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Supporting Information

Expanding dimensionality of bis(tetrazolyl)alkane-based Fe(II) coordination polymers by an application of dinitrile coligands

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Table S1. Crystallographic data for crystal structures of **1-4** determined at different temperatures.

	80 K [1] ebtz/ADN	**250 K [1] ebtz/ADN**	**80 K [2] hbtz/ADN**	**300 K [2] hbtz/ADN**	**80 K [3] 1ditz/GLN**	**200 K [3] 1ditz/GLN**	**100 K [4] 1ditz/SUN**	**200 K [4] 1ditz/SUN**
CCDC number	2332601	2332602	2332603	2332604	2332605	2332606	2332607	2332608
Chemical formula	$C_{14}H_{20}B_2F_8FeN_{18}$	$C_{14}H_{20}B_2F_8FeN_{18}$	$C_{22}H_{36}B_2F_8FeN_{18}$	$C_{22}H_{36}B_2F_8FeN_{18}$	$C_{84}H_{104}B_8F_{32}Fe_4N_{88}$	$C_{84}H_{104}B_8F_{32}Fe_4N_{88}$	$C_{22}H_{32}B_2F_8FeN_{20}$	$C_{22}H_{32}B_2F_8FeN_{20}$
Formula mass	669.95	669.95	782.16	782.16	3264.43	3264.43	806.14	806.14
Crystal system, space group	triclinic *P-1*	triclinic *P -1*	triclinic *P -1*	triclinic *P -1*	triclinic *P -1*	triclinic *P -1*	monoclinic *P* $2_1/c$	monoclinic *P* $2_1/c$
Z	1	1	1	1	2	2	4	4
***a* [Å]**	7.9876(3)	8.0729(6)	8.7981(15)	9.0817(13)	18.9389(4)	19.0780(4)	13.8300(3)	14.0436(4)
***b* [Å]**	8.6965(3)	8.7297(7)	9.9945(19)	10.250(2)	19.4264(3)	19.7385(3)	13.8083(3)	14.0679(3)
***c* [Å]**	10.1798(3)	10.3033(6)	11.413(2)	11.7285(19)	19.5685(3)	19.8266(3)	18.0906(4)	18.3229(5)
***α* [°]**	90.210(3)	89.886(6)	109.557(18)	110.127(13)	90.0460(10)	90.0320(10)	90	90
***β* [°]**	91.318(3)	91.310(5)	99.300(16)	99.834(13)	104.760(2)	104.745(2)	94.9421(19)	94.555(3)
***γ* [°]**	110.791(3)	111.316(7)	112.580(17)	113.172(13)	103.591(2)	103.643(2)	90	90
V [Å^3]	660.86(4)	676.25(9)	823.2(3)	881.1(3)	6752.9(2)	7001.9(2)	3441.90(12)	3608.53(16)
Absorption coefficient [mm^{-1}]	0.672	0.657	0.552	0.516	0.545	0.526	0.532	0.508
No. of measured reflections	5551	5635	6790	8921	27531	79839	52164	54481
No. of independent reflections	2685	2760	3363	4549	27531	28534	16003	16742
Theta range for data collection [º]	2.978 to 26.366	3.181 to 26.372	3.202 to 26.369	3.335 to 28.699	1.768 to 26.372	1.750 to 26.372	2.983 to 36.488	3.103 to 36.526
Index ranges	$-8 \le h \le 9$ $-10 \le k \le 10$ $-12 \le l \le 12$	$-9 \le h \le 10$ $-10 \le k \le 10$ $-12 \le l \le 12$	$-8 \le h \le 10$ $-12 \le k \le 12$ $-14 \le l \le 14$	$-12 \le h \le 12$ $-8 \le k \le 13$ $-15 \le l \le 15$	$-23 \le h \le 22$ $-24 \le k \le 24$ $0 \le l \le 24$	$-23 \le h \le 23$ $-24 \le k \le 24$ $-24 \le l \le 24$	$-23 \le h \le 22$ $-23 \le k \le 16$ $-27 \le l \le 29$	$-23 \le h \le 23$ $-23 \le k \le 16$ $-28 \le l \le 29$
R_{int}	0.0166	0.0258	0.0275	0.0285	0.0492	0.0493	0.0389	0.0424

Final R_1 values ($I > 2\sigma(I)$)	0.0244	0.0355	0.0355	0.0493	0.0687	0.0928	0.0472	0.0500
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.0591	0.0904	0.0862	0.1243	0.1691	0.2338	0.1079	0.1150
Final R_1 values (all data)	0.0273	0.0409	0.0456	0.0689	0.0954	0.1280	0.0717	0.1010
Final $wR(F^2)$ values (all data)	0.0606	0.0954	0.0912	0.1387	0.1815	0.2531	0.1251	0.1463
Goodness-of-fit on F^2	1.048	1.056	1.041	1.023	1.026	1.053	1.036	1.017
Largest diff. peak and hole [eÅ^{-3}]	0.372 and -0.234	0.367 and -0.503	0.482 and -0.262	0.382 and -0.336	1.318 and -0.675	1.346 and -1.311	1.103 and -0.908	0.505 and -0.701

Table S2. Fe-N distances, as well as selected Fe…Fe distances and torsional angles for **1** (ebtz/ADN) at 80 K and 240 K.

	80 K [1] ebtz/ADN	**250 K [1] ebtz/ADN**
Fe1-N4 [Å]	2.1584(12)	2.1680(15)
Fe1-N8 [Å]	2.1908(12)	2.1983(16)
Fe1-N9 [Å]	2.1568(12)	2.1681(15)
Fe1-ebtz-Fe1[a] [Å]	8.6965(3)	8.7297(7)
Fe1-ADN-Fe1[b] [Å]	10.1798(3)	10.3033(6)
Fe1···Fe1[c] [Å]	9.4930(4)	9.4944(10)
Fe1···Fe1[d] [Å]	7.9876(2)	8.0729(6)
Fe1···Fe1[e] [Å]	13.3644(4)	13.5175(9)
Fe1···Fe1[f] [Å]	13.4130(4)	13.4910(9)
Fe1-N9-C5 [°]	160.91(11)	161.76(15)
C5-C6-C7-C7[g] [°]	59.87(19)	60.8(4)
N1-N2-C3-C4 [°]	-74.74(16)	-75.8(2)
N3-N2-C3-C4 [°]	100.55(15)	101.3(2)
N2-C3-C4-N6[h] [°]	66.90(15)	67.6(2)
C3-C4-N6[h]-N5[h] [°]	62.83(17)	62.8(2)
C3-C4-N6[h] -N7[h] [°]	-120.67(14)	-120.01(19)

[a] 1-x, 1-y, 1-z; [b] x, y, 1+z; [c] 2-x, 1-y, 1-z; [d] 1+x, y, z [e] 1-x, -1-y, -z; [f] 1-x, 1-y, -z; [g] 1-x, -y, -z; [h] x, -1+y, z; [i] x, 1+y, z.

