Electronic Supplementary Information

Optimal diamagnetic dilution concentration for suppressing dipole-dipole interaction in single-ion magnets

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Table S1. Crystallographic data and structural refinement parameters for complexes **Co_1**, **$Co_{0.2}Zn_{0.8}$** and **$Co_{0.1}Zn_{0.9}$**

	Co_1	**$Co_{0.2}Zn_{0.8}$**	**$Co_{0.1}Zn_{0.9}$**
Formula	$C_{20}H_{18}CoN_2O_3$	$C_{20}H_{18}Co_{0.2}Zn_{0.8}N_2O_3$	$C_{20}H_{18}Co_{0.1}Zn_{0.9}N_2O_3$
f w	393.28	398.47	399.73
T / K	296(2)	296(2)	296(2)
λ / Å	0.71073	0.71073	0.71073
Crystal system	Orthorhombic	Orthorhombic	Orthorhombic
Space group	*Pbcn*	*Pbcn*	*Pbcn*
a / Å	7.2787(16)	7.225(3)	7.2405(12)
b / Å	9.128(2)	9.060 (4)	9.0605(13)
c / Å	25.164(6)	25.026(10)	25.074(4)
α / º	90	90	90
β / º	90	90	90
γ / º	90	90	90
V / Å^3	1671.8(7)	1638.2(5)	1644.9(5)
Z	4	4	4
D_c / g cm^{-3}	1.559	1.621	1.610
μ / mm^{-1}	1.050	1.523	1.454
F(000)	808	824	822
θ / °	1.618 to 25.013	3.256 to 27.528	3.249 to 27.59
Reflns collected	8606	9607	12053
Reflns unique	1475	1867	1903
R_{int}	0.1132	0.0732	0.0744
GOF on F^2	1.010	1.062	1.086
R_1 [I> 2σ(I)][a]	0.0421	0.0479	0.0509
wR_2 [I>2σ(I))][b]	0.1036	0.1266	0.1108
R_1 (all data)[a]	0.0638	0.0668	0.0742
wR_2 (all data)[b]	0.1149	0.1374	0.1205
Largest diff. Peak, hole / (e Å^{-3})	0.35 and -0.602	1.054 and -1.478	0.531 and -0.890

[a]$R_1 = \Sigma$ ||Fo| – |Fc||/Σ|Fo|; [b]$wR_2 = [\Sigma w(Fo^2 - Fc^2)^2/\Sigma w(Fo^2)^2]^{½}$.

Table S2: ICP analyses for metals in the samples used for SQUID measurements.

	% Co found	% Co calcd	Yield %	SQUID Mass mg		% Co Experiment	% Co Theory	Yield %	SQUID Mass mg
$Co_{0.67}Zn_{0.33}$	65.67(11)	66.67	27.21	22.37	**$Co_{0.14}Zn_{0.86}$**	13.88(13)	14.29	29.11	26.02
$Co_{0.50}Zn_{0.50}$	51.13(9)	50	29.32	15.06	**$Co_{0.125}Zn_{0.875}$**	11.89(10)	12.5	28.87	44.77
$Co_{0.33}Zn_{0.67}$	31.28(13)	33.33	28.45	28.59	**$Co_{0.11}Zn_{0.89}$**	11.45(14)	11.11	27.98	22.53
$Co_{0.25}Zn_{0.75}$	22.82(19)	25	28.66	22.37	**$Co_{0.10}Zn_{0.90}$**	8.96(8)	10	28.56	23.35
$Co_{0.20}Zn_{0.80}$	21.33(15)	20	28.37	36.67	**$Co_{0.05}Zn_{0.95}$**	4.32(11)	5	28.77	74.50
$Co_{0.17}Zn_{0.83}$	15.25(11)	16.66	29.54	33.17					

Table S3 Bond lengths [A] and angles [deg] for **Co_1**.

Co(1)-O(1)	2.001(2)	C(1)-C(2)	1.431(4)
Co(1)-O(1)A	2.001(2)	C(6)-C(7)	1.412(5)
Co(1)-O(2)	2.110(3)	C(6)-C(5)	1.408(5)
Co(1)-N(1)A	2.120(3)	C(2)-C(3)	1.381(4)
Co(1)-N(1)	2.120(3)	C(9)-C(8)	1.420(5)
O(1)-C(2)	1.333(4)	C(9)-C(10)	1.487(5)
N(1)-C(1)	1.362(4)	C(3)-C(4)	1.408(4)
N(1)-C(9)	1.326(4)	C(7)-C(8)	1.356(5)
O(1)-Co(1)-O(1)A	124.37(13)	N(1)-C(1)-C(6)	122.8(3)
O(1)-Co(1)-O(2)	117.82(7)	N(1)-C(1)-C(2)	116.3(3)
O(1)A-Co(1)-O(2)	117.82(7)	C(6)-C(1)-C(2)	120.8(3)
O(1)A-Co(1)-N(1)A	80.59(9)	C(7)-C(6)-C(1)	116.0(3)
O(1)A-Co(1)-N(1)	99.40(9)	C(5)-C(6)-C(1)	119.3(3)
O(1)-Co(1)-N(1)	80.59(9)	C(5)-C(6)-C(7)	124.7(3)
O(1)-Co(1)-N(1)A	99.40(9)	O(1)-C(2)-C(1)	118.3(3)
O(2)-Co(1)-N(1)	90.00(7)	O(1)-C(2)-C(3)	124.4(3)
O(2)-Co(1)-N(1)A	90.00(7)	C(3)-C(2)-C(1)	117.3(3)
N(1)A-Co(1)-N(1)	179.99(14)	N(1)-C(9)-C(8)	120.1(3)
C(2)-O(1)-Co(1)	114.25(19)	N(1)-C(9)-C(10)	118.4(3)
Co(1)-O(2)-H(2)	109.5	C(8)-C(9)-C(10)	121.4(3)
C(1)-N(1)-Co(1)	110.2(2)	C(8)-C(7)-C(6)	120.2(3)
C(9)-N(1)-Co(1)	129.7(2)	C(7)-C(8)-C(9)	120.9(3)
C(9)-N(1)-C(1)	120.0(3)	C(5)-C(4)-C(3)	122.0(3)

Symmetry codes: A) $-x + 1, y, -z + 3/2$

Table S4 Bond lengths [A] and angles [deg] for **$Co_{0.1}Zn_{0.9}$**.

Zn(1)-O(1)A	2.020(2)	C(2)-C(1)	1.433(5)
Zn(1)-O(1)	2.020(2)	C(9)-C(8)	1.420(5)
Zn(1)-O(2)	2.098(3)	C(9)-C(10)	1.490(5)
Zn(1)-N(1)A	2.117(3)	C(1)-C(6)	1.420(5)
Zn(1)-N(1)	2.117(3)	C(3)-C(4)	1.413(5)
O(1)-C(2)	1.328(4)	C(8)-C(7)	1.358(5)
N(1)-C(9)	1.330(5)	C(6)-C(7)	1.412(5)
N(1)-C(1)	1.364(4)	C(6)-C(5)	1.419(5)
C(2)-C(3)	1.385(5)	C(4)-C(5)	1.365(5)
O(1)A-Zn(1)-O(1)	124.63(14)	O(1)-C(2)-C(1)	118.4(3)
O(1)A-Zn(1)-O(2)	117.69(7)	C(3)-C(2)-C(1)	117.8(3)
O(1)-Zn(1)-O(2)	117.69(7)	N(1)-C(9)-C(8)	120.6(3)
O(1)A-Zn(1)-N(1)A	80.49(10)	N(1)-C(9)-C(10)	117.7(3)
O(1)-Zn(1)-N(1)A	99.41(10)	C(8)-C(9)-C(10)	121.6(3)
O(2)-Zn(1)-N(1)A	90.11(8)	N(1)-C(1)-C(6)	122.4(3)

O(1)A-Zn(1)-N(1)	99.41(10)	N(1)-C(1)-C(2)	116.5(3)
O(1)-Zn(1)-N(1)	80.49(10)	C(6)-C(1)-C(2)	121.1(3)
O(2)-Zn(1)-N(1)	90.11(8)	C(2)-C(3)-C(4)	120.7(3)
N(1)A-Zn(1)-N(1)	179.78(16)	C(7)-C(8)-C(9)	120.3(3)
C(2)-O(1)-Zn(1)	113.9(2)	C(7)-C(6)-C(5)	124.5(3)
C(9)-N(1)-C(1)	119.8(3)	C(7)-C(6)-C(1)	116.5(3)
C(9)-N(1)-Zn(1)	129.6(2)	C(5)-C(6)-C(1)	119.0(3)
C(1)-N(1)-Zn(1)	110.3(2)	C(8)-C(7)-C(6)	120.3(3)
O(1)-C(2)-C(3)	123.9(3)	C(5)-C(4)-C(3)	122.2(3)

Symmetry codes: A) $-x+1, y, -z+1/2$

Table S5. Deviation parameters calculated by SHAPE from each ideal polyhedron for complex **Co_1**. The best matches are displayed in red.

