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Supporting Information

Nickel(I) Monomers and Dimers with Cyclopentadienyl and Indenyl Ligands

Jianguo Wu,[a] Ainara Nova,*[b] David Balcells,[b] Gary W. Brudvig,[a] Wei Dai,[a] Ms. Louise M. Guard,[a] Nilay Hazari,*[a] Po-Heng Lin,[c] Ravi Pokhrel,[a] and Michael K. Takase[a]

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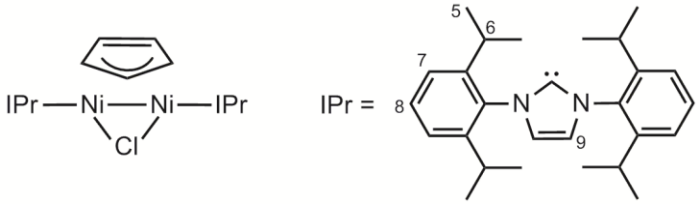
Experimental Procedures and Characterizing Data for New Compounds

(μ-Cp)(μ-Cl)Ni_2(IPr)$_2$ (1a)

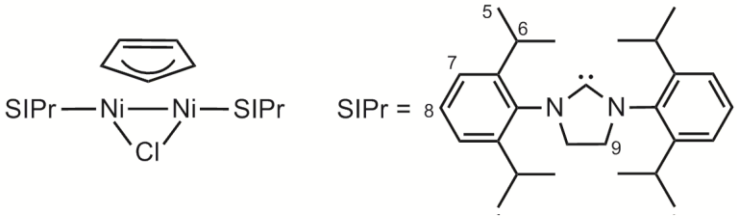
To a solution of (μ-Cl)$_2$$Ni_2(IPr)_2$ (83.4 mg, 0.086 mmol) in 4 mL diethyl ether at -35ºC, NaCp (7.0 mg, 0.08 mmol) was added. The mixture was stirred at -35ºC for 6 hours. The volatiles were removed under vacuum. The resulting residue was extracted with pentane (2 x 5 mL). The combined solution from the extractions was filtered through celite and dried under reduced pressure to give **1a** as a green solid which was purified by recrystallization from a benzene/pentane solution. Single crystals for X-ray analysis were grown from a pentane solution at -35ºC. Yield: 44.7 mg (52%).



Anal. calcd (found) for $C_{59}H_{77}N_4ClNi_2$: C, 71.21 (71.18); H, 7.80 (7.89); N, 5.63 (5.39). ^{1}H NMR (400 MHz, C_7D_8, 223 K): 7.16-6.97 (m, 12H, H8 and H7), 6.43 (s, 4H, H9), 3.93 (s, 5H, Cp), 3.17 (septet, *J* = 6.4 Hz, 8H, H6), 1.43 (d, *J* = 6.2 Hz, 24H, H5), 1.13 (d, *J* = 6.4 Hz, 24H, H5). ^{13}C{^{1}H} NMR (125.7 MHz, C_7D_8, 223 K): 188.1, 145.8, 137.6, 123.6, 122.6, 79.5, 28.7, 25.8, 23.1. UV-Vis λ_{max} (ε): 272 nm (88016), 326 nm (63095), 384 nm (71111), 608 nm (786, br).

(μ-Cp)(μ-Cl)Ni_2(SIPr)$_2$ (1b)

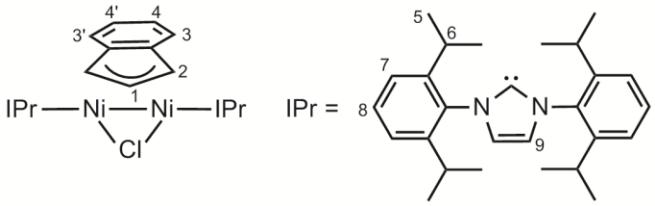
To a solution of (μ-Cl)$_2$$Ni_2$(SIPr)$_2$ (82.4 mg, 0.084 mmol) in 8 mL diethyl ether at -35ºC, NaCp (7.4 mg, 0.084 mmol) was added. The mixture was stirred at -35ºC for 8 hours. The volatiles were removed under vacuum. The resulting residue was extracted with pentane (2 x 5 mL). The combined solution from the extractions was filtered through celite and dried under reduced pressure to give **1b** as a green solid which was purified by recrystallization from a benzene/pentane solution. Yield: 65 mg (77%).



Anal. calcd (found) for $C_{59}H_{77}N_4ClNi_2$: C, 70.93 (70.56); H, 8.17 (7.76); N, 5.61 (5.43). ^{1}H NMR (500 MHz, C_7D_8, 203 K): 7.16 (m, 12H, H8 and H7), 3.91 (s, 5H, Cp), 3.49 (m, 8H, H6), 3.31 (br s, 8H, H9), 1.47 (br s, 24H, H5), 1.24 (br s, 24H, H5). ^{13}C{^{1}H} NMR (125.7 MHz, C_7D_8, 203 K): 214.3, 148.1, 138.4, 188.0, 137.6, 124.7, - 81.1, 54.2, 29.1, 26.9, 24.5. UV-Vis λ_{max} (ε): 280 nm (78464), 331 nm (41531), 382 nm (65693), 633 nm (571, br).

(μ-Ind)(μ-Cl)Ni$_2$(IPr)$_2$ (2a)

To a solution of (μ-Cl)$_2$Ni$_2$(IPr)$_2$ (46 mg, 0.048 mmol) in 6 mL diethyl ether at -35ºC, LiInd (5.8 mg, 0.048 mmol) was added. The mixture was stirred at -35ºC for 6 hours. The volatiles were removed under vacuum. The resulting residue was extracted with benzene (2 x 3 mL). The combined solution from the extractions was filtered through celite and dried under reduced pressure to give **2a** as a green solid which was purified by recrystallization from a benzene/pentane solu

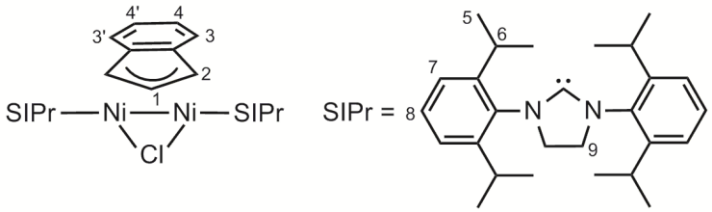


Anal. calcd (found) for $C_{63}H_{79}ClN_4Ni_2$: C, 72.40 (72.41); H, 7.62 (8.06); N, 5.36 (5.14). ^{1}H NMR (400 MHz, C_6D_6): 7.22 (t, *J* = 7.3 Hz, 4H, H8), 7.10 (d, *J* = 7.3 Hz, 8H, H7), 6.76 (m, 2H, H3/H3' or H4/H4'), 6.57 (s, 4H, H9), 6.55 (m, 2H, H3/H3' or H4/H4'), 3.59 (d, *J* = 4.0 Hz, 2H, H2), 3.31 (septet, *J* = 6.8 Hz, 4H, H6), 3.03 (septet, *J* = 6.8 Hz, 4H, H6), 2.48 (t, *J* = 4.3 Hz, 1H, H1), 1.34 (d, *J* = 6.8 Hz, 12H, H5), 1.15 (d, *J* = 6.8 Hz, 12H, H5), 1.12 (d, *J* = 6.8 Hz, 12H, H5), 1.08 (d, *J* = 6.8 Hz, 12H, H5). ^{13}C{^{1}H} NMR (125.7 MHz, C_6D_6): 190.7, 148.9, 146.7, 146.2, 138.1, 124.5, 124.3, 124.2, 122.3, 121.5, 52.1, 34.6, 29.2, 26.5, 25.9, 23.9. UV-Vis λ_{max} (ε): 265 nm (18798, sh), 316 nm (18224, sh), 344 nm (20066), 382 nm (19392), 614 nm (867, br).

(μ-Ind)(μ-Cl)Ni$_2$(SIPr)$_2$ (2b)

To a solution of (μ-Cl)$_2$Ni$_2$(SIPr)$_2$ (97.3 mg, 0.099 mmol) in 8 mL diethyl ether at -35ºC, LiInd (12.1 mg, 0.099 mmol) was added. The mixture was stirred at -35ºC for 6 hours. The volatiles were removed under vacuum. The resulting residue was washed with pentane and extracted with benzene (3 x 4 mL). The combined solution from the extractions was filtered

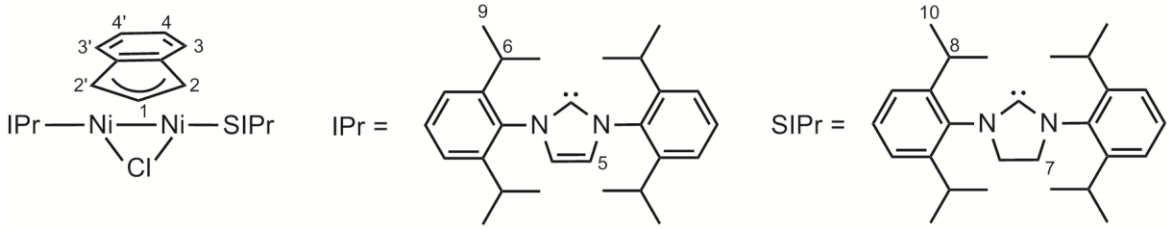
through celite and the volume of solution was reduced to ~1 mL under reduced pressure. Approximately 4 mL pentane was then added, which caused a solid to precipitate. The resulting precipitate was collected by filtration, washed with pentane and dried in vacuo to give **2b** as a green solid. Single crystals for X-ray analysis were grown from a benzene/pentane solution at room temperature. Yield: 81 mg (78%).



Anal. calcd (found) for $C_{63}H_{81}ClN_4Ni_2$: C, 72.26 (72.03); H, 7.80 (7.92); N, 5.35 (4.91). ^{1}H NMR (400 MHz, C_6D_6): 7.20 (d, *J* = 8 Hz, 4H, H8), 7.08 (t, *J* = 8 Hz, 8H, H7), 6.76 (m, 2H, H3/H3' or H4/H4'), 6.52 (m, 2H, H3/H3' or H4/H4'), 3.74 (d, *J* = 4 Hz, 2H, H2), 3.50 (s, 8H, H9), 3.47 (septet, *J* = 6.8 Hz, 8H, H6), 2.49 (t, *J* = 4 Hz, 1H, H1), 1.37 (d, *J* = 6.8 Hz, 12H, H5), 1.19 (d, *J* = 6.8 Hz, 12H, H5), 1.18 (d, *J* = 6.8 Hz, 12H, H5), 1.13 (d, *J* = 6.8 Hz, 12H, H5). ^{13}C{^{1}H} NMR (125.7 MHz, C_6D_6): 217.3, 149.1, 147.7, 147.2, 138.9, 125.0, 124.7, 122.5, 121.6, 54.4, 35.4, 29.2, 29.1, 26.6, 24.8, 24.5. UV-Vis λ_{max} (ε): 316 nm (12802), 344 nm (11622, sh), 384 nm (10863), 604 nm (572, br).

(μ-Ind)(μ-Cl)Ni$_2$(IPr)(SIPr) (2ab)

To a solution of **4b** (20.3 mg, 0.036 mmol) in 1 mL benzene, (μ-Cl)$_2$Ni$_2$(IPr)$_2$ (17.3 mg, 0.018 mmol) was added. The solution changed from red to green. The mixture was stirred for 0.5 hours and the volatiles were removed under vacuum. The resulting residue was washed with pentane and dried in vacuo to give crude **2ab** as a green solid which was purified by recrystallization from a benzene/pentane solution. Yield: 31 mg (82 %).



Anal. calcd (found) for $C_{63}H_{81}ClN_4Ni_2$: C, 72.26 (71.83); H, 7.80 (8.12); N, 5.35 (4.94). ^{1}H NMR (400 MHz, C_6D_6): 7.23-7.02 (m, 12H, Ph), 6.82 (t, *J* = 5.6 Hz, 1H), 6.78 (m, 2H, H3/H3' or H4/H4'), 6.68 (t, 2H, H3/H3' or H4/H4'), 6.58 (s, 2H, H5), 3.65 (d, *J* = 2.4 Hz,

1H, H2 or H2'), 3.57 (d, J = 2.4 Hz, 1H, H2 or H2'), 3.47 (m, 4H, H8), 3.29 (septet, *J* = 5.2 Hz, 2H, H6), 3.02 (septet, *J* = 5.2 Hz, 2H, H6), 2.38 (t, *J* = 5.2 Hz, 1H, H1), 1.41 (d, *J* = 5.2 Hz, 6H, H9 or H10), 1.35 (d, *J* = 5.2 Hz, 6H, H9 or H10), 1.23 (d, *J* = 5.2 Hz, 12H, H9 or H10), 1.20 (d, *J* = 5.2 Hz, 6H, H9 or H10), 1.10 (d, *J* = 5.2 Hz, 6H, H9 or H10), 1.04 (d, *J* = 5.2 Hz, 6H, H9 or H10), 0.98 (d, *J* = 5.2 Hz, 6H, H9 or H10). $^{13}C\{^{1}H\}$ NMR (125.7 MHz, C_6D_6): 217.5, 190.9, 149.3, 149.2, 147.9, 146.7, 146.6, 146.4, 138.2, 129.7, 128.9, 128.7, 125.4, 124.5, 124.4, 124.2, 122.6, 122.3, 121.6, 121.5, 54.6, 52.8, 29.2, 29.1, 26.8, 26.6, 26.4, 26.0, 24.8, 24.7, 23.8, 23.6. UV-Vis λ_{max} (ε): 346 nm (385895), 379 nm (406667), 601 nm (4451, br).

(η^5-Cp)Ni(IPr) (3a)

NaCp (11 mg, 0.12 mmol) was added to a solution of $(\mu\text{-Cl})_2Ni_2(IPr)_2$ (58.5 mg, 0.06 mmol) in 6 mL diethyl ether. The mixture was stirred at room temperature for 2 hours. The volatiles were removed under vacuum. The resulting residue was extracted with benzene (2 x 2 mL). The combined solution from the extractions was filtered through celite, dried under reduced pressure, washed with pentane (~2 x 1 mL) and further dried under vacuum to give **3a** as a yellow solid. Single crystals for X-ray analysis were grown from a benzene/pentane solution at -35ºC. Yield: 52 mg (83%).

Anal. calcd (found) for $C_{33}H_{41}N_2Ni$: C, 75.01 (74.20); H, 8.07 (7.95); N, 5.47 (5.25). ^{1}H NMR (400 MHz, C_6D_6): 26.29 (2H), 7.27 (4H), 5.28 (4H), 3.68 (2H), 2.12 (12H), 1.45 (12H), -39.85 (5H). Magnetic susceptibility (C_6D_6): 1.87 μ_B. UV-Vis λ_{max} (ε): 267 nm (8095), 333 nm (6358), 416 nm (5723).

(η^5-Cp)Ni(SIPr) (3b)

NaCp (35.6 mg, 0.40 mmol) was added to a solution of $(\mu\text{-Cl})_2Ni_2(SIPr)_2$ (195.2 mg, 0.20 mmol) in 12 mL diethyl ether. The mixture was stirred at room temperature for 2 hours. The volatiles were removed under vacuum. The resulting residue was extracted with benzene (2 x 4 mL). The combined solution from the extractions was filtered through celite, dried under reduced pressure, washed with pentane (~2 x 1 mL) and further dried under vacuum to give **3b** as a yellow solid. Yield: 153 mg (73%).

Anal. calcd (found) for $C_{33}H_{43}N_2Ni$: C, 74.72 (73.37); H, 8.43 (8.39); N, 5.45 (4.91). ^1H NMR (400 MHz, C_6D_6): 7.26 (4H), 6.29 (6H), 3.95 (10H), 2.83 (2H), 1.92 (13H), -11.49 (4H), -44.96 (5H). Magnetic susceptibility (C_6D_6): 1.71 $\mu_{B.}$ UV-Vis λ_{max} (ε): 247 nm (14700), 329 nm (5870), 411 nm (5314).

(η^5-Ind)Ni(IPr) (4a)

LiInd (21.5 mg, 0.176 mmol) was added to a solution of $(\mu\text{-Cl})_2Ni_2(IPr)_2$ (84.9 mg, 0.088 mmol) in 10 mL diethyl ether. The mixture was stirred at room temperature for 2 hours. The volatiles were removed under vacuum. The resulting residue was extracted with pentane (4 x 5 mL). The combined solution from the extractions was filtered through celite and reduced to ~0.5 mL under reduced pressure. The mixture was left to stand at -35ºC for 6 hours. The resulting precipitate was collected and dried under vacuum to give **4a** as an orange solid. Single crystals for X-ray analysis were grown from a pentane solution at -35ºC. Yield: 70 mg (72%).

Anal. calcd (found) for $C_{36}H_{43}N_2Ni$: C, 76.88 (75.07); H, 7.71 (7.66); N, 4.98 (4.79). ^1H NMR (400 MHz, C_6D_6): 19.27 (2H), 6.08 (5H), 4.51 (4H), 2.58 (12H), 1.92 (14H), 1.34 (1H), 1.13 (2H), 0.94 (1H), -14.56 (2H). Magnetic susceptibility (C_6D_6): 1.61 $\mu_{B.}$ UV-Vis λ_{max} (ε): 247 nm (20933), 333 nm (6358), 416 nm (5723).

(η^5-Ind)Ni(SIPr) (4b)

LiInd (35.4 mg, 0.29 mmol) was added to a solution of $(\mu\text{-Cl})_2Ni_2(SIPr)_2$ (140.3 mg, 0.14 mmol) in 14 mL diethyl ether. The mixture was stirred at room temperature for 2 hours. The volatiles were removed under vacuum. The resulting residue was extracted with pentane (4 x 8 mL). The combined solution from the extractions was filtered through celite and reduced to ~0.5 mL under reduced pressure. The mixture was left to stand at -35ºC for 6 hours. The resulting precipitate was collected and further dried under vacuum to give **4b** as an orange solid. Single crystals for X-ray analysis were grown from a saturated pentane solution at -35ºC. Yield: 123 mg (76%).

Anal. calcd (found) for $C_{36}H_{45}N_2Ni$: C, 76.60 (75.87); H, 8.04 (7.95); N, 4.96 (4.82). ^1H NMR (400 MHz, C_6D_6): 7.26 (6H), 6.43 (5H), 4.48 (12H), 3.33 (3H), 1.91 (13H), -3.80 (4H),

-15.61 (2H). Magnetic susceptibility (C_6D_6): 1.57 μ_B. UV-Vis λ_{max} (ε): 312 nm (8438, sh), 362 nm (7720), 451 nm (1729, sh), 524 nm (1158, br).

(η^5-Cp)Ni(IPr)(THF)PF_6

$FcPF_6$ (30 mg, 0.09 mmol) was added to a solution of (η^5-Cp)Ni(IPr) (47.6 mg, 0.09 mmol) in 2 mL THF. The mixture was stirred for 16 hours at room temperature. The solution was reduced to ~0.2 mL under vacuum and a solid formed upon addition of diethyl ether. The resulting precipitate was collected by filtration, washed with diethyl ether and dried under vacuum to give **(η^5-Cp)Ni(IPr)(THF)PF_6** as a red solid. Yield: 42 mg (63%).

