

# Synthesis, Structure and Properties of [Co(NCS)<sub>2</sub>(4-(4-Chlorobenzyl)pyridine)<sub>2</sub>]<sub>n</sub>, that shows Slow Magnetic Relaxations and a Metamagnetic Transition

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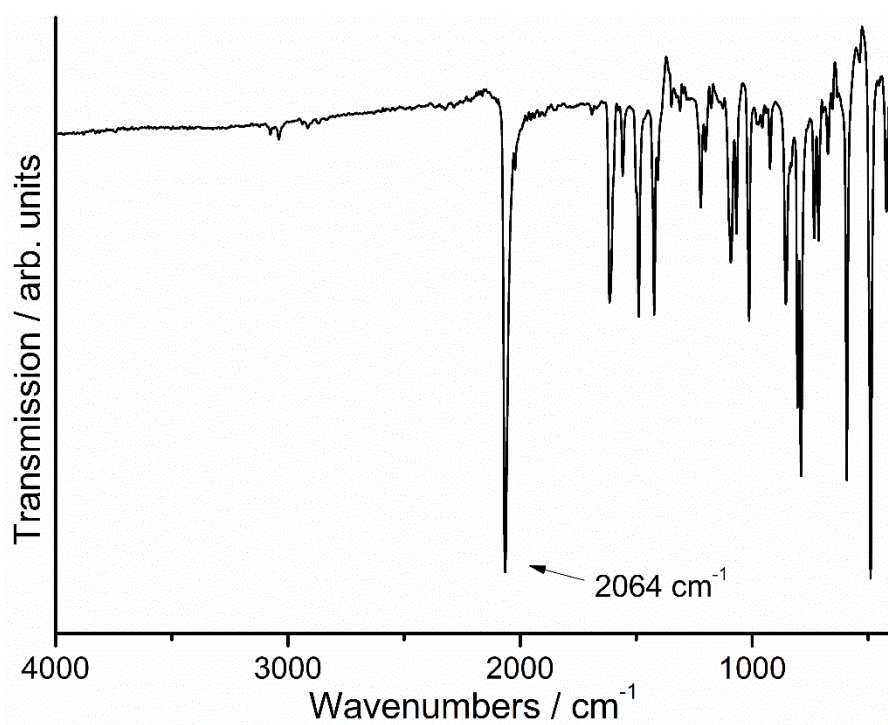


Figure S1: IR spectra of **1**. The CN stretching vibration is given.

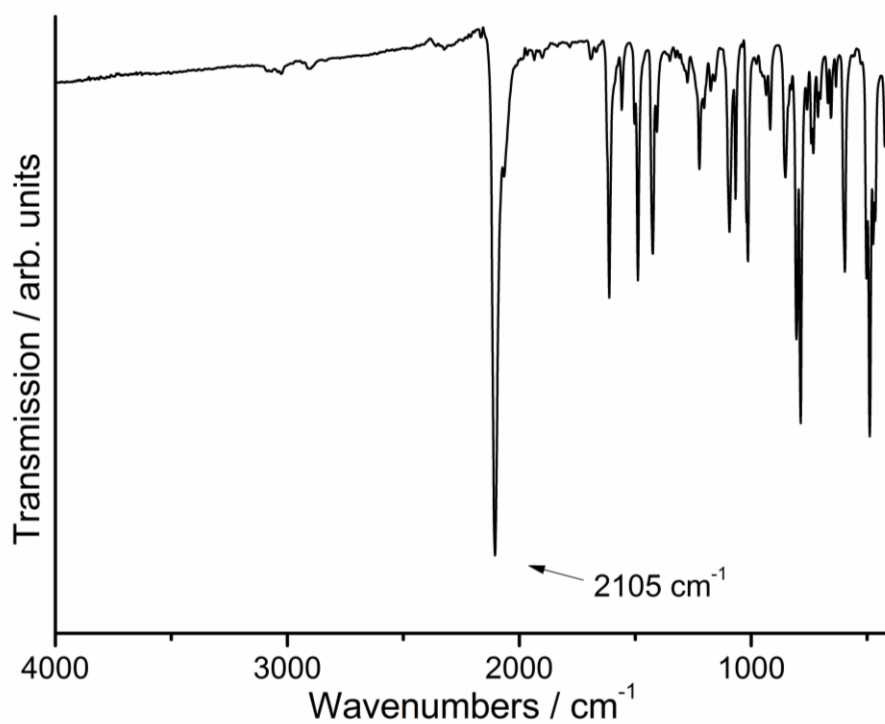


Figure S2: IR spectra of **2**. The CN stretching vibration is given.