Geometry	Symmetry	Co1
PP-5	D_{5h}	28.530
vOC-5	C4v	21.425
TBPY-5	**D_{3h}**	**14.803**
SPY-5	C_{4v}	17.727
JTBPY-5	D_{3h}	25.417

PP-5=Pentagon, vOC-5=Vacant octahedron, TBPY-5=Trigonal bipyramid, SPY-5=Spherical square pyramid, JTBPY-5=Johnson trigonal bipyramid.

Table S6: The fit parameters obtained from analyses of the ac susceptibilities of **$Co_{0.20}Zn_{0.80}$** under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	7.07E+00	2.86E+00	-6.08965	2.03E-01	1.29E-01
2	6.73E+00	2.69E+00	-6.22201	2.01E-01	2.00E-02
2.2	6.37E+00	2.50E+00	-6.30377	1.75E-01	1.96E-02
2.4	5.94E+00	2.30E+00	-6.41539	1.56E-01	1.13E-02
2.6	5.60E+00	2.11E+00	-6.50353	1.52E-01	8.76E-03
2.8	5.23E+00	1.96E+00	-6.62876	1.28E-01	3.26E-03
3	4.95E+00	1.81E+00	-6.69474	1.23E-01	4.20E-03
3.3	4.57E+00	1.68E+00	-6.81289	1.08E-01	4.48E-03
3.6	4.25E+00	1.54E+00	-6.96711	1.15E-01	5.55E-03
3.9	3.97E+00	1.45E+00	-7.09549	1.16E-01	5.43E-03
4.2	3.73E+00	1.35E+00	-7.22729	9.40E-02	2.63E-03
4.5	3.51E+00	1.27E+00	-7.41289	8.95E-02	3.15E-03
4.8	3.31E+00	1.20E+00	-7.6156	7.62E-02	1.11E-03
5.1	3.14E+00	1.11E+00	-7.84851	7.39E-02	2.67E-10
5.4	2.99E+00	1.02E+00	-8.15696	7.32E-02	7.71E-04
5.7	2.84E+00	1.05E+00	-8.36324	3.70E-02	6.95E-04

6	2.71E+00	1.07E+00	-8.67661	8.10E-03	5.40E-04
6.3	2.59E+00	8.85E-01	-9.08153	3.66E-02	7.66E-04
6.6	2.49E+00	5.79E-01	-9.54259	9.08E-02	5.26E-04

Table S7 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.17}Zn_{0.83}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	6.06E-01	2.00E-01	-5.41057	1.28E-01	8.52E-05
2	5.71E-01	1.79E-01	-5.54196	1.53E-01	1.16E-04
2.2	5.32E-01	1.63E-01	-5.63263	1.62E-01	2.00E-04
2.4	4.90E-01	1.49E-01	-5.7508	1.65E-01	9.88E-05
2.6	4.57E-01	1.35E-01	-5.86622	1.76E-01	1.73E-04
2.8	4.29E-01	1.28E-01	-5.96926	1.64E-01	5.91E-05
3	4.03E-01	1.21E-01	-6.08197	1.55E-01	7.85E-05
3.2	3.82E-01	1.14E-01	-6.15603	1.50E-01	2.80E-05
3.4	3.60E-01	1.08E-01	-6.26069	1.36E-01	7.56E-05
3.6	3.37E-01	1.03E-01	-6.34395	1.20E-01	9.06E-05
3.8	3.31E-01	9.94E-02	-6.4394	1.29E-01	1.55E-04
4	3.09E-01	9.65E-02	-6.55999	9.94E-02	4.62E-05
4.3	2.89E-01	9.26E-02	-6.72094	8.79E-02	5.87E-05
4.6	2.72E-01	8.83E-02	-6.94244	7.35E-02	6.25E-05
4.9	2.59E-01	8.48E-02	-7.17298	6.58E-02	6.13E-05
5.2	2.44E-01	8.17E-02	-7.44604	4.12E-02	6.23E-05
5.5	2.29E-01	7.89E-02	-7.78079	1.83E-02	2.46E-05
5.8	2.19E-01	7.46E-02	-8.15442	2.44E-02	2.86E-05
6.1	2.13E-01	8.82E-02	-8.51581	2.67E-02	7.68E-05
6.4	2.01E-01	5.43E-02	-8.91247	5.79E-02	1.47E-05
6.7	1.89E-01	6.99E-02	-9.30913	2.46E-03	5.78E-06
7	1.86E-01	8.45E-02	-9.75995	2.53E-03	4.71E-06

Table S8 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.14}Zn_{0.86}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	6.01E-01	1.15E-01	-5.40252	1.38E-01	2.86E-03
1.9	5.72E-01	1.10E-01	-5.4757	1.28E-01	8.39E-04
2	5.46E-01	1.09E-01	-5.52986	1.11E-01	6.24E-04
2.2	5.17E-01	1.02E-01	-5.64696	1.10E-01	8.53E-04
2.4	4.90E-01	9.44E-02	-5.73527	1.02E-01	1.37E-03
2.6	4.57E-01	9.60E-02	-5.85137	6.27E-02	1.37E-03
2.8	4.24E-01	8.48E-02	-5.9573	7.61E-02	8.80E-04
3	3.97E-01	8.14E-02	-6.04982	7.36E-02	6.76E-04
3.2	3.76E-01	7.71E-02	-6.12809	7.36E-02	6.48E-04
3.4	3.55E-01	7.20E-02	-6.22612	7.05E-02	3.49E-04
3.6	3.38E-01	7.34E-02	-6.31586	4.48E-02	3.01E-04

3.8	3.23E-01	6.92E-02	-6.39418	5.89E-02	2.52E-04
4	3.02E-01	6.68E-02	-6.53182	3.85E-02	2.05E-04
4.3	2.81E-01	6.36E-02	-6.72893	2.90E-02	8.87E-05
4.6	2.62E-01	5.85E-02	-6.93714	3.48E-02	4.40E-05
4.9	2.52E-01	5.84E-02	-7.13869	3.71E-02	5.07E-05
5.2	2.38E-01	5.51E-02	-7.41719	1.76E-02	4.61E-05
5.5	2.26E-01	5.60E-02	-7.70902	1.34E-02	2.30E-05
5.8	2.16E-01	5.29E-02	-8.04696	1.90E-02	1.33E-05
6.1	2.03E-01	4.95E-02	-8.48405	2.73E-03	1.41E-05
6.4	1.96E-01	5.39E-02	-8.95828	3.41E-03	2.59E-05
6.7	1.88E-01	5.41E-02	-9.46019	6.69E-04	1.15E-05

