

Electronic Supplementary Information

Magnetic investigations of monocrystalline $[\text{Co}(\text{NCS})_2(\text{L})_2]_n$: new insights into single-chain relaxations

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1 – $[\text{Co}(\text{NCS})_2(4-(3\text{-phenylpropyl})\text{pyridine})_2]_n$

1-Cd – $[\text{Co}_x\text{Cd}_{1-x}(\text{NCS})_2(4-(3\text{-phenylpropyl})\text{pyridine})_2]_n$

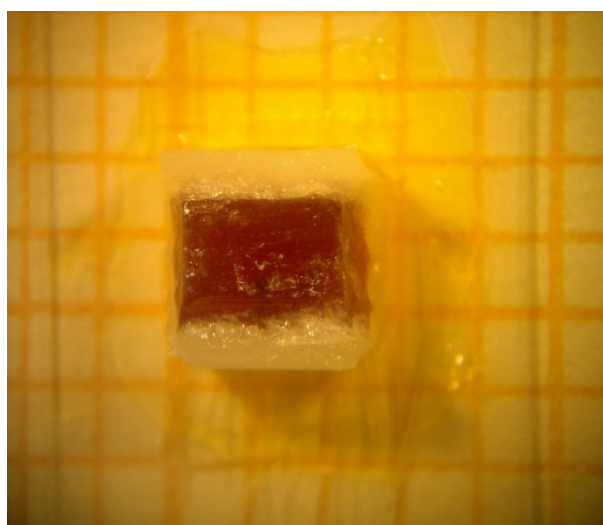
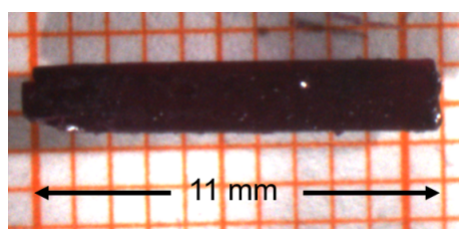


Figure S1. The photos of a single crystal sample of **1**. Left: the whole single crystal. Right: a fragment cut for magnetic measurements, 5.37 mg. The crystal is glued to the holder made of delrin by small amount of Eicosane.

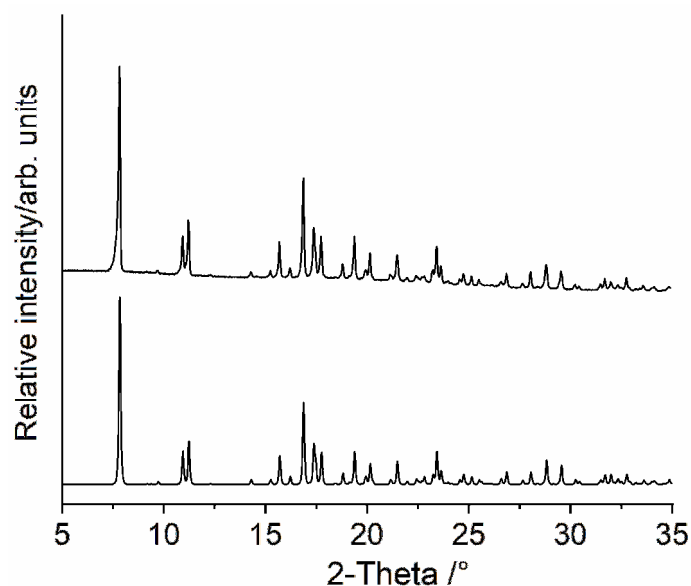


Figure S2a. Experimental (top) and calculated powder pattern (bottom) for **1-Cd** measured with Cu K α radiation. Please note: The crystal structure reported in literature was measured at low temperatures. Therefore, for the calculation of the PXRD pattern the unit cell parameters obtained from a Pawley fit of a powder pattern of to Co compound measured at room-temperature were used.

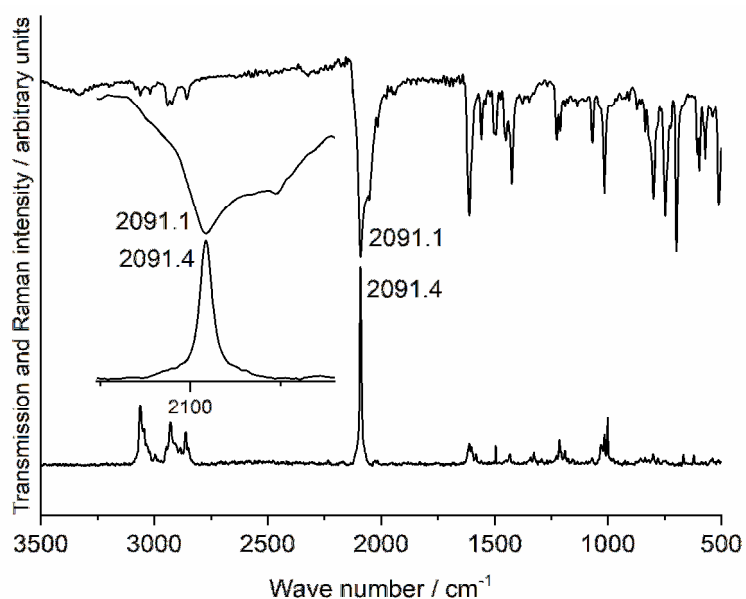


Figure S2b. IR (top) and Raman spectra (bottom) of **1-Cd** ($x = 0.013$). Given are the values of the CN stretching vibration and the inset shows an enlargement of this region. The spectra are very close to that of the pure Cd compound.

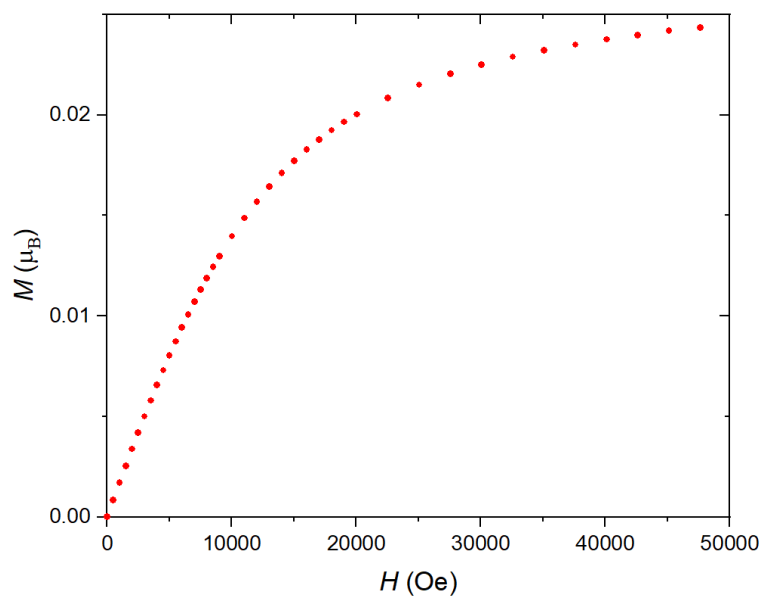


Figure S3. The magnetization curve for **1-Cd** measured at 2 K. The content of Co(II) ions, x , estimated on the basis of comparison of the values of magnetization in 50 kOe for **1-Cd** including diamagnetic corrections and **1** (see Figure 6 in ref. [S1]) and is in agreement with the value from AAS (0.013).

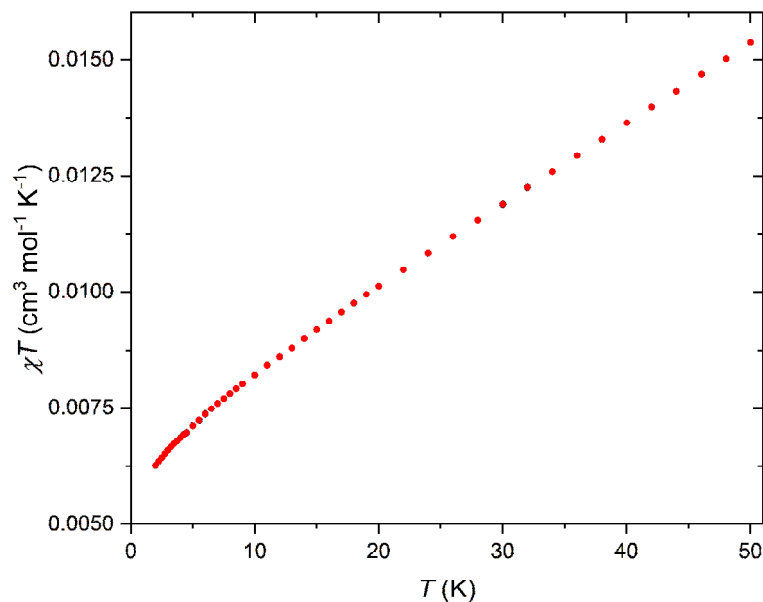


Figure S4. The magnetic susceptibility for **1-Cd** ($x = 0.013$), measured at 100 Oe. The almost linear decrease of χT with decreasing temperature means that Co moments do not interact, i.e. are well diluted. Any presence of **1** would manifest as increase around 3.3 K.

