Supporting Information

for

Stereochemical Control of Fe(II) Complexes Containing a Diphosphine Ligand with a Pendant Nitrogen Base

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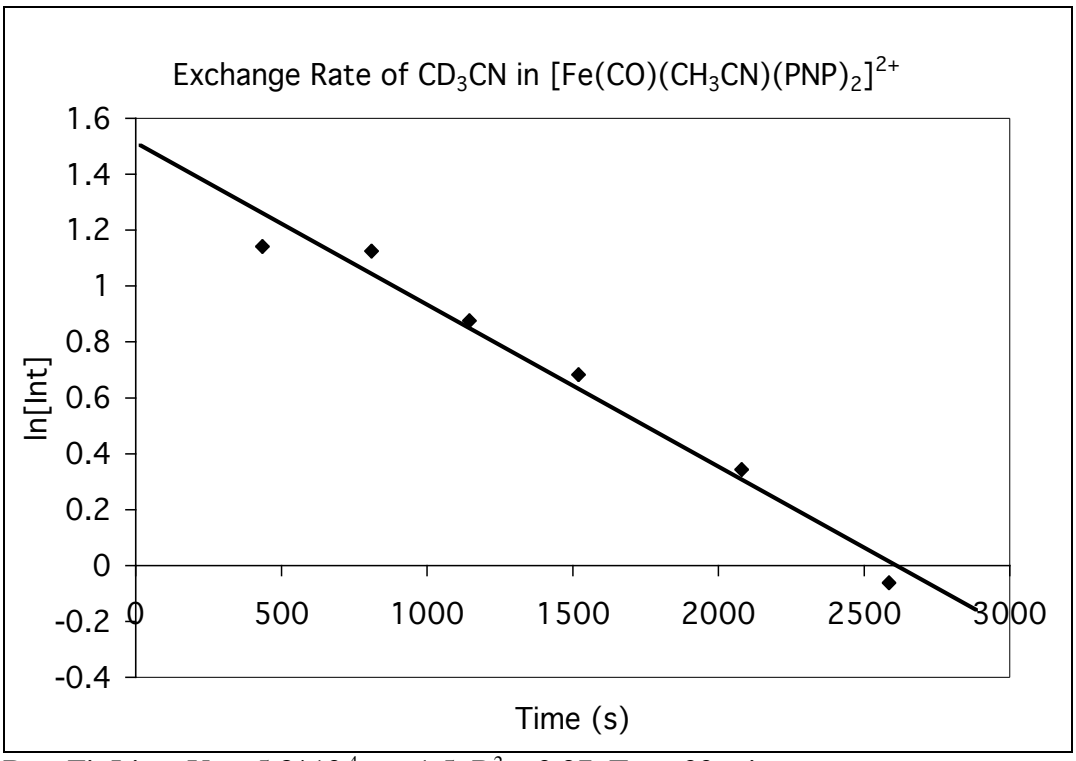

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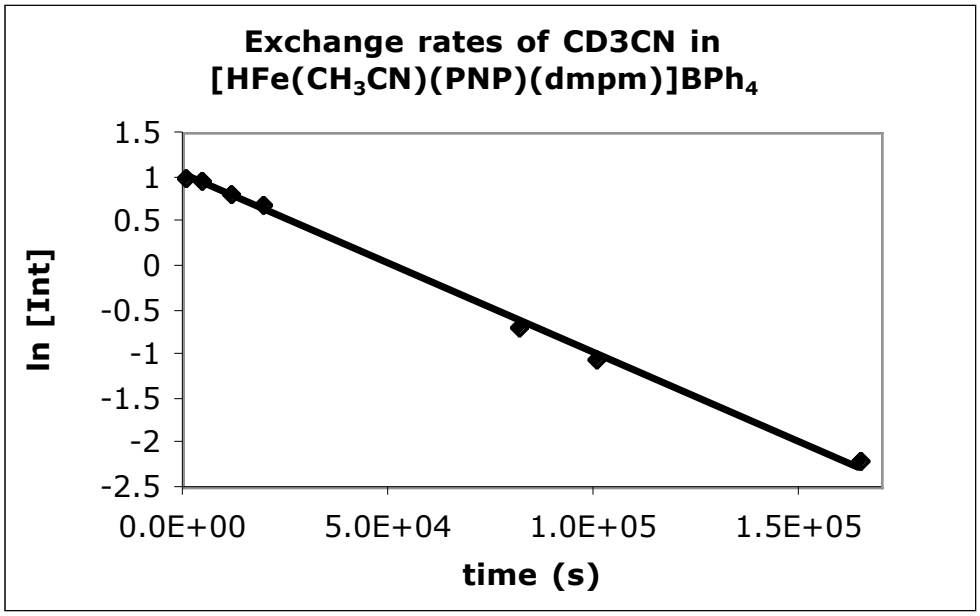

Best Fit Line: $Y = -2.0*10^{-5} x + 1.02$, $R^2 = 0.97$, $T_{1/2} = 9.6$ h

Figure S1. (a) Top graph shows first order kinetics plot for CH_3CN exchange in $[Fe(CO)(CH_3CN)(PNP)_2](BPh_4)_2$ in CD_3CN. (b) Bottom graph shows first order kinetics plot for CH_3CN exchange in $[HFe(PNP)(dmpm)(CH_3CN)](BPh_4)_2$ in CD_3CN. See Experimental Section for details.

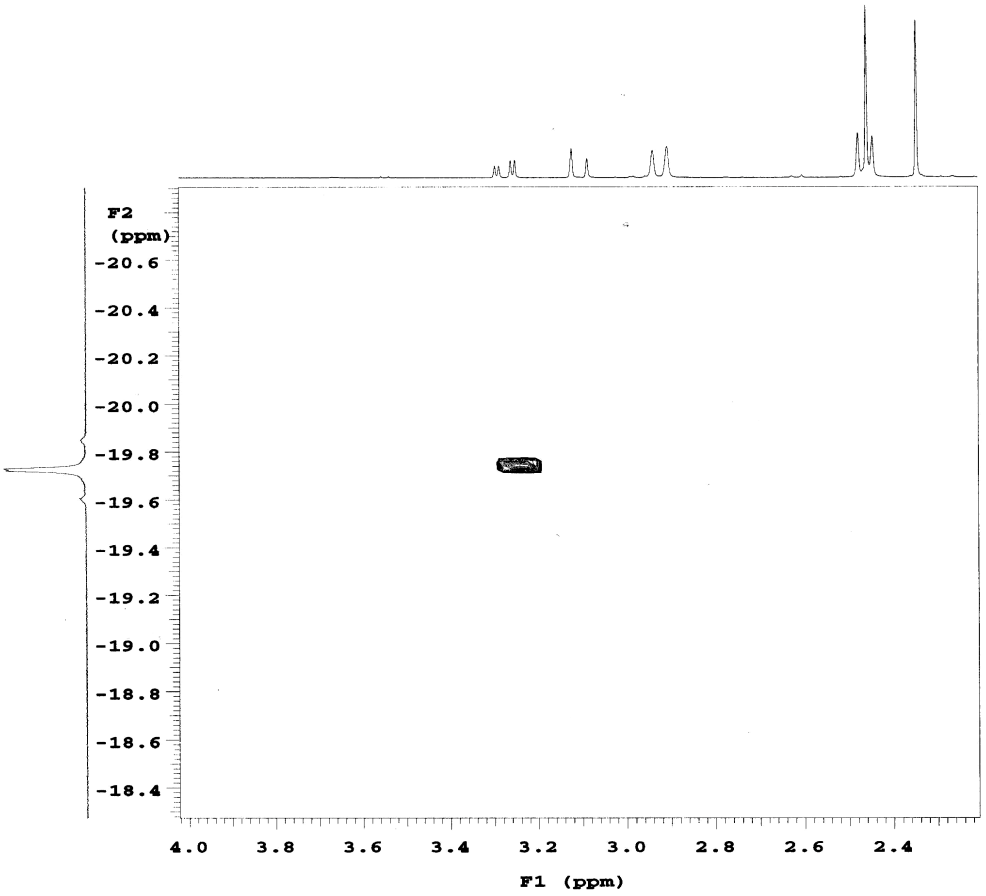

Figure S2. Broadband ^{31}P-decoupled ^{1}H-gCOSY NMR spectrum of [HFe(PNP)(dmpm)(CH_3CN)]$^+$

