Electronic Supplementary Information for Dalton Transactions

New iridium and rhodium chiral di-*N*-heterocyclic carbene (NHC) complexes and their application in enantioselective catalysis.

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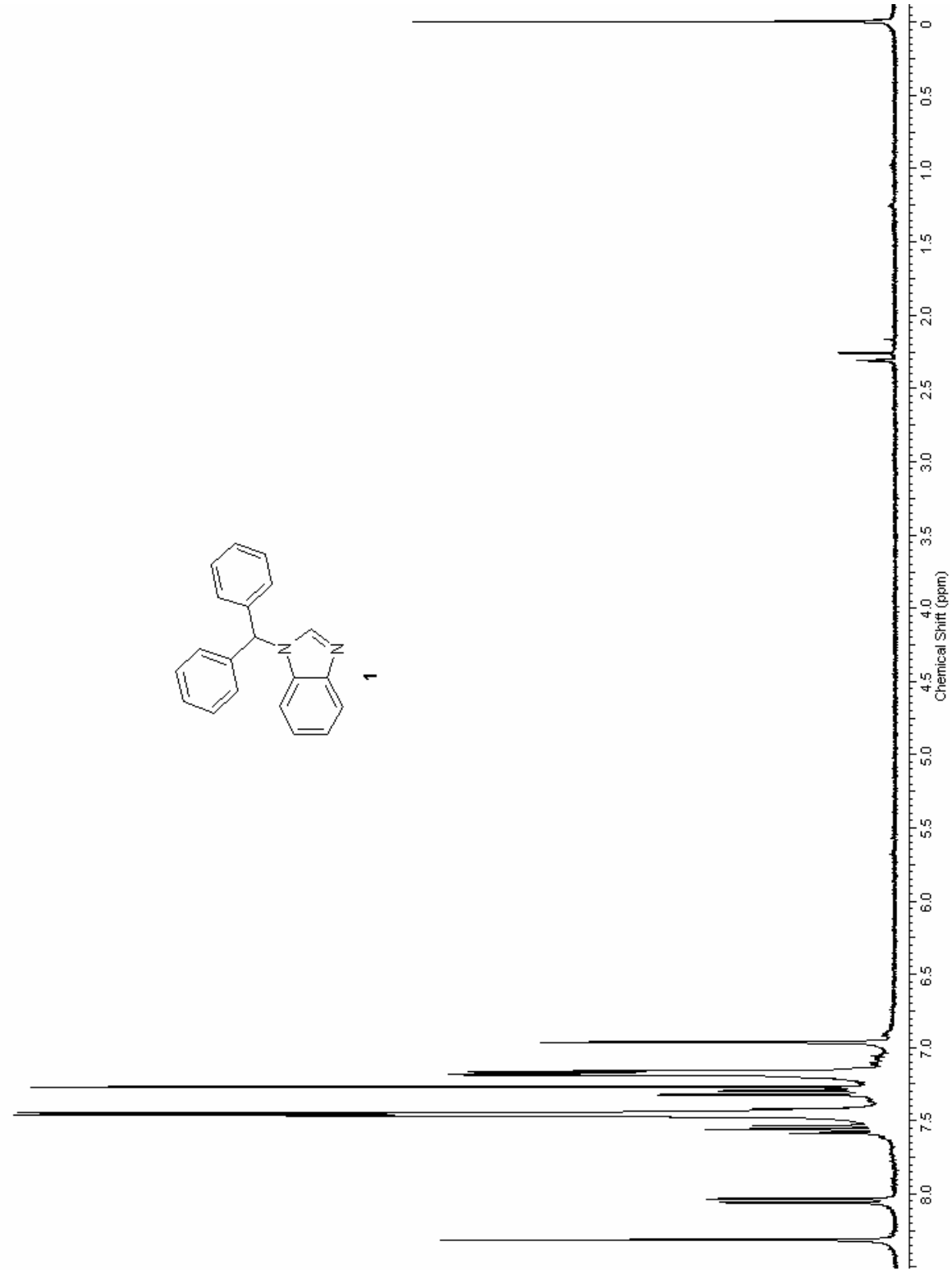

Figure S1: ^{1}H NMR spectrum of 1-diphenylmethanebenzimidazole (**1**) in $CDCl_3$.

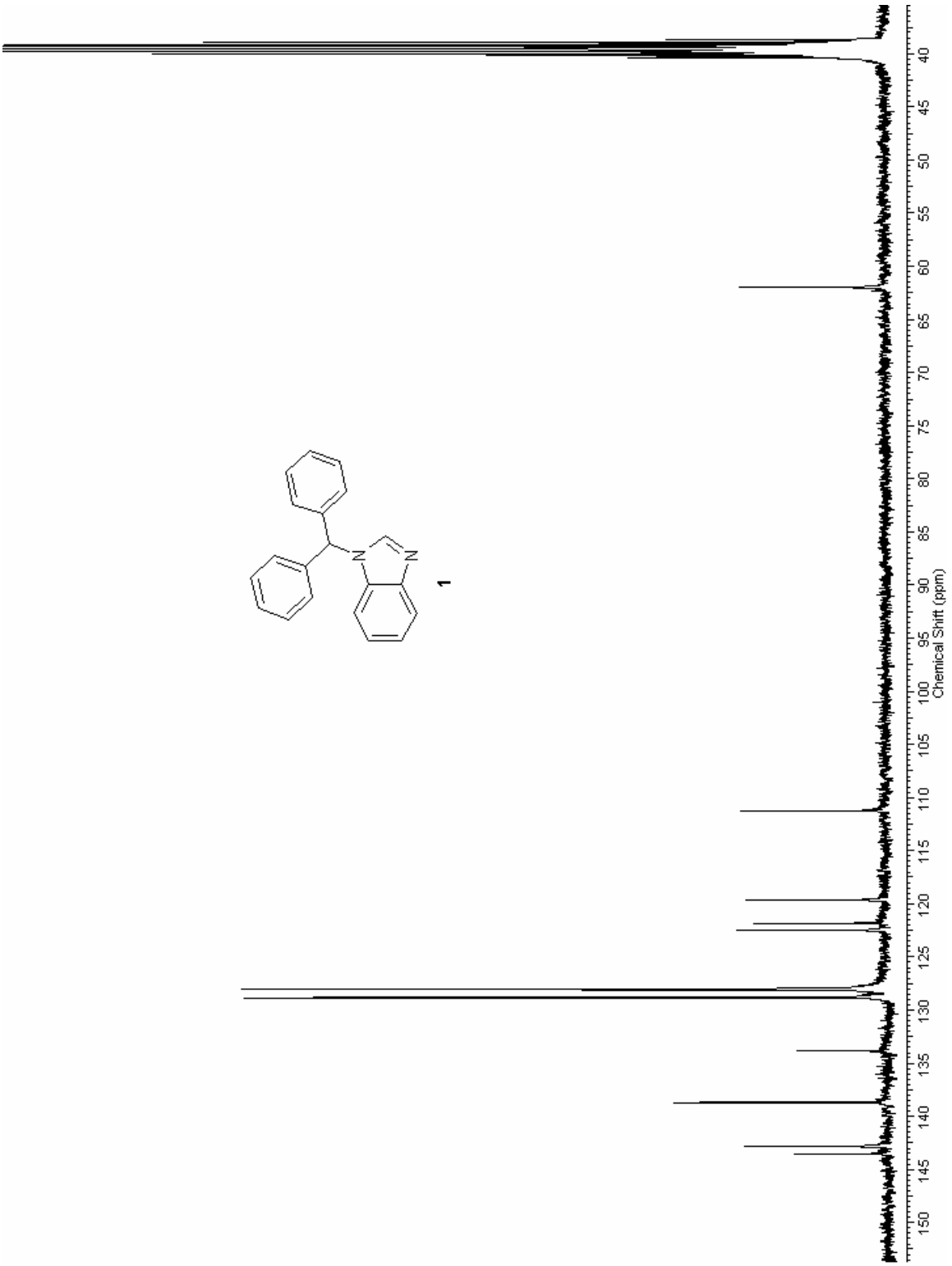

Figure S2: $^{13}C\{^{1}H\}$ NMR spectrum of 1-diphenylmethanebenzimidazole (**1**) in $(CD_3)_2SO$.

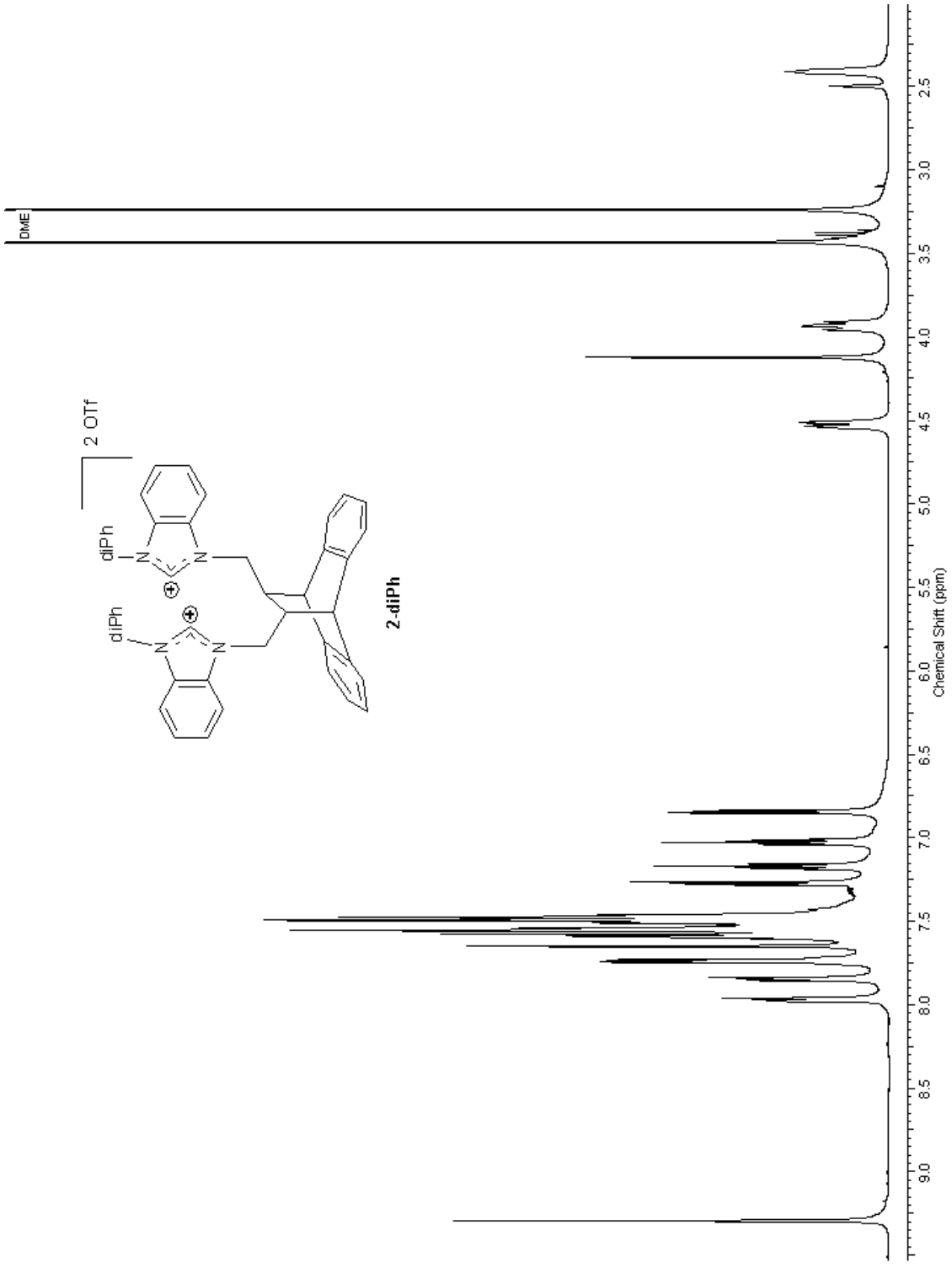

Figure S3: ^{1}H NMR spectrum of (DEAM-diPhBI)$(OTf)_2$ (**2-diPh**) in $(CD_3)_2SO$.

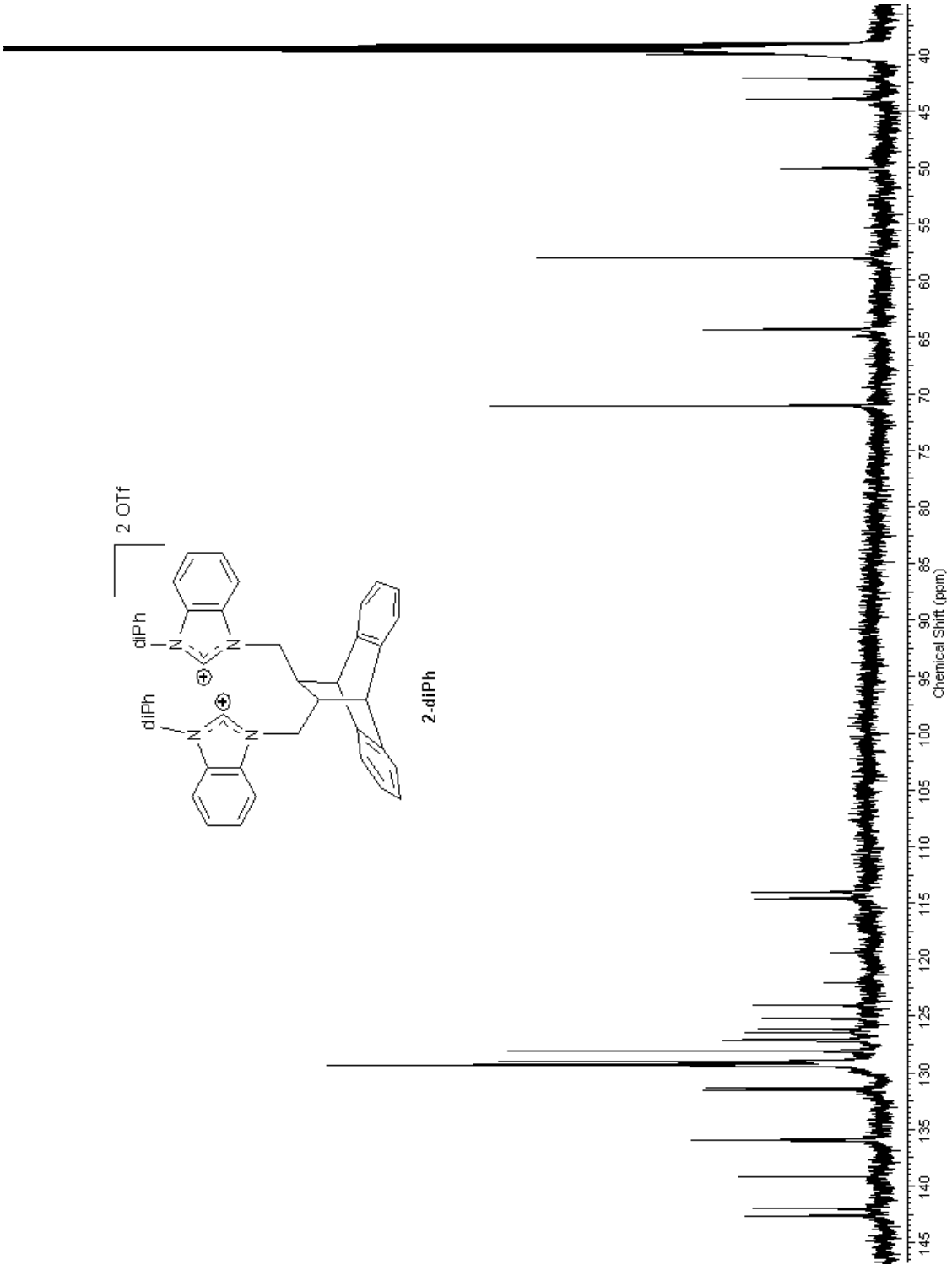

Figure S4: $^{13}C\{^{1}H\}$ NMR spectrum of (DEAM-diPhBI)$(OTf)_2$ (**2-diPh**) in $(CD_3)_2SO$.

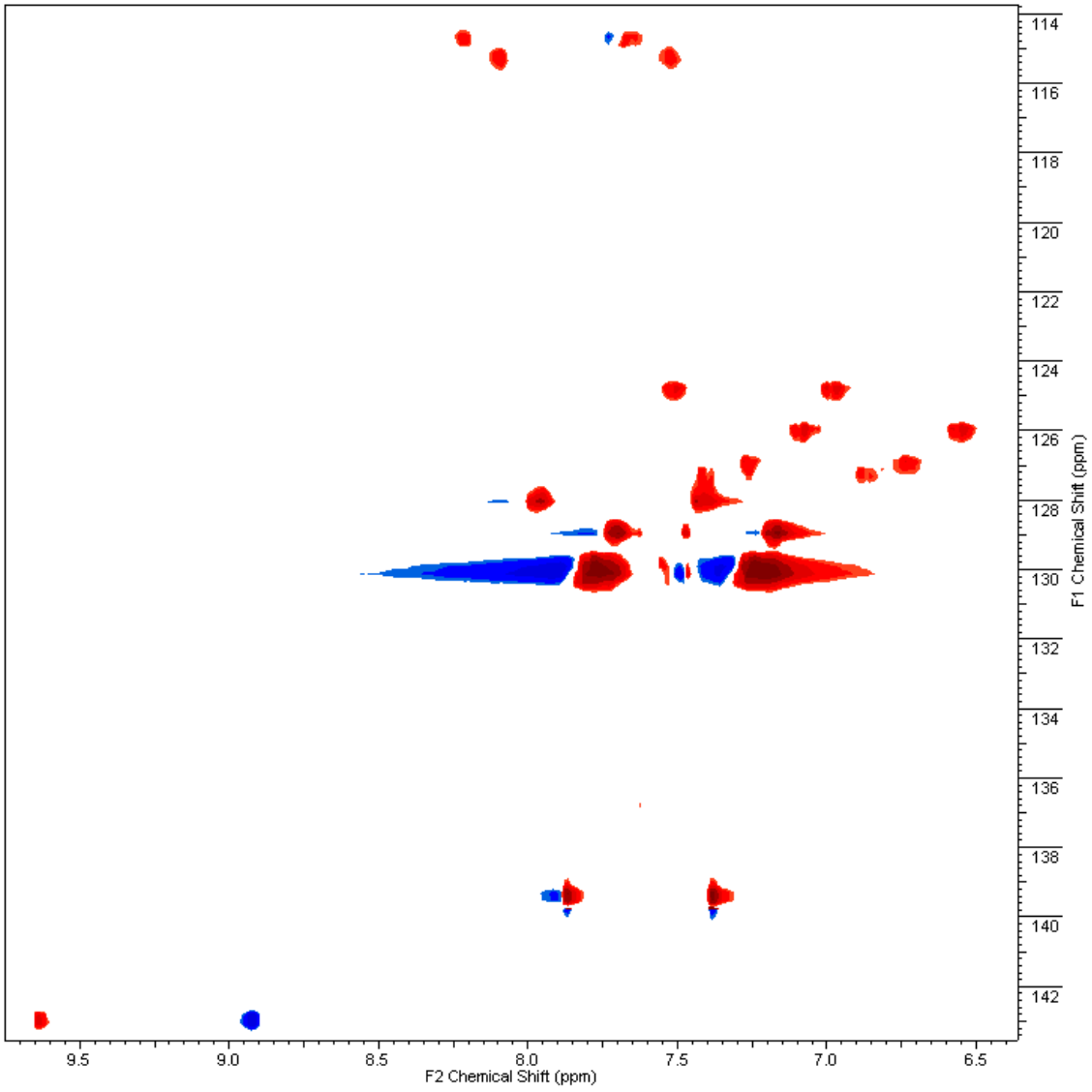

Figure S5: gHMQC NMR spectrum of (DEAM-diPhBI)$(OTf)_2$ (**2-diPh**) in $(CD_3)_2SO$.

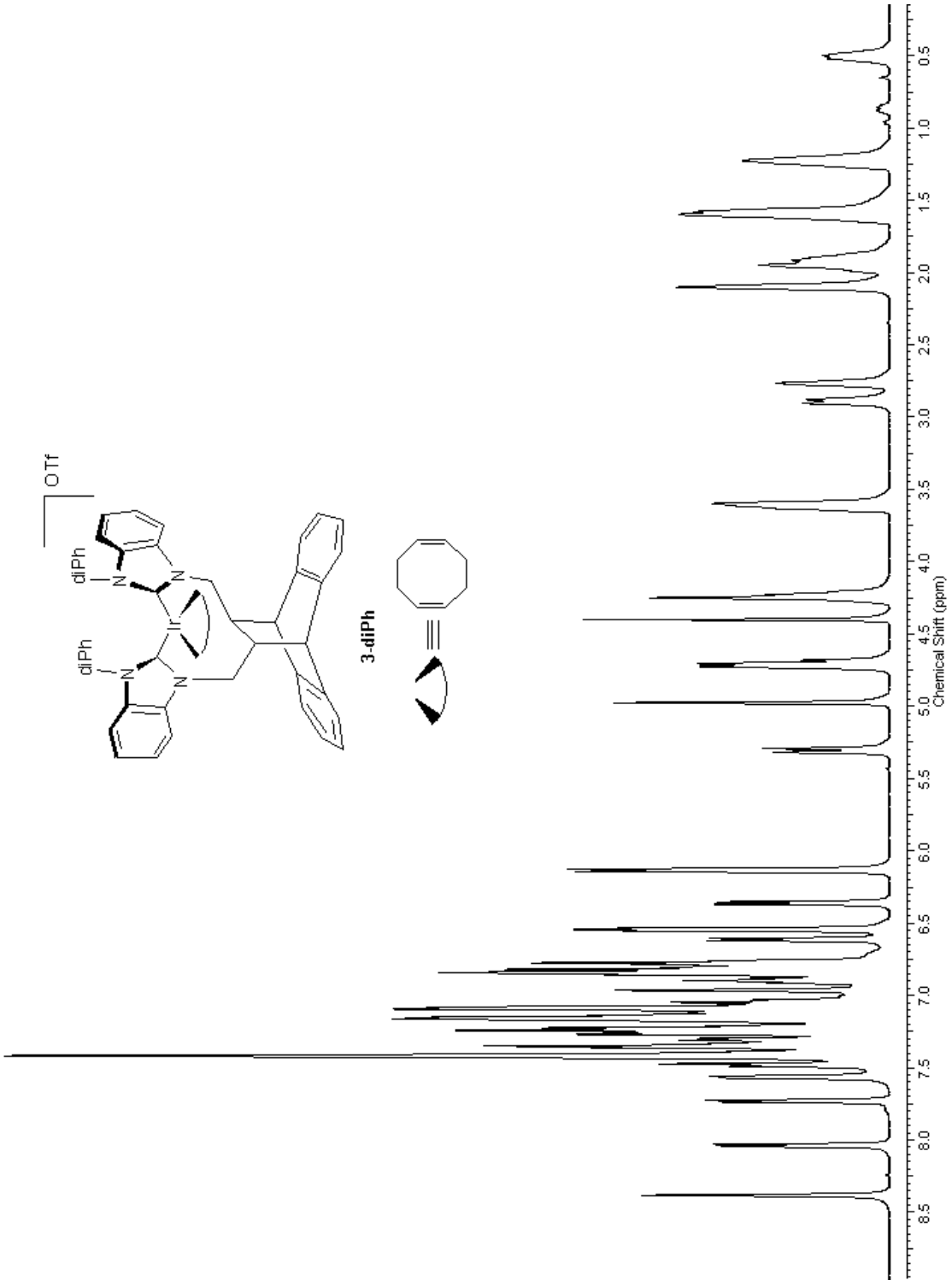

Figure S6: ^{1}H NMR spectrum of [(DEAM-diPhBY)Ir(cod)](OTf) (**3-diPh**) in $CDCl_3$.

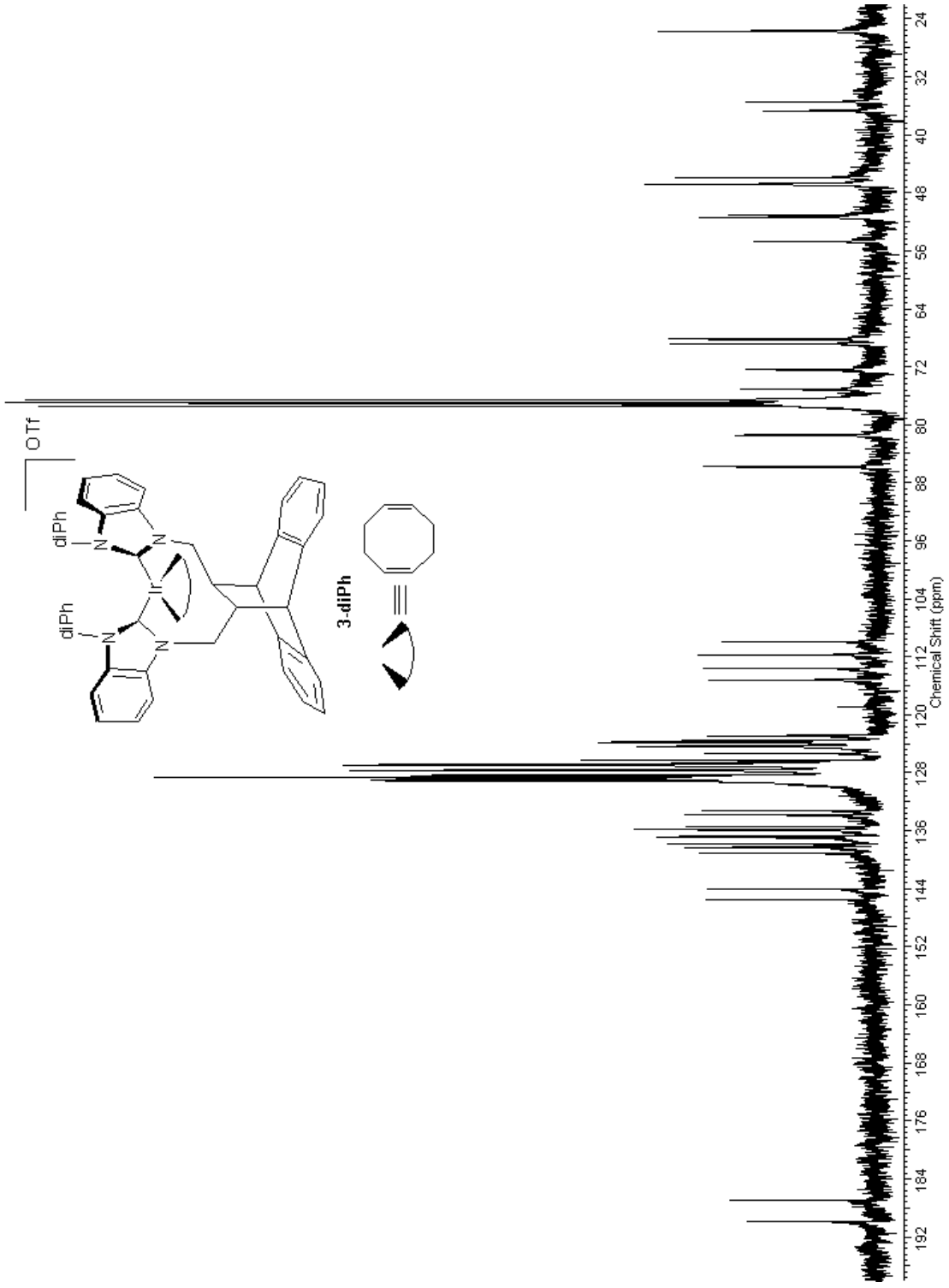

Figure S7: $^{13}C\{^{1}H\}$ NMR spectrum of [(DEAM-diPhBY)Ir(cod)](OTf) (**3-diPh**) in $CDCl_3$.