Table S3. Fe-N distances, as well as selected Fe…Fe distances and torsional angles for **2** (hbtz/ADN) at 80 K and 300 K.

	80 K [2] hbtz/ADN	**300 K [2] hbtz/ADN**
Fe1-N4 [Å]	1.9660(18)	2.1564(19)
Fe1-N8 [Å]	1.9796(16)	2.1702(17)
Fe1-N9 [Å]	1.9216(17)	2.1200(19)
Fe1-hbtz-Fe1[a] [Å]	11.413(2)	11.7285(19)
Fe1-hbtz-Fe1[b] [Å]	12.401(3)	12.645(3)
Fe1-ADN-Fe1[c] [Å]	10.477(3)	10.690(2)
Fe1···Fe1[d] [Å]	8.7981(15)	9.0817(13)
Fe1···Fe1[e] [Å]	9.9945(19)	10.250(2)
Fe1···Fe1[f] [Å]	11.455(5)	11.521(4)
Fe1···Fe1[g] [Å]	16.941(6)	17.086(5)
Fe1-N9-C9 [°]	176.3(3)	177.82(19)
C9-C10-C11-C11[h] [°]	-61.2(3)	-62.0(7)
C9-C10-C11A-C11A[h] [°]	-	62.2(9)
N1-N2-C2-C3 [°]	-66.3(2)	-66.1(4)
N3-N2-C2-C3 [°]	109.3(2)	109.5(3)
N2-C2-C3-C4 [°]	-56.3(2)	-55.6(4)
N2-C2-C3-C4A [°]	-	-68.9(9)
N5-N6-C6-C7 [°]	73.7(2)	73.4(3)
N7-N6-C6-C7 [°]	-100.3(2)	-101.1(3)
N6-C6-C7-C8 [°]	61.5(2)	63.1(4)

[a] x, y, 1+z; [b] x, 1+y, 1+z; [c] 1+x, 1+y, z; [d] 1-x, -y, 1-z; [e] x, 1+y, z; [f] 1+x, 1+y,1+ z; [g] 1+x, 2+y, 1+z; [h] 1-x, 1-y, 1-z.

Table S4. Fe-N distances, as well as selected Fe…Fe distances and torsional angles for **3** (1ditz/GLN) at 80 K and 200 K.

	80 K [3] 1ditz/GLN	**200 K [3] 1ditz/GLN**
Fe1-N4 [Å]	2.046(4)	2.176(5)
Fe1-N12 [Å]	2.028(4)	2.170(5)
Fe1-N20 [Å]	2.032(4)	2.160(5)
Fe1-N28 [Å]	2.043(4)	2.187(5)
Fe1-N33 [Å]	2.001(4)	2.173(6)
Fe1-N35 [Å]	2.002(5)	2.174(5)
Fe2-N8[a] [Å]	2.039(4)	2.178(5)
Fe2-N32 [Å]	2.048(4)	2.173(5)
Fe2-N40 [Å]	2.030(4)	2.170(5)
Fe2-N48 [Å]	2.023(4)	2.161(5)
Fe2-N53 [Å]	1.988(4)	2.169(5)
Fe2-N55 [Å]	1.965(4)	2.144(5)
Fe3-N44 [Å]	2.067(4)	2.185(5)
Fe3-N52[b] [Å]	2.079(4)	2.188(5)
Fe3-N60[Å]	2.061(4)	2.168(5)
Fe3-N68 [Å]	2.070(4)	2.189(5)
Fe3-N73 [Å]	2.023(4)	2.151(5)
Fe3-N75 [Å]	2.016(4)	2.158(6)
Fe4-N16[a] [Å]	2.170(4)	2.193(5)
Fe4-N24[c] [Å]	2.161(4)	2.188(5)
Fe4-N64[a] [Å]	2.158(4)	2.193(5)
Fe4-N72 [Å]	2.158(4)	2.169(5)
Fe4-N77 [Å]	2.136(4)	2.146(5)
Fe4-N79 [Å]	2.153(4)	2.176(6)
Fe1-1ditz-Fe2 [Å]	9.8108(9)	9.9448(11)
Fe1-1ditz-Fe2[d] [Å]	9.7666(9)	9.8865(11)
Fe1-1ditz-Fe4[d] [Å]	9.6251(9)	9.7577(10)
Fe1-1ditz-Fe4[e] [Å]	9.8030(9)	9.9870(10)
Fe2-1ditz-Fe3 [Å]	9.7937(8)	9.9573(10)
Fe2-1ditz-Fe3[f] [Å]	9.6341(9)	9.7848(10)
Fe2-1ditz-Fe1[a] [Å]	9.7666(9)	9.8865(11)
Fe3-1ditz-Fe4 [Å]	9.8171(9)	9.9583(12)
Fe3-1ditz-Fe4[d] [Å]	9.7531(9)	9.8683(11)
Fe1···Fe4 [Å]	21.8334(9)	22.1980(11)
Fe1···Fe1[a] [Å]	19.5685(3)	19.8266(2)
Fe1···Fe3 [Å]	13.6397(9)	13.9456(11)
Fe1···Fe3[e] [Å]	13.9487(9)	14.0433(11)
Fe1···Fe3[f] [Å]	13.9224(9)	14.1223(11)
Fe2···Fe4 [Å]	13.9327(9)	14.1212(11)
Fe2···Fe4[e] [Å]	13.6538(9)	13.8665(11)
Fe2···Fe4[f] [Å]	13.6410(9)	13.9690(11)
Fe1-N33-C13 [°]	172.7(4)	170.5(5)
Fe1-N35-C18 [°]	176.0(4)	172.9(6)
Fe2-N53-C29 [°]	173.2(4)	168.8(5)

