Appendix

1.1 Crystal Structure Determination Details

Table 1
Experimental details of the crystal structure determination of **1**$(BF_4)_2 \cdot H_2O$

	1$(BF_4)_2 \cdot H_2O$
Formula	$C_{27}H_{30}B_2F_8FeN_{10}O$
Formula Weight	740.08
Crystal System	Monoclinic
Space group	*C* 2/c
a (Å)	17.1632(16)
b (Å)	20.9906(19)
c (Å)	19.1771(17)
α (°)	90
β (°)	96.605(5)
γ (°)	90
V (Å^3)	6863.0(11)
D_{calc} (g/cm^3)	1.433
Z	8
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.71073
No. Of reflections	37765
Unique reflections	4550
R_{int}	0.0556
R_1 [$I \geq 2\sigma(I)$][a]	0.068
wR_2 (full set)[b]	0.2322
goodness-of-fit on F^2	1.033
Largest peak (*e* Å^{-3})	0.968
Deepest hole (*e* Å^{-3})	-0.54

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 2
Experimental details of the crystal structure determination of **1**$(ClO_4)_2$.

	1$(ClO_4)_2$
Formula	$C_{26}H_{26}Cl_2FeN_{10}O_8$
Formula Weight	733.32
Crystal System	Trigonal
Space group	$R3_2$
a (Å)	18.6378(12)
b (Å)	18.6378(12)
c (Å)	24.3461(14)
α (°)	90
β (°)	90
γ (°)	120
V (Å^3)	7324.0(8)
D_{calc} (g/cm^3)	1.496
Z	9
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.69
No. Of reflections	23056
Unique reflections	4972
R_{int}	0.0297
R_1 [$I \geq 2\sigma(I)$][a]	0.0607
wR_2 (full set)[b]	0.1708
goodness-of-fit on F^2	1.058
Largest peak (e Å^{-3})	0.678
Deepest hole (e Å^{-3})	-0.547

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 3
Experimental details of the structure determination of **1**$(PF_6)_2$.

	1$(PF_6)_2$
Formula	$C_{26}H_{26}F_{12}FeN_{10}P_2$
Formula Weight	824.36
Crystal System	Monoclinic
Space group	*C*2/*c*
a (Å)	34.124(3)
b (Å)	12.3128(11)
c (Å)	17.6817(17)
α (°)	90
β (°)	114.441(6)
γ (°)	90
V (Å^3)	6763.4(11)
D_{calc} (g/cm^3)	1.619
Z	8
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.643
No. Of reflections	10895
Unique reflections	7634
R_{int}	0.0821
R_1 [$I \geq 2\sigma(I)$][a]	0.0476
wR_2 (full set)[b]	0.1315
goodness-of-fit on F^2	1.023
Largest peak (*e* Å^{-3})	0.778
Deepest hole (*e* Å^{-3})	-0.618

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 4
Experimental details for the structure determination of **2**$(PF_6)_2 \cdot 2CH_3CN$.

	2$(PF_6)_2 \cdot 2CH_3CN$
Formula	$C_{38}H_{48}F_{12}FeN_{12}P_2$
Formula Weight	1018.28
Crystal System	Monoclinic
Space group	$P2_1/n$
a (Å)	20.494(2)
b (Å)	23.257(3)
c (Å)	20.609(2)
α (°)	90
β (°)	101.271(6)
γ (°)	90
V (Å^3)	9633.3(19)
D_{calc} (g/cm^3)	1.389
Z	8
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.71073
No. Of reflections	524484
Unique reflections	17909
R_{int}	0.047
R_1 [$I \geq 2\sigma(I)$][a]	0.074
wR_2 (full set)[b]	0.241
goodness-of-fit on F^2	1.052
Largest peak (e Å^{-3})	1.475
Deepest hole (e Å^{-3})	-1.338

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 5
Experimental details for the structure determination of **3**$(BF_4)_2$.

