

Supporting Information

Experimental and Theoretical Insights into The Co-Ln Magnetic Exchange and The Rare Slow-Magnetic Relaxation Behavior of $[\text{Co}^{\text{II}}_2\text{Pr}]^{2+}$ in a Series of Linear $[\text{Co}^{\text{II}}_2\text{Ln}]^{2+}$ Complexes

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Table S1. Crystallographic parameters for complexes **1-3**

Complex	1	2	3
Formula	$\text{Co}_2\text{LaC}_{60}\text{H}_{56}\text{N}_7\text{O}_{21}$	$\text{Co}_2\text{CeC}_{60}\text{H}_{56}\text{N}_7\text{O}_{23.25}$	$\text{Co}_2\text{PrC}_{60}\text{H}_{56}\text{N}_7\text{O}_{21}$
Size [mm]	0.15 x 0.11 x 0.08	0.21 x 0.13 x 0.02	0.20 x 0.20 x 0.20
System	Triclinic	Triclinic	Triclinic
Space group	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1
<i>a</i> [Å]	12.704(2)	12.691(3)	12.746(3)
<i>b</i> [Å]	13.2508(19)	13.200(3)	13.250(3)
<i>c</i> [Å]	21.871(4)	21.832(5)	21.758(6)
α [°]	85.942(7)	86.095(10)	92.052(11)
β [°]	87.626(7)	87.771(9)	92.012(1)
γ [°]	61.386(4)	61.324(5)	118.729(16)
<i>V</i> [Å ³]	3223.8(9)	3201.2(13)	3214.5(15)
<i>Z</i>	2	2	2
ρ_{calcd} [g/cm ⁻³]	1.565	1.593	1.572
$2\theta_{\text{max}}$	58.38	58.3	58.28
radiation	Mo K_{α}	Mo K_{α}	Mo K_{α}
λ [Å]	0.71070	0.71070	0.71070
<i>T</i> [K]	100	100	100
reflns	50130	43736	55892
Ind. reflns	17267	17092	17178
reflns with $I > 2\sigma(I)$	15459	14301	15342
<i>R</i> 1	0.0473	0.0617	0.0571
<i>wR</i> 2	0.1034	0.1590	0.1205

Table S2. Continuous Shape Measurement (CShM) for Co^{II} sites in **1-3**

Metal Site	HP-6	PPY-6	OC-6	TPR-6	JPPY-6
Complex 1					
Co1	33.345	19.840	3.383	6.049	23.669
Co2	32.880	19.712	3.896	5.587	23.407
Complex 2					
Co1	33.288	19.748	3.435	5.995	23.561
Co2	32.946	19.559	3.990	5.468	23.457
Complex 3					
Co1	33.024	19.500	4.104	5.395	23.383
Co2	33.150	19.657	3.461	6.038	23.452

* HP-6 = Hexagonal planar (D_{6h}), PPY-6 = Pentagonal pyramid (C_{5v}), OC-6 Octahedron (O_h), TPR-6 = Trigonal prism (D_{3h}), JPPY-6 = Johnson pentagonal pyramid J2 (C_{5v})

Table S3. Selected bond lengths, bond angles, dihedral angles, and interatomic distances in complexes **1-3**.

Bonds	1	2	3
Bond Distance (Å)			
Co1-N11	2.054(3)	2.082(4)	2.048(4)
Co1-O11	2.049(2)	2.043(3)	2.046(3)
Co1-O13	2.187(3)	2.236(3)	2.189(3)
Co1-N31	2.084(3)	2.055(4)	2.082(4)
Co1-O31	2.043(3)	2.055(3)	2.048(3)
Co1-O33	2.229(3)	2.188(3)	2.250(3)
Co2-N51	2.097(3)	2.095(4)	2.046(4)
Co2-O51	2.018(3)	2.024(3)	2.066(3)
Co2-O53	2.186(3)	2.187(3)	2.231(3)
Co2-N71	2.046(3)	2.043(4)	2.100(4)
Co2-O71	2.065(2)	2.072(3)	2.023(3)
Co2-O73	2.218(3)	2.220(3)	2.191(3)
Ln1-O11	2.054(3)	2.417(3)	2.564(3)
Ln1-O12	2.590(3)	2.695(3)	2.680(3)
Ln1-O31	2.695(3)	2.567(3)	2.392(3)
Ln1-O32	2.439(2)	2.692(3)	2.676(3)
Ln1-O51	2.717(3)	2.420(3)	2.515(3)
Ln1-O52	2.443(2)	2.667(3)	2.640(3)
Ln1-O71	2.679(3)	2.527(3)	2.401(3)
Ln1-O72	2.552(3)	2.654(3)	2.649(3)
Ln1-O92	2.572(3)	2.552(3)	2.534(4)
Ln1-O93	2.585(3)	2.559(3)	2.555(3)
Bond Angle (°)			
Co1-O11-Ln	105.17(9)	111.04(12)	105.49(18)
Co1-O31-Ln	110.92(9)	105.11(11)	111.82(19)
Co2-O51-Ln	112.27(9)	112.03(13)	106.90(12)
Co2-O71-Ln	106.56(9)	106.43(12)	112.73(19)
Dihedral Angle (°)			
Co1-O11-Ln1-O31	13.03	12.75	12.31
Co2-O51-Ln1-O71	11.47	11.60	11.49
Interplaner twist Angle (°)			

	29.51	29.36	31.53
	Intermetallic separation (Å)		
Co1-Ln1	3.699(4)	3.683(6)	3.689(10)
Co2-Ln1	3.771(4)	3.692(6)	3.682(10)
Co1-Co2	7.388(8)	7.353(12)	7.348(20)

Table S4. The average Bailar twist angle (°) for Co(II) sites in complexes **1-6**.

Metal site	Co ₂ La (1)	Co ₂ Ce (2)	Co ₂ Pr (3)	Co ₂ Gd (4)	Co ₂ Tb (5)	Co ₂ Dy (6)
Co1	32.82	33.74	30.86	33.36	32.12	32.02
Co2	33.31	32.99	31.15	35.45	33.40	29.52

Table S5. The Continuous Shape Measurement (CSHM) for Ln^{III} site in **1-3**

Metal Site	JBCCU-10	JBCSAPR-10	JMBIC-10	JATDI-10	JSPC-10	SDD-10	TD-10	HD-10
La (1)	13.911	4.270	8.428	16.762	3.022	5.217	4.443	11.213
Ce (2)	13.822	4.238	8.517	16.823	3.032	5.221	4.428	11.275
Pr (3)	13.669	4.123	8.503	16.851	3.004	5.263	4.449	11.183

* JBCCU-10 = Bicapped cube J15 (D_{4h}), JBCSAPR-10 = Bicapped square antiprism J17 (D_{4d}), JMBIC-10 = Metabidiminished icosahedron J62 (C_{2v}), JATDI-10 = Augmented tridiminished icosahedron J64 (C_{3v}), JSPC-10 = Sphenocorona J87 (C_{2v}), SDD-10 = Staggered Dodecahedron (2:6:2) (D_2), TD-10 = Tetradecahedron (2:6:2) (C_{2v}), HD-10 = Hexadecahedron (2:6:2) or (1:4:4:1) (D_{4h})

