Supporting information

for

Single-chain magnet based on 1D polymeric azido-bridged seven-coordinate Fe(II) complex with a pyridine-based macrocyclic ligand

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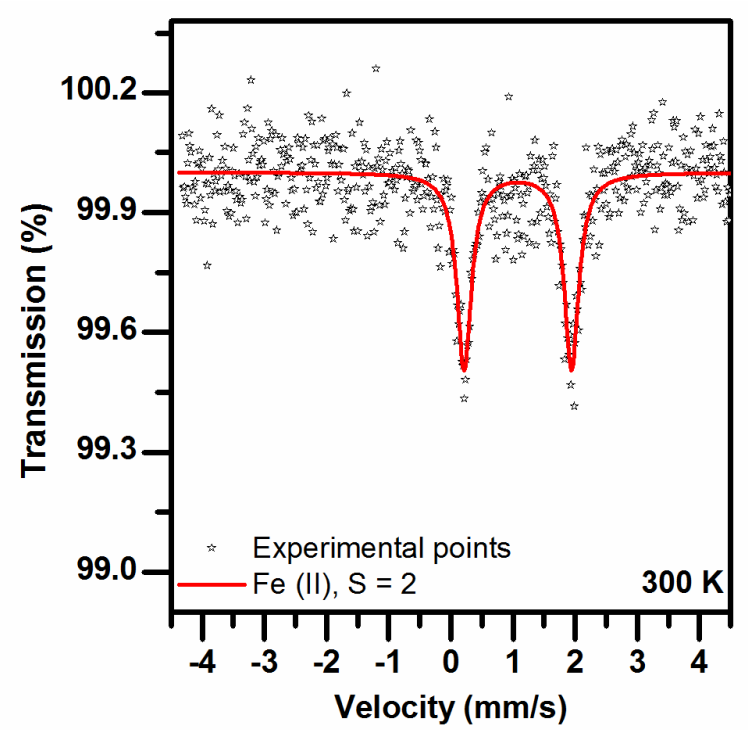

Figure S1. ^{57}Fe Mössbauer spectra of **1** recorded at room temperature and without an external magnetic field. Red line corresponds to the fit with the parameters listed in Table S1.