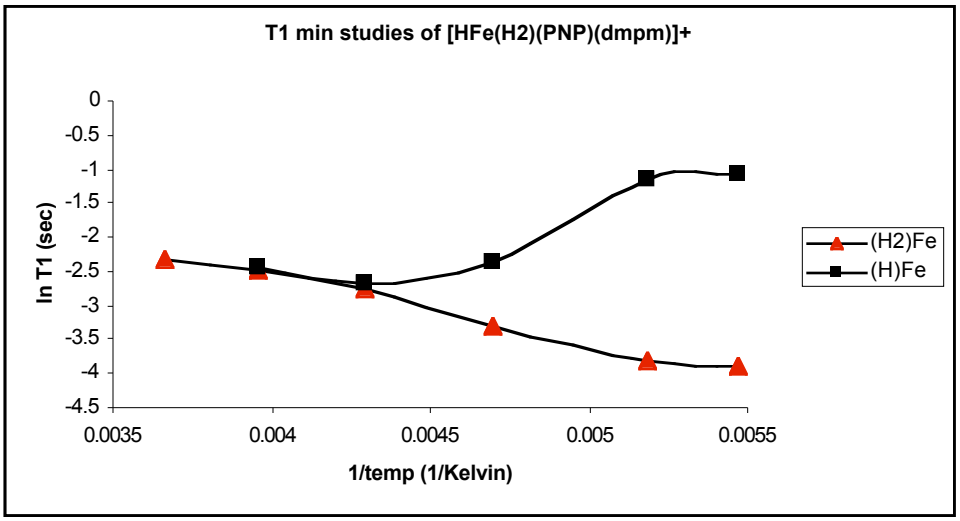

Figure S3. Plot of lnT_1 versus 1/°K for [(H_2)Fe(PNP)(dmpm)(H)]$^+$

Thermodynamic Cycle used for the calculation of the hydride donor ability of $[HFe(PNP)(dmpm)(CH_3CN)]^+$

	$\Delta G°$
$[HFe(PNP)(dmpm)(CH_3CN)]^+ + BH^+ \rightarrow$ $Fe(PNP)(dmpm)(CH_3CN)_2]^{2+} + B + H_2$	$\Delta G°(1) < 0$
$B + H^+ \rightarrow BH^+$	$-1.37pK_a = -15.5$ kcal/mol
$H_2(g) \rightarrow H^+(CH_3CN) + H^-(CH_3CN)$	76.0 kcal/mol[a,b]

$[HFe(PNP)(dmpm)(CH_3CN)]^+ \rightarrow H^- + Fe(PNP)(dmpm)(CH_3CN)_2]^{2+}$

$\Delta G°_{H^-} = \Delta G°(1) - 1.37pK_a + 76.0 = 60.5 + \Delta G°(1)$

Because reaction 1 is spontaneous, $\Delta G°(1)$ is less than 0, and the hydride donor ability ($\Delta G°_{H^-}$) of $[HFe(PNP)(dmpm)(CH_3CN)]^+$ is less than 61 kcal/mol. The pK_a value of anisidinium (BH^+) is 11.3 (second reaction in above scheme).

[a]A value of 76.3 kcal/mol for the heterolytic cleavage of gaseous H_2 in acetonitrile comes from data in Wayner, D. D. M.; Parker, V. D. *Acc. Chem. Res.* **1993**, *26*, 287-294.
[b]The value of 76.3 kcal/mol from Wayner et. al. was modified slightly to 76.0 kcal/mol based on experimental data presented in the supporting information of Ellis, W. W.; Raebiger, J. W.; Curtis, C. J.; Bruno, J. W.; DuBois, D. L. *J. Am. Chem. Soc.* **2004**, *126*, 2738-2743.

Table S1. Crystal data and structure refinement for $[(H)Fe(PNP)(dmpm)(CH_3CN)]BPh_4$.

Identification code	tric	
Empirical formula	$C_{44}H_{68}B\ FeN_3P_4$	
Formula weight	829.55	
Temperature	153(2) K	
Wavelength	0.71073 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 11.7581(5) Å	α = 105.7990(10)°.
	b = 12.0235(6) Å	β = 95.9700(10)°.
	c = 17.3035(8) Å	γ = 98.8340(10)°.
Volume	2298.40(18) $Å^3$	
Z	2	
Density (calculated)	1.199 Mg/m^3	
Absorption coefficient	0.500 mm^{-1}	
F(000)	888	
Crystal size	0.97 x 0.49 x 0.25 mm^3	
Theta range for data collection	1.79 to 27.48°.	
Index ranges	$-15 \le h \le 15, -14 \le k \le 15, -19 \le l \le 22$	
Reflections collected	17440	
Independent reflections	10414 [R(int) = 0.0468]	
Completeness to theta = 27.48°	98.6 %	
Absorption correction	None	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	10414 / 0 / 493	
Goodness-of-fit on F^2	1.016	
Final R indices [I>2sigma(I)]	R1 = 0.0540, wR2 = 0.1403	
R indices (all data)	R1 = 0.0658, wR2 = 0.1515	
Largest diff. peak and hole	0.864 and -0.667 $e.Å^{-3}$	

Table S2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for [(H)Fe(PNP)(dmpm)(CH_3CN)]BPh_4. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Fe(1)	2738(1)	3904(1)	2820(1)	22(1)
P(1)	4135(1)	3185(1)	2188(1)	27(1)
P(2)	4192(1)	5411(1)	3021(1)	29(1)
P(3)	1607(1)	4805(1)	3631(1)	24(1)
P(4)	1577(1)	2205(1)	2655(1)	23(1)
N(2)	-337(2)	3018(2)	3252(1)	29(1)
N(1)	1906(2)	4236(2)	1920(1)	25(1)
N(3)	3736(4)	-1414(5)	-4787(5)	216(4)
B(1)	1770(2)	2315(2)	-1768(2)	25(1)
C(1)	1452(2)	4529(2)	1408(1)	26(1)
C(2)	888(2)	4924(2)	769(1)	33(1)
C(3)	4861(2)	2036(2)	2401(2)	46(1)
C(4)	4070(2)	2853(3)	1094(2)	45(1)
C(5)	5254(2)	4527(2)	2607(2)	39(1)
C(6)	4925(2)	6387(2)	4003(2)	42(1)
C(7)	4164(2)	6417(3)	2404(2)	49(1)
C(8)	1692(2)	4755(2)	4684(1)	33(1)
C(9)	2896(2)	5171(3)	5185(2)	52(1)
C(10)	1680(2)	6375(2)	3708(2)	31(1)
C(11)	873(2)	7049(2)	4211(2)	44(1)
C(12)	24(2)	4260(2)	3303(1)	29(1)
C(13)	-1610(2)	2729(2)	3212(2)	42(1)
C(14)	3(2)	2247(2)	2540(1)	28(1)
C(15)	1686(2)	1477(2)	3456(1)	30(1)
C(16)	2885(2)	1193(3)	3662(2)	41(1)
C(17)	1543(2)	968(2)	1742(1)	30(1)
C(18)	1113(2)	1182(2)	943(1)	34(1)
C(19)	2288(2)	3733(2)	-1349(1)	28(1)
C(20)	1780(2)	4571(2)	-1619(2)	34(1)
C(21)	2124(2)	5779(2)	-1257(2)	38(1)

C(22)	3003(2)	6203(2)	-593(2)	39(1)
C(23)	3528(2)	5413(2)	-303(2)	38(1)
C(24)	3173(2)	4206(2)	-676(1)	32(1)
C(25)	2514(2)	1483(2)	-1390(1)	27(1)
C(26)	3726(2)	1611(2)	-1378(2)	35(1)
C(27)	4380(2)	876(3)	-1112(2)	43(1)
C(28)	3848(2)	-38(2)	-853(2)	39(1)
C(29)	2657(2)	-200(2)	-864(2)	37(1)
C(30)	2013(2)	546(2)	-1126(1)	30(1)
C(31)	1771(2)	1881(2)	-2756(1)	27(1)
C(32)	1316(2)	687(2)	-3168(2)	34(1)
C(33)	1188(2)	217(2)	-4001(2)	38(1)
C(34)	1531(2)	931(2)	-4475(2)	37(1)
C(35)	2012(2)	2106(2)	-4095(2)	36(1)
C(36)	2131(2)	2568(2)	-3251(1)	30(1)
C(37)	406(2)	2137(2)	-1601(1)	26(1)
C(38)	143(2)	2216(2)	-815(1)	30(1)
C(39)	-993(2)	2130(2)	-642(2)	35(1)
C(40)	-1921(2)	1960(2)	-1257(2)	39(1)
C(41)	-1696(2)	1888(2)	-2033(2)	38(1)
C(42)	-555(2)	1972(2)	-2202(2)	31(1)
C(43)	3986(3)	-628(4)	-4237(3)	97(2)
C(44)	4300(3)	367(3)	-3553(3)	77(1)

Table S3. Bond lengths [Å] and angles [°] for [(H)Fe(PNP)(dmpm)(CH_3CN)]BPh_4.

