Electronic Supporting Information for

Two mononuclear iron(II) complexes of 4-phenylpyrazole-5-carbaldehyde derived ligands are stabilised in different spin states

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Table S1. Crystal data and structure refinement for $[Fe^{II}(H_2L^1)_2](BF_4)_2 \cdot ¼(Et_2O) \cdot ¼(H_2O) \cdot MeOH$. This complex was synthesised from nitromethane, so the methanol solvent molecules found in the crystal structure are probably from the ligand, which was isolated as a methanol solvate according to the ^1H NMR data.

Identification code	jo619c2c	
Empirical formula	$C_{42}\ H_{39}\ B_2\ F_8\ Fe\ N_{12}\ O_{1.50}$	
Formula weight	965.32	
Temperature	84(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	*C*2/*c*	
Unit cell dimensions	a = 23.047(10) Å	α = 90°.
	b = 23.329(10) Å	β = 95.974(13)°.
	c = 17.388(7) Å	γ = 90°.
Volume	9298(7) Å3	
Z	8	
Density (calculated)	1.379 Mg/m^3	
Absorption coefficient	0.405 mm^{-1}	
F(000)	3960	
Crystal size	0.11 x 0.05 x 0.04 mm^3	
Theta range for data collection	1.25 to 23.81°.	
Index ranges	$-25 \le h \le 26$, $-23 \le k \le 26$, $-19 \le l \le 19$	
Reflections collected	25950	
Independent reflections	7138 [R(int) = 0.1815]	
Completeness to theta = 23.81°	99.9 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7454 and 0.5238	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	7138 / 166 / 586	
Goodness-of-fit on F^2	1.026	
Final R indices [I>2sigma(I)]	R1 = 0.1208, wR2 = 0.3067	
R indices (all data)	R1 = 0.2899, wR2 = 0.3960	
Largest diff. peak and hole	1.237 and -0.616 e.$Å^{-3}$	

Table S2. Selected bond lengths [Å] and angles [°] for $[Fe^{II}(H_2L^1)_2](BF_4)_2 \cdot ¼\ (Et_2O) \cdot ¼\ (H_2O) \cdot MeOH$.

Fe(1)-N(1)	1.923(11)
Fe(1)-N(11)	1.926(11)
Fe(1)-N(4)	1.938(10)
Fe(1)-N(7)	1.954(10)
Fe(1)-N(5)	1.964(10)
Fe(1)-N(9)	1.965(10)
N(1)-Fe(1)-N(11)	90.3(4)
N(1)-Fe(1)-N(4)	89.3(5)
N(11)-Fe(1)-N(4)	94.9(4)
N(1)-Fe(1)-N(7)	92.1(4)
N(11)-Fe(1)-N(7)	169.5(4)
N(4)-Fe(1)-N(7)	95.4(4)
N(1)-Fe(1)-N(5)	169.4(4)
N(11)-Fe(1)-N(5)	86.6(4)
N(4)-Fe(1)-N(5)	80.8(5)
N(7)-Fe(1)-N(5)	92.8(4)
N(1)-Fe(1)-N(9)	93.6(4)
N(11)-Fe(1)-N(9)	89.7(5)
N(4)-Fe(1)-N(9)	174.5(5)
N(7)-Fe(1)-N(9)	79.9(4)
N(5)-Fe(1)-N(9)	96.5(4)

Table S3. Hydrogen bonds for $[Fe^{II}(H_2L^1)_2](BF_4)_2 \cdot ¼\ (Et_2O) \cdot ¼\ (H_2O) \cdot MeOH$ [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(2)-H(2X)...F(13)#2	0.88	2.12	2.933(13)	152.7
N(6)-H(6)...F(26)#2	0.88	1.78	2.660(17)	172.8
N(6)-H(6)...F(21)#2	0.88	2.08	2.909(18)	155.6
N(6)-H(6)...F(22)#2	0.88	2.42	3.187(18)	145.5
N(8)-H(8X)...F(12)	0.88	1.97	2.847(12)	171.3
N(12)-H(12X)...F(24)	0.88	1.99	2.788(12)	150.1
N(12)-H(12X)...F(23)	0.88	2.50	3.105(18)	126.1