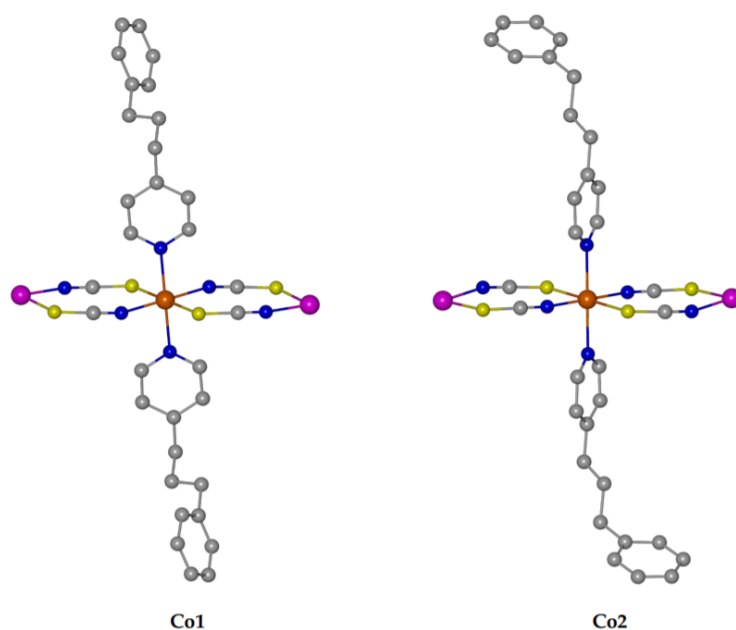


Figure S5. *Ab initio* computational models for **1** (Co – orange; Zn – pink). Hydrogen atoms have been omitted for clarity.

Table S1. Relative CASSCF and CASPT2 energies (in cm^{-1}) of all quartet and the nine lowest doublet states for the mononuclear cobalt(II) computational models (Co1 and Co2) for **1**.

2S + 1	Term	Subterm	Co1 CASSCF	Co1 CASPT2	Co2 CASSCF	Co2 CASPT2
4	^4F	$^4\text{T}_{1g}$	0	0	0	0
			505	619	669	588
			1114	1051	1131	1155
		$^4\text{T}_{2g}$	5606	6668	5308	5590
			7862	9033	7807	7916
			8489	9554	8841	8936
		$^4\text{A}_{2g}$	15471	17666	15445	15850
	^4P	$^4\text{T}_{1g}$	21170	18651	21398	21488
			22865	20756	22703	22696
			25961	23765	26183	26529
2	^2G		12118	9476	11999	9362
			15870	13141	16225	13538
			18060	15956	17599	15639
			18591	16510	18508	16467
			19295	16733	19375	16847
			19955	17261	20011	17329
			20861	18210	20951	18312
			21342	18510	21385	18624
			24321	16534	24305	20847

Table S2. Relative RASSI-SO energies (in cm⁻¹) of the ⁴T_{1g} ground state for the mononuclear Co(II) computational models (Co1 and Co2) for **1**.

Kramers doublet	Co1	Co2
1	0	0
2	161	140
3	753	823
4	1007	1081
5	1403	1393
6	1508	1525

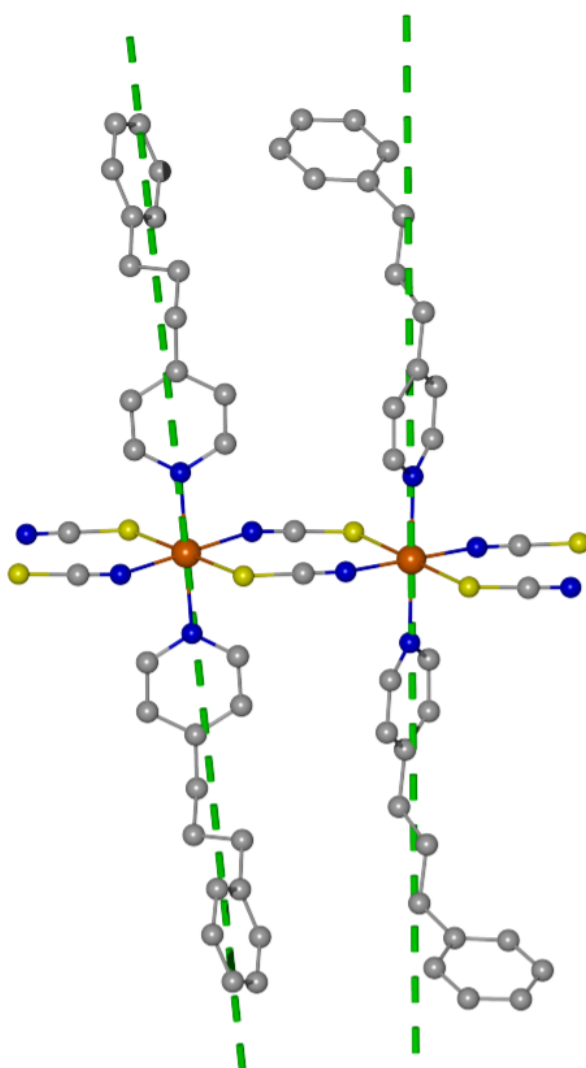


Figure S6. *Ab initio* calculated easy-axes of magnetization ($S_{eff} = 1/2$) of the ground state Kramers doublet for the individual centers Co1 (left) and Co2 (right) projected onto the smallest repeating sequence of the periodic chain for **1** (angle between the axes: 9.3°). Hydrogen atoms have been omitted for clarity.

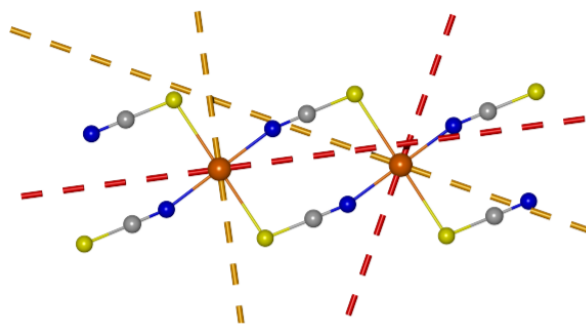


Figure S7. *Ab initio* calculated hard-axes of magnetization ($S_{eff} = 1/2$; red dashed line: g_x ; orange dashed line: g_y) of the ground state Kramers doublet for the individual centers Co1 (left) and Co2 (right) from a top view for **1**. Apical 4-(3-phenylpropyl)pyridine ligands have been omitted for clarity.

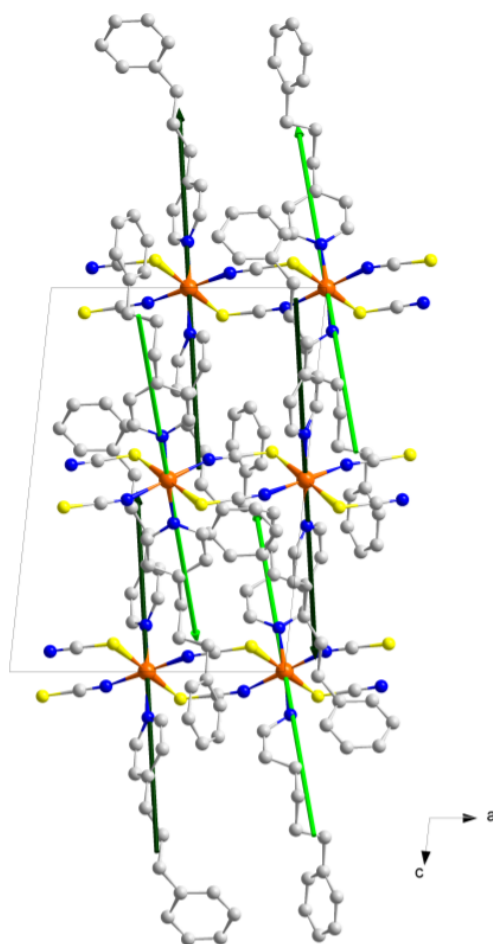


Figure S8. *Ab initio* calculated easy-axes of magnetization ($S_{eff} = 1/2$) of the ground state Kramers doublet for the individual centers Co1 (green) and Co2 (dark green) projected onto the periodic chain structure of **1**. View along $\vec{b}$.