Fe(1)-N(1)	1.9186(18)	C(19)-C(24)	1.402(3)
Fe(1)-P(4)	2.2044(6)	C(19)-C(20)	1.405(3)
Fe(1)-P(2)	2.2159(6)	C(20)-C(21)	1.392(3)
Fe(1)-P(3)	2.2173(6)	C(21)-C(22)	1.386(4)
Fe(1)-P(1)	2.2217(6)	C(22)-C(23)	1.381(4)
Fe(1)-H(1)	1.53(2)	C(23)-C(24)	1.395(3)
P(1)-C(4)	1.816(3)	C(25)-C(30)	1.399(3)
P(1)-C(3)	1.828(3)	C(25)-C(26)	1.408(3)
P(1)-C(5)	1.837(2)	C(26)-C(27)	1.392(4)
P(1)-P(2)	2.6588(8)	C(27)-C(28)	1.384(4)
P(2)-C(6)	1.818(3)	C(28)-C(29)	1.381(4)
P(2)-C(7)	1.821(3)	C(29)-C(30)	1.395(3)
P(2)-C(5)	1.838(3)	C(31)-C(36)	1.397(3)
P(3)-C(8)	1.832(2)	C(31)-C(32)	1.408(3)
P(3)-C(10)	1.844(2)	C(32)-C(33)	1.381(3)
P(3)-C(12)	1.849(2)	C(33)-C(34)	1.385(4)
P(4)-C(15)	1.834(2)	C(34)-C(35)	1.386(4)
P(4)-C(17)	1.844(2)	C(35)-C(36)	1.396(3)
P(4)-C(14)	1.851(2)	C(37)-C(42)	1.405(3)
N(2)-C(14)	1.459(3)	C(37)-C(38)	1.406(3)
N(2)-C(12)	1.463(3)	C(38)-C(39)	1.394(3)
N(2)-C(13)	1.474(3)	C(39)-C(40)	1.391(4)
N(1)-C(1)	1.151(3)	C(40)-C(41)	1.377(4)
N(3)-C(43)	1.120(6)	C(41)-C(42)	1.397(3)
B(1)-C(25)	1.643(3)	C(43)-C(44)	1.403(5)
B(1)-C(31)	1.645(3)		
B(1)-C(19)	1.649(3)	N(1)-Fe(1)-P(4)	94.68(5)
B(1)-C(37)	1.651(3)	N(1)-Fe(1)-P(2)	93.78(5)
C(1)-C(2)	1.455(3)	P(4)-Fe(1)-P(2)	168.28(2)
C(10)-C(11)	1.529(3)	N(1)-Fe(1)-P(3)	89.63(6)
C(15)-C(16)	1.530(3)	P(4)-Fe(1)-P(3)	89.58(2)
C(17)-C(18)	1.525(3)	P(2)-Fe(1)-P(3)	98.58(2)
		N(1)-Fe(1)-P(1)	97.75(6)
		P(4)-Fe(1)-P(1)	97.18(2)
		P(2)-Fe(1)-P(1)	73.62(2)

P(3)-Fe(1)-P(1)	169.52(2)	C(17)-P(4)-Fe(1)	119.48(8)
N(1)-Fe(1)-H(1)	177.7(10)	C(14)-P(4)-Fe(1)	114.93(7)
P(4)-Fe(1)-H(1)	87.5(9)	C(14)-N(2)-C(12)	111.47(18)
P(2)-Fe(1)-H(1)	83.9(9)	C(14)-N(2)-C(13)	109.16(18)
P(3)-Fe(1)-H(1)	91.2(10)	C(12)-N(2)-C(13)	110.10(19)
P(1)-Fe(1)-H(1)	81.1(10)	C(1)-N(1)-Fe(1)	174.49(17)
C(4)-P(1)-C(3)	101.75(15)	C(25)-B(1)-C(31)	105.92(18)
C(4)-P(1)-C(5)	104.56(13)	C(25)-B(1)-C(19)	112.71(17)
C(3)-P(1)-C(5)	103.37(13)	C(31)-B(1)-C(19)	113.36(18)
C(4)-P(1)-Fe(1)	121.84(10)	C(25)-B(1)-C(37)	112.01(17)
C(3)-P(1)-Fe(1)	125.62(10)	C(31)-B(1)-C(37)	108.41(17)
C(5)-P(1)-Fe(1)	95.89(8)	C(19)-B(1)-C(37)	104.49(17)
C(4)-P(1)-P(2)	117.15(11)	N(1)-C(1)-C(2)	178.8(2)
C(3)-P(1)-P(2)	132.77(10)	P(1)-C(5)-P(2)	92.68(11)
C(5)-P(1)-P(2)	43.68(8)	C(9)-C(8)-P(3)	115.24(19)
Fe(1)-P(1)-P(2)	53.090(19)	C(11)-C(10)-P(3)	118.80(18)
C(6)-P(2)-C(7)	102.06(15)	N(2)-C(12)-P(3)	113.06(15)
C(6)-P(2)-C(5)	105.79(12)	N(2)-C(14)-P(4)	113.36(15)
C(7)-P(2)-C(5)	102.69(14)	C(16)-C(15)-P(4)	114.75(16)
C(6)-P(2)-Fe(1)	125.91(10)	C(18)-C(17)-P(4)	114.55(15)
C(7)-P(2)-Fe(1)	120.68(10)	C(24)-C(19)-C(20)	114.8(2)
C(5)-P(2)-Fe(1)	96.04(8)	C(24)-C(19)-B(1)	124.7(2)
C(6)-P(2)-P(1)	135.41(9)	C(20)-C(19)-B(1)	120.30(19)
C(7)-P(2)-P(1)	114.61(12)	C(21)-C(20)-C(19)	123.2(2)
C(5)-P(2)-P(1)	43.64(8)	C(22)-C(21)-C(20)	119.8(2)
Fe(1)-P(2)-P(1)	53.292(19)	C(23)-C(22)-C(21)	119.1(2)
C(8)-P(3)-C(10)	104.41(11)	C(22)-C(23)-C(24)	120.2(2)
C(8)-P(3)-C(12)	98.71(11)	C(23)-C(24)-C(19)	122.9(2)
C(10)-P(3)-C(12)	99.76(10)	C(30)-C(25)-C(26)	114.6(2)
C(8)-P(3)-Fe(1)	120.36(8)	C(30)-C(25)-B(1)	124.28(19)
C(10)-P(3)-Fe(1)	115.13(8)	C(26)-C(25)-B(1)	120.9(2)
C(12)-P(3)-Fe(1)	115.35(8)	C(27)-C(26)-C(25)	122.8(2)
C(15)-P(4)-C(17)	101.16(11)	C(28)-C(27)-C(26)	120.6(2)
C(15)-P(4)-C(14)	100.25(10)	C(29)-C(28)-C(27)	118.4(2)
C(17)-P(4)-C(14)	98.00(11)	C(28)-C(29)-C(30)	120.5(2)
C(15)-P(4)-Fe(1)	119.30(8)	C(29)-C(30)-C(25)	123.1(2)

C(36)-C(31)-C(32)	115.0(2)
C(36)-C(31)-B(1)	127.7(2)
C(32)-C(31)-B(1)	117.23(19)
C(33)-C(32)-C(31)	123.6(2)
C(32)-C(33)-C(34)	119.8(2)
C(33)-C(34)-C(35)	118.7(2)
C(34)-C(35)-C(36)	120.7(2)
C(35)-C(36)-C(31)	122.2(2)
C(42)-C(37)-C(38)	115.5(2)
C(42)-C(37)-B(1)	123.5(2)
C(38)-C(37)-B(1)	120.85(19)
C(39)-C(38)-C(37)	122.5(2)
C(40)-C(39)-C(38)	120.1(2)
C(41)-C(40)-C(39)	119.0(2)
C(40)-C(41)-C(42)	120.5(2)
C(41)-C(42)-C(37)	122.3(2)
N(3)-C(43)-C(44)	179.3(8)

Symmetry transformations used to generate equivalent atoms:

Table S4. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for $[(H)Fe(PNP)(dmpm)(CH_3CN)]BPh_4$. The anisotropic displacement factor exponent takes the form: $-2\pi^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Fe(1)	21(1)	21(1)	23(1)	6(1)	4(1)	5(1)
P(1)	25(1)	28(1)	30(1)	6(1)	8(1)	7(1)
P(2)	23(1)	25(1)	37(1)	7(1)	3(1)	3(1)
P(3)	23(1)	24(1)	23(1)	4(1)	3(1)	7(1)
P(4)	25(1)	21(1)	24(1)	6(1)	5(1)	5(1)
N(2)	25(1)	30(1)	31(1)	5(1)	8(1)	4(1)
N(1)	24(1)	22(1)	27(1)	5(1)	5(1)	3(1)
N(3)	75(3)	155(5)	284(8)	-150(5)	-8(4)	31(3)
B(1)	24(1)	26(1)	26(1)	8(1)	6(1)	5(1)
C(1)	25(1)	24(1)	29(1)	7(1)	4(1)	4(1)
C(2)	38(1)	31(1)	32(1)	12(1)	0(1)	10(1)
C(3)	40(1)	44(2)	64(2)	19(1)	21(1)	24(1)
C(4)	36(1)	63(2)	30(1)	5(1)	12(1)	1(1)
C(5)	24(1)	36(1)	50(2)	3(1)	7(1)	4(1)
C(6)	31(1)	35(1)	48(2)	-5(1)	-1(1)	3(1)
C(7)	40(1)	46(2)	70(2)	34(2)	11(1)	1(1)
C(8)	41(1)	36(1)	23(1)	7(1)	7(1)	13(1)
C(9)	46(2)	81(2)	28(1)	5(1)	1(1)	30(2)
C(10)	31(1)	23(1)	38(1)	5(1)	4(1)	9(1)
C(11)	48(2)	33(1)	51(2)	5(1)	13(1)	20(1)
C(12)	24(1)	29(1)	32(1)	5(1)	5(1)	7(1)
C(13)	27(1)	45(2)	50(2)	8(1)	15(1)	2(1)
C(14)	25(1)	27(1)	31(1)	7(1)	4(1)	4(1)
C(15)	35(1)	30(1)	30(1)	13(1)	10(1)	8(1)
C(16)	41(1)	49(2)	46(2)	28(1)	10(1)	16(1)
C(17)	37(1)	20(1)	32(1)	3(1)	7(1)	5(1)
C(18)	40(1)	31(1)	27(1)	2(1)	7(1)	3(1)
C(19)	25(1)	30(1)	30(1)	9(1)	8(1)	3(1)
C(20)	30(1)	30(1)	39(1)	7(1)	3(1)	6(1)
C(21)	37(1)	29(1)	50(2)	10(1)	12(1)	11(1)
C(22)	35(1)	27(1)	48(2)	1(1)	16(1)	2(1)

C(23)	31(1)	40(1)	36(1)	3(1)	5(1)	-2(1)
C(24)	30(1)	35(1)	31(1)	8(1)	4(1)	3(1)
C(25)	27(1)	29(1)	26(1)	6(1)	6(1)	7(1)
C(26)	29(1)	40(1)	41(1)	15(1)	11(1)	9(1)
C(27)	28(1)	52(2)	51(2)	12(1)	8(1)	18(1)
C(28)	43(1)	38(1)	38(1)	8(1)	3(1)	20(1)
C(29)	43(1)	31(1)	39(1)	12(1)	7(1)	13(1)
C(30)	29(1)	29(1)	34(1)	8(1)	7(1)	8(1)
C(31)	22(1)	30(1)	28(1)	8(1)	5(1)	5(1)
C(32)	37(1)	32(1)	32(1)	9(1)	9(1)	1(1)
C(33)	37(1)	36(1)	34(1)	1(1)	7(1)	0(1)
C(34)	31(1)	51(2)	27(1)	7(1)	5(1)	7(1)
C(35)	28(1)	50(2)	34(1)	21(1)	8(1)	5(1)
C(36)	25(1)	31(1)	33(1)	11(1)	4(1)	3(1)
C(37)	25(1)	20(1)	33(1)	8(1)	6(1)	5(1)
C(38)	31(1)	23(1)	35(1)	8(1)	9(1)	7(1)
C(39)	39(1)	25(1)	45(1)	9(1)	19(1)	8(1)
C(40)	28(1)	27(1)	62(2)	11(1)	17(1)	6(1)
C(41)	26(1)	28(1)	57(2)	9(1)	1(1)	7(1)
C(42)	30(1)	28(1)	36(1)	10(1)	4(1)	6(1)
C(43)	47(2)	65(2)	140(4)	-34(3)	4(2)	19(2)
C(44)	62(2)	54(2)	96(3)	-6(2)	-7(2)	24(2)

Table S5. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for $[(H)Fe(PNP)(dmpm)(CH_3CN)]BPh_4$.

	x	y	z	U(eq)
H(1)	3440(20)	3670(20)	3533(16)	30(6)
H(2A)	785	5736	996	46
H(2B)	127	4414	541	46
H(2C)	1375	4889	339	46
H(3A)	4337	1270	2176	64
H(3B)	5064	2199	2990	64
H(3C)	5571	2024	2150	64
H(4A)	4859	2864	957	63
H(4B)	3726	3445	907	63
H(4C)	3590	2072	829	63
H(5A)	5625	4794	2184	54
H(5B)	5853	4464	3031	54
H(6A)	5670	6816	3937	59
H(6B)	5066	5923	4376	59
H(6C)	4435	6949	4224	59
H(7A)	3631	6953	2587	69
H(7B)	3896	5967	1833	69
H(7C)	4949	6875	2460	69
H(8A)	1162	5245	4956	46
H(8B)	1403	3934	4679	46
H(9A)	3438	4714	4912	73
H(9B)	2867	5062	5724	73
H(9C)	3161	6007	5242	73
H(10A)	2490	6788	3935	43
H(10B)	1525	6437	3150	43
H(11A)	60	6696	3974	61
H(11B)	1029	7873	4211	61
H(11C)	1012	7010	4771	61
H(12A)	-401	4738	3692	40
H(12B)	-197	4367	2764	40

H(13A)	-1995	2869	2726	58
H(13B)	-1840	3226	3698	58
H(13C)	-1841	1899	3185	58
H(14A)	-206	2518	2062	39
H(14B)	-445	1440	2434	39
H(15A)	1482	1989	3954	42
H(15B)	1103	734	3285	42
H(16A)	3093	675	3175	58
H(16B)	2865	798	4088	58
H(16C)	3467	1925	3855	58
H(17A)	2339	795	1725	42
H(17B)	1034	261	1791	42
H(18A)	283	1216	911	48
H(18B)	1230	539	488	48
H(18C)	1551	1930	916	48
H(20)	1172	4299	-2071	40
H(21)	1757	6311	-1464	45
H(22)	3243	7026	-340	46
H(23)	4131	5693	152	46
H(24)	3547	3681	-465	39
H(26)	4115	2224	-1558	42
H(27)	5198	1004	-1108	52
H(28)	4292	-542	-673	47
H(29)	2275	-825	-691	44
H(30)	1195	412	-1126	36
H(32)	1086	177	-2855	41
H(33)	865	-594	-4248	46
H(34)	1439	621	-5050	45
H(35)	2264	2604	-4411	43
H(36)	2469	3375	-3006	35
H(38)	763	2332	-386	35
H(39)	-1134	2188	-103	42
H(40)	-2697	1895	-1144	46
H(41)	-2322	1781	-2456	46
H(42)	-424	1915	-2743	37
H(44A)	3725	877	-3553	107

