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Table S1. Crystallographic data for *trans*-[Cu(cyan-κ*N*)$_2$(H$_2$O)$_2$]·2Na(cyan)·4H$_2$O, **1**; *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**; *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κ*O*^{2})$_2$(NH$_3$)$_4$], **6**; *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**; [Ni(cyan-κ*N*)$_2$(NH$_3$)$_2$(H$_2$O)$_2$], **8**; and [Cu(cyan-κ*N*)(PPh$_3$)$_2$]·2CDCl$_3$, **9**.[a]

	1	**5**	**6**	**7**	**8**	9
formula	$CuC_{12}H_{20}N_{12}O_{18}Na_2$	$CuC_6H_{10}N_8O_6$	$Cu_2C_{12}H_{26}N_{18}O_{12}$	$NiC_6H_{16}N_{10}O_6$	$NiC_6H_{14}N_8O_8$	$CuC_{41}H_{32}N_3P_2O_3Cl_6D_2$
color	blue	dark blue	dark blue	violet	blue-violet	colorless
crystal dimensions (mm)	0.31×0.16×0.12	0.2×0.2×0.06	0.28×0.17×0.14	0.38×0.30×0.15	0.4×0.18×0.14	0.38×0.35×0.20
fw	729.88	353.76	741.59	382.71	384.54	956.89
temp (°C)	26(1)	24(1)	26(1)	25(2)	25(1)	25(1)
radiation	MoKα ($\lambda\bar{\alpha}$ = 0.71073 Å)					
crystal system	triclinic	triclinic	triclinic	orthorhombic	monoclinic	triclinic
space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$	Cmcm	$P2_1/m$	$P\bar{1}$
a (Å)	6.7552(8)	5.031(1)	7.143(1)	12.050(2)	6.334(1)	11.645(1)
b(Å)	10.455(1)	6.989(1)	8.683(2)	7.266(2)	16.022(2)	13.440(2)

c(Å)	10.445(1)	9.120(2)	11.602(2)	15.971(2)	7.017(1)	16.561(2)
α(°)	62.819(6)	90.04(3)	102.12(3)	90	90	102.051(8)
β(°)	71.815(6)	98.01(3)	101.24(3)	90	114.13(2)	97.585(9)
γ(°)	83.688(5)	110.84(3)	106.22(3)	90	90	113.132(9)
V(Å^3)	623.0(1)	296.3(3)	650.44(3)	1398.3(2)	650.0(2)	2264.2(4)
Z	1	1	1	4	2	2
ρ_{calcd}, g/cm^3	1.945	1.982	1.893	1.819	1.967	1.401
μ, cm^{-1}	10.24	18.92	17.31	14.42	15.59	9.47
transmission$_{max,min}$	0.915, 0.867	0.998, 0.778	0.983, 0.919	1.00, 0.77	0.941, 0.582	1.000, 0.846
2θ range (°)	4.0 - 48.0	4.0 - 50.0	4.0 - 50.0	4.0 - 50.0	4.0 - 50.0	4.0 - 45.0
scan method	ω-2θ	ω	ω-2θ	ω-2θ	ω	ω-2θ
No. unique data	1952	1033	[d]2459	676	1186	5894
cutoff	$F_o^2 \geq -3\sigma(F_o^2)$	$F_o^2 \geq -3\sigma(F_o^2)$	$F_o^2 \geq 0$	$F_o^2 \geq 0$	$F_o^2 \geq 0$	$F_o^2 \geq 0$

No. data in refinement	1868	1022	2337	666	1150	4526
No. params.	245	117	202	80	135	549
[a]q.o.f	1.113	1.180	1.121	1.158	1.029	1.078
[b]R1, [c]wR2	0.0319, 0.0779	0.0265, 0.0730	0.0813, 0.3060	0.0265, 0.0750	0.0410, 0.1056	0.0685, 0.1227

[a]quality of fit = $[\sum w(F_{obs}^2 - F_{calc}^2)^2 / \sum (N_{obs} - N_{params})]^{1/2}$

[b]R1 = $\sum ||F_{obs}| - |F_{calc}|| / \sum |F_{obs}|$

[c]wR2 = $[\sum w(F_{obs}^2 - F_{calc}^2)^2 / \sum w(F_{obs}^2)^2]^{1/2}$

[d]Laue-equivalent data were not averaged for **6**, because of twinning.

Table S2. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and their estimated standard deviations for *trans*-[Cu(cyan-κ*N*)$_2$(H$_2$O)$_2$]-·2Na(cyan)·4H$_2$O, **1**.[a]

	x	y	z	U(eq)
Cu(1)	0	0	5000	17(1)
O(1)	2543(4)	-998(3)	5209(3)	39(1)
N(1)	115(4)	670(2)	6438(3)	17(1)
C(2)	984(4)	2007(3)	5866(3)	18(1)
O(2)	2039(3)	2633(2)	4537(2)	26(1)
N(3)	603(4)	2636(3)	6810(3)	19(1)
C(4)	-551(4)	2001(3)	8293(3)	18(1)
O(4)	-885(3)	2598(2)	9103(2)	28(1)
N(5)	-1313(4)	650(3)	8784(3)	19(1)
C(6)	-1039(4)	-31(3)	7893(3)	17(1)
O(6)	-1827(3)	-1243(2)	8406(2)	26(1)
N(7)	5912(4)	2575(2)	6230(3)	18(1)
C(8)	6728(4)	3914(3)	5674(3)	18(1)
O(8)	7841(3)	4566(2)	4355(2)	26(1)
N(9)	6341(4)	4562(3)	6610(3)	21(1)
C(10)	5134(4)	3968(3)	8064(3)	19(1)
O(10)	4828(3)	4563(2)	8881(2)	28(1)
N(11)	4317(4)	2639(3)	8551(3)	23(1)
C(12)	4704(4)	1913(3)	7681(3)	18(1)
O(12)	3929(3)	681(2)	8253(2)	24(1)
Na(1)	-2407(2)	4015(1)	10297(1)	29(1)
O(13)	-25(4)	-6030(3)	11689(3)	35(1)
O(14)	4262(4)	-1787(3)	7545(3)	34(1)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S3. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(H$_2$O)$_2$]·2Na(cyan)·4H$_2$O, **1**.

Cu(1)-O(1)	1.929(3)
Cu(1)-N(1)	1.949(2)
O(1)-H(1)	0.68(4)
O(1)-H(2)	0.95(5)
N(1)-C(6)	1.350(4)
N(1)-C(2)	1.362(4)
C(2)-O(2)	1.232(3)
C(2)-N(3)	1.367(4)
N(3)-C(4)	1.366(4)
N(3)-H(3)	0.69(3)
C(4)-O(4)	1.221(3)
C(4)-N(5)	1.362(4)
O(4)-Na(1)	2.316(2)
N(5)-C(6)	1.370(4)
N(5)-H(5)	0.74(3)
C(6)-O(6)	1.236(3)
N(7)-C(8)	1.352(4)
N(7)-C(12)	1.354(4)
C(8)-O(8)	1.239(3)
C(8)-N(9)	1.375(4)
N(9)-C(10)	1.354(4)
N(9)-H(9)	0.71(3)
C(10)-O(10)	1.227(3)
C(10)-N(11)	1.357(4)
N(11)-C(12)	1.382(4)
N(11)-H(11)	0.73(3)
C(12)-O(12)	1.241(3)
O(13)-H(13)	0.78(5)
O(13)-H(14)	0.80(5)
O(14)-H(15)	0.75(5)
O(14)-H(16)	0.84(5)

Table S4. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(H$_2$O)$_2$]·2Na(cyan)·4H$_2$O, **1**.