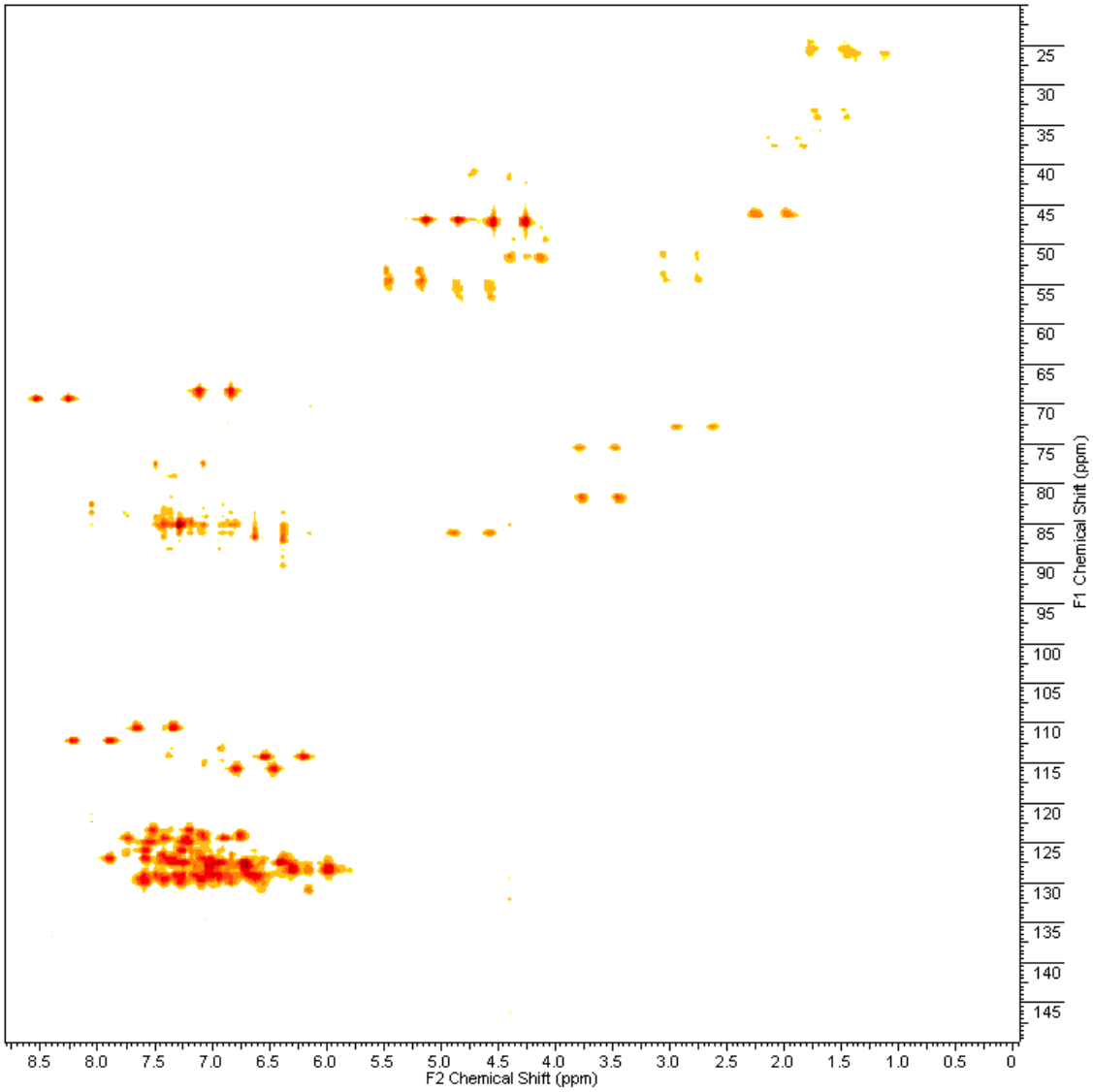

Figure S8: gHMQC NMR spectrum of [(DEAM-diPhBY)Ir(cod)](OTf) (**3-diPh**) in $CDCl_3$.

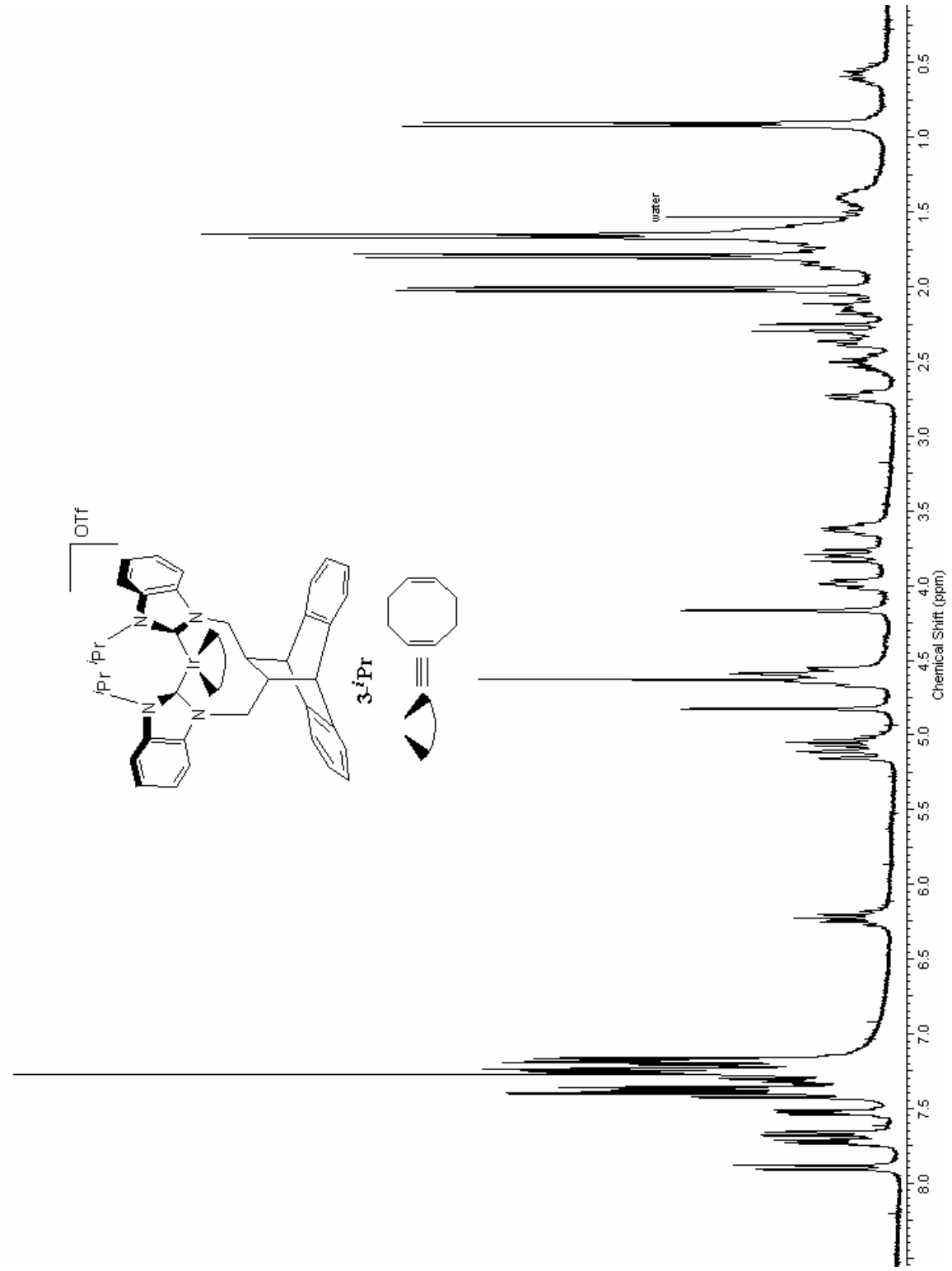

Figure S9: ^{1}H NMR spectrum of [(DEAM-IBY)Ir(cod)](OTf) (**3-*i*Pr**) in $CDCl_3$.

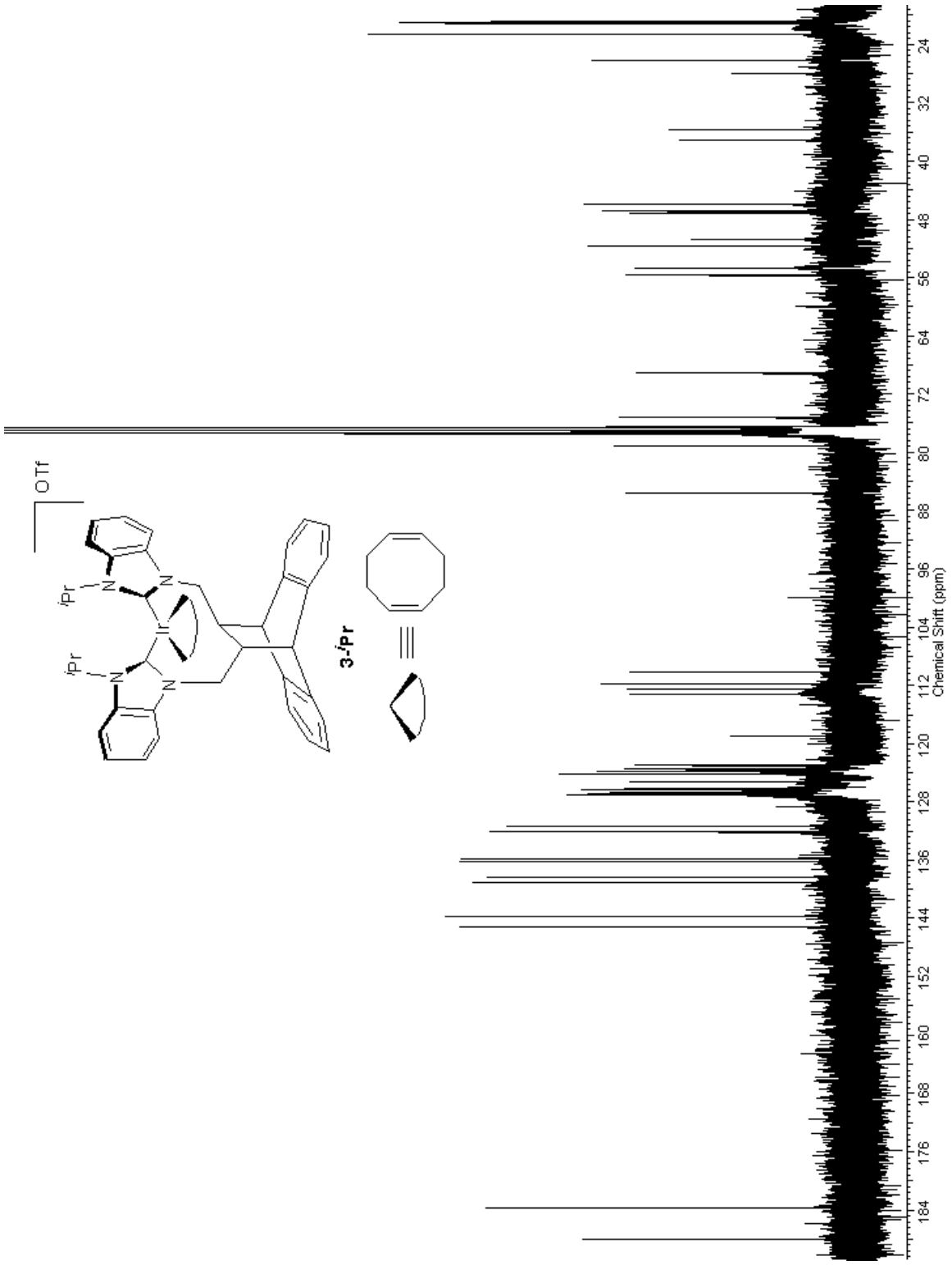

Figure S10: $^{13}C\{^{1}H\}$ NMR spectrum of [(DEAM-IPY)Ir(cod)](OTf) (**3-*i*Pr**) in $CDCl_3$.

X–ray experimental details for [(DEAM-IBY)Ir(cod)](OTf) (3-^{i}Pr):

Data were collected at 173 K on a Siemens SMART PLATFORM equipped with A CCD area detector and a graphite monochromator utilizing MoKα radiation (λ = 0.71073 Å). Cell parameters were refined using up to 8192 reflections. A full sphere of data (1850 frames) was collected using the ω-scan method (0.3° frame width). The first 50 frames were re-measured at the end of data collection to monitor instrument and crystal stability (maximum correction on I was < 1 %). Absorption corrections by integration were applied based on measured indexed crystal faces.

The structure was solved by the Direct Methods in *SHELXTL6,* and refined using full-matrix least squares. The non-H atoms were treated anisotropically, whereas the hydrogen atoms were calculated in ideal positions and were riding on their respective carbon atoms. The asymmetric unit consists of two iridium complex cations, two triflate anions, two toluene solvent molecules and two chloroform solvent molecules. The solvent molecules were disordered and could not be modeled properly, thus program SQUEEZE, a part of the PLATON package of crystallographic software, was used to calculate the solvent disorder area and remove its contribution to the overall intensity data. Space group P2/n was checked to confirm the correct space group. The complexes are not related by a two-fold rotation symmetry or an inversion center. Thus the correct space group is the reported Pn. A total of 1064 parameters were refined in the final cycle of refinement using 19094 reflections with I > 2σ(I) to yield R_1 and wR_2 of 7.50% and 11.54%, respectively. Refinement was done using F^2.

P. van der Sluis & A.L. Spek (1990). SQUEEZE, Acta Cryst. A46, 194-201

SHELXTL6 (2000). Bruker-AXS, Madison, Wisconsin, USA.

Spek, A.L. (1990). PLATON, Acta Cryst. A46, C-34

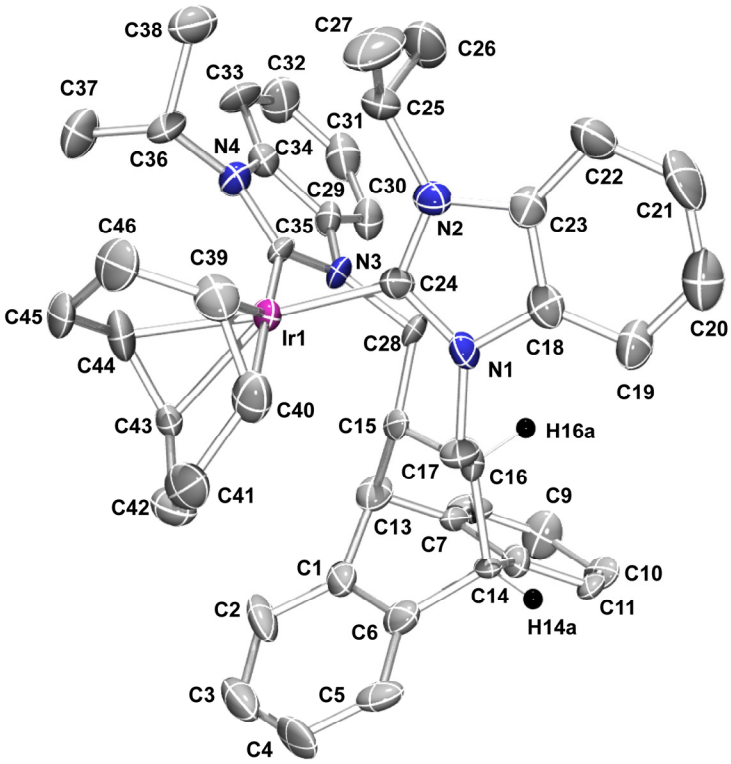

Figure S11. Molecular Structure of [(DEAM-IBY)Ir(cod)](OTf) (**3-^{i}Pr**) with ellipsoids drawn at the 50% probability level. Hydrogen atoms and OTf counter ion have been omitted for clarity.

Table S1. Crystal data, structure solution and refinement for [(DEAM-IBY)Ir(cod)](OTf) (**3-^{i}Pr**).

identification code	mj25
empirical formula	$C_{119}H_{117}Cl_6N_8Ir_2F_6O_6S_2$
formula weight	2530.43
T (K)	173(2)
λ (Å)	0.71073
crystal system	monoclinic
space group	Pn
A (Å)	13.2280(13)
B (Å)	20.306(2)
c (Å)	18.9627(19)
α (deg)	90
β (deg)	97.224(2)
γ (deg)	90
V (Å^3)	5053.2(9)
Z	2
ρ_{calcd} (Mg mm^{-3})	1.663
crystal size (mm^3)	0.11 x 0.11 x 0.02
abs coeff (mm^{-1})	2.906
F(000)	2554
θ range for data collection	1.48 to 27.50
limiting indices	$-17 \le h \le 16, -24 \le k \le 26, -24 \le \ell \le 23$
no. of reflns collcd	34294
no. of ind reflns (R_{int})	19094 (0.0691)
completeness to θ = 27.45°	99.6 %
absorption corr	Integration
refinement method	Full–matrix least–squares on F^2
data / restraints / parameters	19094 / 2 / 1064
$R1$,[a] $wR2$[b] [I > 2σ]	0.0560, 0.1154
$R1$,[a] $wR2$[b] (all data)	0.0750, 0.1212
GOF[c] on F^2	0.980
largest diff. peak and hole	1.551 and –1.072 eÅ^{-3}

$R1 = \Sigma(||F_o| - |F_c||) / \Sigma|F_o|$

$wR2 = [\Sigma[w(F_o^2 - F_c^2)^2] / \Sigma[w(F_o^2)^2]]^{1/2}$

$S = [\Sigma[w(F_o^2 - F_c^2)^2] / (n-p)]^{1/2}$

$w = 1/[\sigma^2(F_o^2)+(m*p)^2+n*p]$, $p = [\max(F_o^2,0)+ 2* F_c^2]/3$, m & n are constants.

Table S2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^2 x 10^3) for [(DEAM-IBY)Ir(cod)](OTf) (**3-*i*Pr**). U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

Atom	X	Y	Z	U(eq)
Ir1	8099(1)	7009(1)	6254(1)	28(1)
Ir2	8567(1)	1026(1)	6564(1)	25(1)
N1	6115(6)	6244(4)	6029(4)	28(2)
N2	6870(6)	6200(4)	5088(4)	31(2)
N3	6793(5)	8173(4)	5562(4)	32(2)
N4	8304(6)	8166(4)	5204(4)	32(2)
N5	6593(5)	1183(4)	5552(4)	29(2)
N6	7893(5)	1418(4)	5009(4)	29(2)
N7	8322(5)	2491(4)	6938(4)	28(2)
N8	9917(5)	2252(4)	6944(4)	24(2)
O1	4140(7)	5464(6)	7643(7)	97(4)
F1	3060(10)	5737(5)	8831(5)	129(4)
S1	3069(2)	5449(2)	7482(2)	54(1)
O2	2666(9)	5815(7)	6885(6)	117(5)
F2	2799(9)	6540(4)	8171(6)	122(4)
O3	2616(8)	4832(5)	7518(6)	85(3)
F3	1657(7)	5824(5)	8233(6)	117(4)
C93	2630(9)	5907(6)	8203(6)	50(3)
S2	2508(2)	743(1)	5543(1)	31(1)
O4	1491(5)	789(4)	5158(5)	51(2)
F4	3093(5)	156(4)	4435(3)	64(2)
O5	3188(5)	1277(4)	5410(4)	41(2)
F5	2488(6)	-506(3)	5174(4)	58(2)
O6	2553(5)	563(3)	6279(3)	39(2)
F6	3995(5)	-83(4)	5428(4)	66(2)
C94	3030(8)	30(6)	5129(6)	47(3)
C1	6059(6)	8012(5)	7820(5)	36(2)
C2	6801(7)	8266(7)	8323(6)	55(3)
C3	7021(8)	7934(8)	8984(6)	63(4)
C4	6534(7)	7369(7)	9105(6)	54(3)
C5	5779(7)	7122(6)	8597(5)	41(3)

C6	5545(7)	7425(5)	7963(5)	36(2)
C7	4590(6)	8383(5)	7011(4)	28(2)
C8	4086(9)	8964(5)	6828(5)	41(2)
C9	3038(8)	8987(5)	6791(7)	47(3)
C10	2505(6)	8438(5)	6958(5)	34(2)
C11	3004(6)	7839(5)	7144(5)	27(2)
C12	4043(6)	7813(5)	7188(5)	29(2)
C13	5737(7)	8264(5)	7072(5)	33(2)
C14	4737(6)	7220(5)	7368(4)	21(2)
C15	5944(6)	7706(4)	6564(4)	23(2)
C16	5250(6)	7110(4)	6684(5)	26(2)
C17	5812(7)	6441(5)	6722(5)	34(2)
C18	5493(7)	5867(5)	5549(6)	34(2)
C19	4570(7)	5544(5)	5603(6)	40(2)
C20	4182(8)	5165(6)	5034(7)	52(3)
C21	4667(9)	5112(6)	4438(7)	61(3)
C22	5592(9)	5435(6)	4360(6)	50(3)
C23	5971(7)	5809(5)	4940(5)	33(2)
C24	6943(7)	6448(5)	5735(5)	29(2)
C25	7618(8)	6334(6)	4571(5)	47(3)
C26	7058(9)	6724(7)	3940(6)	62(4)
C27	8114(10)	5727(7)	4361(8)	75(4)
C28	5857(6)	7899(5)	5790(5)	29(2)
C29	6823(6)	8686(5)	5110(5)	31(2)
C30	6080(7)	9167(5)	4888(6)	39(2)
C31	6340(8)	9644(6)	4404(6)	51(3)
C32	7308(8)	9648(6)	4181(6)	51(3)
C33	8027(8)	9172(6)	4394(5)	50(3)
C34	7774(7)	8700(5)	4873(5)	35(2)
C35	7713(6)	7837(5)	5633(5)	27(2)
C36	9333(7)	7950(6)	5082(5)	41(3)
C37	10142(8)	8474(6)	5308(7)	58(3)
C38	9350(9)	7774(6)	4302(6)	54(3)
C39	9077(8)	6152(5)	6397(6)	43(3)
C40	8469(8)	6140(6)	6957(6)	44(3)
C41	8886(8)	6337(6)	7712(6)	50(3)

C42	8675(8)	7064(6)	7838(6)	47(3)
C43	8750(6)	7511(5)	7217(5)	32(2)
C44	9510(8)	7508(7)	6756(6)	57(3)
C45	10412(7)	7015(7)	6840(7)	55(4)
C46	10165(8)	6336(7)	6432(7)	54(3)
C47	5915(6)	1370(5)	7983(5)	31(2)
C48	6246(6)	974(5)	8574(5)	33(2)
C49	5748(7)	403(5)	8667(5)	37(2)
C50	4918(7)	195(5)	8202(6)	39(2)
C51	4546(8)	588(6)	7619(6)	39(3)
C52	5053(6)	1168(5)	7509(5)	26(2)
C53	5540(6)	2541(5)	7682(5)	33(2)
C54	5569(7)	3148(6)	7994(6)	42(3)
C55	4758(8)	3574(6)	7832(6)	50(3)
C56	3926(8)	3354(7)	7364(6)	56(4)
C57	3889(7)	2748(6)	7056(6)	38(2)
C58	4691(6)	2342(5)	7209(5)	33(2)
C59	6368(6)	1995(5)	7777(4)	27(2)
C60	4777(6)	1649(5)	6901(5)	33(2)
C61	6680(6)	1892(4)	7021(4)	24(2)
C62	5736(6)	1695(5)	6492(4)	24(2)
C63	5932(6)	1048(5)	6104(5)	27(2)
C64	6196(7)	1340(5)	4859(5)	32(2)
C65	5194(7)	1346(5)	4534(5)	34(2)
C66	5058(7)	1499(6)	3813(5)	42(3)
C67	5890(8)	1681(7)	3452(6)	56(3)
C68	6885(7)	1657(5)	3787(5)	37(2)
C69	7028(6)	1503(5)	4502(5)	29(2)
C70	7630(6)	1258(5)	5636(5)	28(2)
C71	8947(7)	1612(6)	4877(5)	42(3)
C72	9321(7)	1242(6)	4283(6)	49(3)
C73	8975(8)	2369(6)	4779(6)	49(3)
C74	7209(6)	2467(4)	6743(4)	26(2)
C75	8844(6)	3071(5)	7113(5)	32(2)
C76	8536(8)	3698(6)	7276(6)	45(3)
C77	9299(9)	4173(6)	7430(6)	48(3)

C78	10297(7)	4012(5)	7428(6)	37(2)
C79	10644(7)	3413(6)	7257(5)	39(2)
C80	9871(6)	2913(5)	7112(5)	32(2)
C81	8955(6)	1981(5)	6812(4)	22(2)
C82	10861(6)	1900(5)	6783(5)	29(2)
C83	11281(7)	2221(5)	6162(6)	39(2)
C84	11651(7)	1853(6)	7435(6)	43(3)
C85	9120(7)	150(5)	6114(6)	42(3)
C86	8179(9)	3(5)	6332(6)	42(3)
C87	8112(9)	-420(5)	7009(6)	47(3)
C88	8069(8)	3(6)	7648(6)	49(3)
C89	8669(7)	633(6)	7649(5)	36(2)
C90	9656(7)	680(6)	7470(5)	37(2)
C91	10312(7)	129(5)	7287(6)	41(3)
C92	10166(8)	-17(6)	6510(7)	51(3)

Table S3. Bond lengths (in Å) for [(DEAM–IBY)Ir(cod)](OTf) (**3-^{i}Pr**).