Table S9 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.125}Zn_{0.875}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	6.00E-01	1.72E-01	-5.15369	1.99E-01	7.15E-04
2	5.70E-01	1.61E-01	-5.30665	1.78E-01	9.72E-04
2.2	5.36E-01	1.44E-01	-5.45567	1.85E-01	1.28E-03
2.4	4.99E-01	1.34E-01	-5.58841	1.66E-01	7.98E-04
2.6	4.68E-01	1.26E-01	-5.68501	1.54E-01	7.04E-04
2.8	4.35E-01	1.16E-01	-5.8106	1.55E-01	3.91E-04
3	4.09E-01	1.08E-01	-5.91042	1.52E-01	5.30E-04
3.2	3.81E-01	1.02E-01	-6.02796	1.44E-01	2.83E-04
3.4	3.63E-01	9.76E-02	-6.12295	1.33E-01	1.78E-04
3.6	3.47E-01	9.35E-02	-6.22761	1.24E-01	2.31E-04
3.8	3.28E-01	8.73E-02	-6.31777	1.29E-01	1.07E-04
4	3.08E-01	8.50E-02	-6.40471	1.05E-01	7.08E-05
4.3	2.91E-01	8.06E-02	-6.5625	1.01E-01	4.88E-05
4.6	2.77E-01	7.81E-02	-6.76376	7.70E-02	9.19E-05
4.9	2.60E-01	7.45E-02	-7.04391	7.10E-02	4.00E-05
5.2	2.46E-01	6.93E-02	-7.357	6.44E-02	3.08E-05
5.5	2.34E-01	7.03E-02	-7.68794	4.63E-02	3.04E-05
5.8	2.19E-01	6.84E-02	-8.13393	2.72E-02	6.72E-06
6.1	2.12E-01	8.16E-02	-8.64666	1.48E-17	2.56E-05
6.4	1.99E-01	6.95E-02	-9.19204	2.16E-17	1.93E-05

Table S10 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.11}Zn_{0.99}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	8.69E-01	2.28E-01	-5.34107	1.39E-01	1.61E-03
2	8.13E-01	2.12E-01	-5.46077	1.13E-01	2.51E-03
2.2	7.57E-01	2.02E-01	-5.59151	9.62E-02	2.80E-03
2.4	7.07E-01	1.82E-01	-5.65198	1.06E-01	2.80E-03
2.6	6.62E-01	1.72E-01	-5.72016	1.15E-01	1.77E-03

2.8	6.15E-01	1.59E-01	-5.82603	1.19E-01	1.17E-03
3	5.68E-01	1.48E-01	-5.94978	1.11E-01	4.95E-04
3.2	5.34E-01	1.40E-01	-6.05998	1.00E-01	2.05E-04
3.4	5.07E-01	1.34E-01	-6.15718	9.11E-02	1.36E-04
3.6	4.79E-01	1.31E-01	-6.28747	6.30E-02	1.93E-04
3.8	4.57E-01	1.30E-01	-6.35988	4.52E-02	2.40E-04
4	4.45E-01	1.29E-01	-6.44669	3.85E-02	5.09E-04
4.2	4.26E-01	1.27E-01	-6.5845	3.00E-02	2.80E-04
4.4	4.06E-01	1.15E-01	-6.6963	3.43E-02	2.95E-04
4.6	3.87E-01	1.13E-01	-6.86051	2.31E-02	1.80E-04
4.8	3.77E-01	1.11E-01	-7.01142	2.68E-02	1.41E-04
5	3.54E-01	1.02E-01	-7.24224	2.74E-02	7.16E-05
5.2	3.45E-01	9.83E-02	-7.42513	3.30E-02	8.21E-05
5.4	3.28E-01	1.01E-01	-7.63423	1.10E-02	7.61E-05
5.6	3.19E-01	9.65E-02	-7.84954	1.11E-02	5.50E-05
5.8	3.09E-01	9.71E-02	-8.07648	7.29E-03	1.23E-05
6	2.97E-01	9.72E-02	-8.31914	4.29E-16	6.84E-05
6.3	2.87E-01	8.97E-02	-8.75181	7.13E-16	5.57E-05
6.6	2.76E-01	1.02E-01	-9.23361	9.97E-16	3.61E-05
6.9	2.58E-01	2.00E-04	-9.75351	1.12E-15	2.74E-10

Table S11 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.1}Zn_{0.90}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	9.66E-02	4.16E-02	-5.71306	8.24E-02	2.17E-06
2	8.74E-02	3.73E-02	-5.85759	8.82E-02	2.31E-06
2.2	8.08E-02	3.42E-02	-5.97172	6.68E-02	1.88E-06
2.4	7.77E-02	3.06E-02	-6.07128	1.47E-01	9.57E-07
2.6	6.95E-02	2.80E-02	-6.18784	1.06E-01	7.16E-06
2.8	6.71E-02	2.64E-02	-6.25161	1.17E-01	1.46E-06
3	6.22E-02	2.55E-02	-6.35353	8.06E-02	2.26E-06
3.2	5.87E-02	2.33E-02	-6.46147	1.09E-01	8.89E-07
3.4	5.55E-02	2.22E-02	-6.55885	9.66E-02	5.30E-07
3.6	5.31E-02	2.14E-02	-6.62868	9.54E-02	8.12E-07
3.8	4.99E-02	2.07E-02	-6.74936	7.32E-02	8.29E-07
4	4.80E-02	1.88E-02	-6.88446	9.80E-02	3.57E-07
4.3	4.44E-02	1.83E-02	-7.06182	6.15E-02	6.16E-07
4.6	4.15E-02	1.78E-02	-7.25291	3.65E-02	5.77E-07
4.9	3.98E-02	1.55E-02	-7.47936	7.71E-02	3.01E-07
5.2	3.76E-02	1.64E-02	-7.69869	3.50E-02	6.35E-07
5.5	3.56E-02	1.57E-02	-7.98215	2.67E-02	4.98E-07
5.8	3.40E-02	1.36E-02	-8.42135	5.42E-02	4.97E-07
6.1	3.26E-02	1.70E-02	-8.87059	3.74E-02	3.32E-07
6.4	3.10E-02	2.13E-02	-9.51173	1.12E-02	3.23E-07

6.7	2.93E-02	6.99E-16	-10.32382	3.73E-02	8.93E-07
7	2.85E-02	4.79E-16	-10.94978	7.86E-02	3.74E-07

Table S12 The fit parameters obtained from analyses of the ac susceptibilities of $\mathbf{Co_{0.05}Zn_{0.95}}$ under 1.5 kOe bias dc field.

T / K	χ_T/cm^3 mol^{-1}	χ_S /cm^3 mol^{-1}	ln(τ / s)	a	R^2
1.8	8.13E-01	1.50E-01	-4.23068	1.41E-01	4.29E-04
2	7.38E-01	1.36E-01	-4.33914	1.37E-01	2.28E-04
2.2	6.81E-01	1.23E-01	-4.42667	1.37E-01	2.42E-04
2.4	6.27E-01	1.15E-01	-4.52939	1.24E-01	4.85E-04
2.6	5.79E-01	1.05E-01	-4.64557	1.27E-01	3.51E-04
2.8	5.41E-01	9.79E-02	-4.76065	1.25E-01	3.18E-04
3	5.03E-01	9.13E-02	-4.9009	1.21E-01	2.48E-04
3.2	4.74E-01	8.73E-02	-5.02335	1.08E-01	3.12E-04
3.4	4.45E-01	8.31E-02	-5.16331	9.80E-02	2.88E-04
3.6	4.23E-01	7.90E-02	-5.29757	9.64E-02	2.60E-04
3.8	4.07E-01	7.44E-02	-5.42889	9.90E-02	2.14E-04
4	3.86E-01	7.23E-02	-5.61321	8.28E-02	4.26E-04
4.3	3.64E-01	6.79E-02	-5.90362	7.35E-02	4.74E-04
4.6	3.39E-01	6.36E-02	-6.24059	6.19E-02	1.14E-04
4.9	3.16E-01	6.15E-02	-6.61942	3.79E-02	4.90E-05
5.2	2.99E-01	6.15E-02	-6.99063	1.97E-02	1.64E-04
5.5	2.84E-01	5.80E-02	-7.40018	2.25E-02	4.50E-05
5.8	2.68E-01	5.64E-02	-7.81517	1.42E-02	9.92E-06
6.1	2.56E-01	5.02E-02	-8.26565	1.94E-02	1.16E-05
6.4	2.46E-01	4.96E-02	-8.72165	3.75E-02	1.43E-05
6.7	2.35E-01	1.85E-02	-9.39054	5.67E-02	2.18E-05

Table S13 The fit parameters to the frequency dependence ac-susceptibilities of $\mathbf{Co_xZn_{1-x}}$ using equation 1.