O(1)-Cu(1)-N(1)	89.71(10)	O(6)-C(6)-N(5)	121.0(3)
Cu(1)-O(1)-H(1)	123(3)	N(1)-C(6)-N(5)	117.7(2)
Cu(1)-O(1)-H(2)	122(3)	C(8)-N(7)-C(12)	119.7(2)
H(1)-O(1)-H(2)	114(4)	O(8)-C(8)-N(7)	122.1(2)
C(6)-N(1)-C(2)	121.6(2)	O(8)-C(8)-N(9)	119.0(3)
C(6)-N(1)-Cu(1)	119.6(2)	N(7)-C(8)-N(9)	118.9(3)
C(2)-N(1)-Cu(1)	117.2(2)	C(10)-N(9)-C(8)	124.5(3)
O(2)-C(2)-N(1)	120.9(2)	C(10)-N(9)-H(9)	117(3)
O(2)-C(2)-N(3)	121.5(3)	C(8)-N(9)-H(9)	119(3)
N(1)-C(2)-N(3)	117.5(3)	O(10)-C(10)-N(9)	123.3(3)
C(4)-N(3)-C(2)	124.4(3)	O(10)-C(10)-N(11)	122.7(3)
C(4)-N(3)-H(3)	120(3)	N(9)-C(10)-N(11)	114.0(3)
C(2)-N(3)-H(3)	115(3)	C(10)-N(11)-C(12)	124.2(3)
O(4)-C(4)-N(5)	122.9(3)	C(10)-N(11)-H(11)	119(3)
O(4)-C(4)-N(3)	122.9(3)	C(12)-N(11)-H(11)	117(3)
N(5)-C(4)-N(3)	114.2(2)	O(12)-C(12)-N(7)	122.4(2)
C(4)-O(4)-Na(1)	161.2(2)	O(12)-C(12)-N(11)	118.9(3)
C(4)-N(5)-C(6)	124.5(3)	N(7)-C(12)-N(11)	118.7(3)
C(4)-N(5)-H(5)	123(2)	H(13)-O(13)-H(14)	109(5)
C(6)-N(5)-H(5)	113(2)	H(15)-O(14)-H(16)	107(5)
O(6)-C(6)-N(1)	121.3(2)		

Table S5. Anisotropic displacement parameters ($Å^2$ x 10^3) for *trans*-[Cu(cyan-$\kappa N)_2(H_2O)_2$]·2Na(cyan)·$4H_2O$, **1**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Cu(1)	22(1)	16(1)	14(1)	-9(1)	-3(1)	-1(1)
O(1)	46(2)	57(2)	44(2)	-43(2)	-30(1)	30(1)
N(1)	21(1)	17(1)	15(1)	-9(1)	-3(1)	-2(1)
C(2)	19(2)	18(2)	19(2)	-10(1)	-7(1)	2(1)
O(2)	35(1)	22(1)	16(1)	-9(1)	4(1)	-7(1)
N(3)	24(2)	14(1)	19(1)	-8(1)	-2(1)	-6(1)
C(4)	17(2)	20(2)	21(2)	-11(1)	-8(1)	4(1)
O(4)	37(1)	29(1)	24(1)	-20(1)	-3(1)	-2(1)
N(5)	24(1)	19(1)	11(1)	-7(1)	0(1)	-4(1)
C(6)	17(2)	17(2)	18(2)	-9(1)	-6(1)	2(1)
O(6)	35(1)	18(1)	22(1)	-10(1)	-1(1)	-7(1)
N(7)	21(1)	17(1)	16(1)	-10(1)	-2(1)	-2(1)
C(8)	17(2)	19(2)	19(2)	-10(1)	-7(1)	3(1)
O(8)	35(1)	22(1)	15(1)	-7(1)	3(1)	-8(1)
N(9)	27(2)	16(1)	21(1)	-10(1)	-2(1)	-7(1)
C(10)	16(2)	20(2)	23(2)	-12(1)	-4(1)	2(1)
O(10)	34(1)	29(1)	25(1)	-20(1)	-3(1)	-2(1)
N(11)	27(2)	23(1)	15(1)	-12(1)	5(1)	-6(1)
C(12)	19(2)	18(2)	20(2)	-9(1)	-8(1)	2(1)
O(12)	30(1)	20(1)	21(1)	-10(1)	1(1)	-7(1)
Na(1)	34(1)	29(1)	29(1)	-20(1)	-5(1)	1(1)
O(13)	44(2)	31(2)	27(1)	-10(1)	-9(1)	-1(1)
O(14)	34(2)	36(2)	35(1)	-18(1)	-9(1)	-3(1)

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S6. Hydrogen Atom Coordinates ($\times 10^4$) and Isotropic Displacement Parameters ($Å^2 \times 10^3$) for *trans*-[Cu(cyan-κ*N*)$_2$(H$_2$O)$_2$]·2Na(cyan)·4H$_2$O, **1**.

	x	y	z	U(eq)
H(1)	2909(55)	-1155(38)	5804(43)	24(11)
H(2)	3161(69)	-1525(49)	4659(52)	74(14)
H(3)	1115(50)	3292(36)	6505(38)	19(10)
H(5)	-1967(47)	219(32)	9561(36)	12(9)
H(9)	6849(49)	5239(35)	6335(36)	16(9)
H(11)	3629(50)	2275(34)	9322(38)	18(9)
H(13)	476(82)	-6774(58)	11882(59)	85(20)
H(14)	-558(72)	-5965(49)	12453(57)	65(16)
H(15)	5367(76)	-1958(50)	7254(55)	66(18)
H(16)	4297(69)	-1123(51)	7775(52)	70(16)

Table S7. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and their estimated standard deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**.[a]

	x	y	z	U(eq)
Cu(1)	0	0	0	20(1)
N(1)	260(5)	1279(4)	-1972(2)	22(1)
C(2)	-1953(6)	729(4)	-3113(3)	21(1)
O(2)	-4340(4)	-540(3)	-3010(2)	31(1)
N(3)	-1457(5)	1630(4)	-4447(3)	23(1)
C(4)	1102(6)	3043(4)	-4684(3)	22(1)
O(4)	1445(4)	3770(3)	-5898(2)	29(1)
N(5)	3203(5)	3533(4)	-3494(3)	26(1)
C(6)	2858(6)	2696(4)	-2128(3)	23(1)
O(6)	4877(4)	3227(3)	-1109(2)	33(1)
N(6)	453(7)	-2474(5)	-853(3)	33(1)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S8. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**.

Cu(1)-N(6)	1.994(3)
Cu(1)-N(1)	2.009(2)
N(1)-C(6)	1.357(4)
N(1)-C(2)	1.357(4)
C(2)-O(2)	1.229(3)
C(2)-N(3)	1.383(3)
N(3)-C(4)	1.361(4)
N(3)-H(3)	0.84(4)
C(4)-O(4)	1.228(3)
C(4)-N(5)	1.354(4)
N(5)-C(6)	1.383(4)
N(5)-H(5)	0.88(4)
C(6)-O(6)	1.225(4)
N(6)-H(61)	0.84(4)
N(6)-H(62)	0.75(5)
N(6)-H(63)	0.94(5)

Table S9. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**.

N(6)-Cu(1)-N(6)#1	180.0
N(1)-Cu(1)-N(1)#1	180.0
N(6)-Cu(1)-N(1)	90.56(11)
C(6)-N(1)-C(2)	121.0(2)
C(6)-N(1)-Cu(1)	115.7(2)
C(2)-N(1)-Cu(1)	123.2(2)
O(2)-C(2)-N(1)	122.8(2)
O(2)-C(2)-N(3)	119.3(3)
N(1)-C(2)-N(3)	117.8(2)
C(4)-N(3)-C(2)	124.1(2)
C(4)-N(3)-H(3)	115(2)
C(2)-N(3)-H(3)	121(2)
O(4)-C(4)-N(5)	123.6(3)
O(4)-C(4)-N(3)	121.6(3)
N(5)-C(4)-N(3)	114.8(2)
C(4)-N(5)-C(6)	124.2(2)
C(4)-N(5)-H(5)	117(2)
C(6)-N(5)-H(5)	118(2)
O(6)-C(6)-N(1)	122.1(3)
O(6)-C(6)-N(5)	119.9(3)
N(1)-C(6)-N(5)	118.0(2)
Cu(1)-N(6)-H(61)	106(2)
Cu(1)-N(6)-H(62)	113(4)
H(61)-N(6)-H(62)	106(4)
Cu(1)-N(6)-H(63)	106(3)
H(61)-N(6)-H(63)	102(4)
H(62)-N(6)-H(63)	122(5)

Symmetry transformations used to generate equivalent atoms:
#1: -x,-y,-z

Table S10. Anisotropic displacement parameters ($Å^2$ x 10^3) for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Cu(1)	20(1)	24(1)	12(1)	6(1)	1(1)	4(1)
N(1)	21(1)	26(1)	15(1)	6(1)	2(1)	4(1)
C(2)	24(1)	24(1)	16(1)	5(1)	6(1)	7(1)
O(2)	21(1)	36(1)	23(1)	10(1)	3(1)	-4(1)
N(3)	18(1)	28(1)	14(1)	7(1)	-1(1)	0(1)
C(4)	20(1)	25(1)	18(1)	2(1)	4(1)	4(1)
O(4)	24(1)	39(1)	15(1)	10(1)	4(1)	0(1)
N(5)	17(1)	33(1)	19(1)	10(1)	2(1)	-1(1)
C(6)	24(1)	26(1)	17(1)	4(1)	3(1)	7(1)
O(6)	25(1)	44(1)	21(1)	7(1)	-5(1)	3(1)
N(6)	45(2)	38(2)	17(1)	6(1)	2(1)	18(1)

