

## Supporting Information

### Magnetic behaviours dominated by the interplay of magnetic anisotropy and exchange coupling in the local Co<sup>II</sup><sub>7</sub> discs

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**Table S1.** Selected bond distances (Å) and angles (°) for **1**<sup>a</sup>

Co(1)–O(9)	1.998(3)	Co(1)–O(13)	2.101(3)
Co(1)–O(22)	2.004(3)	Co(1)–O(6)	2.176(3)
Co(1)–N(6)	2.042(3)	Co(1)–O(26)	2.285(4)
Co(2)–O(3)	2.006(3)	Co(2)–O(17)	2.094(3)
Co(2)–O(21)	2.038(3)	Co(2)–O(25)	2.171(3)
Co(2)–N(7)	2.077(3)	Co(2)–O(6)	2.208(3)
Co(3)–O(14)	2.073(3)	Co(3)–O(22)	2.105(3)
Co(3)–O(24)	2.099(3)	Co(3)–O(30) <sup>#1</sup>	2.122(3)
Co(3)–O(15) <sup>#2</sup>	2.101(2)	Co(3)–O(10)	2.170(3)
Co(4)–O(23)	1.961(3)	Co(4)–O(20) <sup>#3</sup>	2.075(3)
Co(4)–O(11) <sup>#1</sup>	1.980(3)	Co(4)–O(7) <sup>#1</sup>	2.183(3)
Co(4)–N(1)	2.016(3)	Co(5)–O(5)	2.086(3)
Co(5)–O(22)	2.061(3)	Co(5)–O(24)	2.105(3)
Co(5)–O(23)	2.070(3)	Co(5)–O(8) <sup>#1</sup>	2.114(3)
Co(5)–O(21)	2.081(3)	Co(6)–O(16) <sup>#2</sup>	2.070(3)
Co(6)–O(24)	1.968(3)	Co(6)–O(7) <sup>#1</sup>	2.165(3)
Co(6)–O(31) <sup>#1</sup>	1.997(3)	Co(6)–N(2)	2.028(3)
Co(7)–O(18)	2.072(2)	Co(7)–O(4)	2.117(3)
Co(7)–O(21)	2.082(3)	Co(7)–O(23)	2.129(3)
Co(7)–O(19) <sup>#3</sup>	2.104(3)	Co(7)–O(12) <sup>#1</sup>	2.181(3)
O(9)–Co(1)–O(22)	99.00(11)	O(9)–Co(1)–O(13)	90.49(11)
O(9)–Co(1)–O(6)	83.65(11)	O(22)–Co(1)–O(13)	90.01(11)
O(9)–Co(1)–O(2)	81.10(14)	O(22)–Co(1)–N(6)	101.01(12)
N(6)–Co(1)–O(6)	89.40(11)	O(22)–Co(1)–O(6)	96.17(10)
N(6)–Co(1)–O(13)	94.31(11)	O(13)–Co(1)–O(2)	83.19(14)
N(6)–Co(1)–O(2)	79.60(15)	O(6)–Co(1)–O(2)	90.60(14)
O(3)–Co(2)–O(21)	96.84(11)	O(21)–Co(2)–N(7)	96.59(11)
O(3)–Co(2)–O(17)	90.09(11)	O(21)–Co(2)–O(17)	90.68(10)
N(7)–Co(2)–O(17)	93.14(11)	N(7)–Co(2)–O(25)	84.13(12)
O(3)–Co(2)–O(25)	82.60(12)	O(17)–Co(2)–O(25)	86.88(12)
O(21)–Co(2)–O(6)	91.28(10)	O(3)–Co(2)–O(6)	87.15(11)
O(25)–Co(2)–O(6)	91.14(11)	N(7)–Co(2)–O(6)	89.16(11)