^{1}H NMR (400 MHz, CD_2Cl_2, 203 K): 7.57 (t, *J* = 7.7 Hz, 2H, Ph), 7.42 (d, *J* = 7.8 Hz, 4H, Ph), 7.30 (s, 2H, C*H*=C*H* from IPr), 4.54 (s, 5H, Cp), 2.88 (m, 4H, C*H*Me_2), 2.67 (br s, 4H, THF), 1.35 (br s, 16H, CH_3 from IPr and THF), 1.08 (d, *J* = 6.3 Hz, 12H, CH_3 from IPr). ^{13}C{^{1}H} NMR (125.7 MHz, C_6D_6, 203 K): 157.2, 144.5, 135.0, 130.6, 126.8, 124.5, 92.56, 80.9, 67.8, 28.7, 25.6, 25.3, 21.9.

Exposure of **(η^5-Cp)Ni(IPr)(THF)PF_6** to high vacuum resulted in the formation of a new compound which was insoluble except in the presence of THF, when it reverted back to **(η^5-Cp)Ni(IPr)(THF)PF_6**. Elemental analysis on the new compound was consistent with **(η^5-Cp)Ni(IPr)PF_6** Anal. calcd (found) for $C_{33}H_{41}N_2PF_6Ni$: C, 59.22 (58.94); H, 6.17 (6.14); N, 4.19 (3.98).

*Reaction of **(η^5-Cp)Ni(IPr)(THF)PF_6** with tetrabutylammonium chloride*

To a suspension of **(η^5-Cp)Ni(IPr)(THF)PF_6** (10 mg, 0.012 mmol) in 0.7 mL C_6D_6, tetrabutylammonium chloride (3.8 mg, 0.012 mmol) was added. After 5 min, an orange-red solution formed. ^{1}H NMR spectroscopy indicated quantitative conversion into **(η^5-Cp)Ni(IPr)(Cl)**.[1]

*Reaction of **3a** with methyl iodide*

To a solution of **3a** (22 mg, 0.042 mmol) in 2 mL C_6H_6, methyl iodide (1.3 μL, 0.021 mmol) was added. After 5 min, the volatiles were removed under vacuum. ^{1}H NMR spectroscopy of the residue in C_6D_6 indicated the quantitative formation of 1:1 mixture of **(η^5-**

Cp)Ni(IPr)(Me) and **(η^5-Cp)Ni(IPr)(I)**, which have been synthesized by alternative routes and fully characterized.

(η^5-Cp)Ni(IPr)(Me)

To a suspension of CpNi(IPr)Cl (200 mg, 0.36 mmol) in 8 mL diethyl ether at -35ºC, methylmagnesium chloride (3M in THF, 0.12 mL, 0.36 mmol) was added. The mixture was slowly warmed to room temperature. The volatiles were removed under vacuum. The resulting residue was extracted with benzene (2 x 3 mL). The combined solution from the extractions was filtered through Celite and dried under reduced pressure to give **(η^5-Cp)Ni(IPr)(Me)** as an olive green solid. Yield: 126 mg (64%).

^{1}H NMR (400 MHz, C_6D_6): 7.28 (t, *J* = 7.8 Hz, 2H, Ph), 7.18 (d, *J* = 7.4 Hz, 4H, Ph), 6.55 (s, 2H, C*H*=C*H* from IPr), 4.88 (s, 5H, Cp), 3.04 (septet, 4H, C*H*Me$_2$), 1.42 (d, *J* = 6.4 Hz, 12H, C*H*$_3$ from IPr), 1.00 (d, *J* = 6.4 Hz, 12H, C*H*$_3$ from IPr), -0.71 (s, 3H, C*H*$_3$). ^{13}C{^{1}H} NMR (125.7 MHz, C_6D_6): 190.8, 146.8, 138.0, 130.2, 124.4, 124.2, 90.3, 29.4, 26.3, 23.0, -34.4.

(η^5-Cp)Ni(IPr)(I)

To a solution of CpNi(IPr)Cl (92 mg, 0.17 mmol) in 4 mL THF, sodium iodide (252 mg, 1.7 mmol) was added. The mixture was stirred at room temperature for 16 hours. The volatiles were removed under vacuum. The resulting residue was extracted with toluene (2 x 4 mL). The combined solution from the extractions was filtered through celite and the volume of solution was reduced to ~0.5 mL under reduced pressure. Pentane was then added until a precipitate formed. The resulting precipitate was collected by filtration, washed with pentane and dried in vacuo to give **(η^5-Cp)Ni(IPr)(I)** as a purple solid. Yield: 78 mg (72%).

Anal. calcd (found) for $C_{33}H_{41}N_2NiI$: C, 60.86 (60.65); H, 6.35 (6.42); N, 4.30 (4.20). ^{1}H NMR (400 MHz, C_6D_6): 7.29 (t, *J* = 8.4 Hz, 2H, Ph), 7.20 (d, *J* = 8.1 Hz, 4H, Ph), 6.64 (s, 2H, C*H*=C*H* from IPr), 4.84 (s, 5H, Cp), 3.07 (br s, 4H, C*H*Me$_2$), 1.45 (d, *J* = 6.7 Hz, 12H, C*H*$_3$ from IPr), 0.95 (d, *J* = 6.8 Hz, 12H, C*H*$_3$ from IPr). ^{13}C{^{1}H} NMR (125.7 MHz, C_6D_6): 176.8, 147.1, 137.8, 130.8, 126.4, 124.7, 92.8, 29.4, 26.8, 23.4.

$(CO)_3$Ni(IPr)

After three freeze-pump-thaw cycles, excess 1 atm CO was introduced via a dual manifold Schlenk line to a solution of **3a** (6 mg, 0.01 mmol) in 0.5 mL C_6D_6 at room temperature. ^{1}H NMR spectroscopy indicated quantitative conversion into **$(CO)_3$Ni(IPr)**.[2]

(allylether)Ni(IPr)

To a solution of **3a** (8 mg, 0.019 mmol) in 0.5 mL C_6D_6 at room temperature, allylether (2.3 μL, 0.019 mmol) was added. The mixture was heated at 50ºC for 29 hours. ^{1}H NMR spectroscopy indicated quantitative conversion into **(allylether)Ni(IPr)**.[3]

(dvse)Ni(IPr)

To a solution of **3a** (8 mg, 0.015 mmol) in 0.5 mL C_6D_6 at room temperature, dvse (3.4 μL, 0.015 mmol) was added. The mixture was heated at 50ºC for 92 hours. ^{1}H NMR spectroscopy indicated 96% conversion into **(dvse)Ni(IPr)**.[3]

General procedure for the Suzuki-Miyaura Reaction

To a suspension of *p*-chlorotoluene (6.4 mg, 0.05 mmol), phenylboronic acid (6.7 mg, 0.05 mmol) and KO^{t}Bu (16.8 mg, 0.15 mmol) in 0.5 mL benzene in a J. Young NMR tube, the appropriate Ni precatalyst (0.005 mmol) was added. The mixture was heated at 70ºC for 3.5 hours. The solvent was removed under vacuum. The residue was extracted with 0.8 mL $CDCl_3$ and filtered through celite. To this extraction, 1,3,5-trimethoxybenzene (8.4 mg, 0.05 mmol) was added as an internal standard. The yield of product was determined by ^{1}H NMR spectroscopy.

Procedure for EPR Spectroscopy and Spectra of Selected Compounds

Samples for EPR spectroscopy were prepared in a nitrogen filled glovebox by dissolving each complex in toluene. For each sample, 200 μL volume of 1 mM complex was pipetted into an EPR tube and the EPR tube was sealed in the glovebox. X-band EPR spectra were acquired on a Bruker ELEXSYS E500 EPR spectrometer equipped with a SHQ resonator and an Oxford ESR-900 helium-flow cryostat. EPR scans were acquired at 7 K with the following instrumental parameters: microwave frequency 9.39 GHz, modulation frequency 100 kHz, modulation amplitude 5 G, and microwave power 0.1 mW. Simulations of the EPR spectra were done using MATLAB 7.8 software and the EasySpin 4.0.0 package. Table S1 gives the Gaussian FWHM broadenings along the x-,y-,and z-axis used for each simulation.

Table S1: Gaussian FWHM broadenings along the x-,y-,and z-axis used for EPR simulation of **3a**, **3b**, **4a** and **4b**.

Compound/Gaussian broadening	Along x-axis FWHM (MHz)	Along y-axis FWHM (MHz)	Along z-axis FWHM (MHz)
3a	280	85	75
3b	120	210	48
4a	115	140	100
4b	120	210	48

Figure S1: X-band CW EPR spectrum of **3b** (blue) in toluene at 7K and the corresponding simulation (green). The g-values from the simulation are g_1 = 2.349, g_2 = 2.267 and g_3 = 2.062.

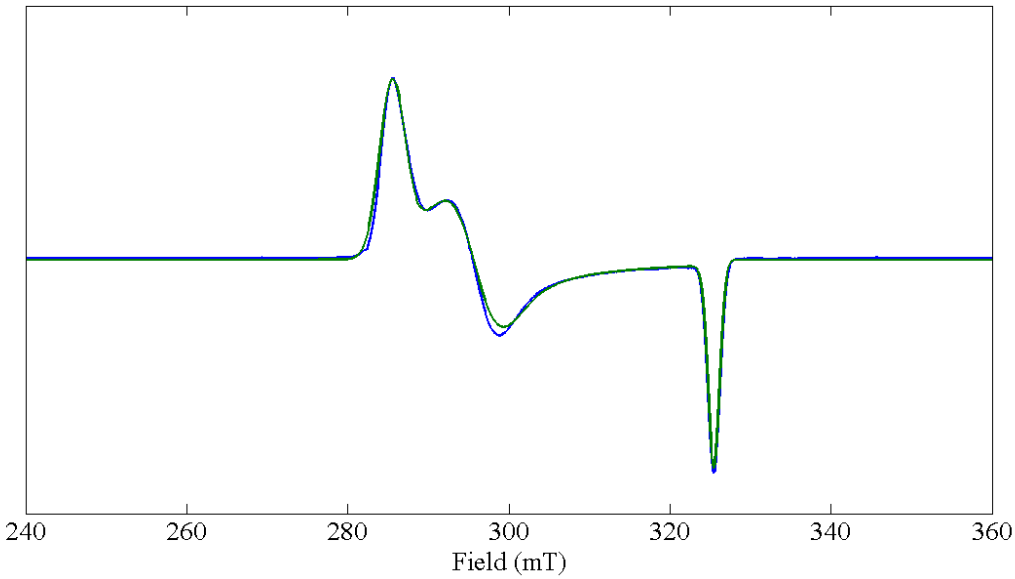


Figure S2: X-band CW EPR spectrum of **4a** (blue) in toluene at 7K and the corresponding simulation (green). The g-values from the simulation are g_1 = 2.328, g_2 = 2.242 and g_3 = 2.085.

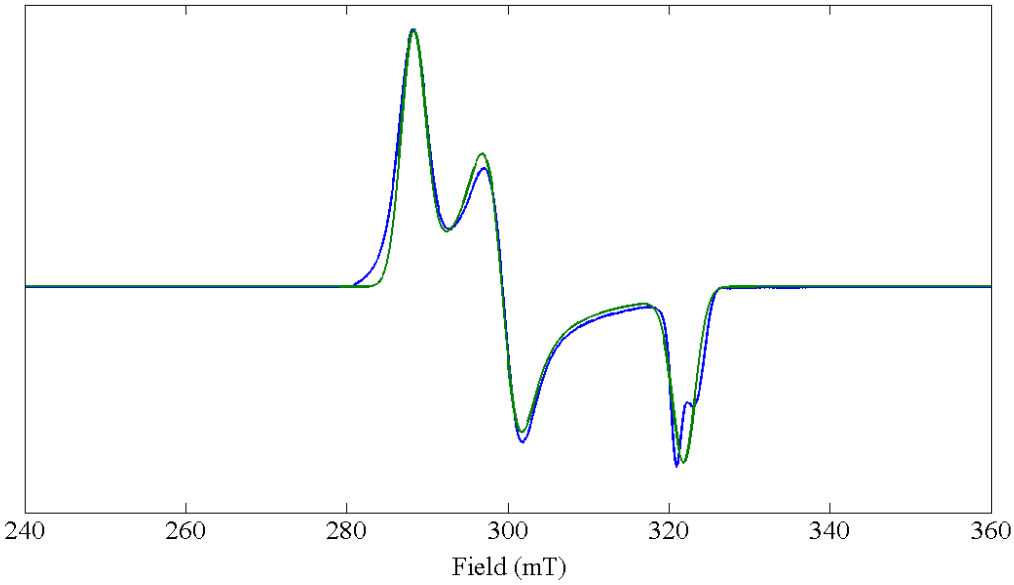


Figure S3: X-band CW EPR spectrum of **4b** (blue) in toluene at 7K and the corresponding simulation (green). The g-values from the simulation are g_1 = 2.340, g_2 = 2.207 and g_3 = 2.072.

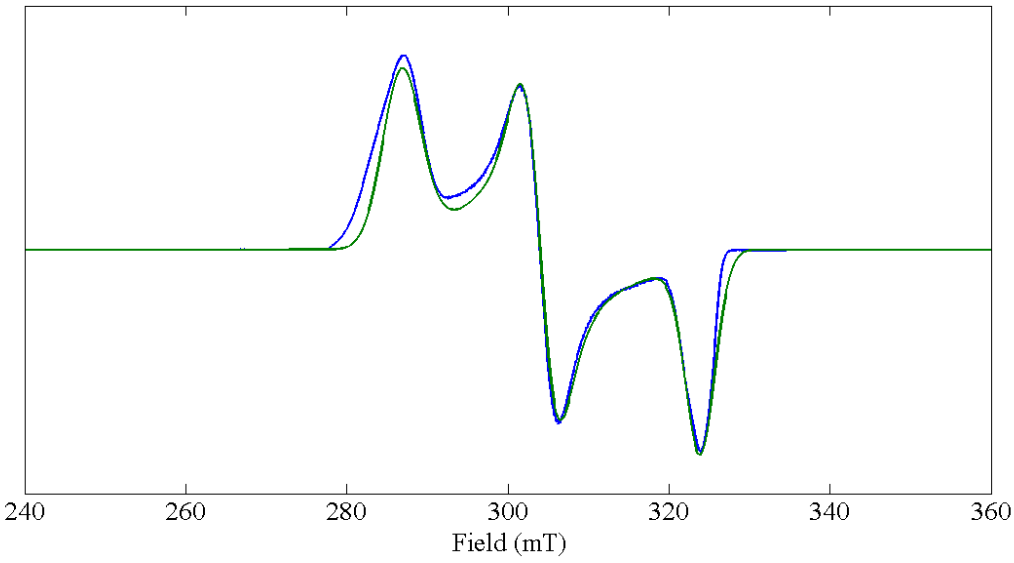


Figure S4: Overlay of X-band CW EPR spectra of **3a** (IPrNiCp, black), **3b** (SIPrNiCp, blue), **4a** (IPrNiInd, red) and **4b** (SIPrNiInd, magenta) in toluene at 7K.

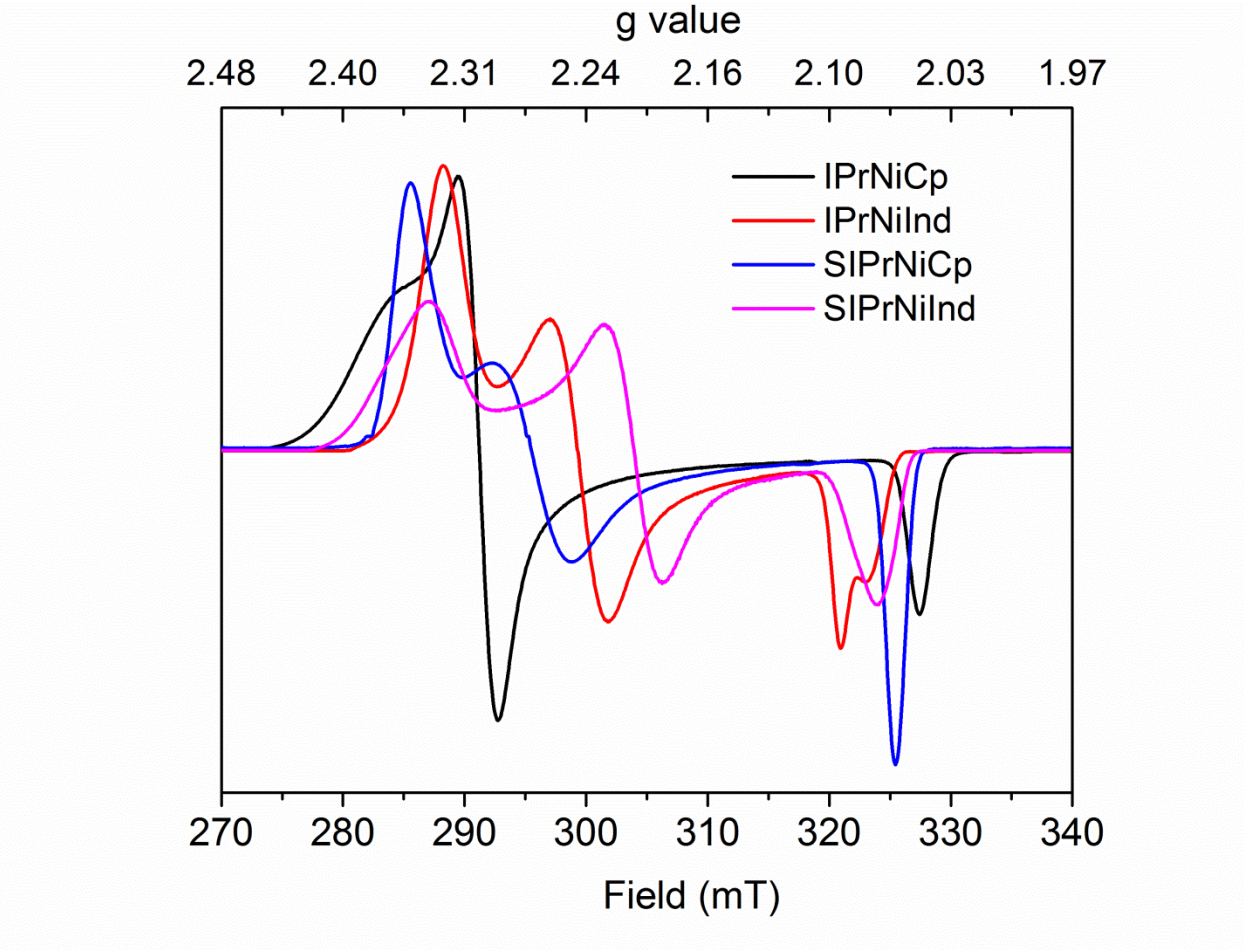


Table S2: Simulated g-values for the EPR spectra of **3a**, **3b**, **4a** and **4b**.

Compound/g-value	g_1	g_2	g_3
3a	2.362	2.306	2.049
3b	2.349	2.267	2.062
4a	2.328	2.242	2.085
4b	2.340	2.207	2.072

Procedure for SQUID Experiments and Data of Selected Compounds

The magnetic susceptibility measurements were obtained using a Quantum Design SQUID magnetometer MPMS operating between 2 and 300 K for dc-applied fields at 0.1 T. Dc analyses were performed on polycrystalline samples of 39.8, 12.7, 38.9 and 14.9 mg for **3a**, **3b**, **4a** and **4b,** respectively, wrapped in a polyethylenemembrane.