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S11. Hydrogen Atom Coordinates ($\times 10^4$) and Isotropic Displacement Parameters ($Å^2 \times 10^3$) for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$], **5**.

	x	y	z	U(eq)
H(3)	-2754(77)	1309(53)	-5192(41)	31(9)
H(5)	4906(80)	4422(54)	-3613(38)	34(9)
H(61)	1821(79)	-2636(52)	-293(41)	27(9)
H(62)	902(103)	-2332(72)	-1608(57)	64(15)
H(63)	-1134(116)	-3609(82)	-650(56)	79(16)

Table S12. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and their estimated standard deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κ*O*^{2})$_2$(NH$_3$)$_4$], **6**.[a]

	x	y	z	U(eq)
Cu(1)	0	-5000	0	16(1)
N(1)	1012(11)	-2503(8)	445(6)	19(2)
O(2)	892(12)	-2353(7)	2398(6)	36(2)
C(2)	1352(13)	-1626(11)	1632(8)	22(2)
Cu(2)	5000	5000	5000	22(1)
N(3)	2245(12)	87(8)	1951(6)	23(2)
C(4)	2803(14)	962(11)	1168(8)	24(2)
O(4)	3555(11)	2472(7)	1447(6)	29(2)
N(5)	2462(12)	8(9)	-2(7)	24(2)
O(6)	1405(13)	-2490(8)	-1436(5)	33(2)
C(6)	1641(15)	-1705(11)	-365(9)	25(2)
N(7)	2304(12)	-689(9)	6869(6)	25(2)
O(8)	3942(12)	1682(8)	8407(6)	34(2)
C(8)	3434(15)	999(11)	7276(8)	26(2)
N(9)	3870(13)	1878(9)	6484(7)	25(2)
O(10)	3539(12)	1759(8)	4469(6)	32(2)
C(10)	3183(15)	1049(10)	5266(8)	22(2)
N(11)	2086(13)	-654(9)	4875(7)	27(2)
O(12)	680(10)	-3073(7)	5312(5)	24(1)
C(12)	1596(13)	-1572(10)	5663(7)	19(2)
N(13)	-2839(12)	-4993(8)	-30(8)	26(2)
N(14)	6565(12)	4842(9)	3754(6)	24(2)
N(15)	7312(12)	5019(10)	6318(7)	27(2)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S13. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κ*O*^{2})$_2$(NH$_3$)$_4$], **6**.

Cu(1)-N(1)	2.000(7)
Cu(1)-N(13)	2.023(8)
N(1)-C(6)	1.353(12)
N(1)-C(2)	1.365(11)
O(2)-C(2)	1.237(10)
C(2)-N(3)	1.379(10)
N(3)-C(4)	1.350(11)
C(4)-O(4)	1.214(11)
C(4)-N(5)	1.369(12)
N(5)-C(6)	1.373(11)
O(6)-C(6)	1.240(11)
Cu(2)-N(14)	1.998(7)
Cu(2)-N(15)	2.014(8)
Cu(2)-O(10)	2.602(6)
N(7)-C(12)	1.363(11)
N(7)-C(8)	1.387(11)
O(8)-C(8)	1.255(12)
C(8)-N(9)	1.340(12)
N(9)-C(10)	1.364(11)
O(10)-C(10)	1.242(10)
C(10)-N(11)	1.393(11)
N(11)-C(12)	1.367(10)
O(12)-C(12)	1.221(10)

Table S14. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κ*O*^{2})$_2$(NH$_3$)$_4$], **6**.

N(1)-Cu(1)-N(1)#1	180.0
N(1)-Cu(1)-N(13)	90.3(3)
N(13)-Cu(1)-N(13)#1	180.0
C(6)-N(1)-C(2)	120.1(7)
C(6)-N(1)-Cu(1)	120.5(6)
C(2)-N(1)-Cu(1)	118.7(6)
O(2)-C(2)-N(1)	120.7(8)
O(2)-C(2)-N(3)	121.3(8)
N(1)-C(2)-N(3)	118.0(7)
C(4)-N(3)-C(2)	124.5(7)
O(4)-C(4)-N(3)	124.3(8)
O(4)-C(4)-N(5)	121.0(8)
N(3)-C(4)-N(5)	114.7(8)
C(4)-N(5)-C(6)	123.5(8)
O(6)-C(6)-N(1)	120.9(8)
O(6)-C(6)-N(5)	120.0(8)
N(1)-C(6)-N(5)	119.0(8)
N(14)-Cu(2)-N(14)#2	180.0
N(14)-Cu(2)-N(15)	91.8(3)
N(15)#2-Cu(2)-N(15)	180.0
N(14)-Cu(2)-O(10)	90.9(3)
N(15)-Cu(2)-O(10)	89.9(3)
C(12)-N(7)-C(8)	123.4(7)
O(8)-C(8)-N(9)	121.6(8)
O(8)-C(8)-N(7)	117.3(8)
N(9)-C(8)-N(7)	121.1(8)
C(8)-N(9)-C(10)	117.7(7)
C(10)-O(10)-Cu(2)	121.2(5)
O(10)-C(10)-N(9)	122.1(8)
O(10)-C(10)-N(11)	117.4(7)
N(9)-C(10)-N(11)	120.5(7)
C(12)-N(11)-C(10)	122.9(7)
O(12)-C(12)-N(7)	123.1(7)
O(12)-C(12)-N(11)	122.4(8)
N(7)-C(12)-N(11)	114.4(7)

Symmetry transformations used to generate equivalent atoms:
#1: -x,-y-1,-z #2: -x+1,-y+1,-z+1

Table S15. Anisotropic displacement parameters ($Å^2$ x 10^3) for *trans*-[Cu(cyan-κ*N*)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κ*O*^{2})$_2$(NH$_3$)$_4$], **6**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Cu(1)	19(1)	9(1)	18(1)	4(1)	4(1)	0(1)
N(1)	24(4)	13(3)	14(3)	2(3)	2(3)	0(3)
O(2)	61(5)	15(3)	20(3)	-2(2)	17(3)	-4(3)
C(2)	24(4)	20(4)	18(4)	8(3)	3(3)	1(3)
Cu(2)	25(1)	21(1)	16(1)	5(1)	3(1)	2(1)
N(3)	35(4)	10(3)	18(3)	2(3)	6(3)	0(3)
C(4)	25(4)	23(4)	21(4)	6(3)	8(4)	4(4)
O(4)	46(4)	11(3)	20(3)	2(2)	4(3)	-2(3)
N(5)	28(4)	15(3)	24(3)	9(3)	5(3)	-2(3)
O(6)	65(5)	14(3)	14(3)	0(2)	18(3)	2(3)
C(6)	31(5)	16(4)	30(5)	8(4)	11(4)	7(4)
N(7)	33(4)	15(3)	17(3)	2(3)	6(3)	-4(3)
O(8)	51(4)	20(3)	18(3)	4(3)	8(3)	-6(3)
C(8)	35(5)	16(4)	25(4)	4(3)	10(4)	3(4)
N(9)	34(4)	13(3)	20(4)	3(3)	8(3)	-3(3)
O(10)	55(5)	17(3)	19(3)	7(2)	10(3)	0(3)
C(10)	35(5)	15(4)	20(4)	6(3)	12(4)	8(4)
N(11)	36(4)	16(4)	19(4)	6(3)	7(3)	-5(3)
O(12)	34(3)	11(3)	23(3)	3(2)	4(3)	4(3)
C(12)	24(4)	17(4)	17(4)	8(3)	8(3)	4(3)
N(13)	30(4)	5(3)	37(4)	-2(3)	5(3)	3(3)
N(14)	31(4)	18(3)	13(3)	-3(3)	3(3)	0(3)
N(15)	25(4)	30(4)	20(4)	7(3)	0(3)	2(3)

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S16. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and their estimated standard deviations for *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**.[a]

	x	y	z	U(eq)
Ni(1)	5000	2010(1)	7500	22(1)
N(1)	5000	2045(3)	6069(1)	24(1)
C(2)	4028(2)	2191(3)	5642(1)	24(1)
O(2)	3107(1)	2043(2)	5978(1)	33(1)
N(3)	4053(1)	2516(2)	4784(1)	25(1)
C(4)	5000	2713(4)	4328(2)	23(1)
O(4)	5000	3029(3)	3576(1)	31(1)
N(4)	6187(2)	4076(4)	7500	30(1)
N(5)	6205(2)	-19(4)	7500	29(1)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S17. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**.

