

Supporting Information

The degree of distortion away from ideal geometries for the coordination polyhedra around the Fe atoms (Figures S1-S3) is estimated by the Muetterties-Guggenberger¹ and the Continuous Symmetry Measure² (CSM) methods. The results of those calculations are presented in Tables S1-S3. In Table S1 we compare the dihedral angles associated with each of the 12 edges of the coordination polyhedra of the Fe atoms in **1** and **2**. In Table S2 we present the Bailar-twist angles for each of these polyhedra and in Table S3 we present their symmetry measures $S(G)$ according to the definition of Alvarez et al. [$S(G) = 0$ corresponds to a polyhedron site of perfect G symmetry].

Table S1 Coordination polyhedra edges for Fe atoms in **1** and **2** and associated dihedral angles as defined by Muetterties and Guggenberger.¹

1: Fe		2: Fe(1)		2: Fe(2)	
Edge	Dihedral angle (°)	Edge	Dihedral angle (°)	Edge	Dihedral angle (°)
N(31)-O(61)	58.7	N(61)-O(21)	52.7	N(2)-O(61)	39.92
N(61)-O(1)	56.2	N(22)-O(41)	55.0	N(62)-O(41)	45.954
N(1)-O(31)	58.1	N(42)-O(61)	42.4	N(41)-O(1)	42.06
O(61)-N(61)	84.3	O(61)-N(61)	86.1	N(2)-O(1)	93.403
O(1)-N(1)	82.7	O(21)-N(22)	94.3	N(41)-O(41)	94.691
O(31)-N(31)	85.11	O(41)-N(42)	89.2	N(62)-O(61)	91.05
N(1)-N(61)	76.5	O(61)-O(21)	77.7	O(1)-O(61)	77.91
N(61)-N(31)	73.44	O(41)-O(21)	50.6	O(41)-O(1)	66.22
N(1)-N(31)	75.44	O(41)-O(61)	81.5	O(61)-O(41)	70.58
O(1)-O(61)	66.18	N(61)-N(42)	80.3	N(62)-N(41)	73.455
O(31)-O(1)	65.27	N(61)-N(22)	63.3	N(2)-N(62)	77.394

O(61)-O(31) 66.72 N(22)-N(42) 82.3 N(2)-N(41) 84.373

Table S2 Bailar-twist angles for the coordination polyhedra of the Fe atoms in **1** and **2**.¹

	Bailar-twist angles (°)
1 : Fe	43 ^a
2 : Fe(1)	33, 34, 43
2 : Fe(2)	28, 32, 32

^aIndividual values are very close. Only the average value is given

Table S3 Symmetry measures $S(G)$ for the coordination polyhedra of the Fe atoms in **1** and **2**, according to the definitions of Alvarez et al.²

	1 : Fe	2 : Fe(1)	2 : Fe(2)
$S(O_h)$	2.095	4.285	4.828
$S(itp)$	11.173	9.015	6.026

Based on the above, the values of dihedral angles defined by Muetterties and Guggenberger are quite dispersed between the values corresponding to ideal O_h (70.5° for all edges) and ideal trigonal prismatic (itp, 0, 90 and 120° for the three types of edges) geometries, suggesting intermediate geometries for all Fe atoms. Similarly, the Bailar-twist angles are intermediate between those that correspond to ideal O_h (60°) and itp (0°) geometries. In the case for **2**, these angles also exhibit a significant distribution.

On the other hand, the $S(G)$ value for the Fe atom in **1** suggests a polyhedron closer to trigonally distorted O_h than to itp geometry. However, this measure is less discerning for the Fe atoms in **2**. In particular for Fe(2), the results are controversial; $S(O_h) = 4.8 (> 4.42)$ indicates a distorted trigonal prism, whereas $S(itp) = 5.3 (> 4.42)$ indicates a distorted octahedron.