Sample	U / K	τ*10^{-8} s	R
$\mathbf{Co_{0.20}Zn_{0.80}}$	57(4)	18.2(6)	0.99789
$\mathbf{Co_{0.17}Zn_{0.83}}$	63(4)	18.8(6)	0.998
$\mathbf{Co_{0.14}Zn_{0.86}}$	67(6)	19.4(4)	0.9999
$\mathbf{Co_{0.125}Zn_{0.875}}$	67(2)	19.6(4)	0.9994
$\mathbf{Co_{0.11}Zn_{0.89}}$	72(3)	20.2(5)	0.9988
$\mathbf{Co_{0.10}Zn_{0.90}}$	108(5)	25.3(7)	0.9988
$\mathbf{Co_{0.05}Zn_{0.95}}$	118(11)	26.3(8)	0.9992

Table S14 The fit parameters to the frequency dependence ac-susceptibilities of $\mathbf{Co_xZn_{1-x}}$ using equation 2.

Sample	A	n	U_{eff}	τ_0	τ_{QTM}	R
	K^{-1} s^{-1}		**K**	**10^{-8} s**	**s**	
$\mathbf{Co_{0.20}Zn_{0.80}}$	194.58	1.32	58 (3)	1.577	0	0.9977

$Co_{0.17}Zn_{0.83}$	90.32	1.45	61(2)	1.105	0	0.9991
$Co_{0.14}Zn_{0.86}$	90.88	1.44	65(3)	0.5867	0	0.9976
$Co_{0.125}Zn_{0.875}$	66.11	1.59	72(2)	0.1579	0	0.9992
$Co_{0.11}Zn_{0.89}$	73.24	1.58	70(3)	0.269	0	0.9967
$Co_{0.10}Zn_{0.90}$	102.09	1.65	100(6)	0.00139	0	0.9962
$Co_{0.05}Zn_{0.95}$	12.01	2.36	100(7)	0.00248	0	0.982

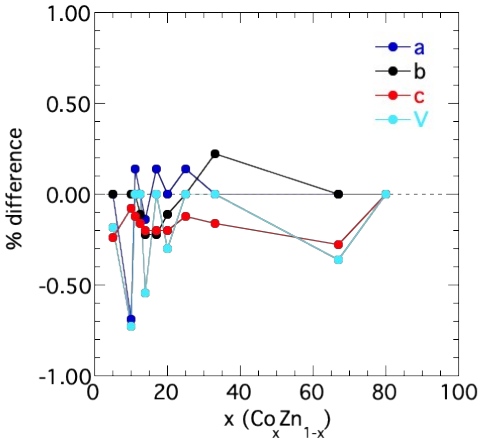


Fig. S1 Relative percentage change of lattice parameters obtained from single crystal diffraction for all the solid solution samples.

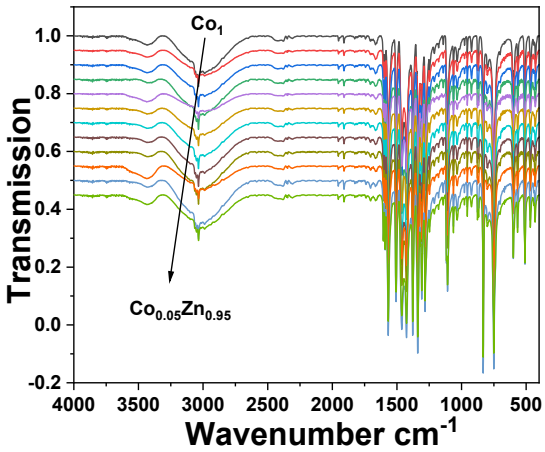


Fig. S2 IR spectra of Co_1 and Co_xZn_{1-x} at variable temperature.

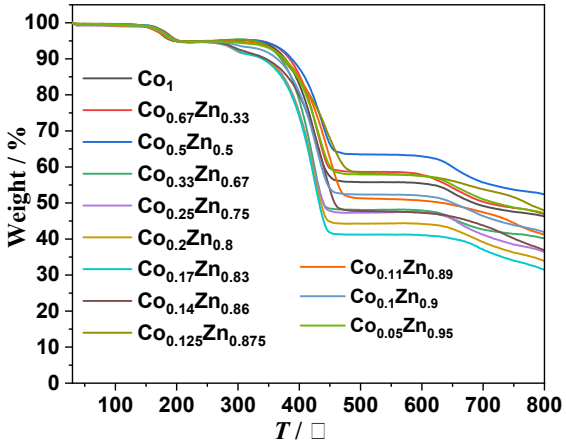


Fig. S3 TG curves for $\mathbf{Co_1}$ and $\mathbf{Co_xZn_{1-x}}$.

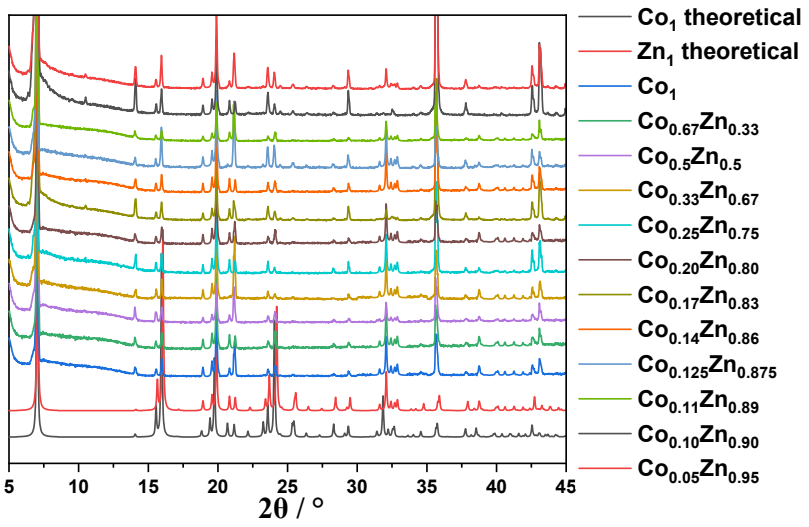


Fig. S4 PXRD patterns of $\mathbf{Co_1}$ and $\mathbf{Co_xZn_{1-x}}$.

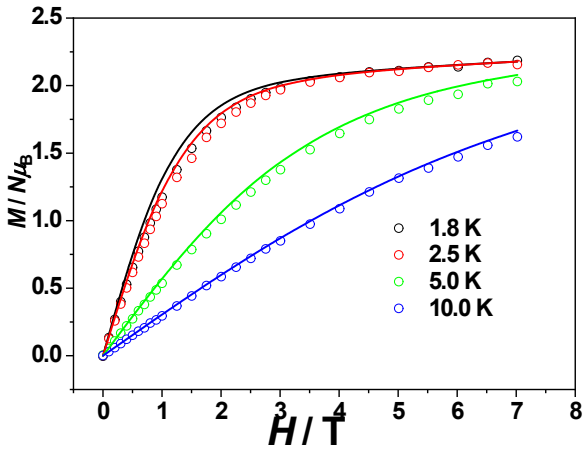


Fig. S5 Isothermal magnetization at different temperatures for $\mathbf{Co_{0.1}Zn_{0.9}}$.

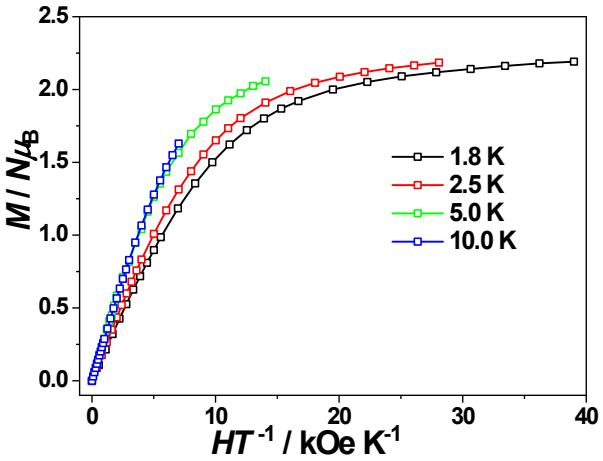


Fig. S6 Experimental M vs H / T plots at different temperatures for complex $\mathbf{Co_1}$.