Ni(1)-N(5)	2.069(3)
Ni(1)-N(4)	2.073(3)
Ni(1)-N(1)	2.285(2)
N(1)-C(2)	1.360(2)
C(2)-O(2)	1.237(2)
C(2)-N(3)	1.391(3)
N(3)-C(4)	1.362(2)
N(3)-H(3)	0.90(3)
C(4)-O(4)	1.222(4)
N(4)-H(41)	0.77(5)
N(4)-H(42)	0.87(8)
N(5)-H(51)	0.86(3)
N(5)-H(52)	0.83(7)

Table S18. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**.

N(5)#1-Ni(1)-N(5)	89.2(2)
N(5)-Ni(1)-N(4)#1	179.02(12)
N(5)-Ni(1)-N(4)	91.81(11)
N(4)#1-Ni(1)-N(4)	87.2(2)
N(5)-Ni(1)-N(1)	90.46(4)
N(4)-Ni(1)-N(1)	89.53(4)
N(1)-Ni(1)-N(1)#2	178.70(12)
N(5)-Ni(1)-N(1)#3	90.46(4)
N(4)-Ni(1)-N(1)#3	89.53(4)
C(2)#4-N(1)-C(2)	119.0(2)
C(2)-N(1)-Ni(1)	120.15(12)
O(2)-C(2)-N(1)	123.3(2)
O(2)-C(2)-N(3)	117.5(2)
N(1)-C(2)-N(3)	119.2(2)
C(4)-N(3)-C(2)	124.3(2)
C(4)-N(3)-H(3)	120(2)
C(2)-N(3)-H(3)	116(2)
O(4)-C(4)-N(3)	123.07(12)
N(3)#4-C(4)-N(3)	113.9(2)
Ni(1)-N(4)-H(41)	117(4)
Ni(1)-N(4)-H(42)	116(5)
H(41)-N(4)-H(42)	97(4)
Ni(1)-N(5)-H(51)	108(2)
Ni(1)-N(5)-H(52)	114(4)
H(51)-N(5)-H(52)	113(3)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+3/2 #2 x,y,-z+3/2 #3 x,y,z #4 -x+1,y,z

Table S19. Anisotropic displacement parameters ($Å^2$ x 10^3) for *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Ni(1)	9(1)	34(1)	22(1)	0	0	0
N(1)	12(1)	42(1)	17(1)	0(1)	0	0
C(2)	18(1)	35(1)	20(1)	-2(1)	-1(1)	0(1)
O(2)	13(1)	64(1)	22(1)	3(1)	-1(1)	-1(1)
N(3)	16(1)	41(1)	18(1)	2(1)	-3(1)	0(1)
C(4)	20(1)	27(1)	23(1)	-1(1)	0	0
O(4)	29(1)	47(1)	17(1)	6(1)	0	0
N(4)	16(1)	43(1)	31(1)	0	0	-4(1)
N(5)	16(1)	42(1)	29(1)	0	0	3(1)

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S20. Hydrogen Atom Coordinates ($\times 10^4$) and Isotropic Displacement Parameters ($Å^2 \times 10^3$) for *trans*-[Ni(cyan-κ*N*)$_2$(NH$_3$)$_4$], **7**.

	x	y	z	U(eq)
H(3)	3391(25)	2622(31)	4533(18)	44(7)
H(41)	6223(39)	4676(75)	7102(28)	144(20)
H(42)	6875(62)	3707(115)	7500	135(26)
H(51)	6642(22)	185(44)	7915(17)	61(8)
H(52)	5951(49)	-1085(95)	7500	106(21)

Table S21. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) and their estimated standard deviations for [Ni(cyan-κ*N*)$_2$(NH$_3$)$_2$(H$_2$O)$_2$], **8**.[a]

	x	y	z	U(eq)
Ni(1)	358(1)	7500	1947(1)	21(1)
N(1)	141(4)	6100(2)	1907(4)	22(1)
C(2)	2091(5)	5661(2)	3059(5)	23(1)
O(2)	4051(4)	5963(1)	3679(4)	35(1)
N(3)	1898(4)	4835(2)	3526(4)	25(1)
C(4)	-119(5)	4409(2)	2885(5)	24(1)
O(4)	-264(4)	3686(2)	3383(4)	34(1)
N(5)	-1987(4)	4859(2)	1613(4)	25(1)
C(6)	-1904(5)	5687(2)	1141(5)	23(1)
O(6)	-3756(4)	6022(1)	-25(4)	31(1)
N(2)	2918(7)	7500	4923(7)	27(1)
N(4)	2608(7)	7500	514(7)	30(1)
O(1)	-2039(6)	7500	3438(6)	31(1)
O(3)	-2377(6)	7500	-950(5)	30(1)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S22. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for $[Ni(cyan\text{-}\kappa N)_2(NH_3)_2(H_2O)_2]$, **8**.

Ni(1)-N(2)	2.055(4)
Ni(1)-N(4)	2.053(4)
Ni(1)-O(3)	2.063(3)
Ni(1)-O(1)	2.168(3)
Ni(1)-N(1)	2.247(3)
N(1)-C(6)	1.354(4)
N(1)-C(2)	1.363(4)
C(2)-O(2)	1.234(4)
C(2)-N(3)	1.381(4)
N(3)-C(4)	1.352(4)
N(3)-H(3)	0.83(5)
C(4)-O(4)	1.225(4)
C(4)-N(5)	1.361(4)
N(5)-C(6)	1.373(4)
N(5)-H(5)	0.86(5)
C(6)-O(6)	1.245(4)
N(2)-H(21)	0.79(6)
N(2)-H(20)	0.83(9)
N(4)-H(40)	0.96(9)
N(4)-H(41)	0.84(5)
O(1)-H(10)	0.89(2)
O(3)-H(30)	0.88(5)

Table S23. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for $[Ni(cyan\text{-}\kappa N)_2(NH_3)_2(H_2O)_2]$, **8**.

N(2)-Ni(1)-N(4)	94.6(2)
N(2)-Ni(1)-O(3)	176.0(2)
N(4)-Ni(1)-O(3)	89.4(2)
N(2)-Ni(1)-O(1)	85.8(2)
N(4)-Ni(1)-O(1)	179.6(2)
O(3)-Ni(1)-O(1)	90.22(14)
N(2)-Ni(1)-N(1)	91.82(6)
N(4)-Ni(1)-N(1)	92.58(6)
O(3)-Ni(1)-N(1)	88.00(6)
O(1)-Ni(1)-N(1)	87.40(6)
N(1)-Ni(1)-N(1)#1	173.43(12)
C(6)-N(1)-C(2)	118.1(3)
C(6)-N(1)-Ni(1)	122.4(2)
C(2)-N(1)-Ni(1)	118.1(2)
O(2)-C(2)-N(1)	122.9(3)
O(2)-C(2)-N(3)	117.8(3)
N(1)-C(2)-N(3)	119.2(3)
C(4)-N(3)-C(2)	124.6(3)
C(4)-N(3)-H(3)	119(4)
C(2)-N(3)-H(3)	117(4)
O(4)-C(4)-N(3)	123.6(3)
O(4)-C(4)-N(5)	122.8(3)
N(3)-C(4)-N(5)	113.6(3)
C(4)-N(5)-C(6)	124.1(3)
C(4)-N(5)-H(5)	118(4)
C(6)-N(5)-H(5)	118(4)
O(6)-C(6)-N(1)	122.8(3)
O(6)-C(6)-N(5)	117.1(3)
N(1)-C(6)-N(5)	120.0(3)
Ni(1)-N(2)-H(21)	113(4)
Ni(1)-N(2)-H(20)	116(6)
H(21)-N(2)-H(20)	95(5)
Ni(1)-N(4)-H(40)	111(5)
Ni(1)-N(4)-H(41)	115(4)
H(40)-N(4)-H(41)	108(5)
Ni(1)-O(1)-H(10)	105(3)
Ni(1)-O(3)-H(30)	106(4)