O(24)–Co(3)–O(22)	79.15(10)	O(24)–Co(3)–O(15) <sup>#2</sup>	94.35(10)
O(14)–Co(3)–O(15) <sup>#2</sup>	92.62(10)	O(14)–Co(3)–O(22)	94.19(10)
O(14)–Co(3)–O(30) <sup>#1</sup>	85.79(11)	O(24)–Co(3)–O(30) <sup>#1</sup>	97.80(11)
O(15) <sup>#2</sup> –Co(3)–O(30) <sup>#1</sup>	86.71(11)	O(24)–Co(3)–O(10)	93.31(11)
O(22)–Co(3)–O(30) <sup>#1</sup>	89.11(11)	O(15) <sup>#2</sup> –Co(3)–O(10)	82.42(10)
O(22)–Co(3)–O(10)	102.90(10)	O(14)–Co(3)–O(10)	84.42(11)
O(23)–Co(4)–O(7) <sup>#1</sup>	95.96(11)	O(23)–Co(4)–O(11) <sup>#1</sup>	106.86(11)
O(23)–Co(4)–N(1)	114.84(12)	O(23)–Co(4)–O(20) <sup>#3</sup>	95.87(11)
N(1)–Co(4)–O(7) <sup>#1</sup>	84.24(11)	N(1)–Co(4)–O(20) <sup>#3</sup>	94.37(11)
O(11) <sup>#1</sup> –Co(4)–N(1)	137.47(12)	O(11) <sup>#1</sup> –Co(4)–O(7) <sup>#1</sup>	83.54(11)
O(11) <sup>#1</sup> –Co(4)–O(20) <sup>#3</sup>	89.23(12)	O(21)–Co(5)–O(8) <sup>#1</sup>	86.02(10)
O(22)–Co(5)–O(21)	101.90(10)	O(21)–Co(5)–O(5)	93.34(11)
O(22)–Co(5)–O(24)	80.02(10)	O(23)–Co(5)–O(24)	97.17(10)
O(22)–Co(5)–O(5)	92.72(11)	O(23)–Co(5)–O(8) <sup>#1</sup>	93.13(11)
O(22)–Co(5)–O(8) <sup>#1</sup>	87.01(11)	O(23)–Co(5)–O(21)	80.91(10)
O(5)–Co(5)–O(24)	85.77(11)	O(24)–Co(6)–O(31) <sup>#1</sup>	110.57(11)
O(24)–Co(5)–O(8) <sup>#1</sup>	94.89(11)	O(24)–Co(6)–O(16) <sup>#2</sup>	94.06(11)
O(24)–Co(6)–N(2)	112.40(12)	O(31) <sup>#1</sup> –Co(6)–O(16) <sup>#2</sup>	88.76(11)
O(31) <sup>#1</sup> –Co(6)–N(2)	136.70(12)	N(2)–Co(6)–O(16) <sup>#2</sup>	93.30(11)
O(24)–Co(6)–O(7) <sup>#1</sup>	93.17(11)	O(31) <sup>#1</sup> –Co(6)–O(7) <sup>#1</sup>	87.85(11)
N(2)–Co(6)–O(7) <sup>#1</sup>	84.78(11)	O(19) <sup>#3</sup> –Co(7)–O(4)	84.11(11)
O(18)–Co(7)–O(19) <sup>#3</sup>	93.37(10)	O(21)–Co(7)–O(23)	79.52(10)
O(18)–Co(7)–O(4)	88.94(10)	O(19) <sup>#3</sup> –Co(7)–O(23)	92.83(10)
O(21)–Co(7)–O(4)	102.37(11)	O(18)–Co(7)–O(21)	94.76(10)
O(4)–Co(7)–O(23)	87.94(10)	O(23)–Co(7)–O(12) <sup>#1</sup>	100.64(10)
O(18)–Co(7)–O(12) <sup>#1</sup>	84.00(10)	O(19) <sup>#3</sup> –Co(7)–O(12) <sup>#1</sup>	81.75(10)
O(21)–Co(7)–O(12) <sup>#1</sup>	92.78(10)		