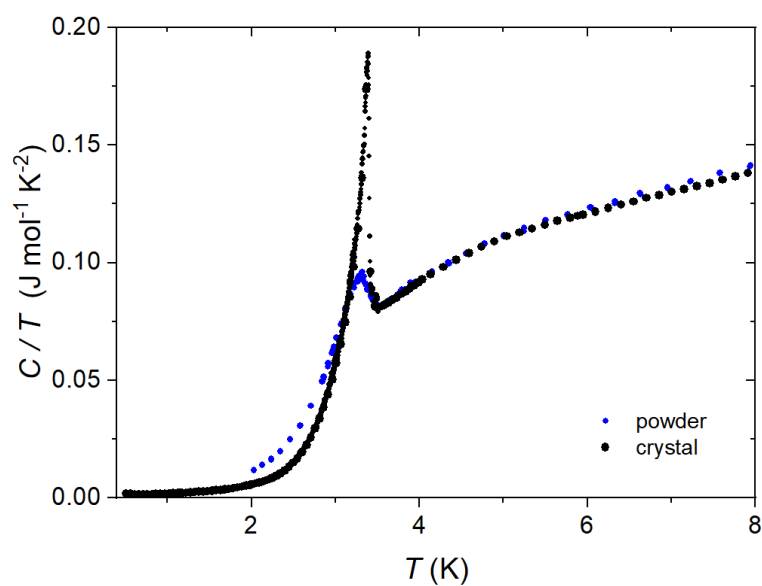


Figure S9. Temperature dependence of the specific heat, C , presented as C/T for a powder and single crystal samples of **1** at $H_{DC} = 0$ Oe.

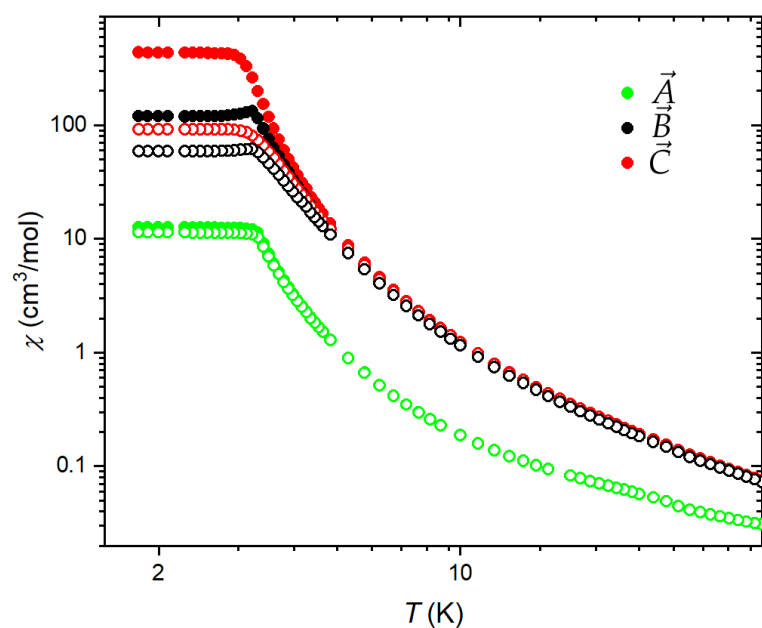


Figure S10. Magnetic susceptibility, χ , measured at $H_{dc} = 100$ Oe for **1** along 3 perpendicular crystallographic directions. Open circles – data as measured, full circles – data corrected for magnetization.^[S3]

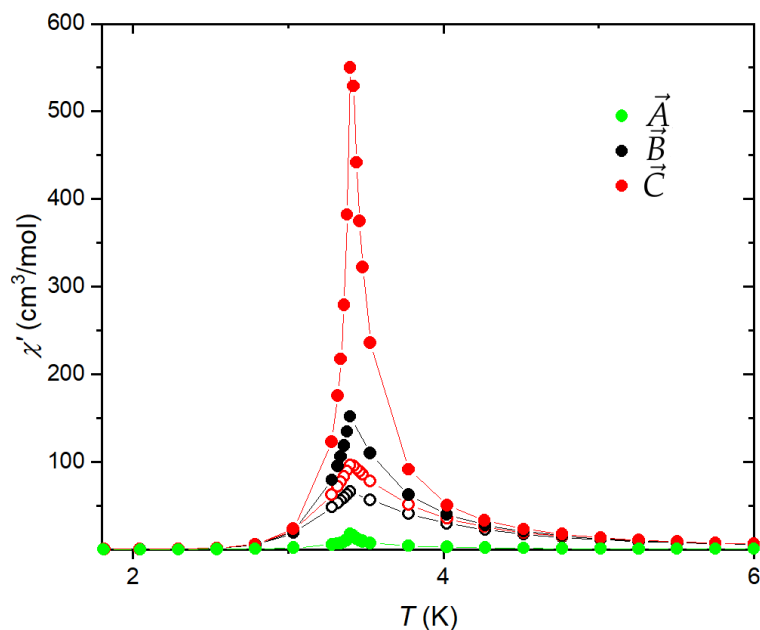


Figure S11. Ac magnetic susceptibility, χ' , measured at $H_{dc} = 0$ Oe, $H_{ac} = 3$ Oe, $f = 10$ Hz for 1 along 3 perpendicular crystallographic directions. Open circles – data as measured, full circles – data corrected for demagnetization.^[S2] The lines are to guide the eye. The demagnetization was included assuming spherical shape of the sample. $\vec{A}$, $\vec{B}$, $\vec{C}$ directions are defined in the main text.

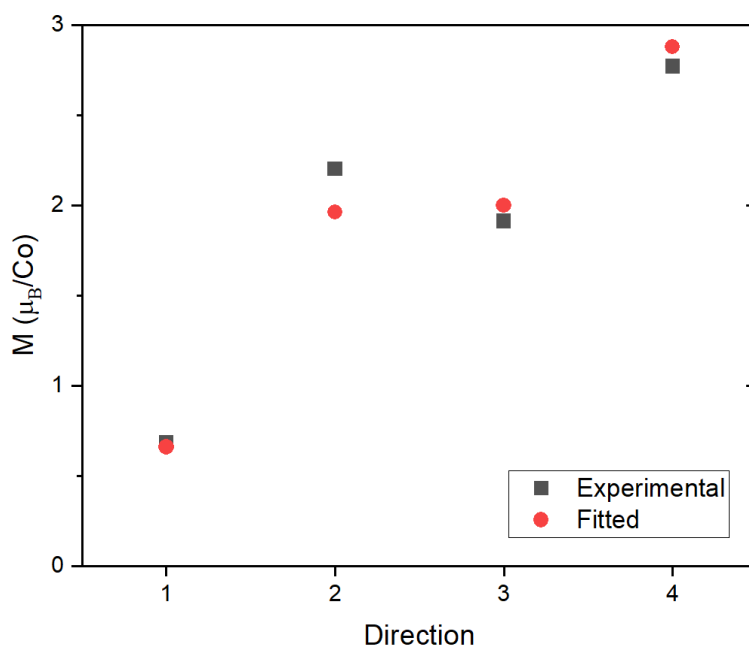


Figure S12. The values of saturation magnetization determined for 4 different directions (1: $\vec{A}$, 2: $\vec{B}$, 3: $\vec{C}$, 4: crystal easy axis) experimentally and fitted.