Symmetry transformations used to generate equivalent atoms:
#1: x,-y+3/2,z

Table S24. Anisotropic displacement parameters (Å^2 x 10^3) for [Ni(cyan-κ*N*)$_2$(NH$_3$)$_2$(H$_2$O)$_2$], **8**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Ni(1)	15(1)	22(1)	20(1)	0	0(1)	0
N(1)	12(1)	21(1)	25(1)	0(1)	0(1)	1(1)
C(2)	18(1)	23(2)	25(2)	-1(1)	4(1)	3(1)
O(2)	14(1)	25(1)	52(2)	7(1)	1(1)	1(1)
N(3)	20(1)	21(1)	29(1)	4(1)	3(1)	2(1)
C(4)	21(2)	28(2)	20(2)	-2(1)	4(1)	0(1)
O(4)	33(1)	22(1)	38(1)	8(1)	7(1)	-2(1)
N(5)	20(1)	19(1)	30(2)	1(1)	4(1)	-3(1)
C(6)	18(1)	25(2)	23(2)	-3(1)	6(1)	0(1)
O(6)	16(1)	26(1)	38(1)	4(1)	-2(1)	0(1)
N(2)	20(2)	24(2)	26(2)	0	-2(2)	0
N(4)	27(2)	28(2)	34(2)	0	12(2)	0
O(1)	27(2)	31(2)	31(2)	0	8(1)	0
O(3)	24(2)	25(2)	29(2)	0	-2(1)	0

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S25. Hydrogen Atom Coordinates ($\times 10^4$) and Isotropic Displacement Parameters ($Å^2 \times 10^3$) for $[Ni(cyan\text{-}\kappa N)_2(NH_3)_2(H_2O)_2]$, **8**.

	x	y	z	U(eq)
H(3)	3115(89)	4584(38)	4215(87)	77(7)
H(5)	-3308(90)	4615(37)	1093(85)	77(7)
H(21)	3031(90)	7068(36)	5499(97)	77(7)
H(20)	4273(147)	7500	5024(136)	77(7)
H(40)	1793(132)	7500	-982(145)	77(7)
H(41)	3548(87)	7101(34)	847(90)	77(7)
H(10)	-1557(83)	7910(24)	4367(68)	77(7)
H(30)	-3222(84)	7060(34)	-972(87)	77(7)

Table S26. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2$ $\times 10^3$) and their estimated standard deviations for [Cu(cyan-κ*N*)(PPh_3)$_2$], **9**.[a]

	x	y	z	U(eq)
Cu(1)	8901(1)	1660(1)	7252(1)	49(1)
N(1)	8033(6)	698(5)	6076(3)	42(2)
C(1)	6822(8)	445(6)	5681(5)	47(2)
O(1)	6112(5)	755(4)	6033(3)	59(2)
N(2)	6396(6)	-188(5)	4836(4)	50(2)
C(2)	7098(8)	-552(7)	4356(5)	52(2)
O(2)	6682(5)	-1126(5)	3614(3)	64(2)
C(3)	8790(9)	384(6)	5648(5)	50(2)
O(3)	9921(6)	671(5)	6005(3)	71(2)
N(3)	8314(6)	-212(6)	4806(4)	53(2)
P(1)	9285(2)	900(2)	8269(1)	51(1)
C(4)	8775(9)	-612(7)	7982(5)	59(2)
C(5)	7575(10)	-1302(8)	7456(6)	95(3)
C(6)	7115(12)	-2460(10)	7264(7)	126(5)
C(7)	7838(15)	-2948(10)	7568(9)	123(5)
C(8)	9035(13)	-2274(10)	8076(7)	109(4)
C(9)	9499(9)	-1115(8)	8286(6)	80(3)
C(10)	8546(7)	1147(7)	9141(4)	51(2)
C(11)	8033(8)	357(8)	9576(5)	69(3)
C(12)	7416(9)	578(9)	10205(6)	84(3)
C(13)	7308(10)	1566(10)	10402(6)	84(3)
C(14)	7799(10)	2352(8)	9990(6)	85(3)
C(15)	8429(8)	2143(7)	9364(5)	68(3)
C(16)	10981(7)	1476(6)	8743(5)	50(2)
C(17)	11532(9)	1948(7)	9596(5)	63(2)
C(18)	12854(10)	2396(7)	9906(6)	78(3)
C(19)	13627(9)	2374(8)	9349(7)	85(3)
C(20)	13097(10)	1907(8)	8488(7)	81(3)
C(21)	11783(9)	1467(7)	8191(5)	67(3)
P(2)	9393(2)	3474(2)	7344(1)	56(1)
C(22)	9018(8)	3691(8)	6322(5)	68(3)
C(23)	9379(9)	3173(7)	5650(6)	81(3)
C(24)	9118(13)	3311(11)	4856(7)	128(6)
C(25)	8482(14)	3929(16)	4714(11)	154(9)
C(26)	8120(12)	4430(13)	5350(10)	136(6)
C(27)	8374(10)	4311(9)	6165(7)	101(4)
C(28)	11054(8)	4511(7)	7792(5)	56(2)
C(29)	11855(10)	4288(8)	8336(6)	81(3)
C(30)	13129(11)	5039(10)	8662(7)	106(4)
C(31)	13628(10)	6010(10)	8455(7)	98(4)
C(32)	12847(12)	6256(8)	7916(6)	89(3)
C(33)	11587(10)	5526(8)	7591(5)	74(3)
C(34)	8463(9)	3988(8)	7952(5)	60(2)
C(35)	8994(10)	4894(8)	8677(6)	85(3)
C(36)	8234(15)	5215(10)	9137(8)	116(4)
C(37)	6960(17)	4639(12)	8886(10)	122(5)
C(38)	6371(11)	3734(12)	8162(10)	119(4)
C(39)	7151(11)	3402(8)	7710(6)	86(3)
C(99)	5918(11)	554(11)	2770(7)	94(4)
Cl(1)	7500(3)	1080(3)	2656(2)	120(1)
Cl(2)	5689(3)	1584(3)	3445(2)	129(1)

Cl(3)	4874(3)	12(3)	1795(2)	159(2)
C(100)	13543(19)	3228(22)	5897(16)	238(25)
Cl(4)	12443(9)	2841(7)	4902(6)	163(4)
Cl(5)	15040(10)	3553(13)	5621(8)	229(8)
Cl(6)	13629(14)	4567(9)	6423(5)	209(6)
C(10A)	13857(18)	3509(21)	5861(14)	59(9)
Cl(4A)	13865(36)	2964(16)	4961(15)	354(21)
Cl(5A)	12572(18)	3436(21)	6063(20)	342(16)
Cl(6A)	14991(18)	4732(12)	6299(16)	371(15)

[a] The equivalent isotropic displacement parameter, U_{eq}, is calculated as:

$$U_{eq} = \frac{1}{3} \sum_{ij} U_{ij} \boldsymbol{a}_i \cdot \boldsymbol{a}_j a^*_i a^*_j$$

Table S27. Full Listing of Bond Distances (Å) and Their Estimated Standard Deviations for [Cu(cyan-κ*N*)(PPh_3)$_2$], **9**.

Cu(1)-N(1)	1.971(5)	C(23)-C(24)	1.377(13)
Cu(1)-P(1)	2.226(2)	C(23)-H(23)	.93
Cu(1)-P(2)	2.241(2)	C(24)-C(25)	1.35(2)
N(1)-C(3)	1.338(8)	C(24)-H(24)	.93
N(1)-C(1)	1.349(8)	C(25)-C(26)	1.34(2)
C(1)-O(1)	1.222(8)	C(25)-H(25)	.93
C(1)-N(2)	1.384(8)	C(26)-C(27)	1.398(14)
N(2)-C(2)	1.364(9)	C(26)-H(26)	.93
N(2)-H(2)	.856(11)	C(27)-H(27)	.93
C(2)-O(2)	1.221(8)	C(28)-C(29)	1.372(10)
C(2)-N(3)	1.358(9)	C(28)-C(33)	1.391(10)
C(3)-O(3)	1.239(8)	C(29)-C(30)	1.376(12)
C(3)-N(3)	1.368(9)	C(29)-H(29)	.93
N(3)-H(3)	.855(10)	C(30)-C(31)	1.343(12)
P(1)-C(16)	1.803(8)	C(30)-H(30)	.93
P(1)-C(4)	1.812(8)	C(31)-C(32)	1.370(13)
P(1)-C(10)	1.814(8)	C(31)-H(31)	.93
C(4)-C(5)	1.377(11)	C(32)-C(33)	1.359(11)
C(4)-C(9)	1.377(10)	C(32)-H(32)	.93
C(5)-C(6)	1.381(12)	C(33)-H(33)	.93
C(5)-H(5)	.93	C(34)-C(39)	1.370(11)
C(6)-C(7)	1.359(14)	C(34)-C(35)	1.381(11)
C(6)-H(6)	.93	C(35)-C(36)	1.374(13)
C(7)-C(8)	1.361(14)	C(35)-H(35)	.93
C(7)-H(7)	.93	C(36)-C(37)	1.33(2)
C(8)-C(9)	1.380(12)	C(36)-H(36)	.93
C(8)-H(8)	.93	C(37)-C(38)	1.381(14)
C(9)-H(9)	.93	C(37)-H(37)	.93
C(10)-C(15)	1.376(10)	C(38)-C(39)	1.393(13)
C(10)-C(11)	1.392(10)	C(38)-H(38)	.93
C(11)-C(12)	1.387(11)	C(39)-H(39)	.93
C(11)-H(11)	.93	C(99)-Cl(3)	1.706(11)
C(12)-C(13)	1.359(11)	C(99)-Cl(2)	1.720(12)
C(12)-H(12)	.93	C(99)-Cl(1)	1.747(12)
C(13)-C(14)	1.356(11)	C(99)-H(99)	.81(7)
C(13)-H(13)	.93	C(100)-Cl(5)	1.77(2)
C(14)-C(15)	1.391(11)	C(100)-Cl(4)	1.79(2)
C(14)-H(14)	.93	C(100)-Cl(6)	1.79(2)
C(15)-H(15)	.93	C(10A)-Cl(4A)	1.52(2)
C(16)-C(17)	1.371(9)	C(10A)-Cl(5A)	1.55(2)
C(16)-C(21)	1.391(10)	C(10A)-Cl(6A)	1.59(2)
C(17)-C(18)	1.388(10)		
C(17)-H(17)	.93		
C(18)-C(19)	1.376(11)		
C(18)-H(18)	.93		
C(19)-C(20)	1.377(11)		
C(19)-H(19)	.93		
C(20)-C(21)	1.379(11)		
C(20)-H(20)	.93		
C(21)-H(21)	.93		
P(2)-C(22)	1.802(9)		
P(2)-C(34)	1.803(8)		
P(2)-C(28)	1.818(8)		
C(22)-C(27)	1.361(12)		
C(22)-C(23)	1.389(11)		

