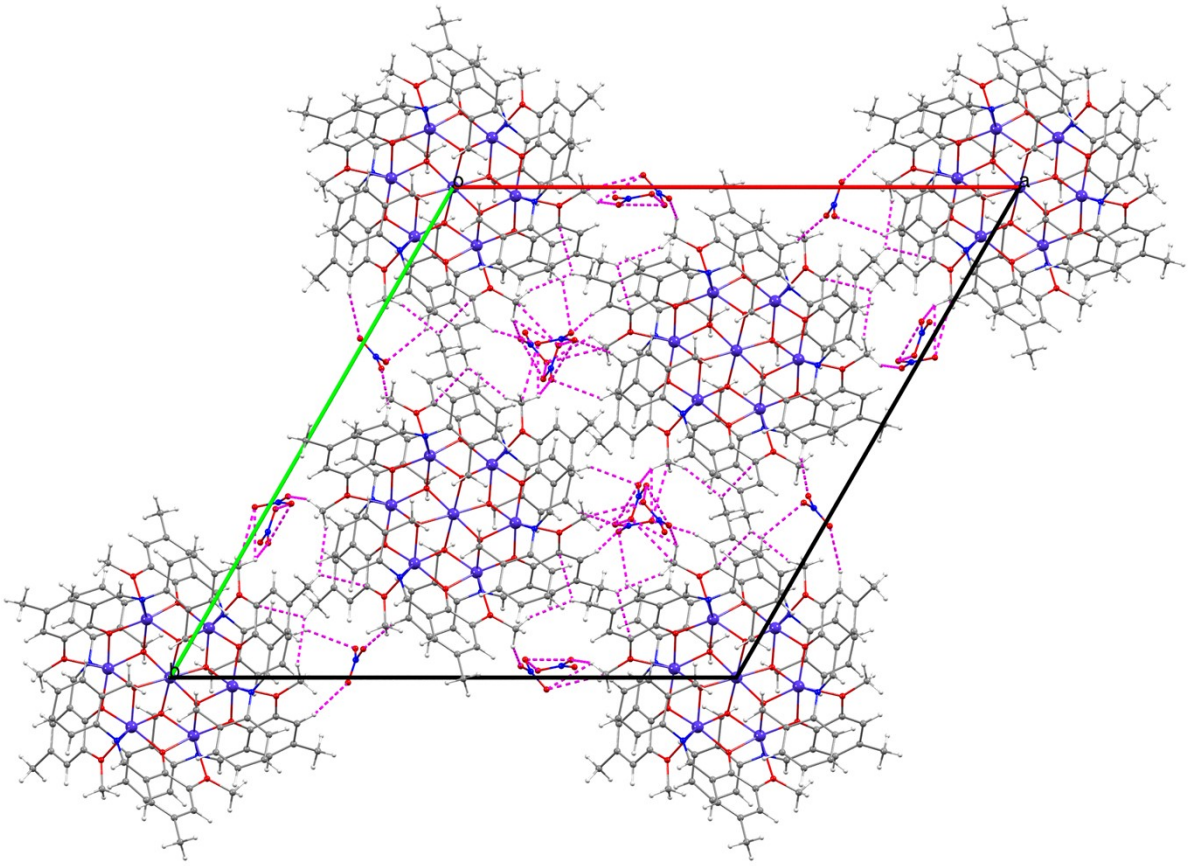


Fig. S5. Crystal packing in complex **1** showing different non-covalent interactions.

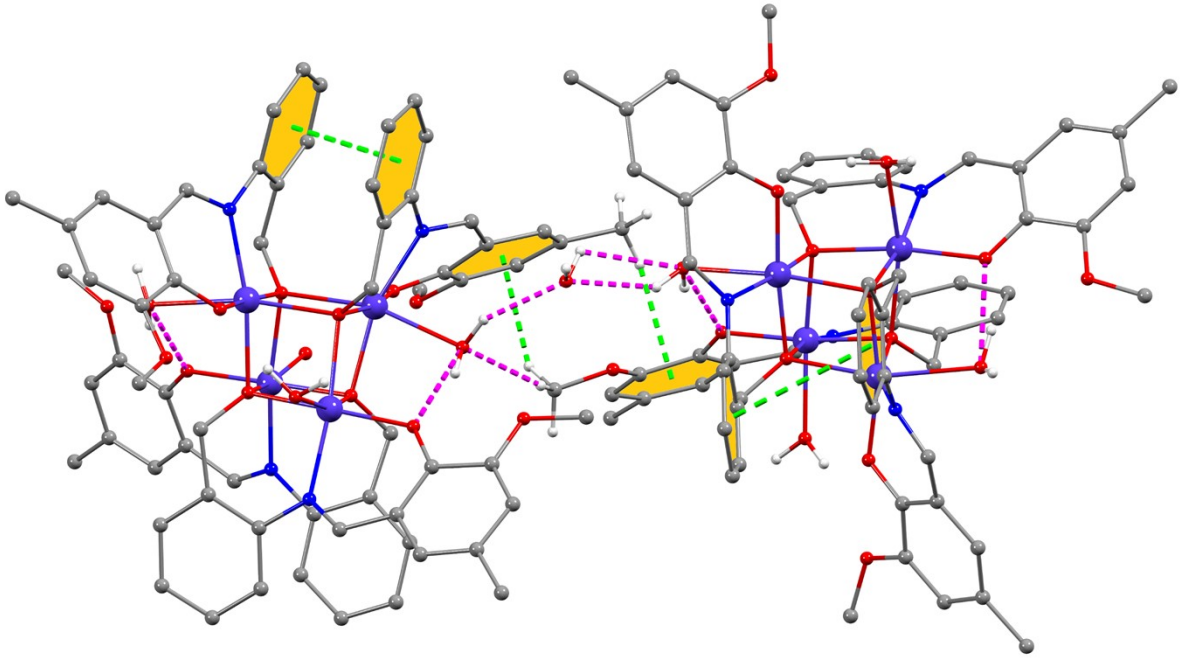


Fig. S6. Crystal packing in complex **2** showing different non-covalent interactions.