DC susceptibility measurements

The measured molar magnetic susceptibility of **3a**-**4b** in a field of 0.1 T are plotted as a function of temperature in Figure S5. At room temperature, the χT product is 0.45, 0.47, 0.51 and 0.55 cm^3K/mol for **3a**, **3b**, **4a** and **4b**, respectively. The theoretical χT value for S=1/2 is 0.375 cm^3K/mol and for S=1, 1 cm^3K/mol if g is equal to 2. All measured χT values are close to the S=1/2 value which is consistent with a one unpaired electron system. The χT values can also be converted by

$\mu_{eff} = \sqrt{8\chi_M \mathrm{T}}$ (no units)

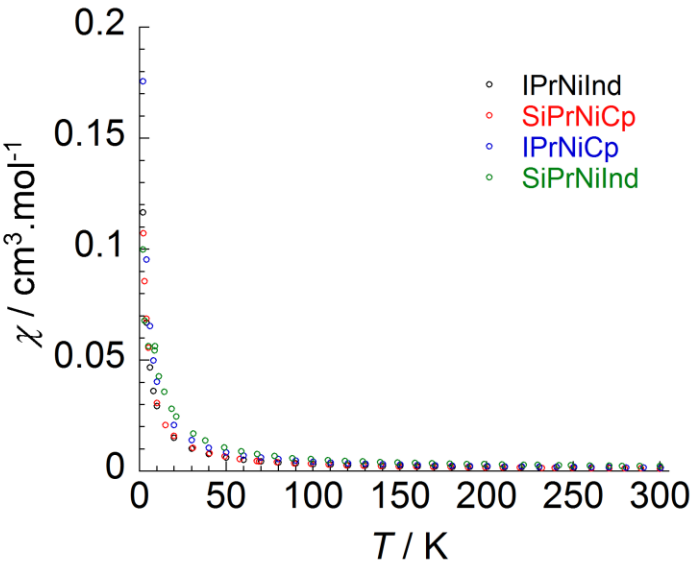
At room temperature, the μ_{eff} values are 1.90, 1.93, 2.0 and 2.09 for **3a**, **3b**, **4a** and **4b**, respectively. Theoretical values, μ_{eff} of S=1/2 is 1.732 and S=1 is 2.828.

The g values can be calculated from the equation:

$$\chi T = \frac{\mathrm{g}^2 * \mathrm{S}(\mathrm{S}+1)}{8}$$

At room temperature, the χT values correspond to g = 2.19, 2.24, 2.33 and 2.42 for **3a**, **3b**, **4a** and **4b**, respectively, when S = 1/2.

Figure S5: Temperature dependence of the χ products at 0.1 T for **3a** (IPrNiCp, blue), **3b** (SIPrNiCp, red), **4a** (IPrNiInd, black) and **4b** (SIPrNiInd, green) (with $\chi = M/H$ normalized per mol).



For mononuclear paramagnetic metal systems, the χT vs T plot should be a horizontal straight line which obeys the Curie Law (Figures S6 and S7). However, the χT values of all complexes decreased slightly with the decrease in temperature. The obvious reason for the decreasing χT values may be a small diamagnetic contribution since the χT value of S = 1/2 is very small; however, this effect is normally considered negligible in high spin systems. The χT values decreased to reach 0.35 (1.67), 0.20 (1.25), 0.23 (1.36) and 0.24 (1.38) cm^3K/mol (μ_{eff}) for **3a**, **3b**, **4a** and **4b**, respectively, at 2.0 K due to the weak intermolecular antiferromagnetic interaction.

Figure S6: Temperature dependence of the χT products at 0.1 T for **3a** (IPrNiCp, red), **3b** (SIPrNiCp, blue), **4a** (IPrNiInd, black) and **4b** (SIPrNiInd, green) (with $\chi = M/H$ normalized per mol).

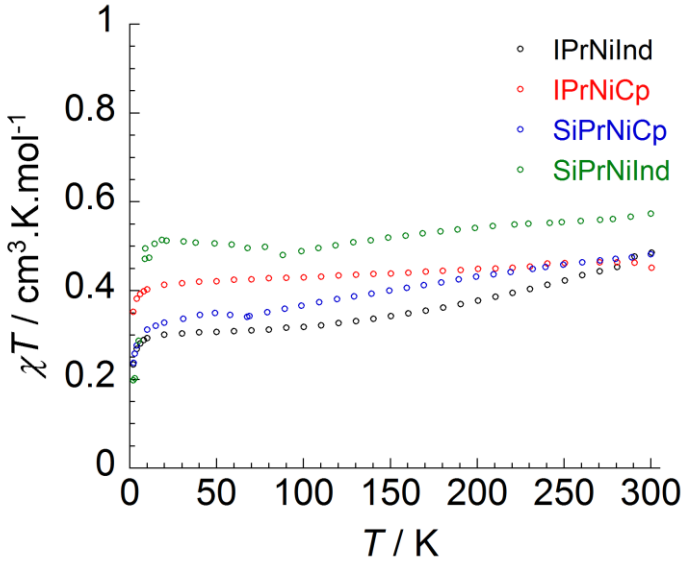
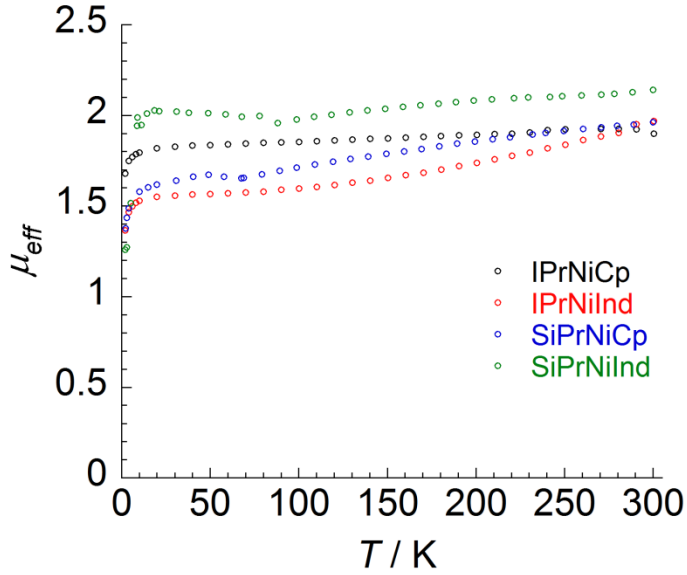


Figure S7: Temperature dependence of μ_{eff} at 0.1 T for **3a** (IPrNiCp, black), **3b** (SIPrNiCp, blue), **4a** (IPrNiInd, red) and **4b** (SIPrNiInd, green) (with $\chi = M/H$ normalized per mol).



Procedure for Electrochemistry and CVs of Selected Compounds

Electrochemistry voltammetric data were collected using an air tight three-electrode system, which was assembled in a nitrogen filled glovebox. The working electrode was a 2 mm diameter platinum electrode. The reference and counter electrodes were 0.8 mm platinum wires. The electrolyte was 0.10 M $^{n}Bu_4NPF_6$ in THF, which was synthesized by the metathesis of Bu_4NBr and HPF_6, recrystallized from hot ethanol, and dried under vacuum overnight. The THF used in the experiment was HPLC grade and dried before use. Ferrocene was used as internal standard. Cyclic voltammetry data were measured with Princeton Applied Research VersaSTAT 4 potentiostatic instrumentation.

Table S3: Tabulated cyclic voltammetry data of 1.6 mM Ni complexes (**3a**, **3b**, **4a** and **4b**) in 0.1 M $^{n}Bu_4NPF_4$ in THF at a Pt working electrode.

	3a	**3b**	**4a**	**4b**
Oxidation Potential vs. Fc^{+}/Fc	-0.66 V	-0.75 V	-0.41 V	-0.64 V
Reduction Potential vs. Fc^{+}/Fc	-2.57 V	-2.73 V	-2.27 V	-2.30 V

Figure S8: CV of **3a** (1.6 mM) at reductive potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

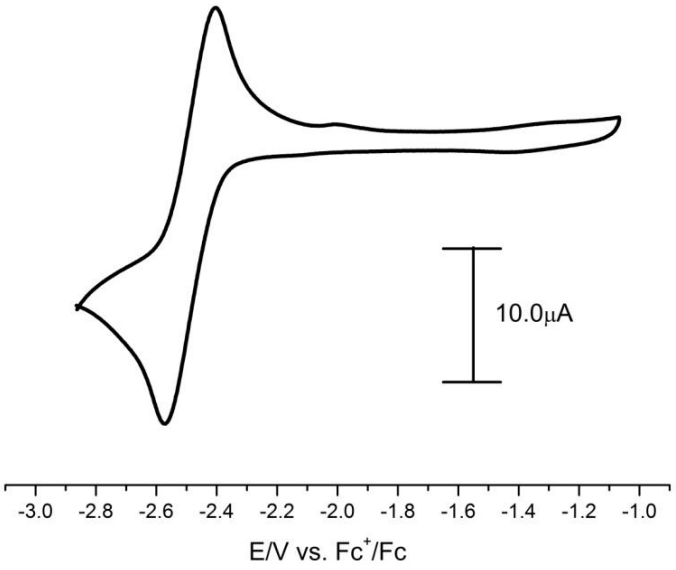


Figure S9: CV of **3b** (1.6 mM) at reductive potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

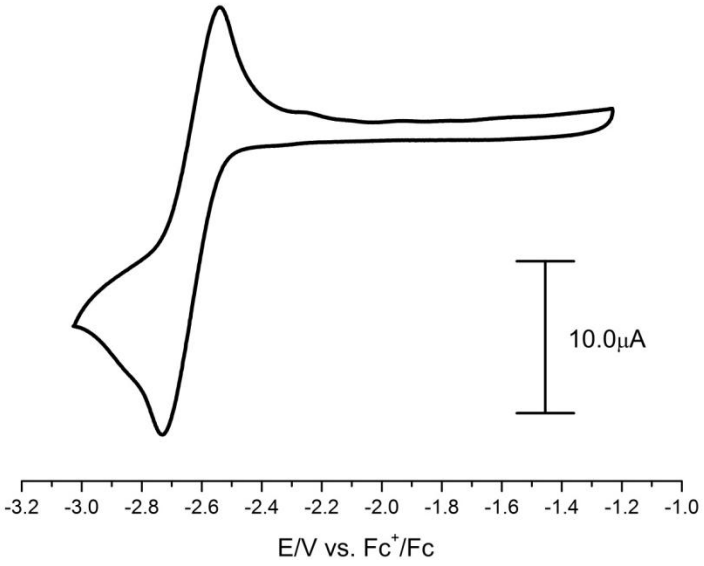


Figure S10: CV of **4a** (1.6 mM) at reductive potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

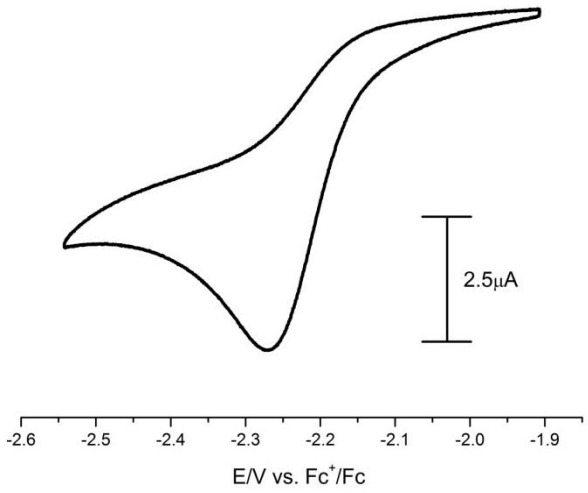


Figure S11: CV of **4b** (1.6 mM) at reductive potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

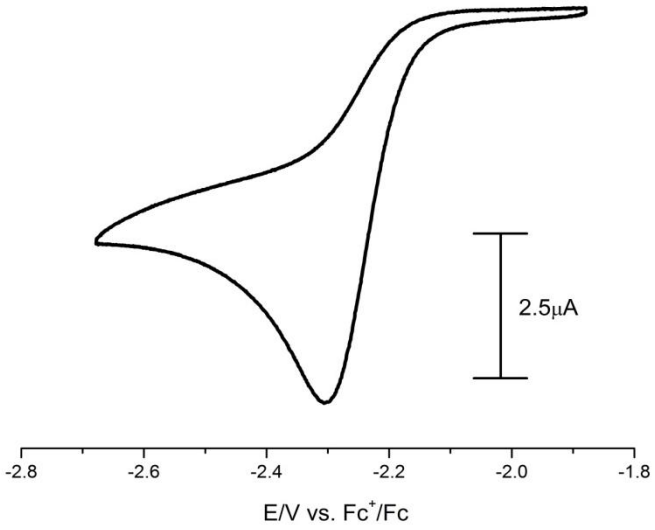


Figure S12: CV of **3a** (1.6 mM) at oxidative potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

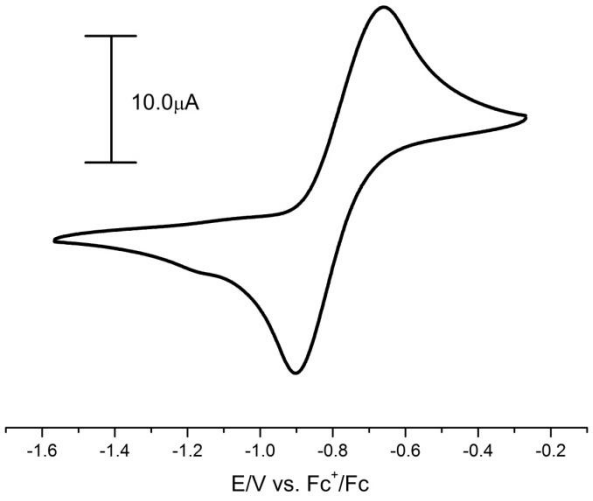


Figure S13: CV of **3b** (1.6 mM) at oxidative potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

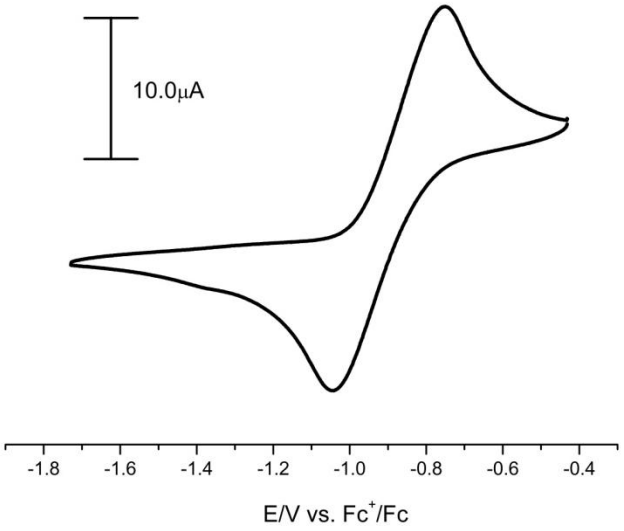


Figure S14: CV of **4a** (1.6 mM) at oxidative potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.

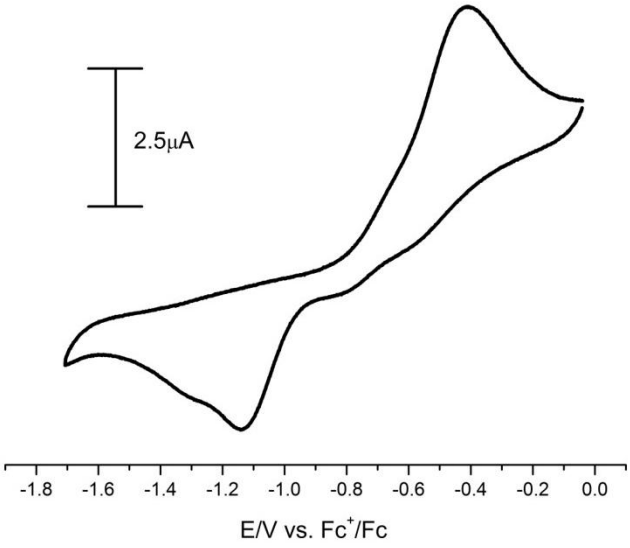
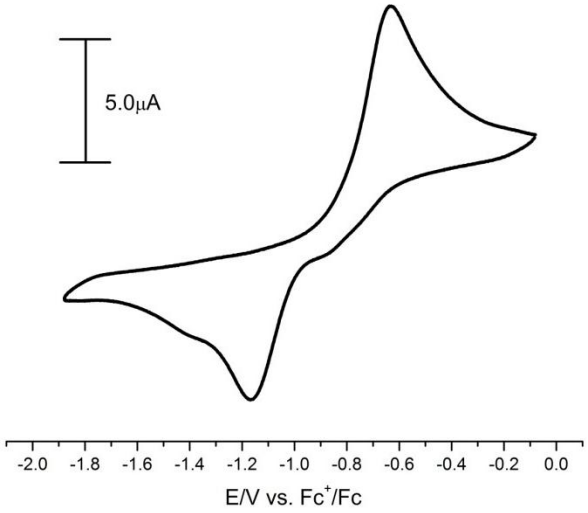


Figure S15: CV of **4b** (1.6 mM) at oxidative potentials in a 0.10 M $^{n}Bu_4NPF_6$ solution of THF under N_2 at room temperature. The working electrode was a 2.0 mm diameter platinum disk, and the scan rate was 0.50 V s^{-1}.