H(44B)	5070	801	-3571	107
H(44C)	4325	119	-3058	107

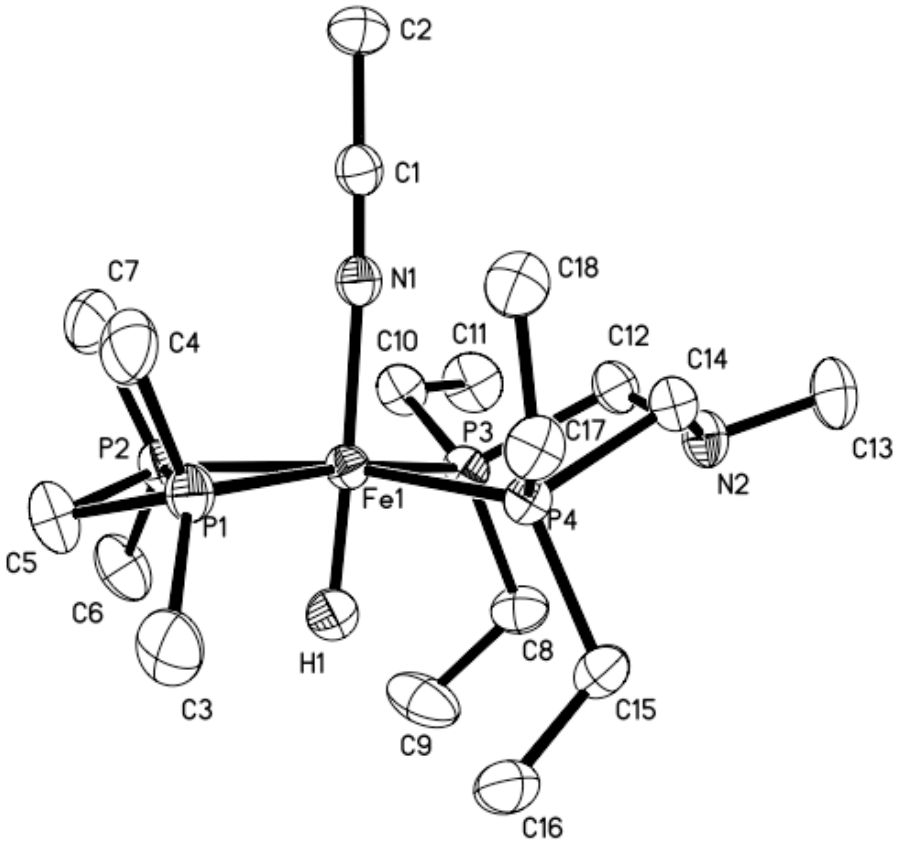

Figure S4. Drawing of the *trans*-[HFe(PNP)(dmpm)(CH_3CN)]$^+$ cation indicating the atom numbering scheme. Thermal ellipsoids are drawn at the 50% probability level.

Table S6. Crystal data and structure refinement for $[Fe(PNP)_2(CH_3CN)(CO)](BPh_4)_2$.

Identification code	p21	
Empirical formula	$C_{73}H_{97}B_2FeN_3OP_4$	
Formula weight	1233.89	
Temperature	154(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P2(1)/n	
Unit cell dimensions	a = 21.873(10) Å	α = 90°.
	b = 13.629(6) Å	β = 109.824(10)°.
	c = 24.754(11) Å	γ = 90°.
Volume	6942(6) $Å^3$	
Z	4	
Density (calculated)	1.181 Mg/m^3	
Absorption coefficient	0.353 mm^{-1}	
F(000)	2640	
Crystal size	0.45 x 0.5 x 0.55 mm^3	
Theta range for data collection	1.73 to 27.48°.	
Index ranges	$-28 \le h \le 28$, $-17 \le k \le 15$, $-31 \le l \le 32$	
Reflections collected	53474	
Independent reflections	15911 [R(int) = 0.3327]	
Completeness to theta = 27.48°	99.8 %	
Absorption correction	None	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	15911 / 0 / 768	
Goodness-of-fit on F^2	0.896	
Final R indices [I>2sigma(I)]	R1 = 0.1074, wR2 = 0.2230	
R indices (all data)	R1 = 0.2638, wR2 = 0.3062	
Largest diff. peak and hole	1.233 and -1.421 e.$Å^{-3}$	

Table S7. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for $[Fe(PNP)_2(CH_3CN)(CO)](BPh_4)_2$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Fe(1)	9881(1)	12447(1)	7596(1)	26(1)
P(1)	10774(1)	13549(1)	7927(1)	34(1)
P(2)	10081(1)	12019(1)	8540(1)	33(1)
P(3)	10436(1)	11149(1)	7392(1)	29(1)
P(4)	9575(1)	12985(1)	6652(1)	32(1)
N(1)	9314(3)	13527(4)	7693(3)	41(2)
N(2)	11154(3)	13167(4)	9079(2)	39(2)
N(3)	10109(3)	11491(4)	6219(2)	34(1)
O(1)	8721(2)	11200(3)	7324(2)	42(1)
B(1)	13706(4)	12301(5)	8912(3)	31(2)
B(2)	9078(4)	7480(6)	8548(3)	34(2)
C(1)	8907(4)	14044(5)	7662(3)	37(2)
C(2)	8378(4)	14725(5)	7679(3)	47(2)
C(3)	9180(3)	11664(5)	7419(3)	32(2)
C(4)	10516(4)	14806(5)	8052(4)	50(2)
C(5)	10930(5)	15379(6)	8581(4)	82(3)
C(6)	11269(3)	13699(5)	7458(3)	41(2)
C(7)	11741(4)	14556(6)	7584(3)	57(2)
C(8)	11416(3)	13299(5)	8621(3)	35(2)
C(9)	11638(4)	13413(6)	9643(3)	50(2)
C(10)	10900(3)	12182(5)	9076(3)	38(2)
C(11)	9588(3)	12753(5)	8860(3)	40(2)
C(12)	8867(3)	12494(5)	8651(3)	41(2)
C(13)	9917(4)	10764(5)	8683(3)	38(2)
C(14)	9978(4)	10481(5)	9294(3)	39(2)
C(15)	10023(3)	9940(4)	7314(3)	36(2)
C(16)	9498(4)	9696(5)	6739(3)	52(2)
C(17)	11244(3)	10858(5)	7921(3)	32(2)
C(18)	11664(4)	10135(5)	7739(3)	44(2)
C(19)	10647(3)	11193(5)	6733(3)	35(2)
C(20)	10227(4)	11129(5)	5707(3)	48(2)

C(21)	10023(3)	12556(5)	6191(3)	36(2)
C(22)	8742(3)	12640(5)	6205(3)	41(2)
C(23)	8190(4)	13084(6)	6347(4)	67(3)
C(24)	9573(4)	14322(5)	6557(3)	45(2)
C(25)	9322(5)	14712(6)	5932(4)	76(3)
C(26)	13447(3)	13324(5)	9120(3)	34(2)
C(27)	13224(3)	14141(5)	8763(3)	39(2)
C(28)	13060(4)	15027(6)	8972(4)	50(2)
C(29)	13119(4)	15130(6)	9539(4)	55(2)
C(30)	13350(4)	14338(7)	9901(4)	63(2)
C(31)	13509(4)	13448(6)	9690(3)	52(2)
C(32)	13334(3)	11308(5)	9039(3)	34(2)
C(33)	12854(4)	11295(6)	9300(3)	53(2)
C(34)	12567(4)	10407(7)	9387(4)	60(3)
C(35)	12733(4)	9521(6)	9205(4)	59(2)
C(36)	13198(4)	9509(5)	8943(3)	46(2)
C(37)	13471(4)	10372(5)	8858(3)	41(2)
C(38)	13542(3)	12294(4)	8210(3)	32(2)
C(39)	12895(3)	12303(5)	7843(3)	37(2)
C(40)	12716(4)	12309(5)	7243(3)	42(2)
C(41)	13189(4)	12320(5)	6996(3)	41(2)
C(42)	13831(4)	12314(5)	7336(3)	42(2)
C(43)	14000(4)	12305(5)	7934(3)	38(2)
C(44)	14486(3)	12231(5)	9265(3)	33(2)
C(45)	14915(3)	12972(5)	9207(3)	38(2)
C(46)	15585(4)	12957(5)	9487(3)	43(2)
C(47)	15852(4)	12216(5)	9875(3)	44(2)
C(48)	15464(4)	11491(5)	9963(3)	41(2)
C(49)	14801(3)	11494(5)	9668(3)	31(2)
C(50)	8689(3)	8453(5)	8680(3)	36(2)
C(51)	8613(3)	8559(5)	9215(3)	39(2)
C(52)	8331(4)	9402(6)	9355(4)	50(2)
C(53)	8095(4)	10158(6)	8959(4)	50(2)
C(54)	8166(3)	10063(5)	8433(4)	43(2)
C(55)	8461(3)	9241(5)	8293(3)	37(2)
C(56)	8834(4)	6494(5)	8822(3)	40(2)

C(57)	9248(4)	5747(6)	9130(4)	53(2)
C(58)	9013(5)	4910(6)	9352(4)	63(3)
C(59)	8357(6)	4836(6)	9256(4)	68(3)
C(60)	7926(6)	5534(6)	8932(5)	74(3)
C(61)	8170(4)	6362(5)	8725(4)	49(2)
C(62)	8879(4)	7262(5)	7857(3)	39(2)
C(63)	8249(4)	7423(5)	7465(4)	49(2)
C(64)	8074(4)	7225(6)	6873(4)	56(2)
C(65)	8534(5)	6888(6)	6652(4)	59(2)
C(66)	9160(4)	6706(5)	7027(4)	47(2)
C(67)	9320(4)	6896(5)	7610(3)	41(2)
C(68)	9849(4)	7745(5)	8871(3)	37(2)
C(69)	10225(4)	8211(5)	8594(3)	37(2)
C(70)	10869(4)	8496(5)	8866(3)	42(2)
C(71)	11176(4)	8318(6)	9448(3)	50(2)
C(72)	10811(4)	7873(6)	9744(4)	57(2)
C(73)	10173(4)	7599(5)	9467(3)	48(2)

Table S8. Bond lengths [Å] and angles [°] for $[Fe(PNP)_2(CH_3CN)(CO)](BPh_4)_2$.