Bond	Distance	Bond	Distance
Ir1-C24	2.056(9)	F3-C93	1.307(14)
Ir1-C35	2.081(10)	S2-O6	1.436(7)
Ir1-C39	2.165(10)	S2-O5	1.451(8)
Ir1-C43	2.172(9)	S2-O4	1.452(8)
Ir1-C40	2.228(11)	S2-C94	1.824(11)
Ir1-C44	2.229(10)	F4-C94	1.354(12)
Ir2-C81	2.046(9)	F5-C94	1.313(13)
Ir2-C70	2.075(9)	F6-C94	1.349(12)
Ir2-C85	2.142(11)	C1-C2	1.380(13)
Ir2-C86	2.17(1)	C1-C6	1.415(14)
Ir2-C89	2.195(9)	C1-C13	1.517(14)
Ir2-C90	2.212(9)	C2-C3	1.421(18)
N1-C24	1.355(11)	C2-H2A	0.9500
N1-C18	1.379(12)	C3-C4	1.349(18)
N1-C17	1.477(11)	C3-H3A	0.9500
N2-C24	1.318(11)	C4-C5	1.392(15)
N2-C23	1.428(12)	C4-H4A	0.9500
N2-C25	1.503(12)	C5-C6	1.350(14)
N3-C29	1.354(12)	C5-H5A	0.9500
N3-C35	1.386(12)	C6-C14	1.512(13)
N3-C28	1.47(1)	C7-C8	1.378(14)
N4-C35	1.370(11)	C7-C12	1.427(13)
N4-C34	1.396(13)	C7-C13	1.527(12)
N4-C36	1.475(11)	C8-C9	1.379(14)
N5-C70	1.369(11)	C8-H8A	0.9500
N5-C64	1.390(12)	C9-C10	1.377(14)
N5-C63	1.47(1)	C9-H9A	0.9500
N6-C70	1.321(11)	C10-C11	1.408(13)
N6-C69	1.409(11)	C10-H10G	0.9500
N6-C71	1.500(11)	C11-C12	1.367(11)
N7-C81	1.372(11)	C11-H11A	0.9500
N7-C75	1.384(12)	C12-C14	1.527(12)
N7-C74	1.472(10)	C13-C15	1.534(12)
N8-C81	1.381(10)	C13-H13E	1.0000
N8-C80	1.382(12)	C14-C16	1.554(11)
N8-C82	1.503(11)	C14-H14A	1.0000
O1-S1	1.412(10)	C15-C28	1.509(12)
F1-C93	1.301(14)	C15-C16	1.553(12)
S1-O3	1.394(10)	C15-H15A	1.0000
S1-O2	1.403(12)	C16-C17	1.545(13)
S1-C93	1.809(13)	C16-H16A	1.0000
F2-C93	1.307(13)	C17-H17D	0.9900

C17-H17E	0.9900	C41-C42	1.528(17)
C18-C23	1.390(14)	C41-H41D	0.9900
C18-C19	1.402(13)	C41-H41E	0.9900
C19-C20	1.371(16)	C42-C43	1.501(14)
C19-H19D	0.9500	C42-H42A	0.9900
C20-C21	1.373(16)	C42-H42B	0.9900
C20-H20C	0.9500	C43-C44	1.414(14)
C21-C22	1.412(16)	C43-H43B	1.0000
C21-H21A	0.9500	C44-C45	1.549(17)
C22-C23	1.378(14)	C44-H44B	1.0000
C22-H22A	0.9500	C45-C46	1.594(17)
C25-C27	1.475(17)	C45-H45A	0.9900
C25-C26	1.544(16)	C45-H45B	0.9900
C25-H25A	1.0000	C46-H46B	0.9900
C26-H26B	0.9800	C46-H46C	0.9900
C26-H26C	0.9800	C47-C48	1.403(13)
C26-H26D	0.9800	C47-C52	1.420(13)
C27-H27D	0.9800	C47-C59	1.478(13)
C27-H27E	0.9800	C48-C49	1.356(14)
C27-H27F	0.9800	C48-H48A	0.9500
C28-H28A	0.9900	C49-C50	1.385(14)
C28-H28B	0.9900	C49-H49A	0.9500
C29-C34	1.388(12)	C50-C51	1.402(14)
C29-C30	1.412(14)	C50-H50A	0.9500
C30-C31	1.407(15)	C51-C52	1.385(14)
C30-H30B	0.9500	C51-H51A	0.9500
C31-C32	1.397(15)	C52-C60	1.520(13)
C31-H31B	0.9500	C53-C54	1.366(14)
C32-C33	1.381(16)	C53-C58	1.406(13)
C32-H32A	0.9500	C53-C59	1.552(13)
C33-C34	1.389(14)	C54-C55	1.381(15)
C33-H33B	0.9500	C54-H54A	0.9500
C36-C38	1.525(14)	C55-C56	1.398(17)
C36-C37	1.532(15)	C55-H55A	0.9500
C36-H36A	1.0000	C56-C57	1.361(16)
C37-H37B	0.9800	C56-H56A	0.9500
C37-H37C	0.9800	C57-C58	1.346(14)
C37-H37D	0.9800	C57-H57A	0.9500
C38-H38D	0.9800	C58-C60	1.534(14)
C38-H38E	0.9800	C59-C61	1.556(11)
C38-H38F	0.9800	C59-H59A	1.0000
C39-C40	1.410(15)	C60-C62	1.57(1)
C39-C46	1.481(15)	C60-H60A	1.0000
C39-H39A	1.0000	C61-C74	1.491(12)
C40-C41	1.522(16)	C61-C62	1.552(11)
C40-H40A	1.0000	C61-H61A	1.0000

C62-C63	1.545(13)	C78-H78A	0.9500
C62-H62A	1.0000	C79-C80	1.442(13)
C63-H63A	0.9900	C79-H79A	0.9500
C63-H63B	0.9900	C82-C83	1.512(13)
C64-C65	1.389(12)	C82-C84	1.518(15)
C64-C69	1.403(12)	C82-H82A	1.0000
C65-C66	1.392(14)	C83-H83A	0.9800
C65-H65A	0.9500	C83-H83B	0.9800
C66-C67	1.417(14)	C83-H83C	0.9800
C66-H66A	0.9500	C84-H84A	0.9800
C67-C68	1.389(14)	C84-H84B	0.9800
C67-H67A	0.9500	C84-H84C	0.9800
C68-C69	1.381(13)	C85-C86	1.392(14)
C68-H68A	0.9500	C85-C92	1.527(15)
C71-C72	1.489(14)	C85-H85A	1.0000
C71-C73	1.550(16)	C86-C87	1.557(15)
C71-H71A	1.0000	C86-H86A	1.0000
C72-H72A	0.9800	C87-C88	1.492(15)
C72-H72B	0.9800	C87-H87A	0.9900
C72-H72C	0.9800	C87-H87B	0.9900
C73-H73A	0.9800	C88-C89	1.506(15)
C73-H73B	0.9800	C88-H88A	0.9900
C73-H73C	0.9800	C88-H88B	0.9900
C74-H74A	0.9900	C89-C90	1.393(13)
C74-H74B	0.9900	C89-H89A	1.0000
C75-C76	1.383(15)	C90-C91	1.483(14)
C75-C80	1.395(12)	C90-H90A	1.0000
C76-C77	1.400(15)	C91-C92	1.491(15)
C76-H76A	0.9500	C91-H91A	0.9900
C77-C78	1.360(15)	C91-H91B	0.9900
C77-H77A	0.9500	C92-H92A	0.9900
C78-C79	1.355(15)	C92-H92B	0.9900

Table S4. Bond angles in (°) for [(DEAM–IBY)Ir(cod)](OTf) (**3-^{i}Pr**).

Bond Angle	Angle	Bond Angle	Angle
C24-Ir1-C35	93.5(4)	C70-N5-C64	109.8(7)
C24-Ir1-C39	90.6(4)	C70-N5-C63	128.0(7)
C35-Ir1-C39	145.2(4)	C64-N5-C63	121.8(7)
C24-Ir1-C43	148.8(3)	C70-N6-C69	111.2(7)
C35-Ir1-C43	98.0(4)	C70-N6-C71	124.7(8)
C39-Ir1-C43	96.0(4)	C69-N6-C71	123.1(7)
C24-Ir1-C40	86.7(4)	C81-N7-C75	112.7(7)
C35-Ir1-C40	177.4(4)	C81-N7-C74	123.0(7)
C39-Ir1-C40	37.4(4)	C75-N7-C74	122.8(7)
C43-Ir1-C40	80.6(4)	C81-N8-C80	111.3(7)
C24-Ir1-C44	171.4(4)	C81-N8-C82	122.9(8)
C35-Ir1-C44	90.4(4)	C80-N8-C82	124.8(7)
C39-Ir1-C44	81.8(5)	O3-S1-O2	113.2(8)
C43-Ir1-C44	37.4(4)	O3-S1-O1	115.8(7)
C40-Ir1-C44	89.8(5)	O2-S1-O1	115.6(7)
C81-Ir2-C70	94.9(4)	O3-S1-C93	104.1(5)
C81-Ir2-C85	142.2(3)	O2-S1-C93	102.3(6)
C70-Ir2-C85	92.7(4)	O1-S1-C93	103.5(7)
C81-Ir2-C86	178.3(4)	F1-C93-F3	104.2(11)
C70-Ir2-C86	86.8(4)	F1-C93-F2	104.3(12)
C85-Ir2-C86	37.6(4)	F3-C93-F2	107.7(10)
C81-Ir2-C89	98.4(4)	F1-C93-S1	114.3(9)
C70-Ir2-C89	146.9(3)	F3-C93-S1	112.2(10)
C85-Ir2-C89	95.1(4)	F2-C93-S1	113.4(7)
C86-Ir2-C89	80.1(4)	O6-S2-O5	114.1(4)
C81-Ir2-C90	90.2(4)	O6-S2-O4	115.3(4)
C70-Ir2-C90	172.4(4)	O5-S2-O4	114.8(5)
C85-Ir2-C90	79.9(4)	O6-S2-C94	104.2(5)
C86-Ir2-C90	88.1(4)	O5-S2-C94	104.0(5)
C89-Ir2-C90	36.9(3)	O4-S2-C94	102.2(5)
C24-N1-C18	110.1(8)	F5-C94-F6	109.1(10)
C24-N1-C17	127.4(8)	F5-C94-F4	108.8(9)
C18-N1-C17	122.1(8)	F6-C94-F4	105.6(8)
C24-N2-C23	110.9(7)	F5-C94-S2	113.0(7)
C24-N2-C25	124.1(8)	F6-C94-S2	110.1(8)
C23-N2-C25	124.9(8)	F4-C94-S2	110.0(8)
C29-N3-C35	110.1(7)	C2-C1-C6	120.0(11)
C29-N3-C28	124.7(8)	C2-C1-C13	127.7(10)
C35-N3-C28	123.2(8)	C6-C1-C13	112.2(8)
C35-N4-C34	110.7(8)	C1-C2-C3	118.7(12)
C35-N4-C36	123.6(9)	C1-C2-H2A	120.6
C34-N4-C36	125.6(8)	C3-C2-H2A	120.6

C4-C3-C2	120.3(11)	C13-C15-H15A	106.8
C4-C3-H3A	119.8	C16-C15-H15A	106.8
C2-C3-H3A	119.8	C17-C16-C15	113.7(7)
C3-C4-C5	120.3(11)	C17-C16-C14	110.2(7)
C3-C4-H4A	119.8	C15-C16-C14	109.8(7)
C5-C4-H4A	119.8	C17-C16-H16A	107.6
C6-C5-C4	121.1(11)	C15-C16-H16A	107.6
C6-C5-H5A	119.4	C14-C16-H16A	107.6
C4-C5-H5A	119.4	N1-C17-C16	112.3(8)
C5-C6-C1	119.5(10)	N1-C17-H17D	109.1
C5-C6-C14	126.9(10)	C16-C17-H17D	109.1
C1-C6-C14	113.6(8)	N1-C17-H17E	109.1
C8-C7-C12	120.7(8)	C16-C17-H17E	109.1
C8-C7-C13	126.8(9)	H17D-C17-H17E	107.9
C12-C7-C13	112.5(8)	N1-C18-C23	107.4(8)
C7-C8-C9	119.5(10)	N1-C18-C19	131.6(10)
C7-C8-H8A	120.2	C23-C18-C19	120.9(9)
C9-C8-H8A	120.2	C20-C19-C18	116.6(10)
C10-C9-C8	120.1(9)	C20-C19-H19D	121.7
C10-C9-H9A	120	C18-C19-H19D	121.7
C8-C9-H9A	120	C19-C20-C21	121.7(11)
C9-C10-C11	121.3(8)	C19-C20-H20C	119.2
C9-C10-H10G	119.3	C21-C20-H20C	119.2
C11-C10-H10G	119.3	C20-C21-C22	123.3(11)
C12-C11-C10	118.9(9)	C20-C21-H21A	118.3
C12-C11-H11A	120.5	C22-C21-H21A	118.3
C10-C11-H11A	120.5	C23-C22-C21	114(1)
C11-C12-C7	119.4(9)	C23-C22-H22A	123
C11-C12-C14	128.0(8)	C21-C22-H22A	123
C7-C12-C14	112.5(7)	C22-C23-C18	123.5(9)
C1-C13-C7	106.5(7)	C22-C23-N2	132.4(9)
C1-C13-C15	106.7(8)	C18-C23-N2	104.1(8)
C7-C13-C15	108.9(7)	N2-C24-N1	107.3(8)
C1-C13-H13E	111.5	N2-C24-Ir1	128.4(6)
C7-C13-H13E	111.5	N1-C24-Ir1	124.3(6)
C15-C13-H13E	111.5	C27-C25-N2	112.1(10)
C6-C14-C12	107.4(8)	C27-C25-C26	113.9(10)
C6-C14-C16	108.7(6)	N2-C25-C26	107.9(8)
C12-C14-C16	104.0(7)	C27-C25-H25A	107.5
C6-C14-H14A	112.1	N2-C25-H25A	107.5
C12-C14-H14A	112.1	C26-C25-H25A	107.5
C16-C14-H14A	112.1	C25-C26-H26B	109.5
C28-C15-C13	114.9(8)	C25-C26-H26C	109.5
C28-C15-C16	111.8(7)	H26B-C26-H26C	109.5
C13-C15-C16	109.1(7)	C25-C26-H26D	109.5
C28-C15-H15A	106.8	H26B-C26-H26D	109.5

H26C-C26-H26D	109.5	C36-C38-H38D	109.5
C25-C27-H27D	109.5	C36-C38-H38E	109.5
C25-C27-H27E	109.5	H38D-C38-H38E	109.5
H27D-C27-H27E	109.5	C36-C38-H38F	109.5
C25-C27-H27F	109.5	H38D-C38-H38F	109.5
H27D-C27-H27F	109.5	H38E-C38-H38F	109.5
H27E-C27-H27F	109.5	C40-C39-C46	127.9(11)
N3-C28-C15	114.8(7)	C40-C39-Ir1	73.7(6)
N3-C28-H28A	108.6	C46-C39-Ir1	111.4(8)
C15-C28-H28A	108.6	C40-C39-H39A	112.4
N3-C28-H28B	108.6	C46-C39-H39A	112.4
C15-C28-H28B	108.6	Ir1-C39-H39A	112.4
H28A-C28-H28B	107.5	C39-C40-C41	122.0(11)
N3-C29-C34	108.7(8)	C39-C40-Ir1	68.9(6)
N3-C29-C30	130.3(8)	C41-C40-Ir1	112.4(8)
C34-C29-C30	121.0(9)	C39-C40-H40A	115
C31-C30-C29	116.9(9)	C41-C40-H40A	115
C31-C30-H30B	121.6	Ir1-C40-H40A	115
C29-C30-H30B	121.6	C40-C41-C42	110.5(9)
C32-C31-C30	120.7(11)	C40-C41-H41D	109.6
C32-C31-H31B	119.7	C42-C41-H41D	109.6
C30-C31-H31B	119.7	C40-C41-H41E	109.6
C33-C32-C31	122.1(10)	C42-C41-H41E	109.6
C33-C32-H32A	119	H41D-C41-H41E	108.1
C31-C32-H32A	119	C43-C42-C41	115.5(9)
C32-C33-C34	117.4(9)	C43-C42-H42A	108.4
C32-C33-H33B	121.3	C41-C42-H42A	108.4
C34-C33-H33B	121.3	C43-C42-H42B	108.4
C29-C34-C33	121.9(10)	C41-C42-H42B	108.4
C29-C34-N4	105.3(8)	H42A-C42-H42B	107.5
C33-C34-N4	132.8(9)	C44-C43-C42	127.2(10)
N4-C35-N3	105.3(8)	C44-C43-Ir1	73.5(6)
N4-C35-Ir1	127.8(7)	C42-C43-Ir1	108.5(7)
N3-C35-Ir1	126.9(6)	C44-C43-H43B	113.3
N4-C36-C38	110.3(9)	C42-C43-H43B	113.3
N4-C36-C37	112.1(9)	Ir1-C43-H43B	113.3
C38-C36-C37	109.8(8)	C43-C44-C45	122.5(12)
N4-C36-H36A	108.2	C43-C44-Ir1	69.1(5)
C38-C36-H36A	108.2	C45-C44-Ir1	110.1(8)
C37-C36-H36A	108.2	C43-C44-H44B	115.4
C36-C37-H37B	109.5	C45-C44-H44B	115.4
C36-C37-H37C	109.5	Ir1-C44-H44B	115.4
H37B-C37-H37C	109.5	C44-C45-C46	113.6(9)
C36-C37-H37D	109.5	C44-C45-H45A	108.9
H37B-C37-H37D	109.5	C46-C45-H45A	108.9
H37C-C37-H37D	109.5	C44-C45-H45B	108.9

C46-C45-H45B	108.9	C53-C59-C61	104.7(7)
H45A-C45-H45B	107.7	C47-C59-H59A	111.6
C39-C46-C45	112.3(9)	C53-C59-H59A	111.6
C39-C46-H46B	109.1	C61-C59-H59A	111.6
C45-C46-H46B	109.1	C52-C60-C58	108.8(8)
C39-C46-H46C	109.1	C52-C60-C62	106.4(7)
C45-C46-H46C	109.1	C58-C60-C62	103.7(7)
H46B-C46-H46C	107.9	C52-C60-H60A	112.5
C48-C47-C52	119.1(9)	C58-C60-H60A	112.5
C48-C47-C59	127.3(8)	C62-C60-H60A	112.5
C52-C47-C59	113.6(8)	C74-C61-C62	110.4(7)
C49-C48-C47	119.0(9)	C74-C61-C59	114.3(7)
C49-C48-H48A	120.5	C62-C61-C59	110.2(6)
C47-C48-H48A	120.5	C74-C61-H61A	107.2
C48-C49-C50	122.6(9)	C62-C61-H61A	107.2
C48-C49-H49A	118.7	C59-C61-H61A	107.2
C50-C49-H49A	118.7	C63-C62-C61	111.0(7)
C49-C50-C51	120(1)	C63-C62-C60	112.3(7)
C49-C50-H50A	120	C61-C62-C60	109.3(7)
C51-C50-H50A	120	C63-C62-H62A	108
C52-C51-C50	118.3(10)	C61-C62-H62A	108
C52-C51-H51A	120.8	C60-C62-H62A	108
C50-C51-H51A	120.8	N5-C63-C62	109.3(7)
C51-C52-C47	121.0(9)	N5-C63-H63A	109.8
C51-C52-C60	125.9(9)	C62-C63-H63A	109.8
C47-C52-C60	113.1(8)	N5-C63-H63B	109.8
C54-C53-C58	120.8(9)	C62-C63-H63B	109.8
C54-C53-C59	127.8(8)	H63A-C63-H63B	108.3
C58-C53-C59	111.4(8)	C65-C64-N5	130.4(9)
C53-C54-C55	119.4(10)	C65-C64-C69	123.1(9)
C53-C54-H54A	120.3	N5-C64-C69	106.5(7)
C55-C54-H54A	120.3	C64-C65-C66	115.9(8)
C54-C55-C56	118.0(12)	C64-C65-H65A	122.1
C54-C55-H55A	121	C66-C65-H65A	122.1
C56-C55-H55A	121	C65-C66-C67	121.4(9)
C57-C56-C55	123.0(11)	C65-C66-H66A	119.3
C57-C56-H56A	118.5	C67-C66-H66A	119.3
C55-C56-H56A	118.5	C68-C67-C66	121.3(10)
C58-C57-C56	118.4(10)	C68-C67-H67A	119.4
C58-C57-H57A	120.8	C66-C67-H67A	119.4
C56-C57-H57A	120.8	C69-C68-C67	117.5(8)
C57-C58-C53	120.5(10)	C69-C68-H68A	121.2
C57-C58-C60	125.0(9)	C67-C68-H68A	121.2
C53-C58-C60	114.5(8)	C68-C69-C64	120.6(8)
C47-C59-C53	109.9(7)	C68-C69-N6	134.1(7)
C47-C59-C61	106.9(7)	C64-C69-N6	105.1(8)

N6-C70-N5	107.2(8)	N7-C81-Ir2	127.9(5)
N6-C70-Ir2	128.5(6)	N8-C81-Ir2	128.2(6)
N5-C70-Ir2	123.7(6)	N8-C82-C83	110.4(8)
C72-C71-N6	113.4(9)	N8-C82-C84	111.6(8)
C72-C71-C73	113.1(9)	C83-C82-C84	112.2(7)
N6-C71-C73	108.5(8)	N8-C82-H82A	107.4
C72-C71-H71A	107.1	C83-C82-H82A	107.4
N6-C71-H71A	107.1	C84-C82-H82A	107.4
C73-C71-H71A	107.1	C82-C83-H83A	109.5
C71-C72-H72A	109.5	C82-C83-H83B	109.5
C71-C72-H72B	109.5	H83A-C83-H83B	109.5
H72A-C72-H72B	109.5	C82-C83-H83C	109.5
C71-C72-H72C	109.5	H83A-C83-H83C	109.5
H72A-C72-H72C	109.5	H83B-C83-H83C	109.5
H72B-C72-H72C	109.5	C82-C84-H84A	109.5
C71-C73-H73A	109.5	C82-C84-H84B	109.5
C71-C73-H73B	109.5	H84A-C84-H84B	109.5
H73A-C73-H73B	109.5	C82-C84-H84C	109.5
C71-C73-H73C	109.5	H84A-C84-H84C	109.5
H73A-C73-H73C	109.5	H84B-C84-H84C	109.5
H73B-C73-H73C	109.5	C86-C85-C92	126.5(10)
N7-C74-C61	115.8(7)	C86-C85-Ir2	72.3(6)
N7-C74-H74A	108.3	C92-C85-Ir2	108.8(8)
C61-C74-H74A	108.3	C86-C85-H85A	113.7
N7-C74-H74B	108.3	C92-C85-H85A	113.7
C61-C74-H74B	108.3	Ir2-C85-H85A	113.7
H74A-C74-H74B	107.4	C85-C86-C87	120.6(10)
C76-C75-N7	133.2(8)	C85-C86-Ir2	70.1(6)
C76-C75-C80	121.7(9)	C87-C86-Ir2	113.4(7)
N7-C75-C80	105.1(8)	C85-C86-H86A	115
C75-C76-C77	117.1(10)	C87-C86-H86A	115
C75-C76-H76A	121.4	Ir2-C86-H86A	115
C77-C76-H76A	121.4	C88-C87-C86	111.3(9)
C78-C77-C76	120.6(11)	C88-C87-H87A	109.4
C78-C77-H77A	119.7	C86-C87-H87A	109.4
C76-C77-H77A	119.7	C88-C87-H87B	109.4
C79-C78-C77	125(1)	C86-C87-H87B	109.4
C79-C78-H78A	117.5	H87A-C87-H87B	108
C77-C78-H78A	117.5	C87-C88-C89	114.7(8)
C78-C79-C80	115.1(9)	C87-C88-H88A	108.6
C78-C79-H79A	122.4	C89-C88-H88A	108.6
C80-C79-H79A	122.4	C87-C88-H88B	108.6
N8-C80-C75	107.1(8)	C89-C88-H88B	108.6
N8-C80-C79	132.5(8)	H88A-C88-H88B	107.6
C75-C80-C79	120.3(9)	C90-C89-C88	124.5(10)
N7-C81-N8	103.7(7)	C90-C89-Ir2	72.2(6)

C88-C89-Ir2	109.7(6)	C92-C91-H91A	109.2
C90-C89-H89A	114.2	C90-C91-H91B	109.2
C88-C89-H89A	114.2	C92-C91-H91B	109.2
Ir2-C89-H89A	114.2	H91A-C91-H91B	107.9
C89-C90-C91	126.9(10)	C91-C92-C85	116.0(9)
C89-C90-Ir2	70.9(5)	C91-C92-H92A	108.3
C91-C90-Ir2	113.6(6)	C85-C92-H92A	108.3
C89-C90-H90A	112.7	C91-C92-H92B	108.3
C91-C90-H90A	112.7	C85-C92-H92B	108.3
Ir2-C90-H90A	112.7	H92A-C92-H92B	107.4
C90-C91-C92	112.0(9)		
C90-C91-H91A	109.2		

Table S5. Anisotropic displacement parameters ($Å^2$ x10^3) for [(DEAM–IBY)Ir(cod)](OTf) (**3-^{i}Pr**). 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}
Ir1	19(1)	43(1)	23(1)	-4(1)	2(1)	0(1)
Ir2	20(1)	34(1)	20(1)	-1(1)	3(1)	-4(1)
N1	26(4)	30(5)	26(4)	-9(3)	0(3)	-4(3)
N2	31(4)	36(5)	26(4)	-6(3)	6(3)	5(3)
N3	21(4)	47(5)	29(4)	0(4)	8(3)	-15(4)
N4	27(4)	42(5)	28(4)	3(4)	6(3)	-5(3)
N5	16(3)	41(5)	29(4)	-4(3)	2(3)	4(3)
N6	17(4)	49(6)	23(4)	6(4)	5(3)	-6(3)
N7	10(3)	42(5)	31(4)	3(4)	4(3)	-2(3)
N8	16(3)	34(5)	22(4)	0(3)	-1(3)	-2(3)
O1	50(6)	97(8)	150(11)	-2(8)	37(6)	-3(5)
F1	219(13)	87(7)	72(6)	7(5)	-17(7)	27(7)
S1	40(2)	63(2)	60(2)	9(2)	17(1)	1(1)
O2	115(9)	168(13)	76(8)	33(8)	39(7)	52(9)
F2	196(11)	49(6)	143(9)	5(5)	107(8)	-5(6)
O3	95(7)	70(7)	96(8)	-32(6)	38(6)	-23(5)
F3	79(6)	121(8)	168(11)	-7(7)	76(7)	10(5)
C93	62(7)	41(7)	53(7)	29(6)	25(6)	4(5)
S2	24(1)	41(2)	27(1)	1(1)	2(1)	3(1)
O4	27(4)	60(5)	63(6)	5(5)	-3(4)	1(4)
F4	79(5)	79(5)	37(4)	-11(3)	20(3)	-6(4)
O5	32(4)	48(4)	42(4)	-5(3)	5(3)	4(3)
F5	79(5)	41(4)	51(4)	-7(3)	-2(4)	-8(4)
O6	45(4)	42(4)	28(4)	4(3)	3(3)	-1(3)
F6	40(3)	77(5)	78(5)	-8(4)	-3(3)	28(3)
C94	43(6)	55(8)	41(6)	-5(6)	5(5)	7(6)
C1	19(4)	61(7)	30(5)	-13(5)	7(4)	-1(4)
C2	28(5)	91(10)	45(7)	-43(7)	-3(5)	0(6)
C3	30(6)	125(13)	32(6)	-29(7)	-3(5)	13(7)
C4	27(5)	105(11)	26(6)	-4(6)	-11(4)	21(6)
C5	41(6)	56(7)	31(5)	4(5)	22(4)	9(5)

C6	24(4)	51(7)	33(5)	3(5)	11(4)	5(4)
C7	31(4)	39(6)	16(4)	-2(4)	8(4)	-12(4)
C8	59(6)	37(6)	29(5)	-4(5)	19(5)	4(5)
C9	40(6)	35(6)	65(8)	11(6)	5(5)	11(5)
C10	19(4)	51(7)	33(5)	2(5)	10(4)	12(4)
C11	19(4)	42(6)	23(5)	-8(4)	13(3)	-7(4)
C12	19(4)	43(6)	26(5)	-6(4)	3(3)	0(4)
C13	31(5)	35(6)	37(6)	-9(4)	14(4)	5(4)
C14	17(4)	33(5)	13(4)	-4(4)	6(3)	-1(4)
C15	11(3)	36(5)	22(4)	5(4)	0(3)	3(3)
C16	17(4)	34(5)	28(5)	-1(4)	0(3)	-6(3)
C17	34(5)	46(6)	26(5)	-6(4)	14(4)	-2(4)
C18	33(5)	31(6)	36(6)	-2(4)	-1(4)	2(4)
C19	33(5)	35(6)	52(7)	-3(5)	8(5)	-6(4)
C20	42(6)	43(7)	67(8)	3(6)	-3(6)	-5(5)
C21	63(8)	58(8)	59(8)	-25(7)	-11(6)	-15(6)
C22	62(7)	61(8)	26(6)	-1(5)	1(5)	-13(6)
C23	32(5)	29(5)	39(6)	-5(4)	7(4)	0(4)
C24	34(5)	33(6)	20(5)	-1(4)	6(4)	-10(4)
C25	39(6)	78(9)	25(5)	-14(5)	9(4)	-13(5)
C26	61(7)	93(10)	29(6)	16(6)	-5(5)	-27(7)
C27	74(8)	90(11)	73(9)	-35(8)	49(8)	-6(7)
C28	18(4)	41(6)	29(5)	2(4)	9(4)	-12(4)
C29	18(4)	45(6)	30(5)	2(5)	1(4)	-5(4)
C30	29(5)	40(6)	46(6)	1(5)	-1(4)	-7(4)
C31	42(6)	53(7)	56(8)	-6(6)	-4(6)	-5(5)
C32	51(7)	53(8)	49(7)	18(6)	4(5)	-14(6)
C33	47(6)	76(9)	32(6)	9(6)	23(5)	-17(6)
C34	28(5)	49(6)	28(5)	-6(5)	3(4)	-7(4)
C35	21(4)	40(6)	22(5)	-4(4)	11(4)	-12(4)
C36	32(5)	61(7)	35(6)	-10(5)	19(4)	-2(5)
C37	41(6)	70(9)	64(8)	1(7)	15(6)	-23(6)
C38	60(7)	62(8)	44(7)	-10(6)	24(6)	-12(6)
C39	40(6)	46(7)	44(7)	-18(5)	7(5)	12(5)
C40	33(6)	48(7)	48(7)	10(5)	-2(5)	3(5)
C41	35(6)	74(9)	37(6)	19(6)	-4(5)	13(6)

