Electronic Supplementary Information

Spin-crossover in a *trans*-[$FeL_2(NCS)_2$] family (L = triaryltriazole): Remote substituent effects on spin transition modes and temperature

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Table S1 Dihedral angles (°) between the aryl ring and triazole ring of triaryltriazoles in **1**-**4**.

Complexes	Py/Trz	*p*-Cl-Ph/Trz	*p*-CH_3-Ph/Trz	*p*-CH_3O-Ph/Trz	*p*-F-Ph/Trz
1	13.4(3)/14.3(3)	85.8(3)			
2	11.6(3)/14.8(3)		82.9(3)		
3 $0.5H_2O$	9.9(3)		75.5(3)	36.3(3)	
4	9.3(3)		80.4(3)		36.9(3)

Table S2 Hydrogen-bond geometry and π-stacking interaction (Å, °) for **1** and **2**.

Complexes	D−H ··A	d(D−H)	d(H ··A)	d(D ··A)	∠D−H ··A
1	C9−H9 ··Cl1^{i}	0.93	2.780(1)	3.528(4)	138
	C2−H2 ··C17^{ii}	0.93	2.744(1)	3.627(4)	159
	C14−H14 ··N3^{iii}	0.93	2.506(2)	3.403(4)	162
	C4−H4 ··π (C13-18)	0.93	2.899(1)	3.707(4)	146
	C19 ··C12^{iii}			3.309(3)	
	π–π interaction		d(cent ··cent)	d(π ··π)	dihedral angle
	π(N4) ··π^{iii} (N4^{iii})		3.816	3.685	0.0
	π(N5) ··π^{iv} (N5^{iv})		3.790	3.547	0.0
2	C9−H9 ··C19^{i}	0.93	2.889(1)	3.631(4)	138
	C2−H2 ··C15^{v}	0.93	2.732(1)	3.621(4)	160
	C18−H18 ··N3^{vi}	0.93	2.523(2)	3.433(4)	167
	C4−H4 ··π (C13-18)	0.93	2.887(1)	3.708(4)	148
	C20 ··C12^{vi}			3.357(3)	
	π–π interaction		d(cent ··cent)	d(π ··π)	dihedral angle
	π(N4) ··π^{vi} (N4^{vi})		4.001	3.841	0.0
	π(N5) ··π^{vii} (N5^{vii})		3.885	3.593	0.0

Symmetry codes: (*i*) *x*, *y*, *z*-1; (*ii*) -*x*, 1-*y*, 2-*z*; (*iii*) 1-*x*, 1-*y*, 1-*z*; (*iv*) 1-*x*, -*y*, 1-*z*; (*v*) -*x*, -*y*, 1-*z*; (*vi*) 1-*x*, -*y*, -*z*; (*vii*) 1-x, -1-y, -z.

Table S3 Hydrogen-bond geometry and π-stacking interaction (Å, °) for **3** $0.5H_2O$ and **4**.

Complexes	D−H ··A	d(D−H)	d(H ··A)	d(D ··A)	∠D−H ··A
3 $0.5H_2O$	C16−H16 ··O1W	0.93	2.550	3.136(4)	121
	C20^{viii}−H20A^{viii} ··O1	0.93	2.530	3.169(6)	126
	C14A−H14E ··S1^{ix}	0.96	2.826	3.470(4)	125
	C16−H16 ··π^{iv} (Py)	0.93	3.045	3.883(3)	151
	C4−H4 ··π (C15-20)	0.93	2.890	3.728(4)	151
4	C15−H15 ··F1^{x}	0.93	2.490	3.329(3)	150
	C3−H3B ··S1^{xi}	0.93	2.938	3.656(3)	135
	C19−H19 ··π^{xi} (Py)	0.93	2.976	3.788(3)	147
	C4−H4 ··π (C14-19)	0.93	2.903	3.746(3)	151

Symmetry codes: (*viii*) *x*, -1/2-*y*, 1/2+*z*; (*ix*) *x*-1, -1/2-*y*, *z*-1/2; (*x*) *x*, 1/2-*y*, *z*-1/2; (*xi*) 2-*x*, -*y*, -*z*.

