Supplementary Information

Table S1. Selected bond lengths (Å) and angles (°) for **1a**-**7d**.

	1a•C_7H_8		
Nd1-N1	2.396(4)	Nd1-C1	2.786(5)
Nd1-N2	2.356(4)	P2-C1	1.728(5)
Nd1-I1	3.1203(6)	P1-N1	1.621(4)
Nd1-I2	3.1387(7)	P2-N2	1.610(4)
Nd1-O1	2.497(3)	P1-C1	1.725(5)
P1-C1-P2	134.0(3)	N1-P1-C1	108.4(2)
N1-Nd1-N2	116.28(14)	N2-P2-C1	107.8(2)
P1-N1-Nd1	102.78(19)	P2-N2-Nd1	102.49(19)
I1-Nd1-I2	156.167(13)	O1-Nd1-C1	151.54(13)
	1b•OC_4H_8		
Gd1-N1	2.412(7)	Gd1-C1	2.657(9)
Gd1-N2	2.383(7)	P2-C1	1.759(9)
Gd1-I1	3.0442(7)	P1-N1	1.603(8)
Gd1-I2	3.0234(8)	P2-N2	1.608(8)
Gd1-O1	2.372(6)	P1-C1	1.752(8)
P1-C1-P2	122.9(5)	N1-P1-C1	107.2(4)
N1-Gd1-N2	93.6(2)	N2-P2-C1	108.0(4)
P1-N1-Gd1	99.0(3)	P2-N2-Gd1	99.6(3)
I1-Gd1-I2	97.20(2)	O1-Gd1-C1	86.5(2)
	1c•OC_4H_8		
Tb1-N1	2.394(3)	Tb1-C1	2.609(4)
Tb1-N2	2.351(3)	P2-C1	1.747(4)
Tb1-I1	2.9948(5)	P1-N1	1.599(3)
Tb1-I2	3.0192(5)	P2-N2	1.599(3)
Tb1-O1	2.367(3)	P1-C1	1.747(4)
P1-C1-P2	122.9(2)	N1-P1-C1	106.42(17)
N1-Tb1-N2	93.40(10)	N2-P2-C1	107.05(17)
P1-N1-Tb1	98.55(14)	P2-N2-Tb1	99.71(15)
I1-Tb1-I2	96.364(9)	O1-Tb1-C1	87.30(10)
	2•2C_7H_8		
La1-C1	2.778(8)	P1-C1	1.722(8)
La1-N1	2.426(6)	P2-C1	1.718(8)
La1-N2	2.460(6)	P1-N1	1.610(6)
La1-I1	3.1144(7)	P2-N2	1.603(7)
La1-I2	3.2253(8)	La1-I2a	3.3114(6)
P2-N2-La1	101.6(3)	N1-La1-N2	118.3(2)
I1-La1-I2	159.35(2)	P1-C1-P2	138.3(5)
La1-I2-La1a	101.699(16)	C1-La1-I1	118.37(16)
C1-La1-I2	81.26(16)	P1-N1-La1	103.3(3)

