Supporting Information for

Two 2-D multifunctional cobalt(II) compounds: Field-induced single-ion magnetism and catalytic oxidation of benzylic C-H bonds

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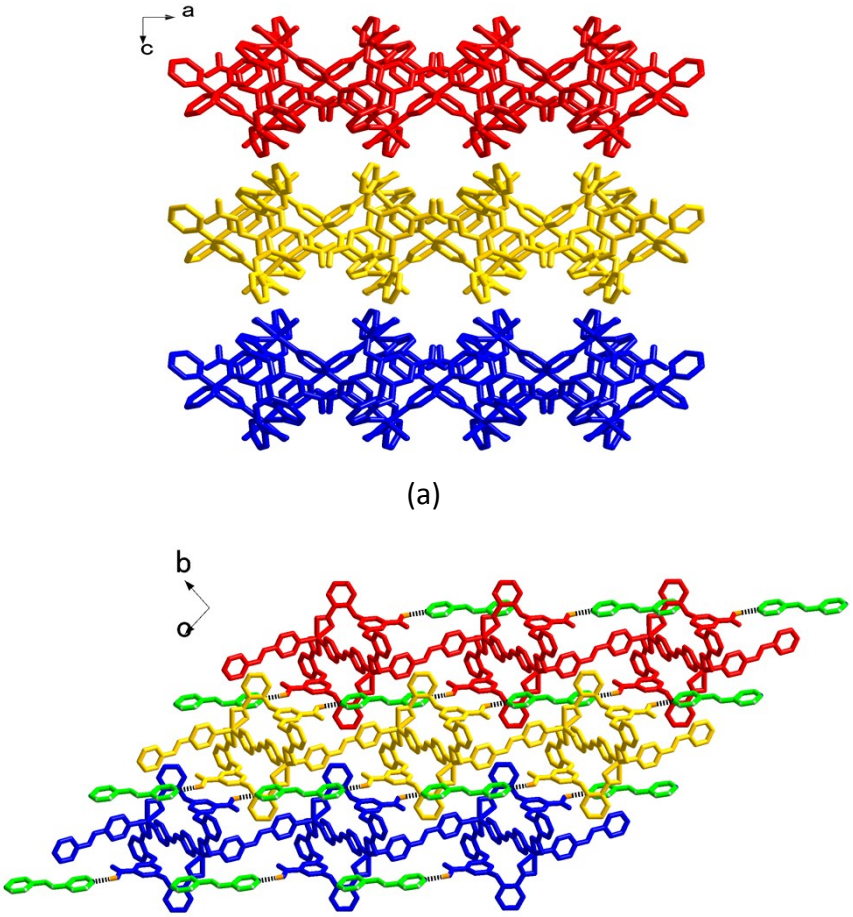
Table S1. Selected bond distances and angles of compounds **1** and **2**.

		1	
Co(1)-O(1)	2.135(4)	Co(1)-O(7)	2.081(3)
Co(1)-O(8)	2.134(4)	Co(1)-O(9)	2.099(3)
Co(1)-N(1)	2.170(4)	Co(1)-N(2)	2.121(4)
O(7)-Co(1)-O(9)	94.85(14)	O(7)-Co(1)-N(2)	86.75(14)
O(7)-Co(1)-O(8)	89.25(13)	O(9)-Co(1)-N(2)	88.43(15)
O(9)-Co(1)-O(8)	90.50(14)	O(7)-Co(1)-O(1)	89.22(14)
N(2)-Co(1)-O(8)	175.76(14)	O(9)-Co(1)-O(1)	174.86(14)
N(2)-Co(1)-O(1)	94.94(15)	O(9)-Co(1)-N(1)	89.98(14)
O(8)-Co(1)-O(1)	86.41(14)	N(2)-Co(1)-N(1)	89.71(15)
O(7)-Co(1)-N(1)	173.93(15)	O(8)-Co(1)-N(1)	94.39(15)
O(1)-Co(1)-N(1)	86.17(15)		
		2	
Co(1)-O(1)	2.068(3)	Co(1)-O(2)	2.257(3)
Co(1)-O(7A)	2.144(3)	Co(1)-O(8A)	2.163(3)
Co(1)-N(1)	2.093(3)	Co(1)-N(2)	2.089(3)
O(1)-Co(1)-N(2)	95.88(13)	O(1)-Co(1)-O(7A)	106.52(12)
O(1)-Co(1)-N(1)	96.94(13)	O(1)-Co(1)-O(8A)	165.12(12)
O(7A)-Co(1)-O(2)	92.93(12)	O(8A)-Co(1)-O(2)	110.56(13)
N(1)-Co(1)-O(2)	91.46(13)	N(1)-Co(1)-O(8A)	95.04(13)
N(1)-Co(1)-O(7A)	155.11(14)	N(2)-Co(1)-N(1)	91.92(14)
N(2)-Co(1)-O(7A)	93.81(13)	N(2)-Co(1)-O(2)	156.30(13)
N(2)-Co(1)-O(8A)	92.49(13)		

Symmetry code for compound **2**, A: -x+2, -y+1, -z+2.

