Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Insights into the Crystal-Packing Effects in the Spin Crossover of $[Fe^{II}(1\text{-bpp})]^{2+}$-based Materials

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- Supporting Information -

S1 - Model Systems and Unit Cell parameters for 1a-1h.

The initial (crystal structures reported in the literature) and final (optimized) lattice parameters of the super-cells are collected in Table S1. For the distorted super-cells (**1a**, **1c**, **1e** and **1g**) the crystalline unit cells, which contained 4 SCO units and 8 counterions, were directly adopted as initial structure. For the regular ones (**1b**, **1d**, **1f** and **1h**), the unit cells, which contained 2 SCO units and 4 counterions, were doubled in order to include the same number of molecules as in the distorted set of super-cells. One must note that we have chosen to double along the 'a' direction because is the one that holds the main difference between both sets of unit cells (see Figure S1). We have selected model systems with 4 SCO and 8 counterions because it is a balanced choice between computational cost and versatility, that is, we provide enough degrees of freedom to the system to break its symmetry in any direction, if necessary.

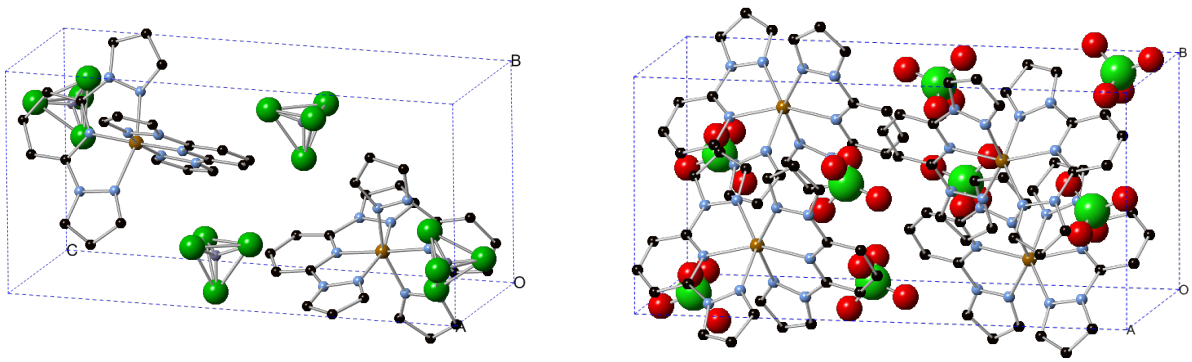

Figure S1. Representation of the (left) regular and (right) distorted sets of initial unit cells. Note that they include a different number of SCO units and Counterions.

Table S1. Initial and final cell parameters for the **1a-1h** variable-cell geometry optimizations. All distances indicated in Å and angles in degrees.

	1a		**1b**		**1c**		**1d**	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
a	14.386	14.258	17.092	16.953	14.386	17.022	17.008	16.791
b	9.246	9.028	8.559	8.389	9.246	9.238	8.566	8.530
c	20.103	20.054	19.153	19.138	20.103	20.128	18.496	18.606
α	90.0	90.0	90.0	89.979	90.0	88.107	90.0	90.014
β	99.438	100.703	95.64	98.122	99.438	100.423	98.411	99.463
γ	90.0	90.0	90.0	89.991	90.0	89.850	90.0	89.956
	1e		**1f**		**1g**		**1h**	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
a	14.386	13.839	17.092	16.780	14.386	17.568	17.008	16.590
b	9.246	9.094	8.559	8.278	9.246	8.788	8.566	8.397
c	20.103	19.864	19.153	18.768	20.103	17.384	18.496	18.259
α	90.0	90.0	90.0	90.0	90.0	90.047	90.0	89.984
β	99.438	100.031	95.64	97.291	99.438	92.599	98.411	98.949
γ	90.0	90.0	90.0	90.0	90.0	90.012	90.0	90.010