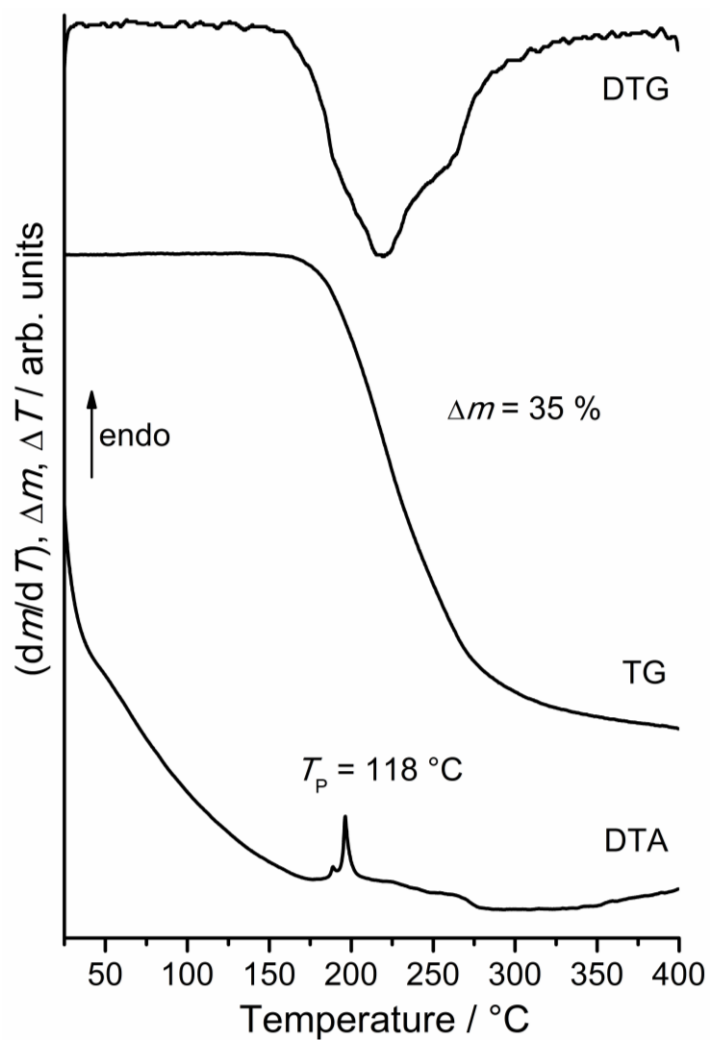


Figure S3. DTG, TG and DTA curves for **1**. Heating rate = 2 °C/min. Given are the mass change in % and the peak temperature in  $T_p$  in °C.

Table S 1: Selected bond length (Å) and angles (°) for **1**.

Co(1)-N(1)	2.0943(15)	Co(1)-N(31)	2.2412(15)
Co(1)-N(11)	2.1641(15)	N(11)-Co(1)-N(11A)	180.00(5)
N(1)-Co(1)-N(1A)	180.00(8)	N(1)-Co(1)-N(31A)	88.66(6)
N(1)-Co(1)-N(11)	89.21(6)	N(11)-Co(1)-N(31)	90.35(6)
N(1)-Co(1)-N(11A)	90.79(6)	N(11)-Co(1)-N(31A)	89.65(6)
N(1)-Co(1)-N(31)	91.34(6)		