Fe2-N55-C34 [°]	173.0(4)	168.2(6)
Fe3-N73-C45 [°]	160.3(4)	155.0(5)
Fe3-N75-C50 [°]	167.2(4)	165.3(5)
Fe4-N77-C55 [°]	149.4(4)	154.8(5)
Fe4-N79-C60 [°]	170.7(4)	167.2(6)
Σ_{Fe1} [°]	6.2	10.8
Σ_{Fe2} [°]	7.9	11.0
Σ_{Fe3} [°]	18.1	19.9
Σ_{Fe4} [°]	12.2	13.5

[a] x,y,1+z; [b] x,-1+y,z; [c] x,-1+y,1+z; [d] x,y,-1+z; [e] x, 1+y, -1+z; [f] x, 1+y, z

Table S5. Fe-N distances, as well as selected Fe…Fe distances and torsional angles for **4** (1ditz/SUN) at 100 K and 200 K.

	100 K [4] 1ditz/SUN	**200 K [4] 1ditz/SUN**
Fe1-N4 [Å]	1.9954(11)	2.1667(12)
Fe1-N8 [Å]	1.9997(11)	2.1723(13)
Fe1-N12 [Å]	2.0004(11)	2.1731(13)
Fe1-N16 [Å]	1.9995(11)	2.1709(13)
Fe1-N17 [Å]	1.9371(12)	2.1599(14)
Fe1-N18 [Å]	1.9351(12)	2.1370(14)
Fe1-1ditz-Fe1[a] [Å]	9.7422(3)	9.9124(3)
Fe1-1ditz -Fe1[b] [Å]	9.8015(3)	9.9661(3)
Fe1-SUN-Fe1[c] [Å]	11.9229(4)	12.2279(5)
Fe1-SUN-Fe1[d] [Å]	11.8685(4)	12.1574(5)
Fe1···Fe1[e] [Å]	9.2963(2)	9.3711(3)
Fe1···Fe1[f] [Å]	13.8300(3)	14.0436(4)
Fe1···Fe1[g] [Å]	13.8083(3)	14.0679(3)
Fe1···Fe1[h] [Å]	14.7593(4)	15.1746(4)
Fe1···Fe1[i] [Å]	16.0042(3)	16.2667(5)
Fe1···Fe1[j] [Å]	19.5432(3)	19.8779(3)
Fe1···Fe1[k] [Å]	19.6863(4)	20.1756(5)
Fe1-N17-C7 [°]	177.01(11)	172.84(13)
Fe1-N18-C11 [°]	172.61(11)	173.16(13)
C7-C8-C9-C10 [°]	-65.34(16)	-65.1(2)
C8-C9-C10-C10[d] [°]	-178.97(14)	-179.37(18)
C11-C12-C13-C14 [°]	-61.42(17)	-61.8(2)
C12-C13-C14-C14[c] [°]	-173.21(15)	-174.30(17)
N1-C3-N5[m]-N6[m] [°]	-74.54(17)	-99(2)
N1-C3-N5[m] -C2[m] [°]	110.49(16)	99.5(2)
C1-N1-C3-N5[m] [°]	102.63(16)	98.78(19)
N2-N1-C3-N5[m] [°]	-78.71(16)	-83.17(19)
N9-C6-N13[a]-N14[a] [°]	95.42(16)	98.30(19)
N9-C6-N13[a] -C5[a] [°]	-91.33(17)	-88.0(2)
C4-N9-C6-N13[a] [°]	-97.41(17)	-90.9(2)
N10-N9-C6-N13[a] [°]	88.50(16)	88.0(7)

[a] 2-x, -1/2+y, 1/2-z; [b] 1-x, -1/2+y, 1/2-z; [c] 2-x, -y, 1-z; [d] 1-x, -y, -z; [e] x, 1/2-y, 1/2+z; [f] 1+x, y, z; [g] x, -1+y, z; [h] x, -1/2-y, 1/2+z; [i] 1+x, 1/2-y, 1/2+z; [j] -1+x, -1+y, z; [k] 1+x, -1/2-y, 1/2+z; [l] 1+x, -1+y, z; [m] 1-x, 1/2+y, 1/2-z.

Table S6. Selected C-H···N and C-H···F interatomic contacts [Å] and angles [°] for **1** (ebtz/ADN) at 80 K and 250 K. D-H…A distances longer than 3.5 Å and angles lesser than 110° were included only for facilitate comparison of changes.

	80 K [1] ebtz/ADN		**250 K [1] ebtz/ADN**	
	d(D..A)	<DHA	d(D..A)	<DHA
C1-H1...F2[a]	3.0534(16)	121.0	3.059(2)	123.1
C2-H2...F2[b]	3.1360(17)	147.4	3.155(3)	144.2
C3-H3A...F4[c]	3.3666(17)	134.2	3.438(3)	134.3
C3-H3B...F4[b]	3.2737(17)	129.9	3.333(3)	129.2
C4-H4A...F1[c]	3.2985(17)	161.4	3.351(3)	160.6
C4-H4A...F3[c]	3.2166(17)	128.8	3.249(3)	131.6
C4-H4B...F1[d]	3.2474(17)	163.1	3.295(3)	163.8
C4-H4B...F4 [d]	3.4953(17)	142.4	3.541(3)	143.3
C6-H6B...F3[e]	3.1547(16)	128.2	-	-
C6-H6D...F3[e]	-	-	3.177(2)	141.2
C6-H6C...F1[d]	-	-	3.242(3)	131.5

[a] 1-x, -y, 1-z; [b] -1+x,-1+y, z; [c] 1-x,-y,-z; [d] x,-1+y,z; [e] 2-x,1-y,-z.