	3$(BF_4)_2$
Formula	$C_{34}H_{34}B_2F_8FeN_{10}$
Formula Weight	812.18
Crystal System	Cubic
Space group	$I\ \bar{4}_3d$
a (Å)	22.8650(18)
b (Å)	22.8650(18)
c (Å)	22.8650(18)
α (°)	90
β (°)	90
γ (°)	90
V (Å^3)	11954.0(16)
D_{calc} (g/cm^3)	1.354
Z	12
T (K)	150
μ Mo-Kα (mm^{-1})	0.454
No. Of reflections	87527
Unique reflections	2127
R_{int}	0.0409
R_1 [$I \geq 2\sigma(I)$]	0.048
wR_2 (full set)	0.1362
goodness-of-fit on F^2	1.102
Largest peak (e Å^{-3})	0.355
Deepest hole (e Å^{-3})	-0.212

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 6
Experimental details for the structure determination of **4**$(BF_4)_2$.

	4$(BF_4)_2$
Formula	$C_{50}H_{42}B_2F_8FeN_{10}$
Formula Weight	1012.3
Crystal System	Orthorhombic
Space group	*Pbca*
a (Å)	15.4334(17)
b (Å)	14.0656(17)
c (Å)	43.884(5)
α (°)	90
β (°)	90
γ (°)	90
V (Å^3)	9526.3(19)
D_{calc} (g/cm^3)	1.412
Z	8
T (K)	296(2)
μ Mo-Kα (mm^{-1})	0.71073
No. Of reflections	162270
Unique reflections	9530
R_{int}	0.049
R_1 [$I \geq 2\sigma(I)$]	0.0449
wR_2 (full set)	0.1291
goodness-of-fit on F^2	1.019
Largest peak (e Å^{-3})	0.996
Deepest hole (e Å^{-3})	-0.771

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 7
Details of the crystal structure determination of **5**$(ClO_4)_2 \cdot 2(C_2H_5)_2O \cdot CH_3NO_2$.

	5$(ClO_4)_2 \cdot 2(C_2H_5)_2O \cdot CH_3NO_2$
Formula	$C_{31}H_{45}Cl_2FeN_{15}O_{12}$
Formula Weight	946.57
Crystal System	Orthorhombic
Space group	*Pbca*
a (Å)	14.5138(7)
b (Å)	23.2609(11)
c (Å)	12.8419(6)
α (°)	90
β (°)	90
γ (°)	90
V (Å^3)	4335.5(4)
D_{calc} (g/cm^3)	1.45
Z	4
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.71073
No. Of reflections	58012
Unique reflections	5826
R_{int}	0.0402
R_1 [$I \geq 2\sigma(I)$]	0.0335
wR_2 (full set)	0.0918
goodness-of-fit on F^2	1.03
Largest peak (e Å^{-3})	0.438
Deepest hole (e Å^{-3})	-0.399

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w F_o^4]^{1/2}$.

Table 8
Details of the crystal structure determination of **6**$(BF_4)_2 \cdot 3CH_3CN$.

	6$(BF_4)_2 \cdot 3CH_3CN$
Formula	$C_{48}H_{63}B_2F_8FeN_{17}O_4$
Formula Weight	1171.62
Crystal System	Triclinic
Space group	*P*-1
a (Å)	12.8890(4)
b (Å)	13.3521(4)
c (Å)	19.1548(6)
α (°)	104.759(2)
β (°)	100.581(2)
γ (°)	108.536(2)
V (Å^3)	2893.71(17)
D_{calc} (g/cm^3)	1.345
Z	2
T (K)	173(2)
μ Mo-Kα (mm^{-1})	0.343
No. Of reflections	61657
Unique reflections	20578
R_{int}	0.052
R_1 [$I \geq 2\sigma(I)$]	0.0539
wR_2 (full set)	0.1534
goodness-of-fit on F^2	1.035
Largest peak (*e* Å^{-3})	1.28
Deepest hole (*e* Å^{-3})	-0.89

[a] $R = \Sigma[||F_o| - |F_c||]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 9
Details of the crystal structure determination of **6**$(ClO_4)_2 \cdot 3CH_3CN$.