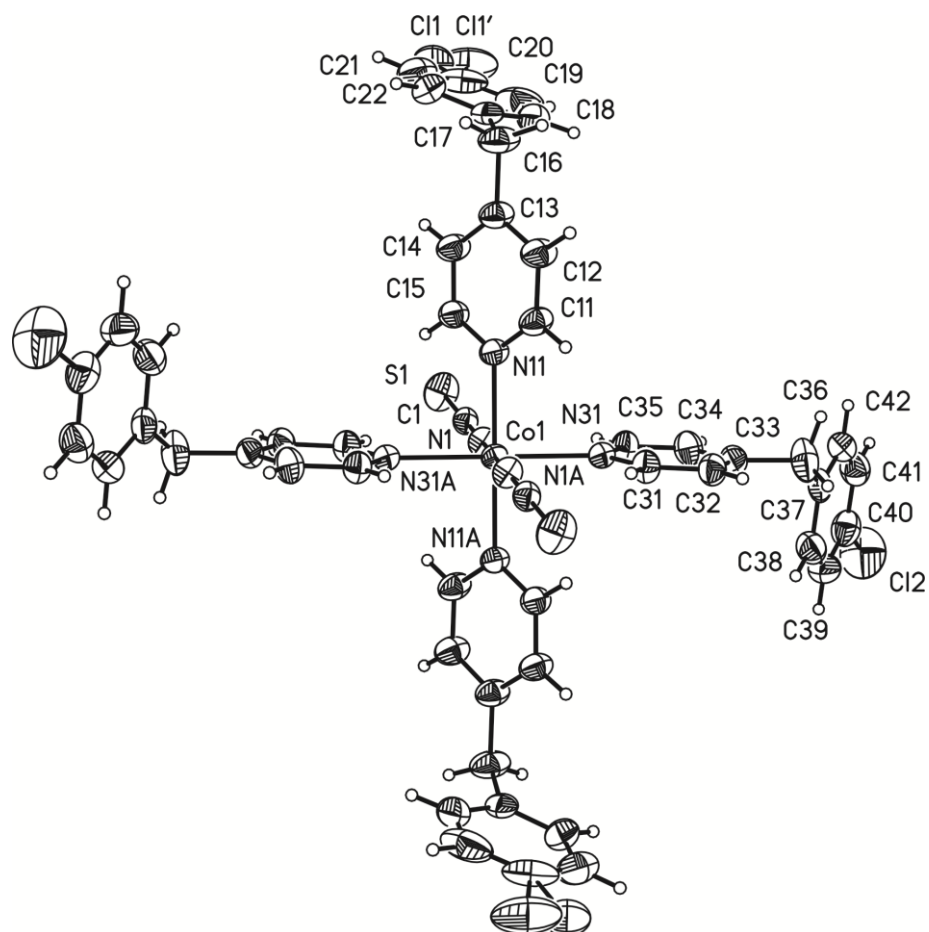


Figure S4: ORTEP plot of **1** with labelling and displacement ellipsoids drawn at 50 % probability level. Symmetry transformation used to generate equivalent atoms: A:  $-x+3/2, -y+1/2, -z+1$ .

Table S2: Selected bond length (Å) and angles (°) for **2**.

Co(1)-N(2)	2.060(2)	Co(2)-N(1A)	2.066(2)
Co(1)-S(1)	2.5694(10)	Co(2)-S(2)	2.6066(10)
Co(1)-N(11)	2.165(2)	Co(2)-N(31)	2.145(2)
N(2)-Co(1)-N(2A)	180.00(9)	N(1B)-Co(2)-N(1A)	180.00(9)
N(2)-Co(1)-N(11)	89.93(8)	N(1A)-Co(2)-N(31)	89.83(8)
N(2)-Co(1)-N(11A)	90.07(8)	N(1A)-Co(2)-N(31C)	90.17(8)
N(2)-Co(1)-S(1)	86.73(6)	N(1A)-Co(2)-S(2)	94.23(6)
N(2)-Co(1)-S(1A)	93.27(6)	N(1A)-Co(2)-S(2C)	85.77(6)