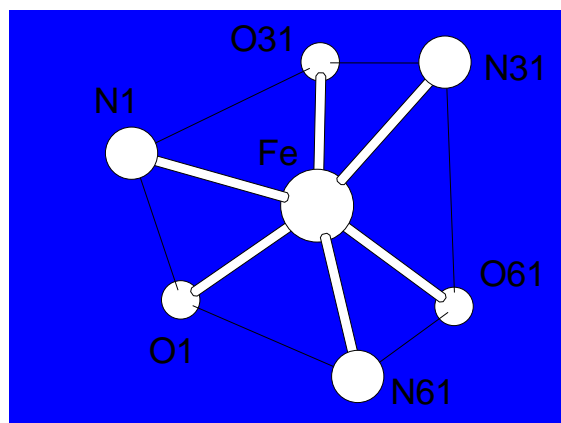


Fig. S1 Coordination polyhedron of Fe in **1**.

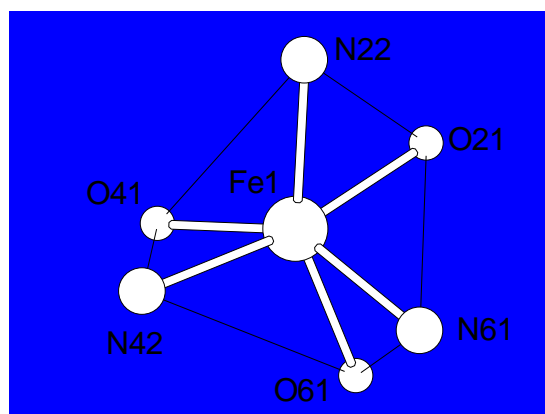


Fig. S2 Coordination polyhedron of Fe(1) in **2**.

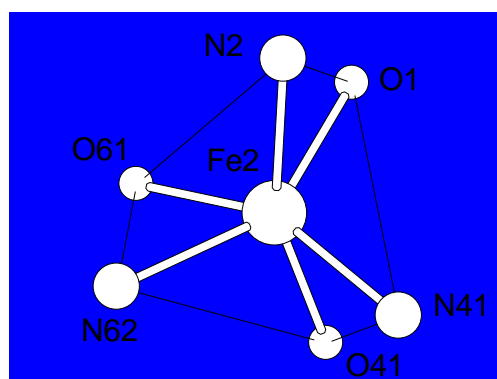


Fig. S3 Coordination polyhedron of Fe(2) in **2**.

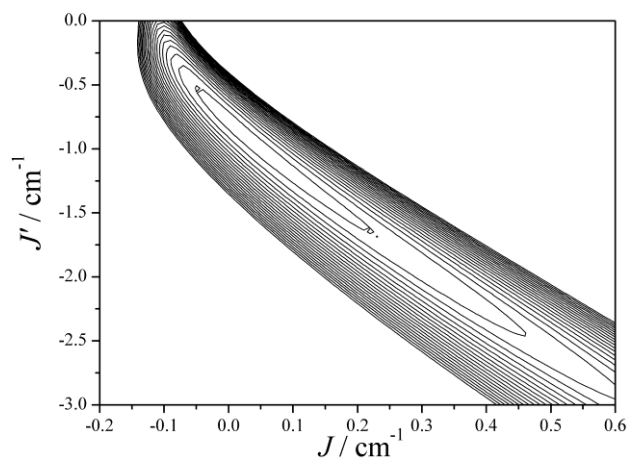


Fig. S4 Error contour-plot of J vs J' ($g = 1.98$) for the magnetic data of complex **2**, confirming the uniqueness of the best-fit solution described in the text.

¹ E. L. Muetterties and L. J. Guggenberger, *J. Am. Chem. S.* 1974, **96**, 1748.

² S. Alvarez, D. Anvir, M. Lulunell and M. Pinsky, *New J. Chem.* 2002, **26**, 996. We have used the internet interactive version of the SHAPE program (<http://telem.openu.ac.il/csm-calc/>)