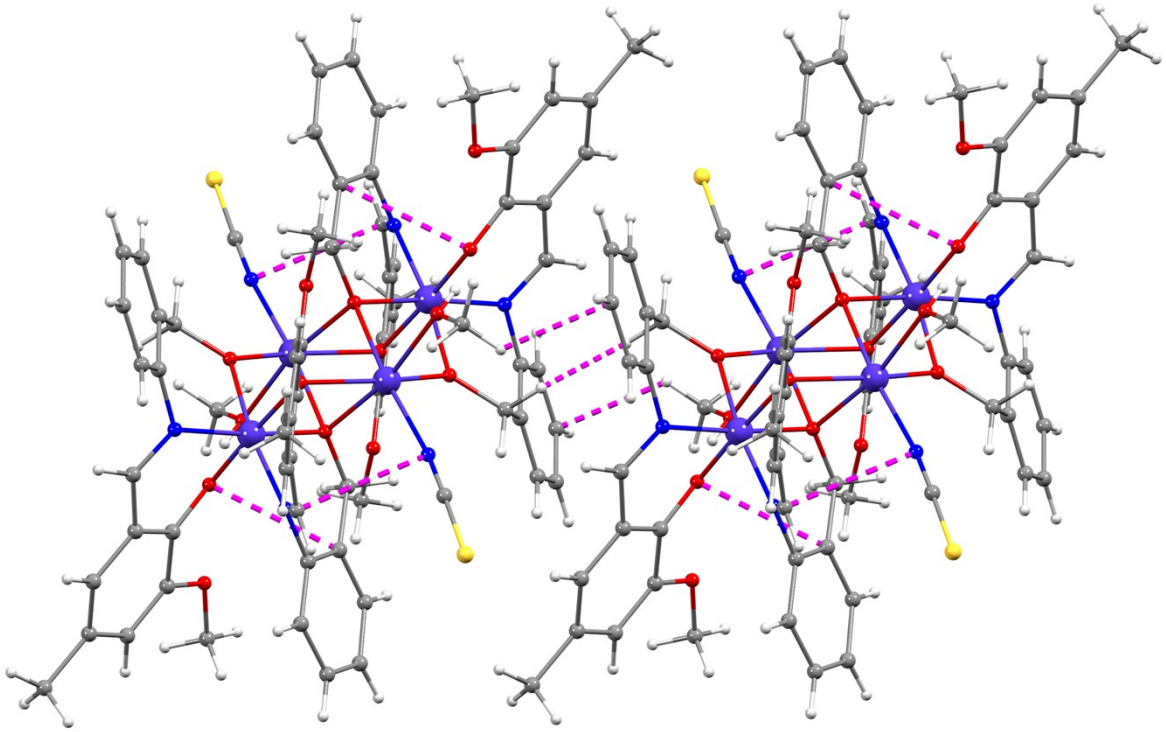


Fig. S7. Crystal packing in complex **3** showing different non-covalent interactions.

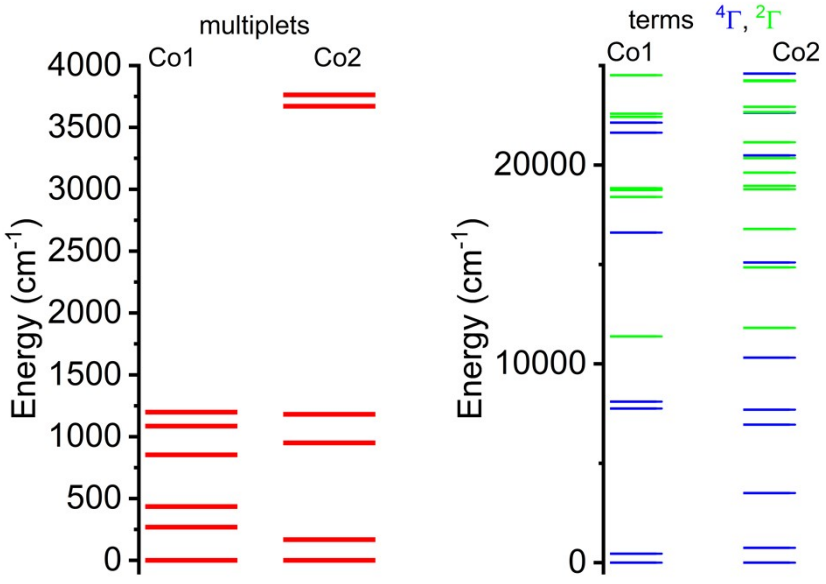


Fig. S8. Energies of ligand field terms and the lowest six multiplets of **1** obtained from SA-CASSCF/NEVPT2 calculations.

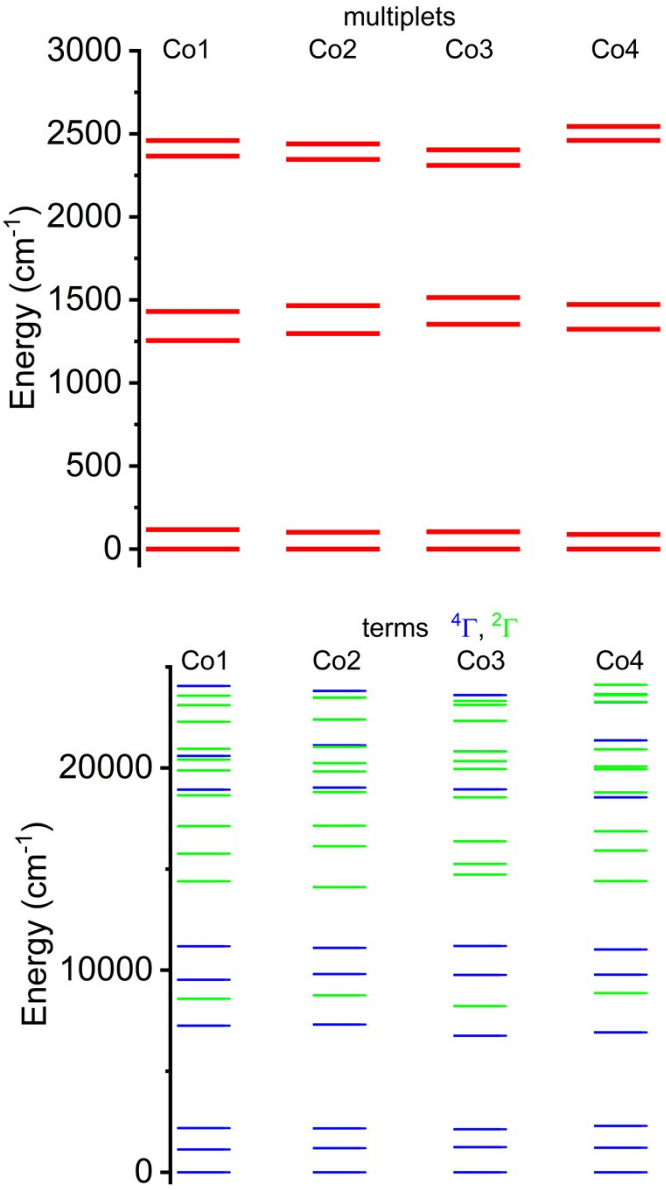


Fig. S9. Energies of ligand field terms (lower panel) and the lowest six multiplets (upper panel) of **2** obtained from SA-CASSCF/NEVPT2 calculations.

