Supporting Information for

Dynamic Nuclear Polarization of ^{1}H, ^{13}C, and ^{59}Co in a Co(III) tris-ethylenediamine crystalline lattice doped with Cr(III)

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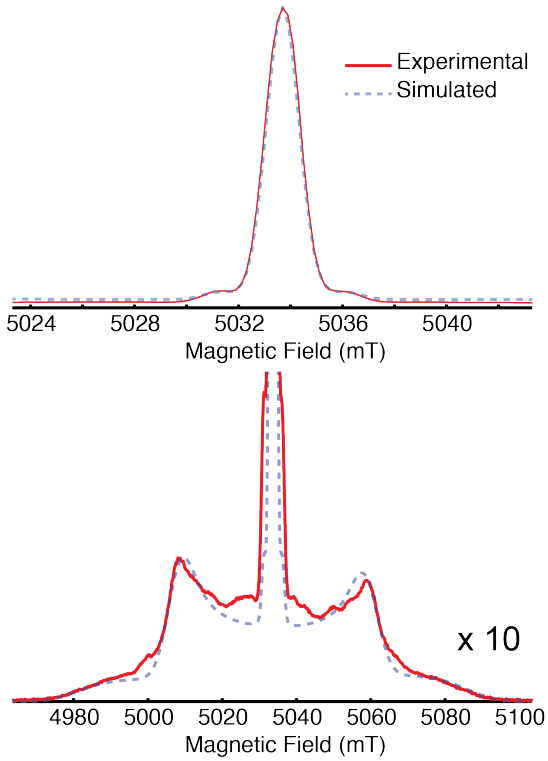

Figure S1: Cr(III) EPR spectrum (red solid) and simulation (blue dashed) of the Cr(III)-doped $[Co(en)_3Cl_3]_2{\cdot}NaCl{\cdot}6H_2O$ molecular complex at 139.997 GHz at 80 K. Data was simulated using a g = 1.98714, D = 740 MHz, S = 3/2, and ^{53}Cr natural abundance (9.5%) with a hyperfine coupling of 45 MHz. Experimental and simulation of the full spectrum (top) and a 10× vertical magnification to emphasize the $m_S = \pm 3/2 \leftrightarrow \pm 1/2$ transitions (bottom). Simulation and experiment have been scaled differently in top and bottom figure. This was necessary due to different transition moments of the central transition (CT) and satellite transitions (ST) resulting in different flip angles of pulses.

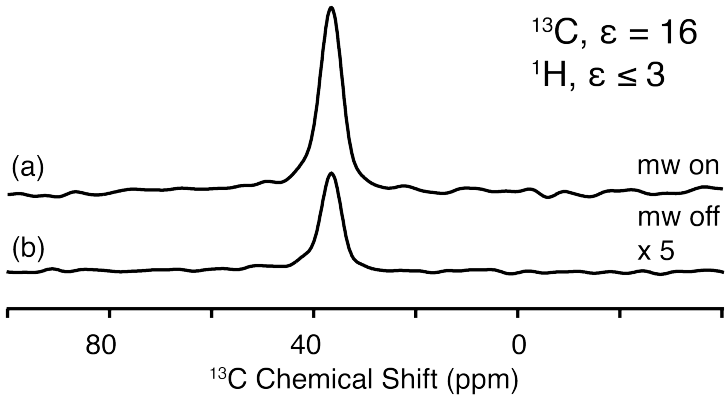

Figure S2: ^{13}C DNP MAS NMR of 3% Cr(III)-doped $[Co(en)_3Cl_3]_2{\cdot}NaCl{\cdot}6H_2O$, (a) microwave on and (b) microwave off (with a 5× vertical magnification). The indirect enhancement (*i.e.,* through ^{1}H's) is ε ≤ 3 (not shown).