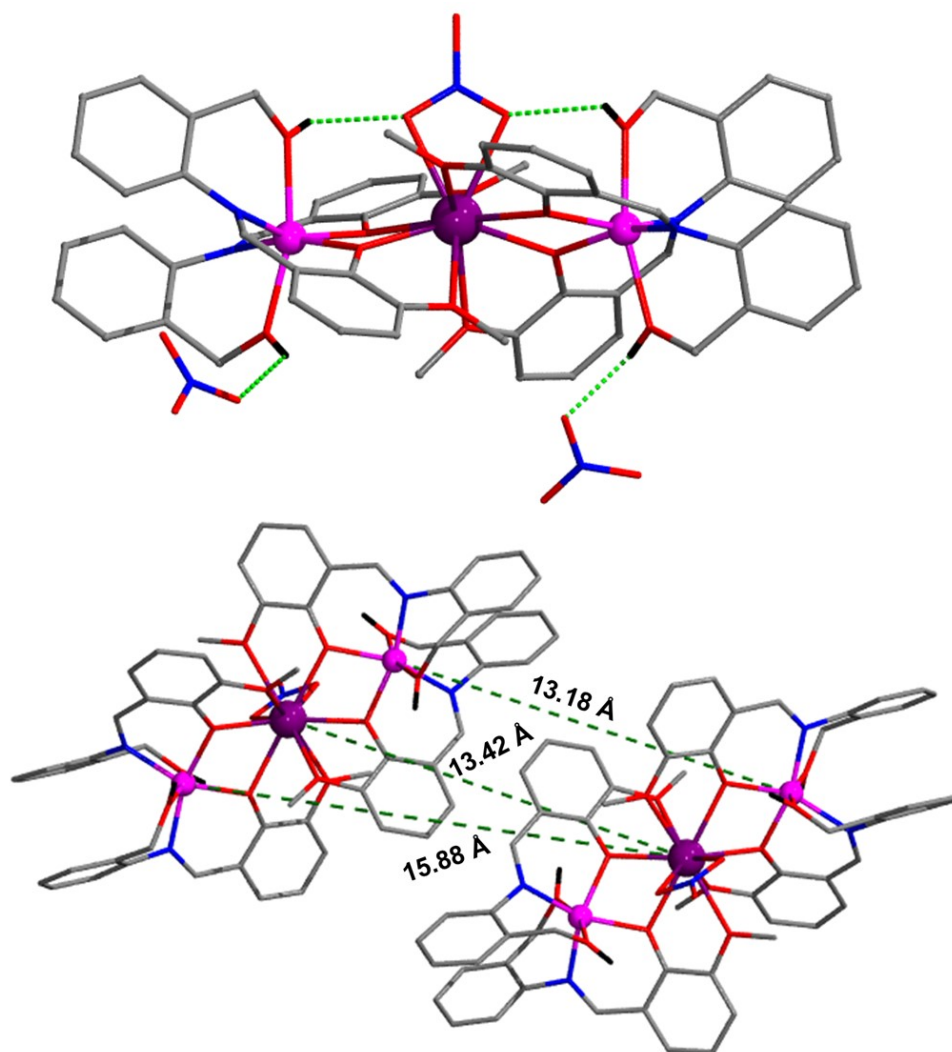


Figure S1. Hydrogen bonding (top) and intermolecular metallic separation (bottom) present in complex **3**.

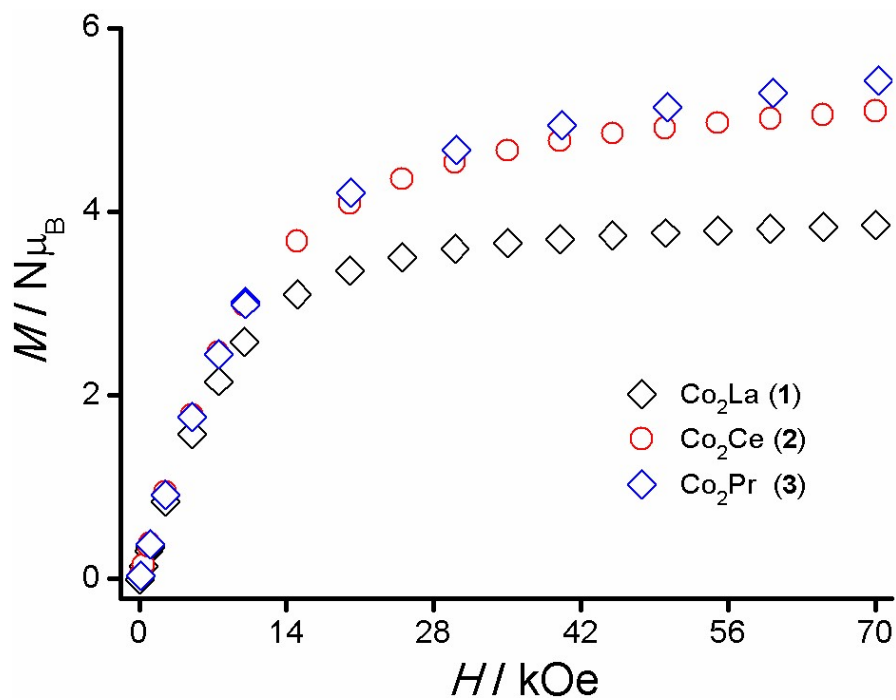


Figure S2. Magnetization plot for complexes 1-3 at 2 K.