Table S28. Full Listing of Bond Angles (°) and Their Estimated Standard Deviations for [Cu(cyan-κ*N*)(PPh_3)$_2$], **9**.

N(1)-Cu(1)-P(1)	120.2(2)	C(14)-C(13)-H(13)	119.6(6)
N(1)-Cu(1)-P(2)	110.8(2)	C(12)-C(13)-H(13)	119.6(6)
P(1)-Cu(1)-P(2)	128.94(8)	C(13)-C(14)-C(15)	119.2(9)
C(3)-N(1)-C(1)	120.4(6)	C(13)-C(14)-H(14)	120.4(6)
C(3)-N(1)-Cu(1)	114.3(5)	C(15)-C(14)-H(14)	120.4(6)
C(1)-N(1)-Cu(1)	125.0(5)	C(10)-C(15)-C(14)	121.6(8)
O(1)-C(1)-N(1)	123.0(7)	C(10)-C(15)-H(15)	119.2(5)
O(1)-C(1)-N(2)	120.0(7)	C(14)-C(15)-H(15)	119.2(6)
N(1)-C(1)-N(2)	117.0(7)	C(17)-C(16)-C(21)	118.2(8)
C(2)-N(2)-C(1)	126.3(7)	C(17)-C(16)-P(1)	124.9(7)
C(2)-N(2)-H(2)	116(4)	C(21)-C(16)-P(1)	116.9(6)
C(1)-N(2)-H(2)	118(4)	C(16)-C(17)-C(18)	121.1(8)
O(2)-C(2)-N(3)	123.9(7)	C(16)-C(17)-H(17)	119.5(5)
O(2)-C(2)-N(2)	124.4(7)	C(18)-C(17)-H(17)	119.5(5)
N(3)-C(2)-N(2)	111.7(7)	C(19)-C(18)-C(17)	119.7(9)
O(3)-C(3)-N(1)	119.9(7)	C(19)-C(18)-H(18)	120.2(6)
O(3)-C(3)-N(3)	120.8(8)	C(17)-C(18)-H(18)	120.2(5)
N(1)-C(3)-N(3)	119.2(7)	C(18)-C(19)-C(20)	120.3(9)
C(2)-N(3)-C(3)	125.3(7)	C(18)-C(19)-H(19)	119.8(6)
C(2)-N(3)-H(3)	115(4)	C(20)-C(19)-H(19)	119.8(6)
C(3)-N(3)-H(3)	119(4)	C(19)-C(20)-C(21)	119.2(9)
C(16)-P(1)-C(4)	103.5(4)	C(19)-C(20)-H(20)	120.4(6)
C(16)-P(1)-C(10)	105.6(3)	C(21)-C(20)-H(20)	120.4(6)
C(4)-P(1)-C(10)	103.3(4)	C(20)-C(21)-C(16)	121.4(8)
C(16)-P(1)-Cu(1)	111.6(3)	C(20)-C(21)-H(21)	119.3(6)
C(4)-P(1)-Cu(1)	118.1(3)	C(16)-C(21)-H(21)	119.3(5)
C(10)-P(1)-Cu(1)	113.4(3)	C(22)-P(2)-C(34)	104.0(4)
C(5)-C(4)-C(9)	118.0(8)	C(22)-P(2)-C(28)	103.3(4)
C(5)-C(4)-P(1)	118.2(8)	C(34)-P(2)-C(28)	105.3(4)
C(9)-C(4)-P(1)	123.8(7)	C(22)-P(2)-Cu(1)	112.5(3)
C(4)-C(5)-C(6)	120.1(10)	C(34)-P(2)-Cu(1)	111.9(3)
C(4)-C(5)-H(5)	120.0(6)	C(28)-P(2)-Cu(1)	118.4(3)
C(6)-C(5)-H(5)	120.0(7)	C(27)-C(22)-C(23)	118.0(9)
C(7)-C(6)-C(5)	121.4(12)	C(27)-C(22)-P(2)	123.9(8)
C(7)-C(6)-H(6)	119.3(8)	C(23)-C(22)-P(2)	118.1(8)
C(5)-C(6)-H(6)	119.3(7)	C(24)-C(23)-C(22)	120.7(11)
C(6)-C(7)-C(8)	119.0(12)	C(24)-C(23)-H(23)	119.7(9)
C(6)-C(7)-H(7)	120.5(8)	C(22)-C(23)-H(23)	119.7(6)
C(8)-C(7)-H(7)	120.5(8)	C(25)-C(24)-C(23)	120(2)
C(7)-C(8)-C(9)	120.3(11)	C(25)-C(24)-H(24)	119.8(11)
C(7)-C(8)-H(8)	119.9(8)	C(23)-C(24)-H(24)	119.8(9)
C(9)-C(8)-H(8)	119.9(7)	C(26)-C(25)-C(24)	120(2)
C(4)-C(9)-C(8)	121.2(10)	C(26)-C(25)-H(25)	120.1(11)
C(4)-C(9)-H(9)	119.4(5)	C(24)-C(25)-H(25)	120.1(11)
C(8)-C(9)-H(9)	119.4(7)	C(25)-C(26)-C(27)	121(2)
C(15)-C(10)-C(11)	117.8(8)	C(25)-C(26)-H(26)	119.5(11)
C(15)-C(10)-P(1)	118.8(7)	C(27)-C(26)-H(26)	119.5(9)
C(11)-C(10)-P(1)	123.3(7)	C(22)-C(27)-C(26)	120.0(12)
C(12)-C(11)-C(10)	120.2(9)	C(22)-C(27)-H(27)	120.0(6)
C(12)-C(11)-H(11)	119.9(6)	C(26)-C(27)-H(27)	120.0(9)
C(10)-C(11)-H(11)	119.9(5)	C(29)-C(28)-C(33)	117.0(8)
C(13)-C(12)-C(11)	120.3(9)	C(29)-C(28)-P(2)	119.7(7)
C(13)-C(12)-H(12)	119.8(6)	C(33)-C(28)-P(2)	123.3(7)
C(11)-C(12)-H(12)	119.8(6)	C(28)-C(29)-C(30)	121.1(9)
C(14)-C(13)-C(12)	120.8(10)	C(28)-C(29)-H(29)	119.4(6)

