Supporting Information

Table S1. Ranges of the Mössbauer parameters for the complex **1**

	Site 1	Site 2 & 3	Site 4
η	0 - 0.2	0 - 0.5	0.4 - 0.7
β_{efg}	75 - 105	100 - 110	85 - 95
γ_{efg}	free	free	70 - 110
A_x (KG)	–80 - (–70)	– 101 - (–95)	15 - 0
A_y (KG)	–80 - (–30)	– 77 - (–37)	60 - 50
A_z (KG)	–67 - (–66)	– 71 - (–69)	28.5 - 29.5

Table S2. Fe-Fe distances in all-ferrous cluster in *Av*2 and complex **1**

	Fe_iFe_j (Å)	
i, j	protein [a]	complex **1**
1,4	2.793	2.719
2,4	2.674	2.709
3,4	2.693	2.764
av.	**2.720**	**2.731**
1,2	2.573	2.602
1,3	2.630	2.674
2,3	2.516	2.612
av.	**2.573**	**2.629**

[a] The sites of the nitrogenase cluster labeled Fe_1, Fe_2, Fe_3, and Fe_4 correspond with the sites Fe(1), Fe(4), Fe(2), and Fe(3) in the PDB file 1G1M with the structure of the all-ferrous Fe-protein of *A. vinelandii*, respectively.

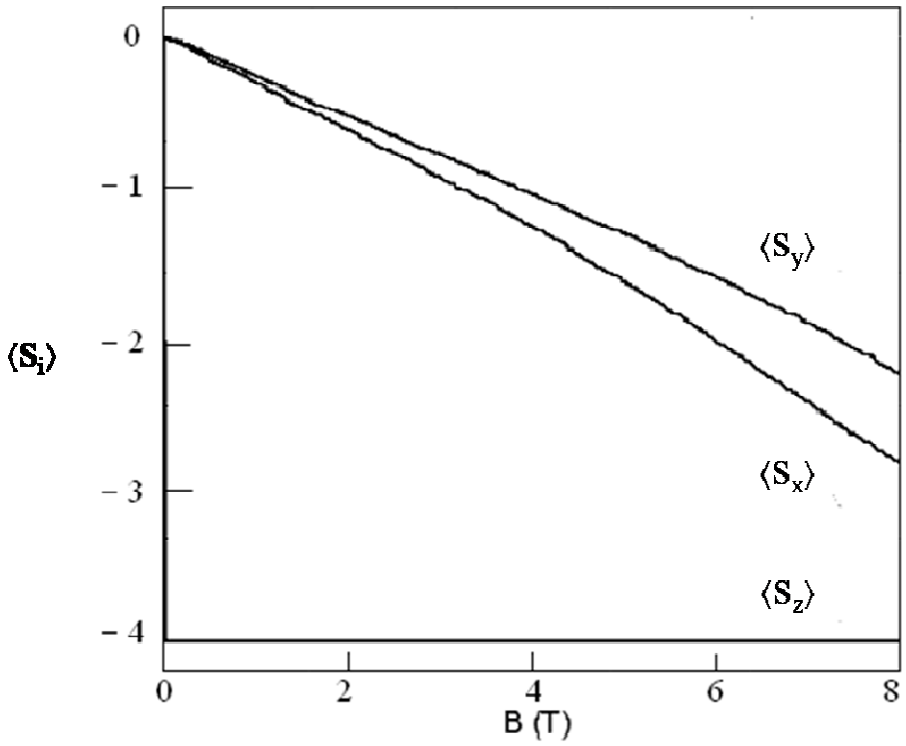

Figure S1. Spin expectation values, $\langle S_i \rangle$, i =x, y, and z, for the lowest state of an $S = 4$ multiplet obtained for a system with $D = -1.9$ cm^{-1} and $E/D = 0.07$, as a function of the applied magnetic field indicated.

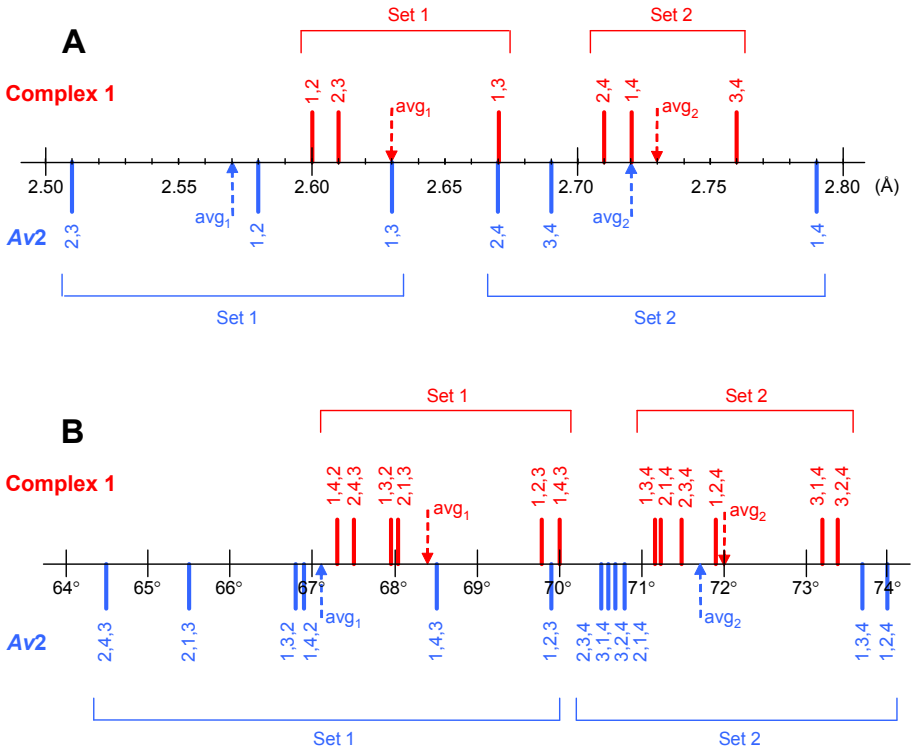

Figure S2. (**A**) Graphic representation of the Fe–Fe distances (Table S2) in the all-ferrous clusters of complex **1** (red) and *Av*2 (blue). (**B**) Graphic representation of the Fe–S–Fe angles (Table 8) in the all-ferrous clusters of complex **1** (red) and *Av*2 (blue). The averages (avg_i) for the subsets (sets 1 and 2, see section 5.3) have been indicated by dashed arrows.