Fe(1)-C(3)	1.795(7)	B(2)-C(50)	1.668(10)
Fe(1)-N(1)	1.990(5)	B(2)-C(56)	1.674(11)
Fe(1)-P(3)	2.297(2)	C(1)-C(2)	1.495(10)
Fe(1)-P(2)	2.302(2)	C(4)-C(5)	1.529(12)
Fe(1)-P(4)	2.321(2)	C(6)-C(7)	1.520(9)
Fe(1)-P(1)	2.379(2)	C(11)-C(12)	1.525(9)
P(1)-C(8)	1.845(7)	C(13)-C(14)	1.524(9)
P(1)-C(6)	1.848(7)	C(15)-C(16)	1.532(10)
P(1)-C(4)	1.861(7)	C(17)-C(18)	1.516(9)
P(2)-C(13)	1.807(6)	C(22)-C(23)	1.497(10)
P(2)-C(11)	1.837(6)	C(24)-C(25)	1.550(11)
P(2)-C(10)	1.846(7)	C(26)-C(31)	1.382(10)
P(3)-C(19)	1.842(7)	C(26)-C(27)	1.401(9)
P(3)-C(17)	1.850(7)	C(27)-C(28)	1.407(10)
P(3)-C(15)	1.857(7)	C(28)-C(29)	1.373(11)
P(4)-C(21)	1.835(6)	C(29)-C(30)	1.384(11)
P(4)-C(24)	1.837(7)	C(30)-C(31)	1.410(10)
P(4)-C(22)	1.842(7)	C(32)-C(33)	1.409(9)
N(1)-C(1)	1.118(8)	C(32)-C(37)	1.417(9)
N(2)-C(8)	1.444(8)	C(33)-C(34)	1.412(11)
N(2)-C(10)	1.452(8)	C(34)-C(35)	1.380(11)
N(2)-C(9)	1.475(9)	C(35)-C(36)	1.380(11)
N(3)-C(21)	1.462(8)	C(36)-C(37)	1.367(9)
N(3)-C(20)	1.462(8)	C(38)-C(43)	1.391(9)
N(3)-C(19)	1.469(8)	C(38)-C(39)	1.399(9)
O(1)-C(3)	1.142(7)	C(39)-C(40)	1.402(10)
B(1)-C(44)	1.636(11)	C(40)-C(41)	1.368(9)
B(1)-C(38)	1.650(10)	C(41)-C(42)	1.372(10)
B(1)-C(26)	1.651(10)	C(42)-C(43)	1.400(10)
B(1)-C(32)	1.663(10)	C(44)-C(45)	1.418(9)
B(2)-C(62)	1.642(11)	C(44)-C(49)	1.419(9)
B(2)-C(68)	1.642(11)	C(45)-C(46)	1.393(9)
		C(46)-C(47)	1.380(10)
		C(47)-C(48)	1.366(10)
		C(48)-C(49)	1.386(9)

C(50)-C(51)	1.395(9)	N(1)-Fe(1)-P(1)	87.93(17)
C(50)-C(55)	1.412(10)	P(3)-Fe(1)-P(1)	97.36(8)
C(51)-C(52)	1.403(10)	P(2)-Fe(1)-P(1)	87.62(7)
C(52)-C(53)	1.395(11)	P(4)-Fe(1)-P(1)	94.02(7)
C(53)-C(54)	1.368(11)	C(8)-P(1)-C(6)	100.5(3)
C(54)-C(55)	1.394(9)	C(8)-P(1)-C(4)	101.1(3)
C(56)-C(57)	1.402(11)	C(6)-P(1)-C(4)	106.1(3)
C(56)-C(61)	1.403(10)	C(8)-P(1)-Fe(1)	118.7(2)
C(57)-C(58)	1.433(10)	C(6)-P(1)-Fe(1)	116.2(2)
C(58)-C(59)	1.377(12)	C(4)-P(1)-Fe(1)	112.2(3)
C(59)-C(60)	1.387(14)	C(13)-P(2)-C(11)	104.3(3)
C(60)-C(61)	1.416(11)	C(13)-P(2)-C(10)	100.5(3)
C(62)-C(67)	1.397(9)	C(11)-P(2)-C(10)	101.1(3)
C(62)-C(63)	1.407(11)	C(13)-P(2)-Fe(1)	117.1(2)
C(63)-C(64)	1.409(11)	C(11)-P(2)-Fe(1)	111.3(3)
C(64)-C(65)	1.376(11)	C(10)-P(2)-Fe(1)	120.3(2)
C(65)-C(66)	1.391(12)	C(19)-P(3)-C(17)	99.6(3)
C(66)-C(67)	1.389(10)	C(19)-P(3)-C(15)	101.3(3)
C(68)-C(69)	1.389(9)	C(17)-P(3)-C(15)	101.4(3)
C(68)-C(73)	1.419(10)	C(19)-P(3)-Fe(1)	118.7(2)
C(69)-C(70)	1.394(10)	C(17)-P(3)-Fe(1)	117.3(2)
C(70)-C(71)	1.389(10)	C(15)-P(3)-Fe(1)	115.6(2)
C(71)-C(72)	1.393(10)	C(21)-P(4)-C(24)	102.4(3)
C(72)-C(73)	1.382(11)	C(21)-P(4)-C(22)	100.0(3)
		C(24)-P(4)-C(22)	102.6(3)
C(3)-Fe(1)-N(1)	87.4(3)	C(21)-P(4)-Fe(1)	119.6(2)
C(3)-Fe(1)-P(3)	87.7(2)	C(24)-P(4)-Fe(1)	115.4(3)
N(1)-Fe(1)-P(3)	173.4(2)	C(22)-P(4)-Fe(1)	114.3(2)
C(3)-Fe(1)-P(2)	87.1(2)	C(1)-N(1)-Fe(1)	166.5(7)
N(1)-Fe(1)-P(2)	88.7(2)	C(8)-N(2)-C(10)	111.3(5)
P(3)-Fe(1)-P(2)	95.42(7)	C(8)-N(2)-C(9)	111.5(5)
C(3)-Fe(1)-P(4)	90.7(2)	C(10)-N(2)-C(9)	111.5(5)
N(1)-Fe(1)-P(4)	84.8(2)	C(21)-N(3)-C(20)	110.5(5)
P(3)-Fe(1)-P(4)	90.89(7)	C(21)-N(3)-C(19)	111.6(5)
P(2)-Fe(1)-P(4)	173.22(7)	C(20)-N(3)-C(19)	109.4(5)
C(3)-Fe(1)-P(1)	173.0(2)	C(44)-B(1)-C(38)	112.2(5)