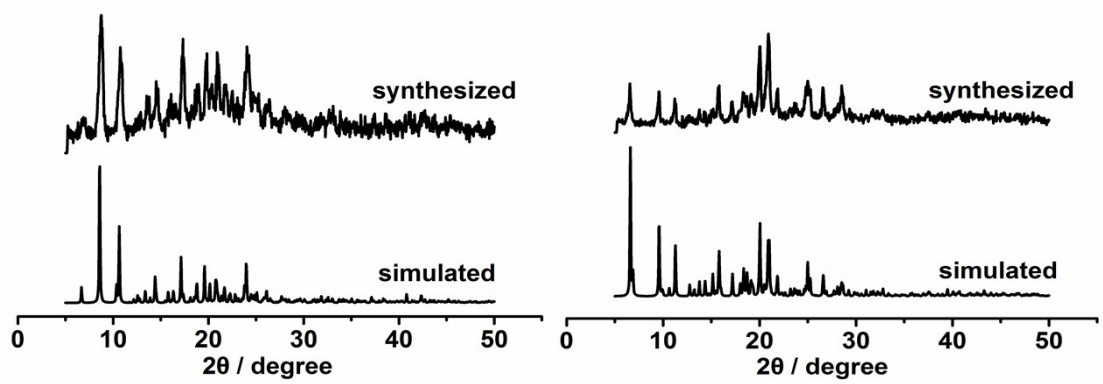
Fig. S1. 3D stacking interactions of compounds **1** (a) and **2** (b).



(a)

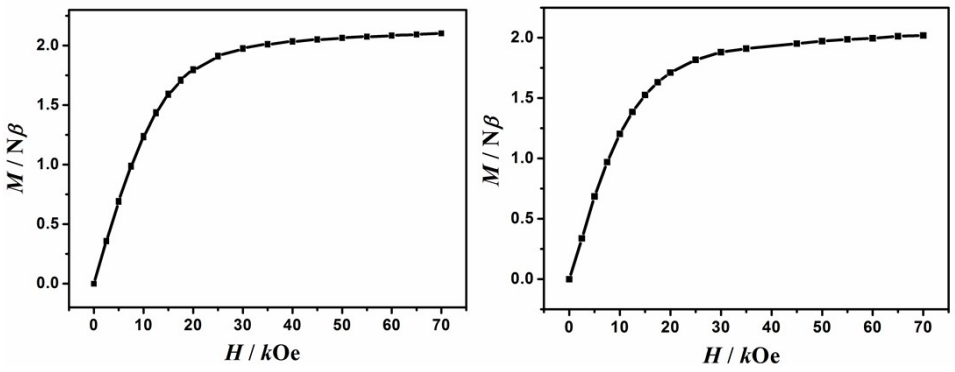
(b)

Fig. S2. Powder XRD of compounds **1** (a) and **2** (b).



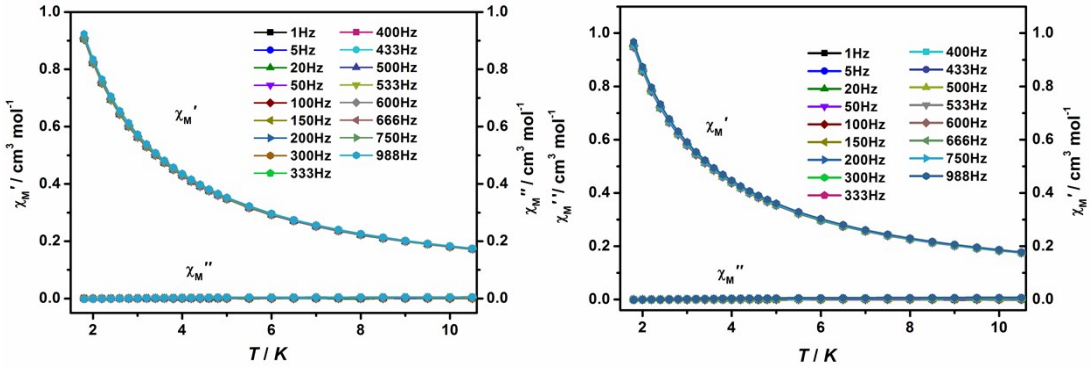
(a) (b)

Fig. S3. M-H data of compounds **1** (a) and **2** (b) at 2.0K.



(a) (b)

Fig. S4. The temperature dependence of the in-phase (χ') and out-of-phase (χ'') ac susceptibilities at different frequencies with an oscillation of zero Oe dc field for complex **1** ((a)) and **2** (b)), respectively.



(a) (b)

Table S2. The relaxation fitting parameters from Least-Squares Fitting of $\chi(\omega)$ data for compound **1** from 1.8 to 6.0 K under 2 kOe dc field.

