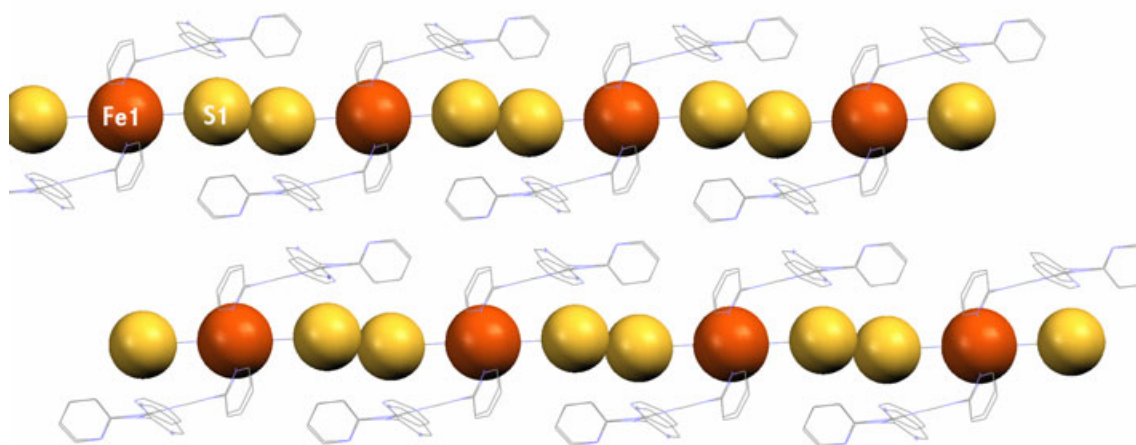


## Spin transition in a triazine-based Fe(II) complex: variable-temperature structural, thermal, magnetic and spectroscopic studies

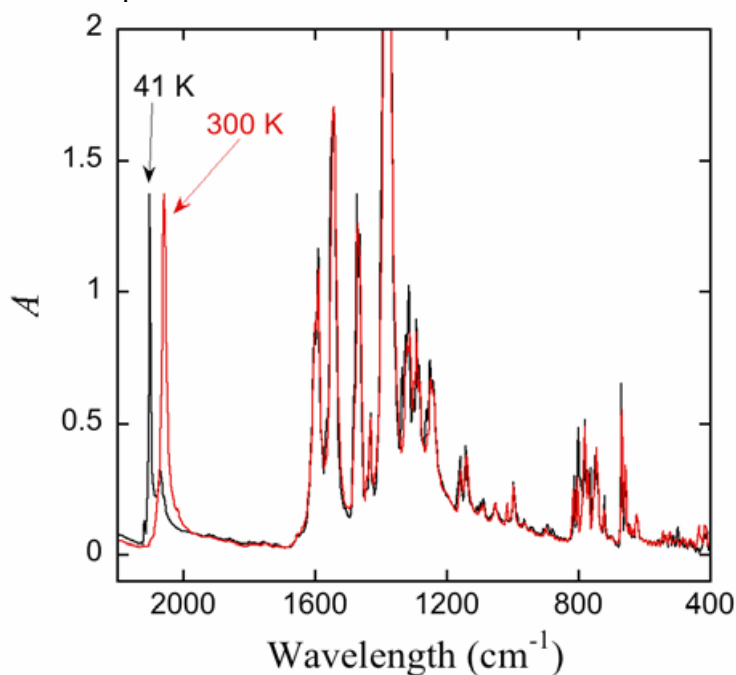
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### Supplementary Material