C42	45(6)	68(8)	29(6)	-7(5)	5(5)	9(6)
C43	19(4)	52(7)	23(5)	-9(5)	0(4)	-3(4)
C44	29(5)	90(10)	48(7)	-12(7)	-11(5)	-17(6)
C45	15(5)	105(11)	44(7)	-14(7)	1(4)	-3(5)
C46	28(5)	79(9)	54(8)	-3(7)	4(5)	21(5)
C47	26(4)	46(6)	24(5)	1(4)	8(4)	-2(4)
C48	21(4)	48(6)	32(5)	7(5)	8(4)	-2(4)
C49	36(5)	61(7)	15(5)	16(5)	15(4)	8(5)
C50	37(5)	38(6)	44(6)	-1(5)	13(5)	2(4)
C51	30(5)	50(7)	36(6)	6(5)	5(4)	-5(5)
C52	18(4)	47(6)	16(4)	8(4)	9(3)	0(4)
C53	24(4)	45(6)	31(5)	-4(4)	10(4)	-5(4)
C54	28(5)	59(8)	39(6)	5(5)	8(4)	-11(5)
C55	51(7)	47(7)	57(7)	8(6)	31(6)	16(5)
C56	27(6)	95(11)	50(7)	13(7)	15(5)	1(6)
C57	25(5)	50(7)	41(6)	9(5)	9(4)	0(4)
C58	23(4)	48(6)	33(5)	-6(5)	17(4)	-3(4)
C59	20(4)	40(5)	20(4)	-12(4)	2(3)	1(4)
C60	17(4)	58(7)	25(5)	11(5)	11(4)	-10(4)
C61	14(4)	38(5)	20(4)	-6(4)	0(3)	-3(3)
C62	20(4)	41(5)	14(4)	8(4)	11(3)	-5(4)
C63	14(4)	49(6)	19(4)	5(4)	5(3)	-11(4)
C64	27(5)	46(6)	23(5)	-11(4)	-1(4)	7(4)
C65	25(4)	41(6)	37(6)	-2(5)	9(4)	1(4)
C66	26(5)	66(8)	33(6)	5(5)	-3(4)	7(5)
C67	42(6)	85(10)	39(7)	15(6)	3(5)	10(6)
C68	23(4)	59(7)	30(5)	-2(5)	7(4)	-4(4)
C69	17(4)	49(6)	23(5)	-6(4)	5(3)	0(4)
C70	28(5)	40(6)	18(4)	6(4)	6(4)	-6(4)
C71	25(5)	81(9)	21(5)	15(5)	0(4)	-10(5)
C72	26(5)	76(8)	46(7)	-11(6)	10(5)	-1(5)
C73	46(6)	47(7)	54(7)	10(6)	3(5)	-2(5)
C74	27(4)	33(5)	17(4)	-2(4)	6(3)	-4(4)
C75	17(4)	40(6)	37(5)	0(5)	2(4)	2(4)
C76	28(5)	51(7)	51(7)	10(6)	-13(5)	-12(5)
C77	72(8)	33(6)	36(6)	-1(5)	0(6)	4(6)

C78	36(5)	29(6)	46(6)	3(5)	1(5)	-7(4)
C79	21(4)	63(8)	30(5)	-1(5)	-3(4)	-10(5)
C80	22(4)	46(6)	29(5)	3(4)	7(4)	-2(4)
C81	13(4)	36(5)	19(4)	9(4)	5(3)	-3(4)
C82	14(4)	34(6)	39(6)	-2(4)	7(4)	-2(4)
C83	18(4)	51(7)	49(7)	0(5)	6(4)	-1(4)
C84	33(5)	47(7)	53(7)	-3(5)	18(5)	5(5)
C85	42(6)	44(7)	42(6)	10(5)	14(5)	5(5)
C86	47(6)	38(6)	41(7)	4(5)	6(5)	-14(5)
C87	59(7)	28(6)	58(7)	-7(5)	27(6)	-12(5)
C88	39(6)	66(8)	43(6)	21(6)	10(5)	-6(6)
C89	44(6)	54(7)	12(4)	-12(4)	6(4)	-12(5)
C90	28(5)	61(7)	20(5)	-14(5)	-6(4)	0(5)
C91	31(5)	47(7)	45(6)	3(5)	6(5)	-2(5)
C92	42(6)	48(7)	64(8)	-1(6)	8(6)	3(5)

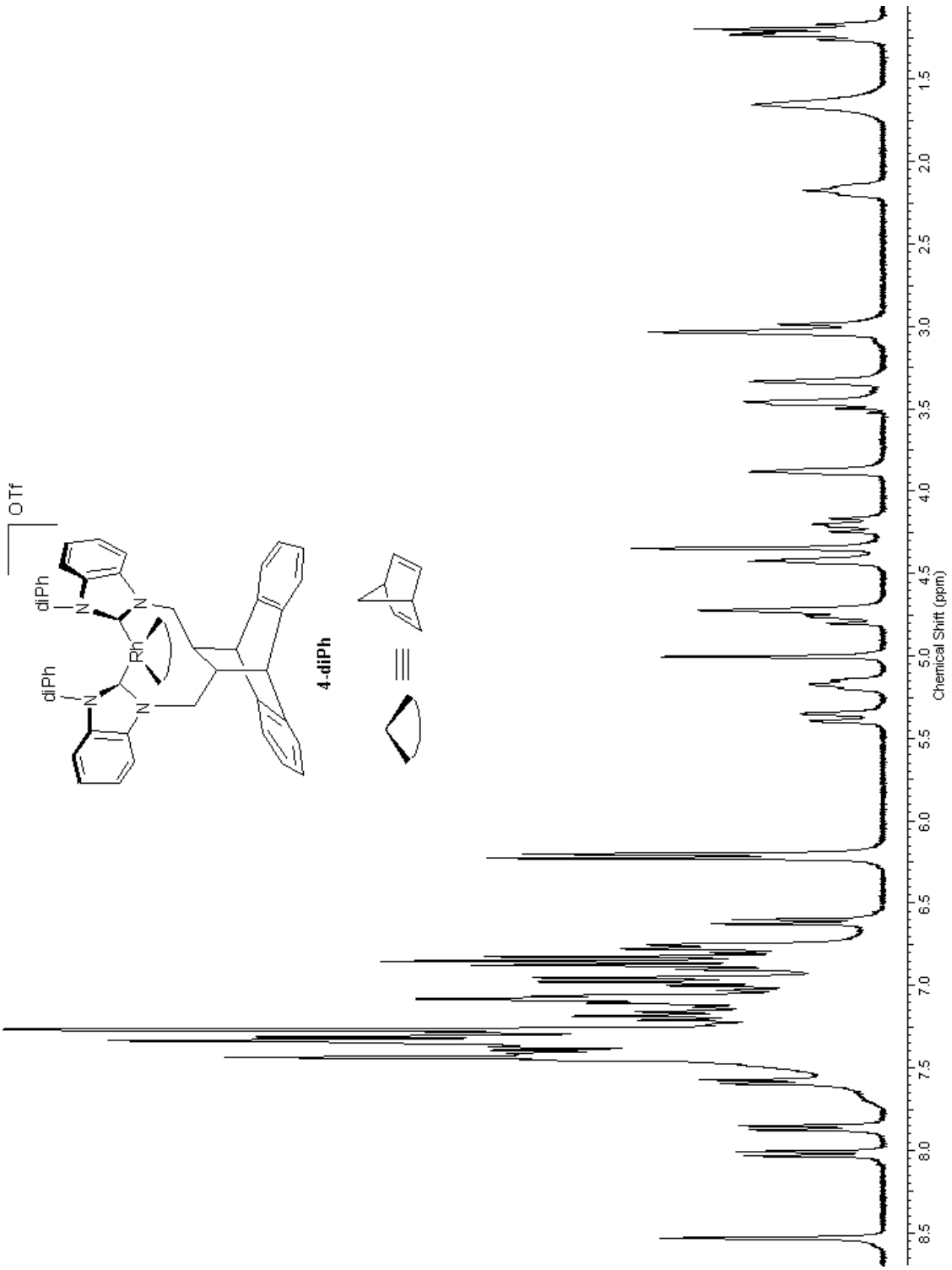

Figure S12: ^{1}H NMR spectrum of [(DEAM-diPhBY)Rh(nbd)](OTf) (**4-diPh**) in $CDCl_3$.

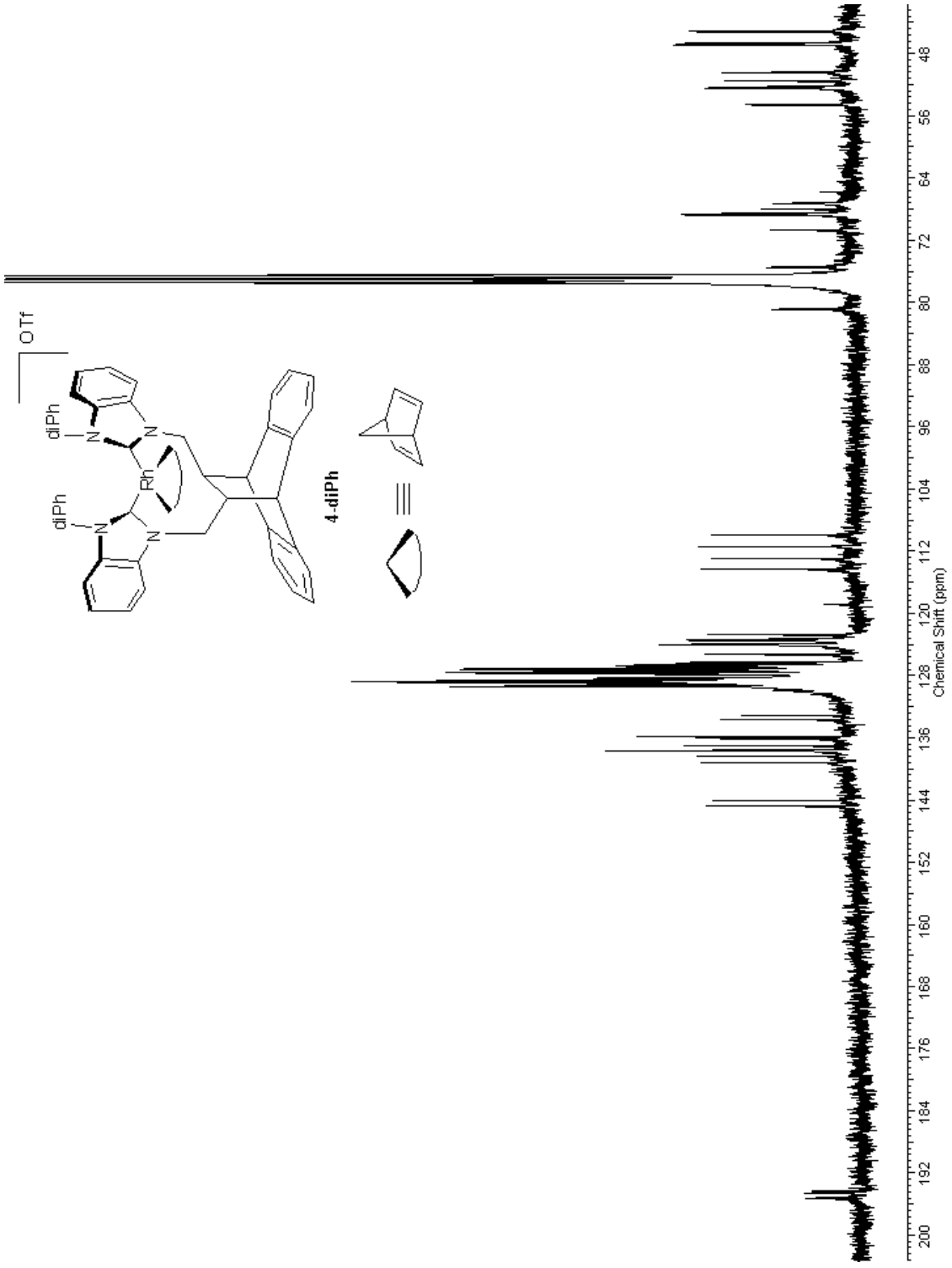

Figure S13: $^{13}C\{^{1}H\}$ NMR spectrum of [(DEAM-diPhBY)Rh(nbd)](OTf) (**4-diPh**) in $CDCl_3$.

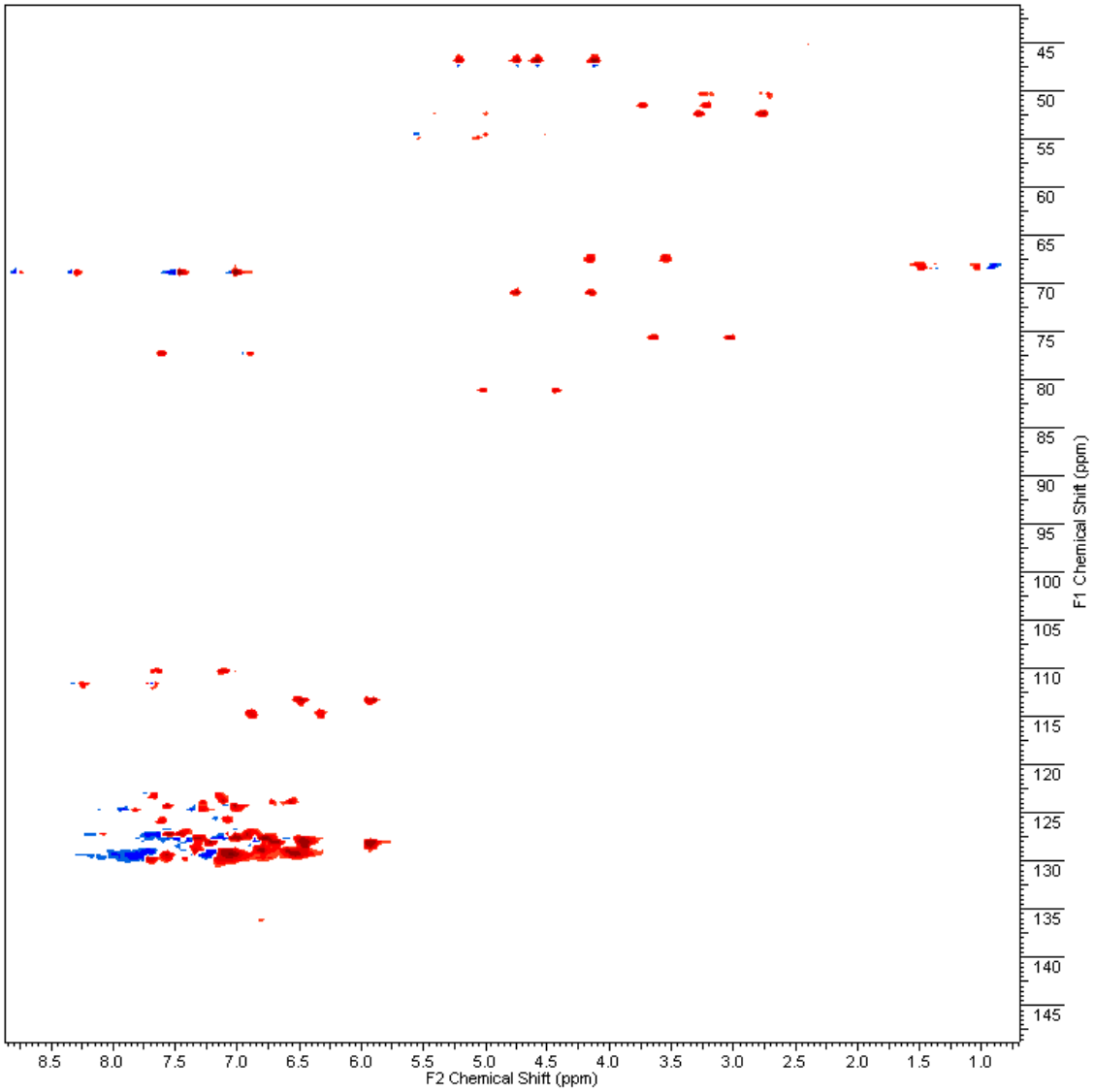

Figure S14: gHMQC NMR spectrum of [(DEAM-diPhBY)Rh(nbd)](OTf) (**4-diPh**) in $CDCl_3$.

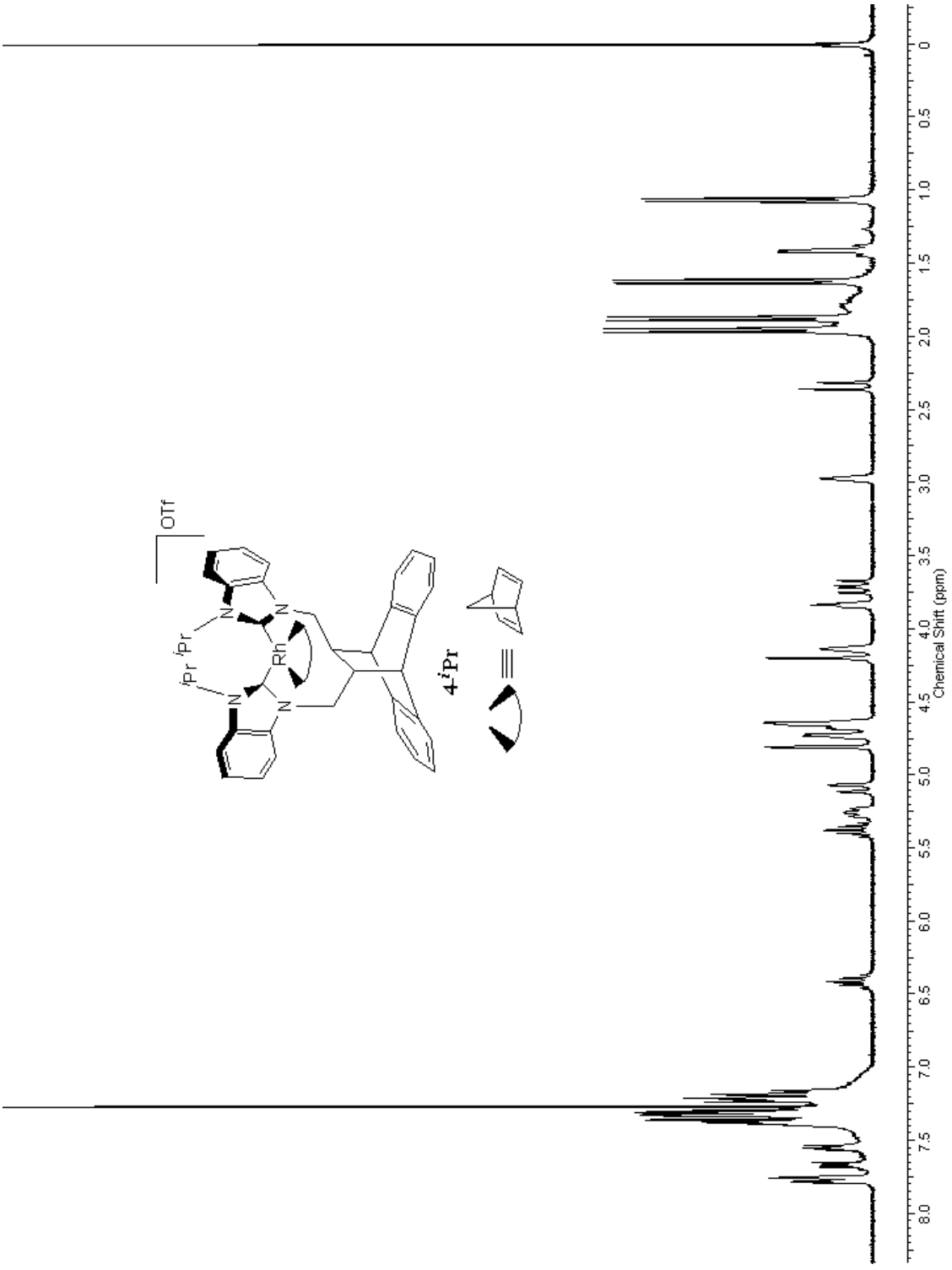

Figure S15: ^{1}H NMR spectrum of [(DEAM-IBY)Rh(nbd)](OTf) (**4-*i*Pr**) in $CDCl_3$.

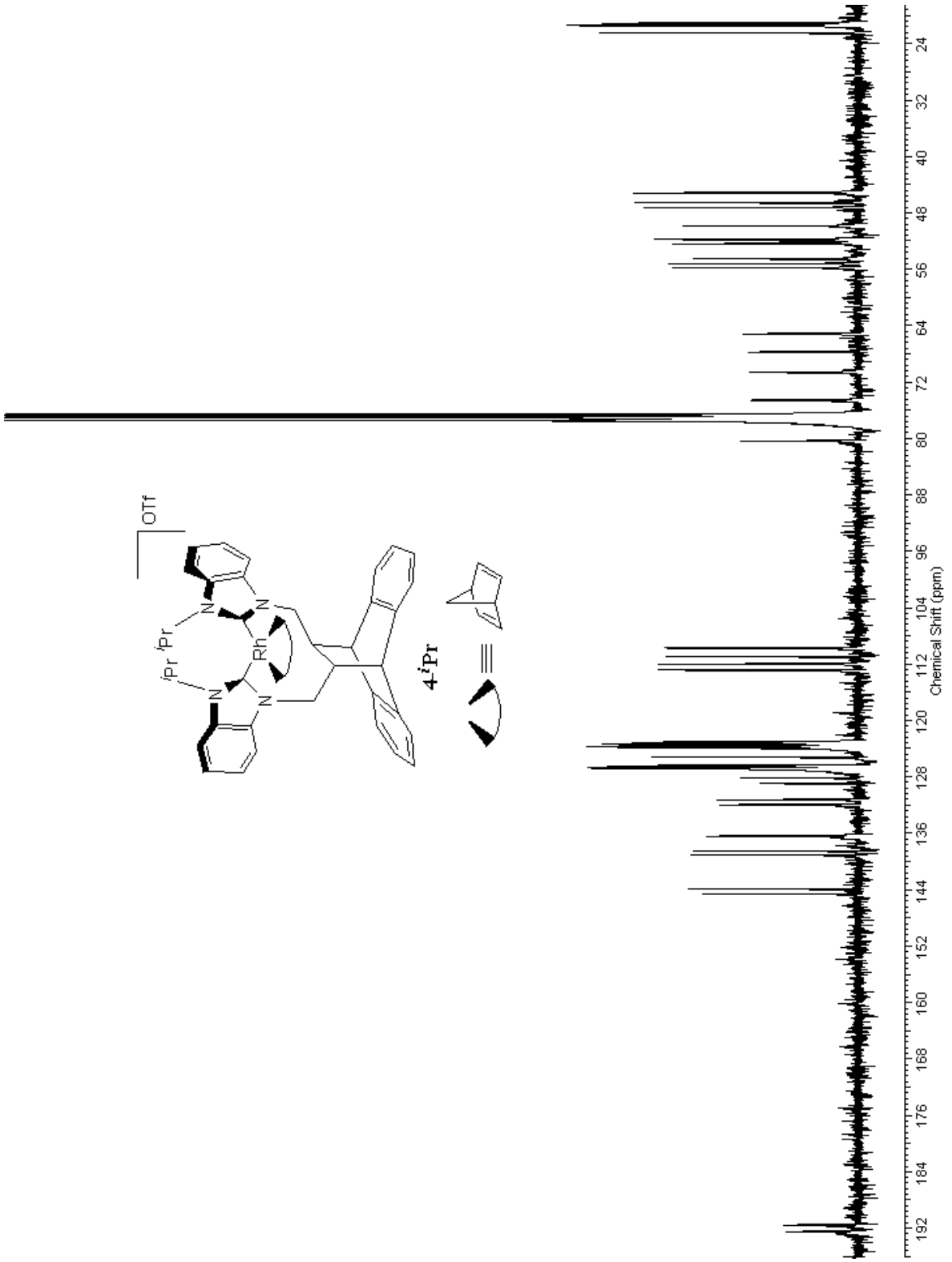

Figure S16: $^{13}C\{^{1}H\}$ NMR spectrum of [(DEAM-IBY)Rh(nbd)](OTf) (**4-*i*Pr**) in $CDCl_3$.

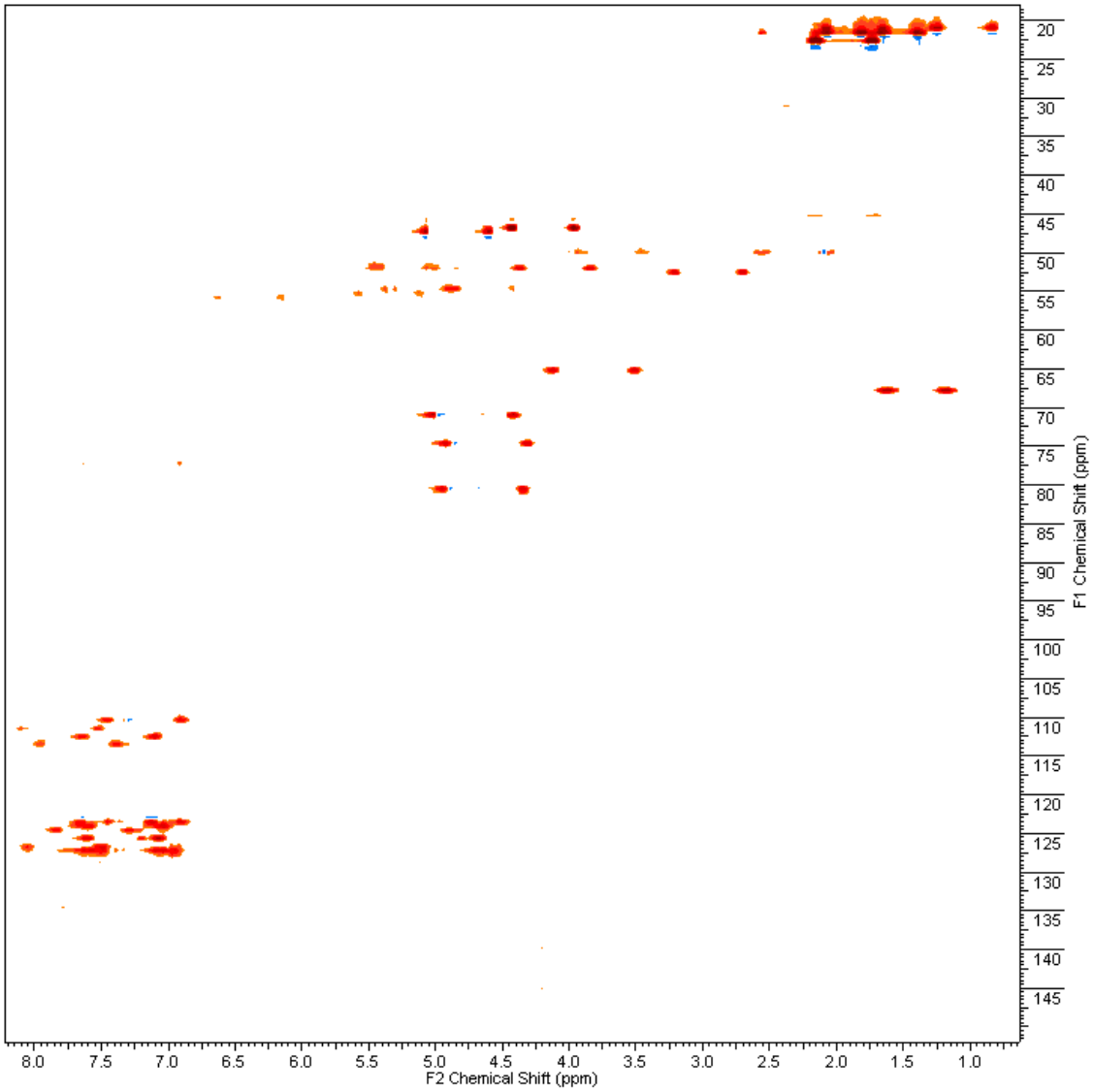

Figure S17: gHMQC NMR spectrum of [(DEAM-IBY)Rh(nbd)](OTf) (**4-^{i}Pr**) in $CDCl_3$.