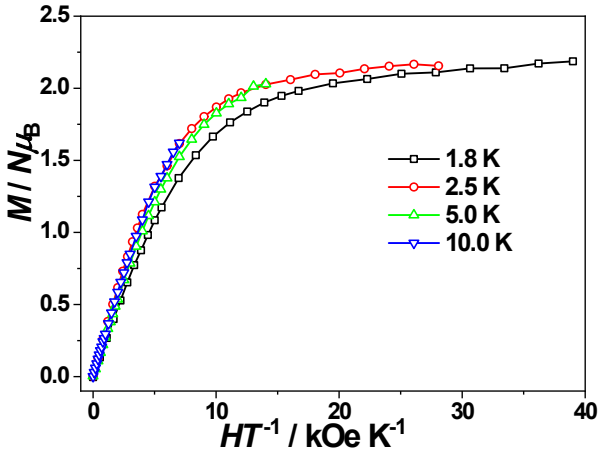
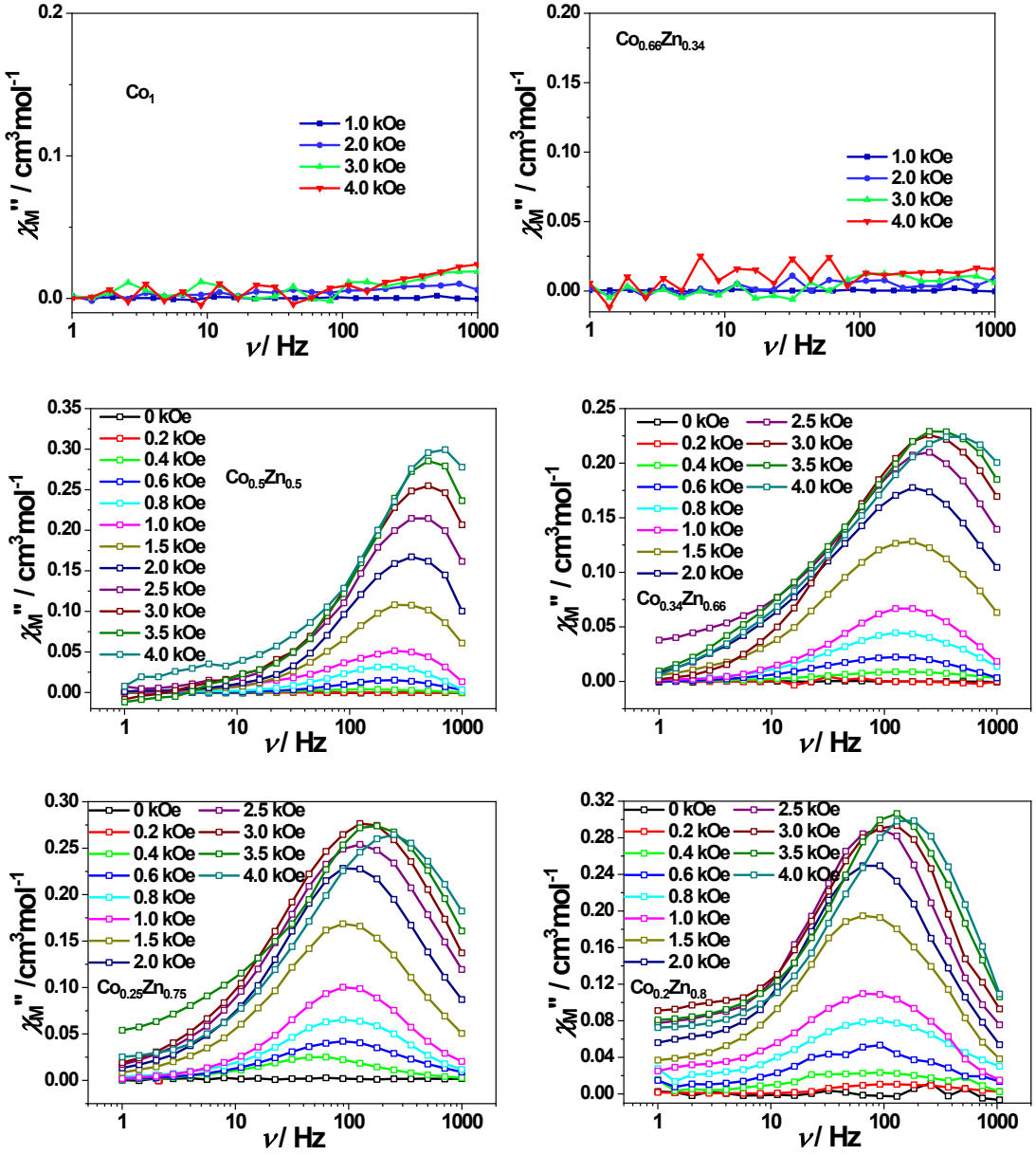


Fig. S7 Experimental M vs H / T plots at different temperatures for complex $\mathbf{Co_{0.1}Zn_{0.9}}$.



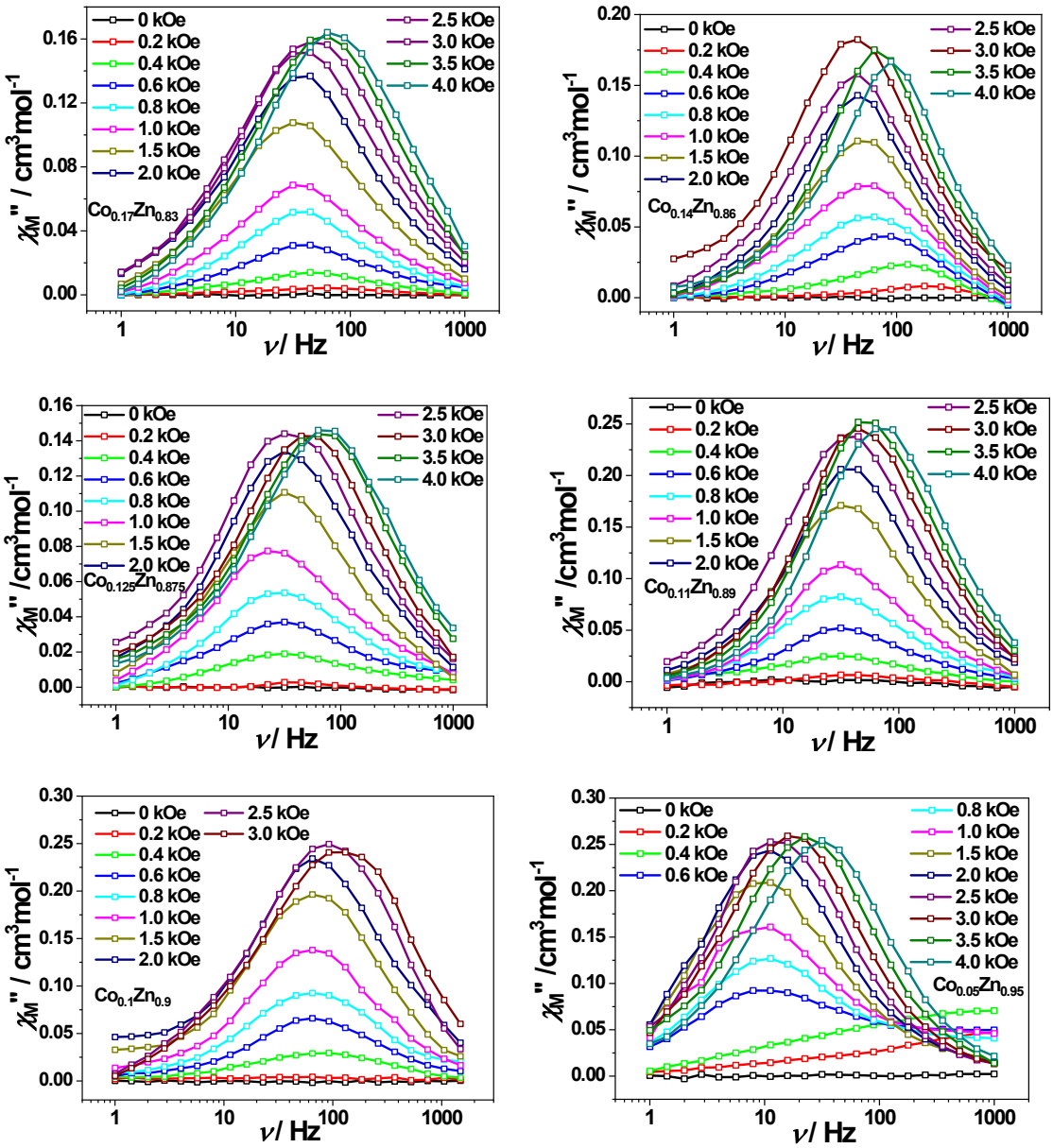
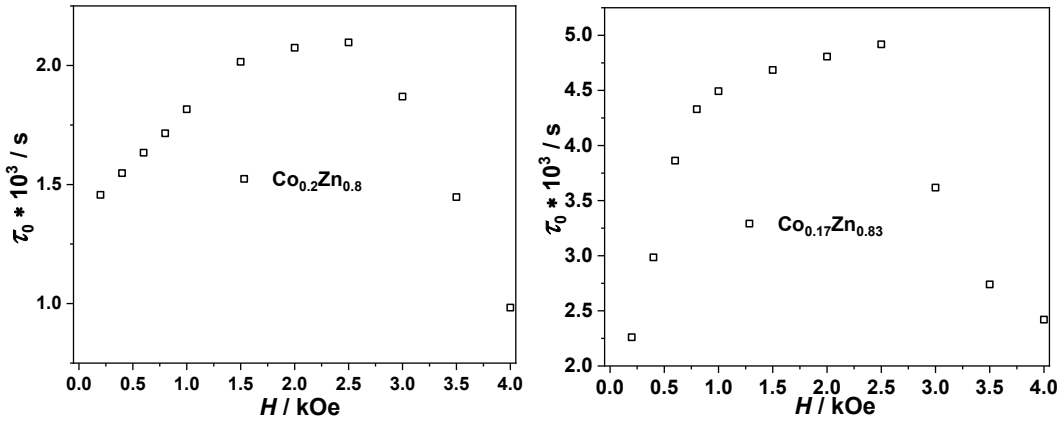