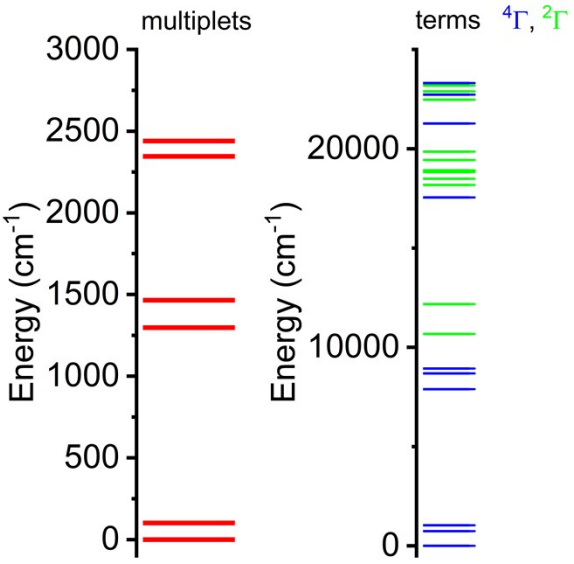
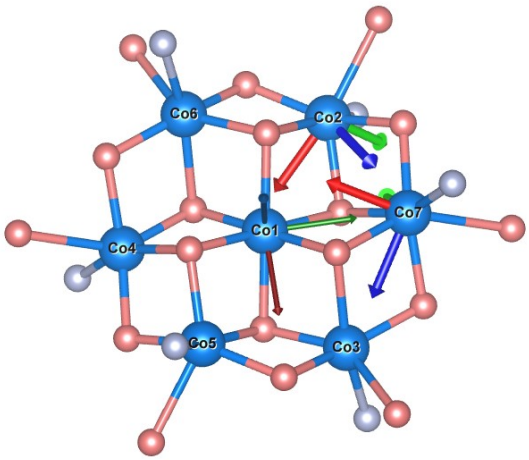
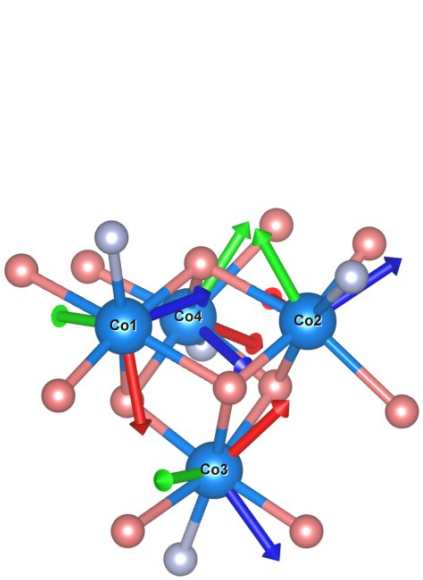


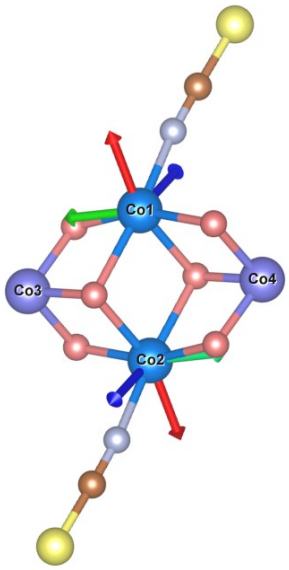
Fig. S10. Energies of ligand field terms and the lowest six multiplets of **3** obtained from SA-CASSCF/NEVPT2 calculations.



a) **1**



b) **2**



c) **3**

Fig S11. The orientation of the *D*-tensor axis frame of outer Co^{II} ions in **1**, **2**, and **3** (panels a, b, and c) resulting from SA-CASSCF/NEVPT2 calculations done in ORCA for **1** is shown by arrows. The color coding is red – D_x, green - D_y, and blue – D_z. Plotted using VESTA software. In the case of the central Co1 ion in **1**, the axis frame of the g-tensor of the ground state Kramers doublet is shown with the following color coding: wine red – g_x, olive green - g_y, and navy blue – g_z.

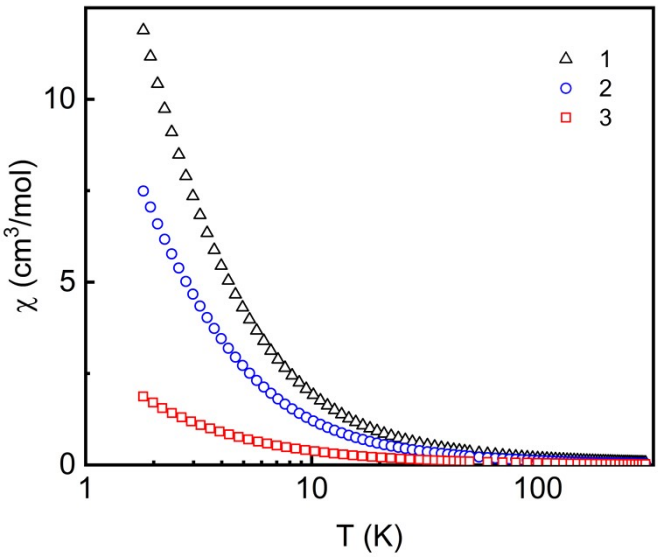


Fig S12. Temperature dependence of susceptibility of **1**, **2**, and **3** measured in an applied field of 1 kOe.

