Electronic Supporting Information (**ESI**)

Chiral Magnetic Metal-Organic Frameworks of Dimetal Sub-Units: Magnetism Tuning by Mixed-Metal Compositions of the Solid Solutions

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Experimental Section

Synthesis of 1,4-di (1-imidazolylmethyl) benzene.

Although the synthesis of 1,4-dimb was previously reported, we prepared this compound in a different way. 1,4-Dichloromethylbenzene (10.20 g, 0.06 mol) was dissolved in DMSO (20.0 mL), followed by addition of KOH (10.11 g, 0.18 mol). After the mixture is stirred at 50~60^{o}C for 2 hours, a DMSO solution (50.0 mL) of imidazole (10.50 g, 0.15 mol) was added dropwise, which was continued for ca. 1 hour. The mixture was then allowed to stir at 40~50^{o}C until the reactant 1,4-dichloro-methylbezene was no longer detected by TLC. Upon removal of the solvent under reduced pressure, the crude product was obtained. After recrystallization from ethyl acetate-petroleum, pale-yellow crystals (8.29 g) of 1,4-dimb were obtained. The yield was 62.5%, mp. 86-87 ^{o}C. ^{1}H NMR ($CDCl_3$, δ, ppm): 5.11 (s, 4H, $C\underline{H}_2PhC\underline{H}_2$), 6.88 (s, 2H, imidazole-H), 7.09 (s, 2H, imidazole-H), 7.14 (s, 4H, Ph-H), 7.54 (s, 2H, 3-imidazole-H).

Table S1. Elemental analysis (%) For **1~6**:

	Calculated			Found		
	C%	H%	N%	C%	H%	N%
1	54.30	5.63	7.44	54.28	5.66	7.42
2	54.26	5.63	7.44	54.23	5.65	7.41
3	54.28	5.63	7.45	54.26	5.66	7.42
4	54.29	5.63	7.45	54.27	5.65	7.43
5	54.27	5.63	7.45	54.28	5.64	7.43
6	54.28	5.63	7.45	54.25	5.64	7.42

Table S2. Main IR bands (cm^{-1})

1	**2**	**3**	**4**	**5**	**6**
2969.0*s*	2967.9*s*	2967.9*s*	2969.3*s*	2968.3*s*	2970*s*
1598.3*vs*	1597.6*vs*	1597.6*vs*	1597.7*vs*	1597.7*vs*	1598*vs*
1522.8*s*	1520.7*s*	1520.7*s*	1521.7*s*	1521.3*s*	1441*s*
1403.7*vs*	1408.3*vs*	1408.3*vs*	1404.3*vs*	1407.1*vs*	1408*vs*
1289.0*m*	1288.1*m*	1288.1*m*	1288.8*m*	1288.5*m*	1292.9*m*
1112.4*m*	1111.8*m*	1111.8*m*	1112.0*m*	1112.3*m*	1114*m*
804.4*m*	804.2*m*	804.2*m*	804.2*m*	804.1*m*	802*m*
756.8*m*	756.9*m*	756.9*m*	757.6*m*	757.4*m*	759*m*
656.1*m*	655.6*m*	655.6*m*	655.8*m*	656.0*m*	658*m*
517.0*w*	512.7*w*	512.7*w*	515.3*w*	513.4*w*	516*w*
480.7*w*	481.0*w*	481.0*w*	480.5*w*	481.0*w*	482*w*

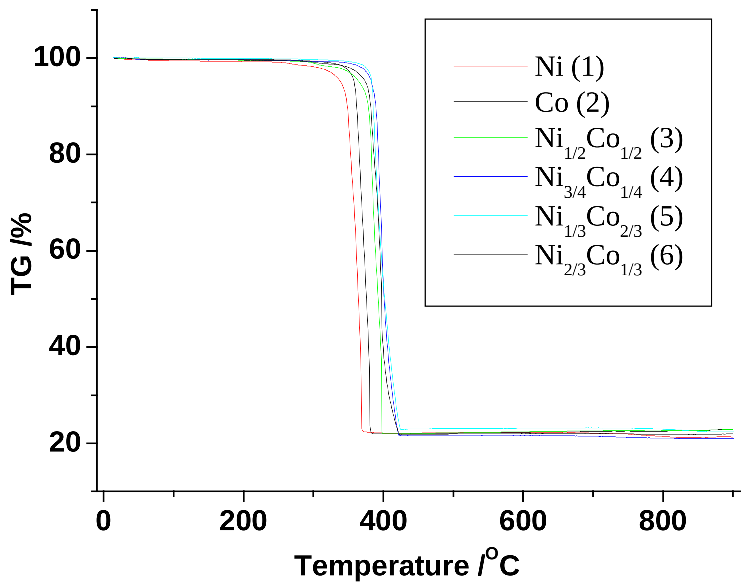


Figure S1. The thermal gravimetric carve of **1~6** in air.