C(44)-B(1)-C(26)	106.7(6)	C(32)-C(33)-C(34)	121.3(8)
C(38)-B(1)-C(26)	110.9(6)	C(35)-C(34)-C(33)	121.4(7)
C(44)-B(1)-C(32)	109.9(5)	C(34)-C(35)-C(36)	118.8(7)
C(38)-B(1)-C(32)	104.4(5)	C(37)-C(36)-C(35)	119.5(8)
C(26)-B(1)-C(32)	112.8(5)	C(36)-C(37)-C(32)	125.1(7)
C(62)-B(2)-C(68)	114.4(6)	C(43)-C(38)-C(39)	114.8(6)
C(62)-B(2)-C(50)	112.1(6)	C(43)-C(38)-B(1)	125.5(6)
C(68)-B(2)-C(50)	103.8(5)	C(39)-C(38)-B(1)	119.7(6)
C(62)-B(2)-C(56)	105.6(6)	C(38)-C(39)-C(40)	123.0(6)
C(68)-B(2)-C(56)	113.3(6)	C(41)-C(40)-C(39)	119.5(7)
C(50)-B(2)-C(56)	107.7(5)	C(40)-C(41)-C(42)	119.9(7)
N(1)-C(1)-C(2)	174.8(8)	C(41)-C(42)-C(43)	119.8(7)
O(1)-C(3)-Fe(1)	176.2(6)	C(38)-C(43)-C(42)	123.0(7)
C(5)-C(4)-P(1)	119.2(6)	C(45)-C(44)-C(49)	113.4(7)
C(7)-C(6)-P(1)	117.2(5)	C(45)-C(44)-B(1)	120.7(6)
N(2)-C(8)-P(1)	112.1(5)	C(49)-C(44)-B(1)	125.9(6)
N(2)-C(10)-P(2)	113.0(5)	C(46)-C(45)-C(44)	124.0(7)
C(12)-C(11)-P(2)	114.7(5)	C(47)-C(46)-C(45)	118.8(7)
C(14)-C(13)-P(2)	118.5(5)	C(48)-C(47)-C(46)	120.1(7)
C(16)-C(15)-P(3)	118.3(5)	C(47)-C(48)-C(49)	120.7(7)
C(18)-C(17)-P(3)	117.3(5)	C(48)-C(49)-C(44)	122.9(6)
N(3)-C(19)-P(3)	114.1(4)	C(51)-C(50)-C(55)	115.9(6)
N(3)-C(21)-P(4)	111.9(4)	C(51)-C(50)-B(2)	120.1(6)
C(23)-C(22)-P(4)	117.9(6)	C(55)-C(50)-B(2)	123.8(6)
C(25)-C(24)-P(4)	116.9(6)	C(50)-C(51)-C(52)	121.6(7)
C(31)-C(26)-C(27)	115.9(7)	C(53)-C(52)-C(51)	121.2(7)
C(31)-C(26)-B(1)	119.8(6)	C(54)-C(53)-C(52)	117.7(7)
C(27)-C(26)-B(1)	123.9(6)	C(53)-C(54)-C(55)	121.5(7)
C(26)-C(27)-C(28)	121.9(7)	C(54)-C(55)-C(50)	122.0(7)
C(29)-C(28)-C(27)	121.0(8)	C(57)-C(56)-C(61)	116.1(7)
C(28)-C(29)-C(30)	118.0(8)	C(57)-C(56)-B(2)	124.6(7)
C(29)-C(30)-C(31)	120.8(8)	C(61)-C(56)-B(2)	119.2(6)
C(26)-C(31)-C(30)	122.3(7)	C(56)-C(57)-C(58)	122.5(9)
C(33)-C(32)-C(37)	113.9(6)	C(59)-C(58)-C(57)	118.7(9)
C(33)-C(32)-B(1)	125.8(6)	C(58)-C(59)-C(60)	120.9(9)
C(37)-C(32)-B(1)	120.3(6)	C(59)-C(60)-C(61)	119.3(10)

C(56)-C(61)-C(60)	122.4(8)
C(67)-C(62)-C(63)	114.4(7)
C(67)-C(62)-B(2)	122.7(7)
C(63)-C(62)-B(2)	122.9(6)
C(62)-C(63)-C(64)	123.0(7)
C(65)-C(64)-C(63)	120.0(8)
C(64)-C(65)-C(66)	118.8(8)
C(67)-C(66)-C(65)	120.2(7)
C(66)-C(67)-C(62)	123.6(8)
C(69)-C(68)-C(73)	114.1(7)
C(69)-C(68)-B(2)	122.8(7)
C(73)-C(68)-B(2)	122.9(6)
C(68)-C(69)-C(70)	124.0(7)
C(71)-C(70)-C(69)	120.4(7)
C(70)-C(71)-C(72)	117.4(8)
C(73)-C(72)-C(71)	121.3(8)
C(72)-C(73)-C(68)	122.8(7)

Symmetry transformations used to generate equivalent atoms:

Table S9. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for $[Fe(PNP)_2(CH_3CN)(CO)](BPh_4)_2$.
The anisotropic displacement factor exponent takes the form: $-2\pi^2[\ h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
Fe(1)	26(1)	34(1)	12(1)	-2(1)	-3(1)	-1(1)
P(1)	30(1)	44(1)	21(1)	-5(1)	0(1)	-3(1)
P(2)	36(1)	40(1)	18(1)	1(1)	0(1)	-1(1)
P(3)	29(1)	38(1)	12(1)	1(1)	-3(1)	4(1)
P(4)	35(1)	37(1)	18(1)	1(1)	0(1)	4(1)
N(1)	17(3)	32(3)	68(5)	6(3)	8(3)	7(2)
N(2)	39(4)	53(4)	16(3)	-1(3)	-1(3)	-12(3)
N(3)	49(4)	40(3)	2(3)	3(2)	-6(3)	13(3)
O(1)	38(3)	54(3)	26(3)	-7(2)	0(3)	-9(2)
B(1)	35(5)	36(4)	20(5)	3(3)	7(4)	-3(3)
B(2)	36(5)	43(5)	18(4)	-1(4)	2(4)	5(4)
C(1)	43(5)	50(5)	12(4)	5(3)	2(4)	1(4)
C(2)	46(5)	50(5)	33(5)	-6(4)	-3(4)	9(4)
C(3)	30(4)	43(4)	12(4)	3(3)	-7(3)	-2(3)
C(4)	47(5)	39(4)	54(6)	-12(4)	5(5)	-3(3)
C(5)	124(10)	56(6)	42(6)	-6(5)	-2(6)	12(5)
C(6)	36(5)	49(4)	24(4)	0(3)	-6(4)	-1(3)
C(7)	49(6)	90(6)	21(5)	6(4)	0(4)	-21(4)
C(8)	37(4)	40(4)	13(4)	-8(3)	-10(3)	-5(3)
C(9)	47(5)	65(5)	27(5)	-9(4)	1(4)	-13(4)
C(10)	29(4)	44(4)	29(5)	6(3)	-6(4)	1(3)
C(11)	40(5)	57(5)	27(5)	-2(3)	16(4)	-3(3)
C(12)	44(5)	49(4)	28(4)	-3(4)	9(4)	0(3)
C(13)	47(5)	40(4)	23(4)	5(3)	8(4)	5(3)
C(14)	52(5)	32(4)	27(5)	6(3)	4(4)	-5(3)
C(15)	47(5)	32(4)	20(4)	-4(3)	-1(4)	5(3)
C(16)	69(6)	43(4)	32(5)	-16(4)	1(5)	-6(4)
C(17)	27(4)	44(4)	15(4)	2(3)	-7(3)	1(3)
C(18)	44(5)	63(5)	15(4)	4(4)	-3(4)	21(4)
C(19)	33(4)	41(4)	22(4)	-1(3)	0(3)	10(3)
C(20)	67(6)	57(5)	13(4)	2(3)	2(4)	20(4)

C(21)	39(4)	51(4)	12(4)	2(3)	-2(3)	5(3)
C(22)	36(5)	55(5)	22(4)	1(3)	-4(4)	-6(3)
C(23)	32(5)	85(6)	63(7)	16(5)	-11(5)	3(4)
C(24)	54(5)	47(4)	29(5)	8(4)	9(4)	8(4)
C(25)	133(10)	50(5)	39(6)	24(4)	25(6)	22(5)
C(26)	30(4)	48(4)	18(4)	2(3)	-2(3)	0(3)
C(27)	33(5)	57(5)	22(4)	5(4)	2(4)	-5(3)
C(28)	37(5)	58(5)	45(6)	10(4)	3(4)	-1(4)
C(29)	47(6)	57(5)	52(6)	-9(5)	6(5)	-1(4)
C(30)	62(7)	93(7)	20(5)	-12(5)	-4(5)	9(5)
C(31)	60(6)	67(5)	14(4)	0(4)	-5(4)	20(4)
C(32)	22(4)	57(5)	14(4)	13(3)	-5(3)	4(3)
C(33)	44(5)	84(6)	28(5)	26(4)	8(4)	17(4)
C(34)	42(6)	97(7)	45(6)	31(5)	21(5)	6(5)
C(35)	38(5)	71(6)	49(6)	31(5)	-8(5)	-1(4)
C(36)	44(5)	55(5)	21(5)	8(4)	-14(4)	3(4)
C(37)	44(5)	50(5)	18(4)	7(3)	-2(4)	-12(4)
C(38)	30(4)	41(4)	20(4)	1(3)	2(3)	0(3)
C(39)	20(4)	60(5)	28(5)	-3(3)	6(4)	-4(3)
C(40)	39(5)	62(5)	13(4)	6(3)	-8(4)	-9(3)
C(41)	55(6)	47(4)	14(4)	4(3)	5(4)	-7(3)
C(42)	38(5)	56(5)	33(5)	6(4)	11(4)	-2(3)
C(43)	34(5)	51(5)	25(4)	5(3)	6(4)	6(3)
C(44)	40(5)	38(4)	12(4)	3(3)	-4(3)	3(3)
C(45)	40(5)	45(4)	20(4)	9(3)	-2(4)	-4(3)
C(46)	30(5)	60(5)	33(5)	1(4)	2(4)	-12(3)
C(47)	28(4)	66(5)	25(5)	-4(4)	-8(4)	3(4)
C(48)	40(5)	41(4)	28(5)	6(3)	-5(4)	10(3)
C(49)	27(4)	37(4)	25(4)	-4(3)	3(3)	2(3)
C(50)	33(4)	42(4)	28(5)	-11(3)	5(4)	-9(3)
C(51)	34(5)	44(4)	40(5)	-12(4)	14(4)	-10(3)
C(52)	47(5)	71(6)	34(5)	-18(4)	16(5)	-16(4)
C(53)	42(5)	50(5)	49(6)	-19(4)	6(5)	-3(4)
C(54)	31(5)	41(4)	39(5)	1(4)	-11(4)	-7(3)
C(55)	39(5)	44(4)	22(4)	-4(3)	3(4)	-3(3)
C(56)	58(6)	39(4)	18(4)	-10(3)	7(4)	6(4)