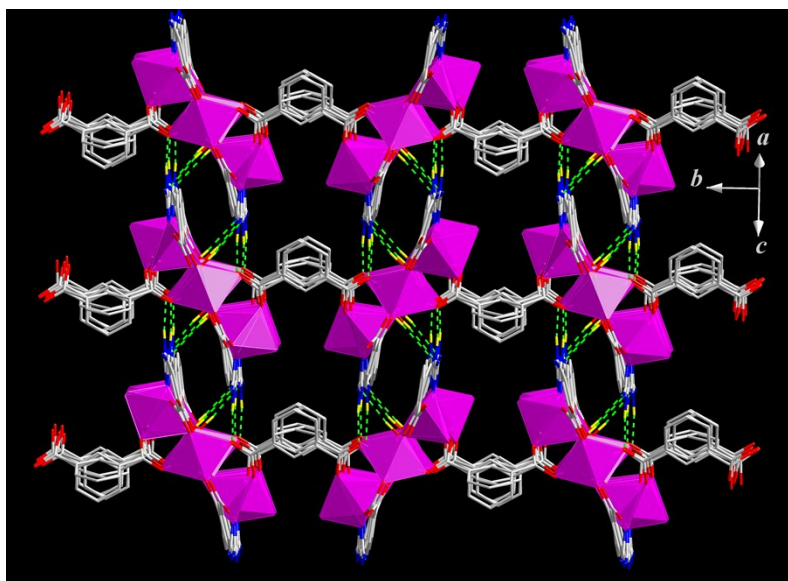
<sup>a</sup> Symmetry codes: <sup>#1</sup> 1 – x, y – 1/2, 1/2 – z; <sup>#2</sup> – x, –y, –z; <sup>#3</sup> 2 – x, –y, 1 – z.

**Table S2.** Hydrogen-bonding parameters (Å, deg) for **1** and **2**<sup>a</sup>

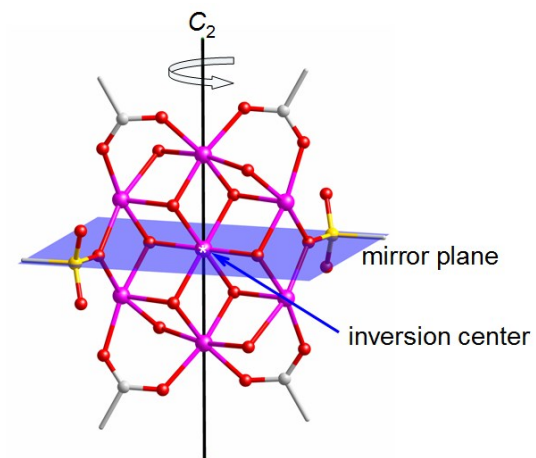
D–H···A	<i>d</i> (D–H)	<i>d</i> (H···A)	<i>d</i> (D..A)	∠DHA
<b>1</b>				
N10–H10B···O29A <sup>#1</sup>	0.86	2.06	2.898(8)	165.2
N5–H5B···O28	0.86	2.14	2.949(6)	158.1
O21–H21···N9 <sup>#2</sup>	0.82	2.46	3.275(1)	178.0
O24–H24A···N4 <sup>#1</sup>	0.82	2.36	3.178(2)	174.0
<b>2</b>				
O7W–H7WA···O4 <sup>#1</sup>	0.84	2.26	2.889(17)	131.8
O7W–H7WB···N1 <sup>#2</sup>	0.84	1.74	2.57(3)	166.8
O7W–H7WB···N7 <sup>#3</sup>	0.84	1.92	2.73(14)	161.3

<sup>a</sup> Symmetry codes: for **1** <sup>#1</sup>  $x-1, 1/2-y, z-1/2$ ; <sup>#2</sup>  $1-x, -y, 1-z$ . For **2**: <sup>#1</sup>  $2-x, 1-y, 1-z$ ; <sup>#2</sup>  $x+1/2, y+$

$1/2, z+1/2$ ; <sup>#3</sup>  $x+1/2, 3/2-y, z+1/2$ .