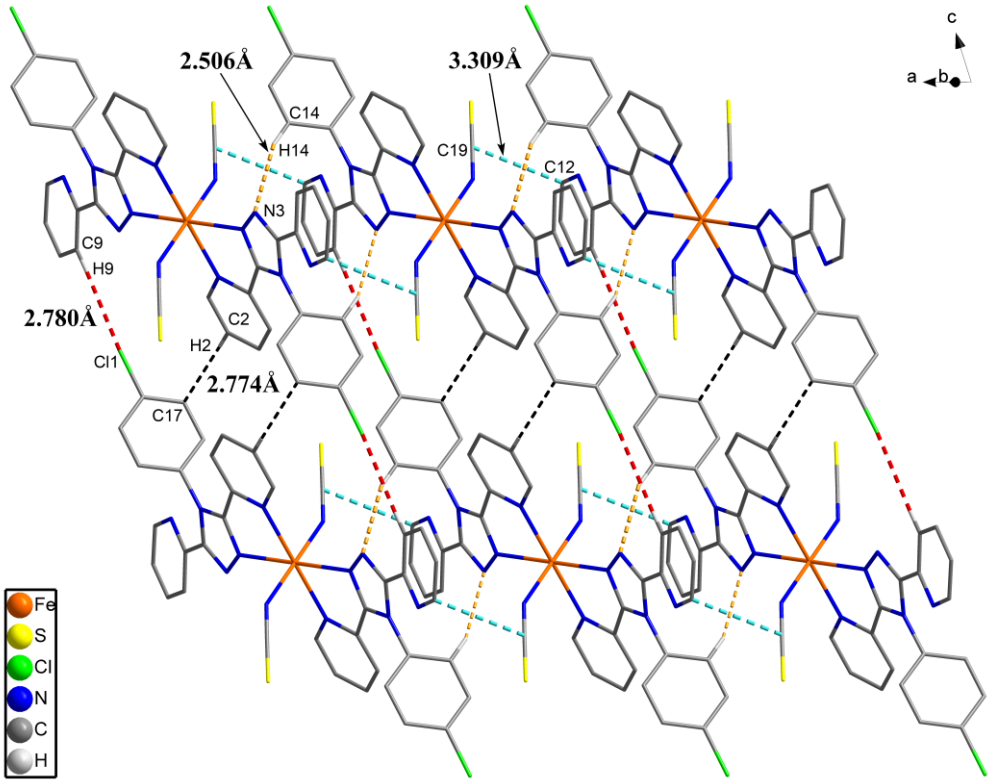


Figure S1. View of the hydrogen bonds and C ··C contacts in **1**.

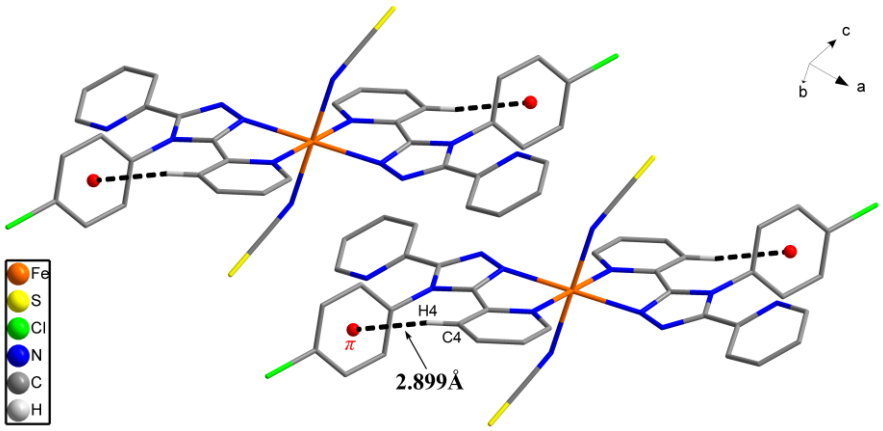


Figure S2. View of the intramolecular edge-to-face C4-H4 ··π interaction in **1**.