Table S1. *Cont.*

	3•3C_7H_8		
Gd1-C1	2.731(4)	P1-N1	1.627(3)
Gd1-N1	2.445(3)	P2-N2	1.625(3)
Gd1-N2	2.464(3)	Gd1-I1	3.0936(3)
Gd1-O1	2.506(3)	Gd1-I2	3.1000(3)
Gd1-O2	2.458(3)	P1-C1	1.721(4)
N1-Gd1-N2	113.23(10)	P2-C1	1.725(4)
I1-Gd-I2	155.765(9)	N2-P2-C1	103.51(16)
P1-C1-P2	134.3(2)	P1-N1-Gd1	103.62(14)
N1-P1-C1	104.29(17)	P2-N2-Gd1	100.65(14)
	5a•4C_7H_8		
Dy1-C1	2.676(2)	P2-C1	1.752(2)
Dy1-N1	2.3849(19)	P2-N2	1.631(2)
Dy1-N3	2.3584(17)	P4-N4	1.627(2)
Dy1-C44	2.679(2)	P4-C44	1.733(2)
Dy1-N2	2.3445(18)	P6-C87	1.711(2)
Dy1-N4	2.3866(19)	P6-N6	1.586(2)
P1-C1	1.735(2)	P3-C44	1.746(2)
P1-N1	1.633(2)	P5-C87	1.707(2)
P3-N3	1.6365(19)	P5-N5	1.597(2)
Dy1-N1-P1	102.63(9)	N1-Dy1-N2	105.14(6)
Dy1-N2-P2	103.92(9)	N3-Dy1-N4	104.76(6)
Dy1-N3-P3	103.03(9)	P1-C1-P2	128.19(12)
Dy1-N4-P4	103.07(9)	P3-C44-P4	129.11(13)
P5-C87-P6	133.89(15)		
	5b•5C_7H_8		
Er1-C1	2.676(5)	P2-C1	1.746(5)
Er1-N1	2.356(4)	P2-N2	1.648(4)
Er1-N3	2.386(4)	P4-N4	1.628(4)
Er1-C44	2.688(5)	P4-C44	1.740(5)
Er1-N2	2.340(4)	P6-C87	1.711(5)
Er1-N4	2.354(4)	P6-N6	1.582(4)
P1-C1	1.734(5)	P3-C44	1.742(5)
P1-N1	1.632(4)	P5-C87	1.705(5)
P3-N3	1.632(4)	P5-N5	1.589(4)
Er1-N1-P1	103.76(19)	N1-Er1-N2	103.17(14)
Er1-N2-P2	102.93(19)	N3-Er1-N4	105.15(13)
Er1-N3-P3	103.58(19)	P1-C1-P2	127.4(3)
Er1-N4-P4	103.58(19)	P3-C44-P4	129.1(3)
P5-C87-P6	128.7(3)		

Table S1. *Cont.*

	6a•3C_7H_8		
La1-C1	2.822(5)	La1-I2	3.2432(5)
La1-N1	2.564(4)	P1-C1	1.736(5)
La1-N2	2.526(4)	P2-C1	1.727(5)
La1-N3	2.852(5)	P1-N1	1.622(5)
La1-N4	2.844(4)	P2-N2	1.620(4)
La1-I1	3.2026(5)	N1-La1-N2	110.72(13)
I1-La1-12	158.395(12)	P2-C1-P1	134.1(3)
N3-La1-N4	67.43(14)	P1-N1-La1	104.1(2)
P2-N2-La1	101.87(18)	N1-P1-C1	105.0(2)
N2-P2-C1	103.6(2)		
	6b•2C_7H_8		
Gd1-C1	2.678(4)	P1-C1	1.7197(17)
Gd1-N1	2.476(3)	P1-N1	1.623(3)
Gd1-N2	2.755(3)	N1-Gd1-N1a	118.66(12)
Gd1-I1	3.0966(4)	P1-C1-P1a	139.4(3)
Gd1-I2	3.0853(4)	P1-N1-Gd1	102.00(13)
I1-Gd1-12	164.360(11)	N1-P1-C1	102.14(18)
N2-Gd1-N2a	68.67(12)		
	7a•3C_7H_8		
La1-C1	2.725(5)	La1-C44	2.731(4)
La1-N2	2.549(4)	La1-N3	2.523(3)
C1-P1	1.693(4)	C1-P2	1.691(4)
C44-P4	1.700(4)	P1-N1	1.639(3)
P3-N3	1.640(4)	P4-N4	1.635(3)
La1-N1	2.526(3)	C44-P3	1.691(4)
La1-N4	2.552(3)	P2-N2	1.624(4)
P2-N2-La1	100.07(16)	P3-C44-P4	133.5(2)
N2-P2-C1	106.6(2)	N4-P4-C44	106.5(2)
N1-La1-N2	105.28(11)	N1-P1-C1	107.6(2)
P1-C1-P2	133.9(3)	P1-N1-La1	99.63(15)
N3-La1-N4	104.42(11)	N3-P3-C44	107.5(2)
P4-N4-La1	99.74(15)	P3-N3-La1	99.92(16)
	7b•2C_6H_{14}		
Ce1-C1	2.819(11)	C1-P1	1.740(11)
Ce1-N2	2.496(9)	C44-P4	1.671(12)
Ce1-N1	2.561(9)	P3-N3	1.652(9)
Ce1-N4	2.493(10)	P2-N2	1.618(9)
Ce1-C44	2.681(11)	C1-P2	1.693(12)
Ce1-N3	2.480(9)	P1-N1	1.619(9)
C44-P3	1.698(11)	P4-N4	1.651(9)
N4-P4-C44	107.1(5)	P3-N3-Ce1	101.7(4)
N2-P2-C1	105.9(5)	P3-C44-P4	134.3(7)
P2-N2-Ce1	104.5(5)	P1-C1-P2	130.2(7)