Fig. S8 Isothermal field sweep measurement performed on polycrystalline sample of complex $\mathbf{Co_1}$ and complexes $\mathbf{Co_xZn_{1-x}}$ at 2.0 K.



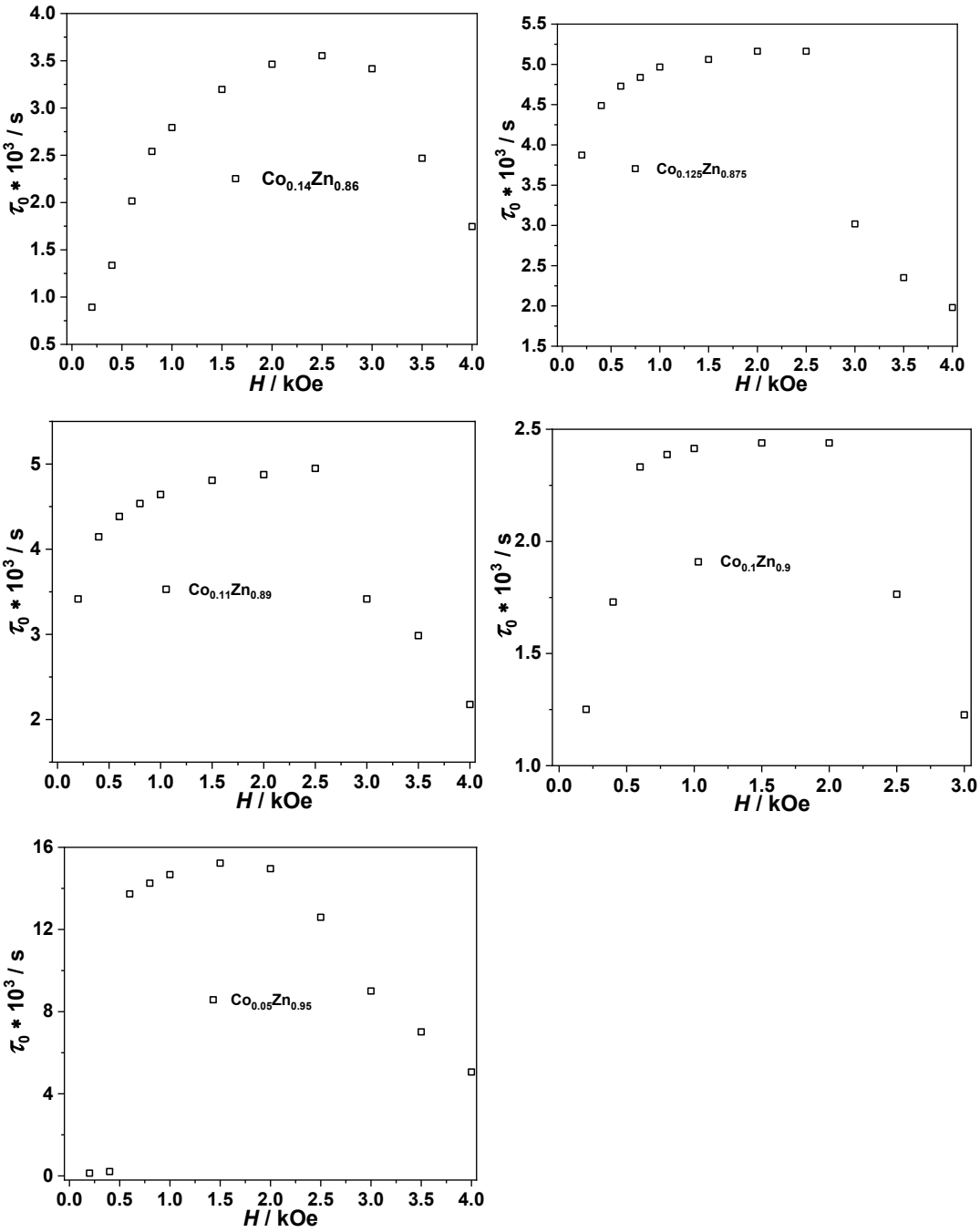
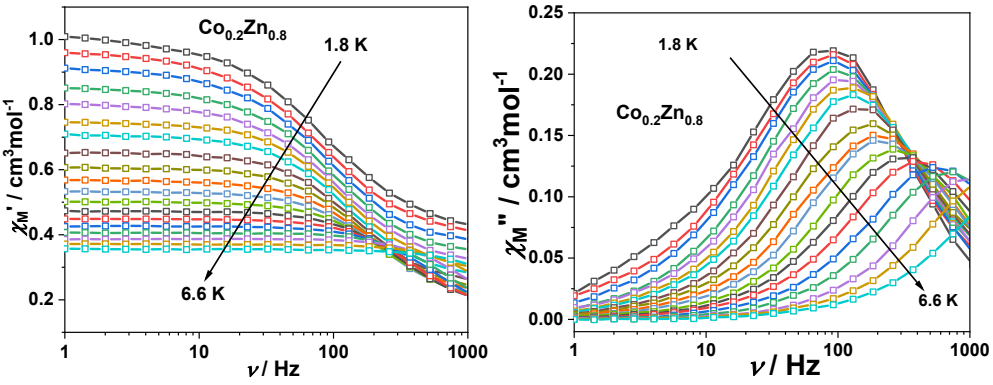
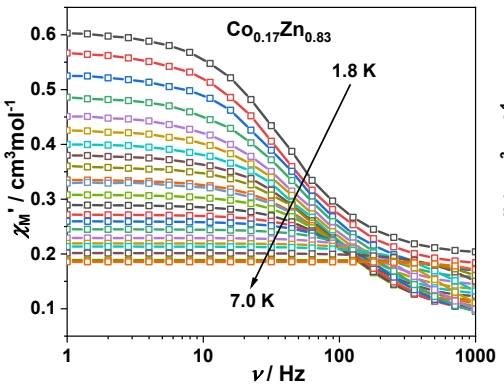
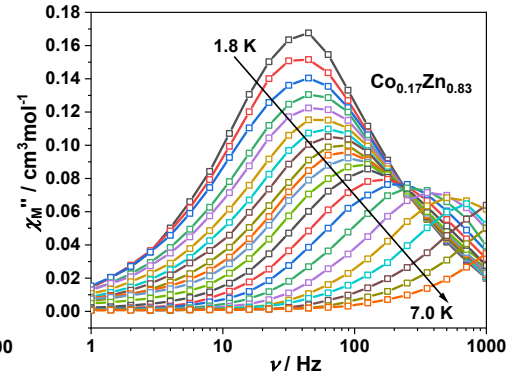
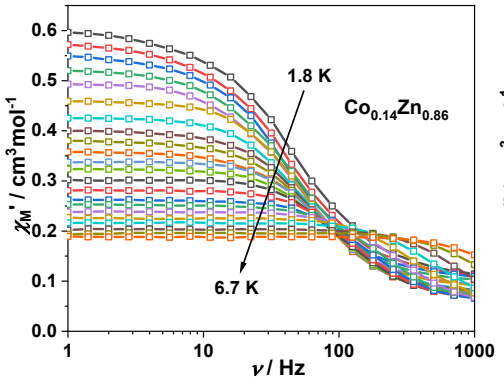