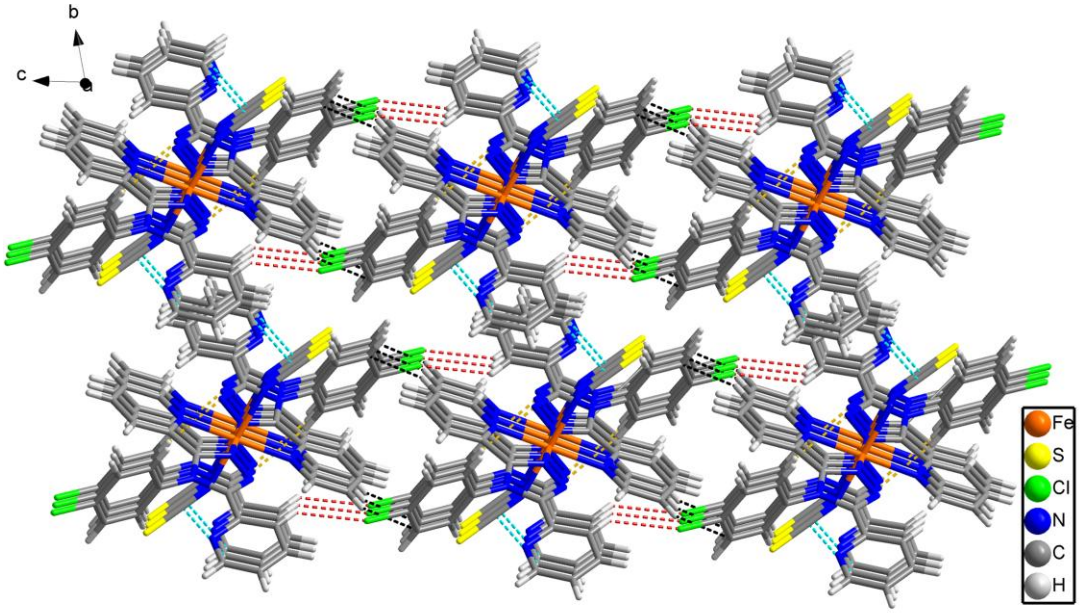


Figure S3. 3D structure of **1**.

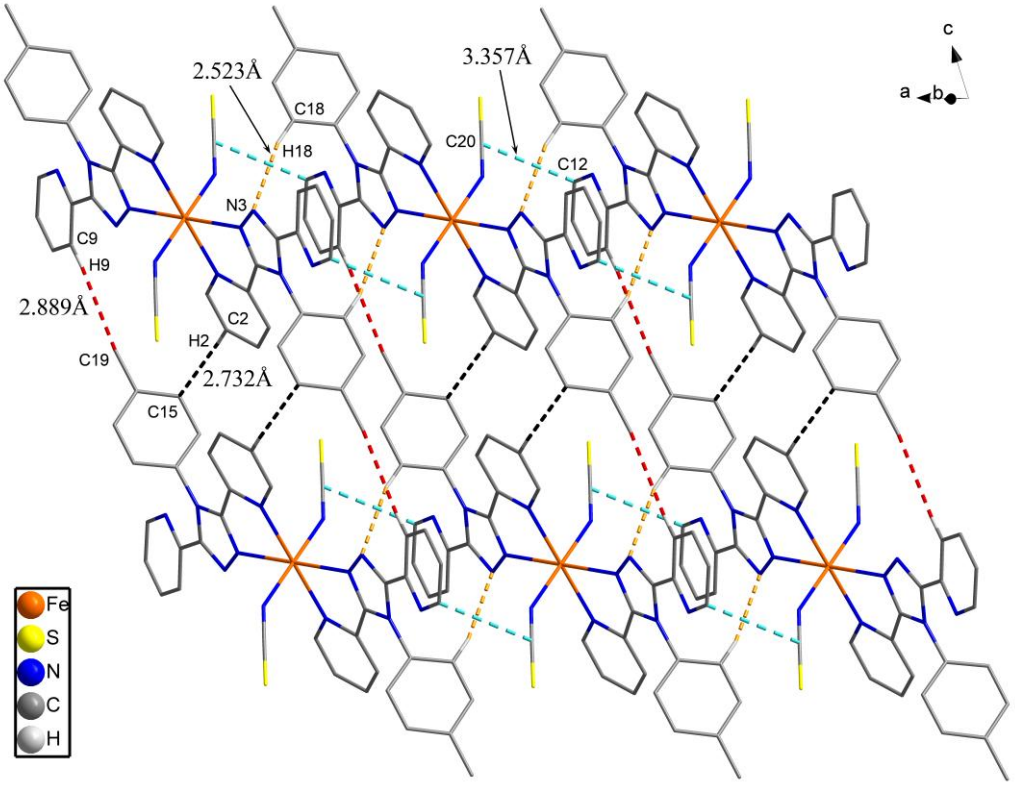


Figure S4. View of the hydrogen bonds and C ··C contacts in **2**.