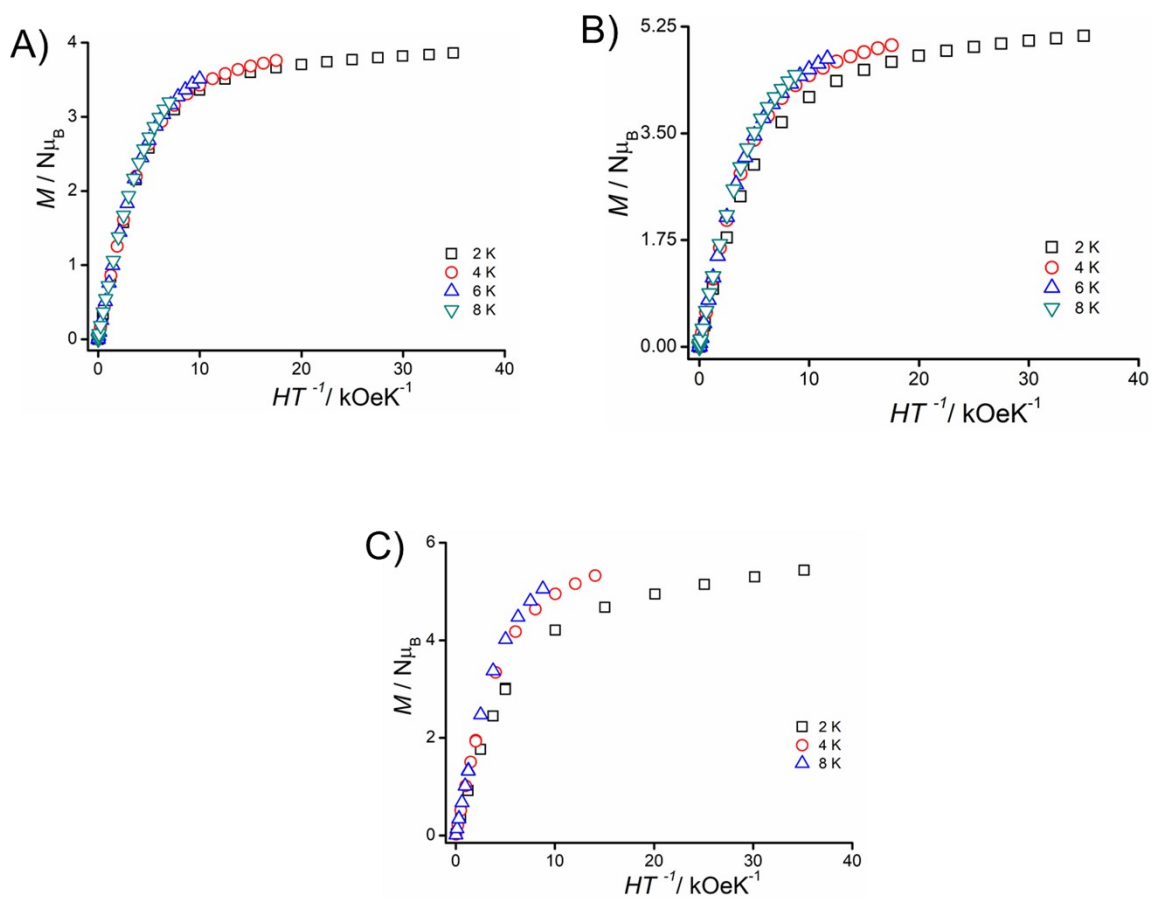


Figure S3. Reduced Magnetization plots for complexes 1-3 at the indicated temperatures.

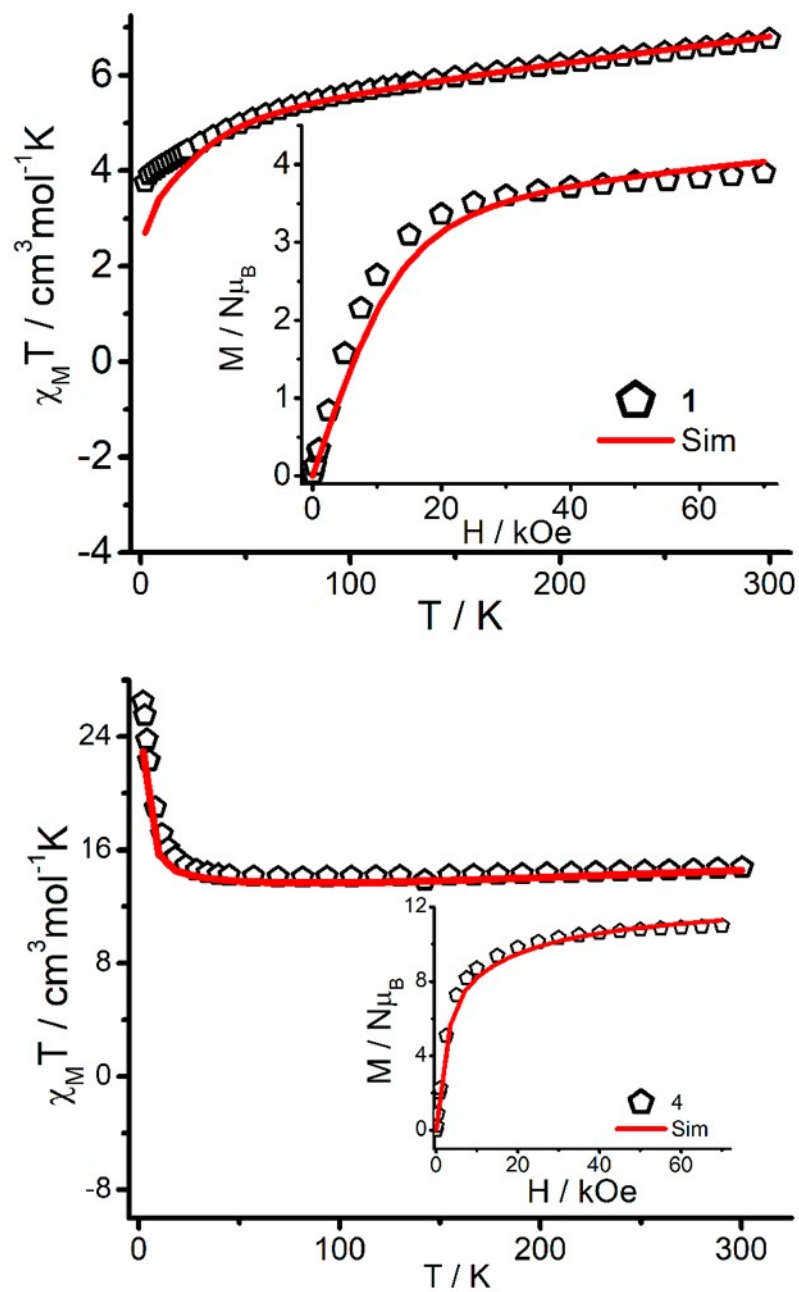


Figure S4. Simulation of dc magnetic data using PHI program for complex **1** (top) and complex **4** (bottom) to extract the SH parameters. Solid lines represent the simulated data.