Table S7. Selected C-H···N and C-H···F interatomic contacts [Å] and angles [°] for **2** (hbtz/ADN) at 80 K and 300 K. D-H…A distances longer than 3.5 Å and angles lesser than 110° were included only for facilitate comparison of changes.

	80 K [2] hbtz/ADN		**300 K [2] hbtz/ADN**	
	d(D..A)	<DHA	d(D..A)	<DHA
C1-H1...F4[a]	3.010(2)	158.7	3.050(7)	166.9
C2-H2A...F1[b]	3.294(3)	175.8	3.366(8)	176.4
C2-H2B...F3[c]	3.244(3)	119.0	3.427(4)	125.5
C2-H2B...F4[c]	3.359(3)	140.3	3.483(8)	152.5
C4-H4B...F1[d]	3.468(3)	143.2	-	-
C5-H5...F2[e]	3.195(3)	143.8	3.508(8)	148.9
C5-H5...F3[e]	3.260(3)	146.0	3.331(4)	152.1
C5-H5...N3[f]	3.283(3)	118.8	3.553(3)	118.3
C6-H6A...N2[g]	3.416(3)	136.6	3.596(3)	136.0
C6-H6B...F3	3.215(3)	119.5	3.534(5)	121.3
C8-H8A...F1[d]	3.241(3)	121.0	3.340(7)	127.9
C10-H10A...F2[h]	3.424(3)	150.2	-	-
C11-H11A...F4	3.241(3)	122.9	-	-

[a] -1+x,-1+y,z; [b] -1+x,y,z; [c] -x,-y,-z; [d] 1-x,-y,-z; [e] 1-x,-y,1-z; [f] -x,-y,1-z; [g] 1+x,y,z; [h] 1-x,1-y,1-z.

Table S8a. Selected C-H···N and C-H···F interatomic contacts [Å] and angles [°] for **3** (1ditz/GLN) at 80 K and 200 K. D-H···A distances longer than 3.5 Å and angles lesser than 110° were included only for facilitate comparison of changes.

	80 K		200 K			80 K		200 K	
Fe1	d(D···A)	<DHA	d(D···A)	<DHA	**Fe3**	d(D···A)	<DHA	d(D···A)	<DHA
Donor – tetrazole ring linked to Fe1					**Donor – tetrazole ring linked to Fe3**				
C1-H1...N56[b] (dinitrile coordinated to Fe2)	3.786(9)	126.2	3.480(2)	136.6	C24-H24...N34[a] (dinitrile coordinated to Fe1)	3.465(7)	133.7	3.391(12)	135.7
C4-H4...N78[a] (dinitrile coordinated to Fe4)	3.256(7)	139.0	3.327(10)	141.0	C42-H42...N36[f] (dinitrile coordinated to Fe1)	3.323(7)	154.3	3.319(11)	158.5
C7-H7...N82[b] (noncoordinated dinitrile)	3.208(7)	161.0	3.253(11)	166.4	C27-H27...N81[b] (noncoordinated dinitrile)	3.267(7)	155.7	3.333(11)	154.0
C10-H10...N83[f] (noncoordinated dinitrile)	3.246(7)	153.5	3.316(11)	156.6	C39-H39...N84[f] (noncoordinated dinitrile)	3.212(7)	162.1	3.267(11)	163.6
Donor - dinitrile coordinated to Fe1					**Donor - dinitrile coordinated to Fe3**				
C14-H14A...N66[a] (Tz coordinated to Fe3)	3.504(7)	137.8	3.474(9)	135.9	C41-H41B...N76[f] (dinitrile coordinated to Fe3)	3.346(8)	163.8	3.410(3)	150.3
C19-H19B...N46[b] (Tz coordinated to Fe2)	3.629(8)	128.4	3.499(12)	141.0	C46-H46B...N30[a] (Tz coordinated to Fe2)	3.111(6)	116.9	3.165(9)	126.8
C19-H19A...N81 (noncoordinated dinitrile)	3.283(9)	121.0	3.338(16)	116.3	C48-H48A...N51[k] (Tz coordinated to Fe3)	3.455(7)	135.6	3.503(11)	135.6
C19-H19B...N82[b] (noncoordinated dinitrile)	3.377(9)	133.3	3.516(16)	125.4	C51-H51B...N10[f] (Tz coordinated to Fe1)	3.245(7)	122.3	3.376(11)	136.6
C15-H15B...F22A	-	-	3.25(4)	155.9	C51-H51B...N11[f] (Tz coordinated to Fe1)	3.116(7)	101.7	3.166(10)	114.7
C21-H21A...F4	3.266(9)	148.7	3.75(3)	142.0	C52-H52B...N10[f] (Tz coordinated to Fe1)	3.391(8)	129.1	3.422(18)	127.6
					C52-H52B...N11[f] (Tz coordinated to Fe1)	3.391(8)	118.6	3.375(18)	126.3
					C46-H46B...N83[j] (noncoordinated dinitrile)	3.209(7)	131.3	3.209(12)	127.7
					C51-H51A...N81[f] (noncoordinated dinitrile)	3.412(8)	128.6	3.410(14)	134.7
					C41-H41A...F18	3.303(13)	139.4	3.224(11)	132.2
					C47-H47B...F10a	3.434(7)	167.3	3.461(13)	172.3
					C48-H48A...F26	3.458(6)	141.3	3.718(14)	144.1

	80 K		200 K			80 K		200 K	
Donor - CH_2 from 1ditz bridging Fe1 and Fe2	d(D···A)	<DHA	d(D···A)	<DHA	**Donor - CH_2 from 1ditz bridging Fe3 and Fe2**	d(D···A)	<DHA	d(D···A)	<DHA
C3-H3A...N56[b] (dinitrile coordinated to Fe2) C3-H3A...N56A[b] ((dinitrile coordinated to Fe2)	3.231(8) -	159.6 -	3.420(2) 3.19(5)	144.1 164.4	C25-H25D...N34[a] (dinitrile coordinated to Fe1)	3.340(8)	135.8	3.365(12)	135.3
C3-H3B...F28[c]	2.911(6)	115.5	2.915(8)	112.6	C25-H25C...F1[h]	2.937(6)	118.9	2.972(8)	112.8
C12-H12A...F20	3.229(11)	125.1	3.258(11)	126.7	C25-H25A...F22	2.990(6)	127.3	3.003(10)	123.7
C12-H12B...F8	2.962(6)	118.2	2.948(8)	119.9	C28-H28B...F14	3.044(6)	132.8	2.942(12)	118.8
					C28-H28B...F13A	-	-	3.093(10)	136.9