C(30)-C(29)-H(29)	119.4(7)
C(31)-C(30)-C(29)	121.0(11)
C(31)-C(30)-H(30)	119.5(7)
C(29)-C(30)-H(30)	119.5(7)
C(30)-C(31)-C(32)	119.1(11)
C(30)-C(31)-H(31)	120.4(7)
C(32)-C(31)-H(31)	120.4(7)
C(33)-C(32)-C(31)	120.6(10)
C(33)-C(32)-H(32)	119.7(6)
C(31)-C(32)-H(32)	119.7(7)
C(32)-C(33)-C(28)	121.2(9)
C(32)-C(33)-H(33)	119.4(6)
C(28)-C(33)-H(33)	119.4(5)
C(39)-C(34)-C(35)	118.0(8)
C(39)-C(34)-P(2)	117.9(8)
C(35)-C(34)-P(2)	124.0(8)
C(36)-C(35)-C(34)	121.3(10)
C(36)-C(35)-H(35)	119.3(8)
C(34)-C(35)-H(35)	119.3(6)
C(37)-C(36)-C(35)	119.6(12)
C(37)-C(36)-H(36)	120.2(9)
C(35)-C(36)-H(36)	120.2(7)
C(36)-C(37)-C(38)	121.8(13)
C(36)-C(37)-H(37)	119.1(9)
C(38)-C(37)-H(37)	119.1(9)
C(37)-C(38)-C(39)	118.0(12)
C(37)-C(38)-H(38)	121.0(9)
C(39)-C(38)-H(38)	121.0(8)
C(34)-C(39)-C(38)	121.1(10)
C(34)-C(39)-H(39)	119.4(6)
C(38)-C(39)-H(39)	119.4(8)
Cl(3)-C(99)-Cl(2)	113.0(7)
Cl(3)-C(99)-Cl(1)	109.9(7)
Cl(2)-C(99)-Cl(1)	110.3(6)
Cl(3)-C(99)-H(99)	99(6)
Cl(2)-C(99)-H(99)	110(6)
Cl(1)-C(99)-H(99)	114(7)
Cl(5)-C(100)-Cl(4)	103(2)
Cl(5)-C(100)-Cl(6)	104.5(14)
Cl(4)-C(100)-Cl(6)	103(2)
Cl(4A)-C(10A)-Cl(5A)	119(2)
Cl(4A)-C(10A)-Cl(6A)	115(2)
Cl(5A)-C(10A)-Cl(6A)	111(2)

Table S29. Anisotropic displacement parameters ($Å^2$ x 10^3) for [Cu(cyan-κ*N*)(PPh_3)$_2$], **9**.

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Cu(1)	51(1)	55(1)	37(1)	8(1)	5(1)	24(1)
N(1)	36(4)	53(4)	33(4)	3(3)	5(3)	21(4)
C(1)	46(6)	52(5)	29(5)	-4(4)	-4(4)	20(5)
O(1)	40(3)	88(4)	43(3)	-3(3)	6(3)	34(3)
N(2)	25(4)	69(5)	51(5)	5(4)	0(4)	24(4)
C(2)	42(6)	72(6)	47(6)	13(5)	4(5)	36(5)
O(2)	54(4)	87(4)	38(3)	-8(3)	-2(3)	35(3)
C(3)	48(6)	57(6)	40(5)	4(4)	4(5)	24(5)
O(3)	55(4)	106(5)	47(4)	-8(3)	1(3)	48(4)
N(3)	48(5)	83(5)	33(4)	-6(4)	4(4)	44(4)
P(1)	59(2)	52(1)	39(1)	9(1)	4(1)	24(1)
C(4)	75(7)	59(6)	35(5)	5(5)	3(5)	28(6)
C(5)	101(9)	58(7)	98(8)	2(6)	-16(7)	27(7)
C(6)	132(11)	58(8)	137(11)	2(8)	-24(8)	20(8)
C(7)	171(15)	54(8)	135(12)	17(8)	30(10)	47(10)
C(8)	151(13)	70(9)	109(10)	23(7)	21(9)	56(9)
C(9)	89(8)	61(7)	86(7)	16(6)	10(6)	36(6)
C(10)	55(6)	52(6)	34(5)	10(4)	-3(4)	17(5)
C(11)	92(7)	76(7)	50(6)	22(5)	16(5)	45(6)
C(12)	109(9)	101(9)	64(7)	40(6)	42(6)	51(7)
C(13)	105(8)	95(8)	68(7)	27(7)	34(6)	54(7)
C(14)	127(9)	74(7)	65(7)	17(6)	41(7)	49(7)
C(15)	86(7)	56(6)	60(6)	21(5)	21(5)	24(5)
C(16)	51(5)	50(5)	43(5)	8(4)	10(5)	20(4)
C(17)	62(7)	68(6)	51(6)	16(5)	11(5)	23(5)
C(18)	67(8)	89(7)	54(6)	4(5)	-7(6)	25(6)
C(19)	63(7)	81(8)	93(8)	18(7)	6(7)	20(6)
C(20)	63(8)	83(7)	91(8)	21(6)	32(6)	23(6)
C(21)	59(7)	78(7)	52(6)	14(5)	8(5)	23(6)
P(2)	67(2)	55(2)	42(1)	14(1)	9(1)	23(1)
C(22)	63(7)	82(7)	50(6)	30(5)	3(5)	21(6)
C(23)	90(8)	76(7)	48(6)	17(5)	16(6)	8(6)
C(24)	125(13)	146(13)	48(8)	32(7)	13(8)	-6(9)
C(25)	78(12)	219(21)	96(12)	86(14)	-19(9)	-14(11)
C(26)	87(10)	177(15)	162(15)	128(13)	19(10)	38(9)
C(27)	100(9)	142(10)	84(8)	78(8)	23(7)	52(8)
C(28)	60(6)	53(6)	44(5)	8(4)	8(5)	19(5)
C(29)	82(8)	75(7)	66(7)	23(6)	-5(6)	21(7)
C(30)	79(9)	90(9)	113(9)	28(8)	-15(7)	13(8)
C(31)	61(8)	94(10)	92(9)	7(7)	-2(7)	2(7)
C(32)	93(9)	67(7)	67(7)	10(6)	15(7)	0(7)
C(33)	80(8)	57(6)	65(6)	20(5)	3(6)	13(6)
C(34)	70(7)	69(6)	52(6)	23(5)	29(5)	34(6)
C(35)	113(9)	72(7)	76(7)	13(6)	46(7)	42(7)
C(36)	130(12)	92(10)	116(10)	16(8)	51(10)	37(10)
C(37)	161(15)	101(11)	145(13)	49(10)	92(12)	73(11)
C(38)	80(9)	133(12)	164(13)	67(10)	62(10)	42(9)
C(39)	79(8)	79(8)	99(8)	25(6)	33(7)	28(7)
C(99)	85(9)	77(9)	90(9)	22(7)	-6(6)	16(7)
Cl(1)	102(2)	130(3)	117(2)	24(2)	15(2)	50(2)
Cl(2)	110(2)	110(2)	147(3)	23(2)	41(2)	31(2)
Cl(3)	117(3)	213(4)	102(2)	22(2)	-27(2)	54(3)

C(100)	363(55)	124(26)	321(52)	110(29)	293(50)	111(30)
Cl(4)	188(8)	148(6)	144(7)	50(5)	14(6)	67(6)
Cl(5)	176(9)	330(20)	265(14)	128(13)	115(9)	154(11)
Cl(6)	293(16)	163(9)	175(7)	-7(6)	64(8)	130(11)
Cl(4A)	574(57)	229(18)	248(24)	101(17)	237(32)	101(30)
Cl(5A)	198(17)	265(25)	588(47)	79(26)	176(23)	122(17)
Cl(6A)	190(17)	144(14)	674(41)	29(19)	1(21)	44(12)

The anisotropic displacement factor takes the form:

$$\exp\left(-2\pi^2\left[h^2a^{*2}U_{11} + k^2b^{*2}U_{22} + \ldots + 2hka^*b^*U_{12}\right]\right)$$

Table S30. Hydrogen Atom Coordinates ($\times 10^4$) and Isotropic Displacement Parameters ($Å^2 \times 10^3$) for [Cu(cyan-κ*N*)(PPh_3)$_2$], **9**.