**Fig. S1** 3D supramolecular network of **1** formed by hydrogen-bonding interactions.



**Fig. S2** Crystallographic symmetry in the local Co<sup>II</sup><sub>7</sub> disc of **2**.

**Table S3.** Selected bond distance (Å) and angles (°) for **2**<sup>a</sup>

Co(1)–O(5)	2.097(11)	Co(3)–O(5) <sup>#3</sup>	2.061(10)
Co(1)–O(6)	2.105(13)	Co(3)–O(2) <sup>#6</sup>	2.067(11)
Co(2)–O(5)	2.018(11)	Co(3)–N(3)	2.17(2)
Co(2)–O(1) <sup>#4</sup>	2.086(12)	Co(3)–O(3)	2.209(12)
Co(2)–O(7)	2.227(11)	Co(3)–O(7) <sup>#3</sup>	2.301(11)
Co(3)–O(6)	2.044(9)	N(9)–Co(3) <sup>#1</sup>	1.976(13)
O(5) <sup>#1</sup> –Co(1)–O(5) <sup>#2</sup>	180.0(4)	O(6)–Co(3)–O(5) <sup>#3</sup>	83.3(5)
O(5) <sup>#1</sup> –Co(1)–O(5)	100.9(6)	O(6)–Co(3)–O(2) <sup>#6</sup>	167.8(5)
O(5) <sup>#2</sup> –Co(1)–O(5)	79.1(6)	O(5) <sup>#3</sup> –Co(3)–O(2) <sup>#6</sup>	91.1(5)
O(5) <sup>#2</sup> –Co(1)–O(6)	80.9(4)	O(6)–Co(3)–N(3)	88.8(9)
O(5)–Co(1)–O(6)	99.1(4)	O(5) <sup>#3</sup> –Co(3)–N(3)	166.7(8)
O(6)–Co(1)–O(6) <sup>#3</sup>	180.0	O(2) <sup>#6</sup> –Co(3)–N(3)	98.6(8)
O(5)–Co(2)–O(5) <sup>#2</sup>	82.9(6)	O(6)–Co(3)–O(3)	81.3(5)
O(5)–Co(2)–O(1) <sup>#4</sup>	168.9(4)	O(5) <sup>#3</sup> –Co(3)–O(3)	86.4(5)
O(5) <sup>#2</sup> –Co(2)–O(1) <sup>#4</sup>	91.1(5)	O(2) <sup>#6</sup> –Co(3)–O(3)	109.2(5)
O(1) <sup>#4</sup> –Co(2)–O(1) <sup>#5</sup>	96.2(7)	N(3)–Co(3)–O(3)	81.8(10)
O(1) <sup>#4</sup> –Co(2)–O(7) <sup>#2</sup>	85.2(5)	O(6)–Co(3)–O(7) <sup>#3</sup>	87.0(5)
O(5)–Co(2)–O(7)	87.3(4)	O(5) <sup>#3</sup> –Co(3)–O(7) <sup>#3</sup>	84.3(4)
O(5) <sup>#2</sup> –Co(2)–O(7)	85.2(4)	O(2) <sup>#6</sup> –Co(3)–O(7) <sup>#3</sup>	81.7(4)
O(1) <sup>#4</sup> –Co(2)–O(7)	101.6(4)	N(3)–Co(3)–O(7) <sup>#3</sup>	106.0(10)
O(7) <sup>#2</sup> –Co(2)–O(7)	170.0(6)	O(3)–Co(3)–O(7) <sup>#3</sup>	165.9(4)

<sup>a</sup> Symmetry codes: <sup>#1</sup>  $x, 1 - y, z$ ; <sup>#2</sup>  $1 - x, y, 1 - z$ ; <sup>#3</sup>  $1 - x, 1 - y, 1 - z$ ; <sup>#4</sup>  $3/2 - x, y + 1/2, 1/2 - z$ ; <sup>#5</sup>  $x - 1/2, y + 1/2, z + 1/2$ ; <sup>#6</sup>  $3/2 - x, 1/2 - y, 1/2 - z$ .