	80 K		200 K			80 K		200 K	
Donor - CH_2 from 1ditz bridging Fe1 and Fe4	d(D···A)	<DHA	d(D···A)	<DHA	**Donor - CH_2 from 1ditz bridging Fe3 and Fe4**	d(D···A)	<DHA	d(D.···A)	<DHA
C6-H6A...N78[a] (dinitrile coordinated to Fe4)	3.449(7)	143.6	3.504(11)	144.5	C41-H41B...N76[f] (dinitrile coordinated to Fe3) C41-H41B...N76A[f] (dinitrile coordinated to Fe3)	3.346(8) -	163.8 -	3.410(3) 3.28(6)	150.3 166.5
C9-H9B...N54[e] (dinitrile coordinated to Fe2) C9-H9B...N54A[e] (dinitrile coordinated to Fe2)	3.137(16) 3.38(3)	144.0 135.2	3.248(12) -	141.1 -	C41-H41A...F18 C41-H41A...F18A	3.303(13)	139.4	3.224(11) 3.13(3)	132.2 124.4
C6-H6B...F23[d] C6-H6B...F23A[d]	3.182(6) -	133.6 -	3.213(11) 3.06(5)	135.9 132.7	C44-H44B...F31	2.951(7)	115.7	3.098(13)	130.5
C9-H9A...F11 C9-H9A...F11A	3.216(6) 3.043(15)	128.3 136.9	3.313(10) 3.20(3)	127.2 132.7					
	80 K		200 K			80 K		200 K	
Fe2	d(D···A)	<DHA	d(D···A)	<DHA	**Fe4**	d(D···A)	<DHA	d(D···A)	<DHA
Donor – tetrazole ring linked to Fe2					**Donor – tetrazole ring linked to Fe4**				
C11-H11...N80[g] (dinitrile coordinated to Fe4)	3.233(7)	153.5	3.277(11)	157.7	C8-H8...N54[e] (dinitrile coordinated to Fe2) C8-H8...N54A[e] (dinitrile coordinated to Fe2)	3.204(14) 3.40(3)	126.3 135.4	3.299(12) -	133.1 -
C26-H26...N74a (dinitrile coordinated to Fe3)	3.390(7)	140.5	3.453(11)	139.6	C5-H5...N85 (noncoordinated dinitrile)	3.322(7)	166.5	3.342(10)	166.5
C2-H2...N88[a] (noncoordinated dinitrile) C2-H2...N88A[a] (noncoordinated dinitrile)	3.24(2) 3.27(2)	147.6 158.7	3.300(3) 3.31(5)	153.7 160.2	C43-H43...N87[i] (noncoordinated dinitrile) C43-H43...N87A[i] (noncoordinated dinitrile)	3.310(3) 3.18(5)	163.8 166.2	3.240(2) 3.35(7)	166.7 161.8
C23-H23A...N86[h] (noncoordinated dinitrile)	3.240(7)	148.7	3.296(10)	147.6					
Donor - dinitrile coordinated to Fe2					**Donor - dinitrile coordinated to Fe4**				
C30-H30B...N62[a] (Tz coordinated to Fe4)	3.406(7)	133.2	3.416(10)	134.3	C56-H56A...N2[a] (Tz coordinated to Fe1)	3.143(8)	123.9	3.163(10)	125.8
C32-H32B...N74[a] (dinitrile coordinated to Fe3)	3.166(8)	112.9	3.223(13)	114.3	C58-H58B...N15[h] (Tz coordinated to Fe4)	3.494(9)	145.2	3.535(10)	138.7
C35-H35A...N22[b] (Tz coordinated to Fe4)	3.164(7)	128.9	3.194(13)	134.0	C56-H56A...N88A	-	-	3.17(4)	123.2
C37-H37A...N7[h] (Tz coordinated to Fe2)	3.461(7)	151.3	3.475(10)	161.2	C61-H61A...N85[h] (noncoordinated dinitrile)	3.379(8)	126.8	3.345(13)	130.2
C39-H39...N84[f] (noncoordinated dinitrile)	3.212(7)	162.1	3.267(11)	163.6	C61-H61B...N86[f] (noncoordinated dinitrile)	3.372(9)	122.9	3.408(15)	118.1
C35-H35B...N86[h] (noncoordinated dinitrile)	3.451(8)	137.3	3.490(2)	144.7	C58-H58B...F5[h]	3.337(8)	135.8	3.434(11)	138.7
C31-H31A...F16[e] C31-H31A...F16A[e]	- -	- -	3.127(16) 3.423(14)	127.4 162.4	C62-H62A...F20A[g]	3.23(2)	154.6	-	-
C36-H36B...F29[g]	3.126(10)	139.2	3.447(19)	133.6	C63-H63B...F13[l]	3.388(9)	142.3	3.790(2)	146.8

	80 K		200 K	
Donor - noncoordinated dinitriles	d(D···A)	<DHA	d(D···A)	<DHA
C66-H66A...N26 (Tz coordinated to Fe1)	3.003(11)	118.3	3.214(18)	127.3
C71-H71C...N34[f] (dinitrile coordinated to Fe1)	3.401(10)	120.9	3.040(3)	129.5
C78-H78A...N70[f] (Tz coordinated to Fe4)	3.167(8)	123.1	3.201(12)	120.2
C83-H83A...N54[m] (dinitrile coordinated to Fe2)	-	-	3.420(7)	130.5
C67-H67A...F11A[b]	-	-	3.25(3)	141.5
C68-H68A...F9	3.298(14)	121.7	3.226(15)	137.4
C76-H76A...F21[f]	3.245(7)	129.1	3.244(12)	125.7
C77-H77A...F23[d]	3.406(7)	132.0	3.426(14)	137.9
C78-H78B...F24A[f]	-	-	3.29(5)	148.3
C81-H81A...F31	3.349(15)	130.5	3.410(3)	147.0

[a] -x,1-y,1-z; [b] 1-x,2-y,1-z; [c] x,1+y,-1+z; [d] x,y,-1+z; [e] -x,2-y,1-z; [f] 1-x,1-y,1-z; [g] 1-x,1-y,2-z; [h] x,y,1+z; [i] -x,-y,2-z; [j] -1+x,y,z; [k] x,-1+y,z; [l] x,-1+y,1+z; [m] -x,1-y,2-z.