The conversion of crystallographic coordinates (x, y, z) to Cartesian coordinates (x_c, y_c, z_c) was done using the following equation:

$$\begin{pmatrix} x_c \\ y_c \\ z_c \end{pmatrix} = \begin{pmatrix} a & 0 & a \cdot \cos \beta \\ 0 & b & 0 \\ 0 & 0 & c \cdot \sin \beta \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} = \begin{pmatrix} 11.27740 & 0 & -1.71896 \\ 0 & 16.03430 & 0 \\ 0 & 0 & 15.72110 \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}$$

a, b, c, β – unit cell parameters

Easy axis vector in Cartesian coordinates for the first chain in a unit cell:

$$\vec{a}_1 = (M_{xc}, M_{yc}, M_{zc});$$

The easy axis of a second chain in a unit cell is related to the first one by the C_2 symmetry represented by a matrix:

$$C_2 = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix};$$

According to that, the easy axis vector for the second chain in a unit cell is:

$$\vec{a}_2 = (-M_{xc}, M_{yc}, -M_{zc}).$$

The function used for fitting:

$$\chi^2 = (M_A - F_A)^2 + (M_B - F_B)^2 + (M_C - F_C)^2 + (M_{ea} - F_{ea})^2,$$

where:

M_A, M_B, M_C, M_{ea} – values of magnetization determined experimentally for directions $\vec{A}, \vec{B}, \vec{C}$ and easy axis, respectively, at 2 K and 2 kOe;

F_A, F_B, F_C – projections of the easy axes vectors on the $\vec{A}, \vec{B}, \vec{C}$ directions:

$$F_A = \frac{1}{2} (|\vec{a}_1 \cdot \vec{A}| + |\vec{a}_2 \cdot \vec{A}|)$$

$$F_B = \frac{1}{2} (|\vec{a}_1 \cdot \vec{B}| + |\vec{a}_2 \cdot \vec{B}|)$$

$$F_C = \frac{1}{2} (|\vec{a}_1 \cdot \vec{C}| + |\vec{a}_2 \cdot \vec{C}|)$$

F_{ea} takes the maximal value from: $||(\vec{a}_1 + \vec{a}_2)/2||$ or $||(\vec{a}_1 - \vec{a}_2)/2||$.

To write the magnetic moment in spherical coordinates (M, θ_M, ϕ_M) , conversion of Cartesian coordinates (M_{xc}, M_{yc}, M_{zc}) was done using the following set of equations:

$$\begin{cases} M = \sqrt{M_{xc}^2 + M_{yc}^2 + M_{zc}^2} \\ \theta_M = \cos^{-1} \left(\frac{M_{zc}}{\sqrt{M_{xc}^2 + M_{yc}^2 + M_{zc}^2}} \right) \\ \phi_M = \tan^{-1} \left(\frac{M_{yc}}{M_{xc}} \right) \end{cases}$$

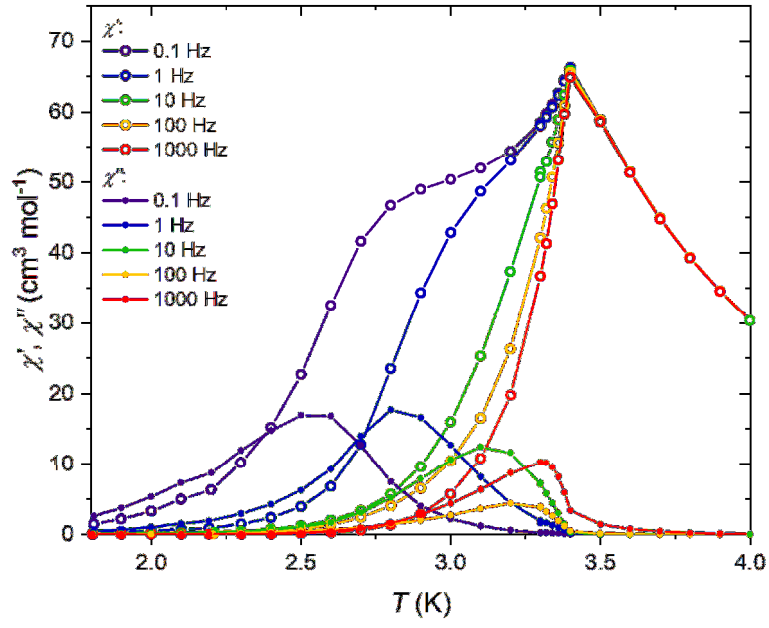


Figure S13. The temperature dependence of ac magnetic susceptibility measured for **1** at $H_{dc} = 0$ and $H_{ac} = 3$ Oe along the $\vec{B} = \vec{b}^* - \vec{c}^*$ direction.

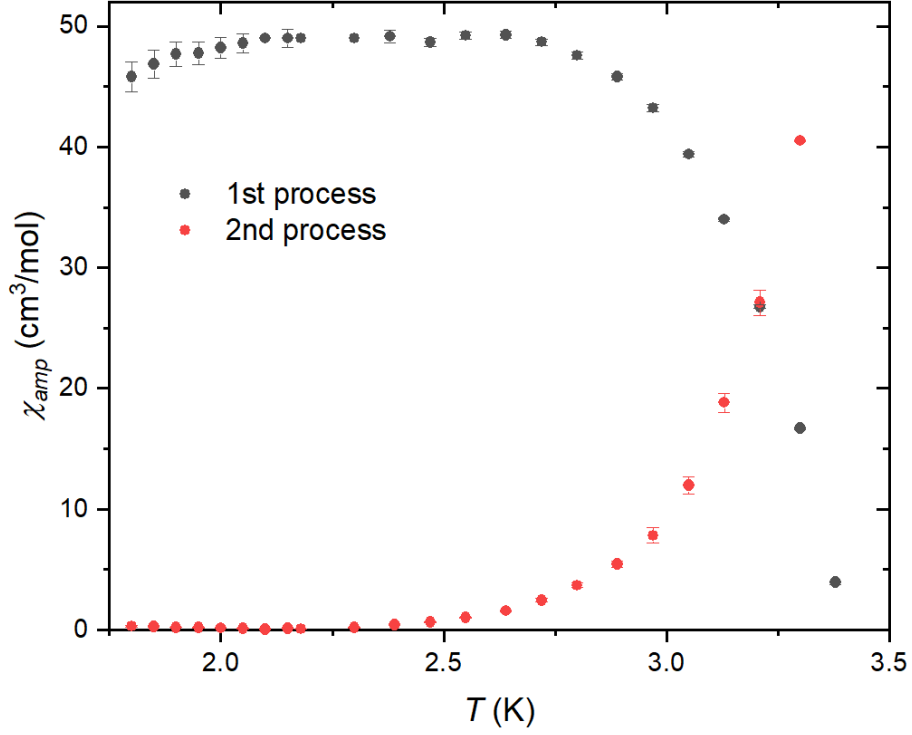


Figure S14. The amplitude of the magnetic susceptibility obtained from the fitting of the Cole-Cole model for both relaxation processes for **1**.

The analysis of the ac magnetic susceptibility was done by using the single-mode Cole-Cole model:

$$\chi_{AC} = \chi_0 + \frac{\chi_1}{1 + (i\omega\tau_1)^{1-\alpha_1}}$$

or double-mode Cole-Cole model:

$$\chi_{AC} = \chi_0 + \frac{\chi_1}{1 + (i\omega\tau_1)^{1-\alpha_1}} + \frac{\chi_2}{1 + (i\omega\tau_2)^{1-\alpha_2}}$$

(χ_0 - the adiabatic limit of the magnetic susceptibility, χ_1 and χ_2 - the amplitudes of the processes, τ - a relaxation time, α - parameter describing the dispersion of the relaxation times).^[S4]

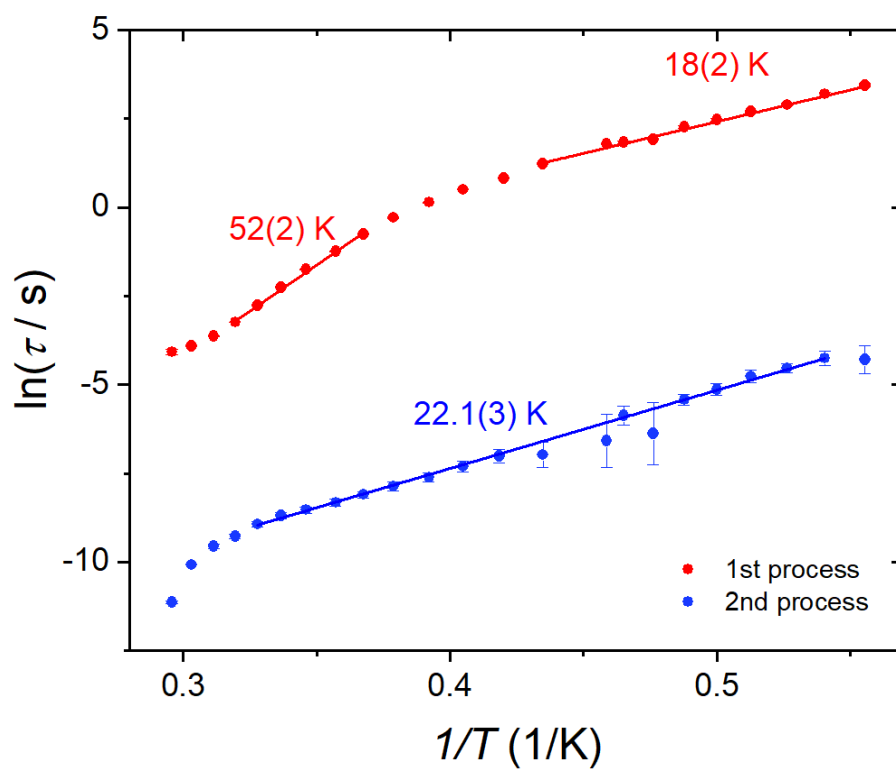


Figure S15. The relaxation times of 1. Red points – SCM relaxation process. Blue points - the faster relaxation process. Lines – the fits of the Arrhenius law.