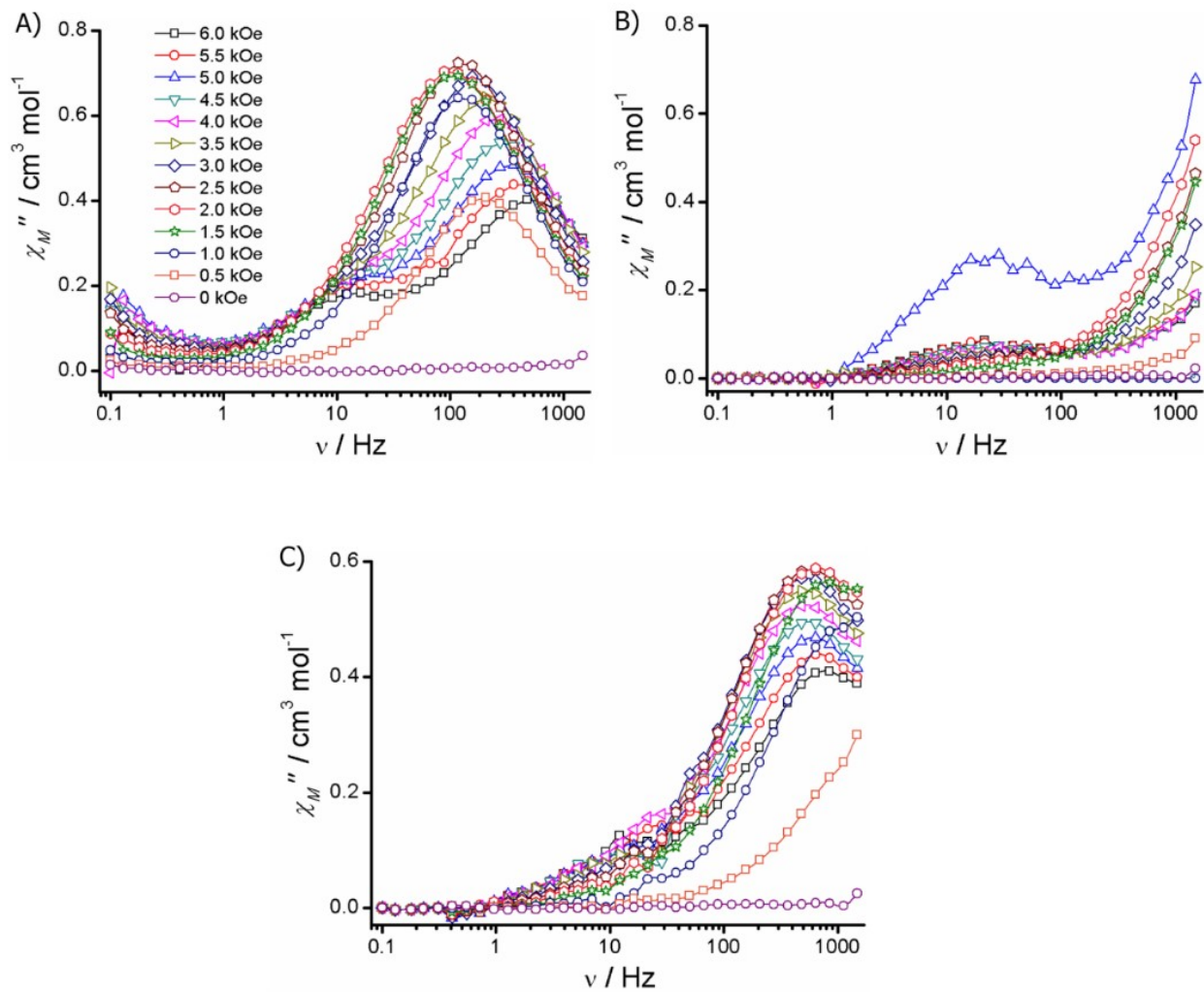


Figure S5. Field dependent ac susceptibility measurement (A) for complex **1**, (B) for **2** and (C) for **3** respectively in the presence of external dc magnetic field 0-6 kOe at 1.8 K.

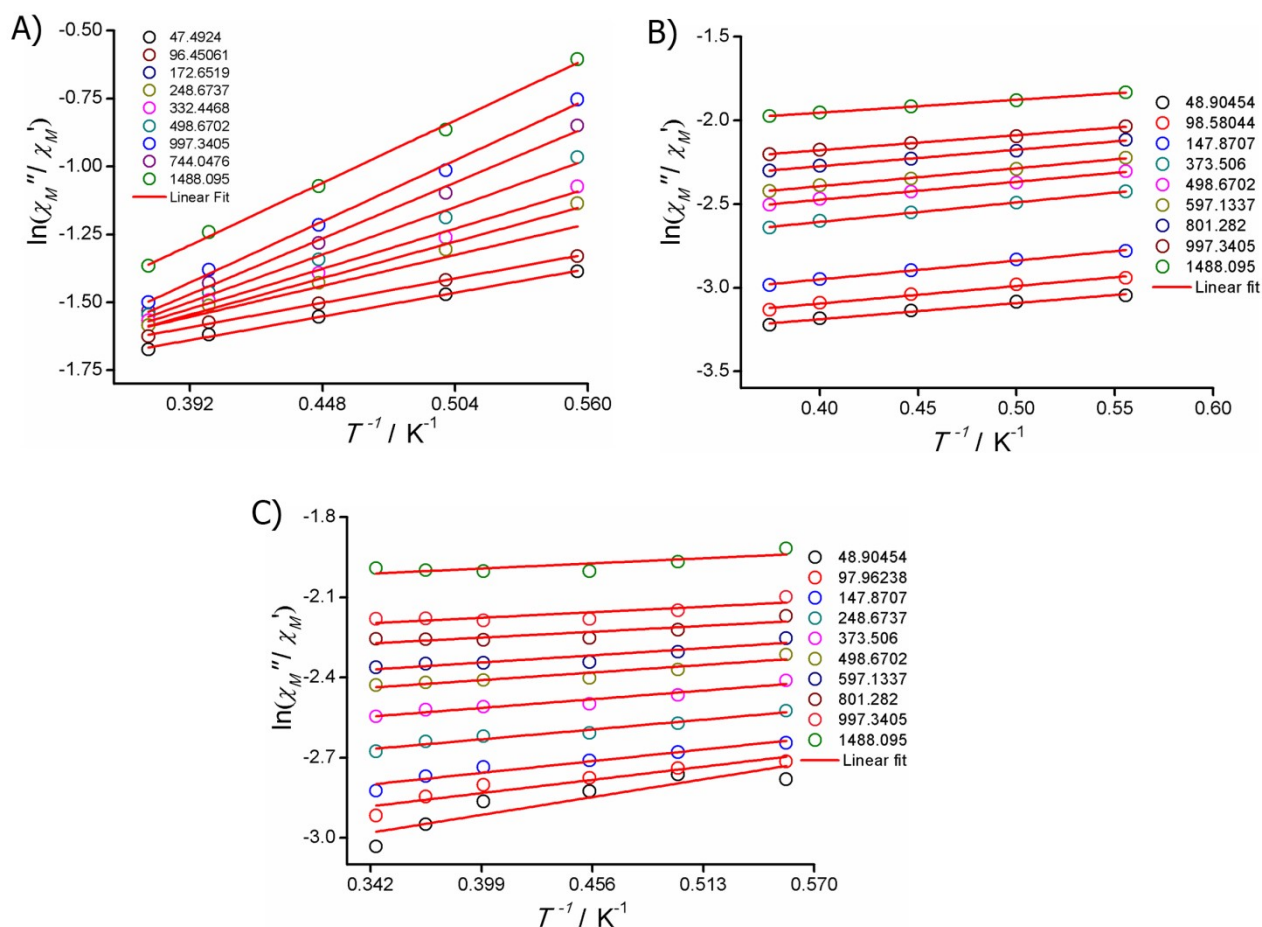


Figure S6. Ac data fit (A-C) using equation $\ln(\chi''/\chi') = \ln(\omega\tau_0) + E_a/k_B T$ to extract the energy barrier for complexes **4-6** respectively.

Computational Details

CASSCF/SO-RASSI/SINGLE_ANISO calculations

During the CASSCF based *ab initio* calculations using MOLCAS 8.0 suit, we have considered the single paramagnetic metal ion at a time by replacing the rest of the centers Co(II) and Ln(III) by their diamagnetic analogue Zn(II) and La(III) respectively. We have taken the active metal fragment Co(II)-La(III)-Zn(II) and Zn(II)-La(III)-Co(II) for Co1 and Co2 ions respectively. The metal fragment Zn(II)-Ln(III)-Zn(II) (where Ln(III) = Ce, Pr, Gd, Tb and Dy) was used to compute the individual lanthanide(III) metal ion for all the complexes **2-6** containing paramagnetic Ln(III) centers. The Spin Hamiltonian (SH) parameters were computed using *ab initio* method. We have employed the basis set [ANO-RCC...3s2p], [ANO-RCC...2s], [ANO-RCC...3s2p1d] for C, H and N or O respectively. The basis sets [ANO-RCC...5s4p2d] and [ANO-RCC...8s7p5d3f2g1h] have been employed for divalent transition and trivalent lanthanide metal ions respectively. The complete active space self-consistent method (CASSCF) was performed to achieve energies of the spin-free

states. The CASSCF were performed by considering the 7 electrons across the five 3d orbitals of Co(II) in all the complexes **1-6**. In case of lanthanide metal ions we have considered the 1 electron across the seven 4f orbitals of Ce (for complex **2**) and 2, 7, 8 and 9 electrons across the seven 4f orbitals of Pr, Gd, Tb and Dy in complexes **3-6** respectively. In the configuration integration procedure for the calculation of roots we computed 10 quartet and 40 doublet excited states for cobalt(II) ions and for lanthanide ions 7 singlet for Ce(III), 21 triplet and 28 singlet for Pr(III), 1 octet, 48 sextet for Gd(III), 7 septet, 140 quintet and 195 triplet excited states for Tb(III) and 21 excited sextet excited states roots for Dy(III) respectively were computed to investigate the anisotropy. The computed 10 quartet and 40 doublet spin states for each Co(II) ion were further mixed by the spin-orbit coupling within the Restricted Active Space State Interaction (RASSI) step to get energies of spin-orbit states for Co1 and Co2 metal fragments in each of the complex. For the lanthanide metal center, due to the hardware limitation only limited number of CASSCF states were mixed by spin orbit coupling in RASSI-SO step. In order to compute the g-tensors and magnetic properties we have performed the SINGLE_ANISO step in which the *ab initio* computed matrix elements of angular momentum and spin-orbit multiplets were used.