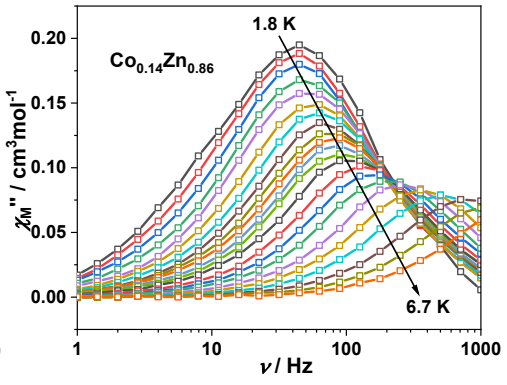
Fig. S9 The plots *τ vs. H* for complexes $\mathbf{Co_xZn_{1-x}}$ at 2.0 K

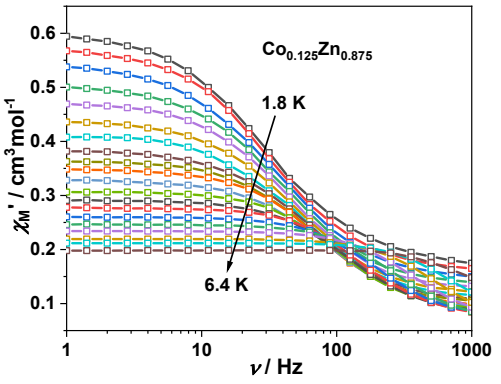


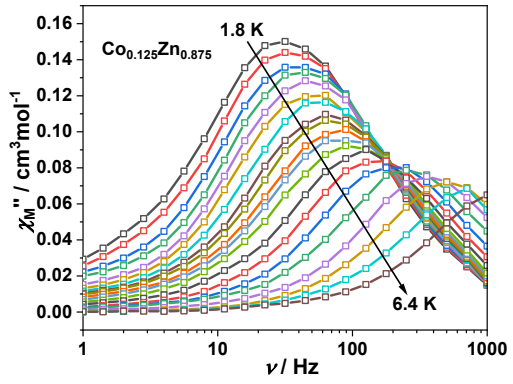

Co0.17Zn0.83
1.8 K
7.0 K
χM' / cm3mol-1
ν / Hz

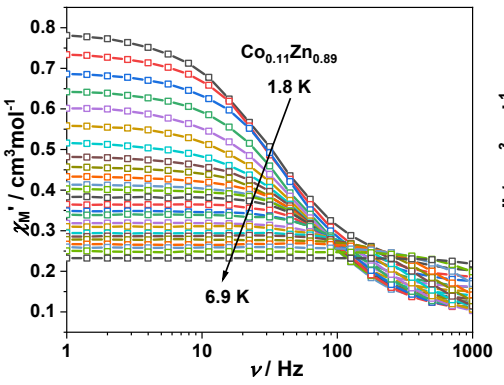

1.8 K
Co0.17Zn0.83
7.0 K
χM'' / cm3mol-1
ν / Hz

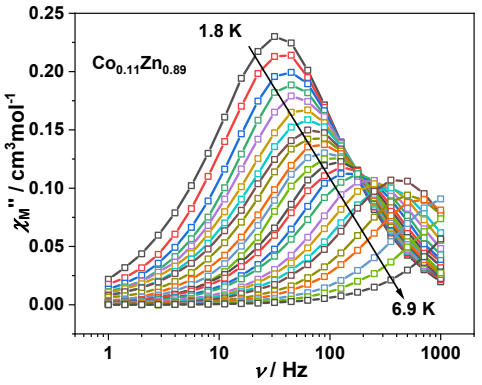

1.8 K
Co0.14Zn0.86
6.7 K
χM' / cm3mol-1
ν / Hz


1.8 K
Co0.14Zn0.86
6.7 K
χM'' / cm3mol-1
ν / Hz


Co0.125Zn0.875
1.8 K
6.4 K
χM' / cm3mol-1
ν / Hz


1.8 K
Co0.125Zn0.875
6.4 K
χM'' / cm3mol-1
ν / Hz


Co0.11Zn0.89
1.8 K
6.9 K
χM' / cm3mol-1
ν / Hz


1.8 K
Co0.11Zn0.89
6.9 K
χM'' / cm3mol-1
ν / Hz

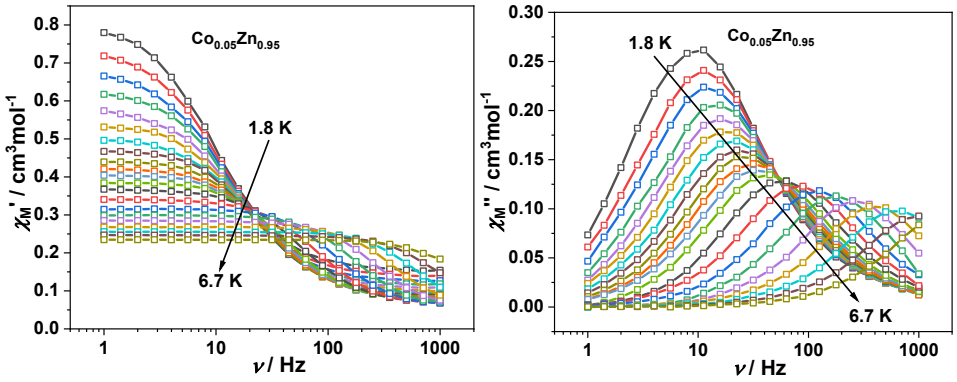


Fig. S10 Frequency-dependent out-of-phase (χ_M'') and in-phase (χ_M') AC susceptibilities for $\mathbf{Co_1Zn_{1-x}}$.