Table S1. *Cont.*

N3-P3-C44	**103.6(5)**	**N3-Ce1-N4**	**106.5(3)**
N1-P1-C1	105.8(5)	P4-N4-Ce1	99.1(4)
P1-N1-Ce1	102.9(4)	N1-Ce1-N2	97.6(3)
	7c•3C_7H_8		
Pr1-C1	2.667(6)	P2-N2	1.633(5)
Pr1-N2	2.503(4)	P3-N3	1.631(5)
Pr1-C44	2.662(6)	C1-P2	1.702(6)
Pr1-N3	2.517(5)	P1-N1	1.624(5)
Pr1-N1	2.515(4)	P4-N4	1.645(5)
Pr1-N4	2.500(5)	C1-P1	1.702(6)
C44-P3	1.696(6)	C44-P4	1.696(6)
N2-P2-C1	106.6(3)	N1-Pr1-N2	105.87(15)
P2-N2-Pr1	99.3(2)	P1-C1-P2	132.9(3)
P3-C44-P4	134.0(4)	N3-Pr1-N4	107.00(15)
N4-P4-C44	107.0(3)	P4-N4-Pr1	98.7(2)
N3-P3-C44	105.5(3)	N1-P1-C1	105.6(3)
P3-N3-Pr1	99.6(2)	P1-N1-Pr1	99.6(2)
	7d•4C_7H_8		
Gd1-C1	2.605(11)	P1-N1	1.621(9)
Gd1-N1	2.494(8)	P2-N2	1.622(9)
Gd1-N2	2.451(8)	P1-C1	1.703(10)
Gd1-N3	2.487(9)	P4-C44	1.685(11)
Gd1-N4	2.431(8)	P4-N4	1.684(9)
Gd1-C44	2.573(10)	P3-C44	1.724(12)
P3-N3	1.633(9)	P2-C1	1.698(10)
P2-N2-Gd1	98.0(4)	P3-C44-P4	131.9(6)
N2-P2-C1	107.4(5)	N4-P4-C44	104.7(5)
N1-Gd1-N2	108.9(3)	N1-P1-C1	106.6(5)
P1-C1-P2	132.5(7)	P1-N1-Gd1	97.9(4)
N3-Gd1-N4	109.2(3)	N3-P3-C44	104.9(5)
P4-N4-Gd1	97.7(4)	P3-N3-Gd1	98.0(4)

Table S2. Crystallographic data for **1**-**7**.