Table S8b. Amount of interatomic contacts for **3** (involving 1ditz and GLN as donor (DH) and acceptor (A)) at 80 K, 200 K related to the Fe1, Fe2, Fe3 and Fe4 surroundings.

	DH		**A**		**IN TOTAL**	
	80 K	**200 K**	**80 K**	**200 K**	**80 K**	**200 K**
Fe1	6	8	10	10	16	18
Fe2	11	12	5	8	15	19
Fe3	16	14	5	5	19	18
Fe4	10	8	7	5	16	13

Table S9. Selected C-H···N and C-H···F interatomic contacts [Å] and angles [°] for **4** (1ditz/SUN) at 100 K and 200 K. D-H…A distances longer than 3.5 Å and angles lesser than 110° were included only for facilitate comparison of changes.

	100 K [4] 1ditz/SUN		**200 K [4] 1ditz/SUN**	
	d(D..A)	<DHA	d(D..A)	<DHA
C1-H1...N20	3.393(2)	146.4	3.394(3)	151.1
C3-H3A...F1	3.303(6)	125.7	3.279(7)	125.1
C5-H5...N19[b]	3.146(2)	138.7	3.235(3)	141.7
C6-H6A...F3[c]	3.056(6)	127.1	3.142(9)	121.6
C6-H6B...F5[c]	2.9774(19)	133.9	3.041(5)	132.9
C8-H8A...N3[d]	3.1945(19)	122.3	3.230(2)	127.7
C8-H8B...N20	3.443(2)	141.7	3.498(3)	136.7
C12-H12A...N20[a]	3.498(2)	151.3	3.564(3)	148.3
C12-H12B...N19[b]	3.216(2)	115.4	3.337(3)	118.1
C13-H13B...N14[a]	3.288(2)	146.7	3.408(2)	147.6
C13-H13B...N15[a]	3.380(2)	129.4	3.397(2)	129.7
C16-H16B...F4[e]	3.310(12)	145.8	3.409(13)	143.1

[a] x,1/2-y,1/2+z; [b] x,3/2-y,1/2+z; [c] x,-1+y,z; [d] x,1/2-y,-1/2+z; [e] x,3/2-y,-1/2+z; [f] 1-x,-1/2+y,1/2-z.

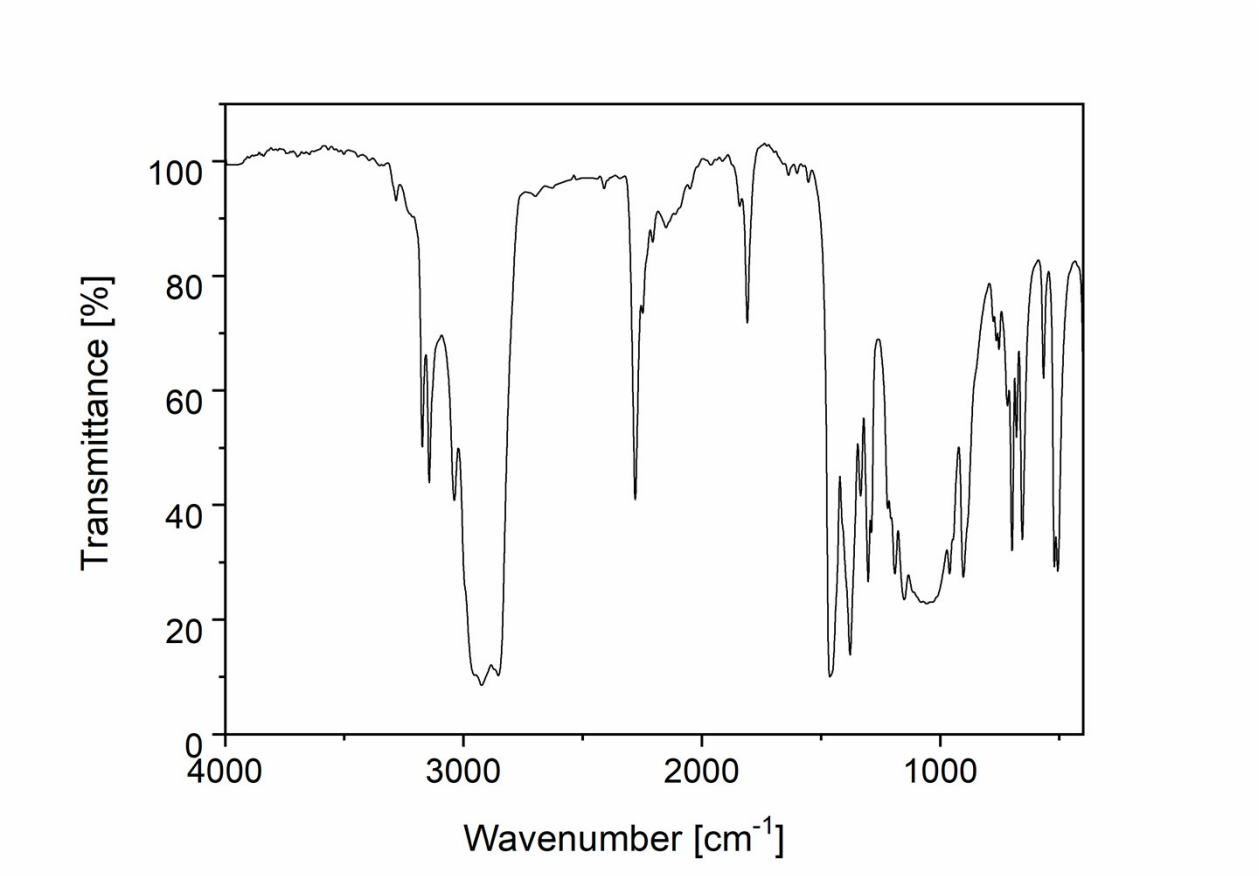

Figure S1. FTIR spectrum of **1** (RT).