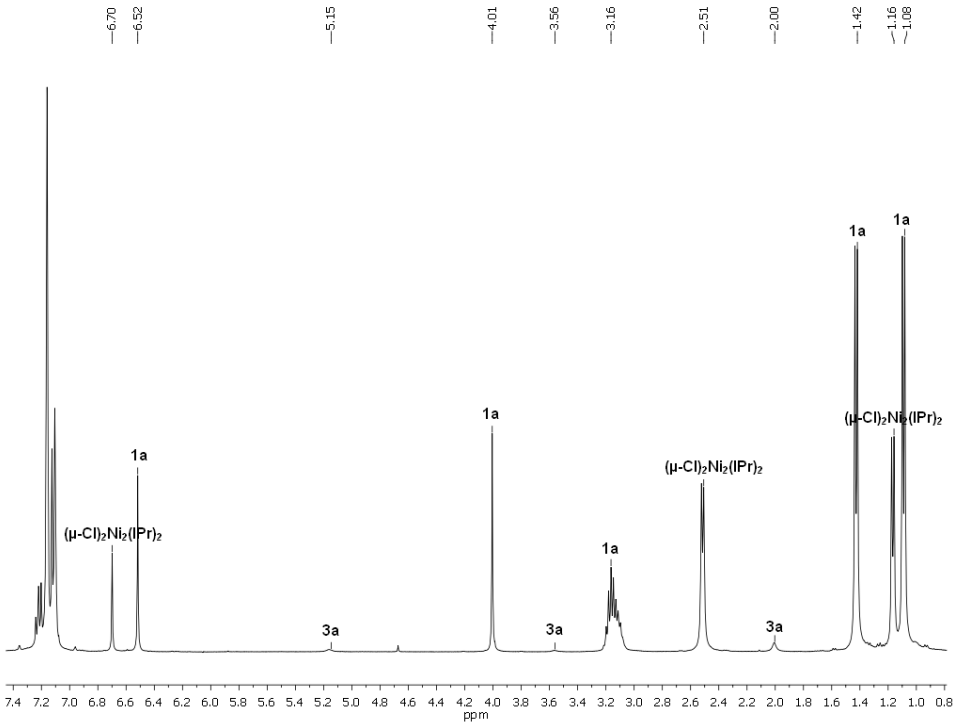
Reactions Relating to Equilibrium Between Ni(I) Monomers and Dimers

*Addition of **3a** to an equilibrated mixture of **1a***

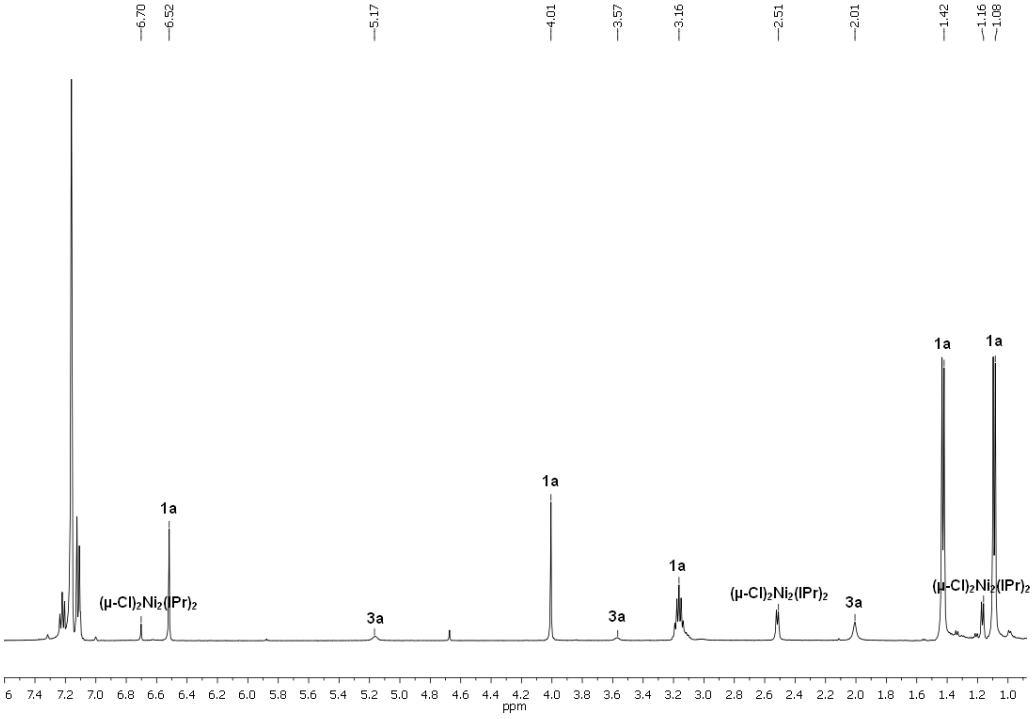
To a solution of **1a** (6.5 mg, 0.0065 mmol) in 0.5 mL C_6D_6 in a J. Young NMR tube, **3a** (5.3 mg, 0.01 mmol) was added. After 10 minutes ^{1}H NMR spectroscopy showed that the reaction equilibrium had shifted to **1a** and **3a**. The NMR spectra are shown in Figure S16.

Figure S16: a) ^{1}H NMR spectrum of an equilibrated mixture of **1a**; **b)** ^{1}H NMR spectrum of equilibrated mixture of **1a** after addition of **3a**.

a)



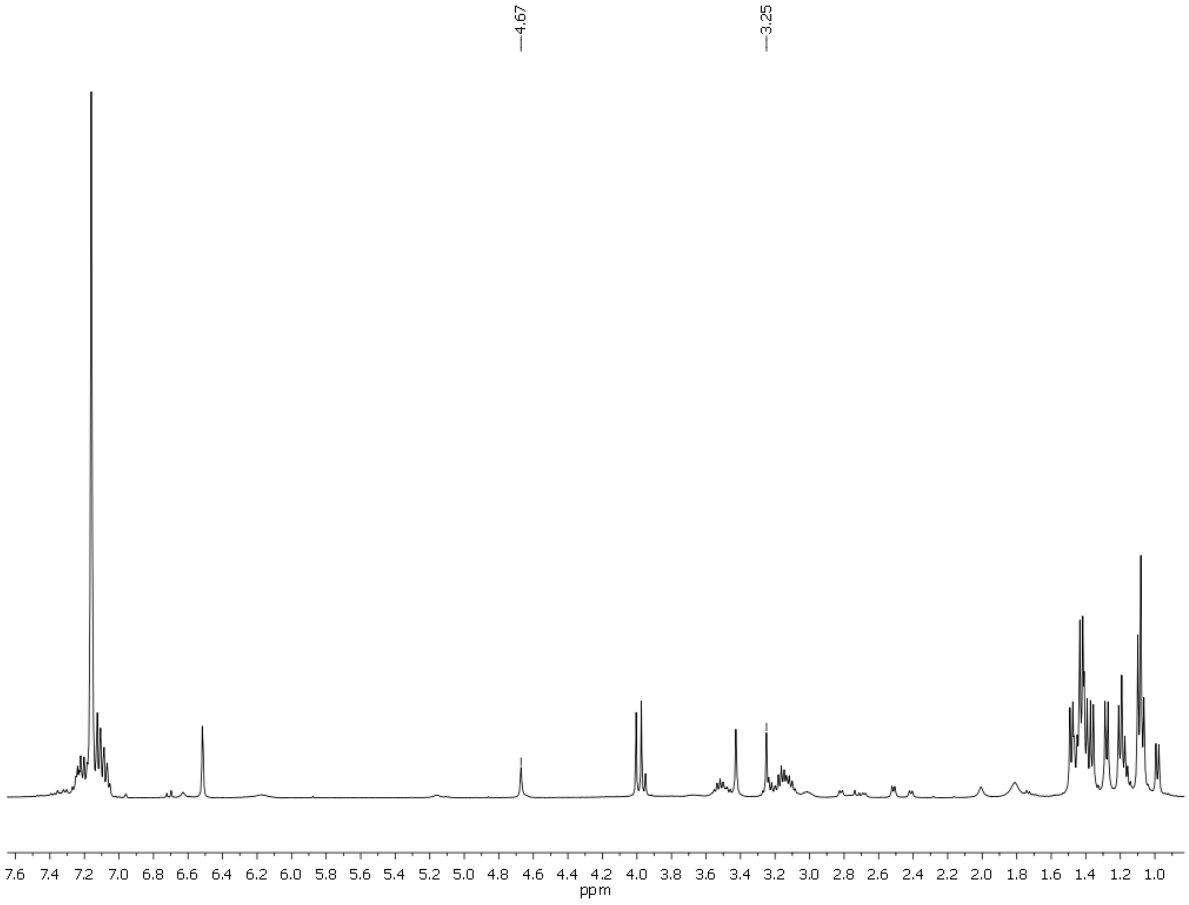
b)



*Reaction of **1a** and **1b***

To a solution of **1a** (6.5 mg, 0.0065 mmol) in 0.5 mL C_6D_6, **1b** (6.5 mg, 0.0065 mmol) was added. ^{1}H NMR spectroscopy indicated that the solution is a mixture of **1a**, **1b**, **3a**, **3b**, (μ-Cl)$_2$Ni$_2$(SIPr)$_2$, (μ-Cl)$_2$Ni$_2$(IPr)$_2$ and a new product which has been assigned as **(μ-Cp)(μ-Cl)Ni$_2$(IPr)(SIPr)**. Although many peaks are overlapping in the spectrum, we believe that the new peaks at 4.67 (s) for Cp and 3.25 (s) for the saturated backbone peak of SIPr are consistent with the formation of **(μ-Cp)(μ-Cl)Ni$_2$(IPr)(SIPr)**. The ^{1}H NMR spectrum of the mixture is shown in Figure S17.

Figure S17: ^{1}H NMR spectrum of reaction between **1a** and **1b**.



Reaction of (μ-Cl)$_2$Ni$_2$(SIPr)$_2$ and (μ-Cl)$_2$Ni$_2$(IPr)$_2$

To a solution of (μ-Cl)$_2$Ni$_2$(IPr)$_2$ (7.6 mg, 0.0078 mmol) in 0.5 mL C_6D_6, (μ-Cl)$_2$Ni$_2$(SIPr)$_2$ (7.6 mg, 0.0078 mmol) was added. After 20 minutes ^{1}H NMR spectroscopy showed a mixture of (μ-Cl)$_2$Ni$_2$(SIPr)$_2$, (μ-Cl)$_2$Ni$_2$(IPr)$_2$ and **(μ-Cl)$_2$Ni$_2$(IPr)(SIPr)**. The ^{1}H NMR spectrum of the mixture is shown in Figure S18.

^{1}H NMR (400 MHz, C_6D_6) for **(μ-Cl)$_2$Ni$_2$(IPr)(SIPr)**: 7.11 (m, 12H) (Ph from IPr and SIPr), 6.72 (s, 2H, C*H*=C*H* from IPr), 3.66 (m, 4H, C*H*Me$_2$ from SIPr overlapping with (μ-Cl)$_2$Ni$_2$(SIPr)$_2$), 3.11 (m, 4H, C*H*Me$_2$ from IPr overlapping with (μ-Cl)$_2$Ni$_2$(IPr)$_2$), 2.74 (s, 4H, CH_2-CH_2 from SIPr), 2.82 (d, *J* = 8 Hz, 12H, Me), 2.41 (d, *J* = 8 Hz, 12H, Me), 1.29 (d, *J* = 8 Hz, 12H, Me), 1.15 (d, *J* = 8 Hz, 12H, Me) (CH_3 from IPr and SIPr).

Figure S18: ^{1}H NMR spectrum of reaction between (μ-Cl)$_2$Ni$_2$(SIPr)$_2$ and (μ-Cl)$_2$Ni$_2$(IPr)$_2$.

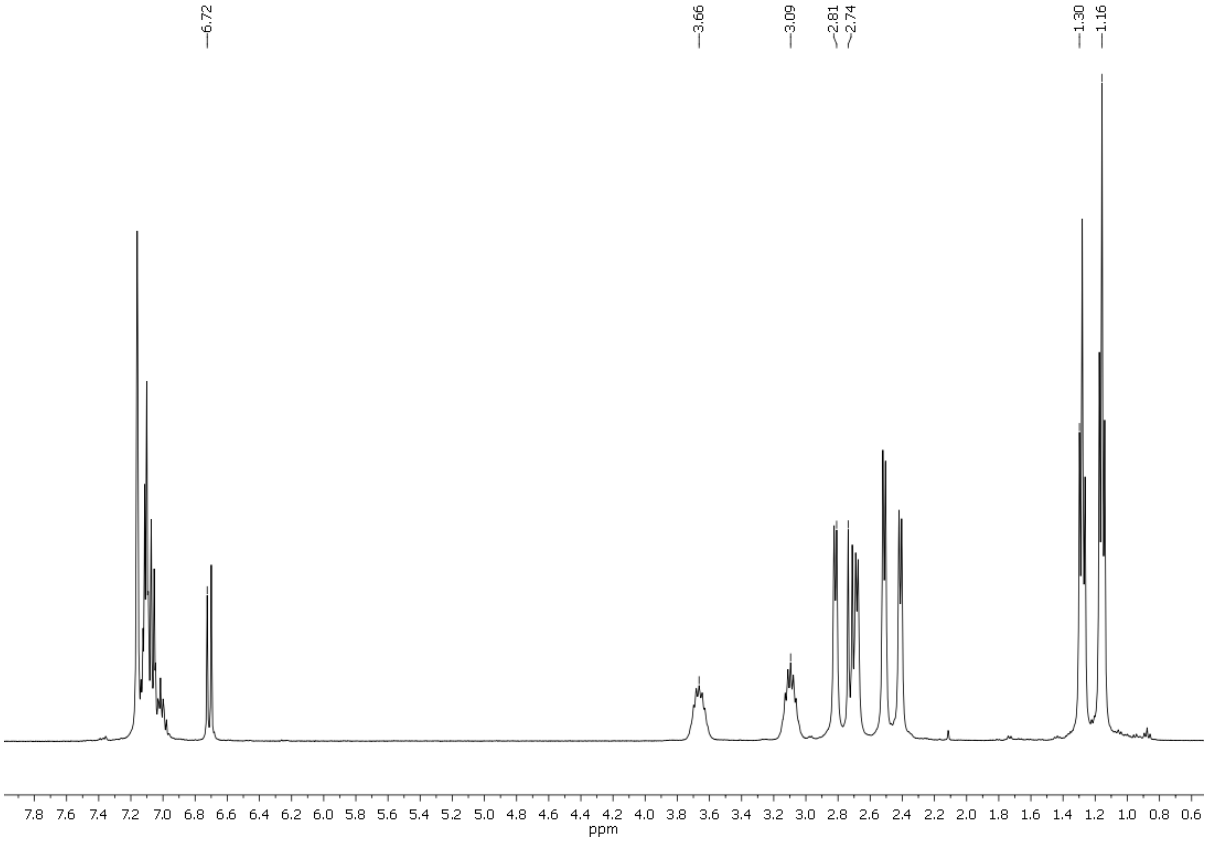


Figure S19: EPR spectrum $(\mu\text{-Cl})_2Ni_2(IPr)_2$ which shows presence of S = ½ compound.

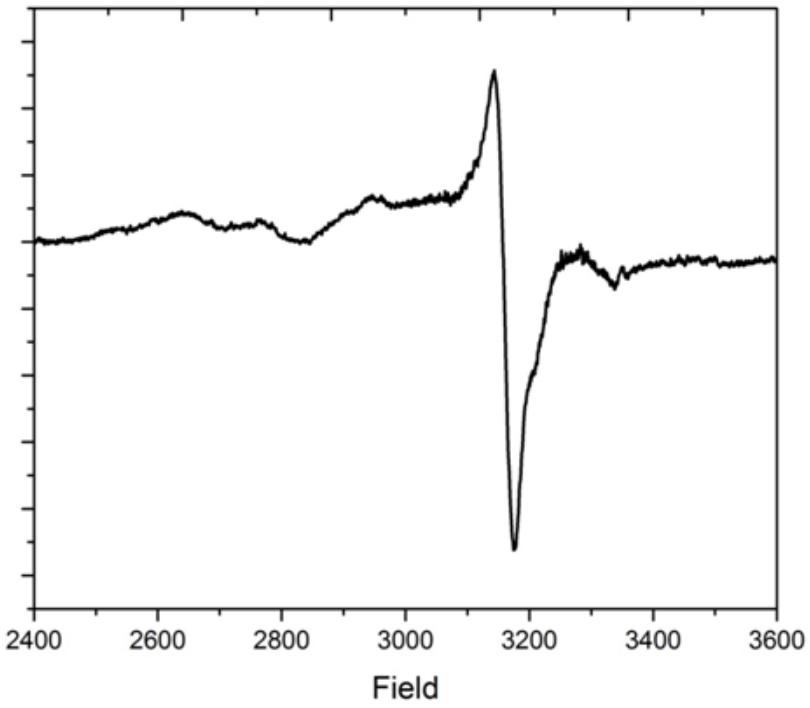
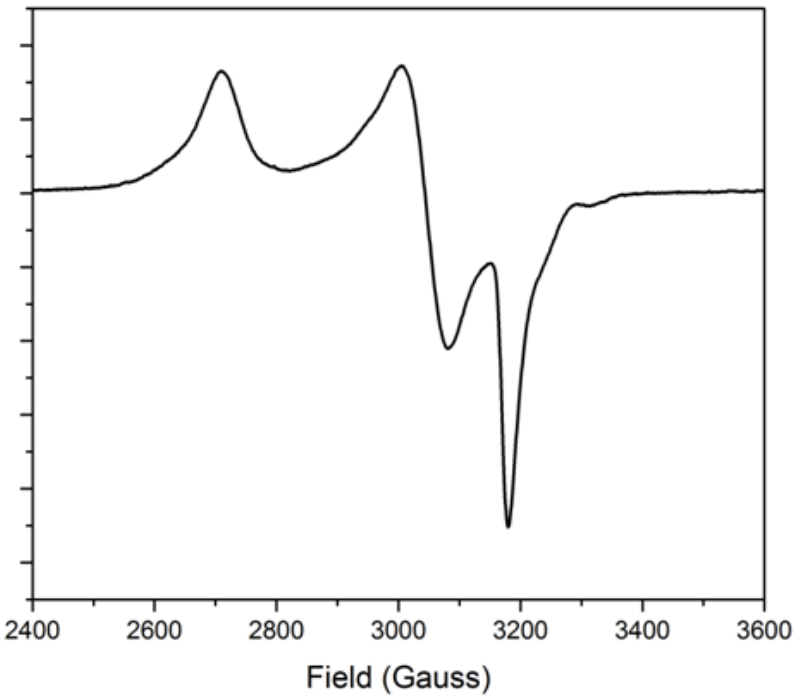


Figure S20: EPR spectrum $(\mu\text{-Cl})_2Ni_2(SIPr)_2$ which shows presence of S = ½ compound.



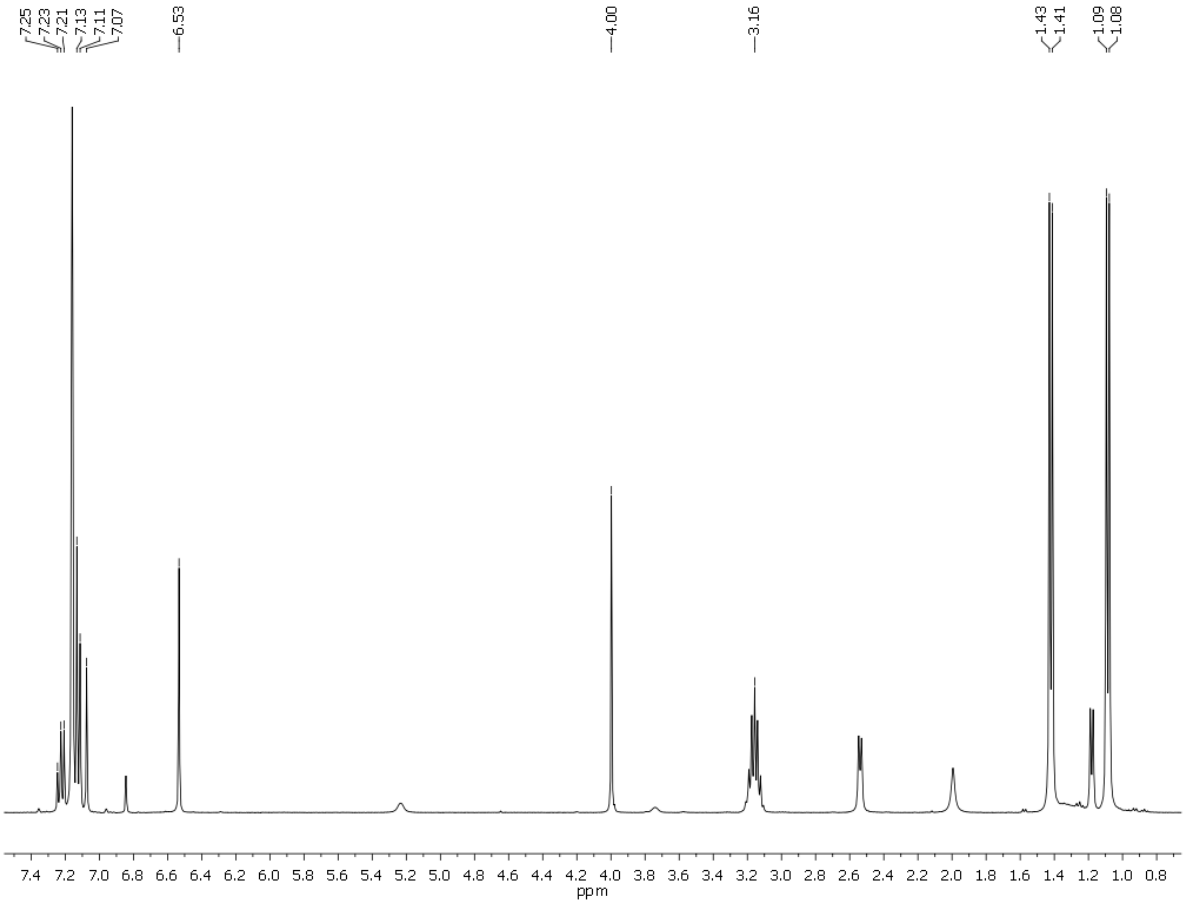
*Reaction of **4a** and $(\mu\text{-Cl})_2Ni_2(IPr)_2$*

To a solution of **4a** (7.5 mg, 0.014 mmol) in 0.5 mL C_6D_6, $(\mu\text{-Cl})_2Ni_2(IPr)_2$ (6.8 mg, 0.007 mmol) was added. Upon addition, the yellow solution turned green. After 20 minutes ^{1}H NMR spectroscopy indicated quantitative conversion to **2a**.