	6$(ClO_4)_2 \cdot 3CH_3CN$
Formula	$C_{48}H_{63}Cl_2FeN_{17}O_{12}$
Formula Weight	1196.9
Crystal System	Triclinic
Space group	*P*-1
a (Å)	12.995(2)
b (Å)	13.3971(19)
c (Å)	19.304(3)
α (°)	104.696(7)
β (°)	100.401(9)
γ (°)	108.320(8)
V (Å^3)	2959.6(8)
D_{calc} (g/cm^3)	1.343
Z	2
T (K)	150.15
μ Mo-Kα (mm^{-1})	0.416
No. Of reflections	67966
Unique reflections	17432
R_{int}	0.1098
R_1 [$I \geq 2\sigma(I)$]	0.0639
wR_2 (full set)	0.1769
goodness-of-fit on F^2	1.025
Largest peak (*e* Å^{-3})	1.00
Deepest hole (*e* Å^{-3})	-0.83

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 10
Details of the crystal structure determination of **12**$(BF_4)_2 \cdot 1.5CF_3CH_2OH \cdot 0.7(C_2H_5)_2O$.

	12$(BF_4)_2 \cdot 1.5CF_3CH_2OH \cdot 0.7(C_3H_7)_2O$
Formula	$C_{41.55}H_{57.08}B_2F_{10.18}FeN_{10}O_{1.08}$
Formula Weight	984.64
Crystal System	Orthorhombic
Space group	$P2_12_12_1$
a (Å)	17.588(2)
b (Å)	19.509(2)
c (Å)	32.399(5)
α (°)	90
β (°)	90
γ (°)	90
V (Å^3)	11117(3)
D_{calc} (g/cm^3)	1.177
Z	8
T (K)	150(2)
μ Mo-Kα (mm^{-1})	0.343
No. Of reflections	124468
Unique reflections	20237
R_{int}	0.0571
R_1 [$I \geq 2\sigma(I)$]	0.0642
wR_2 (full set)	0.1909
goodness-of-fit on F^2	1.043
Largest peak (e Å^{-3})	0.876
Deepest hole (e Å^{-3})	-0.482

[a] $R = \Sigma[||F_o| - |F_c||]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 11
Details of the crystal structure determination of **14**$(BF_4)_2 \cdot 0.25CF_3CH_2OH \cdot 2.6(C_2H_5)_2O$.

	14$(BF_4)_2 \cdot 0.25CF_3CH_2OH \cdot 2.6(C_2H_5)_2O$
Formula	$C_{42.90}H_{64.75}B_2F_{8.75}FeN_{10}O_{2.85}$
Formula Weight	1009.92
Crystal System	Monoclinic
Space group	$P2_1/c$
a (Å)	15.1739(5)
b (Å)	17.9832(6)
c (Å)	19.9978(7)
α (°)	90
β (°)	95.1170(10)
γ (°)	90
V (Å^3)	5435.2(3)
D_{calc} (g/cm^3)	1.234
Z	4
T (K)	110(2)
μ Mo-Kα (mm^{-1})	0.352
No. Of reflections	58452
Unique reflections	13077
R_{int}	0.0365
R_1 [$I \geq 2\sigma(I)$]	0.0783
wR_2 (full set)	0.2626
goodness-of-fit on F^2	1.046
Largest peak (e Å^{-3})	1.239
Deepest hole (e Å^{-3})	-0.556

[a] $R = \Sigma[|F_o| - |F_c|]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 12
Details of the crystal structure determination of $Fe_2(\mathbf{L}^{15a})_2](ClO_4)_4 \cdot 8(CD_3)_2CO$.