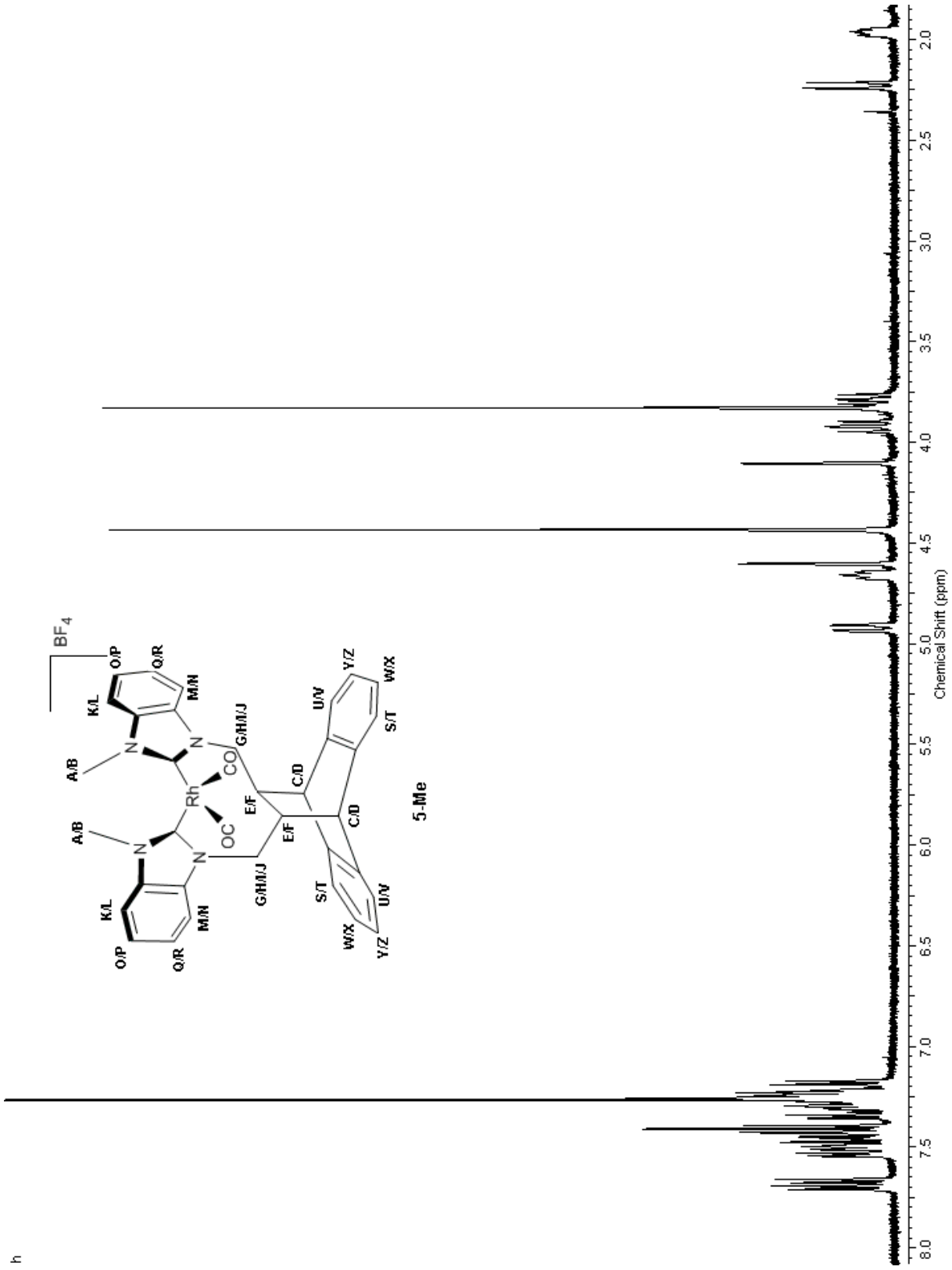

Figure S18: ^{1}H NMR spectrum of [(DEAM-MbBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$.

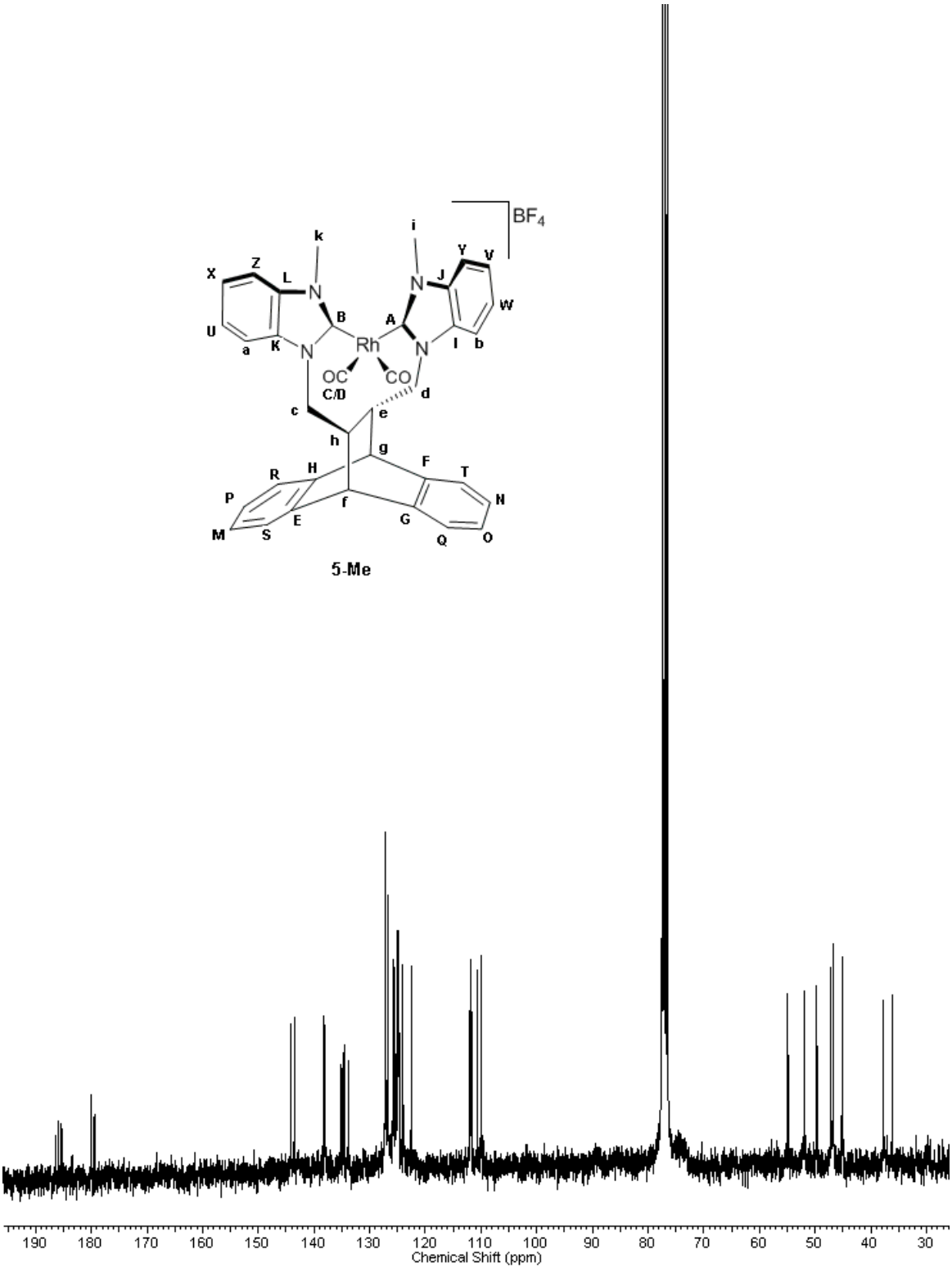

Figure S19: $^{13}C\{^{1}H\}$ NMR spectrum of $[(DEAM\text{-}MbBY)Rh(CO)_2](BF_4)$ (**5-Me**) in $CDCl_3$.

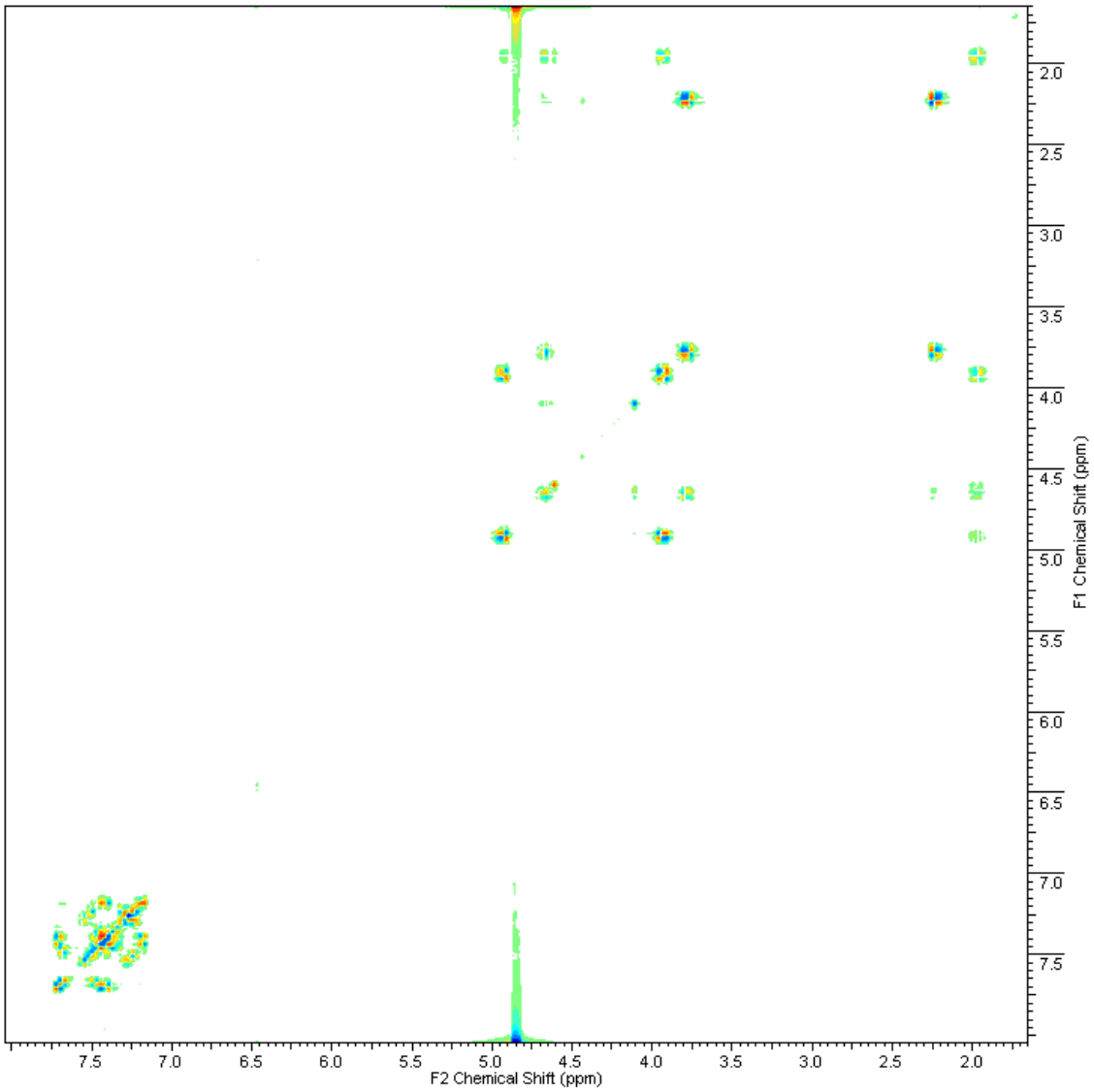

Figure S20. gdqCOSY spectrum of [(DEAM–MbBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$.

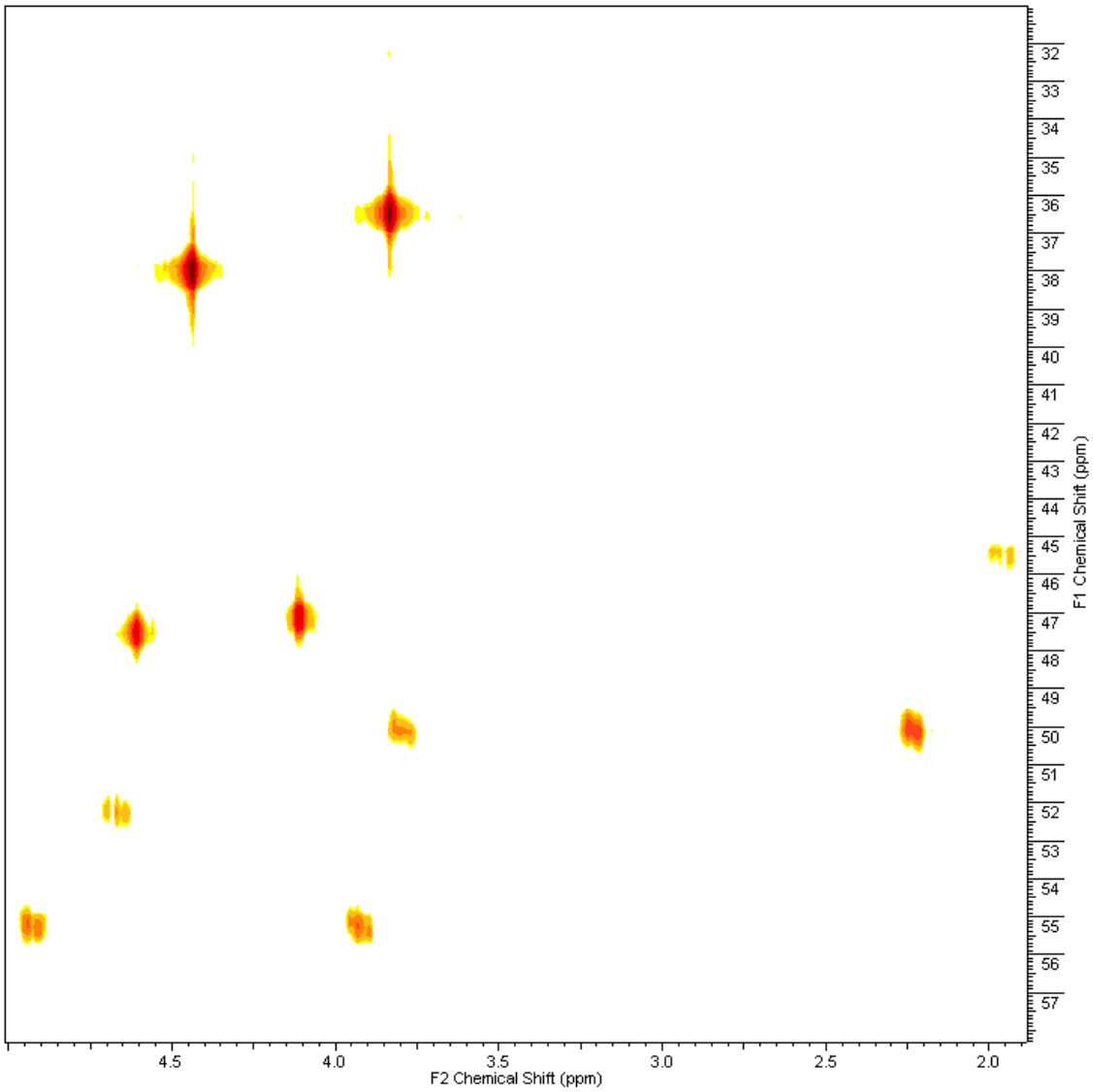

Figure S21. gHMQC spectrum expansion of $[(DEAM–MbBY)Rh(CO)_2](BF_4)$ (**5-Me**) in $CDCl_3$.

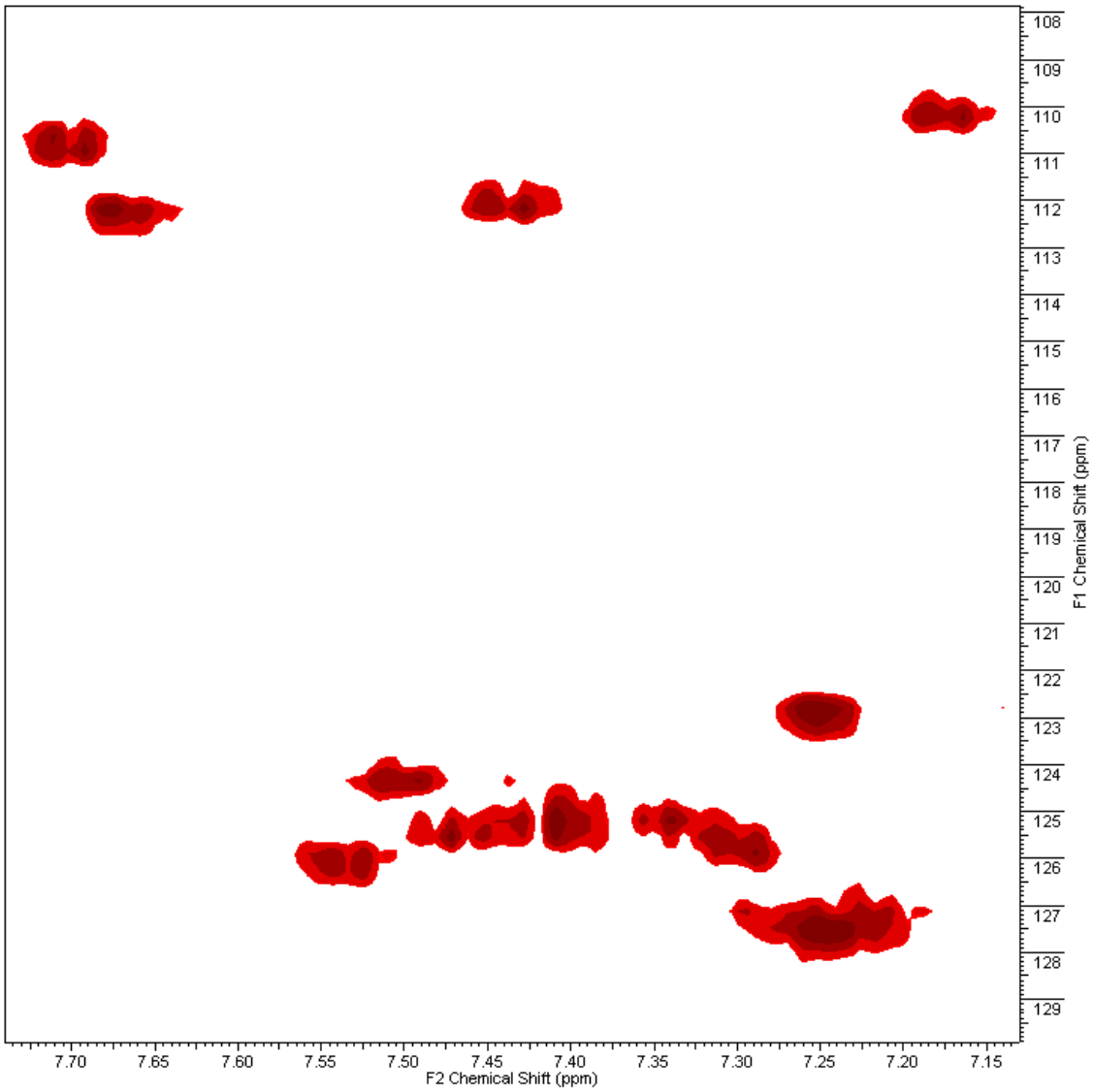

Figure S22. gHMQC spectrum expansion of $[(DEAM–MbBY)Rh(CO)_2](BF_4)$ (**5-Me**) in $CDCl_3$.

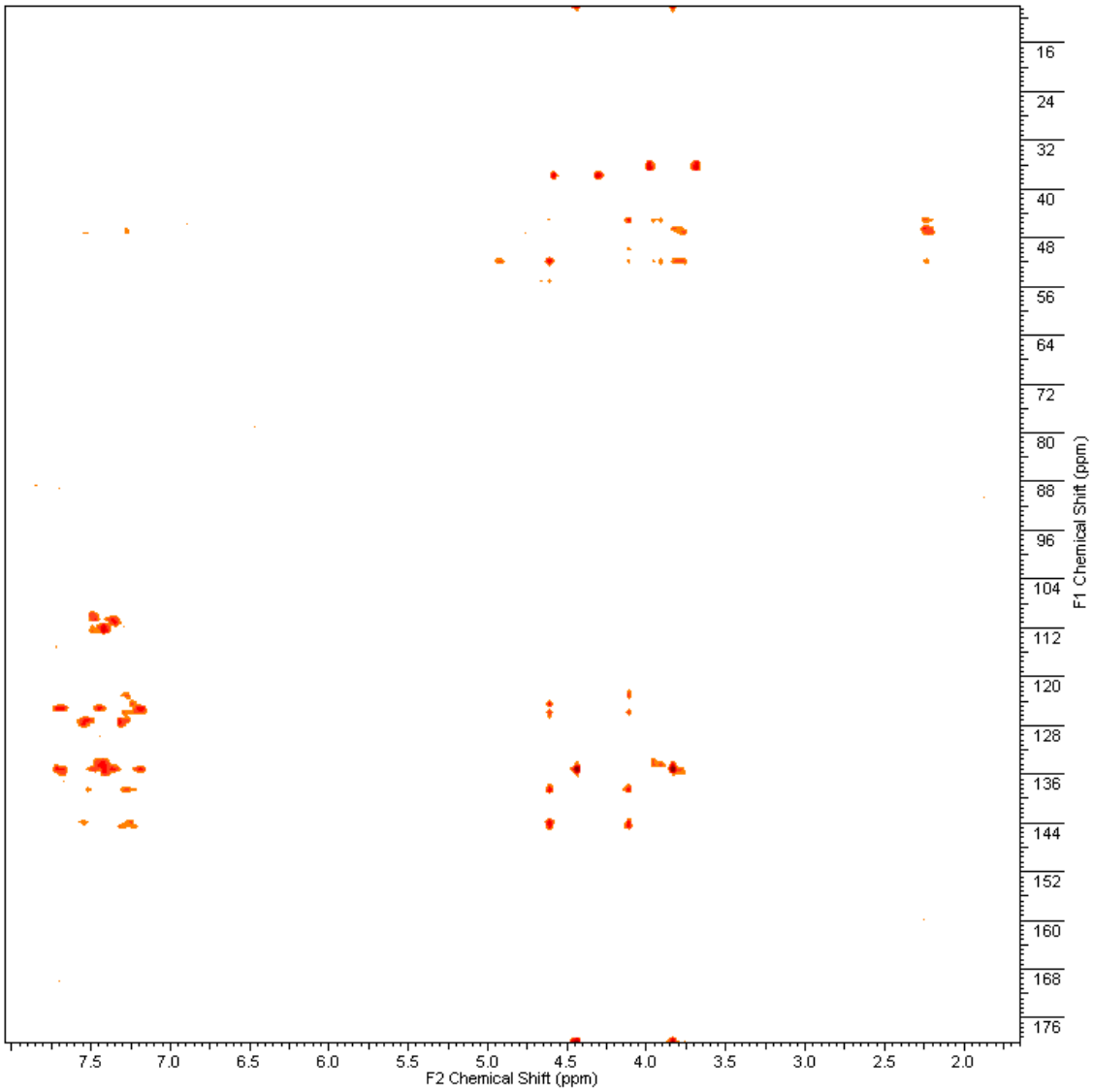

Figure S23. gHMBC spectrum of [(DEAM–MbBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$.

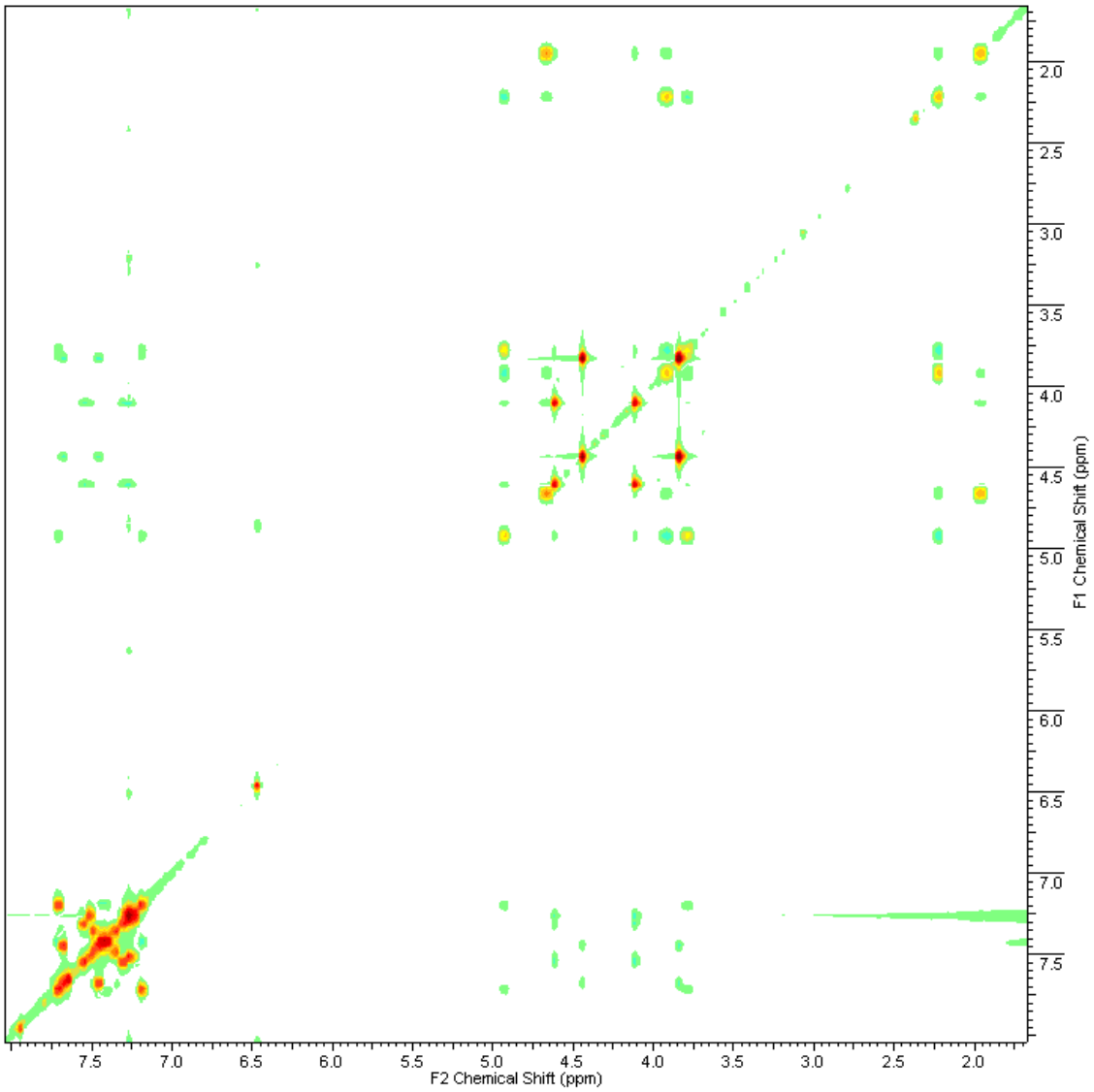

Figure S24. NOESY spectrum of [(DEAM–MbBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$.