*Reaction of **3a** and $(\mu\text{-Cl})_2Ni_2(IPr)_2$*

To a solution of **3a** (7.3 mg, 0.014 mmol) in 0.5 mL C_6D_6, $(\mu\text{-Cl})_2Ni_2(IPr)_2$ (6.8 mg, 0.007 mmol) was added. After 20 minutes, ^{1}H NMR spectroscopy indicated a mixture of **1a** with small amount of **3a** and $(\mu\text{-Cl})_2Ni_2(IPr)_2$, as shown in Figure S21.

Figure S21: ^{1}H NMR spectrum reaction between **3a** and $(\mu\text{-Cl})_2Ni_2(IPr)_2$.



*Reaction of **1a** and **4a***

To a solution of **1a** (7.4 mg, 0.074 mmol) in 0.5 mL C_6D_6, **4a** (4.2 mg, 0.0074 mmol) was added. After 48 hours, ^{1}H NMR spectroscopy indicated full conversion to **2a** and **3a**.

Figure S22: EPR spectrum **1a** which shows presence of **3a**.

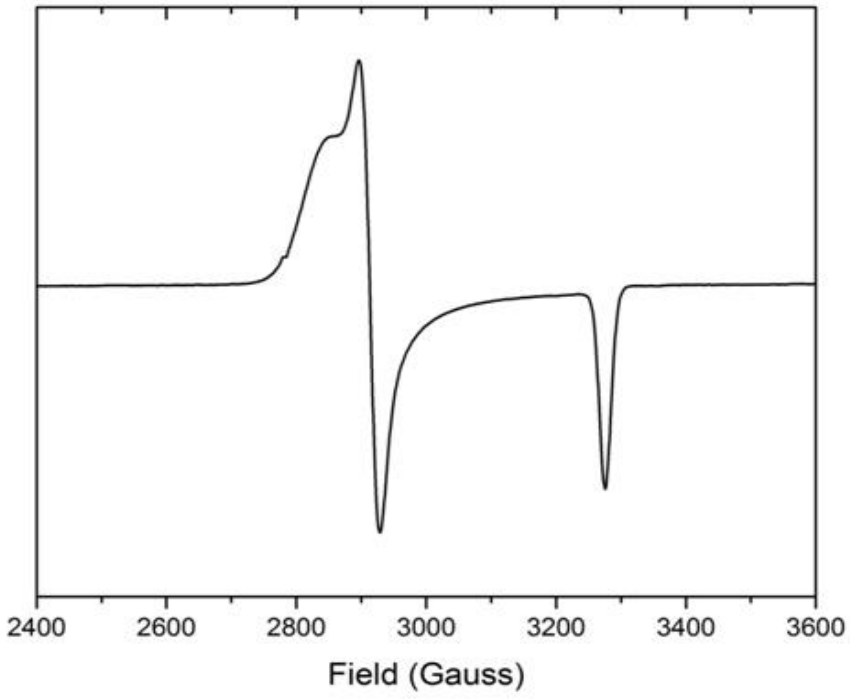


Figure S23: EPR spectrum **1b** which shows presence of **3b**.

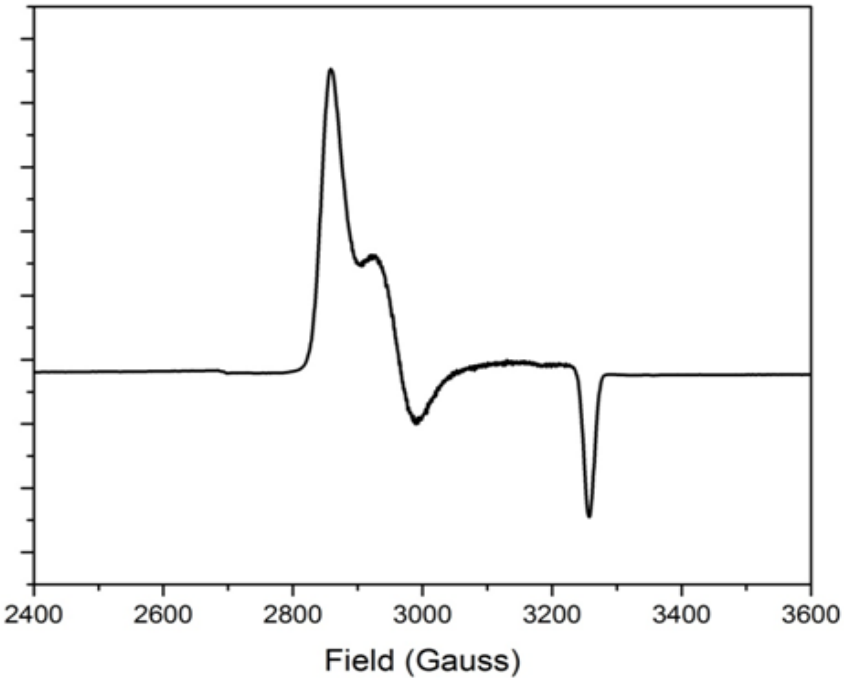


Figure S24: EPR spectra for **2a**, which shows no S = ½ impurities.

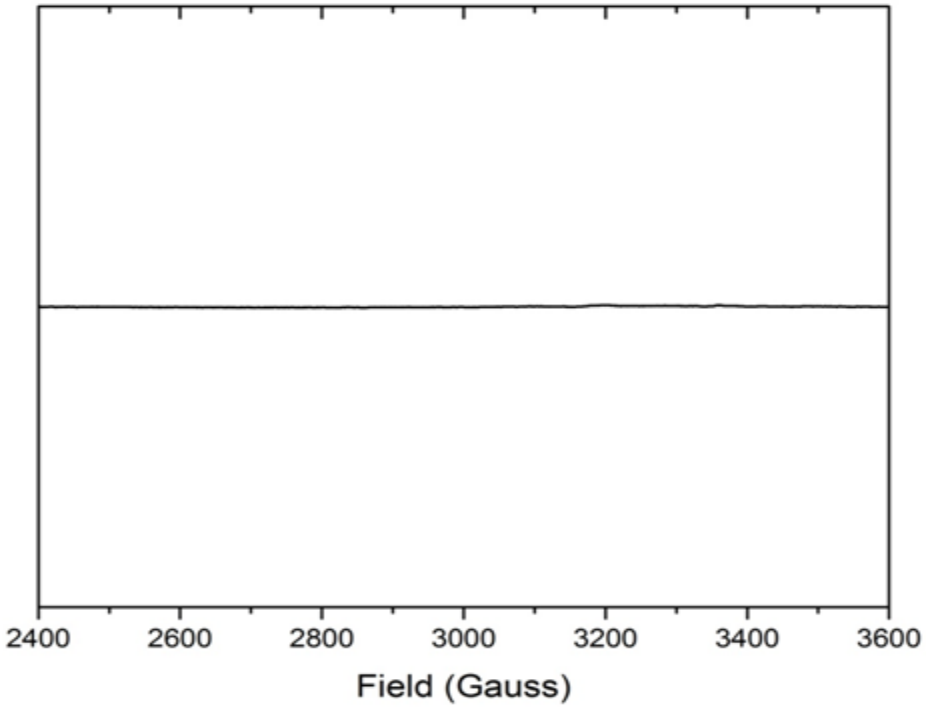
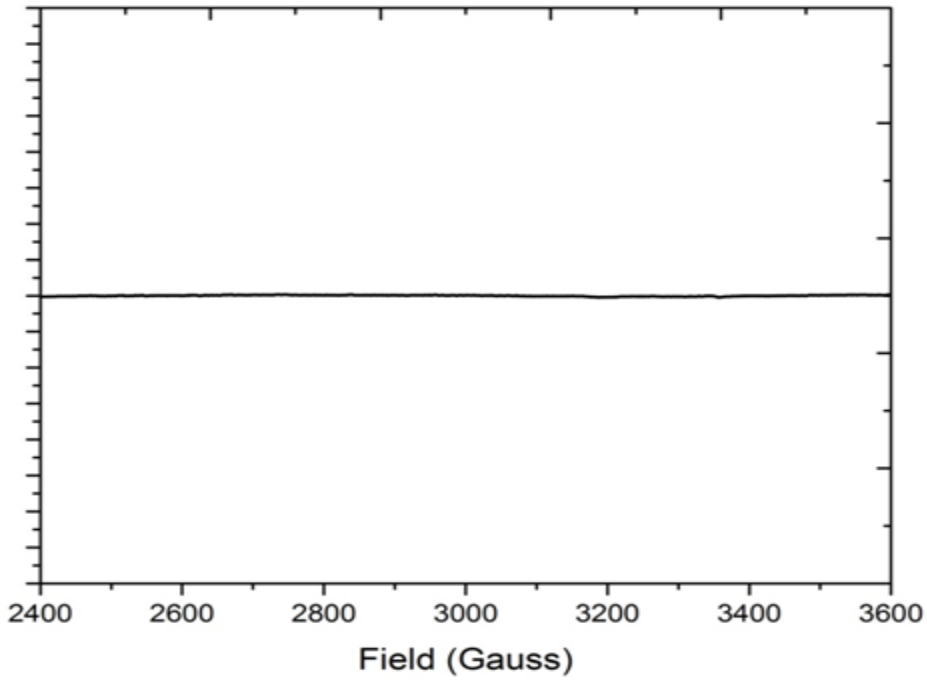


Figure S25: EPR spectra for **2b**, which shows no S = ½ impurities.



*Reaction of **1a** and LiInd*

To a solution of **1a** (23 mg, 0.024 mmol) in 1 mL THF, a solution of LiInd (1.4 mg, 0.012 mmol) in 1 mL THF was added dropwise. After 1 hour, the volatiles were removed under vacuum. The resulting residue was dissolved in 0.8 mL C_6D_6. ^{1}H NMR spectroscopy indicated quantitative conversion to **2a** and **3a**.

*Reaction of **1a** and NaCp*

To a solution of **1a** (35 mg, 0.035 mmol) in 1 mL THF, NaCp (3.1 mg, 0.035 mmol) was added. After 4 hours, the volatiles were removed under vacuum. ^{1}H NMR spectroscopy of the resulting residue in C_6D_6 indicated that only **3a** was present. The residue was extracted with benzene and the solvent removed under vacuum to provide yellow **3a**. Yield: 33 mg (92%).

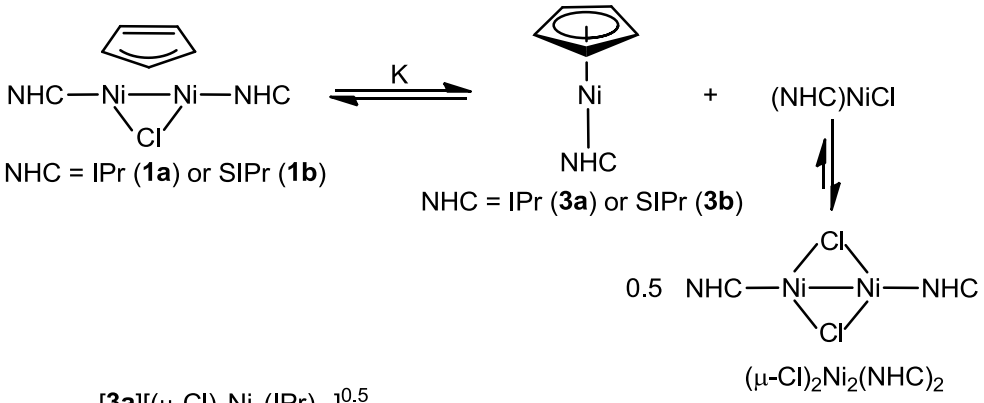
*Reaction of **1b** and NaCp*

To a solution of **1b** (35 mg, 0.035 mmol) in 1 mL THF, NaCp (3.1 mg, 0.035 mmol) was added. After 4 hours, the volatiles were removed under vacuum. The residue was extracted with benzene and the solvent removed under vacuum to provide yellow **3b**. Yield: 31 mg (90%).

General procedure for Van't Hoff analysis

1a (7.3 mg, 0.0073 mmol) or **1b** (5.6 mg, 0.0056 mmol) was dissolved in 0.5 mL C_6D_6 in a J. Young NMR tube. The sample was placed into a 400 MHz NMR spectrometer. ^{1}H NMR spectra were recorded at the following temperatures: 25, 30, 35, 40, 45, 50, 55, 60, 65, 70ºC. The methyl peaks of the NHC ligand in **1a** (or **1b**) and $(\mu\text{-Cl})_2Ni_2(NHC)_2$ were integrated to determine the concentration of the corresponding complexes in the mixture. The concentration of the paramagnetic species **3a** or **3b** was determined from the concentration of $(\mu\text{-Cl})_2Ni_2(NHC)_2$.

Table S4: Experimentally determined equilibrium constants for **1a** and **1b**.



$$K_a = \frac{[\mathbf{3a}][(\mu\text{-Cl})_2Ni_2(IPr)_2]^{0.5}}{[\mathbf{1a}]}$$

$$K_b = \frac{[\mathbf{3b}][(\mu\text{-Cl})_2Ni_2(SIPr)_2]^{0.5}}{[\mathbf{1b}]}$$

Temperature(Kelvin)	1/Temperature	K_a	K_b
298.24	0.003353	0.006021	0.03739
302.28	0.003308	0.009043	0.051617
309.35	0.003233	0.012343	0.069071
314.4	0.003181	0.015863	0.091171
318.44	0.00314	0.022114	0.120831
324.50	0.003082	0.028736	0.153849
328.54	0.003044	0.037052	0.190134
334.60	0.002989	0.04714	0.236962
339.65	0.002944	0.06056	0.294398
344.70	0.002901	0.080575	0.359495

Figure S26: Van't Hoff Plot for **1a**

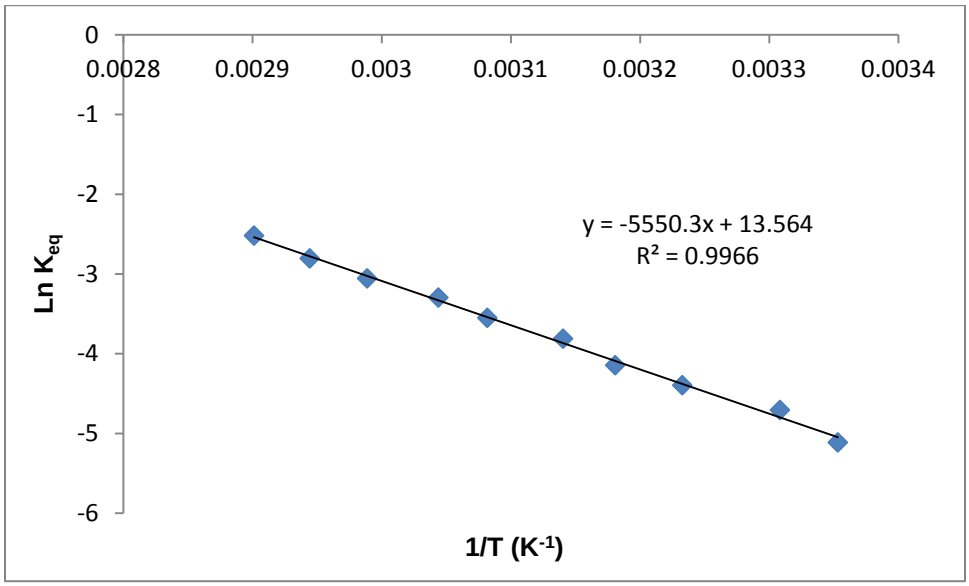
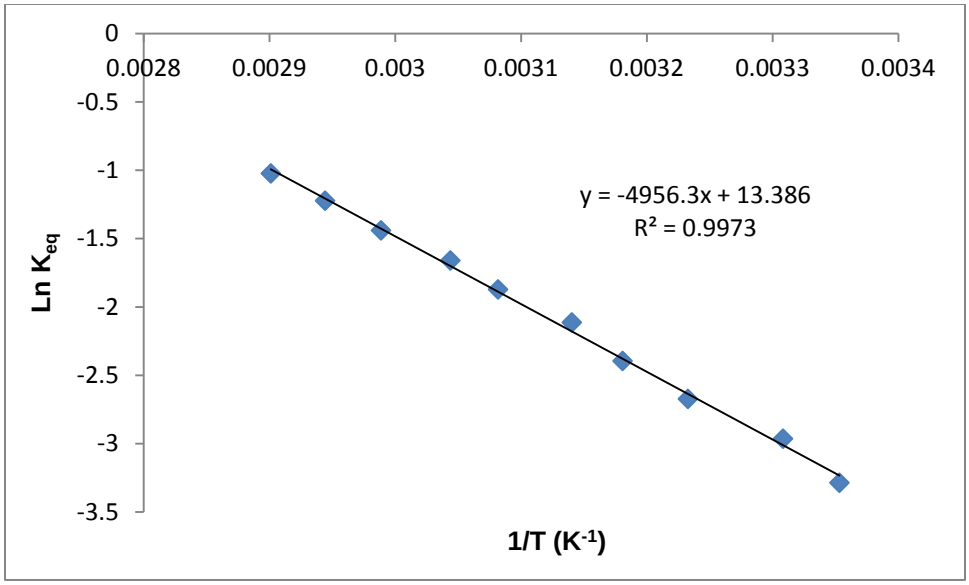


Figure S27: Van't Hoff Plot for **1b**.



DFT Calculations

NBO analysis details

NBO calculations (version 6.0) were carried out with Gaussian09 (revision D.01) on the optimized geometries of **3a** and **4a**. In order to simplify the analysis, the 2,6-diisopropylphenyl substituents on the NHC ligand (IPr) were replaced by hydrogens. The N-H distance of these models was obtained by geometry optimization keeping the rest of the molecule frozen. In addition, 3c-2e interactions were excluded between the Cp/indenyl ligands and Ni, in order to compare only the donor-acceptor interactions with these two ligands. The π(Cp/indenyl) → *d* (Ni) interactions were identified in the β-electron configuration (both **3a** and **4a** have a doublet spin ground state) as donations from π(C=C) and LP(C) (LP = Lone Pair) orbitals to the Ni *d* orbital with a single-electron vacancy. The π(Cp/indenyl) → σ*(Ni-C) interactions were found both in the α and β-electron configurations and involve electron-donation from π(C=C) and LP(C) orbitals to the antibonding σ*(Ni-C) orbital. In the second order perturbation analysis, the π(Cp/indenyl) → *d* (Ni) and π(Cp/indenyl) → σ*(Ni-C) interactions yield several natural localized molecular orbitals (NLMOs). Those shown in Figures 9 and S31 were selected as the most representative examples. The stabilization energies given in the manuscript are the summation of all components contributing to these interactions in the β-electron configuration.

Additional figures

Figure S28: Spin densities of complexes **3b** (left) and **4b** (right).