POLY_ANISO Simulations: Simulation of dc susceptibility data for complexes **1-6** was done using POLY_ANISO program implemented in MOLCAS 8.0 program. During simulation we have considered the SINGLE_ANISO computed wave functions for each paramagnetic metal ion. To generate the low lying exchange state we have taken the ground and first excited state energy level (spin-orbit state) for each Co(II) and Ln(III) in all the cases except complex **4** for which all the eight spin-orbit states are taken.

BS-DFT Calculation: We have used B3LYP functional with all-electron Alhrich's triple- ζ valence (TZV) ^{1, 2} basis sets implemented in Gaussian 09 program ³ for all the elements. We have estimated the J value from the energy difference of high-spin and broken symmetry states following equation $J = E_{BS} - E_{HS}/S(S + 1)$, where S is the total spin ground state, E_{HS} is the energy of high-spin state and E_{BS} is the energy of broken symmetry state respectively.

Table S6. The CASSCF computed g-tensors along with the axial and transverse magnetic anisotropies for Co(II) ion in complexes **1-6**.

	g_x	g_y	g_z	D / cm ⁻¹	E / cm ⁻¹	E/D
Complex 1						
Co1	1.8706879	2.09459	3.2229492	-108.823	13.722	0.126
Co2	1.9099073	2.140170	3.1918984	-104.508	12.439	0.119
Complex 2						
Co1	1.8698972	2.094142	3.2242057	-109.3225	13.574	0.124
Co2	1.9194293	2.145748	3.1883677	-103.5894	12.016	0.115
Complex 3						
Co1	1.8962470	2.123592	3.2022004	-105.630	13.158	0.124
Co2	1.9099929	2.137717	3.1943319	-105.793	11.980	0.113
Complex 4						
Co1	1.593473	2.135621	3.4360036	-122.624	11.455	0.093
Co2	1.7591667	2.276640	3.2817568	-107.950	15.117	0.140
Complex 5						
Co1	1.8874935	2.111067	3.2334645	-110.3753	11.323	0.102
Co2	1.8883271	2.112788	3.2151164	-107.5369	12.456	0.115
Complex 6						
Co1	1.8793162	2.097995	3.2533810	-110.456	11.699	0.105
Co2	1.8857695	2.098968	3.2207568	-110.464	11.697	0.105

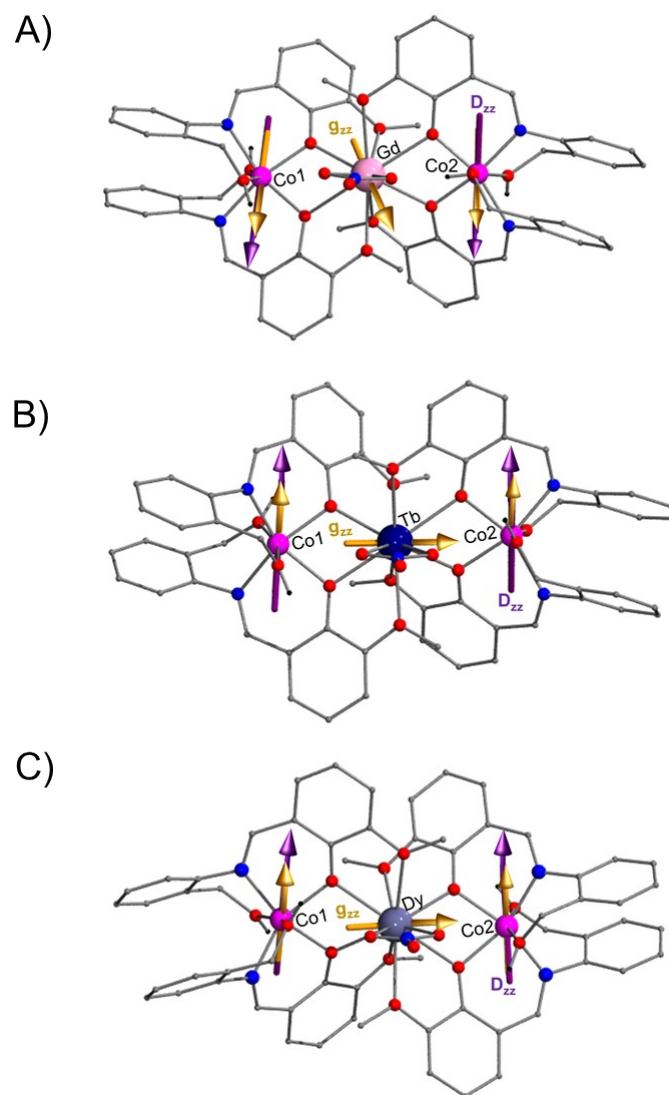


Figure S7. The orientation of main magnetization axes g_{zz} (yellow arrow) on each metal ion in complexes **4-6** (A-C). The local magnetic anisotropy D_{zz} on each Co(II) centre. shown in purple arrow.

Table S6. Energy / cm^{-1} (shown in bold) of the low lying doublets and g -tensors on individual magnetic centers in complexes **1-6**.

Multiplets	1	2	3	4	5	6	7	8
Complex 1								
Co1	0.000	222.778	636.141	943.127	2649.686	2749.038	5633.475	5729.712
	$g_x = 0.7558$	$g_x = 4.2122$	$g_x = 3.5415$	$g_x = 0.2369$	$g_x = 0.5711$	$g_x = 4.1317$	$g_x = 0.2591$	$g_x = 3.6249$
	$g_y = 0.8818$	$g_y = 3.6003$	$g_y = 2.8192$	$g_y = -0.2947$	$g_y = 0.6908$	$g_y = 3.2081$	$g_y = 0.2891$	$g_y = 3.1685$
	$g_z = 9.0551$	$g_z = 2.5780$	$g_z = 0.25103$	$g_z = 3.0399$	$g_z = 6.8468$	$g_z = 2.2260$	$g_z = 6.4595$	$g_z = 2.1426$
	0.000	213.412	693.026	988.307	2815.170	2915.659	5855.137	5950.701

