Supporting Information

Figure S1. INS spectra of a powder sample of deuterated $Fe(Im)_6(NO_3)_2$. Spectra were recorded at FOCUS at various temperatures using an incident wavelength λ of 4.5 Å. The feature marked with an asterisk originates from the aluminium sample can. The labels I and I' correspond to the transitions depicted in the inset of figure 2.

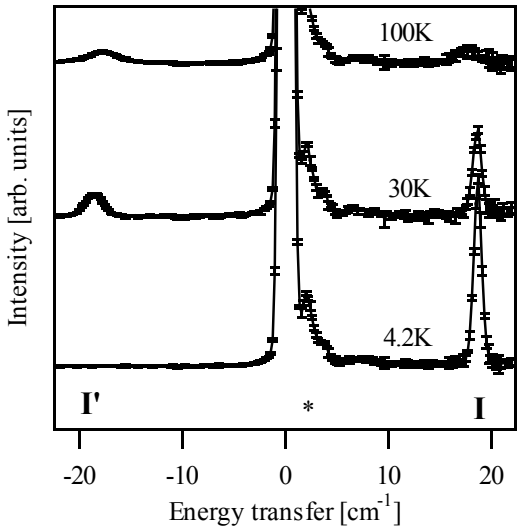

Figure S2. INS spectra of a powder sample of deuterated $Fe(Im)_6(NO_3)_2$ at 1.5K, 4.2K, 30K and 100K. Spectra were recorded at FOCUS using an incident wavelength λ of 3.3 Å. The labels I and I' correspond to the transitions depicted in the inset of figure 2.

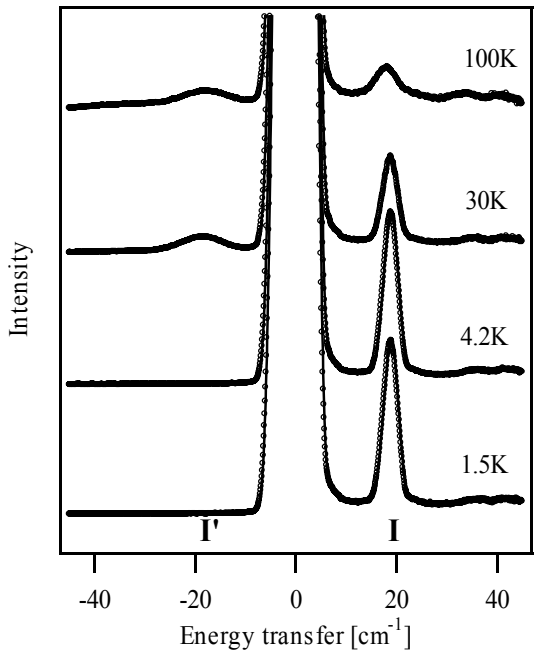

Table S3. Atomic coordinates (Å x 10^4) and equivalent isotropic displacement parameters (Å^2 x 10^3) for $Fe(C_3H_4N_2)_6(NO_3)_2$. [a]

Atom	*x*	*y*	*z*	*U(eq)*
C(1)	1641(2)	-111(2)	6717(1)	26(1)
C(2)	2424(2)	-540(2)	6923(1)	35(1)
C(3)	1715(1)	-1095(1)	5526(1)	20(1)
N(1)	1188(1)	-468(1)	5835(1)	18(1)
N(2)	2471(1)	-1150(1)	6154(1)	27(1)
Fe(1)	0	0	5000	14(1)
N(3)	3333	-3333	5602(1)	18(1)
O(1)	4031(1)	-2180(1)	5604(1)	27(1)

[a] U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

Table S4. Anisotropic displacement parameters ($Å^2$ x 10^3) for $Fe(C_3H_4N_2)_6(NO_3)_2$. The anisotropic displacement factor exponent takes the form: -2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

Atom	U11	U22	U33	U23	U13	U12
C(1)	30(1)	28(1)	24(1)	-7(1)	-9(1)	18(1)
C(2)	42(1)	38(1)	35(1)	-13(1)	-21(1)	28(1)
C(3)	19(1)	19(1)	23(1)	1(1)	-1(1)	10(1)
N(1)	17(1)	18(1)	19(1)	2(1)	0(1)	9(1)
N(2)	25(1)	24(1)	38(1)	-5(1)	-9(1)	17(1)
Fe(1)	13(1)	13(1)	15(1)	0	0	6(1)
N(3)	20(1)	20(1)	13(1)	0	0	10(1)
O(1)	22(1)	16(1)	40(1)	1(1)	-2(1)	7(1)