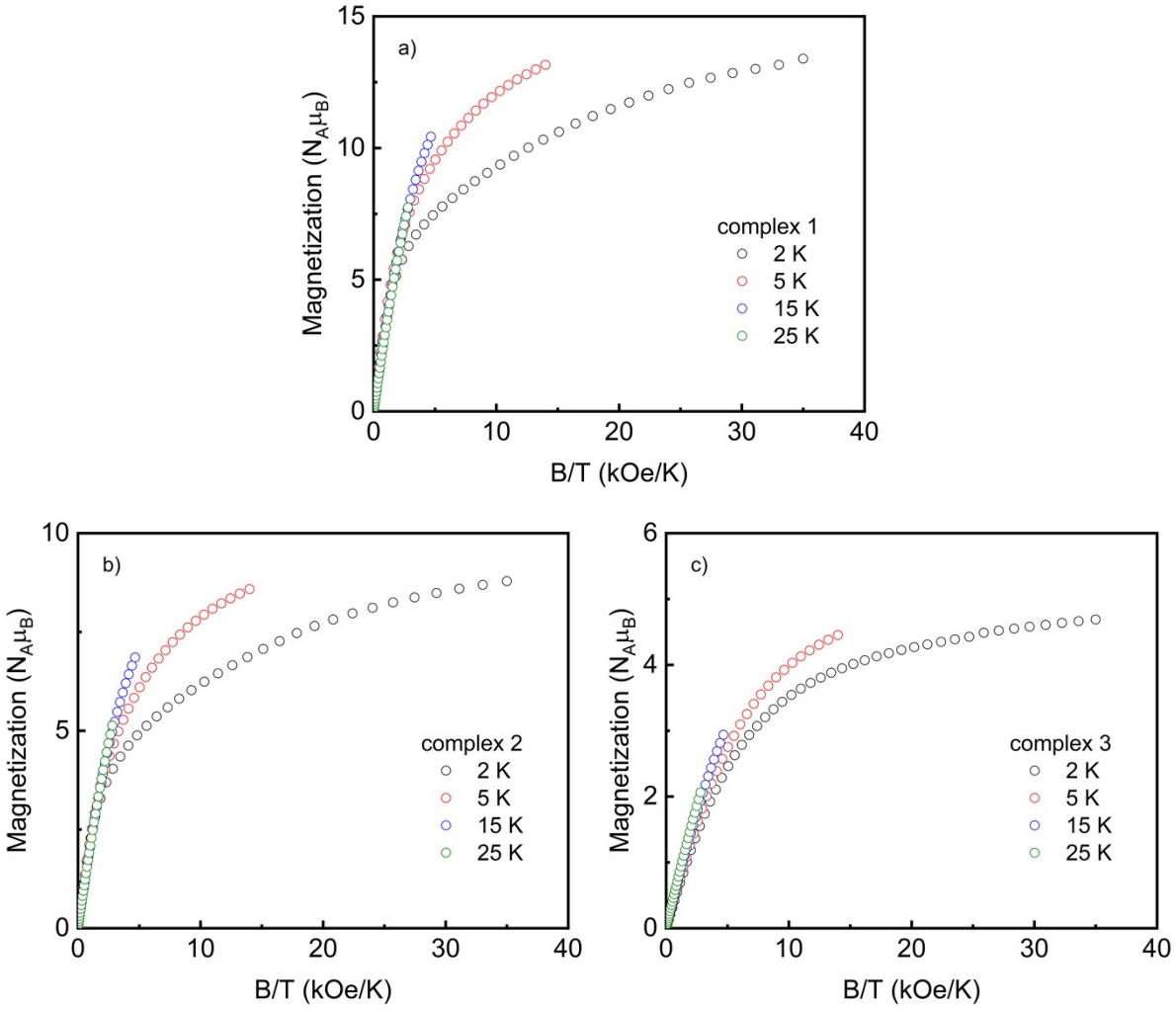


Fig S13. Field dependence of magnetization of **1** (a), **2** (b), and **3** (c) in reduced units measures at different temperatures.

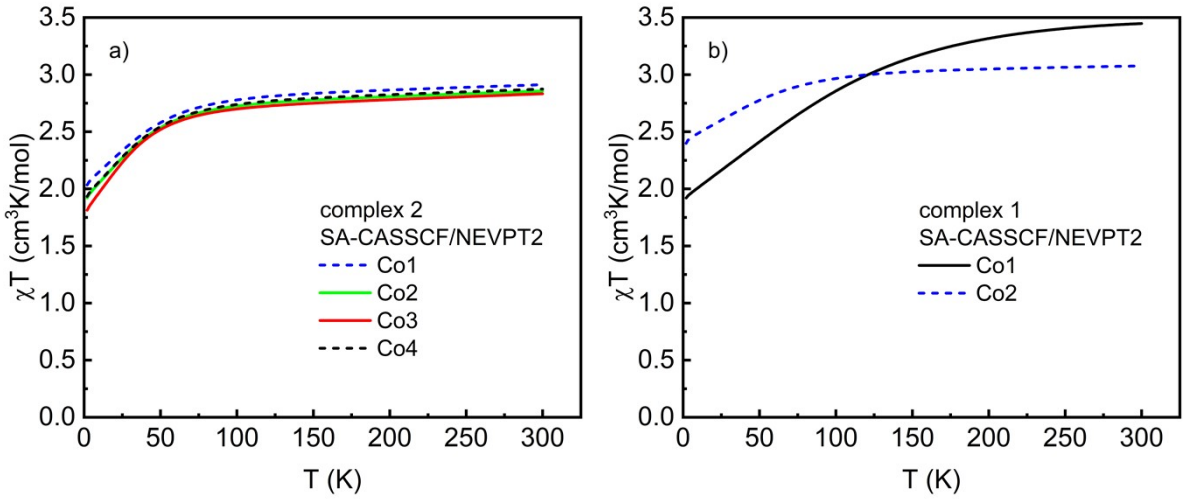


Fig S14. Temperature dependence of χT for each Co^{II} ion in the molecular unit of **2** (a) and **1** (b) as obtained from CAS(7,5) SA-CASSCF/NEVPT2 approach.

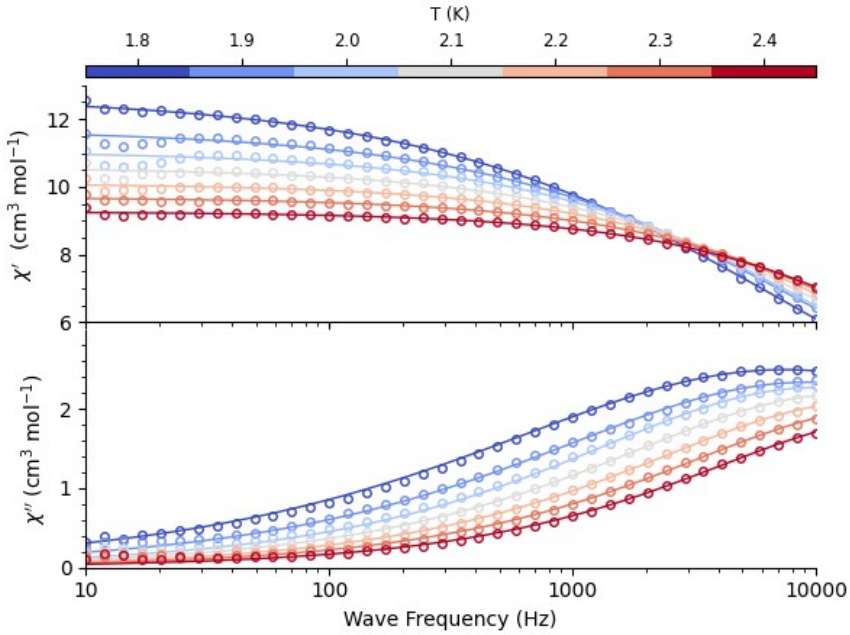


Fig S15. Frequency dependence of real and imaginary components of AC susceptibility of **1** obtained in zero DC magnetic field at different temperatures (open symbols), solid lines represent the fit of the generalized Debye model.