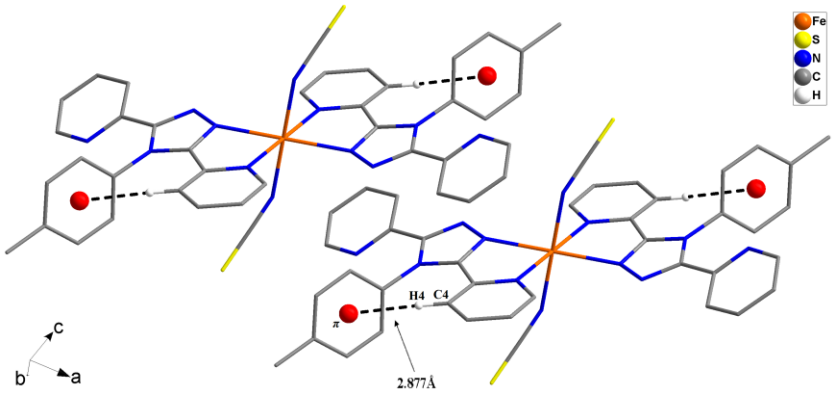


Figure S5. View of the intramolecular edge-to-face C4-H4 $\cdots\pi$ interaction in **2**.

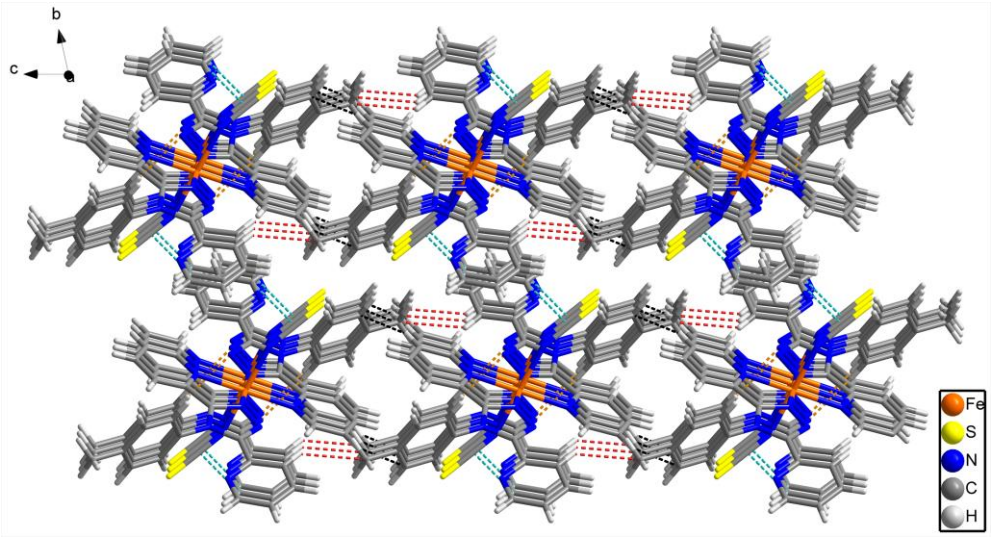
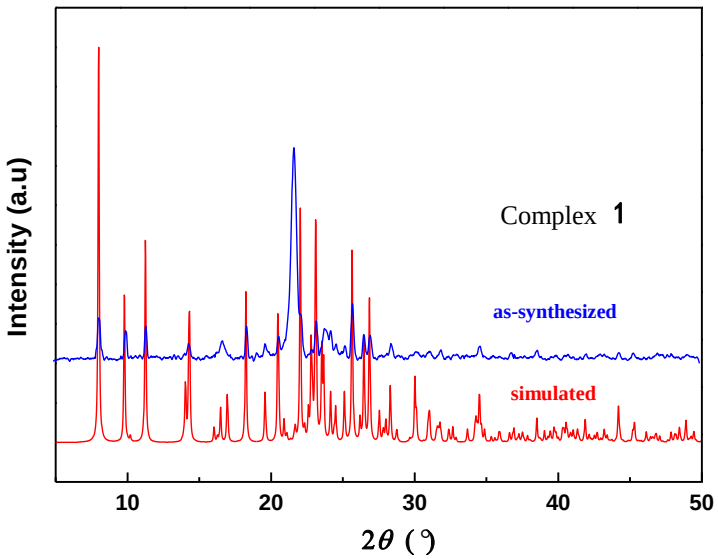