	1a•C_7H_8	**1b•OC_4H_8**	**1c•OC_4H_8**	**2•2C_7H_8**	**3•3C_7H_8**
Formula	$C_{42}H_{55}I_2N_2NdOP_2Si_2$	$C_{39}H_{55}GdI_2N_2O_2P_2Si_2$	$C_{39}H_{55}I_2N_2O_2P_2Si_2Tb$	$C_{76}H_{94}I_4La_2N_4P_4Si_4$	$C_{72}H_{83}GdI_2N_2O_2P_2$
Fw	1120.04	1113.02	1114.69	2085.21	1481.39
cryst size, mm	0.06 x 0.04 x 0.02	0.13 x 0.11 x 0.05	0.57 x 0.33 x 0.18	0.24 x 0.16 x 0.02	0.15 x 0.09 x 0.09
cryst syst	Triclinic	Monoclinic	Monoclinic	Monoclinic	Triclinic
space group	*P*-1	$P2_1/c$	$P2_1/c$	*C*2/c	*P*-1
a, Å	9.839(2)	16.1695(8)	16.142(2)	36.332(2)	12.7851(6)
b, Å	12.426(3)	19.5199(12)	19.415(3)	12.8545(4)	15.1220(8)
c, Å	19.947(5)	14.1982(8)	14.134(2)	21.6820(13)	17.2635(9)
α, °	106.347(5)				85.091(2)
β, °	96.476(4)	99.491(5)	99.341(2)	122.332(9)	76.608(2)
γ, °	98.785(4)				87.266(2)
V, Å^3	2281.3(9)	4420.0(4)	4370.8(11)	8556.3(11)	3233.7(3)
Z	2	4	4	4	2
ρ_{calcd}, g cm^{-3}	1.631	1.673	1.694	1.619	1.521
μ, mm^{-1}	2.647	22.135	3.193	20.494	2.076
no. of reflections measd	20251	28684	26058	25755	40617
no. of unique reflns, R_{int}	10266, 0.0267	8830, 0.1240	9666, 0.0306	8532, 0.076	14563, 0.034
no. of reflns with $F^2 > 2\sigma(F^2)$	9590	6154	8589	6688	12925
transmn coeff range	0.33-0.43	0.33-0.65	0.30-0.43	0.09-0.85	0.35 - 0.43
R, R_w[a] ($F^2 > 2\sigma(F^2)$)	0.0461, 0.0898	0.0589, 0.1300	0.0420, 0.1101	0.0570, 0.0146	0.0419, 0.109
R, R_w[a] (all data)	0.0505, 0.0915	0.0951, 0.1518	0.0468, 0.1144	0.0771, 0.158	0.0475, 0.114
S[a]	1.305	1.023	1.038	1.03	1.14
Parameters	476	472	472	426	663
max., min. diff map, e Å^{-3}	1.50, -0.81	1.95, -1.59	3.34, -3.64	2.27, -1.51	2.25, -1.26

Table S2. *Cont.*

	4•3C$_7$H$_8$	**5a•4C$_7$H$_8$**	**5b•5C$_7$H$_8$**	**6a•3C$_7$H$_8$**	**6b•2C$_7$H$_8$**
Formula	$C_{68}H_{75}I_2N_2OP_2Yb$	$C_{157}H_{161}DyN_6P_6$	$C_{164}H_{169}ErN_6P_6$	$C_{70}H_{83}I_2LaN_4P_2$	$C_{63}H_{75}GdI_2N_4P_2$
Fw	1425.08	2480.24	2577.13	1435.05	1361.26
cryst size, mm	0.21 x 0.08 x0.03	0.16 x 0.12 x 0.11	0.13 x 0.05 x 0.04	0.31 x 0.16 x 0.09	0.22 x 0.22 x 0.09
cryst syst	Triclinic	Triclinic	Triclinic	Triclinic	Orthorhombic
space group	*P*-1	*P*-1	*P*-1	*P*-1	*C*mca
a, Å	12.209(3)	17.2957(11)	16.895(3)	13.0112(11)	26.770(2)
b, Å	13.057(3)	18.9502(15)	19.820(4)	15.2310(11)	18.9186(16)
c, Å	17.560(5)	21.6886(15)	21.149(4)	17.0292(12)	22.445(2)
α, °	80.744(4)	75.525(6)	76.79(3)	82.761(6)	
β, °	75.234(4)	74.875(6)	78.76(3)	74.997(7)	
γ, °	77.456(4)	73.945(6)	83.90(3)	87.043(7)	
V, Å^3	2625.6(12)	6472.4(8)	6748(2)	3233.1(4)	11367.4(17)
Z	2	2	2	2	8
ρ_{calcd}, g cm^{-3}	1.803	1.273	1.268	1.474	1.591
μ, mm^{-1}	3.068	4.210	0.749	13.393	2.353
no. of reflections measd	22426	55800	50018	25965	34384
no. of unique reflns, R_{int}	11730, 0.0744	25787, 0.048	23655, 0.0490	12864, 0.058	6604, 0.0458
no. of reflns with $F^2 > 2\sigma(F^2)$	7643	22999	19011	10928	5170
transmn coeff range	0.75-0.43	1.00-0.37	0.75-0.61	1.00-0.19	0.43-0.35
R, R_w[a] ($F^2 > 2\sigma(F^2)$)	0.1097, 0.2865	0.0387, 0.0896	0.0577, 0.1270	0.058, 0.153	0.0314, 0.0687
R, R_w[a] (all data)	0.1439, 0.3027	0.0461, 0.0939	0.0761, 0.1340	0.067, 0.164	0.0483, 0.0758
S[a]	1.07	1.02	1.11	1.04	1.03
Parameters	502	1553	1617	725	413
max., min. diff map, e Å^{-3}	10.06, -5.31	0.68, -0.88	1.92, -1.48	2.65, -1.94	1.30. -0.63