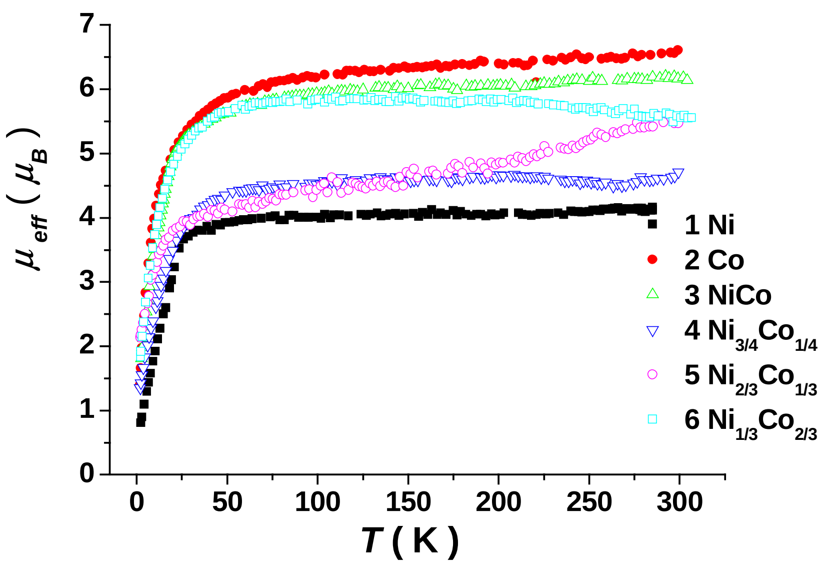


Figure S2. μ_{eff} $-T$ plots for **1** per Ni^{II}_2 at 30 kOe (black), **2** per Co^{II}_2 at 10 kOe (red), **3** per CoNi at 10 kOe (green), **4** per $Ni_{3/2}Co_{1/2}$ at 30 kOe (blue), **5** per $Ni_{4/3}Co_{2/3}$ at 20 kOe (magenta) and **6** per $Ni_{2/3}Co_{4/3}$ at 20 kOe (cyan).

Least-squares fitting of the magnetic data for 1 and 2:

1. Fittied by pure spin system (Ni^{2+}):

The cluster Hamiltonian is given in equation as follows

$$\hat{H}_{ex} = -J\hat{S}_1\hat{S}_2$$

The magnetic susceptibility data were quantitatively analyzed by treating the Ni(II) ions as an interacting dimer (Eq. 1 and 2), where $\chi = J/kT$, N, J is intra-dimer interaction and zJ' is inter-dimer interaction, β, g, k, and T have their usual meanings.

$$\chi_{\text{dimer}} = \frac{2N\beta^2 g^2}{kT}[\frac{5e^{3x} + e^x}{5e^{3x} + 3e^x + 1}] \qquad \text{(Eq.S1)}$$

$$\chi_{\text{dimer}} = \frac{\chi'_{\text{dimer}}}{1\text{-}2zJ'\chi'_{\text{dimer}}/N\beta^2 g^2} \qquad \text{(Eq.S2)}$$