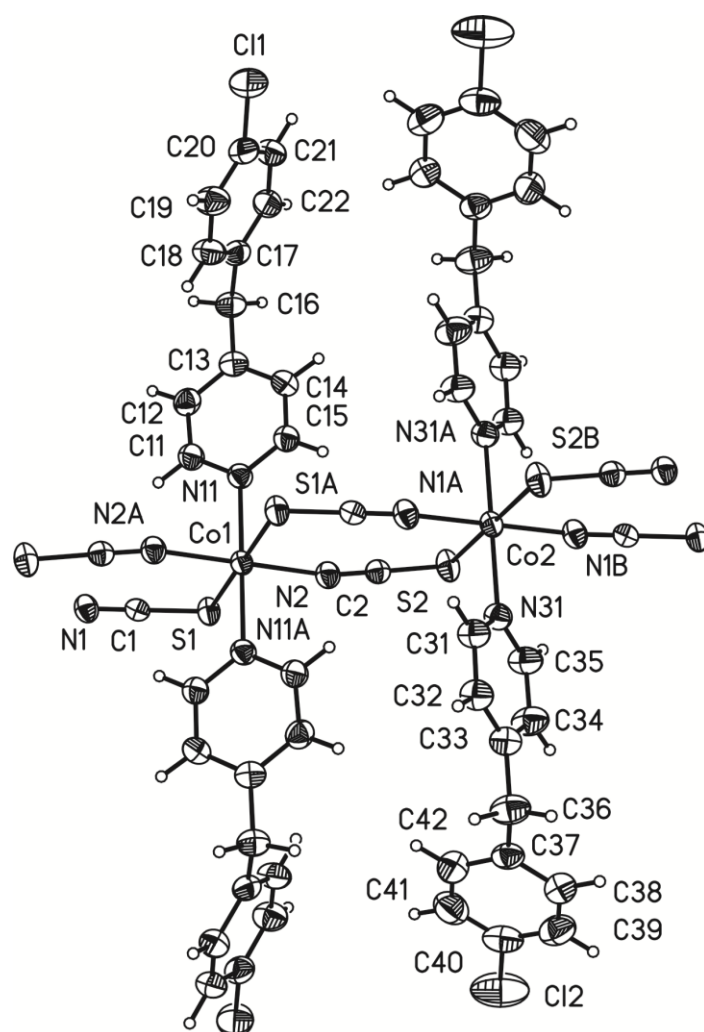


Figure S5: ORTEP plot of **2** with labelling and displacement ellipsoids drawn at 50 % probability level. Symmetry transformation used to generate equivalent atoms: A:  $-x+1, -y+1, -z+1$ , B:  $x-1, y+1, z$ , C:  $-x, -y+2, -z+1$ , D:  $x+1, y-1, z$ .

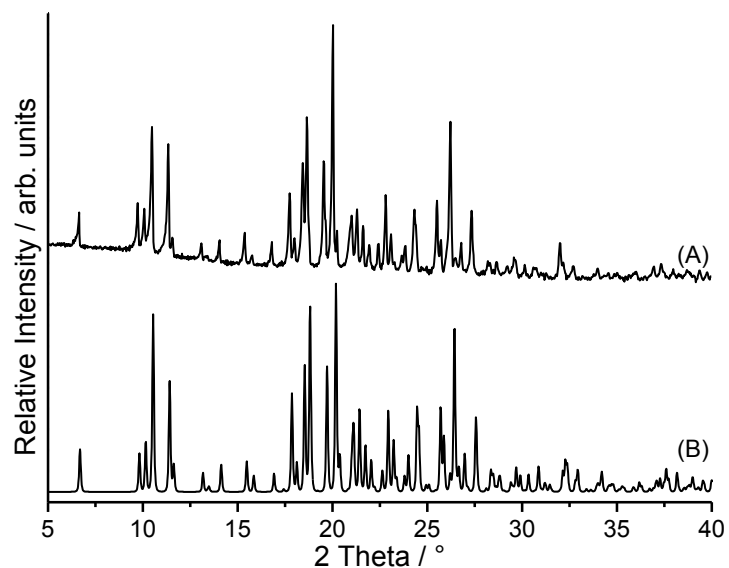


Figure S6: Experimental (top) and calculated (bottom) x-ray powder pattern for compound **1**.