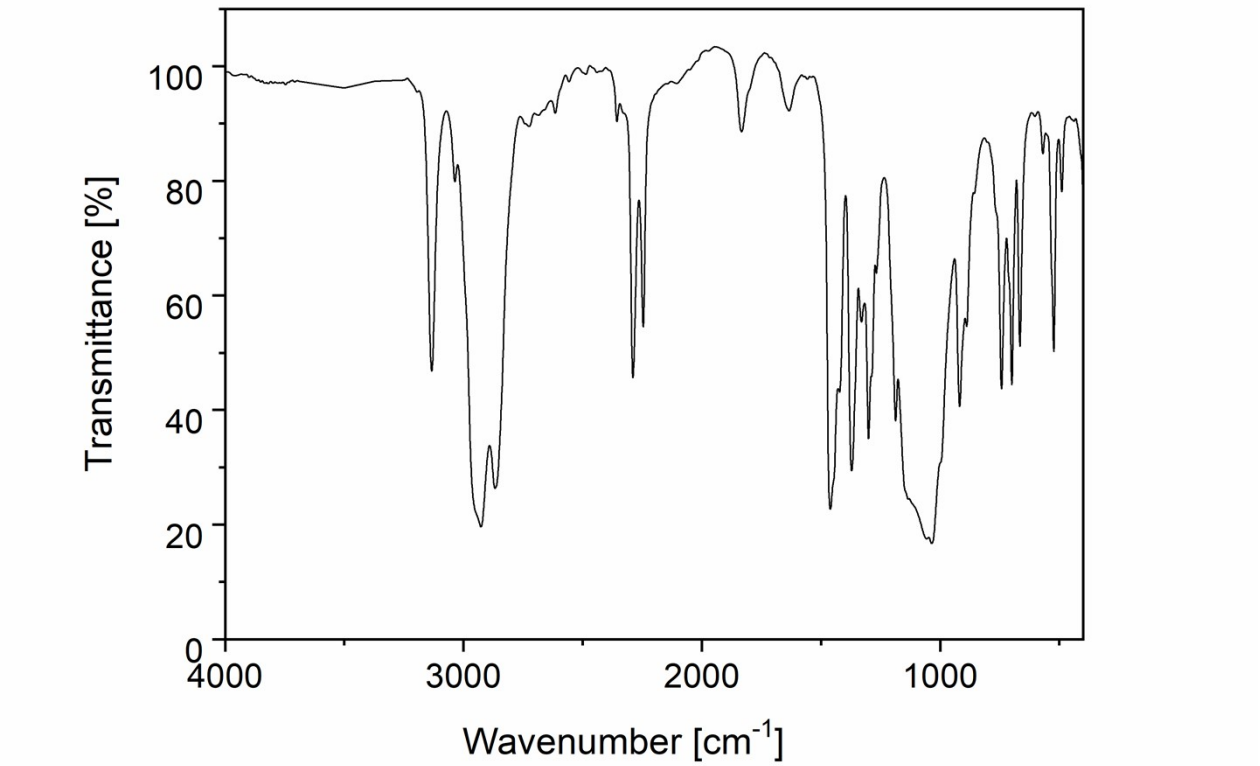

Figure S2. FTIR spectrum of **2** (RT).