Experimental details and data for the degenerate isomerization of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (5-Me).

Experimental:

NMR spectra were recorded on a Varian Inova spectrometer equipped with a 5 mm indirect detection probe, operating at 500 MHz for H1 and at 125 MHz for C13. Chemical shifts are reported in ppm relative to TMS.

The assignment of the proton and carbon chemical shifts, given in Table S6, was based on the proton-proton and proton-carbon (both one-bond and long-range) couplings seen in the gDQCOSY, gHMQC and gHMBC spectra, correspondingly. The NOESY spectrum at 25 ºC taken with a mixing time of 1 s displayed exchange resonances between protons with similar connectivity but different stereochemistry, e.g. H13 – H14 or H15 – H16. Of the two methylene protons in position 17, the one with two large coupling constants, therefore anti-periplanar to H16, exchanges with the proton exhibiting only one large coupling constant on C26. The stereochemical discrimination between C17 and C26 was determined from the nOe's observed in the CYCLENOE spectra at -15 ºC. In one of the methylene groups, assigned therefore to position 17, the proton *anti* to H15 displays an nOe with a doublet from the closest imidazole moiety (H19), while in the other methylene group, assigned as 26, it is the proton gauche to H16 which displays an nOe with H28. The assignment of the anthracene fragment was based on the nOe between 4.92 and 7.54 ppm.

Proton chemical shifts calculated in PERCH[1] are given for representative protons in Table S7, in parentheses. They demonstrate that unusual chemical shifts for H16 (1.97 ppm) and H17 (2.23 ppm) are due to anisotropic shielding from the N3-N4 benzimidazole, and they fully support the assignment.

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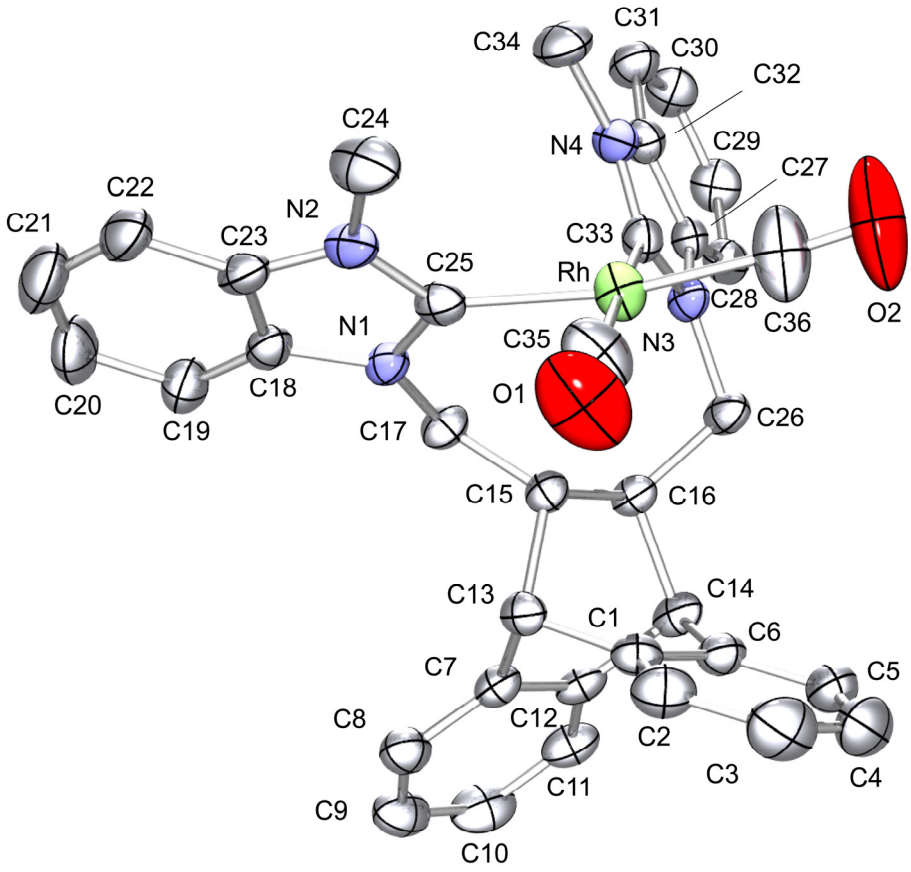


Figure S25. X-ray structure of compound [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**).

Table S6. H1 and C13 chemical shifts in compound [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**). The numbering of the position is given in Fig.**S25**.

Position	**δ C (ppm)**	**δ H (ppm), multiplicity, J (Hz)**
1	143.7	-
2	127.0	7.29, m, 1H
3	127.4	7.25, m, 1H
4	122.8	7.25, m, 1H
5	126.0	7.54 [7.49], d, 8.3 Hz, 1H
6	138.1	-
7	138.5	-
8	125.7	7.30 [7.42], m, 1H
9	127.0	7.25, m, 1H
10	127.4	7.24, m, 1H
11	124.2	7.50 [7.48], d, 8.3 Hz, 1H
12	144.3	-
13	47.0	4.10 [4.18], d, 1.6 Hz, 1H
14	47.4	4.60 [4.45], d, 1.6 Hz, 1H
15	52.1	4.66 [3.04], ddd, 9.6, 7.2, 1.6 Hz, 1H
16	45.4	1.97 [1.86], dddd, 11.0, 7.2, 2.5, 1.6 Hz, 1H
17	49.9	2.23 [2.64], d, 14.4 Hz, 1H 3.79 [3.70], dd, 14.4, 9.6 Hz
18	135.3	-
19	110.1	7.18 [7.62], d, 7.8 Hz, 1H
20	125.2	7.41, t, 7.8 Hz, 1H

21	125.1	7.47, t, 7.8 Hz, 1H
22	112.2	7.67 [7.65], d, 8.3 Hz, 1H
23	135.0	-
24	37.9	4.44 [4.24], s, 3H
25	nm	-
26	55.1	3.92 [4.24], dd, 14.0, 11.0 Hz, 1H 4.92 [4.34], dd, 14.0, 2.5 Hz, 1H
27	134.0	-
28	110.8	7.70 [7.80], d, 7.6 Hz, 1H
29	124.9	7.41, t, 7.8 Hz, 1H
30	124.9	7.34, t, 7.8 Hz, 1H
31	112.0	7.44 [7.50], d, 7.8 Hz, 1H
32	134.8	-
33	nm	-
34	36.4	3.83 [3.85], s, 3H
35	nm	-
36	nm	-

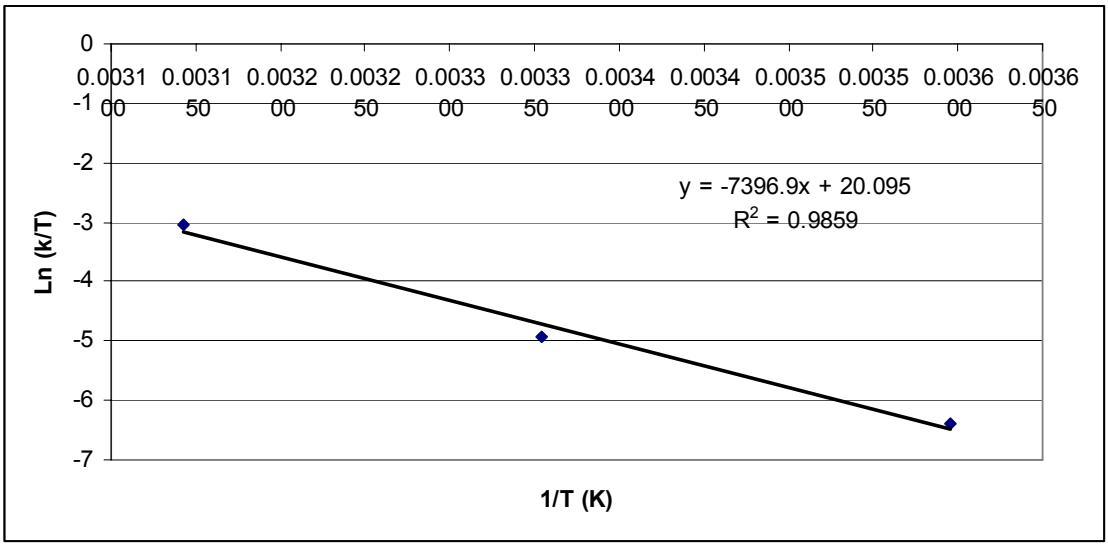
Table S7. Comparison between the experimental and calculated proton chemical shifts for the two branches of compound [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**).

Branch A		Branch B	
Experimental	Calculated	Experimental	Calculated
3.83	3.85	4.44	4.42
3.92	4.24	2.23	2.64
4.92	4.34	3.79	3.70
1.97	1.86	4.66	3.04
4.60	4.45	4.10	4.17
7.70	7.80	7.18	7.62
7.44	7.50	7.67	7.65
7.54	7.49	7.29	7.29
7.50	7.48	7.30	7.42

The ring inversion rates were measured in a chloroform-*d* solution, by the method of Forsen and Hoffman.[2] Limited solubility precluded the use of C13 signals at natural abundance, therefore proton signals were used, and as a consequence the data suffer from the interference of the nOe's. The signal at 3.79, MB(∞), was monitored after the proton at 4.92 was selectively irradiated for 10 s, in an nOe difference experiment at 5, 25, and 45 °C. The time to reach equilibrium was monitored and the temperature used in the calculation was measured by the thermocouple in the probe. The barrier to ring inversion was calculated as 16.9(18) kcal/mol.

Temp (ºC)	T1 (s)	MB(∞)	k=(1-MB(∞))/MB(∞)/T1	1/T	ln(k/T)
5	0.32	0.87	0.47	0.003595	-6.28972
25	0.37	0.56	2.12	0.003354	-4.94454
45	0.40	0.14	15.36	0.003143	-3.03097

ΔH (kcal/mol)	**ΔS (cal/mole K)**	**ΔG (25°C) (kcal /mol)**
14.7 +/- 1.8	-7 +/- 6	16.9 +/- 1.8



[2] Forsen, S.; Hoffman, R.A., Acta Chem. Scand. **17**, 1787 (1963); J. Chem. Phys. **39**, 2892 (1963).

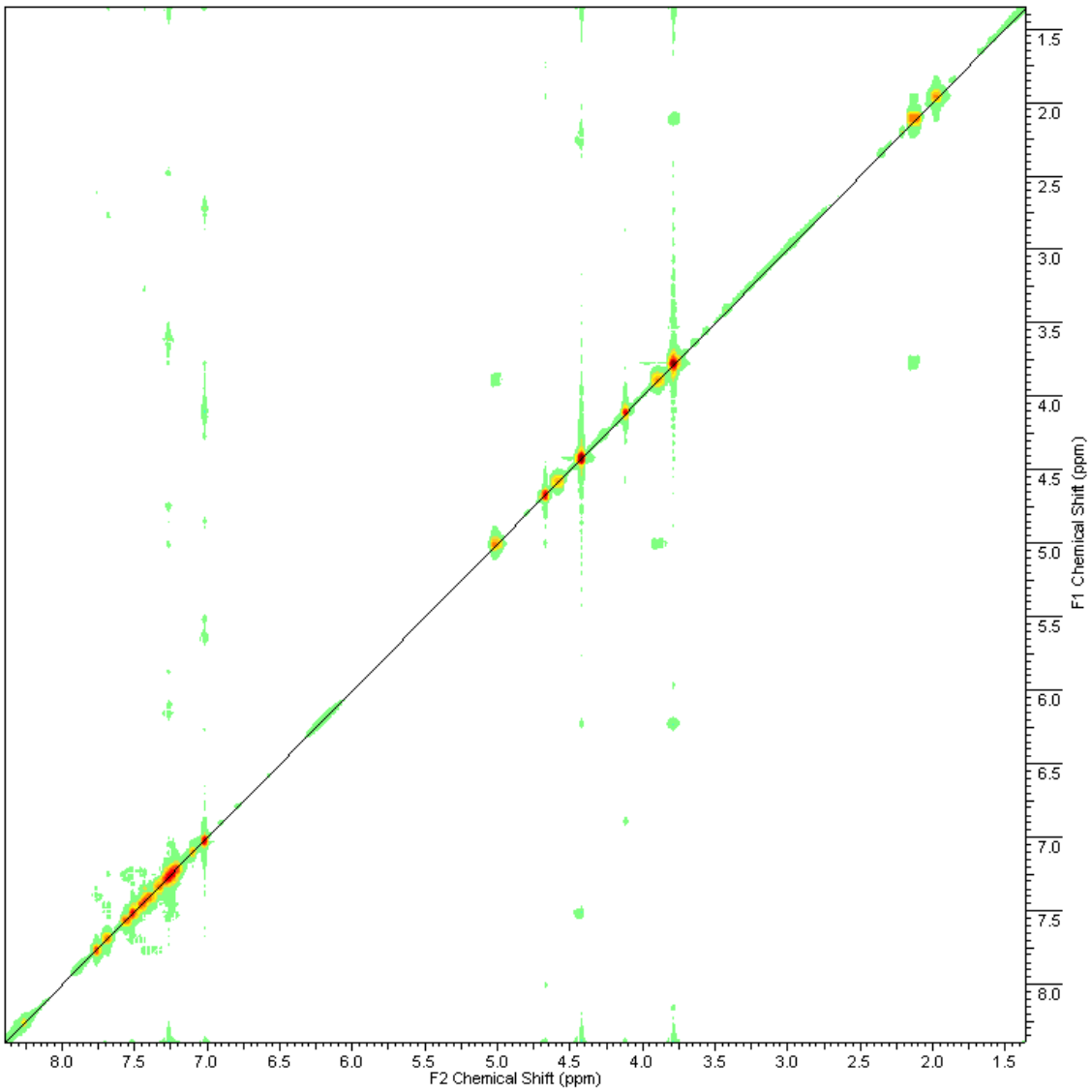


Figure S26. NOESY spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$ at 45 °C.

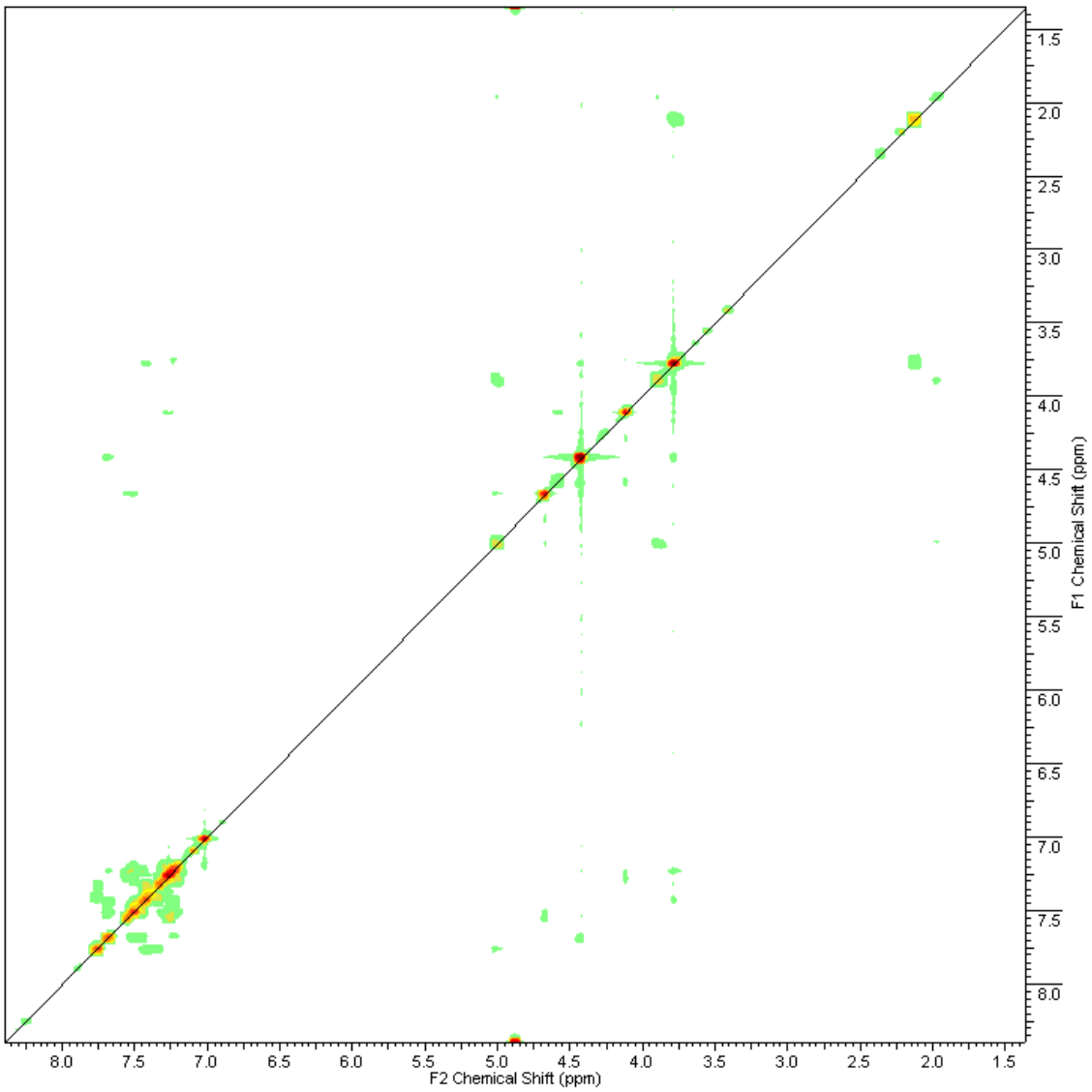


Figure S27. ROESY spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in CDCl$_3$ at 45 °C.

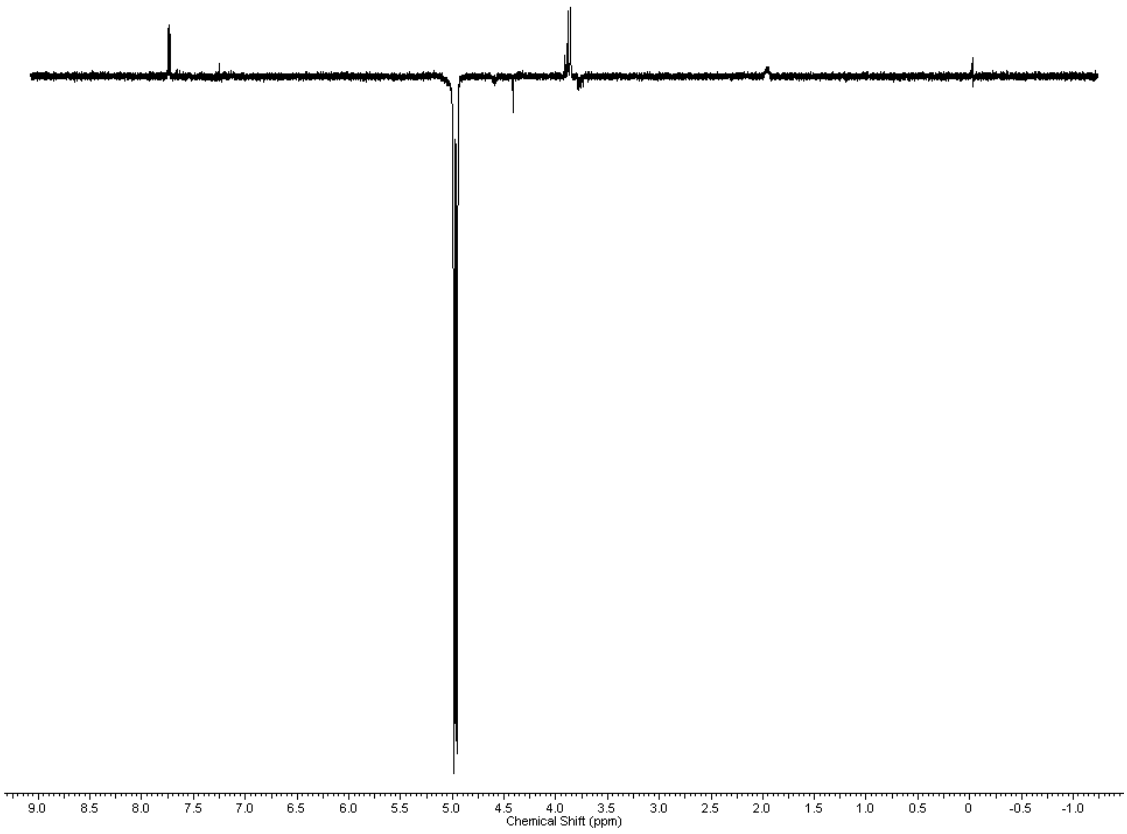


Figure S28. nOe difference spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in $CDCl_3$ at -15 °C.

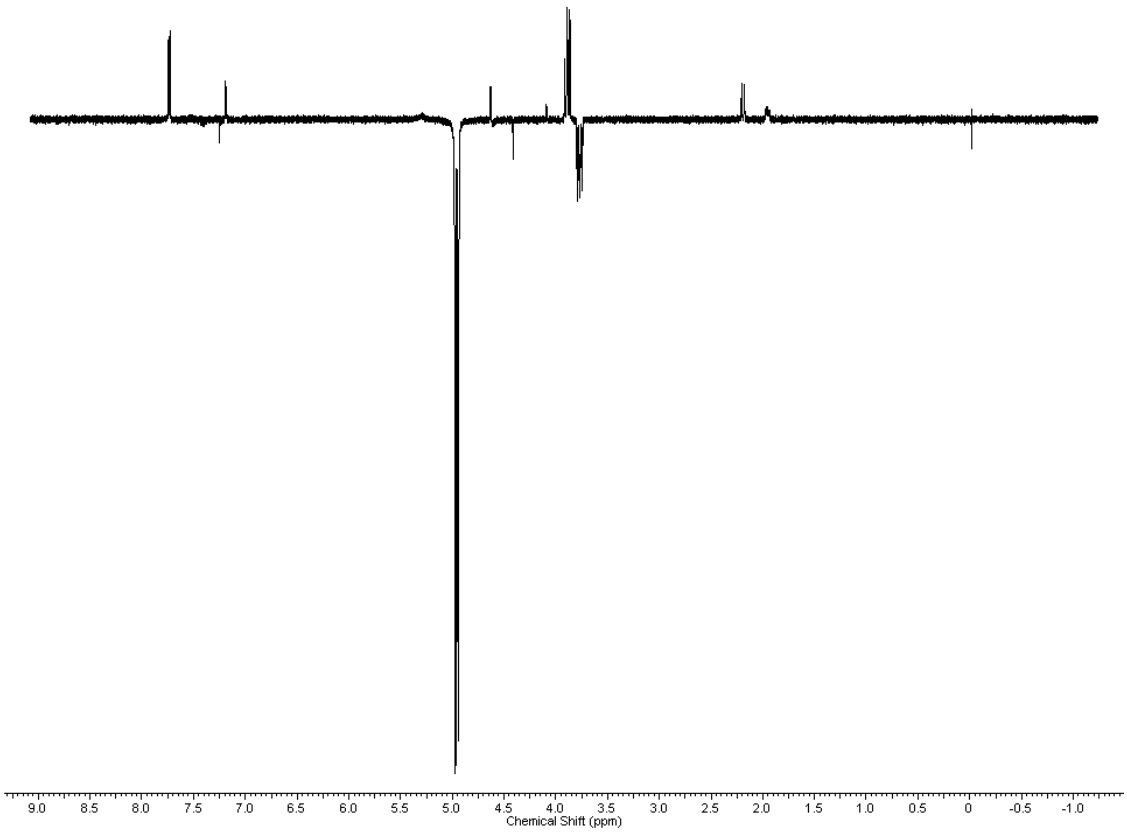


Figure S29. nOe difference spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in $CDCl_3$ at 5 °C.

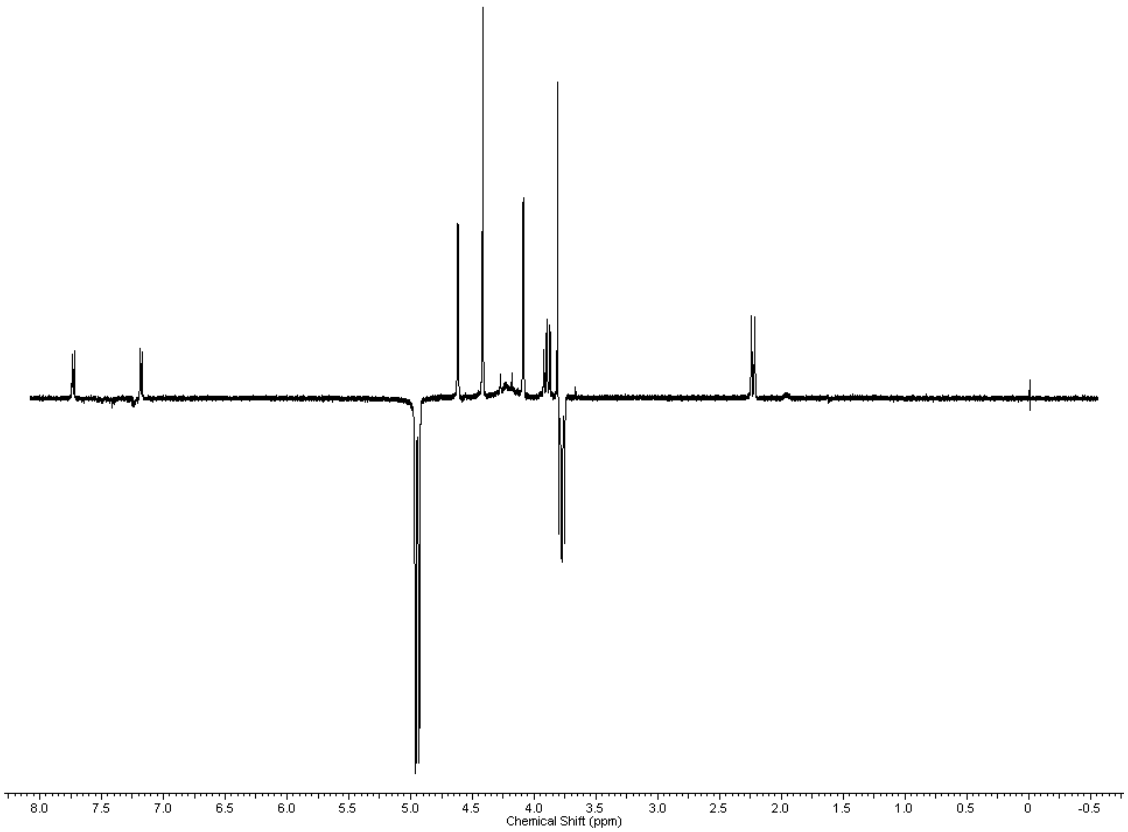


Figure S30. nOe difference spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in $CDCl_3$ at 25 °C.