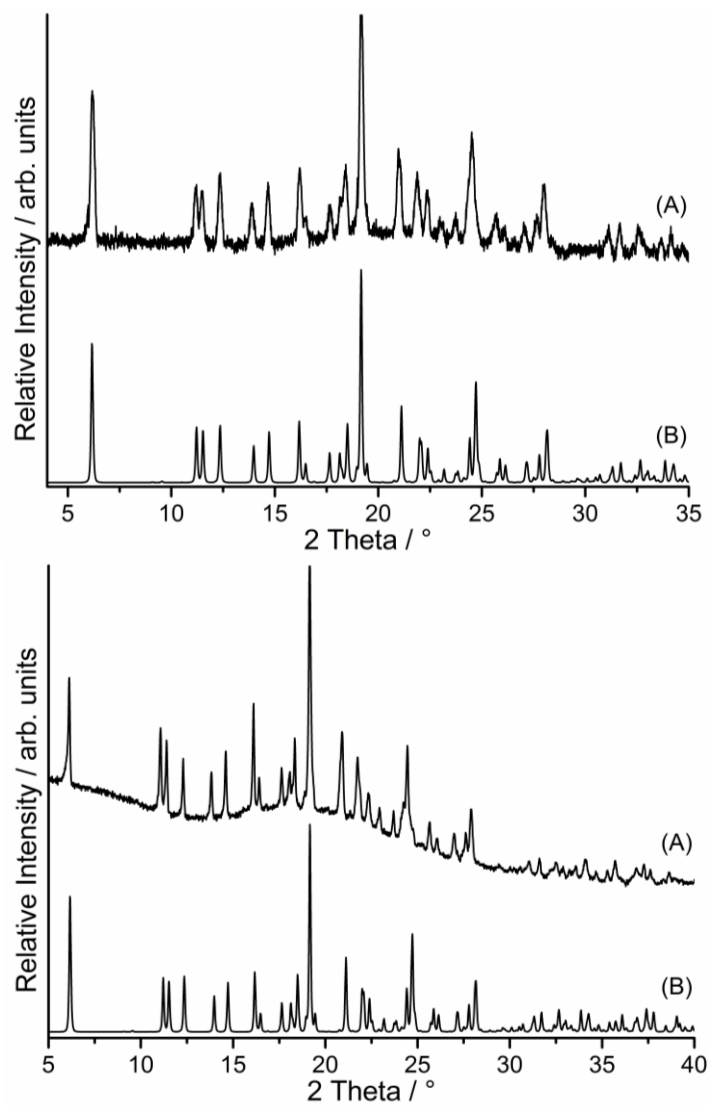


Figure S7: Experimental (top) and calculated (bottom) x-ray powder patterns for two samples of compound **2**.