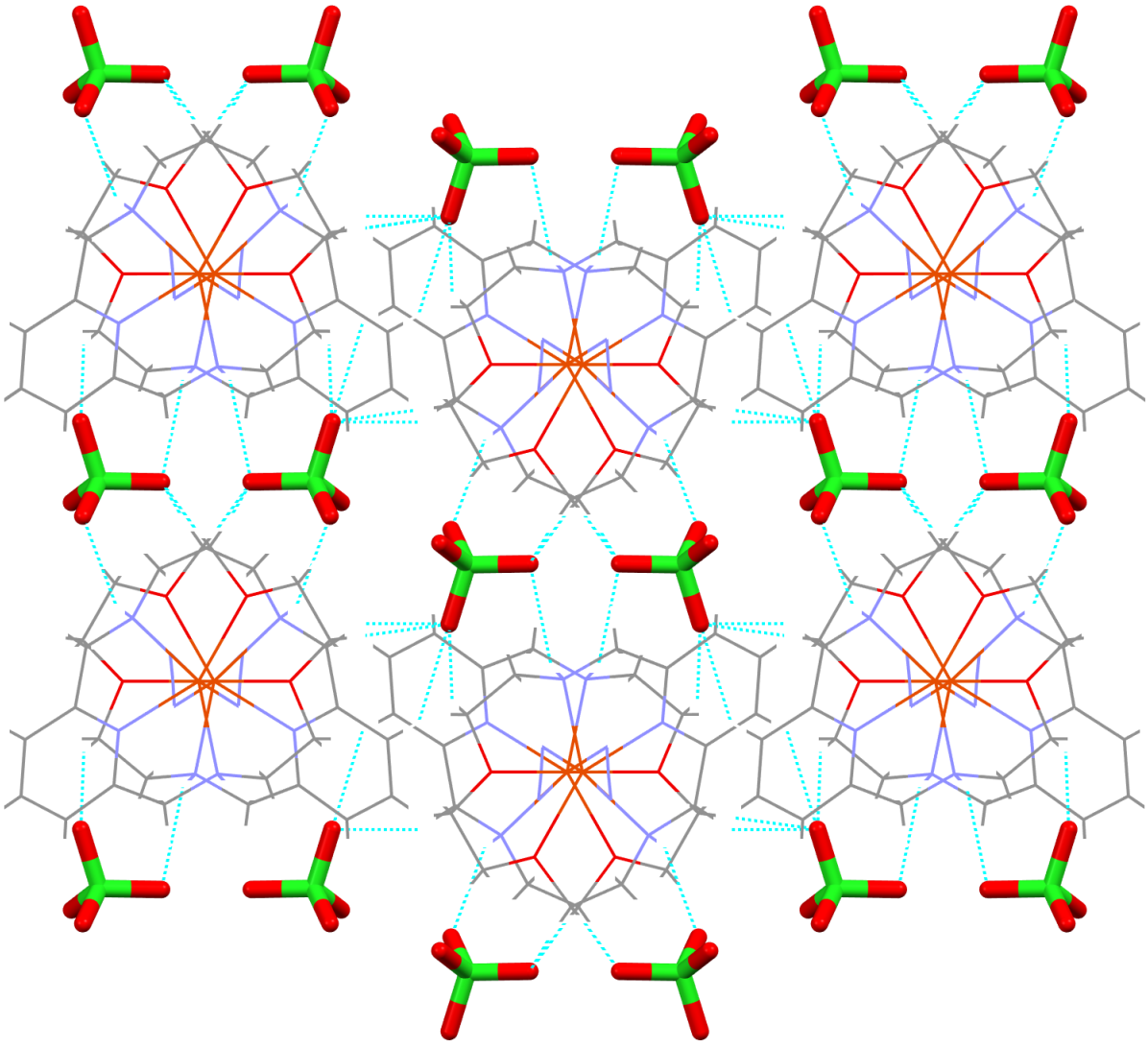

Figure S2. View on the 2D-sheet structure of **1** along the *c* axis, showing some of the H-bonds (blue dashed lines) stabilizing the crystal structure.

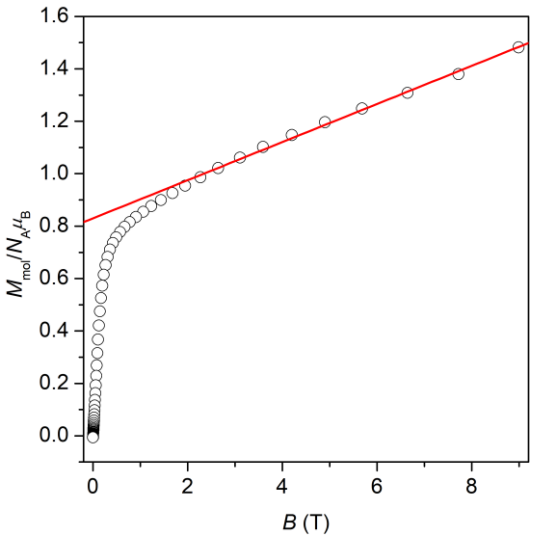

Figure S3. The magnetization data measured at T = 6 K for **1**. The red line corresponds to the extrapolation of the magnetization curve in the high field region down to zero resulting in $M_R/N_A\mu_B$ = 0.83. The canting angle α was estimated using this relationship: $\alpha = \sin^{-1}(M_R/M_S) = \tan^{-1}[0.83/(2.13\cdot 2)]$ = 11°, where M_S is the saturation magnetization calculated as $M_S/N_A\mu_B = g\cdot S$.

Table S1. Values of the Mössbauer hyperfine parameters, derived from the least-square fitting of the Mössbauer spectra of **1**, where T is the temperature of the measurement, δ is the isomer shift, ΔE_Q is the quadrupole splitting, and RA is the relative spectral area of individual spectral components identified during fitting.