	$[Fe_2(L^{15a})_2](ClO_4)_4 \cdot 8(CD_3)_2CO$
Formula	$C_{90}H_{66}D_{58}Cl_4Fe_2N_{24}O_{22}$
Formula Weight	2203.04
Crystal System	triclinic
Space group	P-1
a (Å)	12.8934(4)
b (Å)	14.9852(5)
c (Å)	16.5729(5)
α (°)	114.034(3)
β (°)	99.858(3)
γ (°)	95.295(3)
V (Å^3)	2834.19(16)
D_{calc} (g/cm^3)	1.326
Z	6
T (K)	100.1(6)
μ Mo-Kα (mm^{-1})	0.428
No. Of reflections	14957
Unique reflections	11285
R_{int}	0.0217
R_1 [$I \geq 2\sigma(I)$]	0.0506
wR_2 (full set)	0.1237
goodness-of-fit on F^2	1.059
Largest peak (e Å^{-3})	0.89
Deepest hole (e Å^{-3})	-0.48

[a] $R = \Sigma[||F_o| - |F_c||]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 13
Details of the crystal structure determination of **16**$(ClO_4)_2 \cdot 4C_5H_5N$.

	16$(ClO_4)_2 \cdot 4C_5H_5N$
Formula	$C_{42}H_{40}Cl_2FeN_{16}O_8$
Formula Weight	1022.2
Crystal System	monoclinic
Space group	*I*2/*a*
a (Å)	23.5787(9)
b (Å)	9.7022(4)
c (Å)	21.2623(10)
α (°)	90
β (°)	101.234(4)
γ (°)	90
V (Å^3)	4770.9(4)
D_{calc} (g/cm^3)	1.425
Z	4
T (K)	99.96(18)
μ Cu-Kα (mm^{-1})	4.149
No. Of reflections	5459
Unique reflections	2560
R_{int}	0.0302
R_1 [$I \geq 2\sigma(I)$]	0.0594
wR_2 (full set)	0.1434
goodness-of-fit on F^2	1.042
Largest peak (*e* Å^{-3})	0.9
Deepest hole (*e* Å^{-3})	-0.61

[a] $R = \Sigma[||F_o| - |F_c||]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 14
Details of the crystal structure determination of **18**$(ClO_4)_2 \cdot 2CH_3OH$.

	18$(ClO_4)_2 \cdot 2CH_3OH$
Formula	$C_{24}H_{22}Br_4Cl_2FeN_{10}O_{10}$
Formula Weight	1056.91
Crystal System	triclinic
Space group	P-1
a (Å)	8.9025(4)
b (Å)	12.5712(6)
c (Å)	15.5796(7)
α (°)	78.555(4)
β (°)	86.543(4)
γ (°)	80.986(4)
V (Å^3)	1687.05(13)
D_{calc} (g/cm^3)	2.081
Z	2
T (K)	120.00(11)
μ Mo-Kα (mm^{-1})	5.413
No. Of reflections	18861
Unique reflections	6885
R_{int}	0.0412
R_1 [$I \geq 2\sigma(I)$]	0.039
wR_2 (full set)	0.0882
goodness-of-fit on F^2	1.038
Largest peak (e Å^{-3})	0.9
Deepest hole (e Å^{-3})	-0.76

[a] $R = \Sigma[||F_o| - |F_c||]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 15
Details of the crystal structure determination of the 3:1 mixture of **9**$(BF_4)_x(ClO_4)_{2-x}$.

	9$(BF_4)_x(ClO_4)_{2-x}$ (3:1)
Formula	$C_{26}H_{26}B_{1.5}N_{10}F_6FeCl_{0.5}O_2$
Formula Weight	714.36
Crystal System	tetragonal
Space group	$P4_2/n$
a (Å)	9.6249(16)
b (Å)	9.6249(16)
c (Å)	18.146(3)
α (°)	90
β (°)	90
γ (°)	90
V (Å^3)	1681.1(5)
D_{calc} (g/cm^3)	1.411
Z	2
T (K)	290.15
μ Mo-Kα (mm^{-1})	0.562
No. Of reflections	2868
Unique reflections	1582
R_{int}	0.0373
R_1 [$I \geq 2\sigma(I)$]	0.0953
wR_2 (full set)	0.3174
goodness-of-fit on F^2	1.061
Largest peak (e Å^{-3})	0.52
Deepest hole (e Å^{-3})	-0.7

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 16
Details of the crystal structure determination of [RuCl(**L**^{9})(PPh_3)$_2$]Cl·CH_3OH.