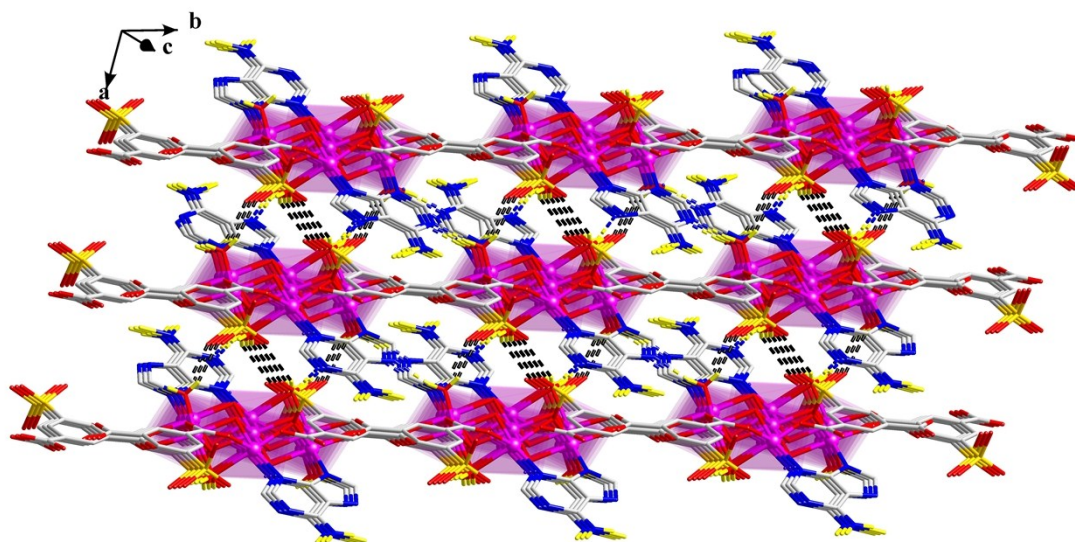
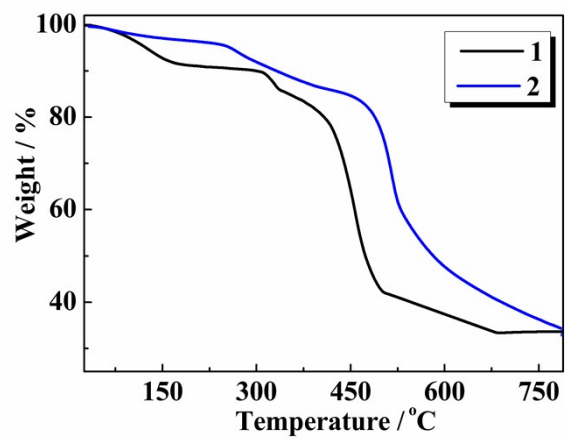
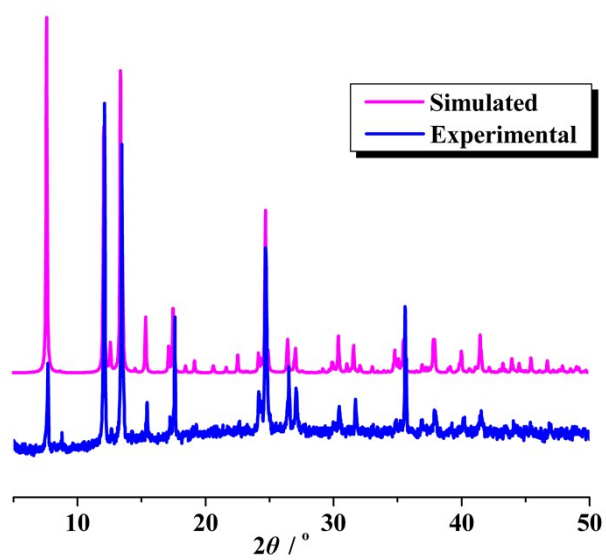
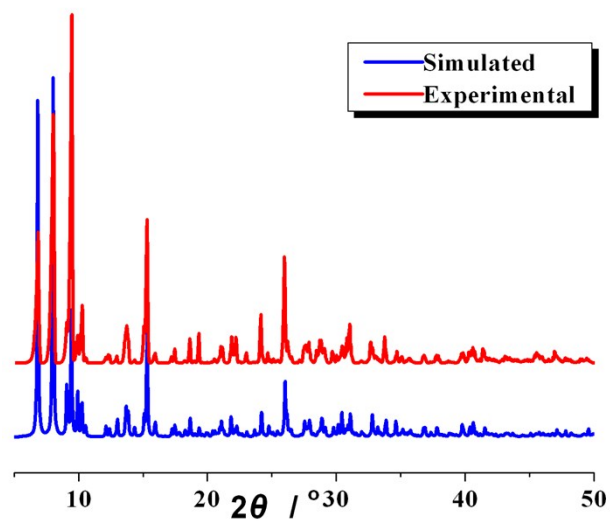