Table S3. Parameters obtained from fitting the double-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1** at $H_{dc} = 0$ Oe from 1.8 to 3.38 K. The first process, assigned with lower index 1, is the SCM relaxation process, the second process, assigned with lower index 2, is the faster relaxation process (spin wave excitations). (fixed parameters do not have any uncertainty given)

T	χ_0	χ_1	χ_2	τ_1	τ_2	α_1	α_2
K	cm ³ /mol	cm ³ /mol	cm ³ /mol	s	s	-	-
1.8	0.104(35)	45.8(1.3)	0.308(65)	31.4(1.6)	0.0137(54)	0.2221(63)	0.23(17)
1.85	0.054(45)	46.9(1.2)	0.269(16)	24.6(1.1)	0.0141(30)	0.2239(66)	0.32981
1.9	0.05387	47.7(99)	0.205(14)	18.18(73)	0.0107(14)	0.2253(61)	0.272(50)
1.95	0.05387	47.74(95)	0.179(18)	14.87(57)	0.0084(15)	0.2213(66)	0.270(66)
2	0.05387	48.20(86)	0.140(15)	11.91(42)	0.0059(11)	0.2205(68)	0.205(80)
2.05	0.05387	48.58(80)	0.116(12)	9.63(32)	0.00442(70)	0.2209(70)	0.090(91)
2.1	0.046(20)	49	0.060(23)	6.813(34)	0.0017(15)	0.1797(19)	0.03552
2.15	0.05387	49.01(77)	0.117(14)	6.31(21)	0.00283(76)	0.2226(81)	0.03552
2.18	0.042(22)	49	0.075(25)	5.922(27)	0.0014(11)	0.1860(18)	0.03552
2.3	0.035(30)	49	0.204(33)	3.422(13)	9.4(3.4)·10 ⁻⁴	0.1737(28)	0.03552
2.38	0.05387	49.14(52)	-	2.274(56)	-	0.2206(81)	-
2.39	0.046(32)	-	0.392(35)	-	8.9(1.8)·10 ⁻⁴	-	0.03552
2.47	0.054(43)	48.65(36)	0.638(46)	1.664(22)	6.7(1.1)·10 ⁻⁴	0.1852(29)	0.03552
2.55	0.051(59)	49.24(31)	1.033(62)	1.149(13)	4.92(60)·10 ⁻⁴	0.1913(30)	0.03552
2.64	0.076(92)	49.28(30)	1.588(93)	0.7466(79)	3.82(44)·10 ⁻⁴	0.1967(35)	0.03552
2.72	0.11(13)	48.68(26)	2.46(13)	0.4681(44)	3.02(29)·10 ⁻⁴	0.1990(36)	0.03552
2.8	0.18(20)	47.58(28)	3.70(19)	0.2872(31)	2.42(23)·10 ⁻⁴	0.1997(46)	0.03552
2.89	0.35(29)	45.81(29)	5.44(28)	0.1743(21)	1.97(18)·10 ⁻⁴	0.1978(55)	0.03552
2.97	0.82(58)	43.21(29)	7.82(65)	0.1043(12)	1.67(17)·10 ⁻⁴	0.1888(59)	0.036(50)
3.05	1.01(65)	39.38(21)	11.95(72)	0.06258(56)	1.30(11)·10 ⁻⁴	0.1721(47)	0.091(32)
3.13	0.54(72)	33.99(15)	18.83(79)	0.03908(27)	9.33(58)·10 ⁻⁵	0.1516(36)	0.152(19)
3.21	1.4(1.0)	26.76(14)	27.1(1.1)	0.02631(21)	7.01(43)·10 ⁻⁵	0.1393(41)	0.163(15)
3.3	1.40374	16.70(11)	40.526(80)	0.01988(21)	4.189(37)·10 ⁻⁵	0.1384(55)	0.1493(54)
3.38	1.40374	3.92(14)	59.34(10)	0.0170(13)	1.457(44)·10 ⁻⁵	0.245(31)	0.011(13)

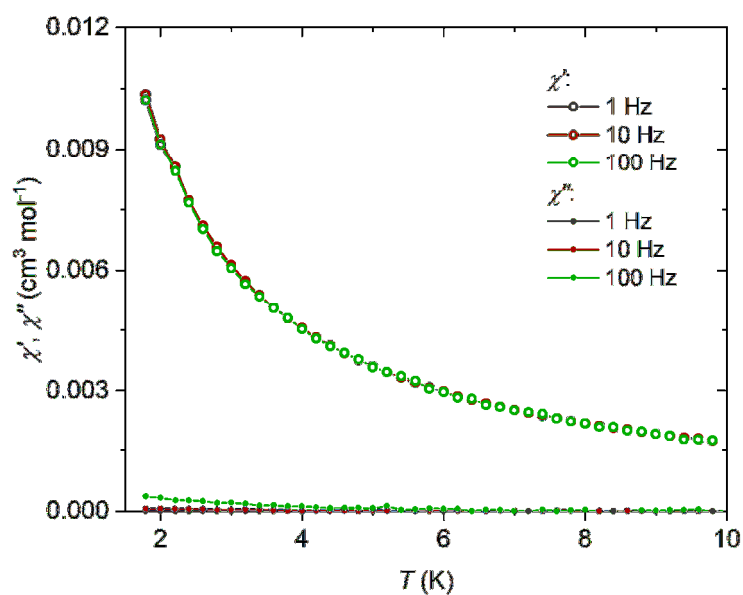


Figure S16. The ac magnetic susceptibility measured for **1-Cd** ($x = 0.013$) at $H_{dc} = 0$ and $H_{ac} = 3$ Oe. No significant difference between the courses of the points indicates good magnetic dilution.

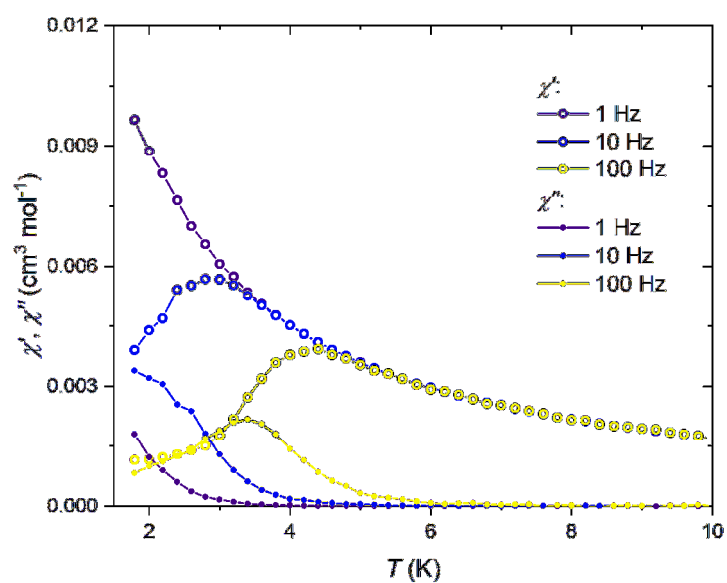


Figure S17. The ac magnetic susceptibility measured for **1-Cd** ($x = 0.013$) at $H_{dc} = 1$ kOe and $H_{ac} = 3$ Oe. The difference for points measured at different frequencies is well visible below 4.5 K.