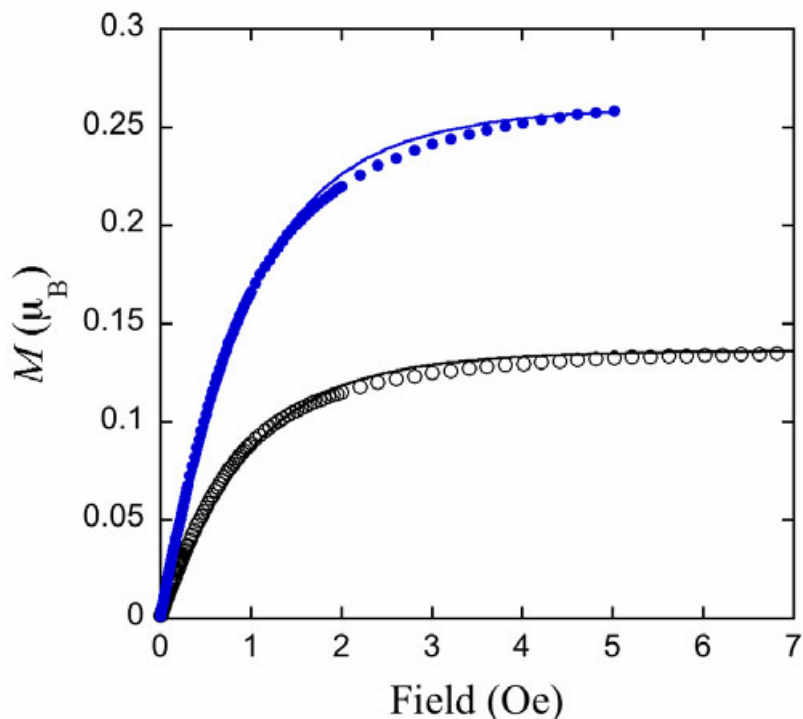
**Figure S1.** Representation of the molecular structure of  $[\text{Fe}(\text{dpyatriz})_2(\text{SCN})_2]$  (**2**), emphasizing the  $[\text{S}\cdots\text{S}]$  interactions between individual molecules, leading to “inorganic wires” protected by an organic shell of dpyatriz ligands.



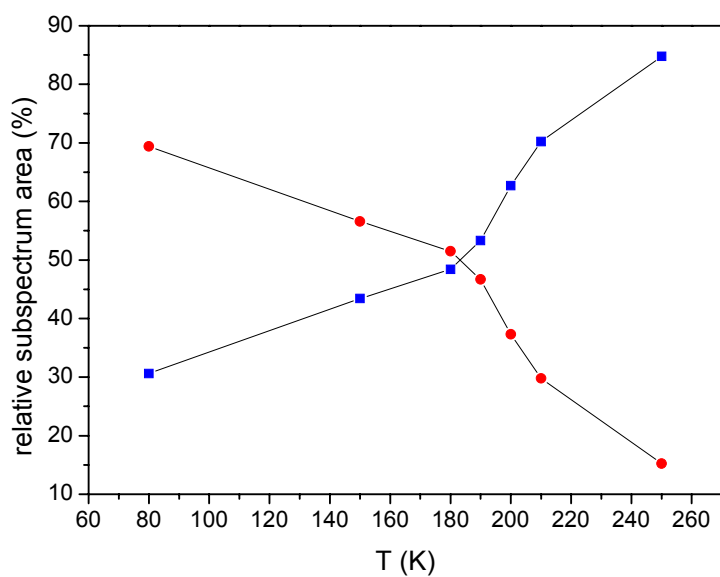
**Figure S2.** Infrared spectra of a ground sample of  $[\text{Fe}(\text{dpyatriz})_2(\text{SCN})_2]$  (**2**) and 41K (black) and 300K (red), emphasizing the differences and similarities between the HS and LS forms of the complex.



**Figure S3.** Isofield magnetization vs field curves of a polycrystalline (black) and ground (blue) sample of  $[\text{Fe}(\text{dpyatriz})_2(\text{SCN})_2]$  (**2**) at 2K, allowing to estimate the residual fraction of the HS form of the complex at this temperature in ca. 3.4 and 6.5 %, respectively. The solid lines are fits to the Brillouin equation.



**Figure S4.** Relative area of the HS (blue squares) versus LS (red circles) forms of  $[\text{Fe}(\text{dpyatriz})_2(\text{SCN})_2]$  (**2**) at different temperatures, as revealed by Mössbauer spectroscopy, showing a gradual and ca. 70 % complete spin transition.



**Figure S5.** Plot of the heat capacity at constant pressure,  $C_p$ , of  $[\text{Fe}(\text{dpyatriz})_2(\text{SCN})_2]$  (**2**) versus  $T$  as derived from Differential Scanning Calorimetry. A heat capacity anomaly is observed, centered near 200K, the temperature where the spin transition of **2** takes place.