Co2	$g_x = 0.70193$ $g_y = 0.8277$ $g_z = 9.0675$	$g_x = 4.0296$ $g_y = 3.8922$ $g_z = 2.9488$	$g_x = 3.7361$ $g_y = 3.2880$ $g_z = 0.0555$	$g_x = 0.1327$ $g_y = 0.1793$ $g_z = 3.0119$	$g_x = 0.6188$ $g_y = 0.7580$ $g_z = 6.9182$	$g_x = 2.2367$ $g_y = 3.1922$ $g_z = 4.2305$	$g_x = 0.2921$ $g_y = 0.3243$ $g_z = 6.4776$	$g_x = 3.6815$ $g_y = 3.1584$ $g_z = 2.1463$
Complex 2								
Co1	0.000 $g_x = 0.7439$ $g_y = 0.8686$ $g_z = 9.0620$	223.644 $g_x = 4.2215$ $g_y = 3.5889$ $g_z = 2.5850$	638.133 $g_x = 3.5205$ $g_y = 2.8325$ $g_z = 0.2561$	945.961 $g_x = 0.2222$ $g_y = 0.2801$ $g_z = 3.0346$	2664.566 $g_x = 0.6041$ $g_y = 0.7402$ $g_z = 6.8565$	2764.808 $g_x = 2.2208$ $g_y = 3.1720$ $g_z = 4.1644$	5630.657 $g_x = 0.2705$ $g_y = 0.3029$ $g_z = 6.4647$	5727.575 $g_x = 3.6304$ $g_y = 3.15329$ $g_z = 2.1432$
Ce	0.000 $g_x = 0.12096$ $g_y = 0.18604$ $g_z = 4.04830$	341.867 $g_x = 1.0017$ $g_y = 1.3273$ $g_z = 3.3897$	570.571 $g_x = 0.1564$ $g_y = 0.9460$ $g_z = 3.5204$	2324.326 $g_x = 0.54962$ $g_y = 0.6126$ $g_z = 7.4403$	2585.06 $g_x = 3.0366$ $g_y = 2.6983$ $g_z = 0.1346$	2800.379 $g_x = 0.6663$ $g_y = 1.0518$ $g_z = 6.7557$	3351.106 $g_x = 1.8783$ $g_y = 3.0401$ $g_z = 5.4688$	-----
Co2	0.000 $g_x = 0.68321$ $g_y = 0.80572$ $g_z = 9.08193$	211.319 $g_x = 4.01525$ $g_y = 3.9270$ $g_z = 3.02295$	709.243 $g_x = 3.7810$ $g_y = 3.3838$ $g_z = 0.0157$	1001.769 $g_x = 0.1152$ $g_y = 0.1599$ $g_z = 2.9994$	2865.111 $g_x = 0.6515$ $g_y = 0.8077$ $g_z = 6.9335$	2966.171 $g_x = 2.2327$ $g_y = 3.1628$ $g_z = 4.2735$	5886.665 $g_x = 0.3012$ $g_y = 0.3347$ $g_z = 6.4835$	5982.548 $g_x = 3.69241$ $g_y = 3.15056$ $g_z = 2.14760$
Complex 3								
Co1	0.000 $g_x = 0.7403$ $g_y = 0.8726$ $g_z = 9.0506$	216.122 $g_x = 4.0861$ $g_y = 3.8079$ $g_z = 2.7843$	663.578 $g_x = 3.7722$ $g_y = 3.0318$ $g_z = 0.1253$	963.366 $g_x = 0.2392$ $g_y = 0.2981$ $g_z = 3.0443$	2692.182 $g_x = 0.6639$ $g_y = 0.8319$ $g_z = 6.8413$	2793.262 $g_x = 2.1998$ $g_y = 3.0988$ $g_z = 4.2133$	5605.578 $g_x = 0.2973$ $g_y = 0.3372$ $g_z = 6.4497$	5704.063 $g_x = 3.6348$ $g_y = 3.1128$ $g_z = 2.1353$
Pr	0.000 / 13.973 $g_x = 0.00$ $g_y = 0.00$ $g_z = 6.1259$	127.363 / 134.187 $g_x = 0.00$ $g_y = 0.00$ $g_z = 4.7051$	188.457 / 261.552 $g_x = 0.00$ $g_y = 0.00$ $g_z = 3.4526$	474.205 / 30.046 $g_x = 0.00$ $g_y = 0.00$ $g_z = 3.5379$	632.820 / 2245.123 $g_x = 0.00$ $g_y = 0.00$ $g_z = 0.1289$	2246.069 / 2354.609 $g_x = 0.00$ $g_y = 0.00$ $g_z = 2.7912$	2366.303 / 2440.288 $g_x = 0.00$ $g_y = 0.00$ $g_z = 2.3600$	2444.904 / 2661.451 $g_x = 0.00$ $g_y = 0.00$ $g_z = 4.9375$
Co2	0.000 $g_x = 0.6674$ $g_y = 0.7873$ $g_z = 9.0863$	215.619 $g_x = 4.0513$ $g_y = 3.8664$ $g_z = 2.9767$	699.781 $g_x = 3.6993$ $g_y = 3.3261$ $g_z = 0.0630$	996.726 $g_x = 0.1014$ $g_y = 0.1473$ $g_z = 3.0021$	2888.743 $g_x = 0.7071$ $g_y = 0.8937$ $g_z = 6.9454$	2991.259 $g_x = 2.2209$ $g_y = 3.1015$ $g_z = 4.3254$	5812.216 $g_x = 0.3277$ $g_y = 0.3697$ $g_z = 6.4905$	5910.573 $g_x = 3.6905$ $g_y = 3.1037$ $g_z = 2.1466$
Complex 4								
Co1	0.000 $g_x = 0.5129$ $g_y = 0.7069$ $g_z = 9.4230$	248.439 $g_x = 1.3028$ $g_y = 1.9508$ $g_z = 5.1578$	574.944 $g_x = 0.7653$ $g_y = 1.4449$ $g_z = 2.3709$	904.958 $g_x = 0.3241$ $g_y = 0.4756$ $g_z = 2.9872$	2357.486 $g_x = 0.6741$ $g_y = 1.0288$ $g_z = 7.7563$	2551.142 $g_x = 2.7023$ $g_y = 2.9972$ $g_z =$ 3.63464	3342.944 $g_x = 0.38102$ $g_y = 1.2175$ $g_z = 5.5778$	3533.176 $g_x = 1.5977$ $g_y = 1.6160$ $g_z = 2.9200$
Gd	0.000 $g_x = 0.00162$ $g_y = 0.00173$ $g_z = 13.9919$	51.869 $g_x = 0.16679$ $g_y = 0.17876$ $g_z = 9.95149$	85.863 $g_x = 3.4501$ $g_y = 3.6250$ $g_z = 5.5553$	105.616 $g_x = 1.5955$ $g_y = 4.2049$ $g_z = 11.2924$	40159.486 $g_x = 0.00704$ $g_y = 0.01753$ $g_z = 11.8093$	40215.601 $g_x = 1.4404$ $g_y = 1.6935$ $g_z = 7.4087$	40255.852 $g_x = 1.86191$ $g_y = 2.43291$ $g_z = 7.89552$	40269.830 $g_x = 0.8552$ $g_y = 1.6585$ $g_z = 8.2719$
	0.000	222.161	639.316	939.238	2316.935	2488.816	3447.845	3619.547