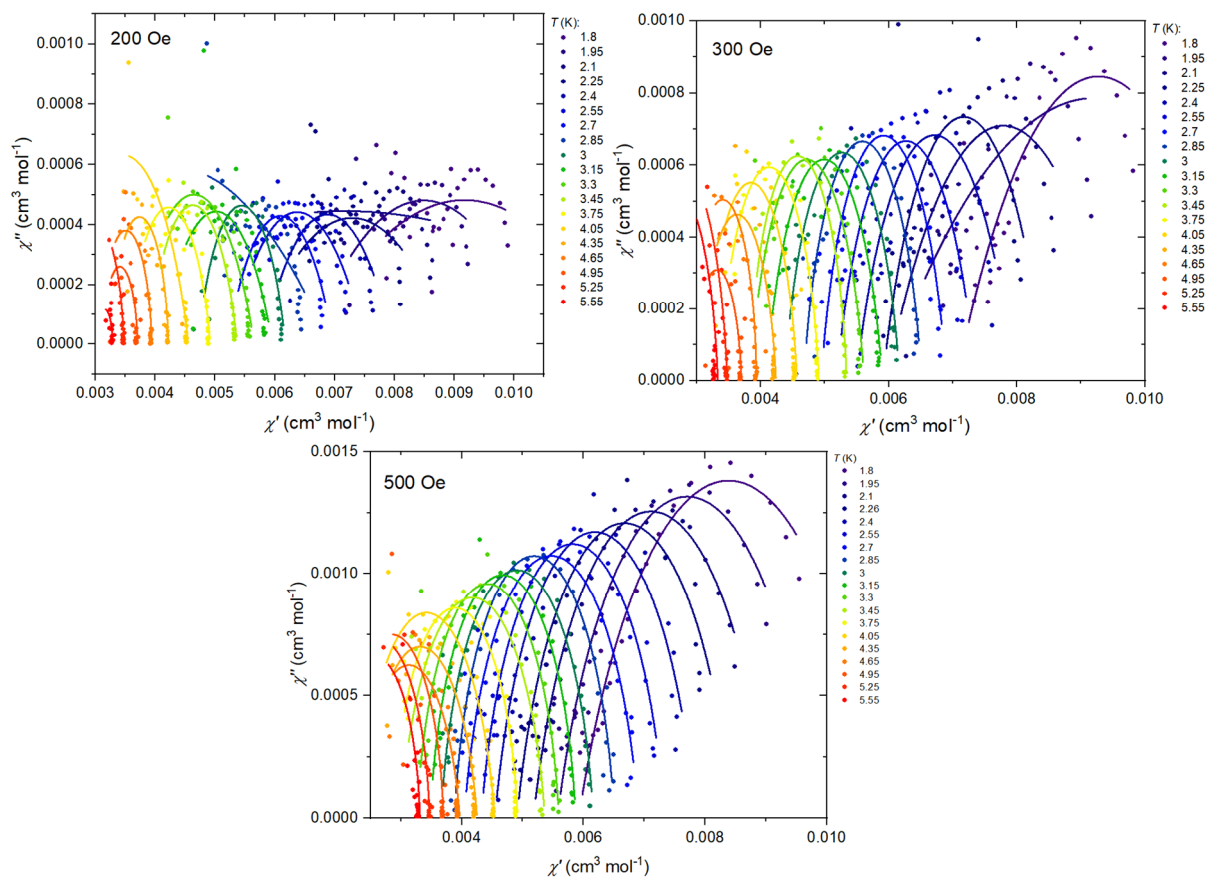


Figure S18. The Argand plots of ac magnetic susceptibility measured at $H_{ac} = 3$ Oe in temperatures below 6 K at 3 different fields (200, 300, 500 Oe) for **1-Cd** ($x = 0.013$). The solid lines are the fits of the single-mode Cole-Cole model.

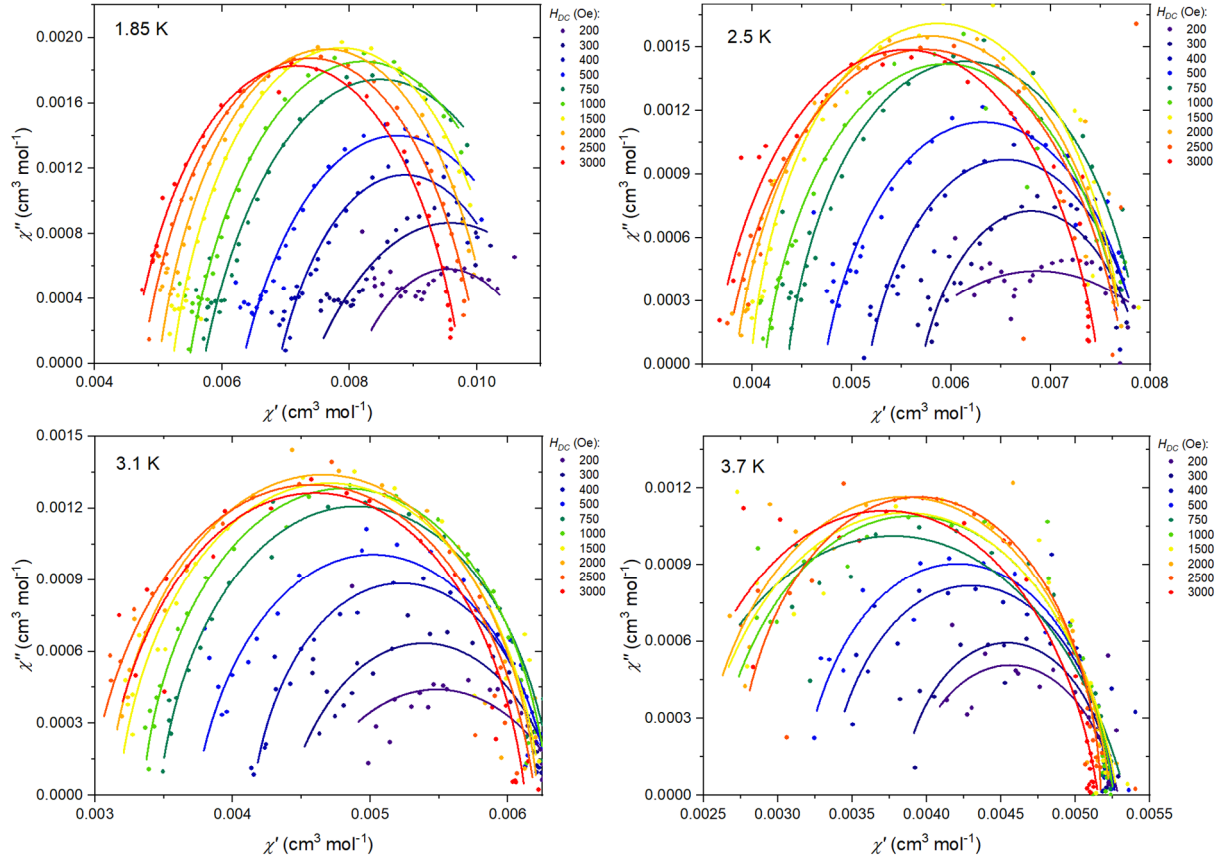


Figure S19. The Argand plot of ac magnetic susceptibility measured at 1.85, 2.5, 3.1 and 3.7 K at $H_{ac} = 3$ Oe and at dc fields up to 3 kOe for **1-Cd** ($x=0.013$). The solid lines are the fits of the single-mode Cole-Cole model.

Table S4. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 1.85 K from 400 to 3000 Oe.

H	χ_0	χ_1	τ_1	α_1
Oe	cm ³ /mol	cm ³ /mol	s	-
400	0.00689(4)	0.00397(16)	0.0373(28)	0.327(23)
500	0.00632(5)	0.00486(25)	0.0458(47)	0.334(27)
750	0.00572(4)	0.00556(23)	0.0566(43)	0.287(21)
1000	0.00548(4)	0.00548(21)	0.0516(33)	0.243(21)
1500	0.00522(5)	0.00531(17)	0.0297(14)	0.197(20)
2000	0.00500(5)	0.00526(12)	0.01406(48)	0.194(17)
2500	0.00475(4)	0.00529(79)	0.00664(16)	0.215(12)
3000	0.00458(4)	0.00517(6)	0.00334(6)	0.2165(84)

Table S5. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 2.5 K from 300 to 3000 Oe.