	x	y	z	U(eq)
H(2)	5613(21)	-396(46)	4586(33)	35(15)
H(3)	8764(47)	-479(46)	4547(33)	35(15)
H(5)	7072(10)	-987(8)	7230(6)	115
H(6)	6294(12)	-2917(10)	6920(7)	151
H(7)	7520(15)	-3729(10)	7429(9)	148
H(8)	9542(13)	-2597(10)	8283(7)	130
H(9)	10315(9)	-665(8)	8640(6)	96
H(11)	8104(8)	-320(8)	9445(5)	83
H(12)	7075(9)	46(9)	10493(6)	101
H(13)	6892(10)	1704(10)	10825(6)	101
H(14)	7715(10)	3024(8)	10125(6)	102
H(15)	8781(8)	2689(7)	9090(5)	82
H(17)	11013(9)	1968(7)	9975(5)	75
H(18)	13214(10)	2710(7)	10487(6)	94
H(19)	14513(9)	2676(8)	9555(7)	102
H(20)	13619(10)	1888(8)	8111(7)	97
H(21)	11425(9)	1158(7)	7609(5)	80
H(23)	9801(9)	2730(7)	5738(6)	97
H(24)	9383(13)	2974(11)	4415(7)	154
H(25)	8295(14)	4007(16)	4174(11)	185
H(26)	7692(12)	4864(13)	5250(10)	163
H(27)	8104(10)	4655(9)	6599(7)	122
H(29)	11533(10)	3619(8)	8486(6)	97
H(30)	13652(11)	4871(10)	9033(7)	127
H(31)	14492(10)	6508(10)	8673(7)	118
H(32)	13182(12)	6929(8)	7773(6)	107
H(33)	11071(10)	5708(8)	7227(5)	88
H(35)	9883(10)	5294(8)	8858(6)	102
H(36)	8608(15)	5830(10)	9621(8)	139
H(37)	6453(17)	4853(12)	9206(10)	147
H(38)	5480(11)	3356(12)	7981(10)	143
H(39)	6774(11)	2773(8)	7236(6)	104
H(99)	5693(80)	-10(64)	2931(51)	68(35)

Treatment of Diffraction Data for *trans*-[Cu(cyan-κN)$_2$(NH$_3$)$_2$]*trans*-[Cu(cyan-κO^2)$_2$(NH$_3$)$_4$], 6.

All of the crystals of compound **6** that were examined were found to be twinned, inasmuch as each sample consisted of at least two crystalline domains joined together with random relative orientations. All of our attempts to produce monocrystalline samples, as well as our attempts to separate crystalline domains within individual samples, were unsuccessful. It proved possible to index the principal components of various crystals, and it was also possible to conduct a structure determination using a twinned sample and treating the data as though the second crystalline domain did not exist. As a first step in determining the structure of compound **6**, such a determination was conducted. The results possessed the expected features -- non-positive-definite anisotropic displacement parameters for a good number of the non-hydrogen atoms, poor location of whatever hydrogen atoms appeared, lack of convergence, and high residuals. The qualitative features of the structure were clear, however, and the distances and angles involving non-hydrogen atoms fell within the ranges expected.

With the aim of obtaining a more satisfactory refinement of the structure, we attempted to separate the diffraction pattern into components derived from the two domains of the crystal, using the following methodology. In a first indexation of 25 reflections obtained in a random search, it was possible to index 17 of the data points using the triclinic unit cell that we report for **6**. The diffraction pattern indexed first with these 17 data will be referred to as the "principal component." The cell dimensions were verified by axial photography, but the photographs clearly showed the presence of a second, minor component in the diffraction pattern. Furthermore, the second diffraction pattern was oriented in the worst possible way with respect to the first -- with overlap at some reciprocal lattice points, without overlap at others, and with apparent partial overlap at still others. The minor pattern was weaker, however, and possessed little observable intensity at higher scattering angles. The eight reflections that had not been indexed for the first component were indexed separately, giving the same unit cell as that of the first component, but a different orientation matrix. This orientation matrix was then refined using 22 reflections. In what follows, the orientation matrices of the first and second components are called [UB1] and [UB2], respectively.

Although the procedure that was actually used to attempt to separate the diffraction patterns was somewhat more involved, the following schematic description gives all of the

important steps.

The orientation matrix, [UB1], of the principal component, relates the indices (h_1,k_1,ℓ_1) of one reflection produced by this component to the coordinates $(d^*_{1,x},d^*_{1,y},d^*_{1,z})$ of the associated scattering vector in the orthogonal reference frame of the diffractometer, when all of the goniometer angles are located at their zero points (Eq. 1). In Equation 1, the

$$\begin{bmatrix} d^*_{1,x} \\ d^*_{1,y} \\ d^*_{1,z} \end{bmatrix} = \begin{bmatrix} a^*_{1,x} & b^*_{1,x} & c^*_{1,x} \\ a^*_{1,y} & b^*_{1,y} & c^*_{1,y} \\ a^*_{1,z} & b^*_{1,z} & c^*_{1,z} \end{bmatrix} \begin{bmatrix} h_1 \\ k_1 \\ l_1 \end{bmatrix} \qquad \textbf{(1)}$$

components of the orientation matrix [UB1] have been given explicitly; they are the coordinates of the reciprocal lattice basis vectors $\mathbf{a}^*$, $\mathbf{b}^*$, and $\mathbf{c}^*$, in the same reference frame and under the same conditions. An analogous equation holds for component 2.

For the present purposes, the equation for the second component is more useful in its inverted form (Eq. 2), in which the reciprocal of the orientation matrix [UB2] multiplies the coordinates of a scattering vector to give the indices (h_2,k_2,ℓ_2) of a second-component reflection.

$$\begin{bmatrix} h_2 \\ k_2 \\ l_2 \end{bmatrix} = \begin{bmatrix} a^*_{2,x} & b^*_{2,x} & c^*_{2,x} \\ a^*_{2,y} & b^*_{2,y} & c^*_{2,y} \\ a^*_{2,z} & b^*_{2,z} & c^*_{2,z} \end{bmatrix}^{-1} \begin{bmatrix} d^*_{2,x} \\ d^*_{2,y} \\ d^*_{2,z} \end{bmatrix} \qquad \textbf{(2)}$$

In order to identify reciprocal lattice points of the principal component which suffer overlap by reciprocal lattice points of the minor component, Equations 1 and 2 are combined. First of all, for a given (h_1,k_1,ℓ_1) of the first component, Eq. 1 is applied, giving the orthogonal coordinates $(d^*_{1,x},d^*_{1,y},d^*_{1,z})$ of the corresponding scattering vector. This vector is then multiplied by $[UB2]^{-1}$ giving the second-component indices (h_2,k_2,ℓ_2) corresponding to the same position in space as the first-component reflection (h_1,k_1,ℓ_1). If the second-

component indices (h_2, k_2, ℓ_2) are all integral or nearly integral, then there is reflection overlap at the point under consideration.

As a practical test for reflection overlap, we calculated the second-component indices -- which are not necessarily integral -- corresponding to each reflection measured for the first component. The nearest reciprocal lattice point of the second component was identified, and the angle subtended by the first- and second-component reciprocal lattice vectors was used as the the discrimination parameter in the test for overlap. This was done both for the angle subtended by the two vectors at the reciprocal lattice origin, and for the angle subtended by the two at the center of the sphere of reflection under the actual experimental conditions. The latter was expected to be more physically meaningful, but in the actual event the two test parameters led to very similar results.

Various values of the discrimination parameter were tested. Moreover, two different approaches to the handling of overlapped data were tested. In the first approach, data considered to have significant overlap were omitted from the data set entirely. In the second approach, if a second-component reflection was found to be quite nearly superimposed upon a measured datum, the respective datum from the second component was included in the data set; in this case, the structure was refined as a twin. As has been our experience with this particular kind of twin, the second approach did not lead to any improvement in the results, as the degree of overlap varies from one affected reflection to another.

For the refinement that we report in this paper, the first approach was used -- namely, the elimination of points considered to have suffered too much from overlap, and the refinement of the structure of a single domain. We found that the use of a strict overlap discrimination factor of 0.1° led to the rejection of a majority of the low-angle reflections. The use of a more lax discrimination factor inevitably led to the inclusion of data that had been affected by the second component, but the compromise of using some such data was necessary in order to achieve a data-to-parameter ratio adequate for refining the structure.

In the end, 43 of the 2504 measured reflections were rejected on the basis of peak overlap between the two components. Another two reflections had been rejected during data reduction because of severe peak asymmetry. This eliminated the worst data, but it is clear that the data set, now with a total of 2459 data, was left with a good number of reflections that were affected by the second component. For the final refinement, 2337 data were used, namely all remaining data with positive measured intensities. Equivalent data were not

merged, as the presence of this type of twinning can lead to inequivalences; and averaging such data would lead to further deterioration in their quality. Hydrogen atoms were left out of the structural model. Otherwise, the geometrical results are in very good accord with all that is known about cyanurates, including cyanurate complexes of copper. We observed the expected effects in the final analysis of variance -- a worse fit of data at low values of 2θ, a systematic trend towards $F_o > F_c$ for the weakest data, and high residual difference densities (ρ(max,min) = +3.59,-2.63 e/Å^3).

As described, we attempted numerous refinements with datasets that had been treated differently in terms of rejection of overlapped data. In the end, though, the bond distances and angles derived from all of the convergent refinements were essentially identical, irrespective of the discrimination factor used, the method of discrimination, and the crystallographic residuals.