Co2	$g_x = 0.7390$ $g_y = 1.0451$ $g_z = 9.1543$	$g_x = 4.3147$ $g_y = 3.5591$ $g_z = 2.5015$	$g_x = 4.0791$ $g_y = 2.3745$ $g_z = 0.3068$	$g_x = 0.4936$ $g_y = 0.6456$ $g_z = 3.1882$	$g_x = 0.7796$ $g_y = 1.2170$ $g_z = 7.3829$	$g_x = 2.4900$ $g_y = 2.7933$ $g_z = 3.8059$	$g_x = 0.4732$ $g_y = 1.0616$ $g_z = 5.8067$	$g_x = 1.7428$ $g_y = 1.8605$ $g_z = 3.0666$
Complex 5								
Co1	0.000 $g_x = 0.6054$ $g_y = 0.7104$ $g_z = 9.1804$	224.20 $g_x = 4.1955$ $g_y = 3.6977$ $g_z = 2.9127$	689.431 $g_x = 3.5240$ $g_y = 3.2442$ $g_z = 0.2053$	993.181 $g_x = 0.0646$ $g_y = 0.1069$ $g_z = 2.9160$	3030.785 $g_x = 0.7916$ $g_y = 1.0304$ $g_z = 6.9609$	3134.319 $g_x = 2.1991$ $g_y = 3.0175$ $g_z = 4.4261$	5854.584 $g_x = 0.3739$ $g_y = 0.4309$ $g_z = 6.4884$	5955.116 $g_x = 3.7112$ $g_y = 3.0391$ $g_z = 2.1393$
Tb	0.000/0.001 $g_x = 0.00$ $g_y = 0.000$ $g_z = 17.932$	180.40/180.1 $g_x = 0.00$ $g_y = 0.00$ $g_z = 14.7431$	336.1/339.1 $g_x = 0.00$ $g_y = 0.00$ $g_z = 11.51$	409.8/428.4 $g_x = 0.00$ $g_y = 0.00$ $g_z = 12.027$	465.5/482.1 $g_x = 0.00$ $g_y = 0.00$ $g_z = 5.109$	504.7 / 641.7 $g_x = 0.00$ $g_y = 0.000$ $g_z = 6.270$	643.7 / 2263.0 $g_x = 0.00$ $g_y = 0.000$ $g_z = 0.505$	2266.6 / 2321.9 $g_x = 0.000$ $g_y = 0.00$ $g_z = 3.131$
Co2	0.000 $g_x = 0.68804$ $g_y = 0.8095$ $g_z = 9.0957$	219.35 $g_x = 4.14730$ $g_y = 3.73335$ $g_z = 2.7954$	665.453 $g_x = 3.67936$ $g_y = 3.03706$ $g_z = 0.17386$	967.261 $g_x = 0.1991$ $g_y = 0.2533$ $g_z = 3.0070$	2812.467 $g_x = 0.7206$ $g_y = 0.9215$ $g_z = 6.87752$	2914.325 $g_x = 2.1950$ $g_y = 3.0501$ $g_z = 4.2931$	5627.131 $g_x = 0.3363$ $g_y = 0.3870$ $g_z = 6.4485$	5727.162 $g_x = 3.6572$ $g_y = 3.0642$ $g_z = 2.1299$
Complex 6								
Co1	0.000 $g_x = 0.6309$ $g_y = 0.7378$ $g_z = 9.2041$	224.600 $g_x = 4.2602$ $g_y = 3.6395$ $g_z = 2.8004$	669.699 $g_x = 3.5794$ $g_y = 3.0464$ $g_z = 0.2784$	976.154 $g_x = 0.1609$ $g_y = 0.2114$ $g_z = 2.8990$	2900.543 $g_x = 0.7492$ $g_y = 0.9636$ $g_z = 6.9345$	3004.065 $g_x = 2.2049$ $g_y = 3.0461$ $g_z = 4.3499$	5691.052 $g_x = 0.3471$ $g_y = 0.4010$ $g_z = 6.4651$	5792.687 $g_x = 3.6635$ $g_y = 3.0538$ $g_z = 2.1349$
Dy	0.000 $g_x = 0.0021$ $g_y = 0.0029$ $g_z = 19.730$	135.660 $g_x = 0.0015$ $g_y = 0.0122$ $g_z = 17.877$	209.172 $g_x = 0.2246$ $g_y = 0.3401$ $g_z = 17.8297$	236.626 $g_x = 0.4845$ $g_y = 1.1504$ $g_z = 11.417$	350.012 $g_x = 0.0633$ $g_y = 1.5211$ $g_z = 9.5477$	442.112 $g_x = 8.0321$ $g_y =$ 6.17961 $g_z = 3.1159$	511.404 $g_x = 1.85125$ $g_y = 3.9588$ $g_z = 9.62535$	586.325 $g_x = 0.03843$ $g_y = 0.73351$ $g_z = 15.9127$
Co2	0.000 $g_x = 0.6326$ $g_y = 0.7382$ $g_z = 9.1204$	224.615 $g_x = 4.2039$ $g_y = 3.6474$ $g_z = 2.8018$	669.655 $g_x = 3.5747$ $g_y = 3.0499$ $g_z = 0.2104$	976.125 $g_x = 0.1600$ $g_y = 0.2118$ $g_z = 2.9908$	2900.488 $g_x = 0.7500$ $g_y = 0.9641$ $g_z = 6.9371$	3004.0 $g_x = 2.2058$ $g_y =$ 3.04804 $g_z =$ 4.35382	5690.869 $g_x = 0.34752$ $g_y = 0.40050$ $g_z = 6.4855$	5792.507 $g_x = 3.6675$ $g_y = 3.0500$ $g_z =$ 2.14138

Table 3. SO-RASSI computed low lying energies (cm^{-1}) of spin-orbit states of the individual magnetic centers in complexes **1-3**.

Complex 1		Complex 2			Complex 3		
Co1	Co2	Co1	Ce	Co2	Co1	Pr	Co2
0	0	0	0	0	0	0	0
0	0	0	0	0	0	13.973	0
222.778	213.412	223.644	341.867	211.319	216.122	127.363	215.619
222.778	213.412	223.644	341.867	211.319	216.122	134.187	215.619
636.141	693.026	638.133	570.571	709.243	663.578	188.457	699.781
636.141	693.026	638.133	570.571	709.243	663.578	261.552	699.781
943.127	988.307	945.961	2324.32	1001.76	963.366	474.205	996.726
943.127	988.307	945.961	2324.32	1001.76	963.366	630.046	996.726
2649.686	2815.17	2664.56	2585.06	2865.11	2692.182	632.82	2888.743
2649.686	2815.17	2664.56	2585.06	2865.11	2692.182	2245.123	2888.743
2749.038	2915.659	2764.80	2800.37	2966.17	2793.262	2246.069	2991.259
2749.038	2915.659	2764.80	2800.37	2966.17	2793.262	2354.609	2991.259
5633.475	5855.137	5630.65	3351.10	2966.17	5605.578	2366.303	5812.216
5633.475	5855.137	5630.65	3351.10	2966.17	5605.578	2440.288	5812.216
5729.712	5950.701	5727.57		5886.66	5704.063	2444.904	5910.573
5729.712	5950.701	5727.57		5886.66	5704.063	2661.451	5910.573

Table 4. SO-RASSI computed low lying energies (cm^{-1}) of spin-orbit states of the individual magnetic centers in complexes **4-6**.