C(57)	64(6)	54(5)	39(6)	-1(4)	14(5)	-10(4)
C(58)	111(9)	45(5)	35(6)	-5(4)	27(6)	-1(5)
C(59)	107(9)	52(6)	64(7)	-6(5)	54(7)	-14(6)
C(60)	104(9)	56(6)	74(8)	-27(5)	46(7)	-27(6)
C(61)	61(6)	40(4)	47(6)	-6(4)	19(5)	-3(4)
C(62)	33(5)	43(4)	38(5)	1(3)	8(4)	-7(3)
C(63)	43(5)	57(5)	40(5)	-10(4)	4(4)	5(4)
C(64)	42(5)	71(6)	38(6)	-4(4)	-9(4)	-9(4)
C(65)	76(7)	65(6)	34(5)	-16(4)	14(5)	-14(5)
C(66)	56(6)	49(5)	40(6)	-6(4)	21(5)	-4(4)
C(67)	38(5)	53(5)	27(5)	-6(4)	6(4)	-2(3)
C(68)	42(5)	42(4)	27(4)	4(3)	11(4)	3(3)
C(69)	47(5)	39(4)	20(4)	0(3)	4(4)	4(3)
C(70)	49(5)	51(5)	25(5)	-4(4)	11(4)	-9(4)
C(71)	39(5)	73(5)	26(5)	-4(4)	-3(4)	-5(4)
C(72)	39(5)	97(7)	23(5)	1(4)	-4(4)	0(4)
C(73)	48(5)	73(5)	21(5)	4(4)	10(4)	-7(4)

Table S10. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for $[Fe(PNP)_2(CH_3CN)(CO)](BPh_4)_2$.

	x	y	z	U(eq)
H(2A)	8380	14780	8075	66
H(2B)	7957	14467	7434	66
H(2C)	8449	15373	7541	66
H(4A)	10484	15205	7710	70
H(4B)	10072	14755	8071	70
H(5A)	10951	15018	8929	115
H(5B)	10734	16025	8585	115
H(5C)	11369	15462	8568	115
H(6A)	11520	13088	7478	57
H(6B)	10969	13767	7057	57
H(7A)	11499	15171	7475	79
H(7B)	12036	14481	7364	79
H(7C)	11995	14569	7996	79
H(8A)	11655	12700	8586	49
H(8B)	11728	13852	8718	49
H(9A)	12002	12952	9732	70
H(9B)	11435	13369	9940	70
H(9C)	11797	14083	9632	70
H(10A)	10884	12031	9462	53
H(10B)	11201	11708	8995	53
H(11A)	9764	12670	9283	56
H(11B)	9634	13454	8776	56
H(12A)	8694	12529	8231	58
H(12B)	8634	12959	8813	58
H(12C)	8811	11827	8775	58
H(13A)	9470	10605	8431	53
H(13B)	10215	10337	8563	53
H(14A)	10426	10587	9549	55
H(14B)	9864	9787	9305	55
H(14C)	9683	10886	9422	55

H(15A)	10361	9425	7393	51
H(15B)	9823	9891	7617	51
H(16A)	9144	10174	6660	73
H(16B)	9328	9036	6757	73
H(16C)	9685	9722	6432	73
H(17A)	11490	11480	8029	45
H(17B)	11178	10598	8271	45
H(18A)	11404	9558	7565	62
H(18B)	12030	9933	8076	62
H(18C)	11828	10446	7459	62
H(19A)	11012	11657	6792	48
H(19B)	10800	10536	6665	48
H(20A)	10666	11313	5725	68
H(20B)	9909	11420	5364	68
H(20C)	10184	10413	5689	68
H(21A)	9785	12751	5789	51
H(21B)	10455	12876	6311	51
H(22A)	8685	12810	5802	58
H(22B)	8705	11917	6223	58
H(23A)	8195	12840	6721	93
H(23B)	7778	12903	6051	93
H(23C)	8235	13800	6362	93
H(24A)	9303	14615	6766	63
H(24B)	10023	14562	6743	63
H(25A)	9597	14458	5723	106
H(25B)	9338	15431	5936	106
H(25C)	8873	14495	5742	106
H(27)	13183	14096	8370	47
H(28)	12904	15562	8716	60
H(29)	13005	15727	9679	66
H(30)	13403	14395	10297	75
H(31)	13663	12916	9948	62
H(33)	12719	11895	9420	64
H(34)	12254	10421	9576	72
H(35)	12530	8931	9259	71
H(36)	13328	8905	8823	55

H(37)	13777	10341	8662	49
H(39)	12561	12305	8008	44
H(40)	12270	12306	7010	51
H(41)	13072	12331	6590	49
H(42)	14161	12316	7166	51
H(43)	14448	12306	8162	45
H(45)	14734	13512	8962	46
H(46)	15854	13448	9411	52
H(47)	16305	12210	10082	53
H(48)	15652	10980	10229	49
H(49)	14545	10979	9739	38
H(51)	8757	8049	9490	47
H(52)	8299	9459	9727	60
H(53)	7892	10719	9051	59
H(54)	8010	10569	8156	51
H(55)	8511	9209	7927	44
H(57)	9700	5796	9193	64
H(58)	9305	4417	9560	76
H(59)	8198	4299	9414	82
H(60)	7471	5457	8849	89
H(61)	7872	6846	8513	59
H(63)	7927	7676	7605	59
H(64)	7639	7323	6626	67
H(65)	8427	6782	6251	71
H(66)	9479	6450	6884	57
H(67)	9752	6771	7854	49
H(69)	10031	8344	8195	45
H(70)	11098	8814	8652	50
H(71)	11618	8493	9636	60
H(72)	11005	7756	10144	68
H(73)	9941	7300	9686	58

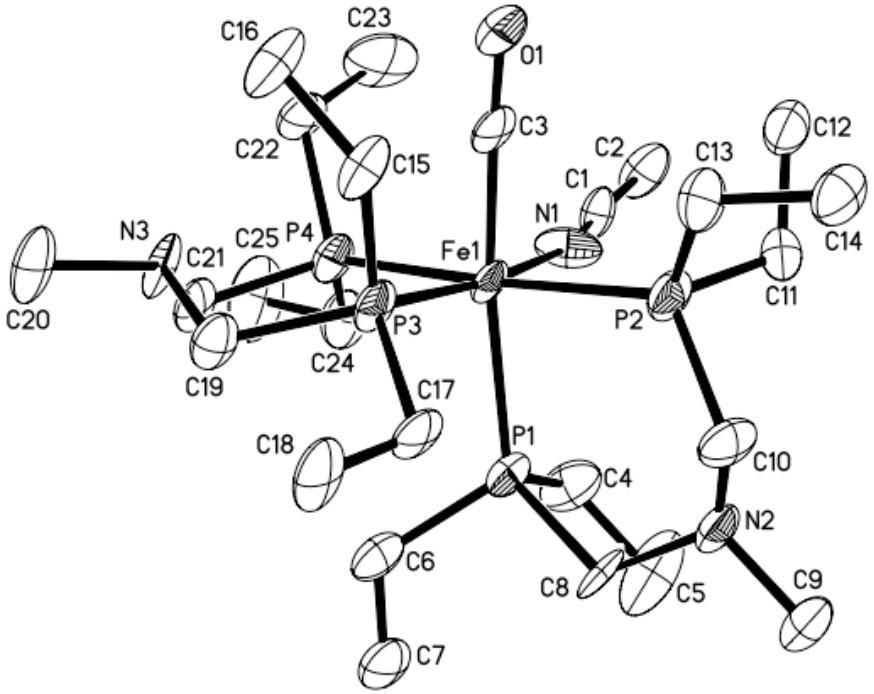

Figure S5. Drawing of the *cis*-[Fe(PNP)2(CH_3CN)(CO)]$^{2+}$ cation indicating the atom numbering scheme. Thermal ellipsoids are drawn at the 50% probability level.