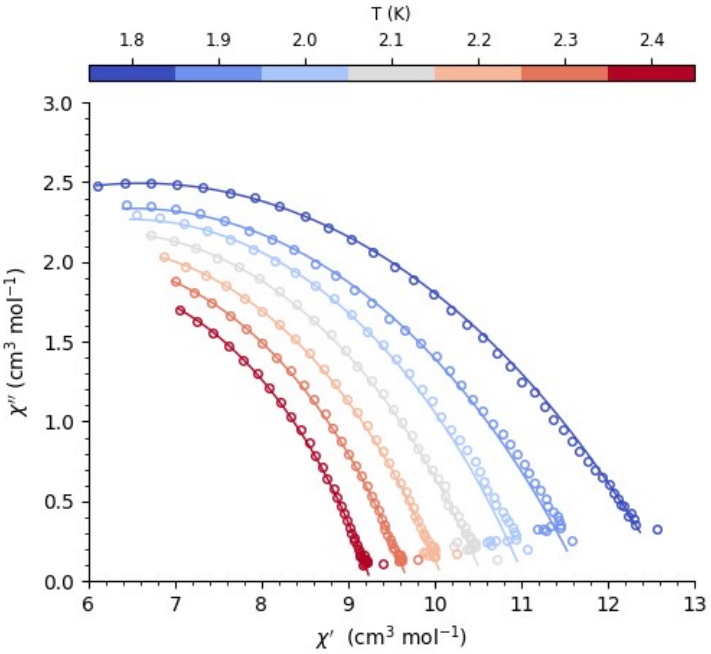


Fig S16. Cole-Cole plots of **1** obtained in zero DC magnetic field at different temperatures (open symbols), solid lines represent the fit of the generalized Debye model.

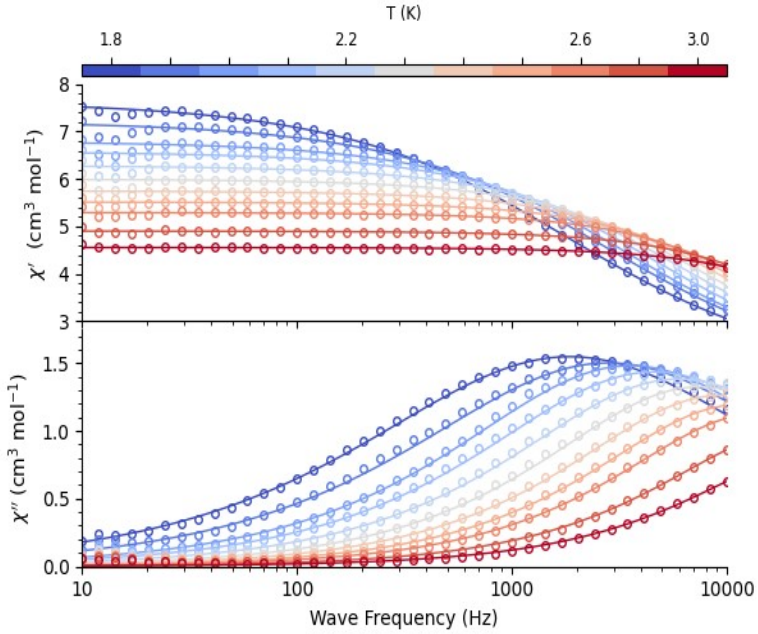


Fig S17. Frequency dependence of real and imaginary components of AC susceptibility of **2** obtained in zero DC magnetic field at different temperatures (open symbols), solid lines represent the fit of the generalized Debye model.

Table S1. Crystal data and structure refinement for complexes **1-3** and Schiff base H_2L**.**

Identification code	**1**	**2**	**3**	Schiff base H_2L
Empirical formula	$C_{96}H_{90}Co_7N_8O_{24}$	$C_{64}H_{71}Co_4N_4O_{17.5}$	$C_{66.5}H_{63.5}N_6O_{14}S_2Co_4$	$C_{32}H_{34}N_2O_6$
Formula weight	2152.26	1411.96	1470.57	542.61
Temperature/K	150.05	150.00	150.00	100.02(10)
Crystal system	trigonal	monoclinic	monoclinic	monoclinic
Space group	$R\bar{3}$	$P2_1/n$	$P2_1/n$	$P2_1/n$
a/Å	27.8412(8)	15.2081(19)	10.281(7)	13.3196(12)
b/Å	27.8412(8)	24.307(2)	16.080(11)	7.1596(6)
c/Å	10.6569(2)	17.027(2)	20.016(13)	28.319(2)
α/°	90	90	90	90
β/°	90	90.524(4)	92.053(17)	95.339(8)
γ/°	120	90	90	90
Volume/Å^3	7153.8(4)	6294.1(12)	3307(4)	2688.9(4)
Z	3	4	2	4
ρ_{calc}g/cm^3	1.499	1.490	1.477	1.340
μ/mm^{-1}	1.265	1.110	1.118	0.093
F(000)	3309.0	2924.0	1513.0	1152.0
2Θ range for data collection/°	5.854 to 50.688	4.924 to 50.81	4.522 to 54.186	3.514 to 59.922
Reflections collected	28952	117475	67508	23623
Independent reflections	2871 [R_{int} = 0.0420, R_{sigma} = 0.0196]	11560 [R_{int} = 0.0638, R_{sigma} = 0.0280]	7282 [R_{int} = 0.0410, R_{sigma} = 0.0224]	6113 [R_{int} = 0.0724, R_{sigma} = 0.0797]
Data/restraints/parameters	2871/48/230	11560/2/831	7282/0/433	6113/0/369
Goodness-of-fit on F^2	1.099	1.056	1.082	1.045
Final R indexes [I>=2σ (I)]	R_1 = 0.0588, wR_2 = 0.1974	R_1 = 0.0447, wR_2 = 0.1092	R_1 = 0.0353, wR_2 = 0.0799	R_1 = 0.0547, wR_2 = 0.1302
Final R indexes [all data]	R_1 = 0.0733, wR_2 = 0.2256	R_1 = 0.0584, wR_2 = 0.1191	R_1 = 0.0518, wR_2 = 0.0892	R_1 = 0.0934, wR_2 = 0.1471
Largest diff. peak/hole / e Å^{-3}	0.77/-0.51	1.65/-1.19	0.61/-0.57	0.30/-0.31