Complex 4			Complex 5			Complex 6		
Co1	Gd	Co2	Co1	Tb	Co2	Co1	Dy	Co2
0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0
248.439	0.463	222.161	224.208	180.403	219.359	224.6	135.66	224.615
248.439	0.463	222.161	224.208	180.403	219.359	224.6	135.66	224.615
574.944	0.762	639.316	689.431	336.110	665.453	669.69	209.17	669.655
574.944	0.762	639.316	689.431	336.110	665.453	669.69	209.17	669.655
904.958	1.11	939.238	993.181	409.874	967.261	976.15	236.62	976.125
904.958	1.11	939.238	993.181	409.874	967.261	976.15	236.62	976.125
2357.486	40664.466	2316.935	3030.785	465.548	2812.46	2900.54	350.01	2900.488
2357.486	40664.466	2316.935	3030.785	465.548	2812.46	2900.54	350.01	2900.488
2551.142	40707.042	2488.816	3134.319	504.747	2914.32	3004.06	442.11	3004.013
2551.142	40707.042	2488.816	3134.319	504.747	2914.32	3004.06	442.11	3004.013
3342.944	40707.042	3447.845	5854.584	641.768	5627.13	5691.05	511.40	5690.869
3342.944	40707.042	3447.845	5854.584	641.768	5627.13	5691.05	511.40	5690.869
3533.176	40739.314	3619.547	5955.116	2263.084	5727.16	5792.68	586.32	5792.507
3533.176	40739.314	3619.547	5955.116	2263.084	5727.16	5792.68	586.32	5792.507

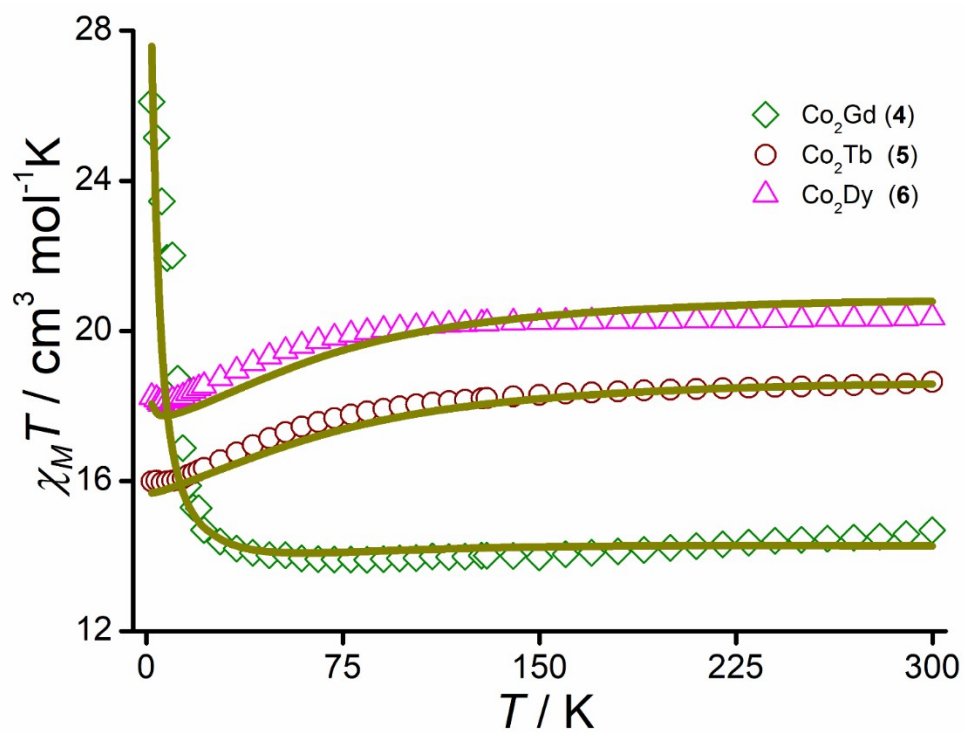


Figure S8. Direct current magnetic susceptibility data for complexes 4-6. Solid lines represent the POLY_ANISO simulated data.

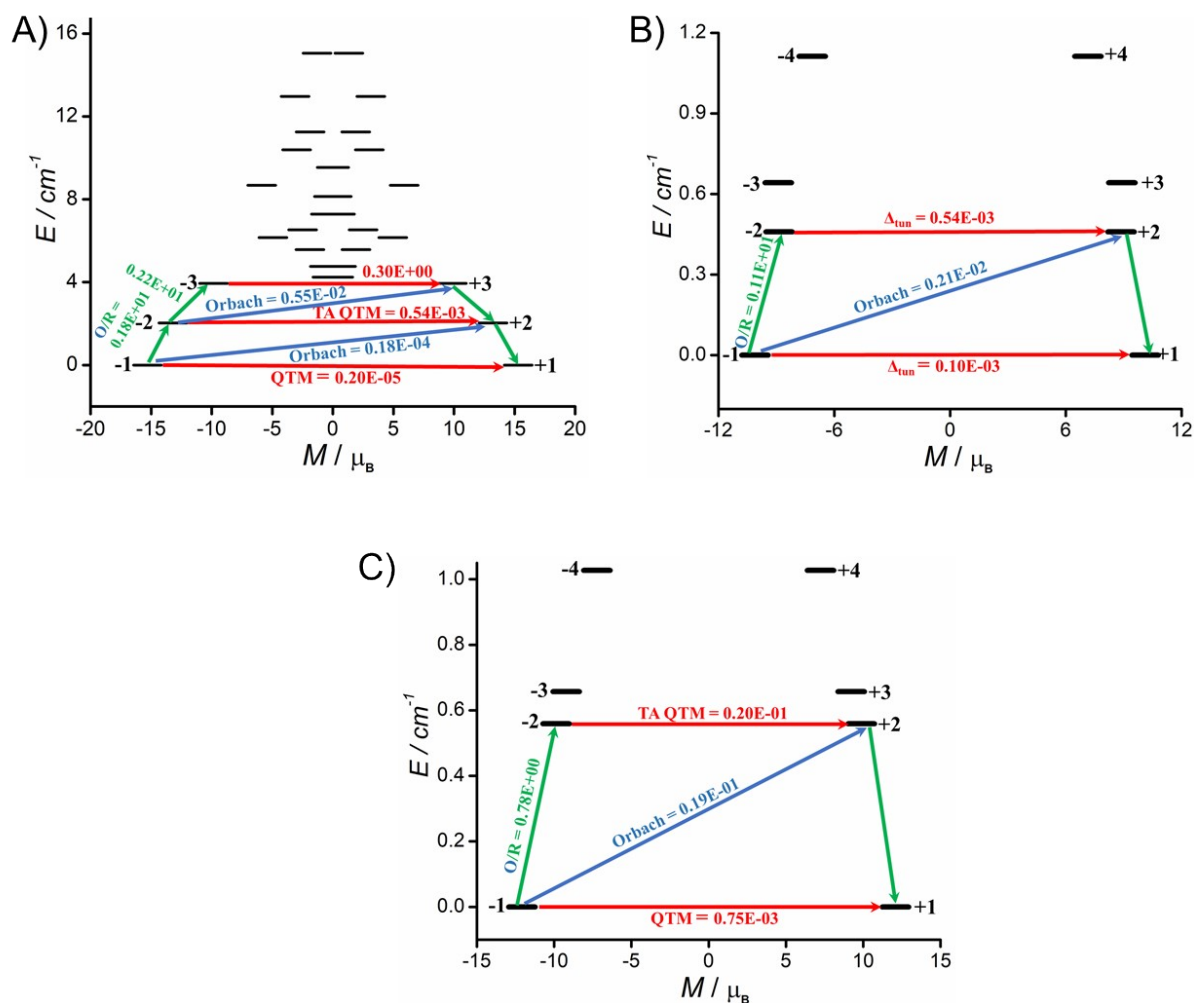


Figure S9. Low-lying exchange spectra (A, B and C) for complexes **4-6** respectively. Exchange states are placed on the diagram in according to their magnetic moment. Red dotted arrows/curved arrows show the tunnelling of magnetization within each doublet, while the green and blue dotted arrows show the possible relaxation through Raman/Orbach process.

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