Figure S6. 3D structure of **2**.



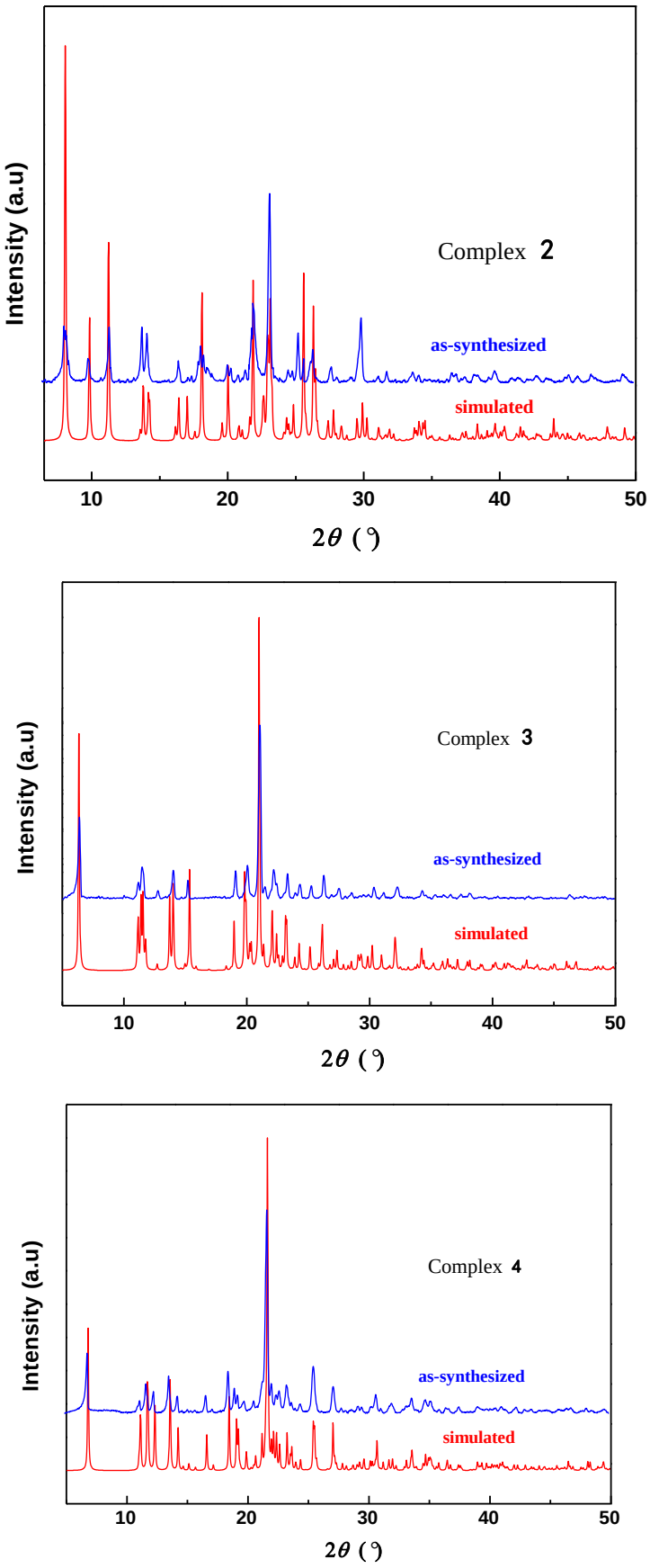


Fig. S7 Experimental and simulated powder XRD patterns of complexes **1**-**4**.