Table S2. Bond distances around metal centers in **1-3**.[#]

Bond Distances (in Å)

	1		**2**		**3**
Co1–O3[1]	2.118(3)	Co2–O10	2.020(2)	Co1–O1	1.9348(17)
Co1–O3[2]	2.118(3)	Co2–O12	2.126(2)	Co1–O3	1.9117(19)
Co1–O3[3]	2.118(3)	Co2–O9	2.154(2)	Co1–O4	1.8889(17)
Co1–O3	2.118(3)	Co2–O3	1.969(2)	Co1–O6	1.8889(18)
Co1–O3[4]	2.118(3)	Co2–O14	2.268(2)	Co1–N1	1.929(2)
Co1–O3[5]	2.118(3)	Co2–N2	2.096(3)	Co1–N2	1.915(2)
Co2–O1[4]	1.992(3)	Co1–O10	2.170(2)	Co2–O1[6]	2.118(2)
Co2–O1	2.039(3)	Co1–O11	2.129(2)	Co2–O3	2.1934(18)
Co2–O2	2.381(4)	Co1–O9	2.004(2)	Co2–O3[6]	2.1126(18)
Co2–N1[4]	2.075(4)	Co1–O1	1.955(2)	Co2–O6	1.9814(19)
Co2–O3[4]	2.043(3)	Co1–O13	2.224(3)	Co2–N3	2.051(2)
Co2–O3[5]	2.186(3)	Co1–N1	2.117(3)	Co2–O7	2.126(2)
		Co3–O10	2.109(2)		
		Co3–O11	2.009(2)		
		Co3–O12	2.129(2)		
		Co3–O5	1.977(2)		
		Co3–O15	2.400(3)		
		Co3–N3	2.073(3)		
		Co4–O11	2.147(2)		
		Co4–O12	2.024(2)		
		Co4–O9	2.084(2)		
		Co4–O7	1.995(2)		
		Co4–N4	2.053(3)		
		Co4–O16	2.301(3)		

Bond Angles (in deg.)

	1		**2**		**3**
Co2–O1–Co2[2]	104.71(14)	Co2–O10–Co1	100.95(9)	Co1–O1–Co2[1]	98.62(8)
Co1–O3–Co2[1]	92.65(12)	Co2–O10–Co3	97.06(9)	Co1–O3–Co2	96.19(7)
Co2[2]–O3–Co1	96.85(13)	Co3–O10–Co1	94.63(8)	Co1–O3–Co2[1]	99.56(8)
Co2[2]–O3–Co2[1]	97.96(13)	Co1–O11–Co4	92.29(9)	Co2[1]–O3–Co2	99.26(8)
		Co3–O11–Co1	98.90(9)	Co1–O6–Co2	104.51(8)
		Co3–O11–Co4	101.32(9)		
		Co2–O12–Co3	93.29(8)		
		Co4–O12–Co2	97.45(9)		
		Co4–O12–Co3	101.43(9)		
		Co1–O9–Co2	102.02(10)		
		Co1–O9–Co4	97.91(10)		
		Co4–O9–Co2	94.79(9)		

symmetry code: [1]1-y,1+x-y,+z; [2]2/3-x,4/3-y,1/3-z; [3]2/3-y+x,1/3+x,1/3-z; [4]-1/3+y,1/3-x+y,1/3-z; [5]+y-x,1-x,+z; [6]2-x,1-y,1-z

Table S3. BVS analysis for the cobalt centers in **1–3** using the empirical equation of $\Sigma v = \Sigma \exp[(R_0 - R)/0.37]$, where r_0 = 1.685 for Co^{II}–O, 1.720 for Co^{II}–N, 1.637 for Co^{III}–O and 1.690 for Co^{III}–N.

Complex	Bond	R	BVS calculated using Co^{II}–N/O R_0 Parameters	BVS calculated using Co^{III}–N/O R_0 Parameters
1	Co1–O3[1]	2.118(3)	0.310283	0.272532
	Co1–O3[2]	2.118(3)	0.310283	0.272532
	Co1–O3[3]	2.118(3)	0.310283	0.272532
	Co1–O3	2.118(3)	0.310283	0.272532
	Co1–O3[4]	2.118(3)	0.310283	0.272532
	Co1–O3[5]	2.118(3)	0.310283	0.272532
			$\Sigma v(Co^{II})$ = 1.862	$\Sigma v(Co^{III})$ = 1.635
	Co2–O1[4]	1.992(3)	0.436167	0.3831
	Co2–O1	2.039(3)	0.384137	0.3374
	Co2–O2	2.381(4)	0.152425	0.13388
	Co2–N1[4]	2.075(4)	0.3831	0.353264
	Co2–O3[4]	2.043(3)	0.380006	0.333772
	Co2–O3[5]	2.186(3)	0.258191	0.226778
			$\Sigma v(Co^{II})$ = 1.994	**$\Sigma v(Co^{III})$ = 1.768**
2	Co2–O10	2.020(2)	0.404378	0.355178
	Co2–O12	2.126(2)	0.303646	0.266702
	Co2–O9	2.154(2)	0.281516	0.247264
	Co2–O3	1.969(2)	0.464141	0.40767
	Co2–O14	2.268(2)	0.206868	0.181699
	Co2–N2	2.096(3)	0.361962	0.333772
			$\Sigma v(Co^{II})$ = 2.022	**$\Sigma v(Co^{III})$ = 1.792**
	Co1–O10	2.170(2)	0.269601	0.2368
	Co1–O11	2.129(2)	0.301194	0.264549
	Co1–O9	2.004(2)	0.422248	0.370874
	Co1–O1	1.955(2)	0.482039	0.423391
	Co1–O13	2.224(3)	0.34199	0.315356
	Co1–N1	2.117(3)	0.232991	0.204643
			$\Sigma v(Co^{II})$ = 2.050	**$\Sigma v(Co^{III})$ = 1.815**
	Co3–O10	2.109(2)	0.322248	0.279242
	Co3–O11	2.009(2)	0.422248	0.365896
	Co3–O12	2.129(2)	0.305292	0.264549
	Co3–O5	1.977(2)	0.460393	0.39895
	Co3–O15	2.400(3)	0.146766	0.127179
	Co3–N3	2.073(3)	0.385176	0.355178
			$\Sigma v(Co^{II})$ = 2.042	**$\Sigma v(Co^{III})$ = 1.791**
	Co4–O11	2.147(2)	0.286892	0.251987
	Co4–O12	2.024(2)	0.40003	0.351359
	Co4–O9	2.084(2)	0.340147	0.298762
	Co4–O7	1.995(2)	0.432645	0.380006
	Co4–N4	2.053(3)	0.40657	0.166195
	Co4–O16	2.301(3)	0.189216	0.374906
			$\Sigma v(Co^{II})$ = 2.055	**$\Sigma v(Co^{III})$ = 1.823**
3	Co1–O1	1.9348(17)	0.509088	0.447148
	Co1–O3	1.9117(19)	0.541884	0.475955
	Co1–O4	1.8889(17)	0.576326	0.506206
	Co1–O6	1.8889(18)	0.576326	0.506206
	Co1–N1	1.929(2)	0.568437	0.524166
	Co1–N2	1.915(2)	0.590357	0.54438
			$\Sigma v(Co^{II})$ = 3.362	**$\Sigma v(Co^{III})$ = 3.004**
	Co2–O1[6]	2.118(2)	0.310283	0.272532
	Co2–O3	2.1934(18)	0.253079	0.222287
	Co2–O3[6]	2.1126(18)	0.314845	0.276538
	Co2–O6	1.9814(19)	0.448843	0.394234
	Co2–N3	2.051(2)	0.303646	0.266702
	Co2–O7	2.126(2)	0.408773	0.376938
			$\Sigma v(Co^{II})$ = 2.039	**$\Sigma v(Co^{III})$ = 1.809**