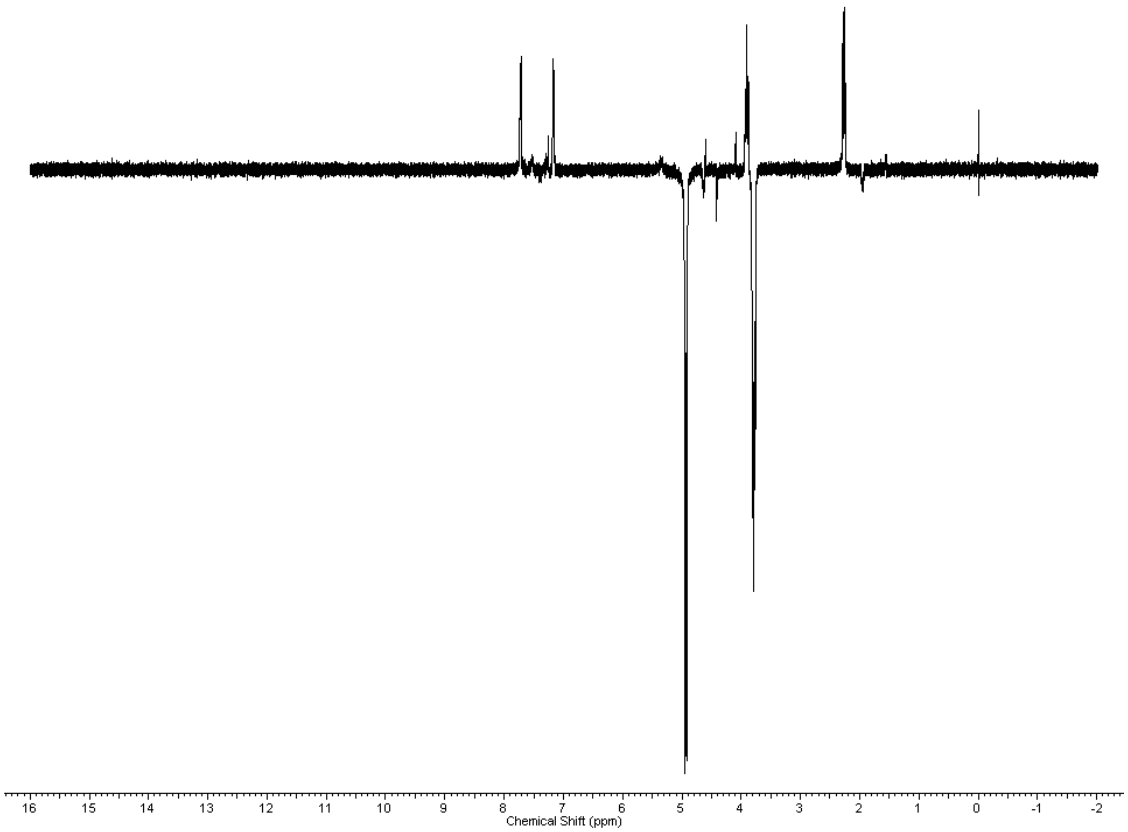


Figure S31. nOe difference spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) in $CDCl_3$ at 45 °C.

X–ray experimental details for [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (5-Me).

Data were collected at 173 K on a Siemens SMART PLATFORM equipped with A CCD area detector and a graphite monochromator utilizing MoKα radiation (λ = 0.71073 Å). Cell parameters were refined using up to 8192 reflections. A full sphere of data (1850 frames) was collected using the ω-scan method (0.3° frame width). The first 50 frames were re-measured at the end of data collection to monitor instrument and crystal stability (maximum correction on I was < 1 %). Absorption corrections by integration were applied based on measured indexed crystal faces.

The structure was solved by the Direct Methods in *SHELXTL6,* and refined using full-matrix least squares. The non-H atoms were treated anisotropically, whereas the hydrogen atoms were calculated in ideal positions and were riding on their respective carbon atoms. The asymmetric unit consists of two chemically equivalent but crystallographically independent complex cations, two tetrafluoroborate anions and four chloroform molecules of crystallization. Both borate anions are disordered by rotation around one of the B-F bonds. Two of the chloroform molecules are disordered. One of the chloroform molecules was disordered in the position of one chlorine atom. The other chloroform was refined in four parts. A total of 1077 parameters were refined in the final cycle of refinement using 11647 reflections with I > 2σ(I) to yield R_1 and wR_2 of 4.86% and 11.78%, respectively. Refinement was done using F^2.

SHELXTL6 (2000). Bruker-AXS, Madison, Wisconsin, USA.

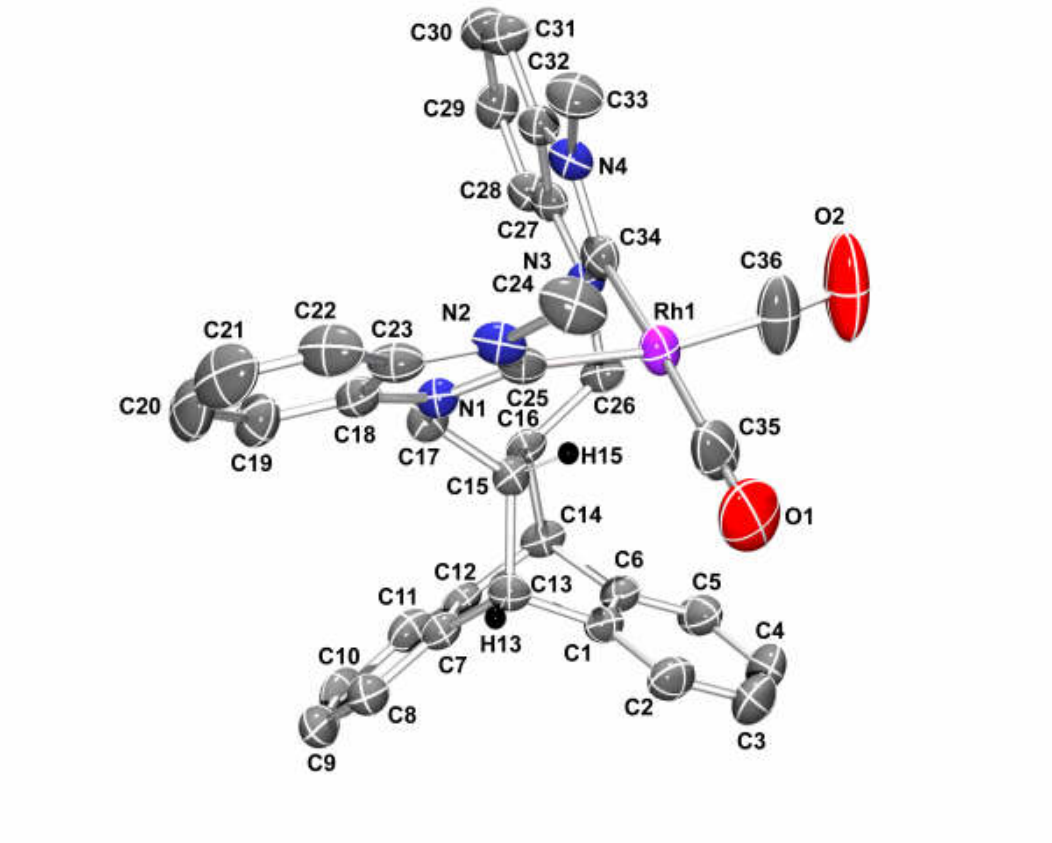


Figure S32. Molecular Structure of [(DEAM-MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) with ellipsoids drawn at the 50% probability level. Hydrogen atoms and BF$_4$ counter ion have been omitted for clarity.

Table S8. Crystal data, structure solution and refinement for [(DEAM–MBY)Rh(CO)$_2$] (BF_4) (**5–Me**).

identification code	tj01
empirical formula	$C_{76}H_{64}N_8Cl_{12}Rh_2B_2F_8O_8$
formula weight	1958.19
T (K)	173(2)
λ (Å)	0.71073
crystal system	Monoclinic
space group	P2(1)/c
a (Å)	20.039(2)
b (Å)	20.668(3)
c (Å)	20.407(2)
α (deg)	90
β (deg)	97.533(2)
γ (deg)	90
V (Å^3)	8379.0(17)
Z	4
ρ_{calcd} (mg mm^{-3})	1.552
crystal size (mm^3)	0.22 x 0.19 x 0.09
abs coeff (mm^{-1})	0.847
F(000)	3936
θ range for data collection	1.02 to 27.50
limiting indices	$-26 \le h \le 25$, $-26 \le k \le 26$, $-26 \le \ell \le 16$
no. of reflns collcd	56208
no. of ind reflns (R_{int})	19074 (0.0799)
completeness to θ = 27.50°	99.1%
absorption corr	Integration
refinement method	Full–matrix least–squares on F^2
data / restraints / parameters	19074 / 52 / 1077
$R1$,[a] $wR2$[b] [I > 2σ]	0.0486, 0.1178
$R1$,[a] $wR2$[b] (all data)	0.0890, 0.1306
GOF[c] on F^2	0.979
largest diff. peak and hole	1.142 and –0.711 eÅ^{-3}

$R1 = \sum(||F_o| - |F_c||) / \sum|F_o|$

$wR2 = [\sum[w(F_o^2 - F_c^2)^2] / \sum[w(F_o^2)^2]]^{1/2}$

$S = [\sum[w(F_o^2 - F_c^2)^2] / (n-p)]^{1/2}$

$w = 1/[\sigma^2(F_o^2)+(m*p)^2+n*p]$, $p = [\max(F_o^2,0)+ 2* F_c^2]/3$, m & n are constants.

Table S9. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$ x 10^3) for [(DEAM-MBY)Rh$(CO)_2$](BF_4) (**5-Me**). U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

Atom	X	Y	Z	U(eq)
Rh1	4274(1)	5852(1)	2437(1)	36(1)
O1	3460(2)	6187(2)	3540(2)	86(1)
O2	3113(2)	5929(3)	1355(2)	112(2)
N1	5708(1)	5873(1)	3238(1)	29(1)
N2	5078(2)	5138(1)	3604(1)	35(1)
N3	5093(1)	6156(1)	1307(1)	29(1)
N4	5068(2)	5126(1)	1479(1)	34(1)
C1	4780(2)	7827(2)	2822(2)	31(1)
C2	4217(2)	7929(2)	3123(2)	43(1)
C3	3683(2)	8280(2)	2789(2)	58(1)
C4	3715(2)	8520(2)	2173(2)	57(1)
C5	4278(2)	8415(2)	1859(2)	45(1)
C6	4813(2)	8072(2)	2185(2)	34(1)
C7	5993(2)	7902(2)	3068(2)	31(1)
C8	6446(2)	8097(2)	3600(2)	39(1)
C9	6936(2)	8549(2)	3503(2)	48(1)
C10	6979(2)	8785(2)	2873(2)	51(1)
C11	6531(2)	8580(2)	2338(2)	42(1)
C12	6028(2)	8149(2)	2437(2)	33(1)
C13	5399(2)	7458(2)	3095(2)	29(1)
C14	5472(2)	7911(2)	1923(2)	33(1)
C15	5447(2)	6893(2)	2599(2)	28(1)
C16	5529(2)	7159(2)	1904(2)	29(1)
C17	5963(2)	6369(2)	2823(2)	33(1)
C18	6102(2)	5564(2)	3757(2)	33(1)
C19	6767(2)	5641(2)	4033(2)	43(1)
C20	6990(2)	5236(2)	4553(2)	57(1)
C21	6583(3)	4780(2)	4790(2)	61(1)
C22	5924(2)	4691(2)	4521(2)	50(1)
C23	5694(2)	5101(2)	3990(2)	36(1)
C24	4508(2)	4736(2)	3701(2)	53(1)

C25	5083(2)	5610(2)	3139(2)	31(1)
C26	5016(2)	6862(2)	1368(2)	34(1)
C27	5458(2)	5872(2)	843(2)	30(1)
C28	5782(2)	6126(2)	341(2)	36(1)
C29	6082(2)	5688(2)	-40(2)	43(1)
C30	6073(2)	5024(2)	80(2)	46(1)
C31	5751(2)	4771(2)	579(2)	43(1)
C32	5445(2)	5211(2)	961(2)	34(1)
C33	4917(2)	4493(2)	1751(2)	47(1)
C34	4858(2)	5697(2)	1692(2)	32(1)
C35	3760(2)	6046(2)	3130(2)	56(1)
C36	3540(2)	5925(3)	1770(2)	65(1)
Rh2	-867(1)	2445(1)	9651(1)	37(1)
O3	-1764(2)	3588(2)	9257(2)	91(1)
O4	-1956(2)	1471(2)	9227(2)	85(1)
N5	526(2)	3103(1)	9647(1)	35(1)
N6	-48(2)	3607(2)	10312(1)	45(1)
N7	-5(1)	1236(1)	9481(1)	28(1)
N8	18(2)	1485(1)	10517(1)	33(1)
C37	-500(2)	2669(2)	7688(2)	28(1)
C38	-1084(2)	3029(2)	7596(2)	38(1)
C39	-1675(2)	2755(2)	7301(2)	52(1)
C40	-1686(2)	2111(2)	7112(2)	54(1)
C41	-1108(2)	1741(2)	7208(2)	40(1)
C42	-513(2)	2016(2)	7493(2)	29(1)
C43	712(2)	2724(2)	7650(2)	33(1)
C44	1199(2)	3148(2)	7487(2)	46(1)
C45	1644(2)	2942(2)	7062(2)	64(1)
C46	1613(2)	2330(2)	6820(3)	71(2)
C47	1141(2)	1889(2)	6992(2)	51(1)
C48	691(2)	2087(2)	7411(2)	35(1)
C49	164(2)	2870(2)	8066(2)	28(1)
C50	153(2)	1680(2)	7666(2)	30(1)
C51	232(2)	2401(1)	8663(2)	27(1)
C52	315(2)	1702(2)	8431(2)	28(1)
C53	771(2)	2598(2)	9225(2)	34(1)

C54	914(2)	3637(2)	9881(2)	42(1)
C55	1539(2)	3863(2)	9752(2)	52(1)
C56	1775(3)	4419(2)	10078(2)	60(1)
C57	1400(3)	4737(2)	10504(2)	69(2)
C58	794(3)	4520(2)	10629(2)	64(1)
C59	547(2)	3959(2)	10304(2)	47(1)
C60	-571(3)	3789(2)	10717(2)	64(1)
C61	-62(2)	3082(2)	9908(2)	38(1)
C62	-145(2)	1243(2)	8758(2)	31(1)
C63	427(2)	795(2)	9838(2)	28(1)
C64	785(2)	272(2)	9634(2)	35(1)
C65	1154(2)	-84(2)	10134(2)	38(1)
C66	1161(2)	77(2)	10798(2)	42(1)
C67	804(2)	595(2)	11000(2)	38(1)
C68	436(2)	954(2)	10498(2)	32(1)
C69	-125(2)	1813(2)	11118(2)	46(1)
C70	-248(2)	1661(2)	9902(2)	31(1)
C71	-1435(2)	3162(2)	9413(2)	54(1)
C72	-1565(2)	1851(2)	9399(2)	54(1)
F1	5148(2)	1482(2)	645(1)	97(1)
B1	4820(3)	1804(2)	132(2)	52(1)
F2	5268(3)	1830(4)	-337(4)	74(2)
F3	4267(3)	1419(4)	-225(4)	107(3)
F4	4523(6)	2362(4)	210(3)	116(5)
F2A	4784(9)	1569(5)	-418(4)	160(8)
F3A	4192(3)	1842(4)	419(4)	87(3)
F4A	5062(4)	2454(4)	202(4)	82(3)
B2	297(2)	4777(1)	7807(1)	42(1)
F5	-22(1)	4338(1)	7367(1)	59(1)
F6	742(1)	5136(1)	7481(1)	63(1)
F7	669(2)	4462(1)	8323(1)	67(1)
F8	-162(2)	5193(1)	8015(1)	72(1)
F6'	-20(17)	4773(19)	8370(10)	260(30)
F7'	959(7)	4602(14)	7970(20)	290(30)
F8'	260(20)	5384(4)	7534(11)	500(60)
C73	2762(2)	3043(2)	4684(3)	72(1)

Cl1	2297(3)	2896(3)	5339(3)	55(1)
Cl2	2323(2)	3656(3)	4196(2)	55(1)
Cl3	2955(2)	2369(2)	4299(4)	55(1)
Cl1A	2062(7)	3407(7)	4885(7)	116(4)
Cl2A	2353(7)	2925(7)	3722(7)	116(4)
Cl3A	2610(7)	2183(7)	4882(7)	116(4)
Cl1B	2393(5)	2758(5)	5253(5)	58(2)
Cl2B	2503(5)	3693(5)	4160(5)	58(2)
Cl3B	2813(5)	2404(3)	4012(7)	58(2)
Cl1C	2210(5)	3038(4)	5364(5)	64(2)
Cl2C	2348(4)	3478(5)	4051(4)	64(2)
Cl3C	2921(3)	2213(4)	4536(6)	64(2)
Cl4	1848(1)	5704(1)	3650(1)	74(1)
Cl5	2064(1)	5494(1)	2296(1)	58(1)
Cl6A	1760(30)	4435(6)	3240(30)	108(10)
Cl6	1521(3)	4472(3)	2985(3)	58(1)
C74	1568(2)	5292(2)	2918(2)	46(1)
Cl7	2503(1)	5030(1)	7374(1)	131(1)
Cl8	2396(1)	5529(1)	8674(1)	85(1)
Cl9	2709(1)	4191(1)	8492(1)	75(1)
C75	2266(2)	4863(2)	8149(2)	63(1)
Cl10	7337(1)	2281(1)	619(1)	133(1)
Cl11	6494(2)	3341(1)	216(1)	190(1)
Cl12	6743(1)	2412(1)	-715(1)	121(1)
C76	6634(2)	2532(2)	107(3)	69(1)

Table S10. Bond lengths (in Å) for [(DEAM–MBY)Rh(CO_2)](BF_4) (**5-Me**).

Bond	Distance	Bond	Distance
Rh1-C36	1.874(5)	C13-C15	1.557(4)
Rh1-C35	1.897(4)	C13-H13A	1.0000
Rh1-C34	2.063(3)	C14-C16	1.558(4)
Rh1-C25	2.078(4)	C14-H14A	1.0000
O1-C35	1.132(5)	C15-C17	1.526(4)
O2-C36	1.122(5)	C15-C16	1.549(4)
N1-C25	1.356(4)	C15-H15A	1.0000
N1-C18	1.391(4)	C16-C26	1.529(5)
N1-C17	1.465(4)	C16-H16A	1.0000
N2-C25	1.362(4)	C17-H17A	0.9900
N2-C23	1.377(5)	C17-H17B	0.9900
N2-C24	1.446(5)	C18-C23	1.382(5)
N3-C34	1.355(4)	C18-C19	1.387(5)
N3-C27	1.400(4)	C19-C20	1.380(5)
N3-C26	1.475(4)	C19-H19A	0.9500
N4-C34	1.344(4)	C20-C21	1.376(6)
N4-C32	1.388(4)	C20-H20A	0.9500
N4-C33	1.468(4)	C21-C22	1.373(6)
C1-C2	1.371(5)	C21-H21A	0.9500
C1-C6	1.405(5)	C22-C23	1.405(5)
C1-C13	1.500(5)	C22-H22A	0.9500
C2-C3	1.395(6)	C24-H24A	0.9800
C2-H2A	0.9500	C24-H24B	0.9800
C3-C4	1.360(6)	C24-H24C	0.9800
C3-H3A	0.9500	C26-H26A	0.9900
C4-C5	1.386(6)	C26-H26B	0.9900
C4-H4A	0.9500	C27-C28	1.385(5)
C5-C6	1.380(5)	C27-C32	1.388(4)
C5-H5A	0.9500	C28-C29	1.380(5)
C6-C14	1.526(5)	C28-H28A	0.9500
C7-C8	1.381(5)	C29-C30	1.395(5)
C7-C12	1.396(5)	C29-H29A	0.9500
C7-C13	1.511(5)	C30-C31	1.380(5)
C8-C9	1.388(5)	C30-H30A	0.9500
C8-H8A	0.9500	C31-C32	1.391(5)
C9-C10	1.387(6)	C31-H31A	0.9500
C9-H9A	0.9500	C33-H33A	0.9800
C10-C11	1.386(6)	C33-H33B	0.9800
C10-H10A	0.9500	C33-H33C	0.9800
C11-C12	1.381(5)	Rh2-C72	1.882(5)
C11-H11A	0.9500	Rh2-C71	1.893(4)
C12-C14	1.507(5)	Rh2-C70	2.064(3)

Rh2-C61	2.095(4)	C52-C62	1.535(4)
O3-C71	1.121(5)	C52-H52A	1.0000
O4-C72	1.131(5)	C53-H53A	0.9900
N5-C61	1.355(4)	C53-H53B	0.9900
N5-C54	1.398(4)	C54-C59	1.376(5)
N5-C53	1.476(4)	C54-C55	1.394(6)
N6-C61	1.361(4)	C55-C56	1.381(5)
N6-C59	1.399(5)	C55-H55A	0.9500
N6-C60	1.466(5)	C56-C57	1.387(7)
N7-C70	1.362(4)	C56-H56A	0.9500
N7-C63	1.395(4)	C57-C58	1.350(7)
N7-C62	1.464(4)	C57-H57A	0.9500
N8-C70	1.347(4)	C58-C59	1.394(5)
N8-C68	1.384(4)	C58-H58A	0.9500
N8-C69	1.463(4)	C60-H60A	0.9800
C37-C38	1.379(5)	C60-H60B	0.9800
C37-C42	1.407(4)	C60-H60C	0.9800
C37-C49	1.506(5)	C62-H62A	0.9900
C38-C39	1.379(5)	C62-H62B	0.9900
C38-H38A	0.9500	C63-C68	1.386(4)
C39-C40	1.384(6)	C63-C64	1.390(5)
C39-H39A	0.9500	C64-C65	1.390(5)
C40-C41	1.381(5)	C64-H64A	0.9500
C40-H40A	0.9500	C65-C66	1.394(5)
C41-C42	1.379(5)	C65-H65A	0.9500
C41-H41A	0.9500	C66-C67	1.380(5)
C42-C50	1.505(4)	C66-H66A	0.9500
C43-C44	1.384(5)	C67-C68	1.394(5)
C43-C48	1.402(5)	C67-H67A	0.9500
C43-C49	1.506(4)	C69-H69A	0.9800
C44-C45	1.389(5)	C69-H69B	0.9800
C44-H44A	0.9500	C69-H69C	0.9800
C45-C46	1.357(7)	F1-B1	1.336(5)
C45-H45A	0.9500	B1-F2A	1.216(8)
C46-C47	1.391(6)	B1-F4	1.319(7)
C46-H46A	0.9500	B1-F2	1.396(8)
C47-C48	1.383(5)	B1-F4A	1.430(9)
C47-H47A	0.9500	B1-F3A	1.460(8)
C48-C50	1.513(5)	B1-F3	1.477(9)
C49-C51	1.549(4)	B2-F8	1.366(3)
C49-H49A	1.0000	B2-F8'	1.371(4)
C50-C52	1.553(4)	B2-F7	1.373(3)
C50-H50A	1.0000	B2-F7'	1.373(4)
C51-C53	1.524(5)	B2-F5	1.375(3)
C51-C52	1.537(4)	B2-F6'	1.384(4)
C51-H51A	1.0000	B2-F6	1.393(3)

C73-Cl1B	1.570(10)	Cl4-C74	1.747(4)
C73-Cl3	1.670(6)	Cl5-C74	1.761(4)
C73-Cl1A	1.689(14)	Cl6A-C74	1.91(2)
C73-Cl2C	1.698(8)	Cl6-C74	1.703(7)
C73-Cl2B	1.752(10)	C74-H74A	1.0000
C73-Cl1	1.754(8)	Cl7-C75	1.742(5)
C73-Cl2	1.772(6)	Cl8-C75	1.742(5)
C73-Cl3C	1.779(9)	Cl9-C75	1.744(5)
C73-Cl3A	1.857(15)	C75-H75A	1.0000
C73-Cl1C	1.884(10)	Cl10-C76	1.719(5)
C73-Cl3B	1.916(11)	Cl11-C76	1.715(5)
C73-Cl2A	2.040(15)	Cl12-C76	1.737(5)
C73-H73A	1.0000	C76-H76A	1.0000

Symmetry transformations used to generate equivalent atoms.

Table S11. Bond angles in (°) for [(DEAM–MBY)Rh(CO_2)](BF_4) (**5-Me**).