H	χ_0	χ_1	τ_1	α_1
Oe	cm ³ /mol	cm ³ /mol	s	-
300	0.0057(4)	0.00222(9)	0.01096(72)	0.262(28)
400	0.00517(5)	0.00278(12)	0.01187(79)	0.225(31)
500	0.00472(6)	0.00320(14)	0.01233(81)	0.210(31)
750	0.00435(5)	0.00357(12)	0.01460(70)	0.140(25)
1000	0.00412(5)	0.00365(12)	0.01438(65)	0.159(23)
1500	0.00399(5)	0.00377(12)	0.01174(49)	0.099(24)
2000	0.00383(5)	0.00394(10)	0.00755(26)	0.150(18)
2500	0.00373(12)	0.00402(24)	0.00455(41)	0.189(44)
3000	0.00361(7)	0.00388(12)	0.00231(11)	0.167(23)

Table S6. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 3.1 K from 300 to 3000 Oe.

H	χ_0	χ_1	τ_1	α_1
Oe	cm ³ /mol	cm ³ /mol	s	-
300	0.00440(7)	0.00198(11)	0.00277(24)	0.275(38)
400	0.00415(3)	0.00216(5)	0.00358(12)	0.127(18)
500	0.00373(6)	0.00259(10)	0.00333(19)	0.159(30)
750	0.00347(3)	0.00286(6)	0.00400(10)	0.108(15)
1000	0.00335(3)	0.00295(5)	0.00408(80)	0.088(12)
1500	0.00317(5)	0.00307(10)	0.00371(15)	0.104(23)
2000	0.00309(5)	0.00313(9)	0.00297(11)	0.099(21)
2500	0.00295(7)	0.00324(12)	0.00208(10)	0.141(26)
3000	0.00307(7)	0.00305(10)	0.00149(7)	0.120(24)

Table S7. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 3.7 K from 200 to 3000 Oe.

H	χ_0	χ_1	τ_1	α_1
Oe	cm ³ /mol	cm ³ /mol	s	-
200	0.00383(10)	0.00145(13)	0.00063(9)	0.224(53)
300	0.00382(6)	0.00145(8)	0.00106(8)	0.124(39)
400	0.00333(6)	0.00194(9)	0.00104(7)	0.107(32)
500	0.00316(4)	0.00210(6)	0.00113(4)	0.096(18)
750	0.0022(3)	0.00312(23)	0.00077(13)	0.266(58)
1000	0.00254(8)	0.00271(11)	0.00106(6)	0.138(27)
1500	0.0024(2)	0.00285(17)	0.00104(9)	0.160(39)
2000	0.00246(7)	0.00278(11)	0.00105(5)	0.111(26)
2500	0.00272(6)	0.00245(9)	0.00097(5)	0.033(27)
3000	0.0023(2)	0.00287(15)	0.00060(5)	0.160(31)

Table S8. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 200 Oe from 1.8 to 5.5 K.

T K	χ_0 cm ³ /mol	χ_1 cm ³ /mol	τ_1 10 ⁻⁴ s	α_1 -
1.80	0.00656	0.00523(59)	0.017(17)	0.768(22)
1.95	0.00656(27)	0.00387(75)	0.0153(76)	0.690(59)
2.10	0.00174(95)	0.0109(17)	0.0010(52)	0.90(16)
2.25	0.00566(38)	0.00327(78)	0.0058(27)	0.678(78)
2.40	0.00581(11)	0.00216(21)	0.0063(12)	0.514(48)
2.55	0.00525(12)	0.00225(22)	0.0040(71)	0.523(46)
2.70	0.00521(8)	0.00176(14)	0.0040(55)	0.423(45)
2.85	0.00161(31)	0.005(34)	0.00003(11)	0.719(96)
3.00	0.00472(5)	0.00145(8)	0.00248(23)	0.277(39)
3.15	0.00409(23)	0.00187(31)	0.00093(34)	0.436(81)
3.30	0.00365(15)	0.00201(19)	0.00056(12)	0.412(43)
3.45	0.00389(6)	0.00148(73)	0.00082(7)	0.284(29)
3.75	0.00354(7)	0.00139(9)	0.00056(6)	0.257(35)
4.05	0.00239(38)	0.00217(41)	0.00014(6)	0.329(35)
4.35	0.00326(10)	0.0001(2)	0.00034(5)	0.076(66)
4.65	0.00308(4)	0.00086(5)	0.00028(2)	0.07641
4.95	0.00312(5)	0.00059(7)	0.00031(5)	0.07641
5.25	0.00257(13)	0.0001(2)	0.00010(2)	0.07641
5.55	0.00303(6)	0.00026(7)	0.00025(8)	0.07641

Table S9. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 300 Oe from 1.8 to 5.5 K.

T K	χ_0 cm ³ /mol	χ_1 cm ³ /mol	τ_1 s	α_1 -
1.80	0.00705(11)	0.00445(64)	0.069(30)	0.537(50)
2.10	0.00597(14)	0.00365(54)	0.030(12)	0.528(63)
2.25	0.00591(5)	0.00253(14)	0.0182(18)	0.332(32)
2.40	0.00549(7)	0.00246(19)	0.0138(20)	0.355(47)
2.55	0.00519(6)	0.00217(12)	0.00950(85)	0.299(37)
2.70	0.00494(4)	0.00195(9)	0.00727(51)	0.224(33)
2.85	0.00467(3)	0.00184(6)	0.00526(25)	0.203(23)
3.00	0.00437(3)	0.00180(6)	0.00353(16)	0.217(21)
3.15	0.00409(4)	0.00182(6)	0.00255(14)	0.241(24)
3.30	0.00383(7)	0.00179(10)	0.00173(15)	0.232(38)
3.45	0.00382(4)	0.00153(5)	0.00157(63)	0.127(21)
3.75	0.00336(4)	0.00155(5)	7.88(35)·10 ⁻⁴	0.167(20)
4.05	0.00313(5)	0.00144(7)	5.31(39)·10 ⁻⁴	0.16715
4.35	0.00303(5)	0.00121(6)	3.90(32)·10 ⁻⁴	0.16715
4.65	0.00289(8)	0.00104(9)	2.68(27)·10 ⁻⁴	0.022(40)
4.95	0.00295(7)	0.00008(8)	2.68(40)·10 ⁻⁴	0.1235
5.25	0.00203(42)	0.00145(44)	6.9(31)·10 ⁻⁵	0.124(69)
5.55	0.00203	0.00130(46)	9.1(22)·10 ⁻⁵	0.1235

Table S10. Parameters obtained from fitting the single-mode Cole-Cole model to the ac magnetic susceptibility, χ' and χ'' , vs. frequency, f , dependence measured for **1-Cd** ($x = 0.013$) at 500 Oe from 1.8 to 5.5 K.

T K	χ_0 cm ³ /mol	χ_1 cm ³ /mol	τ_1 s	α_1 -
1.80	0.00592(5)	0.00494(23)	0.0508(49)	0.351(23)
1.95	0.00557(4)	0.00428(17)	0.0375(27)	0.300(22)
2.10	0.00518(4)	0.00387(13)	0.0279(15)	0.267(20)
2.25	0.00491(4)	0.00355(13)	0.0209(13)	0.239(24)
2.40	0.00455(5)	0.00329(14)	0.01517(95)	0.213(29)
2.55	0.00432(5)	0.00301(13)	0.01221(75)	0.185(31)
2.70	0.00404(3)	0.00287(9)	0.00814(39)	0.182(24)
2.85	0.00388(3)	0.00263(5)	0.00630(15)	0.128(13)
3.00	0.00366(5)	0.00250(10)	0.00446(25)	0.133(30)
3.15	0.00348(4)	0.00239(7)	0.00330(13)	0.117(21)
3.30	0.00326(6)	0.00234(10)	0.00234(14)	0.126(33)
3.45	0.00299(6)	0.00237(10)	0.00159(9)	0.173(29)
3.75	0.00287(5)	0.00205(7)	0.00102(4)	0.111(21)
4.05	0.00230(11)	0.00224(14)	0.00047(5)	0.181(36)
4.35	0.00239(6)	0.00187(8)	0.00037(3)	0.18132
4.65	0.00230(9)	0.00167(10)	0.00026(3)	0.18132
4.95	0.00203(4)	0.00167(35)	0.00016(5)	0.070(83)
5.25	0.00184(12)	0.00163(14)	0.00011(2)	0.07026
5.55	0.00184	0.00147(4)	0.00011(2)	0.07026