T	$\chi_{s,tot}$	$\Delta\chi_1$	τ_1	α_1	$\Delta\chi_2$	τ_2	α_2
1.8	0.114	0.339	0.221	0.439	0.552	0.00225	0.0896
2.2	0.078	0.311	0.070	0.612	0.450	0.00175	0.0619
2.6	0.053	0.227	0.081	0.728	0.442	0.00144	0.0756
3.0	0.79×10^{-7}	0.268	0.120	0.865	0.410	0.00114	0.0677
3.5	0.108×10^{-6}	0.184	0.167	0.917	0.387	0.000827	0.0704
4.0	0.231×10^{-6}	0.138	0.202	0.953	0.356	0.000574	0.0616
4.5	0.137×10^{-6}	0.112	0.427	0.977	0.325	0.000375	0.0470
5.0	0.307×10^{-6}	0.0869	0.654	0.992	0.301	0.000232	0.0359
5.5	0.848×10^{-6}	0.0604	0.864	0.988	0.286	0.000137	0.0296
6.0	0.197×10^{-5}	0.0143	0.634	0.988	0.284	0.000079	0.0265

Table S3. The relaxation fitting parameters from Least-Squares Fitting of $\chi(\omega)$ data for compound **2** from 1.8 to 6.0 K under 2 kOe dc field.

T	$\chi_{s,tot}$	$\Delta\chi_1$	τ_1	α_1	$\Delta\chi_2$	τ_2	α_2
1.8	0.0282	0.182	0.051	0.0668	0.650	0.00096	0.134
2.2	0.212×10^{-7}	0.135	0.075	0.156	0.632	0.00062	0.176
2.6	0.43×10^{-7}	0.0750	0.070	0.142	0.582	0.00044	0.166
3.0	0.138×10^{-6}	0.0293	0.047	0.659×10^{-7}	0.536	0.00030	0.153
3.5	0.150×10^{-6}	0.0205	0.077	0.866×10^{-7}	0.480	0.00018	0.121
4.0	0.120×10^{-6}	0.0125	0.149	0.192×10^{-6}	0.430	0.00010	0.0789
4.5	0.175×10^{-6}	0.0095	0.221	0.483×10^{-6}	0.388	0.000061	0.0331
5.0	0.235×10^{-6}	0.0063	0.238	0.945×10^{-13}	0.353	0.000036	0.0131
5.5	0.447×10^{-6}	0.0091	0.264	0.849×10^{-14}	0.324	0.000022	0.361×10^{-12}
6.0	0.245×10^{-6}	0.0015	0.166	0.235×10^{-13}	0.299	0.000015	0.319×10^{-12}

Table S4. Selective oxidation of diphenylmethane by catalyst using TBHP oxidant[a]

Entry	Solvent	T(°)	t(h)	TBHP/substrate	Conv.(%)	Sele.(%)[b]
1	C_6H_5Cl	80	24	3	90.3	98.1
2	CH_3CN	80	24	3	76.5	94.2
3	C_6H_5CN	80	24	3	85.7	96.6
4	C_6H_5Cl	50	24	3	52.4	96.1
5	C_6H_5Cl	60	24	3	67.8	97.4
6	C_6H_5Cl	70	24	3	78.8	95.7
7	C_6H_5Cl	80	12	3	54.6	98.5
8	C_6H_5Cl	80	16	3	70.4	98.2
9	C_6H_5Cl	80	20	3	85.3	98.7
10	C_6H_5Cl	80	24	0	trace	trace
11	C_6H_5Cl	80	24	2.5	84.9	96.8
12	C_6H_5Cl	80	24	3.75	94.4	90.3
13[c]	C_6H_5Cl	80	24	3	75.5	97.5
14[d]	C_6H_5Cl	80	24	3	90.7	97.6
15[e]	C_6H_5Cl	80	24	-	-	-
16[f]	C_6H_5Cl	80	24	3	34.2	73.1
17[g]	C_6H_5Cl	80	24	3	56.7	89.3

[a] Reaction conditions: diphenylmethane (0.125 mmol), compound **1** (10 μmol), chlorobenzene (0.5 mL), *t*-BuOOH (0.375 mmol), 80 ºC,24 h; [b] Selectivity to ketones. The by-product was benzhydrol; [c] Reaction conditions: diphenylmethane (0.125 mmol), compound **1** (5 μmol), chlorobenzene (0.5 mL), *t*-BuOOH (0.375mmol), 80 ºC,24 h; [d] diphenylmethane (0.125 mmol), compound **1** (15 μmol), chlorobenzene (0.5 mL), *t*-BuOOH (0.375 mmol), 80 ºC,24 h; [e] diphenylmethane (0.125 mmol), compound **1** (10 μmol), chlorobenzene (0.5 mL), 80 ºC,24 h; [f] diphenylmethane (0.125 mmol), chlorobenzene (0.5 mL), *t*-BuOOH (0.375 mmol), 80 ºC,24 h; [g] diphenylmethane (0.125 mmol), $CoCl_2 \cdot 6H_2O$ (10 μmol), chlorobenzene (0.5 mL), *t*-BuOOH (0.375

mmol), 80 ºC,24 h.

Fig. S5. The FT-IR spectra of compound **1** after catalytic cycles.