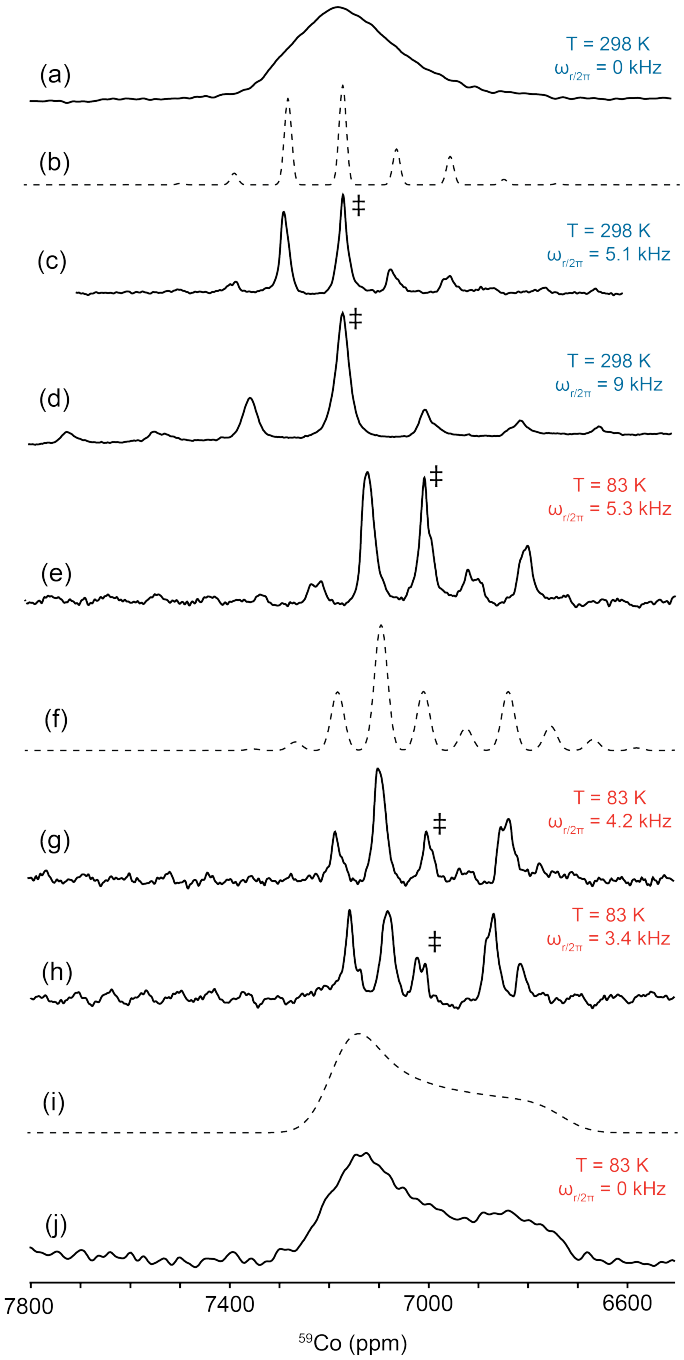

Figure S3: ^{59}Co NMR and DNP NMR spectra of 3% Cr(III)-doped $[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$ at 298 K (a-d) and 80 K (e-j). (a) Non-spinning, (b) simulation of (c), (c) MAS – 5.1 kHz, (d) MAS – 9 kHz, (e) MAS – 5.3 kHz, (f) simulation of (g), (g) MAS – 4.2 kHz, (h) MAS – 3.4 kHz, (i) simulation of (j), (j) non-spinning. CSA parameters are located in Table S3, isotropic chemical shift (δ_{iso}) is marked by "‡".

Table S1: Structural parameters for $[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$ Complex

Parameters	$[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$[1]
Source	X-ray
Crystal system	trigonal
Space group	*P*3
a	11.415
c	8.055
Z	1
Density (g/cm^3)	1.567
Temperature	293 K
	Diamagnetic

Table S2: Nuclear longitudinal spin-lattice (T_{1I}) and build-up time (T_B) constants

Cr(III) doping (%)	**^{1}H** ε	T_B (s)	T_{1I} (s)	**^{13}C** ε	T_B (s)	T_{1I} (s)	**^{59}Co** ε	T_B (s)	T_{1I} (s)
0.1	2.7	20	20	6.6	>1,200	*n.d.*	1.6	50	*n.d.*
0.3	2.4	6.2	6.4	7.2	564	*n.d.*	1.7	20	22
1.0	2.2	1.6	1.5	9.9	111	108	3.6	12	11
3.0	2.3	0.3	0.3	15.4	40	36	10.8	7.1	9.8

Table S3: ^{13}C CP MAS NMR isotropic line widths at 298 K (500 MHz, 10 kHz MAS, Shimmed) and 83 K (212 MHz, 4 kHz MAS, Not Shimmed) of $[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$ with 0 to 3 % Cr doping

Cr(III) (mol%)	Isotropic linewidth (Hz)	Difference (%)	Isotropic linewidth (ppm)	Difference (%)
	298 K, 11.7 T		83 K, 5 T	
0	69	0	151	0
0.1	72	4	154	2
0.3	76	11	157	4
1	77	11	174	15
3	83	20	212	40

Table S4: ^{59}Co NMR Parameters for $[Co(en)_3]Cl_3$ based complexes

Sample	$[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$	$[Co(en)_3Cl_3]_2 \cdot NaCl \cdot 6H_2O$	$[Co(en)_3]Cl_3 \cdot 3H_2O$	$[Co(en)_3]Cl_3$
T (K)	85	290	298	298
δ_{iso} (ppm)	7020	7160	7281	7288
Ω (ppm)	520	380	277	280
κ	>0.9	>0.9	1.0	1.0
C_Q (MHz)	3.2	3.2	-3.05	-2.8
η	<0.1	<0.1	0.0	0.0
Reference	this work	this work	2	2

References

(1) Farrugia, L.J., Peacock, R.D. and Stewart, B., *Acta Cryst.*, **2000,** *C56*, 149-151

(2) Ueda, T., Bernard, G.M., McDonald, R. and Wasylishen, R.E., *Solid State Nucl. Magn. Reson.*, **2003**, *24*, 163-183.