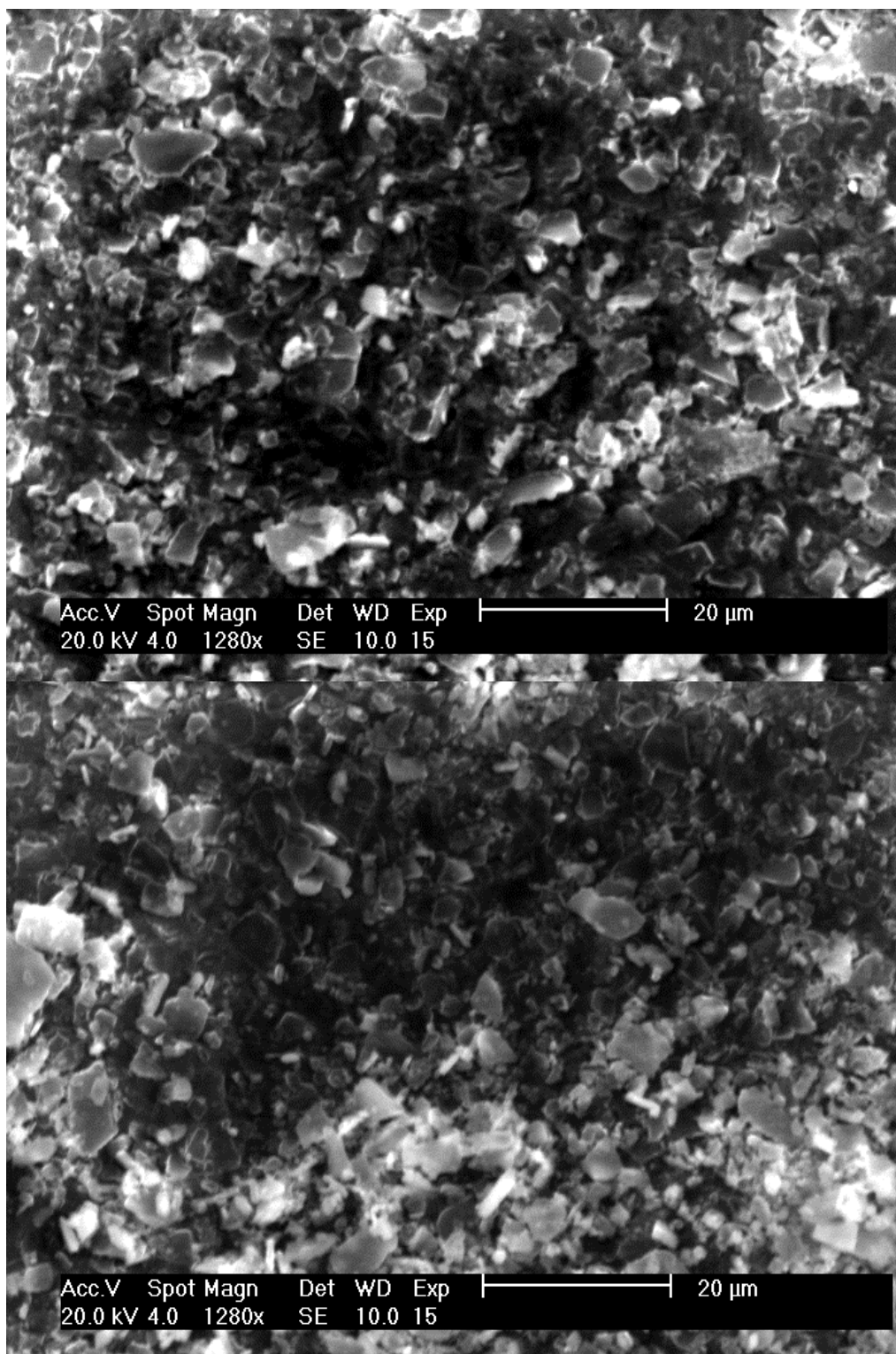


Figure S8: Selected SEM images of the crystals of compound 2.

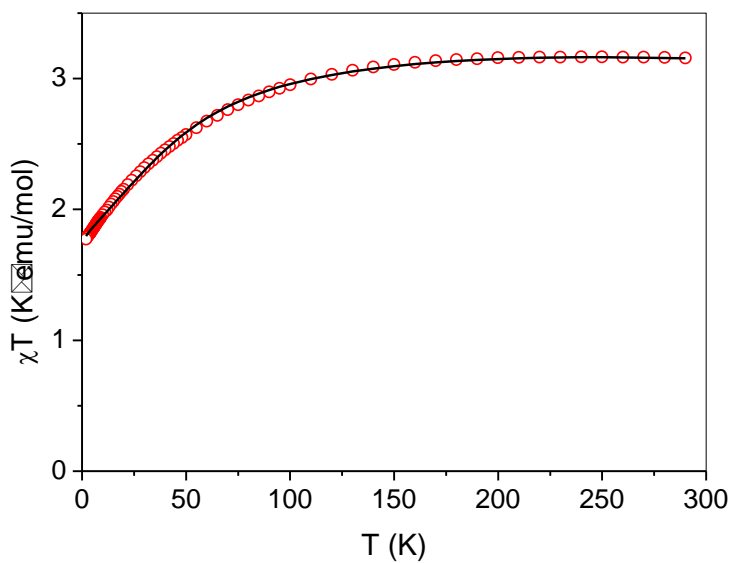


Figure S9. Temperature dependence of  $\chi$  vs.  $T$  (top) and  $\chi T$  vs.  $T$  (bottom) for **1**. The solid line is a fit done by an assumption of the axially distorted octahedral coordination of the Co(II) ion (spin orbit coupling  $\lambda = -100 \pm 3 \text{ cm}^{-1}$ , orbital reduction factor  $\kappa = 1.0 \pm 0.02$ ).