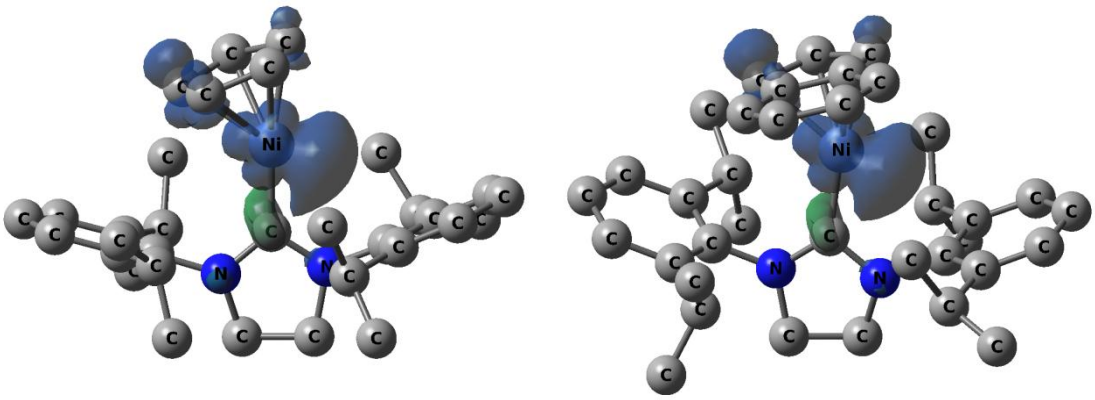


Figure S29: SOMO (left) and spin density (right) of complex **3a**.

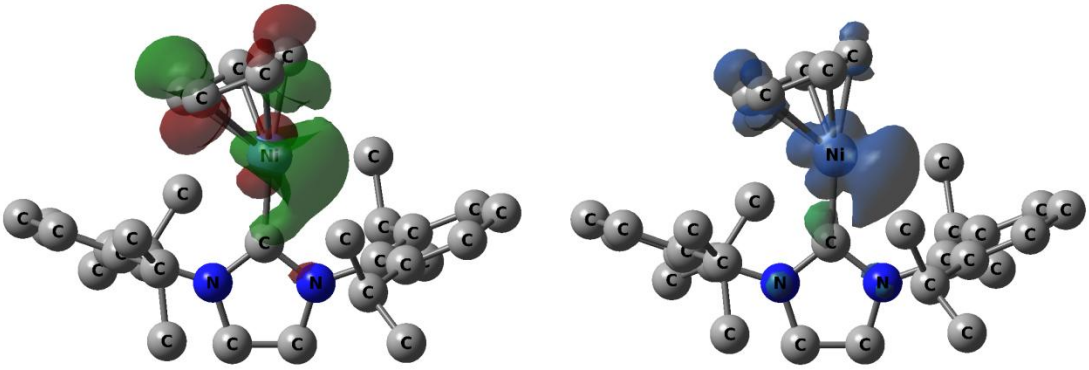


Figure S30: Optimized geometry of the Ni(II) species derived from the one-electron oxidation of complex **3a**.

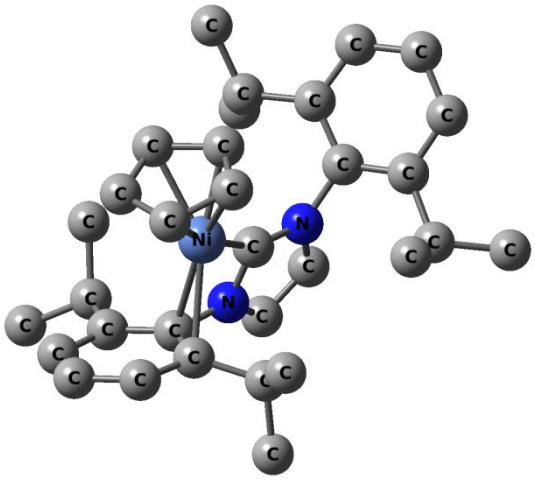


Figure S31: Natural localized molecular orbitals (NLMOs) for the π(L) → σ*(Ni-C) donation. L = Cp (left) or indenyl (right). The substituents on the NHC ligand (IPr) were removed for clarity.

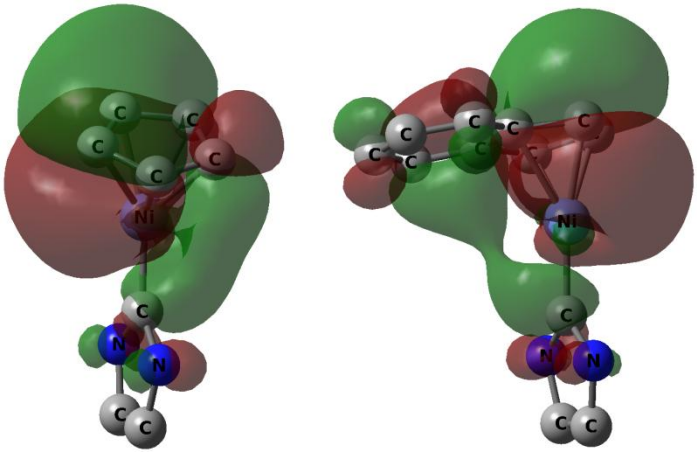
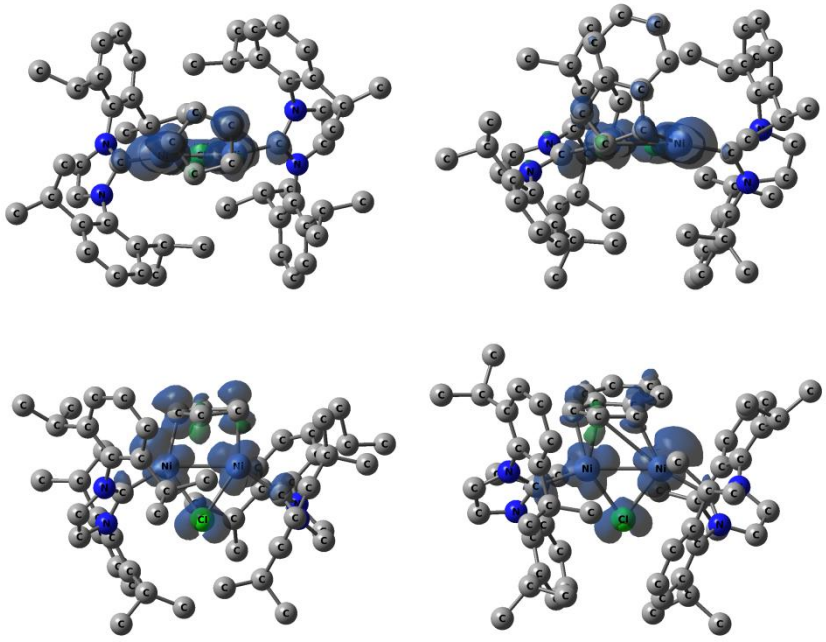


Figure S32: Top (top) and side (bottom) views of the spin densities of the dinuclear complexes **1a** (left) and **2a** (right).



Results with other DFT functionals

In addition to M06L, other pure and hybrid DFT functionals were tested for determining the ground electronic state of **1a**. These calculations showed that the nature of the singlet ground state depends on the functional used (Table S5). With the pure functionals, M06L and BP86* (* = D3 Grimme's corrections for dispersion forces), the singlet is closed-shell, whereas with the hybrid functionals, M06, B3LYP* and PBE0*, the singlet is open-shell, with two antiferromagnetically-coupled unpaired electrons located on the metal centers, *i.e.* Ni(↑)Ni(↓). Furthermore, the triplet state, Ni(↑)Ni(↑), is more stable with the latter functionals. This is due to the Hartree-Fock exchange introduced in hybrid functionals. The best fit between the experimental X-Ray structure and the DFT-optimized geometry of the ground state was given by the closed-shell singlet state determined as the ground state using the BP86* and M06L functionals (Table S6).

Table S5: Relative energies (ΔH), in kJ mol^{-1}, for the closed-shell singlet (**S**), open-shell singlet (**OSS**) and triplet (**T**) states of **1a**, with different DFT functionals. Geometries were fully-optimized in the gas phase for the real systems by using the double-ζ quality basis set.

	S	**OSS**	**T**
M06L	0		32.5
BP86*	0		60.0
M06	8.9	0.0	1.7
B3LYP*	32.2	0.0	5.1
PBE0*	42.4	0.0	5.5

Table S6: Selected bond distances, in Å, for the comparison of the X-ray structure of **1a** with the DFT geometries of the ground state optimized with different functionals.

	Ni1-Ni2	**Ni1-C4**	**Ni2-C4**
X-Ray (exp.)	2.4015(3)	2.2067(18)	2.1879(19)
M06L	2.37	2.18	2.17
BP86*	2.37	2.20	2.20
M06	2.55	2.22	2.65
B3LYP*	2.64	2.23	2.75
PBE0*	2.67	2.26	2.76

In addition to M06L, M06 was also used for modeling complex **2a**. With both functionals, the ground state is a closed shell singlet (Table S7). The energy differences between the **S** and **T** states are 63.3 and 16.6 kJ mol^{-1} with M06L and M06, respectively. When this data is compared to that obtained for **1a** (Table S5), it becomes apparent that the triplet state is less accessible with an indenyl bridging ligand (**2a**) compared with a Cp bridging ligand (**1a**), regardless of the functional used.

Table S7: Relative energies (ΔH), in kJ mol^{-1}, for the closed-shell singlet (**S**) and triplet (**T**) states of **2a**, with the M06L and M06 functionals. Geometries were fully-optimized in the gas phase for the real systems by using the double-ζ quality basis set.

	S	**T**
M06L	0	63.3
M06	0	16.6

DFT results on the (μ-Cl)$_2$Ni$_2$(IPr)$_2$ dimer

All attempts to model the electronic and geometrical structures of the (μ-Cl)$_2$Ni$_2$(IPr)$_2$ dimer were unsuccessful. Both pure and hybrid DFT functionals predicted a triplet ground state for this species (Table S8), which is in contradiction with the diamagnetism observed in the experiments. In addition, there are rather large differences between the X-Ray structure of

this species and the DFT-optimized geometries for all functionals tested (Table S9). These experiment-theory discrepancies may be due to the lack of crystal-packing effects in the gas phase calculations. The DFT models of $(\mu\text{-Cl})_2Ni_2(IPr)_2$ were thus not considered any further.

Table S8: Relative energies (ΔH), in kJ mol^{-1}, for the closed-shell singlet (**S**), open-shell singlet (**OSS**) and triplet (**T**) states of $(\mu\text{-Cl})_2Ni_2(IPr)_2$, with different DFT functionals. Geometries were fully-optimized in gas phase for the real systems by using the double-ζ quality basis set.

	S	**OSS**	**T**
M06L	58.1	50.5	0
M06	114.2	1.5	0
B3LYP*	119.9	0.7	0
PBE0*	141.9	0.4	0

* = D3 Grimme's corrections for dispersion forces.

Table S9: Selected geometrical parameters, in Å and °, for the comparison of the X-ray structure of $(\mu\text{-Cl})_2Ni_2(IPr)_2$ with the DFT geometries of the ground state (**T**) optimized with different functionals.

	Ni-Ni	**Ni1-Cl1**	**Ni1-Cl2**	**N1C1Ni1Cl1**	**N2C2Ni2Cl1**
X-Ray (exp.)	2.5194(2)	2.2085(8)	2.2467(8)	88.8(2)	4.6(3)
M06L	2.77	2.28	2.35	35.6	-8.19
M06	2.73	2.26	2.31	48.4	-6.0
B3LYP*	3.02	2.30	2.37	97.8	2.28
PBE0*	2.88	2.27	2.33	111.4	-21.2

* = D3 Grimme's corrections for dispersion forces.

Optimized energies and geometries of all stationary points

Optimized Cartesian coordinates, in Å, and absolute energies, in hartrees, of all stationary points reported in the text. Enthalpies (H_{Bz}) and Gibbs (G_{THF}) energies were obtained from Eqs S1 and S2, respectively, in which the thermochemistry corrections found in gas phase, (H – E) and (G – E), were added to the single-point energies computed with the continuum SMD model for solvation (solvent = Benzene (Bz) or THF).

$$H_{Bz} = (H - E) + E_{SMD}(Bz) \qquad \text{Eq. S1}$$

$$G_{THF} = (G - E) + E_{SMD}(THF) \qquad \text{Eq. S2}$$

Cp anion

H_{Bz} = –193,510984

C	-0.600748	1.038143	0.000128
H	-1.145737	1.982429	-0.001636
C	0.801683	0.890972	0.003247
H	1.531179	1.701136	0.004464
C	1.095131	-0.487916	0.004212
H	2.091003	-0.931365	0.006667
C	-0.125695	-1.193199	0.001669
H	-0.239560	-2.277363	0.002090
C	-1.173858	-0.249999	-0.000719
H	-2.240174	-0.477060	-0.003153

In anion

H_{Bz} = –347,143563

C	-0.591816	-0.295645	-3.135547
H	-1.515889	0.198576	-3.436685
C	-0.457952	-1.676036	-2.903999
H	-1.240225	-2.425636	-2.991036
C	0.885876	-1.913664	-2.534709
C	1.606049	-3.071841	-2.189068
H	1.101259	-4.041261	-2.176240
C	2.953550	-2.993148	-1.864615
H	3.502844	-3.897080	-1.598294
C	3.622960	-1.748584	-1.876029
H	4.681937	-1.704899	-1.618342
C	2.943356	-0.585617	-2.212002
H	3.474005	0.369977	-2.216698
C	1.577455	-0.627952	-2.546602
C	0.635486	0.356728	-2.922963
H	0.828926	1.421319	-3.027232

1a

H_{Bz} = –3315,04054

Cl	0.009710	-0.296243	1.601543
C	-0.044022	-0.727060	-2.052626
H	0.000356	-1.808633	-2.164285
C	1.107985	0.133381	-2.284167
H	2.035338	-0.219486	-2.730973
C	0.564049	1.463957	-2.553509
H	1.166865	2.353097	-2.716364
C	-0.800799	1.422404	-2.532768
H	-1.463983	2.270359	-2.678053
C	-1.253032	0.059048	-2.248675
H	-2.177600	-0.349142	-2.655380
Ni	1.161256	-0.065847	-0.357922
C	2.991971	-0.041440	-0.012553
N	3.852551	-1.039219	-0.407830
C	3.454982	-2.135501	-1.243083
C	2.605736	-3.122915	-0.709415
C	2.239175	-4.173393	-1.554029
H	1.582183	-4.951860	-1.175905
C	2.141669	-3.093958	0.734166

H	2.102382	-2.048017	1.054342
C	0.732191	-3.641184	0.920549
H	0.012538	-3.117258	0.278525
H	0.404819	-3.483960	1.953428
H	0.665861	-4.717348	0.719273
C	3.147097	-3.808967	1.638544
H	3.235862	-4.870520	1.377989
H	2.831098	-3.744820	2.685947
H	4.145187	-3.362368	1.564902
C	2.693939	-4.238524	-2.865854
H	2.396503	-5.068214	-3.501683
C	3.516695	-3.240848	-3.369348
H	3.851987	-3.288110	-4.402609
C	3.905054	-2.159275	-2.573775
C	4.771484	-1.058199	-3.156276
H	4.706636	-0.189181	-2.487078
C	4.293713	-0.606189	-4.535049
H	4.424967	-1.391070	-5.287598
H	4.873448	0.259353	-4.870470
H	3.236943	-0.324408	-4.530006
C	6.235803	-1.493498	-3.229136
H	6.632241	-1.778539	-2.249949
H	6.864987	-0.689666	-3.624957
H	6.346846	-2.360276	-3.890687
C	5.135107	-0.838870	0.080224
H	5.938608	-1.530304	-0.119883
C	5.099977	0.310121	0.796488
H	5.870957	0.840174	1.333387
N	3.794294	0.780834	0.732831
C	3.336800	1.935120	1.451509
C	3.039052	1.792681	2.816086
C	3.036287	0.436179	3.494342
H	2.856978	-0.317125	2.713600
C	1.908358	0.302458	4.513546
H	2.090600	0.915814	5.403646
H	1.830433	-0.736820	4.849156
H	0.944621	0.588674	4.083226
C	4.379100	0.132304	4.161284
H	5.209845	0.143341	3.450469
H	4.360011	-0.856027	4.632890
H	4.597786	0.870458	4.941839
C	2.726216	2.954810	3.527188
H	2.496560	2.884045	4.587318
C	2.688898	4.191215	2.896016
H	2.447464	5.083471	3.467672
C	2.916368	4.287494	1.527843
H	2.829494	5.253450	1.037839
C	3.230465	3.158335	0.769448
C	3.448063	3.234422	-0.729280
H	3.073225	2.290838	-1.153362
C	2.661807	4.361888	-1.387259
H	1.595029	4.305346	-1.144129
H	2.760929	4.303287	-2.475752
H	3.029188	5.350028	-1.087285
C	4.935453	3.348031	-1.068498
H	5.356941	4.274498	-0.660962

H	5.085805	3.361793	-2.153295
H	5.512667	2.511135	-0.662873
Ni	-1.210248	-0.130556	-0.319314
C	-2.971301	0.102576	0.244455
N	-3.732941	-0.791585	0.956960
C	-3.350018	-2.158655	1.159964
C	-2.858713	-2.554254	2.414774
C	-2.674291	-1.563393	3.546261
H	-2.526808	-0.573823	3.093271
C	-3.919791	-1.509486	4.433504
H	-3.787585	-0.787788	5.246065
H	-4.113855	-2.489253	4.885518
H	-4.814546	-1.220277	3.874524
C	-1.442386	-1.864992	4.394340
H	-1.256995	-1.044361	5.094660
H	-0.549887	-1.981703	3.773040
H	-1.573574	-2.773663	4.993371
C	-2.557555	-3.907031	2.589010
H	-2.175261	-4.246590	3.548208
C	-2.720660	-4.819005	1.553904
H	-2.478115	-5.866782	1.712340
C	-3.173612	-4.392186	0.312449
H	-3.282549	-5.108318	-0.499000
C	-3.491657	-3.051302	0.083970
C	-3.991390	-2.610935	-1.277614
H	-3.941979	-1.517553	-1.321885
C	-5.449673	-3.018546	-1.485542
H	-5.816449	-2.670052	-2.457380
H	-6.100955	-2.598640	-0.711642
H	-5.563086	-4.108654	-1.457704
C	-3.102356	-3.133161	-2.401957
H	-2.056842	-2.854264	-2.234015
H	-3.414524	-2.712087	-3.363747
H	-3.151963	-4.223898	-2.492356
C	-4.972415	-0.263584	1.289802
H	-5.713384	-0.835056	1.826706
C	-5.000299	0.999374	0.800147
H	-5.757867	1.766491	0.844139
N	-3.783199	1.206589	0.166151
C	-3.455609	2.381954	-0.582169
C	-2.466742	3.251899	-0.096759
C	-1.713307	3.006935	1.192964
H	-1.925819	1.989035	1.536384
C	-2.174276	3.976928	2.280678
H	-1.987001	5.016933	1.987545
H	-1.631964	3.792716	3.214082
H	-3.246085	3.879296	2.485909
C	-0.205086	3.096186	0.981490
H	0.121197	2.458421	0.146834
H	0.326301	2.755659	1.876002
H	0.115020	4.126004	0.777477
C	-2.189762	4.395710	-0.850104
H	-1.426903	5.085779	-0.495417
C	-2.863636	4.660250	-2.035564
H	-2.633656	5.557647	-2.603699
C	-3.817386	3.766004	-2.506306