T **(K)**	**Component**	$\boldsymbol{\delta}$ **± 0.01 (mm/s)**	$\boldsymbol{\Delta E_Q}$ **± 0.01 (mm/s)**	**RA ± 1 (%)**	**Assignment**
300	Doublet	1.08	1.73	100	Fe(II), S = 2

Table S2. Selected hydrogen bond parameters (Å, °) for complex **1**.

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1N)...O(6)#1	0.84(3)	2.59(3)	3.249(4)	137(3)
N(2)-H(2N)...O(4)#3	0.86(3)	2.43(3)	3.122(3)	138(3)
C(1)-H(2)...O(3)#1	0.99	2.48	3.355(4)	147.5
C(9)-H(4)...O(3)#4	0.99	2.59	3.275(4)	126.7
C(9)-H(5)...O(1)#1	0.99	2.52	3.486(3)	165.0
C(11)-H(9)...O(6)#3	0.99	2.47	3.460(4)	176.0
C(11)-H(14)...O(6)#5	0.99	2.58	3.400(4)	140.4
C(13)-H(11)...O(3)#6	0.99	2.58	3.570(4)	174.7

Symmetry transformations used to generate equivalent atoms: [#1] x,-y+1/2,z+1/2; [#2] x,-y+1/2,z-1/2; [#3] x-1,y,z; [#4] -x+1,-y,-z+1; [#5] x-1,-y+1/2,z+1/2; [#6] -x+1,y+1/2,-z+1/2.

Table S3 Parameters of one-component Debye model for **1**.

T/K	χ_S/(10^{-6} m^3mol^{-1})	χ_T/(10^{-6} m^3mol^{-1})	α	τ/(s)
4.68	0.733	8.885	0.193	0.071664
5.07	0.709	11.583	0.149	0.019919
5.47	0.716	15.821	0.117	0.006221
5.86	0.637	21.749	0.101	0.002074
6.26	0.729	24.372	0.085	0.000718
6.66	0.815	22.914	0.076	0.000253

Table S4 Energy levels of the ligand field terms in zero magnetic field derived from CASSCF/NEVPT2//def2-TZVP(-f) calculations for **1**.

State	Multiplicity	Energy (cm^{-1})
0	5	0
1	5	547.7
2	5	4193.5
3	5	7542.0
4	5	9392.3
5	3	13176.4
6	3	17236.9
7	3	18480.1
8	3	19313.8
9	3	19582.4
10	3	19911.3
11	3	20513.9
12	3	21010.3
13	3	22038.8
14	3	22531.2
15	3	22694.7
16	3	24213.5
17	3	24356.2
18	3	24760.8
19	3	26181.6
20	3	26499.9
21	3	26830.4
22	3	26938.6
23	3	27192.5
24	3	28639.6
25	3	28956.8
26	3	29402.9
27	3	29538.9
28	3	31703.2
29	3	31784.7
30	3	32212.8

31	3	33103.3
32	3	33467.9
33	3	33931.5
34	3	34059.3
35	3	35151.6
36	3	35492.4
37	3	35636.0
38	3	37724.7
39	3	38598.5
40	3	52448.7
41	3	53425.6
42	3	53508.1
43	3	53534.3
44	3	54933.8
45	3	55484.9
46	3	55532.2
47	3	57876.4
48	3	58220.4
49	3	58539.1

Table S5 Energy levels of the ligand field multiplets in zero magnetic field derived from CASSCF/NEVPT2/def2-TZVP(-f) calculations for **1**.

State	Energy (cm^{-1})
0	0.00
1	0.47
2	29.17
3	38.38
4	47.39
5	588.71
6	600.10
7	626.11
8	673.01
9	675.00
10	4208.40
11	4208.42
12	4262.86
13	4264.54
14	4282.16
15	7620.77
16	7627.98
17	7629.97
18	7653.09
19	7653.10
20	9468.50
21	9468.56
22	9472.43
23	9473.38
24	9474.39
25	13239.92
26	13262.69
27	13263.83
28	17285.09