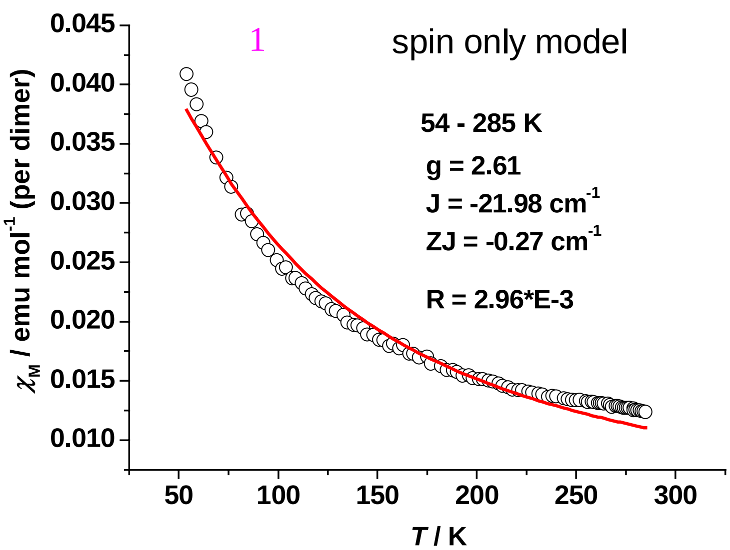


Figure S3. Least-squares fitting of experimental data using eq. S1 for **1**

B. Fittied by pure spin system (Co^{2+}):

The cluster Hamiltonian is given in equation as follows

$$\hat{H}_{ex} = -J\hat{S}_1\hat{S}_2$$

The susceptibility (χ) of **dimmer Co** is given as eq. 3 and 2.

$$\chi'_{\text{dimer}} = \frac{2N\beta^2 g^2}{kT}\left[\frac{14e^{6x} + 5e^{3x} + e^{x}}{7e^{6x} + 5e^{3x} + 3e^{x} + 1}\right] \qquad (3)$$

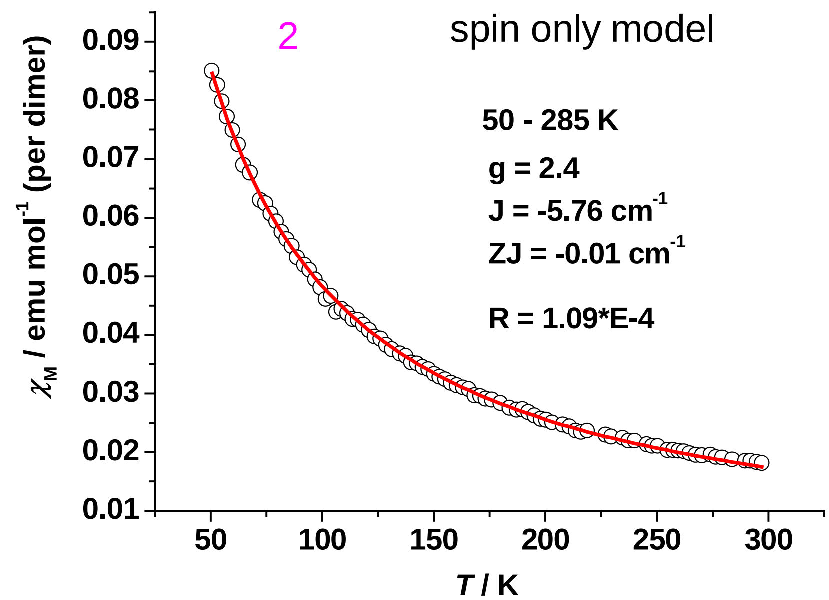
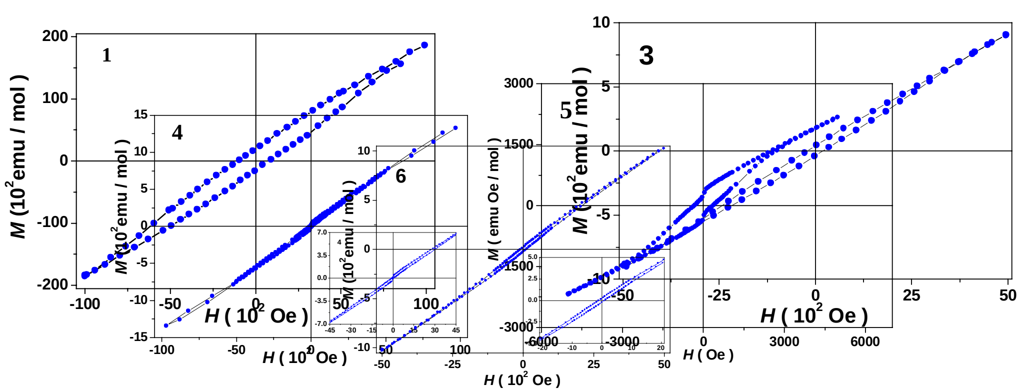


Figure S4. Least-squares fitting of experimental data using eq. S3 for **2**.