symmetry code: [1]1-y,1+x-y,+z; [2]2/3-x,4/3-y,1/3-z; [3]2/3-y+x,1/3+x,1/3-z; [4]-1/3+y,1/3-x+y,1/3-z; [5]+y-x,1-x,+z; [6]2-x,1-y,1-z

Table S4. Shape analysis for Co^{II} or Co^{III} centers in **1-3**.

Label	Shape	Symmetry	**1**		**2**				**3**	
			Co1	Co2	Co1	Co2	Co3	Co4	Co1	Co2
HP-6	Hexagon	D_{6h}	25.801	31.610	30.349	30.705	31.295	29.552	32.539	28.355
PPY-6	Pentagonal pyramid	C_{5v}	27.052	17.728	23.715	22.937	24.312	20.907	28.743	26.127
OC-6	Octahedron	Oh	**0.682**	**6.532**	**1.193**	**1.474**	**1.368**	**2.364**	**0.194**	**1.161**
TPR-6	Trigonal prism	D_{3h}	16.513	**4.426**	12.531	12.072	12.553	10.363	15.542	13.851
JPPY-6	Johnson pentagonal pyramid J2	C_{5v}	30.043	21.371	27.072	25.754	27.061	23.699	31.955	28.800

Table S5 Assignment of high-spin (HS) and BS states a four-spin cluster of **2**. Atom numbers as in Hamiltonian Eq. 2 correspond to Co1-Co4 ions in **2**.

Co1	Co2	Co3	Co4	state	M_S
up	up	up	up	HS	6
down	up	up	up	BS1	2
up	down	up	up	BS2	2
up	up	down	up	BS3	2
up	up	up	down	BS4	2
down	down	up	up	BS5	0
down	up	down	up	BS6	0
down	up	up	down	BS7	0

Table S6: Magneto-structural data of reported Co^{II}_7 disk/wheel-shaped structures.

Complex	Magnetic Interaction	SMM?	U_{eff} [K]	H_{DC} [Oe]	Ref.
$\{[Co_7(C_2H_5OH)1.5(H_2O)0.5(Hdatrz)_2(\mu3\text{-}OH)_4(ip)_5]\cdot 2.5H_2O\cdot C_2H_5OH\}_n$	AFM	Yes	17.65	3000	S1
$[Co_7(H_2O)_4(ade)_2(\mu3\text{-}OH)_6(sip)_2]_n$	FM	No	-	-	
$[Co_7(L^1)_{12}]Cl_2$	AFM	No	-	-	S2
$[Co_7(OH)_4(Hesp)_2(esp)_4(MeCN)_2]\cdot 4MeCN$	AFM	No	-	-	S3
$[Co_7(vab)_6](NO_3)_2\cdot 2H_2O$	FM	Yes	18.8 (estimated)	0	S4
$[Co_7(L^2)_6(\mu_3\text{-}OCH_3)_6](ClO_4)_2\cdot H_2O$	FM	No			S5
$[Co_7(bm)_{12}]\cdot(ClO_4)_2\cdot 13H_2O$	FM	No			S6
$[Co_7(L^3)_6](NO_3)_2\cdot 2CH_3OH$	FM	Yes	-		S7
$[Co_7(mmimp)_6(MeO)_6]\cdot Br_2$	FM	No			S8
$[Co_7(emmp)_6(CH_3O)_6]\cdot(ClO_4)_2$	FM	No			S9
$[Co_7(heb)_6(\mu_3\text{-}OCH3)_6]\cdot(ClO_4)_2$	FM	No			S10
$[Co_7(bhqe)_3(OH)_2(H_2O)_6]\cdot 2C_2H_5OH\cdot H_2O$	FM	Yes	21/13	0	S11
$[Co_7(mmp)_6(CH_3O)_6]\cdot(ClO_4)_2$	FM	No			S12
$[Co_7(\mu_3\text{-}OH)_6(L^4)_6](ClO_4)_2\cdot 12H_2O$	FM	No			S13
$[Co_7(\mu_3\text{-}CH_3O)_6\text{-}(L^4)_6](ClO_4)_2$	FM	No			
$[Co_7(\mu_3\text{-}N_3)_6(L^4)_6](ClO_4)_2$	FM	Yes	-		
$[Co_7(L^5)_9(OH)_2(OAc)_{2.7}(MeO)_{0.3}(H_2O)]\cdot 4.6MeOH\cdot 3.3H_2O$	AFM	No			S14
$[Co_7(eimp)_6(OMe)_6]\cdot 2ClO_4$	FM	No			S15
$[Co_7(\mu\text{-}L^6)_6(\mu\text{-}OMe)_6]Cl_2$	FM	No			S16
$[Co_7(immp)_6(CH_3O)_6]\cdot 2ClO_4$	FM	No			S17
$[Co_7(OH)_6(L^7)_6](NO_3)_2\cdot 2MeOH$	FM	No			S18
$[Co_7(bzp)_6(N_3)_9(CH_3O)_3](ClO_4)_2\cdot 2H_2O$	FM	Yes	-		S19
1	FM	Yes	8.89	0	This Work