Bond Angle	Angle	Bond Angle	Angle
C36-Rh1-C35	94.23(19)	C10-C9-C8	120.0(4)
C36-Rh1-C34	86.77(16)	C10-C9-H9A	120.00
C35-Rh1-C34	176.40(17)	C8-C9-H9A	120.00
C36-Rh1-C25	170.64(19)	C11-C10-C9	120.7(4)
C35-Rh1-C25	89.34(16)	C11-C10-H10A	119.70
C34-Rh1-C25	90.19(13)	C9-C10-H10A	119.70
C25-N1-C18	110.5(3)	C12-C11-C10	119.5(4)
C25-N1-C17	125.5(3)	C12-C11-H11A	120.30
C18-N1-C17	123.8(3)	C10-C11-H11A	120.30
C25-N2-C23	110.5(3)	C11-C12-C7	119.8(4)
C25-N2-C24	125.8(3)	C11-C12-C14	126.6(3)
C23-N2-C24	123.7(3)	C7-C12-C14	113.6(3)
C34-N3-C27	110.6(3)	C1-C13-C7	107.1(3)
C34-N3-C26	126.5(3)	C1-C13-C15	104.9(3)
C27-N3-C26	122.9(3)	C7-C13-C15	108.4(3)
C34-N4-C32	111.1(3)	C1-C13-H13A	112.00
C34-N4-C33	124.8(3)	C7-C13-H13A	112.00
C32-N4-C33	124.1(3)	C15-C13-H13A	112.00
C2-C1-C6	120.1(3)	C12-C14-C6	106.2(3)
C2-C1-C13	126.9(3)	C12-C14-C16	107.1(3)
C6-C1-C13	113.1(3)	C6-C14-C16	107.2(3)
C1-C2-C3	119.0(4)	C12-C14-H14A	112.00
C1-C2-H2A	120.50	C6-C14-H14A	112.00
C3-C2-H2A	120.50	C16-C14-H14A	112.00
C4-C3-C2	121.0(4)	C17-C15-C16	112.1(3)
C4-C3-H3A	119.50	C17-C15-C13	115.7(3)
C2-C3-H3A	119.50	C16-C15-C13	110.6(3)
C3-C4-C5	120.7(4)	C17-C15-H15A	105.90
C3-C4-H4A	119.70	C16-C15-H15A	105.90
C5-C4-H4A	119.70	C13-C15-H15A	105.90
C6-C5-C4	119.0(4)	C26-C16-C15	111.7(3)
C6-C5-H5A	120.50	C26-C16-C14	111.9(3)
C4-C5-H5A	120.50	C15-C16-C14	108.3(2)
C5-C6-C1	120.2(3)	C26-C16-H16A	108.30
C5-C6-C14	126.7(3)	C15-C16-H16A	108.30
C1-C6-C14	113.1(3)	C14-C16-H16A	108.30
C8-C7-C12	120.8(3)	N1-C17-C15	113.3(3)
C8-C7-C13	126.1(3)	N1-C17-H17A	108.90
C12-C7-C13	113.0(3)	C15-C17-H17A	108.90
C7-C8-C9	119.2(4)	N1-C17-H17B	108.90
C7-C8-H8A	120.40	C15-C17-H17B	108.90
C9-C8-H8A	120.40	H17A-C17-H17B	107.70

C23-C18-C19	121.4(3)	C30-C31-H31A	121.70
C23-C18-N1	106.1(3)	C32-C31-H31A	121.70
C19-C18-N1	132.5(3)	C27-C32-N4	106.3(3)
C20-C19-C18	116.1(4)	C27-C32-C31	121.7(3)
C20-C19-H19A	121.90	N4-C32-C31	131.9(3)
C18-C19-H19A	121.90	N4-C33-H33A	109.50
C21-C20-C19	122.6(4)	N4-C33-H33B	109.50
C21-C20-H20A	118.70	H33A-C33-H33B	109.50
C19-C20-H20A	118.70	N4-C33-H33C	109.50
C22-C21-C20	122.3(4)	H33A-C33-H33C	109.50
C22-C21-H21A	118.90	H33B-C33-H33C	109.50
C20-C21-H21A	118.90	N4-C34-N3	106.3(3)
C21-C22-C23	115.5(4)	N4-C34-Rh1	127.3(2)
C21-C22-H22A	122.30	N3-C34-Rh1	126.4(2)
C23-C22-H22A	122.30	O1-C35-Rh1	177.3(4)
N2-C23-C18	106.9(3)	O2-C36-Rh1	175.2(6)
N2-C23-C22	130.9(4)	C72-Rh2-C71	92.33(17)
C18-C23-C22	122.2(4)	C72-Rh2-C70	87.53(15)
N2-C24-H24A	109.50	C71-Rh2-C70	179.54(17)
N2-C24-H24B	109.50	C72-Rh2-C61	177.52(16)
H24A-C24-H24B	109.50	C71-Rh2-C61	89.35(16)
N2-C24-H24C	109.50	C70-Rh2-C61	90.78(14)
H24A-C24-H24C	109.50	C61-N5-C54	111.3(3)
H24B-C24-H24C	109.50	C61-N5-C53	125.4(3)
N1-C25-N2	105.9(3)	C54-N5-C53	123.0(3)
N1-C25-Rh1	128.9(2)	C61-N6-C59	110.9(3)
N2-C25-Rh1	125.1(3)	C61-N6-C60	126.0(4)
N3-C26-C16	112.9(3)	C59-N6-C60	123.1(3)
N3-C26-H26A	109.00	C70-N7-C63	109.9(3)
C16-C26-H26A	109.00	C70-N7-C62	126.3(3)
N3-C26-H26B	109.00	C63-N7-C62	123.8(3)
C16-C26-H26B	109.00	C70-N8-C68	110.8(3)
H26A-C26-H26B	107.80	C70-N8-C69	124.1(3)
C28-C27-C32	121.6(3)	C68-N8-C69	125.2(3)
C28-C27-N3	132.7(3)	C38-C37-C42	119.7(3)
C32-C27-N3	105.7(3)	C38-C37-C49	126.7(3)
C29-C28-C27	116.6(3)	C42-C37-C49	113.1(3)
C29-C28-H28A	121.70	C37-C38-C39	120.2(3)
C27-C28-H28A	121.70	C37-C38-H38A	119.90
C28-C29-C30	122.0(3)	C39-C38-H38A	119.90
C28-C29-H29A	119.00	C38-C39-C40	119.9(4)
C30-C29-H29A	119.00	C38-C39-H39A	120.10
C31-C30-C29	121.4(3)	C40-C39-H39A	120.10
C31-C30-H30A	119.30	C41-C40-C39	120.8(4)
C29-C30-H30A	119.30	C41-C40-H40A	119.60
C30-C31-C32	116.7(3)	C39-C40-H40A	119.60

C42-C41-C40	119.6(3)	C51-C52-H52A	108.80
C42-C41-H41A	120.20	C50-C52-H52A	108.80
C40-C41-H41A	120.20	N5-C53-C51	111.8(3)
C41-C42-C37	119.9(3)	N5-C53-H53A	109.30
C41-C42-C50	126.8(3)	C51-C53-H53A	109.30
C37-C42-C50	113.2(3)	N5-C53-H53B	109.30
C44-C43-C48	120.1(3)	C51-C53-H53B	109.30
C44-C43-C49	126.7(3)	H53A-C53-H53B	107.90
C48-C43-C49	113.2(3)	C59-C54-C55	121.6(4)
C43-C44-C45	119.1(4)	C59-C54-N5	106.1(3)
C43-C44-H44A	120.50	C55-C54-N5	132.3(4)
C45-C44-H44A	120.50	C56-C55-C54	116.5(4)
C46-C45-C44	120.7(4)	C56-C55-H55A	121.70
C46-C45-H45A	119.60	C54-C55-H55A	121.70
C44-C45-H45A	119.60	C55-C56-C57	121.1(5)
C45-C46-C47	121.2(4)	C55-C56-H56A	119.40
C45-C46-H46A	119.40	C57-C56-H56A	119.40
C47-C46-H46A	119.40	C58-C57-C56	122.4(4)
C48-C47-C46	118.9(4)	C58-C57-H57A	118.80
C48-C47-H47A	120.60	C56-C57-H57A	118.80
C46-C47-H47A	120.60	C57-C58-C59	117.3(4)
C47-C48-C43	120.0(3)	C57-C58-H58A	121.40
C47-C48-C50	126.9(3)	C59-C58-H58A	121.40
C43-C48-C50	113.2(3)	C54-C59-C58	121.0(4)
C43-C49-C37	108.4(3)	C54-C59-N6	106.4(3)
C43-C49-C51	108.8(3)	C58-C59-N6	132.6(4)
C37-C49-C51	102.2(2)	N6-C60-H60A	109.50
C43-C49-H49A	112.30	N6-C60-H60B	109.50
C37-C49-H49A	112.30	H60A-C60-H60B	109.50
C51-C49-H49A	112.30	N6-C60-H60C	109.50
C42-C50-C48	108.1(3)	H60A-C60-H60C	109.50
C42-C50-C52	106.5(3)	H60B-C60-H60C	109.50
C48-C50-C52	105.4(3)	N5-C61-N6	105.3(3)
C42-C50-H50A	112.10	N5-C61-Rh2	127.0(2)
C48-C50-H50A	112.10	N6-C61-Rh2	127.4(3)
C52-C50-H50A	112.10	N7-C62-C52	113.5(3)
C53-C51-C52	112.9(3)	N7-C62-H62A	108.90
C53-C51-C49	113.9(3)	C52-C62-H62A	108.90
C52-C51-C49	110.4(3)	N7-C62-H62B	108.90
C53-C51-H51A	106.30	C52-C62-H62B	108.90
C52-C51-H51A	106.30	H62A-C62-H62B	107.70
C49-C51-H51A	106.30	C68-C63-C64	122.3(3)
C62-C52-C51	110.6(3)	C68-C63-N7	106.3(3)
C62-C52-C50	111.2(3)	C64-C63-N7	131.4(3)
C51-C52-C50	108.7(2)	C63-C64-C65	116.0(3)
C62-C52-H52A	108.80	C63-C64-H64A	122.00

C65-C64-H64A	122.00	F8-B2-F7	112.3(3)
C64-C65-C66	121.5(3)	F8'-B2-F7	138.1(8)
C64-C65-H65A	119.20	F8-B2-F7'	139.3(7)
C66-C65-H65A	119.20	F8'-B2-F7'	110.2(4)
C67-C66-C65	122.4(3)	F7-B2-F7'	44(2)
C67-C66-H66A	118.80	F8-B2-F5	110.1(2)
C65-C66-H66A	118.80	F8'-B2-F5	109.9(4)
C66-C67-C68	116.1(3)	F7-B2-F5	110.3(2)
C66-C67-H67A	122.00	F7'-B2-F5	109.7(3)
C68-C67-H67A	122.00	F8-B2-F6'	49(2)
N8-C68-C63	106.5(3)	F8'-B2-F6'	109.4(3)
N8-C68-C67	131.7(3)	F7-B2-F6'	68(2)
C63-C68-C67	121.7(3)	F7'-B2-F6'	109.2(3)
N8-C69-H69A	109.50	F5-B2-F6'	108.3(3)
N8-C69-H69B	109.50	F8-B2-F6	108.2(2)
H69A-C69-H69B	109.50	F8'-B2-F6	48(2)
N8-C69-H69C	109.50	F7-B2-F6	107.6(2)
H69A-C69-H69C	109.50	F7'-B2-F6	66(2)
H69B-C69-H69C	109.50	F5-B2-F6	108.1(2)
N8-C70-N7	106.5(3)	F6'-B2-F6	142.3(7)
N8-C70-Rh2	126.4(2)	Cl1B-C73-Cl3	101.2(5)
N7-C70-Rh2	127.0(2)	Cl1B-C73-Cl1A	60.1(6)
O3-C71-Rh2	178.1(4)	Cl3-C73-Cl1A	137.5(6)
O4-C72-Rh2	175.4(4)	Cl1B-C73-Cl2C	122.1(6)
F2A-B1-F4	119.2(6)	Cl3-C73-Cl2C	101.8(4)
F2A-B1-F1	119.3(6)	Cl1A-C73-Cl2C	67.1(6)
F4-B1-F1	121.6(5)	Cl1B-C73-Cl2B	127.5(7)
F2A-B1-F2	49.4(9)	Cl3-C73-Cl2B	115.0(6)
F4-B1-F2	113.0(7)	Cl1A-C73-Cl2B	67.5(6)
F1-B1-F2	105.3(5)	Cl2C-C73-Cl2B	18.9(4)
F2A-B1-F4A	116.5(9)	Cl1B-C73-Cl1	12.2(4)
F4-B1-F4A	47.0(5)	Cl3-C73-Cl1	113.1(3)
F1-B1-F4A	105.3(5)	Cl1A-C73-Cl1	49.5(6)
F2-B1-F4A	78.1(5)	Cl2C-C73-Cl1	114.6(4)
F2A-B1-F3A	116.2(11)	Cl2B-C73-Cl1	116.9(5)
F4-B1-F3A	58.4(6)	Cl1B-C73-Cl2	115.9(5)
F1-B1-F3A	94.2(4)	Cl3-C73-Cl2	117.5(4)
F2-B1-F3A	159.9(6)	Cl1A-C73-Cl2	56.7(5)
F4A-B1-F3A	102.0(6)	Cl2C-C73-Cl2	15.7(3)
F2A-B1-F3	51.7(9)	Cl2B-C73-Cl2	12.5(3)
F4-B1-F3	102.0(8)	Cl1-C73-Cl2	106.1(3)
F1-B1-F3	112.2(5)	Cl1B-C73-Cl3C	82.8(6)
F2-B1-F3	101.0(5)	Cl3-C73-Cl3C	19.5(3)
F4A-B1-F3	141.1(6)	Cl1A-C73-Cl3C	130.1(6)
F3A-B1-F3	66.1(5)	Cl2C-C73-Cl3C	117.6(4)
F8-B2-F8'	63(2)	Cl2B-C73-Cl3C	133.0(6)

Cl1-C73-Cl3C	95.0(4)	Cl3A-C73-Cl2A	92.2(6)
Cl2-C73-Cl3C	133.0(4)	Cl1C-C73-Cl2A	120.5(5)
Cl1B-C73-Cl3A	51.2(6)	Cl3B-C73-Cl2A	44.1(5)
Cl3-C73-Cl3A	50.0(5)	Cl1B-C73-H73A	108.70
Cl1A-C73-Cl3A	102.0(7)	Cl3-C73-H73A	106.50
Cl2C-C73-Cl3A	126.5(6)	Cl1A-C73-H73A	115.60
Cl2B-C73-Cl3A	145.4(7)	Cl2C-C73-H73A	114.20
Cl1-C73-Cl3A	63.2(6)	Cl2B-C73-H73A	96.40
Cl2-C73-Cl3A	136.4(6)	Cl1-C73-H73A	106.50
Cl3C-C73-Cl3A	31.9(5)	Cl2-C73-H73A	106.50
Cl1B-C73-Cl1C	22.3(4)	Cl3C-C73-H73A	106.90
Cl3-C73-Cl1C	122.4(4)	Cl3A-C73-H73A	117.20
Cl1A-C73-Cl1C	40.3(5)	Cl1C-C73-H73A	105.60
Cl2C-C73-Cl1C	106.7(5)	Cl3B-C73-H73A	116.30
Cl2B-C73-Cl1C	107.3(5)	Cl2A-C73-H73A	131.20
Cl1-C73-Cl1C	10.1(3)	Cl6-C74-Cl4	115.5(3)
Cl2-C73-Cl1C	97.0(4)	Cl6-C74-Cl5	109.7(3)
Cl3C-C73-Cl1C	104.7(4)	Cl4-C74-Cl5	110.6(2)
Cl3A-C73-Cl1C	72.9(6)	Cl6-C74-Cl6A	20.1(19)
Cl1B-C73-Cl3B	110.2(5)	Cl4-C74-Cl6A	97.7(19)
Cl3-C73-Cl3B	18.4(3)	Cl5-C74-Cl6A	110.9(4)
Cl1A-C73-Cl3B	127.2(6)	Cl6-C74-H74A	106.90
Cl2C-C73-Cl3B	83.5(6)	Cl4-C74-H74A	106.90
Cl2B-C73-Cl3B	97.6(6)	Cl5-C74-H74A	106.90
Cl1-C73-Cl3B	120.7(4)	Cl6A-C74-H74A	123.30
Cl2-C73-Cl3B	99.2(5)	Cl7-C75-Cl8	111.5(3)
Cl3C-C73-Cl3B	35.6(5)	Cl7-C75-Cl9	109.7(3)
Cl3A-C73-Cl3B	61.4(6)	Cl8-C75-Cl9	110.7(3)
Cl1C-C73-Cl3B	127.9(4)	Cl7-C75-H75A	108.30
Cl1B-C73-Cl2A	119.9(6)	Cl8-C75-H75A	108.30
Cl3-C73-Cl2A	62.4(5)	Cl9-C75-H75A	108.30
Cl1A-C73-Cl2A	92.6(6)	Cl11-C76-Cl10	110.5(3)
Cl2C-C73-Cl2A	40.3(5)	Cl11-C76-Cl12	107.9(3)
Cl2B-C73-Cl2A	57.0(6)	Cl10-C76-Cl12	110.3(3)
Cl1-C73-Cl2A	121.8(5)	Cl11-C76-H76A	109.40
Cl2-C73-Cl2A	55.8(5)	Cl10-C76-H76A	109.40
Cl3C-C73-Cl2A	77.3(6)	Cl12-C76-H76A	109.40

Symmetry transformations used to generate equivalent atoms.

Table S12. Anisotropic displacement parameters ($Å^2$ x10^3) for [(DEAM–MBY)Rh(CO_2)](BF_4) (**5-Me**). 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}
Rh1	32(1)	46(1)	32(1)	3(1)	8(1)	-5(1)
O1	59(2)	136(3)	70(2)	-25(2)	34(2)	-4(2)
O2	46(2)	228(5)	60(2)	41(3)	-4(2)	-2(3)
N1	36(2)	25(1)	25(2)	4(1)	7(1)	0(1)
N2	51(2)	28(2)	26(2)	1(1)	11(1)	-7(1)
N3	34(2)	29(1)	24(2)	3(1)	6(1)	1(1)
N4	43(2)	31(2)	27(2)	2(1)	4(1)	-5(1)
C1	32(2)	28(2)	33(2)	-4(2)	9(2)	-1(2)
C2	44(2)	44(2)	46(2)	-7(2)	17(2)	1(2)
C3	40(3)	67(3)	68(3)	-15(3)	15(2)	9(2)
C4	42(3)	59(3)	69(3)	-12(2)	-2(2)	16(2)
C5	53(3)	39(2)	43(2)	-4(2)	0(2)	10(2)
C6	38(2)	28(2)	35(2)	-4(2)	6(2)	4(2)
C7	35(2)	25(2)	34(2)	3(2)	10(2)	4(2)
C8	40(2)	36(2)	42(2)	-4(2)	6(2)	3(2)
C9	36(2)	43(2)	63(3)	-13(2)	3(2)	-1(2)
C10	35(2)	39(2)	83(3)	-2(2)	20(2)	-8(2)
C11	45(2)	32(2)	55(3)	3(2)	20(2)	2(2)
C12	36(2)	22(2)	42(2)	0(2)	14(2)	7(2)
C13	39(2)	27(2)	22(2)	1(1)	9(2)	-1(2)
C14	47(2)	27(2)	28(2)	5(2)	15(2)	3(2)
C15	29(2)	26(2)	28(2)	3(1)	6(1)	-1(1)
C16	36(2)	25(2)	28(2)	3(1)	12(2)	4(2)
C17	35(2)	29(2)	36(2)	7(2)	9(2)	-2(2)
C18	42(2)	28(2)	30(2)	0(2)	6(2)	7(2)
C19	44(2)	45(2)	40(2)	6(2)	3(2)	9(2)
C20	51(3)	68(3)	47(3)	4(2)	-8(2)	21(2)
C21	86(4)	56(3)	41(3)	17(2)	5(3)	23(3)
C22	83(3)	34(2)	34(2)	8(2)	6(2)	6(2)
C23	60(3)	22(2)	28(2)	1(2)	11(2)	2(2)
C24	75(3)	45(2)	41(2)	4(2)	18(2)	-19(2)

C25	43(2)	29(2)	24(2)	-4(1)	11(2)	-1(2)
C26	50(2)	27(2)	26(2)	3(1)	8(2)	6(2)
C27	34(2)	32(2)	24(2)	2(2)	-1(2)	1(2)
C28	40(2)	38(2)	31(2)	0(2)	7(2)	-4(2)
C29	44(2)	52(2)	37(2)	2(2)	16(2)	0(2)
C30	53(3)	49(2)	39(2)	-7(2)	13(2)	9(2)
C31	58(3)	34(2)	38(2)	-1(2)	7(2)	7(2)
C32	39(2)	34(2)	27(2)	-1(2)	4(2)	-1(2)
C33	67(3)	28(2)	47(3)	4(2)	8(2)	-8(2)
C34	34(2)	34(2)	28(2)	4(2)	-1(2)	-1(2)
C35	39(2)	77(3)	53(3)	-9(2)	14(2)	-10(2)
C36	36(2)	116(4)	45(3)	24(3)	11(2)	-1(3)
Rh2	43(1)	29(1)	40(1)	5(1)	12(1)	6(1)
O3	64(2)	43(2)	159(4)	7(2)	-6(2)	18(2)
O4	45(2)	48(2)	161(4)	-4(2)	8(2)	3(2)
N5	46(2)	33(2)	25(2)	-3(1)	1(1)	-2(1)
N6	74(2)	38(2)	26(2)	-3(1)	16(2)	-1(2)
N7	38(2)	24(1)	23(2)	4(1)	4(1)	2(1)
N8	46(2)	30(2)	25(2)	4(1)	8(1)	3(1)
C37	35(2)	28(2)	21(2)	4(1)	5(1)	4(2)
C38	40(2)	32(2)	42(2)	5(2)	5(2)	5(2)
C39	38(2)	45(2)	69(3)	9(2)	-5(2)	6(2)
C40	43(2)	54(3)	59(3)	3(2)	-11(2)	-9(2)
C41	53(2)	33(2)	32(2)	-1(2)	-3(2)	-2(2)
C42	39(2)	30(2)	17(2)	3(1)	3(2)	5(2)
C43	37(2)	34(2)	28(2)	9(2)	5(2)	4(2)
C44	42(2)	41(2)	57(3)	14(2)	13(2)	2(2)
C45	52(3)	59(3)	88(4)	30(3)	37(3)	7(2)
C46	71(3)	69(3)	83(4)	28(3)	51(3)	31(3)
C47	68(3)	43(2)	47(3)	11(2)	27(2)	20(2)
C48	41(2)	34(2)	29(2)	7(2)	8(2)	10(2)
C49	32(2)	23(2)	28(2)	3(1)	5(2)	1(1)
C50	46(2)	22(2)	23(2)	-1(1)	5(2)	6(2)
C51	34(2)	24(2)	22(2)	0(1)	3(1)	3(1)
C52	35(2)	24(2)	23(2)	2(1)	0(1)	4(1)
C53	42(2)	32(2)	26(2)	-3(2)	0(2)	-2(2)

C54	62(3)	37(2)	24(2)	1(2)	-4(2)	-6(2)
C55	65(3)	48(2)	41(2)	-1(2)	-1(2)	-16(2)
C56	75(3)	53(3)	50(3)	-5(2)	-3(2)	-26(2)
C57	113(5)	42(2)	51(3)	-4(2)	4(3)	-29(3)
C58	115(4)	38(2)	37(3)	-12(2)	10(3)	-11(3)
C59	81(3)	33(2)	26(2)	0(2)	7(2)	-6(2)
C60	93(4)	55(3)	49(3)	-10(2)	30(3)	6(3)
C61	59(3)	31(2)	23(2)	3(2)	5(2)	2(2)
C62	43(2)	26(2)	23(2)	0(1)	3(2)	0(2)
C63	35(2)	25(2)	24(2)	6(1)	4(2)	-5(2)
C64	37(2)	33(2)	34(2)	7(2)	6(2)	0(2)
C65	37(2)	34(2)	43(2)	13(2)	6(2)	4(2)
C66	38(2)	44(2)	43(2)	13(2)	-5(2)	-1(2)
C67	43(2)	41(2)	27(2)	8(2)	-2(2)	-3(2)
C68	36(2)	31(2)	29(2)	6(2)	6(2)	-4(2)
C69	72(3)	44(2)	25(2)	2(2)	14(2)	5(2)
C70	39(2)	28(2)	27(2)	5(2)	8(2)	-3(2)
C71	48(3)	37(2)	76(3)	4(2)	6(2)	6(2)
C72	38(2)	36(2)	90(4)	7(2)	12(2)	9(2)
F1	145(3)	92(2)	53(2)	26(2)	11(2)	40(2)
B1	84(4)	42(3)	33(3)	11(2)	20(3)	16(3)
F2	67(4)	99(5)	59(5)	16(4)	25(3)	-15(3)
F3	74(5)	148(7)	105(7)	-38(5)	31(4)	-26(4)
F4	225(12)	70(5)	55(4)	4(3)	27(6)	87(8)
F2A	340(20)	110(9)	28(4)	-15(5)	9(10)	126(13)
F3A	59(4)	116(7)	84(5)	-9(5)	2(3)	-4(4)
F4A	89(6)	64(4)	98(6)	22(4)	32(5)	-4(4)
B2	52(3)	34(2)	41(3)	-6(2)	6(2)	5(2)
F5	57(2)	58(2)	60(2)	-22(1)	4(1)	-2(1)
F6	62(2)	57(2)	70(2)	3(2)	10(2)	-20(2)
F7	85(2)	52(2)	57(2)	12(1)	-24(2)	-6(2)
F8	113(3)	53(2)	56(2)	-11(2)	29(2)	29(2)
C73	52(3)	65(3)	96(4)	17(3)	5(3)	5(2)
Cl4	91(1)	76(1)	55(1)	0(1)	10(1)	-35(1)
Cl5	61(1)	50(1)	68(1)	7(1)	30(1)	3(1)
Cl6A	146(18)	48(3)	150(20)	33(6)	93(18)	28(6)

Cl6	68(2)	32(2)	73(2)	12(1)	13(2)	-8(1)
C74	45(2)	39(2)	57(3)	3(2)	15(2)	-2(2)
Cl7	151(2)	171(2)	70(1)	45(1)	14(1)	-13(1)
Cl8	90(1)	60(1)	100(1)	5(1)	-15(1)	1(1)
Cl9	76(1)	67(1)	82(1)	14(1)	14(1)	10(1)
C75	47(3)	73(3)	67(3)	6(3)	-2(2)	-12(2)
Cl10	97(1)	229(2)	69(1)	39(1)	-5(1)	-12(1)
Cl11	368(4)	60(1)	177(2)	-14(1)	172(2)	35(2)
Cl12	101(1)	168(2)	80(1)	-66(1)	-37(1)	71(1)
C76	59(3)	55(3)	92(4)	-13(3)	12(3)	1(2)

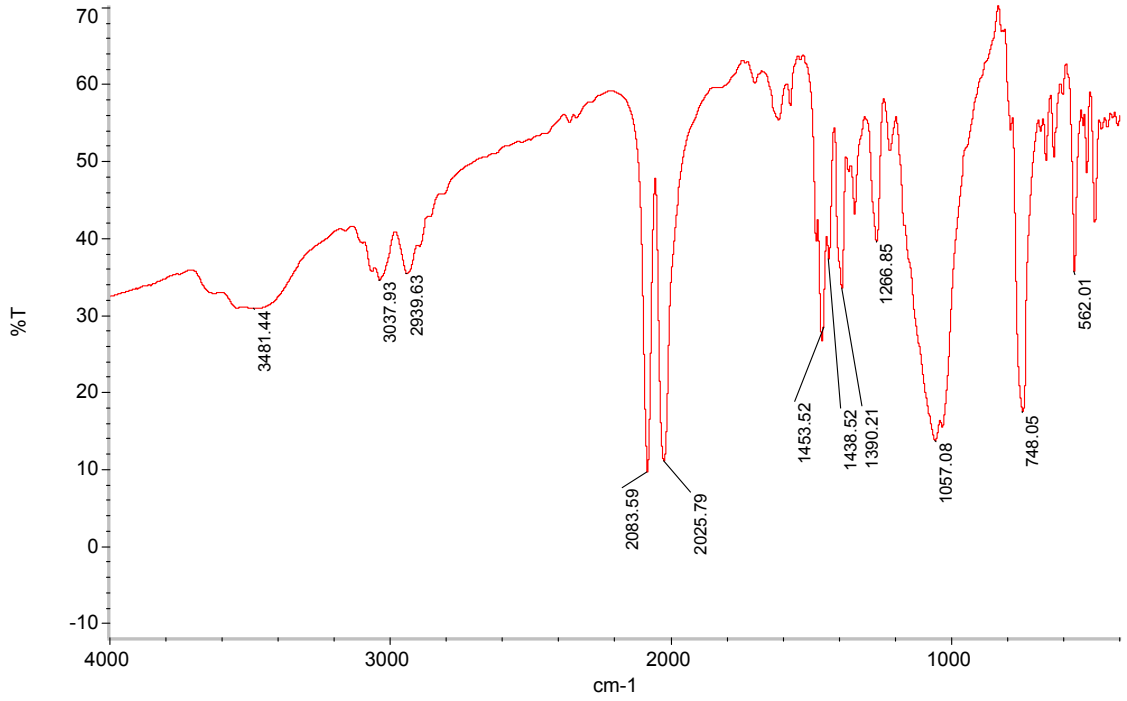


Figure S33. IR spectrum of [(DEAM–MBY)Rh(CO)$_2$](BF$_4$) (**5-Me**) (KBr pellet).