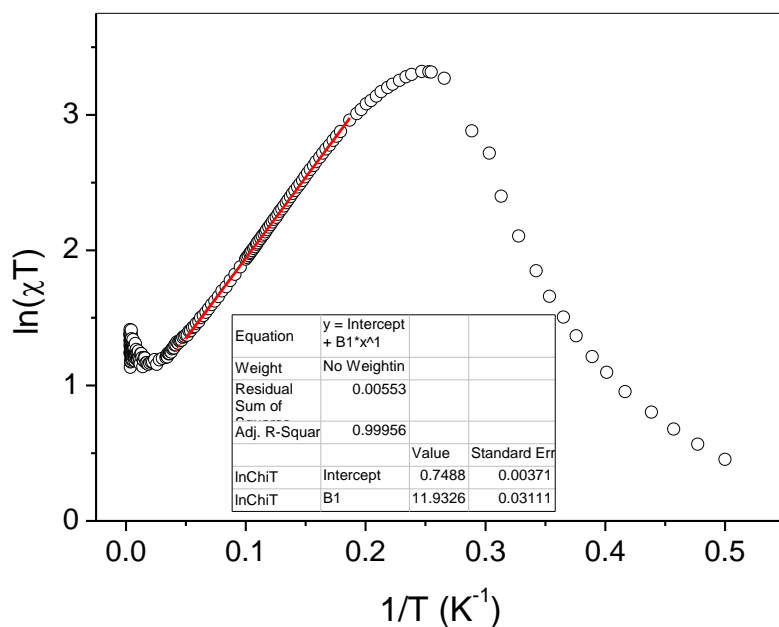


Figure S10:  $\ln(\chi T)$  vs.  $1/T$  dependence for compound **2**.



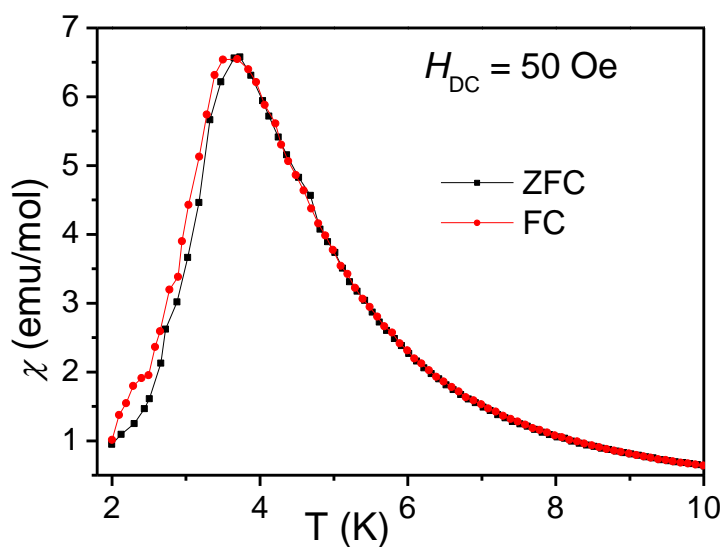


Figure S11. Comparison of  $\chi(T)$  curves registered in FC and ZFC regimes for compound **2**.

Table S3: Fitting results of the AC data for compound **2**.

T (K)	$\chi_0$	$\chi_\infty$	$\tau$ (s)	$\alpha$	Reduced $\chi_{\text{sqr}}$
3.4	4.300	0.0	5.205E-6	0.087	5.713E-4
3.3	3.590	0.0	7.102E-6	0.108	3.873E-4
3.2	2.983	0.0	9.800E-6	0.125	3.112E-4
3.1	2.484	0.0	1.405E-5	0.153	2.185E-4
3.0	2.091	0.110	2.300E-5	0.164	2.859E-4
2.9	1.753	0.129	3.581E-5	0.169	9.762E-5
2.8	1.495	0.122	5.715E-5	0.189	6.379E-5
2.7	1.296	0.109	8.516E-5	0.206	2.891E-4
2.6	1.134	0.095	1.508E-4	0.250	1.472E-4
2.5	1.007	0.104	2.868E-4	0.265	5.969E-5
2.4	0.913	0.106	5.533E-4	0.286	1.172E-5
2.3	0.821	0.112	1.02E-3	0.294	1.690E-5
2.2	0.792	0.105	2.19E-3	0.36	2.596E-5
2.1	0.770	0.110	5.28E-3	0.38	5.864E-6
2.0	0.679	0.117	8.08E-3	0.37	2.3893E-5