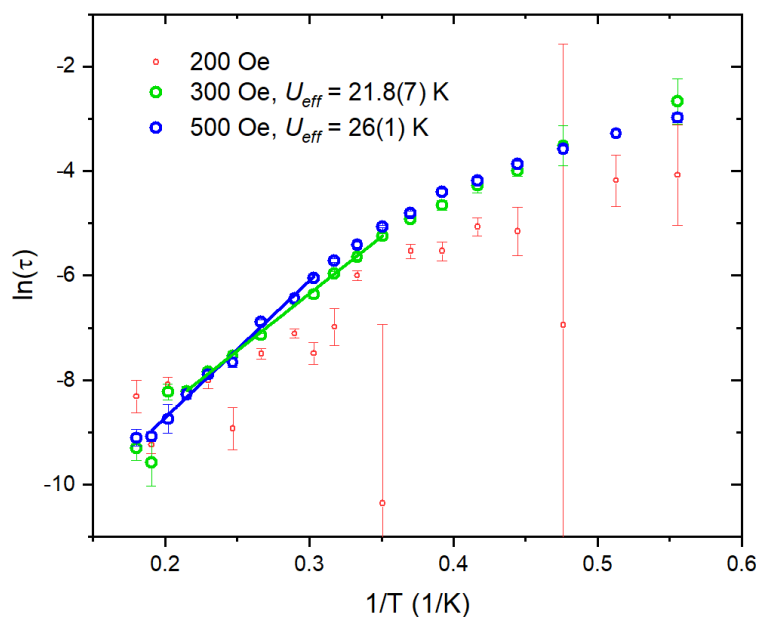


Figure S20. Temperature dependence of the relaxation times for **1-Cd** ($x = 1.3\%$). Solid lines represent a fit of the Arrhenius law describing the Orbach process of relaxation of single spins. The determined effective energy barriers are 21.8(7) K for 300 Oe and 26(1) K for 500 Oe.

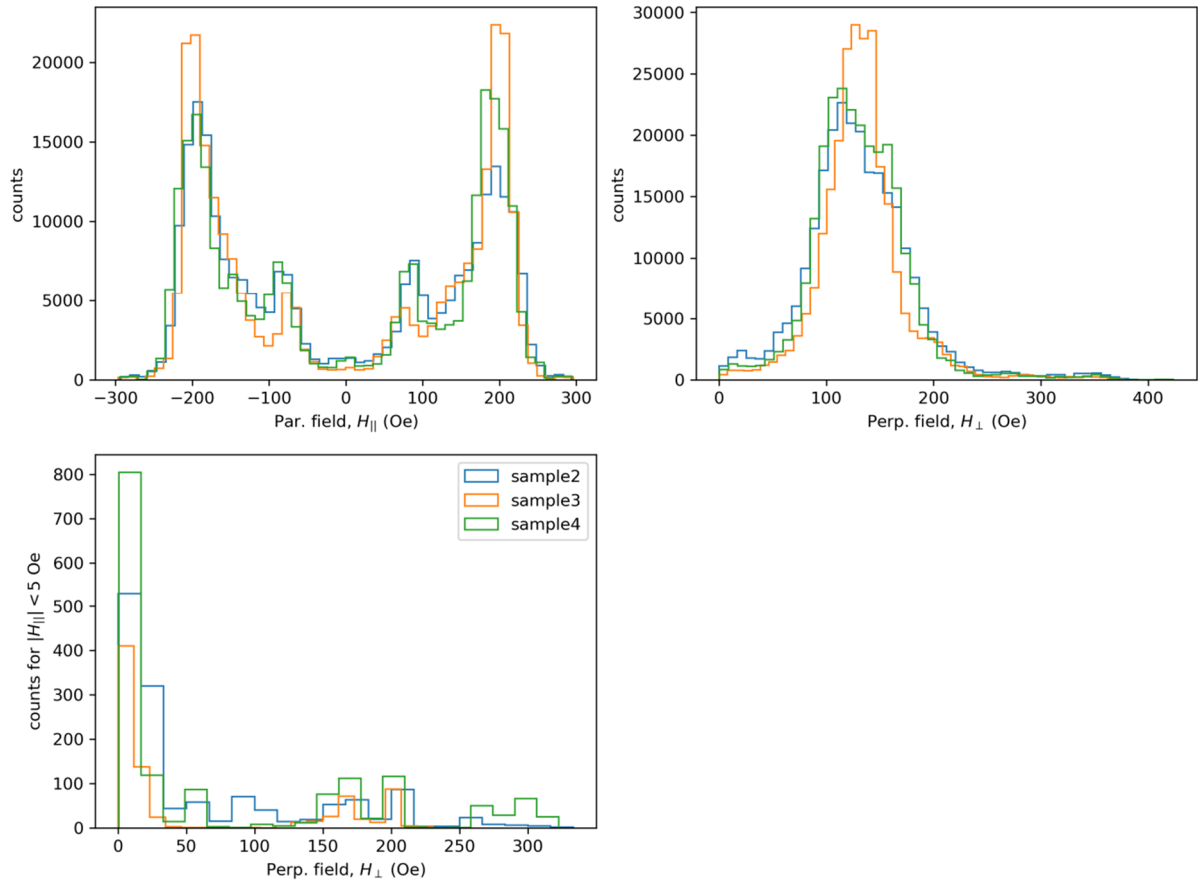


Figure S21. Distribution of dipolar field at Co site obtained from classical Monte-Carlo simulations (see text) for three different domain structures. Top left: Field parallel to easy axis is of order of 200 Oe for most of the spins. Spins at a boundary of the domain experience smaller fields. Top right: Field perpendicular to the easy-axis. Most of spins experience about 120 Oe field. Bottom: The perpendicular field for most of spins that experience a small (< 5 Oe) parallel field is also close to zero.

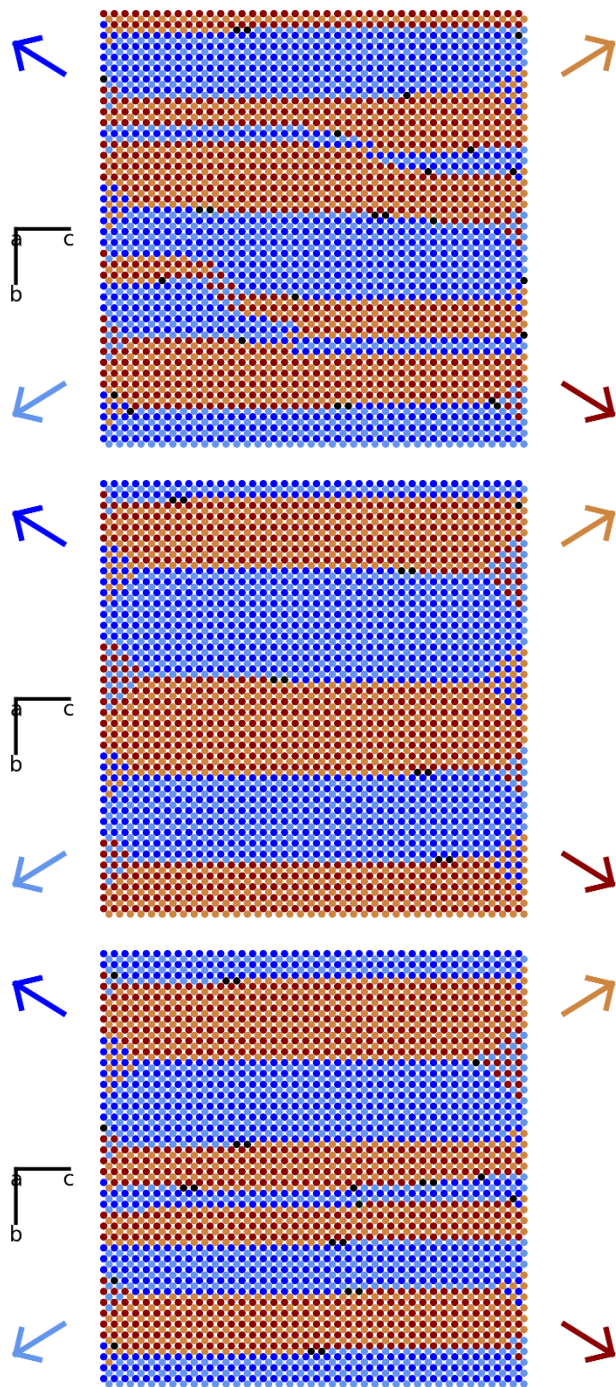


Figure S22. Magnetic structure obtained by Monte Carlo simulations of magnetic moments in **1**. Top: sample 2, middle: sample 3, bottom: sample 4. The view is along the 100 axis, i.e. along the Co chains. Two possible easy axes directions are marked by arrows. Each dot denotes a single chain of spins. The orientation of magnetic moments is marked by colors. Black dots denote chains that reverse by applied field of $H_{ac} < 5$ Oe.

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