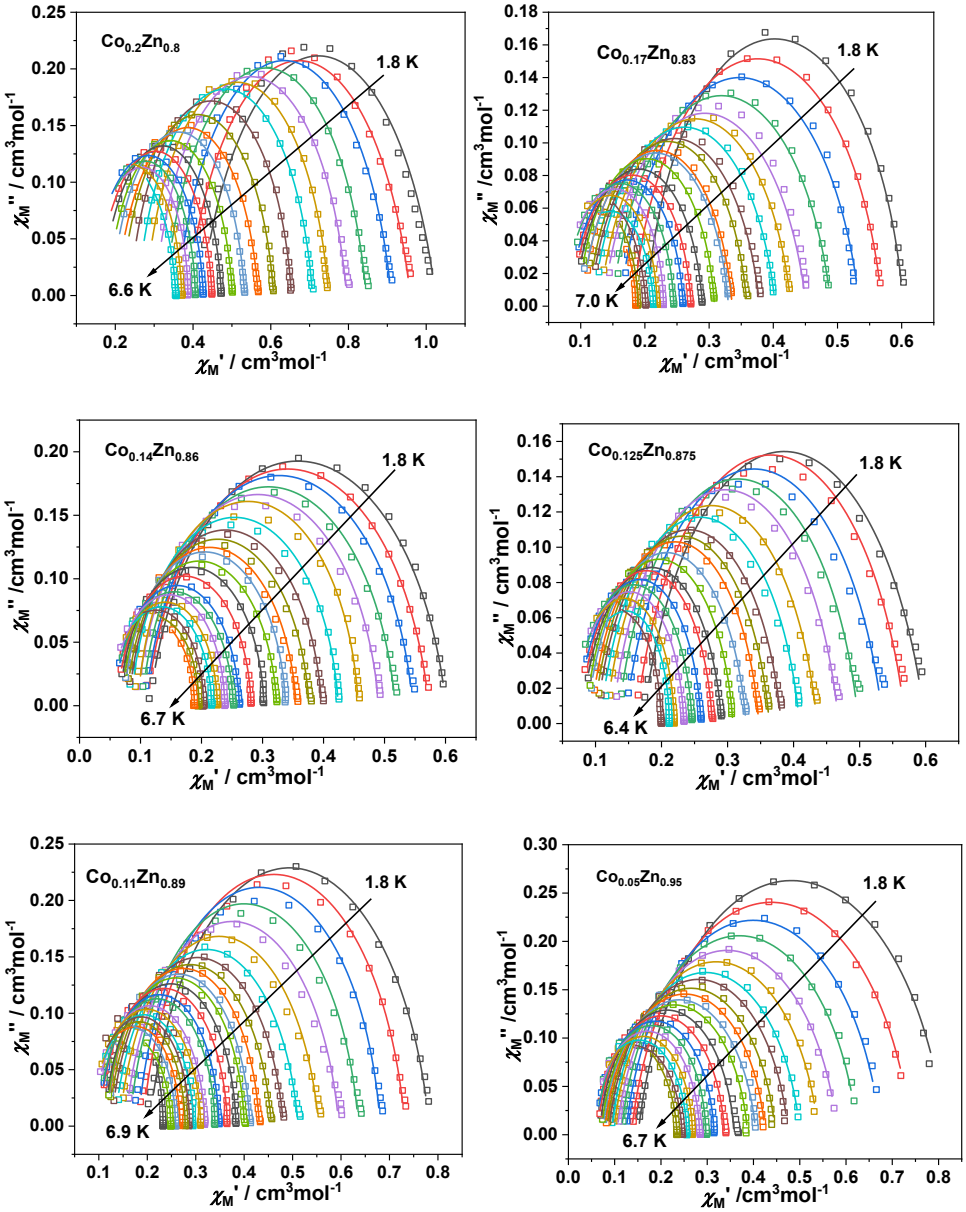


Fig. S11 Cole–Cole plots of $\mathbf{Co_xZn_{1-x}}$ with solid lines denoting the best fits to Debye model.

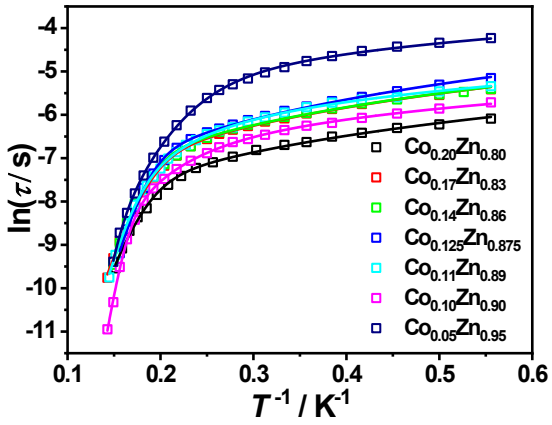
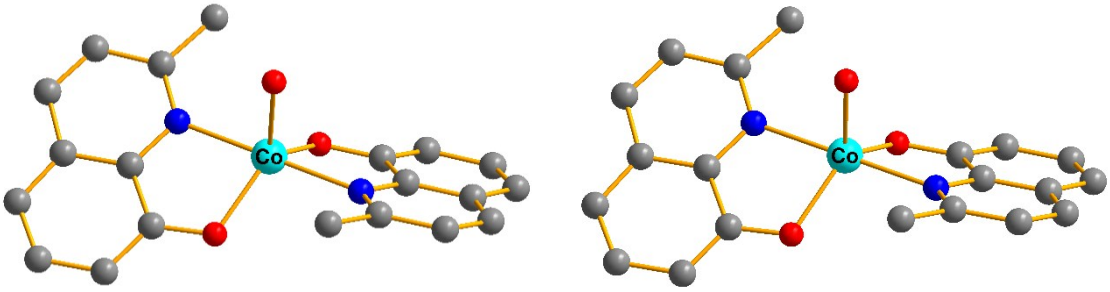
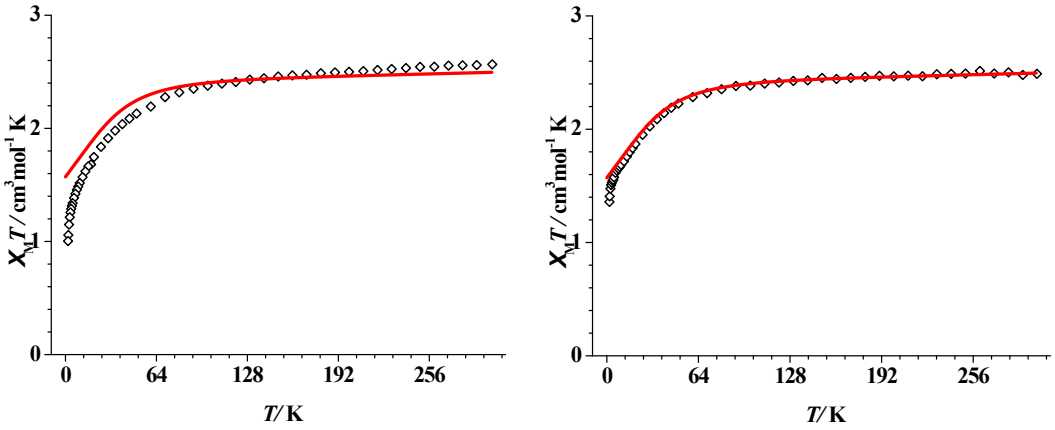


Fig. S12 A plot of ln(τ/s) versus T^{-1} with the solid line representing linear fit to the Arrhenius equation for $\mathbf{Co_xZn_{1-x}}$.



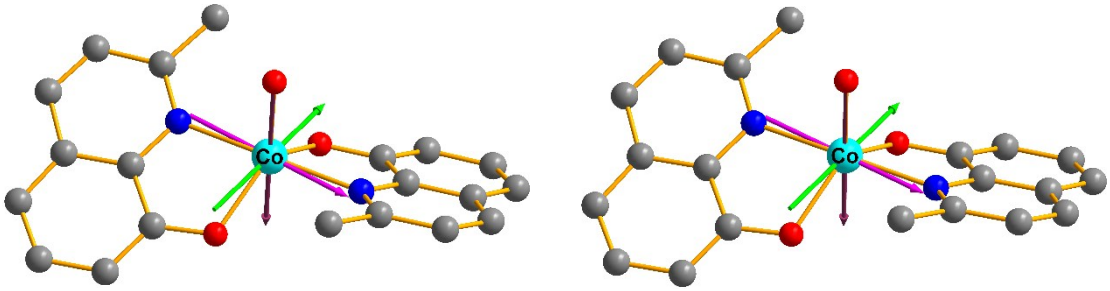
Co_1 **$Co_{0.1}Zn_{0.9}$**

Fig. S13. Calculated molecular structures of complexes **Co_1** and **$Co_{0.1}Zn_{0.9}$**. H atoms are omitted for clarity.



Co_1 **$Co_{0.1}Zn_{0.9}$**

Fig. S14. Calculated (solid line) and experimental (circle dot) data magnetic susceptibilities of complexes **Co_1** and **$Co_{0.1}Zn_{0.9}$**.



1 **$Co_{0.1}Zn_{0.9}$**

Fig. S15. Calculated orientations of the local magnetic axes (g_x: pink; g_y: plum; g_z: green) of the ground doublet on Co^{II} ion of complexes **Co_1** and **$Co_{0.1}Zn_{0.9}$**.

Table S15. Calculated weights of the five most important spin-orbit-free states for the lowest two spin-orbit states of complexes **Co_1** and **$Co_{0.1}Zn_{0.9}$** using CASPT2 with MOLCAS 8.2.

	Spin-orbit states	Energy (cm^{-1})	Spin-free states, Spin, Weights				
Co_1	1	0.0	1,1.5,0.9733	4,1.5,0.0170	5,1.5,0.0045	2,1.5, 0.0016	3,1.5, 0.0015
	2	83.2	1,1.5,0.9875	4,1.5,0.0051	5,1.5,0.0040	2,1.5, 0.0013	3,1.5, 0.0012
$Co_{0.1}$ $Zn_{0.9}$	1	0.0	1,1.5,0.9708	4,1.5,0.0168	5,1.5,0.0048	3,1.5, 0.0034	2,1.5, 0.0023
	2	86.3	1,1.5,0.9866	4,1.5,0.0052	5,1.5,0.0043	2,1.5, 0.0016	3,1.5, 0.0014