H	-4.323907	3.961095	-3.449571
C	-4.123822	2.600945	-1.798703
C	-5.119037	1.614560	-2.379289
H	-5.135324	0.721227	-1.743176
C	-4.702836	1.164607	-3.779294
H	-4.732383	1.994552	-4.494280
H	-5.381508	0.389451	-4.150917
H	-3.685849	0.759870	-3.787978
C	-6.530773	2.198581	-2.397262
H	-6.867769	2.480956	-1.394671
H	-7.248027	1.478423	-2.803723
H	-6.574449	3.098214	-3.021733

1a-triplet

H_{Bz} = –3315,02786

Cl	-0.065574	0.093489	1.135067
C	-0.152049	0.764446	-2.621510
H	-0.579909	1.764005	-2.656629
C	-0.884751	-0.455129	-2.878735
H	-1.960213	-0.512535	-3.045660
C	0.047266	-1.479803	-3.141657
H	-0.189598	-2.527329	-3.296750
C	1.337428	-0.929877	-3.100618
H	2.257596	-1.481324	-3.264037
C	1.244725	0.468310	-2.832529
H	2.020950	1.212070	-2.992877
Ni	-1.366613	0.017925	-0.849124
C	-3.007207	0.169077	0.153328
N	-3.753032	1.319817	0.134334
C	-3.569625	2.352445	-0.848556
C	-2.537865	3.289273	-0.676962
C	-2.393967	4.271448	-1.662028
H	-1.606389	5.012898	-1.553574
C	-1.623871	3.289449	0.529697
H	-1.709906	2.316349	1.028623
C	-0.158572	3.456999	0.140358
H	0.150553	2.696904	-0.587730
H	0.479647	3.335398	1.020001
H	0.047680	4.449713	-0.278569
C	-2.048876	4.373223	1.522163
H	-1.992275	5.368735	1.065898
H	-1.390036	4.371626	2.397535
H	-3.076698	4.227518	1.871687
C	-3.229936	4.309990	-2.769734
H	-3.095204	5.079636	-3.524919
C	-4.237305	3.364085	-2.917049
H	-4.881464	3.396330	-3.791484
C	-4.431954	2.366920	-1.959326
C	-5.481249	1.292674	-2.166664
H	-5.765458	0.896189	-1.182984
C	-4.879780	0.136975	-2.968144
H	-4.564256	0.477158	-3.961242
H	-5.607069	-0.672001	-3.102170
H	-4.000388	-0.276437	-2.462003
C	-6.750892	1.810292	-2.833737

H	-7.163586	2.677981	-2.310458
H	-7.516247	1.028805	-2.849505
H	-6.575022	2.102283	-3.874455
C	-4.715759	1.335640	1.130358
H	-5.387232	2.169196	1.265661
C	-4.592987	0.162452	1.797549
H	-5.155918	-0.251716	2.619114
N	-3.550078	-0.533373	1.200998
C	-3.244287	-1.896689	1.536843
C	-2.587866	-2.183001	2.747723
C	-2.066190	-1.109231	3.682301
H	-2.002669	-0.171248	3.115420
C	-0.658883	-1.444625	4.175337
H	-0.652387	-2.335384	4.813262
H	-0.257707	-0.623727	4.777128
H	0.023539	-1.609946	3.336561
C	-3.005756	-0.900231	4.870514
H	-4.014231	-0.612672	4.557425
H	-2.625924	-0.112581	5.529890
H	-3.095743	-1.817142	5.464820
C	-2.412978	-3.528803	3.084515
H	-1.919075	-3.777851	4.019987
C	-2.835464	-4.549060	2.243181
H	-2.674669	-5.586871	2.522538
C	-3.450006	-4.237515	1.039290
H	-3.766224	-5.034660	0.370001
C	-3.680755	-2.911410	0.665583
C	-4.362559	-2.635750	-0.658028
H	-4.563364	-1.561189	-0.727889
C	-3.429403	-3.007355	-1.810035
H	-2.480423	-2.462413	-1.734617
H	-3.885001	-2.774263	-2.779124
H	-3.200079	-4.079707	-1.797937
C	-5.702773	-3.358491	-0.771719
H	-5.580281	-4.446959	-0.755560
H	-6.196894	-3.103622	-1.714977
H	-6.377773	-3.089725	0.046893
Ni	1.253275	0.152791	-0.836905
C	2.855135	-0.154591	0.193669
N	3.420425	0.641856	1.161519
C	3.187168	2.050554	1.317707
C	2.583921	2.525758	2.493763
C	1.986415	1.613093	3.546556
H	1.901201	0.608422	3.112984
C	2.870288	1.537061	4.791291
H	2.435298	0.860875	5.534949
H	2.974645	2.522950	5.259560
H	3.877903	1.175800	4.562954
C	0.576785	2.070412	3.923468
H	0.127608	1.380589	4.644636
H	-0.073263	2.102775	3.043814
H	0.582373	3.060407	4.393033
C	2.520674	3.912150	2.674760
H	2.074805	4.305343	3.585118
C	2.988601	4.788311	1.707804
H	2.920358	5.861323	1.865590

C	3.530894	4.291926	0.528293
H	3.882753	4.982894	-0.232565
C	3.656088	2.919149	0.310550
C	4.351365	2.402089	-0.934154
H	3.901812	1.435259	-1.189156
C	5.837308	2.170125	-0.648813
H	6.346066	1.761528	-1.529466
H	5.986673	1.467495	0.178444
H	6.334299	3.110333	-0.382119
C	4.163995	3.305327	-2.146831
H	3.107194	3.541708	-2.310779
H	4.541602	2.809006	-3.046656
H	4.709649	4.250897	-2.051027
C	4.396440	-0.038635	1.880520
H	4.957283	0.437527	2.669735
C	4.462345	-1.288705	1.362951
H	5.081432	-2.136388	1.613078
N	3.525375	-1.338696	0.342262
C	3.411995	-2.417057	-0.595293
C	2.368252	-3.342580	-0.463717
C	1.334193	-3.248219	0.636162
H	1.466279	-2.288535	1.149993
C	1.516189	-4.366159	1.662025
H	1.397059	-5.352997	1.198513
H	0.766220	-4.282120	2.456572
H	2.508203	-4.336177	2.126116
C	-0.081104	-3.248785	0.068575
H	-0.205524	-2.451452	-0.675202
H	-0.805853	-3.073263	0.868276
H	-0.332367	-4.205664	-0.408414
C	2.304203	-4.374397	-1.404587
H	1.504440	-5.107658	-1.326609
C	3.230214	-4.467933	-2.435727
H	3.159078	-5.277059	-3.157697
C	4.238305	-3.518197	-2.555764
H	4.943707	-3.578460	-3.382067
C	4.347072	-2.467837	-1.642005
C	5.392288	-1.389603	-1.851127
H	5.381937	-0.719967	-0.983104
C	5.035957	-0.545689	-3.075366
H	4.991209	-1.160617	-3.981790
H	5.783057	0.238603	-3.242230
H	4.058444	-0.066746	-2.951642
C	6.801183	-1.964168	-1.965336
H	7.065468	-2.560657	-1.086411
H	7.537852	-1.159676	-2.061498
H	6.905908	-2.608673	-2.845083

1a-Pd

H_{Bz} = –3229,00887

Cl	7.569146	1.767439	22.586982
C	4.095634	0.647137	20.619807
H	4.322162	-0.392517	20.406037
C	4.230084	1.708920	19.656439
H	4.366414	1.510301	18.595779

C	3.433613	2.829239	20.175493
H	3.366863	3.803120	19.699049
C	2.870913	2.478107	21.367225
H	2.279308	3.119780	22.014647
C	3.253337	1.097409	21.698951
H	2.602849	0.425730	22.256900
C	7.407227	2.688911	19.048626
N	7.815750	1.909207	17.996122
C	7.110929	0.722763	17.600208
C	7.238783	-0.441621	18.377720
C	6.475712	-1.549079	17.995990
H	6.550176	-2.465377	18.576113
C	8.216854	-0.557311	19.531906
H	8.471783	0.452612	19.873694
C	7.637047	-1.293591	20.736846
H	6.712364	-0.817838	21.086811
H	8.344053	-1.265485	21.571846
H	7.430938	-2.348500	20.518860
C	9.503370	-1.229077	19.045721
H	9.304312	-2.248310	18.693650
H	10.236720	-1.292673	19.855993
H	9.962635	-0.678201	18.217663
C	5.628765	-1.498691	16.896651
H	5.042057	-2.371712	16.623860
C	5.532911	-0.337278	16.140646
H	4.866810	-0.308857	15.282618
C	6.269862	0.800976	16.476357
C	6.092651	2.091365	15.696479
H	7.013927	2.679665	15.792491
C	4.961382	2.925134	16.302657
H	4.012946	2.376005	16.267030
H	4.827743	3.862051	15.750633
H	5.163412	3.170026	17.351005
C	5.851516	1.863569	14.208139
H	6.611229	1.213513	13.763644
H	5.866121	2.817530	13.672693
H	4.873083	1.409462	14.018591
C	8.923512	2.444652	17.356620
H	9.384276	1.963777	16.507654
C	9.221912	3.596472	18.004715
H	9.980140	4.340784	17.820561
N	8.294777	3.732948	19.032079
C	8.264208	4.847654	19.941034
C	9.236344	4.933451	20.951153
C	10.263809	3.844463	21.188476
H	9.863449	2.913352	20.764854
C	10.512807	3.597667	22.675445
H	11.071611	4.422512	23.132750
H	11.112210	2.691190	22.807070
H	9.576290	3.457812	23.222172
C	11.589208	4.169290	20.495686
H	11.483628	4.281269	19.413142
H	12.324343	3.378637	20.677125
H	12.006038	5.105743	20.884707
C	9.228238	6.077618	21.756785
H	9.971108	6.173557	22.544381

C	8.280202	7.074905	21.581183
H	8.289655	7.952488	22.222352
C	7.297231	6.937207	20.608085
H	6.534489	7.703641	20.505304
C	7.259138	5.820017	19.773811
C	6.176188	5.659777	18.726054
H	5.888478	4.600785	18.736668
C	4.914543	6.457037	19.029152
H	4.540254	6.255335	20.038593
H	4.126803	6.185379	18.318304
H	5.073121	7.537550	18.934431
C	6.698849	5.999735	17.328545
H	7.057680	7.034982	17.290504
H	5.903122	5.897235	16.583385
H	7.524189	5.347382	17.024880
C	4.713936	1.144115	24.704670
N	5.191932	0.222958	25.597158
C	5.806839	-1.004354	25.172772
C	7.203692	-1.126920	25.232300
C	8.085787	-0.003008	25.737216
H	7.554772	0.940015	25.546087
C	8.328455	-0.132558	27.242992
H	8.945974	0.694263	27.608657
H	8.855736	-1.067128	27.468168
H	7.399388	-0.133390	27.819635
C	9.424275	0.075338	25.008517
H	9.953186	0.987411	25.302759
H	9.290103	0.105278	23.924329
H	10.075061	-0.769058	25.264227
C	7.757551	-2.338566	24.808659
H	8.836224	-2.466724	24.839455
C	6.956863	-3.364853	24.328173
H	7.410032	-4.295967	23.997651
C	5.578500	-3.200117	24.246424
H	4.964465	-4.001213	23.844595
C	4.970651	-2.014617	24.660925
C	3.467454	-1.824904	24.570863
H	3.291721	-0.776225	24.295205
C	2.797368	-2.074147	25.923980
H	1.717889	-1.894226	25.862558
H	3.197710	-1.419514	26.704894
H	2.945339	-3.111096	26.247441
C	2.817734	-2.674805	23.486505
H	3.317694	-2.547563	22.520707
H	1.768657	-2.386677	23.363448
H	2.829941	-3.742312	23.734313
C	4.923386	0.595532	26.907393
H	5.210384	-0.014872	27.748898
C	4.263176	1.777958	26.846847
H	3.868501	2.417290	27.621190
N	4.144269	2.097460	25.501081
C	3.312361	3.149661	24.996135
C	3.895230	4.301317	24.448527
C	5.388251	4.453150	24.251720
H	5.864678	3.487811	24.458151
C	5.981378	5.488080	25.205470

H	5.533008	6.476395	25.045753
H	7.060270	5.583467	25.039835
H	5.821382	5.217421	26.255051
C	5.706052	4.804541	22.799456
H	5.209057	4.115475	22.103131
H	6.782971	4.732899	22.616150
H	5.391642	5.827513	22.554318
C	3.030950	5.320832	24.038426
H	3.455707	6.229346	23.616966
C	1.652370	5.187728	24.146838
H	1.001610	5.997655	23.827867
C	1.103473	4.006183	24.631801
H	0.022709	3.886034	24.673128
C	1.921613	2.955276	25.051371
C	1.304642	1.642155	25.494540
H	2.111401	0.910535	25.623987
C	0.364232	1.087305	24.425653
H	-0.514357	1.727501	24.289653
H	0.002213	0.093105	24.709701
H	0.863513	1.003801	23.455119
C	0.582847	1.780116	26.833294
H	1.255084	2.130380	27.622848
H	0.162211	0.820232	27.151493
H	-0.242083	2.498179	26.760865
Pd	6.158162	2.025802	20.495439
Pd	5.098675	1.231065	22.727317

1a-Pd-triplet

H_{Bz} = –3228,97446

Cl	7.443865	1.988818	22.508241
C	3.179036	0.750159	20.438344
H	2.993687	-0.304284	20.256353
C	3.965520	1.584077	19.546724
H	4.428485	1.216956	18.637980
C	3.687904	2.942359	19.860124
H	3.977065	3.810863	19.277117
C	2.745020	2.958218	20.930720
H	2.367892	3.843720	21.432360
C	2.376545	1.617695	21.241305
H	1.534881	1.334150	21.864305
C	7.256750	2.706799	19.266365
N	7.679204	1.793615	18.336819
C	6.922265	0.643064	17.925269
C	6.887077	-0.490195	18.753097
C	6.124274	-1.578656	18.316940
H	6.079237	-2.468712	18.939471
C	7.647402	-0.582728	20.057242
H	7.941742	0.425785	20.363854
C	6.785194	-1.149626	21.180992
H	5.838766	-0.595340	21.269143
H	7.300873	-1.050892	22.140520
H	6.550457	-2.211251	21.030679
C	8.925199	-1.404973	19.885489
H	8.699530	-2.434679	19.582464
H	9.476290	-1.447876	20.831777

H	9.587945	-0.972740	19.127708
C	5.432312	-1.540701	17.115528
H	4.845772	-2.398811	16.798290
C	5.485792	-0.404607	16.315438
H	4.933829	-0.379335	15.380050
C	6.229754	0.711986	16.701851
C	6.195168	1.986349	15.879343
H	7.147327	2.514298	16.022462
C	5.084093	2.899785	16.402726
H	4.108743	2.404623	16.330114
H	5.034072	3.829840	15.826187
H	5.247186	3.156169	17.455187
C	6.029074	1.740609	14.384421
H	6.768808	1.032032	13.999842
H	6.140042	2.679161	13.833485
H	5.035192	1.347651	14.144427
C	8.904182	2.141584	17.786625
H	9.388510	1.533215	17.038306
C	9.267920	3.308304	18.369205
H	10.128397	3.941173	18.219391
N	8.260211	3.638895	19.266919
C	8.276525	4.839461	20.059698
C	9.245280	4.979177	21.067762
C	10.255829	3.899931	21.401997
H	9.886954	2.951702	20.990682
C	10.417756	3.706076	22.907922
H	10.898824	4.571471	23.378776
H	11.057217	2.838984	23.101657
H	9.452448	3.526547	23.388096
C	11.618658	4.220718	20.782727
H	11.577459	4.335582	19.695510
H	12.341856	3.429921	21.006717
H	12.015462	5.157778	21.191003
C	9.279804	6.192224	21.764510
H	10.021658	6.327566	22.547271
C	8.387046	7.214099	21.481568
H	8.430860	8.145508	22.040080
C	7.425343	7.041792	20.492804
H	6.716700	7.839544	20.288891
C	7.347117	5.856484	19.761799
C	6.325799	5.690283	18.655371
H	5.970171	4.654798	18.709429
C	5.102852	6.584779	18.807050
H	4.647659	6.485725	19.798580
H	4.348451	6.310988	18.061524
H	5.339926	7.643009	18.648468
C	6.977395	5.894703	17.285937
H	7.404394	6.901165	17.206292
H	6.242066	5.780349	16.482549
H	7.782263	5.174183	17.106375
C	4.640209	1.122229	24.450022
N	5.378465	0.322183	25.282934
C	6.054165	-0.878286	24.878928
C	7.459694	-0.935391	24.945693
C	8.311572	0.227094	25.414728
H	7.723361	1.146397	25.296171

C	8.694969	0.059106	26.888191
H	9.282565	0.915434	27.234524
H	9.306971	-0.840394	27.024200
H	7.828881	-0.038284	27.548941
C	9.584612	0.391657	24.586676
H	10.085245	1.325143	24.862147
H	9.364354	0.436909	23.516654
H	10.296881	-0.420595	24.774145
C	8.071941	-2.140597	24.591116
H	9.155659	-2.211578	24.631522
C	7.328640	-3.237463	24.175697
H	7.830717	-4.160181	23.896438
C	5.945386	-3.152468	24.119057
H	5.361342	-4.015558	23.806247
C	5.276094	-1.979651	24.478265
C	3.759601	-1.981420	24.492514
H	3.409541	-0.967746	24.714230
C	3.236213	-2.901397	25.595248
H	2.141576	-2.884752	25.628074
H	3.608492	-2.599349	26.579869
H	3.546315	-3.939666	25.429472
C	3.180415	-2.363737	23.132707
H	3.561258	-1.699855	22.345793
H	2.087820	-2.282538	23.142628
H	3.433394	-3.394060	22.856996
C	5.300842	0.749283	26.601502
H	5.797168	0.224800	27.401432
C	4.518001	1.853845	26.609668
H	4.207062	2.505563	27.411455
N	4.124907	2.066225	25.297050
C	3.225873	3.111297	24.890408
C	3.756130	4.235713	24.234913
C	5.227856	4.307864	23.904360
H	5.531129	3.300117	23.597233
C	6.081929	4.681204	25.116128
H	5.847785	5.691787	25.473522
H	7.141943	4.654482	24.840093
H	5.940710	3.987218	25.951393
C	5.551592	5.214763	22.728941
H	4.941027	4.966676	21.850472
H	6.600398	5.068264	22.449070
H	5.413106	6.280113	22.958349
C	2.863285	5.249859	23.883571
H	3.241319	6.132113	23.373670
C	1.504571	5.137225	24.156748
H	0.826503	5.937979	23.872314
C	1.006725	3.999590	24.779170
H	-0.059825	3.913816	24.970119
C	1.857502	2.958904	25.163641
C	1.302158	1.685276	25.772942
H	2.100113	1.208797	26.356838
C	0.908336	0.703878	24.666468
H	0.100075	1.119520	24.052524
H	0.555532	-0.243850	25.089066
H	1.757428	0.493558	24.002699
C	0.130553	1.932029	26.717118

H	0.376425	2.665021	27.491569
H	-0.161278	1.000859	27.212078
H	-0.753290	2.297589	26.183085
Pd	5.753778	2.347769	20.739906
Pd	4.115167	0.932496	22.465352