Fig. S3 3D supramolecular network of 2 generated by hydrogen-bonding interactions.

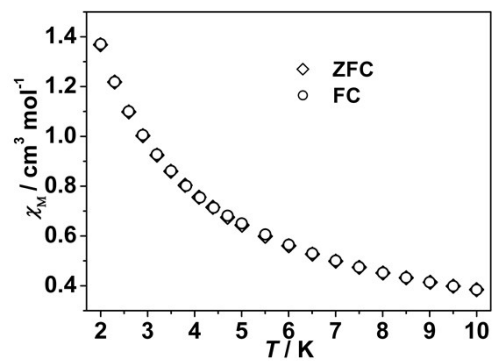


**Fig. S4** TG curves for 1 and 2.

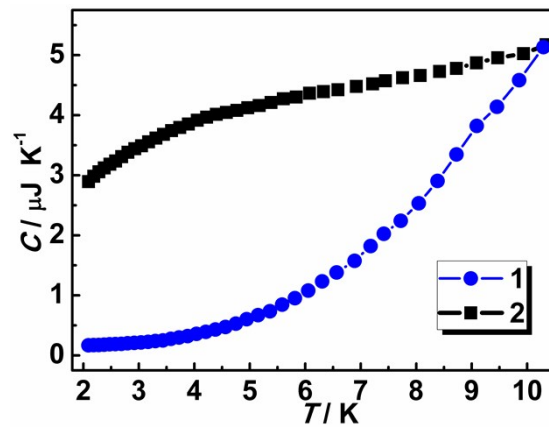




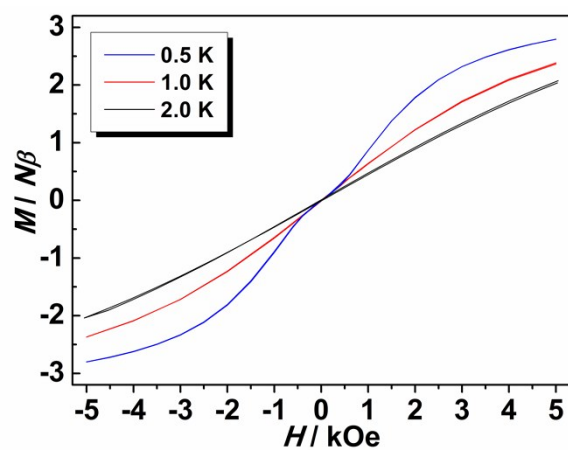
**Fig. S5** Simulated and experimental PXRd patterns for **1** (top) and **2** (bottom).



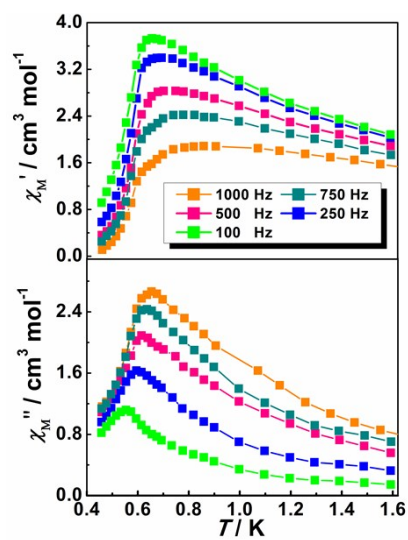
**Fig. S6** FC and ZFC magnetizations for **1** at 50.0 Oe dc field.