29	17293.75
30	17332.34
31	18557.14
32	18559.88
33	18571.53
34	19363.35
35	19368.46
36	19385.08
37	19652.64
38	19663.17
39	19675.35
40	19977.18
41	19984.19
42	20004.53
43	20554.96
44	20582.63
45	20588.80
46	21085.74
47	21098.19
48	21117.52
49	22091.25
50	22126.29
51	22144.23
52	22434.63
53	22450.23
54	22607.20
55	22766.69
56	22954.36
57	22988.99
58	24108.79
59	24157.14
60	24247.87
61	24390.77
62	24520.99
63	24550.80
64	24857.00
65	24915.16
66	24977.59
67	26276.25
68	26283.11
69	26330.16
70	26468.67
71	26494.91
72	26576.19
73	26869.89
74	26900.12
75	26908.98
76	27071.21
77	27076.60
78	27141.61
79	27326.91
80	27379.70
81	27395.76
82	28721.45
83	28733.69
84	28742.67

85	29088.47
86	29110.74
87	29131.20
88	29283.67
89	29309.99
90	29489.53
91	29644.25
92	29893.58
93	29904.50
94	31607.64
95	31676.54
96	31740.38
97	31871.19
98	31999.52
99	32059.52
100	32281.82
101	32372.89
102	32389.30
103	33171.38
104	33173.10
105	33225.39
106	33538.14
107	33540.03
108	33563.69
109	33787.56
110	33812.10
111	34130.22
112	34249.46
113	34437.93
114	34468.77
115	35221.72
116	35226.92
117	35276.27
118	35570.52
119	35641.63
120	35662.74
121	35743.20
122	35778.91
123	35782.16
124	37826.23
125	37827.74
126	37828.18
127	38700.05
128	38701.07
129	38701.55
130	52452.26
131	52507.41
132	52517.92
133	53402.68
134	53469.04
135	53541.26
136	53559.32
137	53566.07
138	53637.82
139	53658.97
140	53797.27

141	53805.78
142	55038.22
143	55044.43
144	55045.73
145	55589.47
146	55590.33
147	55596.00
148	55642.36
149	55645.12
150	55648.92
151	57887.79
152	57926.54
153	57954.22
154	58258.19
155	58378.58
156	58398.62
157	58689.56
158	58730.11
159	58745.07

Table S6. Individual contributions to *D*-tensor for **1** calculated by CASSCF/NEVPT2/def2-TZVP(-f).

Multiplicity	Root	*D*	*E*
5	0	-0.000	-0.000
5	1	-15.618	-0.029
5	2	0.684	0.880
5	3	0.398	-0.229
5	4	1.772	-1.830
3	0	-0.994	-1.032
3	1	0.636	-0.018
3	2	0.027	-0.000
3	3	0.047	-0.040
3	4	0.009	-0.018
3	5	-0.000	-0.002
3	6	-0.088	-0.091
3	7	0.194	0.001
3	8	-0.100	0.085
3	9	1.570	-0.004
3	10	0.742	0.000
3	11	-0.007	-0.038
3	12	-0.006	-0.090
3	13	0.029	-0.014
3	14	-0.011	-0.002
3	15	-0.113	-0.195
3	16	0.168	-0.104
3	17	0.061	-0.072
3	18	-0.356	-0.356
3	19	-0.225	0.201
3	20	-0.030	-0.030
3	21	-0.272	0.255
3	22	-0.186	-0.162
3	23	0.000	-0.000
3	24	-0.002	0.005
3	25	0.008	-0.001
3	26	-0.002	0.002
3	27	-0.003	-0.018
3	28	0.156	0.002
3	29	-0.001	0.007

3	30	-0.003	0.000
3	31	0.219	0.000
3	32	-0.097	-0.107
3	33	-0.080	0.056
3	34	-0.311	0.321
3	35	0.015	0.000
3	36	-0.004	0.004
3	37	-0.009	0.002
3	38	-0.006	-0.005
3	39	-0.011	-0.008
3	40	-0.001	-0.000
3	41	-0.006	-0.005
3	42	-0.007	-0.034
3	43	0.050	-0.009
3	44	0.046	-0.037