	[RuCl(L^{9})(PPh$_3$)$_2$]Cl·CH$_3$OH
Formula	$C_{48}H_{45}Cl_2N_7OP_2Ru$
Formula Weight	969.16
Crystal System	triclinic
Space group	*P*-1
a (Å)	11.9552(4)
b (Å)	12.4611(4)
c (Å)	16.7064(6)
α (°)	79.906(3)
β (°)	78.741(3)
γ (°)	64.853(3)
V (Å^3)	2197.63(13)
D_{calc} (g/cm^3)	1.463
Z	2
T (K)	120.3(6)
μ Cu-Kα (mm^{-1})	5.045
No. Of reflections	16238
Unique reflections	7507
R_{int}	0.0337
R_1 [$I \geq 2\sigma(I)$]	0.0301
wR_2 (full set)	0.0774
goodness-of-fit on F^2	1.026
Largest peak (*e* Å^{-3})	0.87
Deepest hole (*e* Å^{-3})	-0.57

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 17
Details of the crystal structure determination of $[Ru(OH_2)(\mathbf{L^{12}})(PPh_3)_2]Cl_2 \cdot 2CHCl_3$.

	$[Ru(OH_2)(L^{12})(PPh_3)_2]Cl_2 \cdot 2CHCl_3$
Formula	$C_{57}H_{59}Cl_8N_5OP_2Ru$
Formula Weight	1273.08
Crystal System	monoclinic
Space group	$P2/n$
a (Å)	12.1641(5)
b (Å)	10.8190(4)
c (Å)	25.7429(10)
α (°)	90
β (°)	91.863(3)
γ (°)	90
V (Å^3)	3386.1(2)
D_{calc} (g/cm^3)	1.488
Z	2
T (K)	100.0(5)
μ Mo-Kα (mm^{-1})	0.874
No. Of reflections	14378
Unique reflections	6871
R_{int}	0.0322
R_1 [$I \geq 2\sigma(I)$]	0.0477
wR_2 (full set)	0.0935
goodness-of-fit on F^2	1.148
Largest peak (e Å^{-3})	0.55
Deepest hole (e Å^{-3})	-0.71

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.

Table 18
Details of the crystal structure determination of $[Ru(OH_2)(\mathbf{L^{12}})(PPh_3)_2](PF_6)_2$.

	$[Ru(OH_2)(L^{12})(PPh_3)_2](PF_6)_2$
Formula	$C_{55}H_{57}F_{12}N_5OP_4Ru$
Formula Weight	1257.24
Crystal System	monoclinic
Space group	$C2/c$
a (Å)	41.2140(9)
b (Å)	12.9493(3)
c (Å)	21.7724(4)
α (°)	90
β (°)	96.056(2)
γ (°)	90
V (Å^3)	11554.9(4)
D_{calc} (g/cm^3)	1.442
Z	8
T (K)	110.00(10)
μ Cu-Kα (mm^{-1})	3.955
No. Of reflections	24206
Unique reflections	9999
R_{int}	0.0461
R_1 [$I \geq 2\sigma(I)$]	0.0653
wR_2 (full set)	0.1877
goodness-of-fit on F^2	1.039
Largest peak (e Å^{-3})	2.83
Deepest hole (e Å^{-3})	-0.93

[a] $R = \Sigma[\,|F_o| - |F_c|\,]/\Sigma|F_o|$.

[b] $wR = [\Sigma w(F_o^2 - F_c^2)2/\Sigma w F_o^4]^{1/2}$.