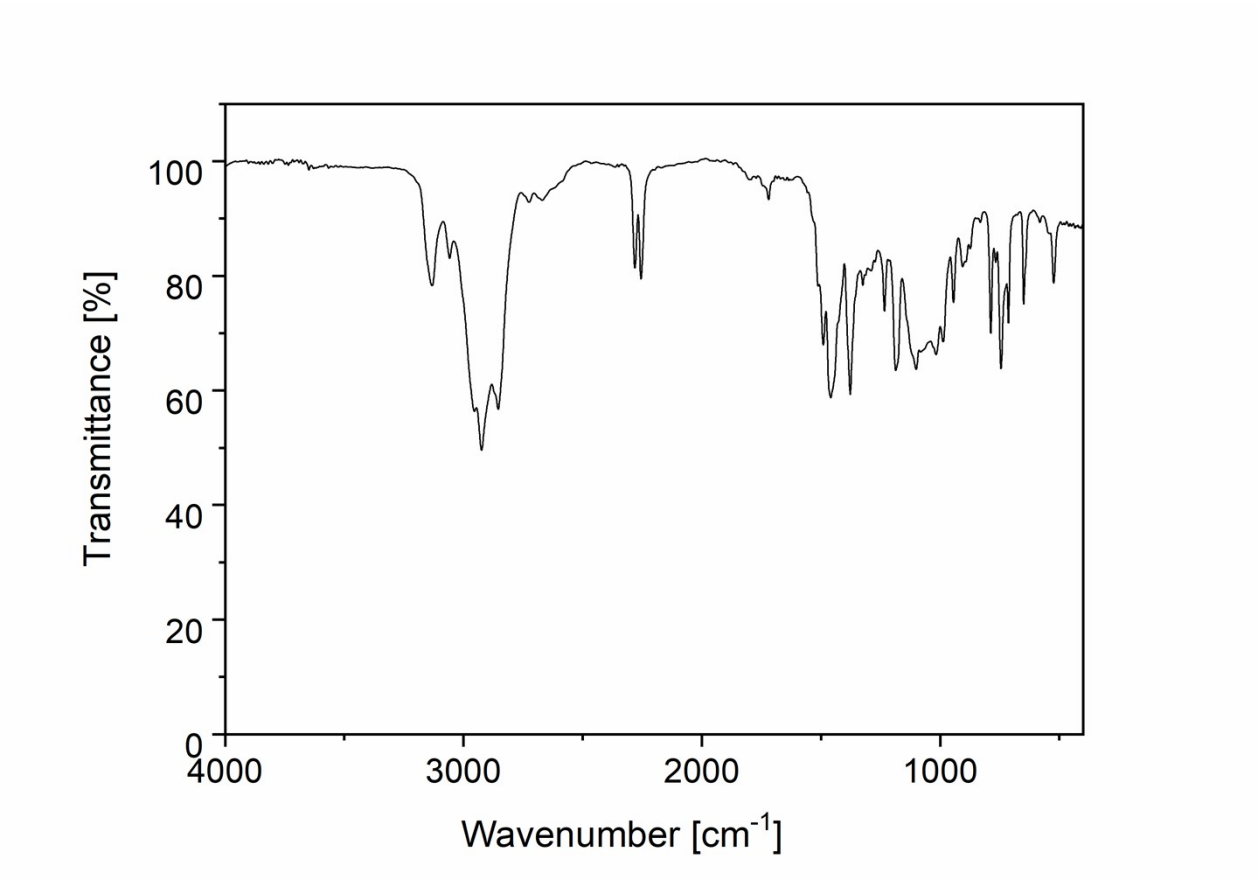

Figure S3. FTIR spectrum of **3** (RT).

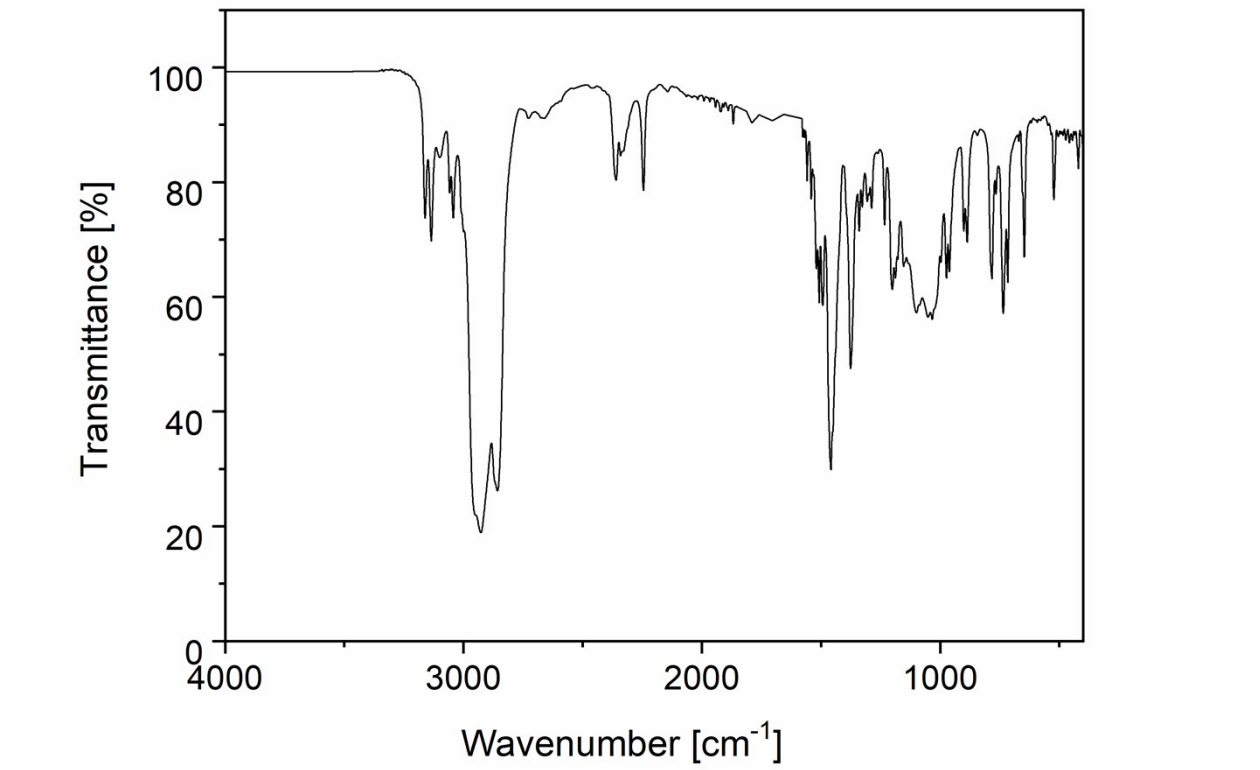

Figure S4. FTIR spectrum of **4** (RT).

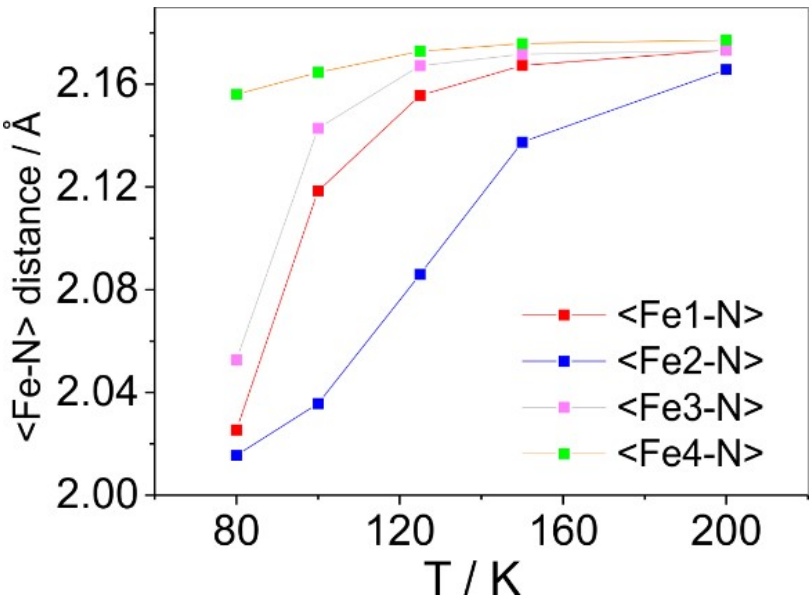

Figure S5. Temperature dependence of average <Fe1-N> (red), <Fe2-N> (blue), <Fe3-N> (pink) and <Fe4-N> (green) distances for individual Fe(II) ions in **3**.