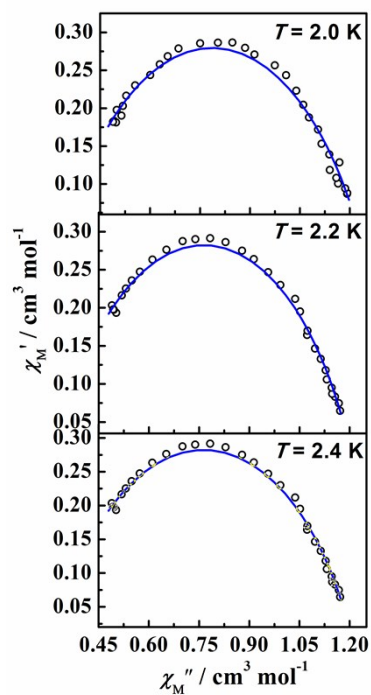
**Fig. S7** Temperature-dependent heat capacity for **1** and **2** measured at zero dc field.



**Fig. S8** Magnetization ( $M$ ) vs. applied dc field ( $H$ ) hysteresis loops for **1** at the indicated temperatures.



**Fig. S9** Temperature dependence of real ( $\chi'$ ) and imaginary ( $\chi''$ ) components of the ac susceptibility of **1** in zero dc field with an oscillating field of 3.5 Oe over frequencies of 100–1500 Hz.

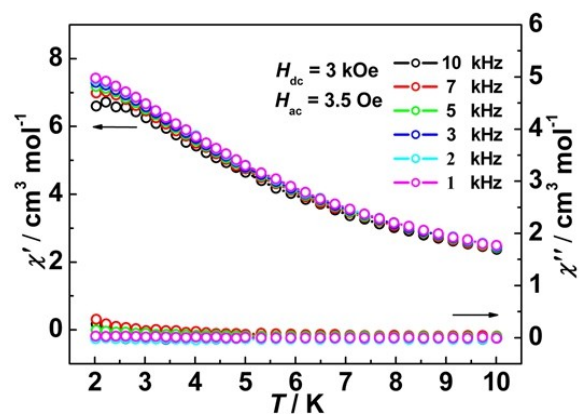


**Fig. S10** Cole–Cole plots of **1** at different temperatures.

**Table S4.** Parameters obtained by fitting the ac magnetic susceptibilities between 2.0 and 2.4 K with generalized Debye-model for **1**.

$T / \text{K}$	$\chi_0 / \text{cm}^3 \text{mol}^{-1}$	$\chi_\infty / \text{cm}^3 \text{mol}^{-1}$	$\tau \text{ (s)}$	$\alpha$	$R$
2.0 K	1.24	0.32	$6 \times 10^{-4}$	0.31	$5.5 \times 10^{-3}$
2.2 K	1.21	0.32	$5 \times 10^{-4}$	0.28	$5.8 \times 10^{-3}$
2.4 K	1.17	0.28	$3 \times 10^{-4}$	0.25	$2.1 \times 10^{-3}$

$$R = \frac{\sum[(\chi'_{\text{obs}} - \chi'_{\text{cal}})^2 + (\chi''_{\text{obs}} - \chi''_{\text{cal}})^2]}{\sum(\chi'_{\text{obs}}{}^2 + \chi''_{\text{obs}}{}^2)}$$



**Fig. S11** Temperature dependence of real ( $\chi'$ ) and imaginary ( $\chi''$ ) components of the ac susceptibility for **2** under  $H_{dc} = 3000$  and  $H_{ac} = 3.5$  Oe over the frequencies of 1–10 kHz.