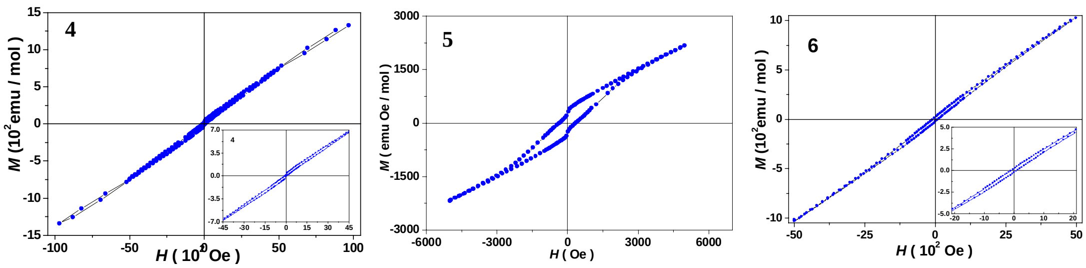


Figure S5. The hysteresis loops for **1**, **3** ~**6** at 1.8 K.

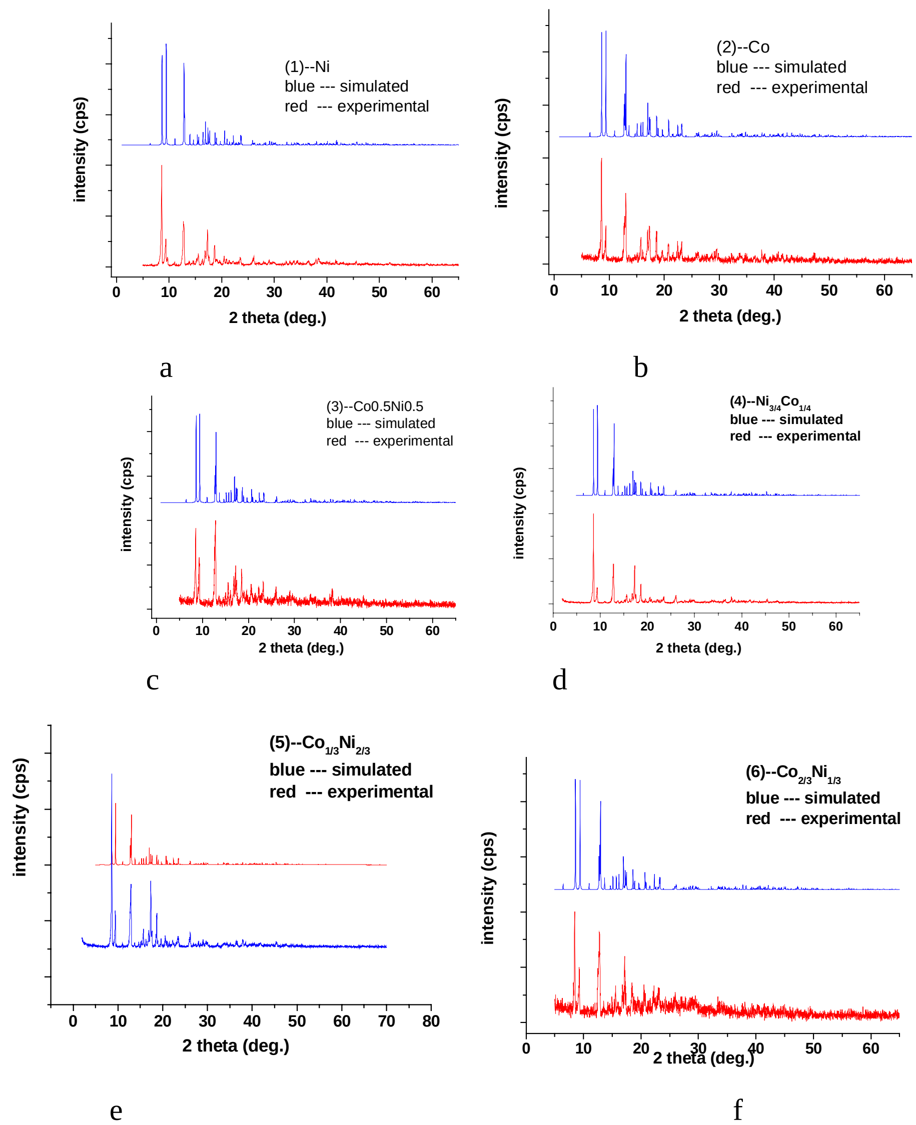


Figure S6. The simulated (upper) and experimental (lower) powder X-ray diffraction patterns of **1** (a), **2** (b), **3** (c), **4** (d), **5** (e) and **6** (f).