Symmetry transformations used to generate equivalent atoms:
#1 x,-y+1,z+1/2

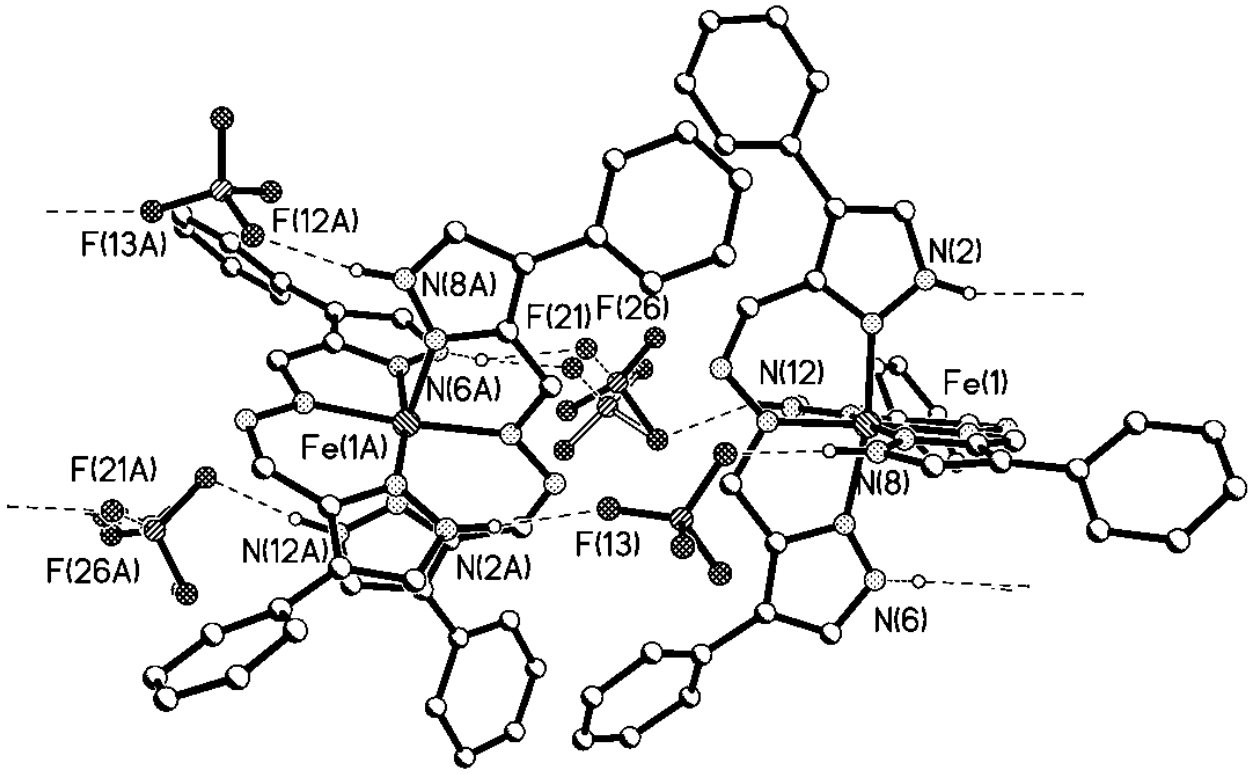

Fig. S1. Ball and stick representation of the supramolecular chains along the *c* axis, formed by the hydrogen bonds between the complex cations and BF_4 anions. Symmetry operation A: x, y-1, z-0.5

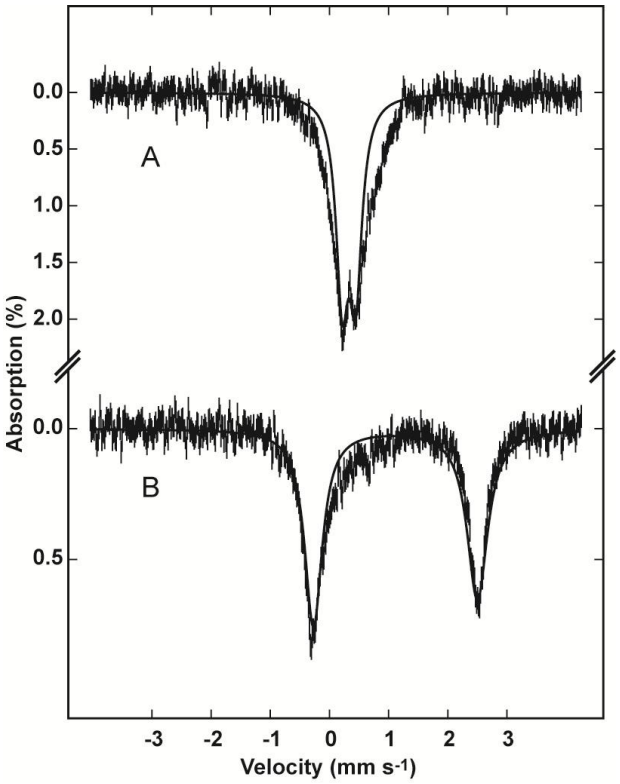

Fig. S2. Mössbauer spectra measured with a low magnetic field (0.47 mT) applied perpendicular to the γ-rays of **A)** $[Fe^{II}(H_2L^{1})_2](BF_4)_2{\cdot}2H_2O$ acquired at 295 K and **B)** $[Fe^{II}(HL^{2})(MeOH)(NCSe)]{\cdot}H_2O$ acquired at 4.6 K.