Table S2. *Cont.*

	7a•3C_7H_8	**7b**•2C_6H_{14}	**7c**•3C_7H_8	**7d**•4C_7H_8
Formula	$C_{107}H_{109}LaN_4P_4$	$C_{98}H_{113}CeN_4P_4$	$C_{107}H_{109}N_4P_4Pr$	$C_{114}H_{117}GdN_4P_4$
Fw	1713.77	1610.92	1715.77	1825.25
cryst size, mm	0.46 x 0.28 x 0.10	0.12 x 0.08 x 0.08	0.28 x 0.21 x 0.05	0.30 x 0.21 x 0.01
cryst syst	Triclinic	Triclinic	Triclinic	Triclinic
space group	*P*-1	*P*-1	*P*-1	*P*-1
a, Å	14.9196(11)	13.6367(15)	14.8733(2)	14.7350(8)
b, Å	15.179(3)	21.346(2)	15.2027(2)	15.2646(10)
c, Å	21.622(2)	29.100(3)	21.6410(3)	21.6185(13)
α, °	71.398(13)	97.388(2)	71.3195(14)	70.916(6)
β, °	75.826(8)	92.382(2)	75.5504(12)	75.272(5)
γ, °	77.368(10)	97.146(2)	77.6266(14)	77.458(5)
V, Å^3	4446.5(10)	8320.8(16)	4440.53(11)	4396.2(4)
Z	2	4	2	2
ρ_{calcd}, g cm^{-3}	1.283	1.286	1.283	1.378
μ, mm^{-1}	4.777	0.674	5.274	5.984
no. of reflections measd	37469	61396	93160	35551
no. of unique reflns, R_{int}	17807, 0.0560	29171, 0.0610	16002, 0.0610	15834, 0.166
no. of reflns with $F^2 > 2\sigma(F^2)$	15935	20928	15114	9354
transmn coeff range	0.71-0.29	0.75-0.60	0.55-0.09	1.18-0.29
R, R_w[a] ($F^2 > 2\sigma(F^2)$)	0.0599, 0.1530	0.1220, 0.2740	0.0676, 0.1780	0.1208, 0.2985
R, R_w[a] (all data)	0.0655, 0.1570	0.1490, 0.2850	0.0708, 0.1820	0.1632, 0.3309
S[a]	1.06	1.10	1.05	1.02
Parameters	972	1710	933	932
max., min. diff map, e Å^{-3}	1.89, -1.58	2.03, -2.83	4.44, -1.23	4.34, -2.77

[a] Conventional $R = \Sigma||F_o| - |F_c||/\Sigma|F_o|$; $R_w = [\Sigma w(F_o^2 - F_c^2)^2/\Sigma w(F_o^2)^2]^{1/2}$; $S = [\Sigma w(F_o^2 - F_c^2)^2/\text{no. data} - \text{no. params})]^{1/2}$ for all data.