Abbreviations: Hdatrz = 3,5-diamino-1,2,4-triazole; **ade**$^-$ = adeninate, **ip**$^{2-}$ = isophthalate; **sip**$^{3-}$: 5-sulfoisophthalate; **HL**^{1} = pyridine-2-ylmethanol; **esp** = α,α,α',α'-tetramethyl-1,3-benzenedipropionate; **H_2vab** = 2-[(2-hydroxymethyl-phenylimino)-methyl]-6-methoxy-phenol; **HL**^{2} = N-(2-hidroxy-3-methoxybenzylidene)-2-methoxy-benzylamine; **Hbm** = (1*H*-Benzimidazol)-methanol; **H_2L^3** = 2-(2-hydroxy-3-methoxybenzylideneamino)phenol; **Hmmimp** = 2-methoxy-6-methyliminomethyl-phenol; **Hemmp** = 2-ethyliminomethyl-6-methoxy-phenol; **Hheb** = 2-hydroxy-3-ethoxy-benzaldehyde; **H_4bhqe** = 2-(hydroxymethyl)quinolin-8-ol; **mmp** = 2-methoxy-6-methyliminomethyl-phenol anion; **HL**^{4} = 2-methoxy-6-[(methylimino)methyl]phenol; H**L**^{5} = 11*H*-indeno[1,2-*b*]quinoxalin-11-one oxime; Heimp = 2-ethoxy-6-(iminomethyl)phenol; **HL**^{6} = 2-{(3-ethoxypropylimino)methyl}-6-methoxyphenol; **immp** = 2-iminomethyl-6-methoxy-phenolic anion; **HL**^{7} = 2-iminomethyl-6-methoxyphenol; **bzp** = 2-benzoyl pyridine.

Table S7: Magneto-structural data of reported Co^{II}_4-cubane and $Co^{III}_2Co^{II}$ defective dicubane types structures

Complex	**Magnetic Interaction**	**SMM?**	**U_{eff} [K]**	**H_{DC} [Oe]**	**Ref.**
$[Co_4(pmab)_4Cl_4]$	FM (Weak)	Yes	-	2000	S20
$[Co_4(pmab)_4(OBz)_2]Cl_2$	AFM				
$[Co_4(hmp)_4(MeOH)_4Cl_4]$	FM	Yes	-		S21
$[C(NH_2)_3]_8\{Co_4(cit)_4\}\cdot 4H_2O$	-	Yes	21	1000	S22
$[C(NH_2)_3]_8[Co_4(cit)_4]\cdot 8H_2O$	AFM	Yes	13.1	Close to zero	S23
$[Co_4(TTA)_4(OMe)_4(EtOH)_4]$	FM	Yes	-		S24
[Co4(TTA)4(OMe)4(MeOH)4] (2)	FM	No			
$[Co_4(L^8)_4(l3\text{-}OMe)_4(MeOH)_4]$	FM	No			S25
[Co4(L^9)4(m_3-OMe)4(MeOH)4]	FM	Yes			
$[Co_4(bm)_4Br_4(CH_3OH)_3(H_2O)]\cdot 2CH_3OH$	FM	No			S26
$[Co_4(bm)_4Br_4(CH_3CH_2OH)_4]$	FM	No			
$[Co_4(mbm)_4Br_4(CH_3OH)_4]$	FM	Yes	26.7	0	
$[Co_4(mbm)_4Br_4(CH_3CH_2OH)_4]$	FM	Yes	21.0	0	
$[Co_4(ntfa)_4(CH_3O)_4(CH_3OH)_4]$	FM	Yes	30.28	2000	S27
$[Co_4(vab)_4(H_2O)_4]\cdot 4THF\cdot 4H_2O$	AFM	No			S4
$[Co_4(heb)_4(m_3\text{-}OCH_3)_4(m_1\text{-}HOCH_3)_4]\cdot(H_2O)_2$	AFM	No			S10
$[Co_4(bm)_4Cl_4(C_3H_7OH)_4]$	FM	No			S6
$[Co_4(bm)_4(m\text{-}HCO_2)_2(m_2\text{-}HCO_2)_2(C_3H_7OH)_2]$	AFM	No			
[CoII4(Cl)4(HL^{10})4]	FM	Yes	55.8	5000	S28
$[Co_4L^{11}_4]\cdot CH_3OH$	FM	Yes	-		S29
$[Co_4(HL^{12})_4]$	FM	Yes	-		S30
$[Co(\mu_3\text{-}Cl)Cl(HL\cdot S)]_4$	FM	Yes	20.1	0	S31
2	FM	Yes	12.86	0	This work

Abbreviations: **Hpmab** = [2-{(2-pyridinylmethylene)amino}benzenemethanol; **hmp** = 2-(hydroxymethyl)pyridine; **cit** = citrate ion; TTA = 4,4,4-trifluoro-1-thienoyl-2,4-butanedione; HL^8 = 3,5-dichloro-2-hydroxy-benzaldehyde; **HL^9** = 2-hydroxy-3-methoxybenzaldehyde; **bm** = (1H-benzo[d]imidazol-2-yl)methanolate; **mbm** = 1-Me-(1H-benzo[d]imidazol-2-yl)methanolate; **ntfa** = 4,4,4-trifluoro-1-(2-naphthyl)-1,3-butanedione; **H_2vab** = 2-[(2-hydroxymethyl-phenylimino)-methyl]-6-methoxy-phenol; **Hheb** = 2-hydroxy-3-ethoxy-benzaldehyde; **Hbm** = (1H-Benzimidazol)-methanol; **H_2L^{10}** = N-substituted diethanolamine ligand; **H_2L^{11}** = 1-(((2-hydroxy-2,3-dihydro-1H-inden-1-yl)imino)methyl)naphthalen-2-ol; **H_3L^{12}** = (*E*)-3-((2-hydroxy-3-methoxybenzylidene)amino)propane-1,2-diol; , **HL·S** = pyridyl-alcohol-type ligand.

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