2a

H_{bz} = -3468,67171

Cl	-0.055433	0.833627	1.490668
C	0.056570	0.543473	-2.091915
H	0.065860	1.604788	-2.338627
C	1.249768	-0.301646	-2.170523
H	2.157342	0.021328	-2.682610
C	0.749215	-1.677821	-2.339148
C	1.425340	-2.890743	-2.433415
H	2.509940	-2.925544	-2.356215
C	0.699689	-4.069580	-2.615284
H	1.227355	-5.018123	-2.687536
C	-0.691881	-4.041680	-2.716048
H	-1.241681	-4.967374	-2.871910
C	-1.382257	-2.833030	-2.615922
H	-2.469401	-2.808208	-2.696499
C	-0.670994	-1.653480	-2.411807
C	-1.143498	-0.273578	-2.254119
H	-2.020200	0.086161	-2.789084
Ni	1.206587	0.240215	-0.327585
C	-2.975509	0.155703	0.179986
N	-3.888724	1.063906	-0.297144
C	-3.662969	1.861068	-1.466926
C	-4.142951	1.378606	-2.697458
C	-4.805292	0.017426	-2.803593
H	-4.383153	-0.606678	-2.001485
C	-6.316975	0.108031	-2.587508
H	-6.779149	0.735625	-3.358252
H	-6.569535	0.535976	-1.613135
H	-6.778154	-0.883777	-2.642831
C	-4.509375	-0.686437	-4.124531
H	-4.882019	-1.715362	-4.091998
H	-3.436600	-0.724931	-4.339101
H	-5.004565	-0.194750	-4.969195
C	-3.940272	2.178089	-3.823240
H	-4.293185	1.835802	-4.792306
C	-3.268777	3.392295	-3.726325
H	-3.119319	3.999215	-4.615447
C	-2.769002	3.820928	-2.503587
H	-2.222463	4.759313	-2.441592
C	-2.947201	3.060183	-1.344212
C	-2.402484	3.545036	-0.016756
H	-2.451696	2.715271	0.699368
C	-3.261768	4.689600	0.523087
H	-2.895639	5.023052	1.499484
H	-4.309768	4.393433	0.638614
H	-3.236765	5.552167	-0.153549
C	-0.937793	3.959723	-0.107099
H	-0.793579	4.819728	-0.772349

H	-0.301504	3.138510	-0.457566
H	-0.567810	4.248488	0.881893
C	-5.035188	1.111721	0.478286
C	-4.870817	0.193317	1.459397
N	-3.624065	-0.387911	1.262371
C	-3.147118	-1.467213	2.081085
C	-3.208227	-2.780599	1.571681
C	-3.607688	-3.061968	0.133466
H	-3.039832	-2.351740	-0.487068
C	-5.098108	-2.832592	-0.135838
H	-5.323019	-3.041361	-1.188801
H	-5.421403	-1.808468	0.068801
H	-5.712550	-3.507525	0.471514
C	-3.227575	-4.464028	-0.323627
H	-3.802655	-5.232642	0.206377
H	-2.161760	-4.668436	-0.183473
H	-3.444922	-4.579832	-1.389712
C	-2.836750	-3.815304	2.432384
H	-2.864212	-4.839946	2.076072
C	-2.407261	-3.559310	3.729026
H	-2.118894	-4.383950	4.375890
C	-2.322400	-2.254232	4.189015
H	-1.960725	-2.059804	5.195653
C	-2.687131	-1.176662	3.375212
C	-2.587578	0.239863	3.904565
H	-2.570104	0.917715	3.040990
C	-3.797951	0.589545	4.773076
H	-3.859362	-0.081965	5.637571
H	-4.742468	0.509798	4.227135
H	-3.719956	1.613381	5.153072
C	-1.302227	0.470023	4.696894
H	-1.319385	-0.057199	5.657768
H	-1.186490	1.535573	4.918505
H	-0.420387	0.145696	4.136321
Ni	-1.182060	0.159082	-0.377183
C	2.949771	0.196575	0.358353
N	3.596609	1.318570	0.824533
C	3.139434	2.653925	0.566435
C	3.269926	3.155018	-0.743781
C	3.919918	2.337697	-1.844166
H	3.705421	1.281019	-1.644242
C	5.438938	2.522010	-1.807955
H	5.709432	3.567056	-1.999048
H	5.926255	1.905467	-2.571218
H	5.858080	2.242371	-0.835513
C	3.369705	2.642343	-3.231635
H	3.642487	3.646632	-3.575181
H	2.278573	2.559374	-3.259128
H	3.777636	1.934789	-3.960883
C	2.847542	4.464091	-0.977908
H	2.931208	4.879810	-1.978316
C	2.333582	5.245972	0.050003
H	2.014251	6.265225	-0.152178
C	2.215854	4.725185	1.330639
H	1.798446	5.339939	2.124295
C	2.599398	3.412309	1.617305

C	2.456113	2.862109	3.021662
H	2.441425	1.766426	2.949907
C	1.154654	3.294227	3.692592
H	1.148014	4.368403	3.910977
H	1.038005	2.774158	4.648686
H	0.286234	3.058794	3.071157
C	3.643923	3.279473	3.892092
H	3.688889	4.371084	3.981903
H	4.601499	2.946512	3.481975
H	3.551843	2.865704	4.901494
C	4.794817	1.002706	1.448025
C	4.921120	-0.343056	1.383024
N	3.803285	-0.819084	0.708666
C	3.684858	-2.186618	0.295940
C	4.616434	-2.664066	-0.645561
C	5.581359	-1.752222	-1.382503
H	5.603293	-0.778052	-0.882308
C	7.008938	-2.290187	-1.389609
H	7.085299	-3.234057	-1.939880
H	7.379137	-2.470394	-0.375184
H	7.685221	-1.579119	-1.874383
C	5.074641	-1.509886	-2.805371
H	4.998045	-2.449813	-3.364546
H	5.751995	-0.847391	-3.355404
H	4.079772	-1.050232	-2.797275
C	4.565255	-4.019313	-0.978696
H	5.270745	-4.413083	-1.706925
C	3.609282	-4.858748	-0.419990
H	3.580863	-5.909599	-0.695806
C	2.671928	-4.347197	0.466498
H	1.902647	-4.998290	0.876330
C	2.686794	-3.002662	0.849085
C	1.637985	-2.491288	1.812316
H	1.790452	-1.416656	1.964756
C	0.238194	-2.661426	1.226983
H	-0.015900	-3.720856	1.088686
H	0.146892	-2.162089	0.252707
H	-0.501704	-2.215723	1.898423
C	1.740036	-3.174418	3.175033
H	1.552776	-4.252505	3.098726
H	0.991877	-2.760664	3.860269
H	2.728222	-3.040495	3.627710
H	5.695445	-1.007562	1.734019
H	5.444474	1.759313	1.858896
H	-5.846897	1.788129	0.260499
H	-5.526426	-0.131027	2.251971

2a-triplet

H_{Bz} = –3468,65017

Cl	0.166103	0.702922	1.160230
C	-0.934903	-1.743151	-1.852732
H	-1.384861	-1.216754	-2.691346
C	0.503675	-1.866192	-1.630273
H	1.196188	-1.722069	-2.468460
C	0.674015	-2.974038	-0.692665

C	1.809266	-3.639932	-0.222051
H	2.799792	-3.362367	-0.581165
C	1.660956	-4.666759	0.705560
H	2.542939	-5.192252	1.065338
C	0.393372	-5.029845	1.184839
H	0.303723	-5.829345	1.916752
C	-0.747894	-4.387487	0.721755
H	-1.734766	-4.683001	1.077834
C	-0.620125	-3.370307	-0.232043
C	-1.603073	-2.588271	-0.934897
H	-2.672012	-2.760520	-0.883646
Ni	1.384566	-0.303050	-0.598979
C	-3.056798	-0.190362	0.304852
N	-4.150912	-0.275783	-0.527656
C	-4.013137	-0.337394	-1.952124
C	-4.404885	-1.494370	-2.645904
C	-5.078277	-2.661154	-1.951593
H	-4.841999	-2.594087	-0.880223
C	-6.598360	-2.565067	-2.101436
H	-6.884573	-2.627639	-3.157484
H	-6.994120	-1.621605	-1.713461
H	-7.096833	-3.382618	-1.571086
C	-4.591389	-4.016976	-2.457899
H	-4.997528	-4.821454	-1.837293
H	-3.500481	-4.091750	-2.442537
H	-4.926375	-4.206169	-3.483448
C	-4.188726	-1.522656	-4.025957
H	-4.474028	-2.406919	-4.589728
C	-3.601226	-0.449024	-4.684068
H	-3.427349	-0.500233	-5.755555
C	-3.246933	0.692595	-3.976081
H	-2.804289	1.539455	-4.497265
C	-3.456787	0.780460	-2.597753
C	-3.140925	2.073312	-1.874364
H	-3.338342	1.934648	-0.806367
C	-4.057175	3.193126	-2.368751
H	-3.866204	4.118820	-1.815175
H	-5.114411	2.935711	-2.243305
H	-3.892661	3.404266	-3.431757
C	-1.675176	2.465014	-2.001600
H	-1.391178	2.636856	-3.046835
H	-1.009897	1.692539	-1.588488
H	-1.478894	3.392071	-1.455498
C	-5.338423	-0.027918	0.144170
C	-5.007025	0.194792	1.438400
N	-3.625038	0.088138	1.519269
C	-2.875520	0.435937	2.694530
C	-2.266393	-0.578198	3.454672
C	-2.362376	-2.037902	3.048128
H	-2.063039	-2.084522	1.987654
C	-3.786770	-2.588987	3.158276
H	-3.811260	-3.632211	2.825333
H	-4.508767	-2.034397	2.553896
H	-4.131872	-2.567480	4.198868
C	-1.415465	-2.941425	3.825476
H	-1.680125	-2.980242	4.889330

H	-0.374906	-2.617323	3.740954
H	-1.470109	-3.961460	3.436271
C	-1.535766	-0.175302	4.574164
H	-1.042654	-0.925086	5.184755
C	-1.405086	1.168382	4.909421
H	-0.820694	1.452907	5.780648
C	-2.000707	2.144792	4.125672
H	-1.870006	3.196670	4.374795
C	-2.746445	1.799462	2.995460
C	-3.332459	2.893599	2.123764
H	-3.886030	2.428416	1.300204
C	-4.320252	3.761453	2.900202
H	-3.829179	4.281521	3.730248
H	-5.136271	3.166473	3.323375
H	-4.760428	4.525011	2.250622
C	-2.225460	3.738855	1.495554
H	-1.681047	4.308133	2.258267
H	-2.642969	4.460615	0.783023
H	-1.496392	3.112006	0.969357
Ni	-1.206765	-0.612978	-0.178533
C	3.040213	0.678252	-0.359596
N	3.302830	2.019264	-0.438032
C	2.331252	3.033775	-0.721425
C	1.946858	3.242306	-2.054430
C	2.455147	2.376414	-3.189844
H	3.216591	1.698469	-2.785946
C	3.116042	3.209123	-4.285279
H	2.399634	3.886443	-4.763190
H	3.522962	2.561009	-5.068396
H	3.935175	3.819304	-3.891632
C	1.336537	1.504840	-3.761102
H	0.564653	2.117014	-4.243485
H	0.851704	0.914882	-2.971543
H	1.730966	0.812327	-4.513330
C	1.066305	4.296253	-2.309378
H	0.746619	4.484867	-3.332634
C	0.586952	5.094007	-1.277797
H	-0.101920	5.906415	-1.496685
C	0.968737	4.846561	0.035739
H	0.575378	5.467124	0.837894
C	1.851939	3.810648	0.346754
C	2.309852	3.584715	1.775293
H	2.724609	2.569978	1.837069
C	1.173123	3.665397	2.788931
H	0.761788	4.679978	2.859458
H	1.541317	3.399000	3.785758
H	0.364425	2.976542	2.535050
C	3.412033	4.579495	2.147636
H	3.031487	5.606891	2.110457
H	4.270931	4.523989	1.472002
H	3.774398	4.395118	3.164299
C	4.635061	2.307196	-0.187214
C	5.242243	1.125895	0.074851
N	4.264962	0.149415	-0.038839
C	4.512044	-1.228717	0.272495
C	5.240000	-2.001274	-0.651246

C	5.572992	-1.453686	-2.026374
H	5.863298	-0.399793	-1.912030
C	6.727788	-2.175196	-2.707709
H	6.461434	-3.204054	-2.972875
H	7.619382	-2.214366	-2.074524
H	6.996042	-1.666240	-3.637912
C	4.323955	-1.484212	-2.913278
H	3.985010	-2.515839	-3.065232
H	4.529643	-1.046418	-3.896731
H	3.494400	-0.927491	-2.462395
C	5.565126	-3.307330	-0.280859
H	6.127494	-3.934639	-0.966383
C	5.177255	-3.816544	0.954806
H	5.444345	-4.834476	1.226753
C	4.429814	-3.040192	1.827967
H	4.101774	-3.456066	2.777949
C	4.070340	-1.727907	1.506287
C	3.251610	-0.908591	2.481298
H	2.899547	-0.007043	1.969697
C	1.997099	-1.656139	2.923372
H	2.234050	-2.560969	3.497426
H	1.392942	-1.951337	2.057595
H	1.377961	-1.003328	3.547946
C	4.100199	-0.469400	3.673327
H	4.471925	-1.332036	4.239110
H	3.508129	0.147556	4.357599
H	4.968707	0.118363	3.355648
H	6.261739	0.883512	0.331436
H	5.018996	3.313850	-0.236084
H	-6.294001	-0.023106	-0.357144
H	-5.611930	0.413329	2.305243

3a

H_{Bz} = –1524,11232

G_{THF} = –1524,80442

Ni	0.158584	-0.006639	1.464490
C	-0.589561	1.210430	3.076036
H	-0.595505	2.293495	3.075307
C	0.482426	0.375106	3.500440
H	1.433142	0.713040	3.895505
C	0.079538	-0.990006	3.346068
H	0.667779	-1.859967	3.609423
C	-1.229957	-0.991267	2.801179
H	-1.817222	-1.865225	2.547900
C	-1.634975	0.363160	2.618059
H	-2.575857	0.686256	2.186346
C	0.066529	0.070551	-0.396721
N	1.148560	0.181865	-1.238041
C	2.482834	0.076790	-0.738266
C	3.164267	1.245960	-0.370947
C	2.532601	2.617331	-0.488581
H	1.561019	2.503186	-0.985185
C	2.267049	3.204356	0.897642
H	1.609609	2.547634	1.480683
H	1.786858	4.185433	0.817963

H	3.200765	3.334395	1.457101
C	3.384605	3.557988	-1.337191
H	4.363892	3.736022	-0.879112
H	2.895411	4.530826	-1.446539
H	3.558806	3.152577	-2.338981
C	4.450172	1.102360	0.156427
H	5.003367	1.990540	0.454489
C	5.021473	-0.154207	0.318310
H	6.021868	-0.244498	0.732857
C	4.313507	-1.296800	-0.037174
H	4.760966	-2.278013	0.107698
C	3.025072	-1.207558	-0.568067
C	2.249072	-2.462785	-0.912152
H	1.293268	-2.162297	-1.357752
C	1.924717	-3.265871	0.347120
H	2.838304	-3.593945	0.856395
H	1.346165	-4.161367	0.093947
H	1.337465	-2.665029	1.053471
C	2.989059	-3.315751	-1.939802
H	3.201828	-2.752444	-2.854016
H	2.392988	-4.192142	-2.214280
H	3.944656	-3.680482	-1.546761
C	0.772541	0.282841	-2.567416
H	1.496525	0.374868	-3.362107
C	-0.581764	0.237363	-2.586809
H	-1.285634	0.279954	-3.403469
N	-0.998843	0.097334	-1.267261
C	-2.362891	-0.023675	-0.846248
C	-3.156632	1.134220	-0.815119
C	-2.574427	2.493018	-1.154160
H	-1.821056	2.350840	-1.940521
C	-1.852101	3.074805	0.064383
H	-1.385409	4.034568	-0.184018
H	-1.077287	2.397768	0.442852
H	-2.562378	3.248930	0.881963
C	-3.608028	3.478644	-1.685973
H	-4.317383	3.781715	-0.908289
H	-4.183893	3.064215	-2.519263
H	-3.115161	4.390306	-2.035782
C	-4.480105	0.999184	-0.387221
H	-5.122188	1.874705	-0.351236
C	-4.979788	-0.236171	0.003523
H	-6.009740	-0.321567	0.339234
C	-4.168004	-1.364595	-0.028003
H	-4.573650	-2.321038	0.287403
C	-2.840614	-1.289152	-0.455957
C	-1.951770	-2.519387	-0.508152
H	-0.982883	-2.228064	-0.075014
C	-2.482183	-3.688927	0.311368
H	-3.389107	-4.117896	-0.130759
H	-2.717138	-3.405275	1.341457
H	-1.735909	-4.487883	0.348534
C	-1.710465	-2.980042	-1.948481
H	-1.040858	-3.847625	-1.964819
H	-1.255697	-2.202215	-2.567662
H	-2.652290	-3.280288	-2.422263

3a-oxidized (Ni(II))
G_{THF} = –1524,649847

Ni	-0.74028300	-0.06721600	1.19499600
C	0.19075700	-0.84448000	2.93919300
H	0.63154300	-1.82756500	3.02869400
C	-1.12998400	-0.47226200	3.23261900
H	-1.91406300	-1.13034000	3.58791300
C	-1.29138600	0.94751800	2.98501200
H	-2.21269100	1.50133200	3.11811300
C	-0.08234300	1.44983500	2.50171000
H	0.13466000	2.46728000	2.20477700
C	0.82022200	0.31876400	2.38378900
H	1.83305200	0.36115200	1.99488100
C	0.02071300	-0.02492200	-0.50278800
N	-1.04517900	0.02135100	-1.33232100
C	-2.26393600	0.01308200	-0.56253800
C	-2.53373200	-1.18041500	0.17289200
C	-1.93362400	-2.52717600	-0.21317500
H	-0.95742000	-2.35269000	-0.68116500
C	-1.71555000	-3.45511600	0.97407800
H	-1.05229900	-3.00350200	1.71873000
H	-1.26100900	-4.39182200	0.64134800
H	-2.65528100	-3.72014300	1.46966700
C	-2.84189800	-3.17823500	-1.26105000
H	-3.83472300	-3.38557200	-0.84817900
H	-2.41681200	-4.12729400	-1.59911300
H	-2.97433700	-2.53718300	-2.13837800
C	-3.66845300	-1.13601700	1.01961200
H	-3.93014900	-2.02393800	1.58840800
C	-4.48083200	-0.02148300	1.07565800
H	-5.35769200	-0.02679800	1.71604300
C	-4.21060400	1.10418500	0.28685800
H	-4.88020100	1.95760700	0.33297100
C	-3.09594600	1.15783900	-0.53939500
C	-2.72899000	2.40001400	-1.32232600
H	-2.28135500	2.07905800	-2.27269300
C	-1.66311600	3.19407400	-0.56024700
H	-2.05994600	3.54025300	0.40140800
H	-1.35244400	4.07307800	-1.13221800
H	-0.77403100	2.58863300	-0.35310600
C	-3.92609200	3.28180400	-1.65079500
H	-4.72992700	2.72159500	-2.13609300
H	-3.62481100	4.08760900	-2.32412300
H	-4.33835000	3.75609200	-0.75435400
C	-0.66106500	0.03486200	-2.66091300
H	-1.35869300	0.05095500	-3.48336400
C	0.69917600	0.02262700	-2.63750200
H	1.42048600	0.03000300	-3.43999400
N	1.10692400	-0.00872300	-1.30287900
C	2.47824200	-0.02732700	-0.85486400
C	3.17188500	-1.24617100	-0.89916500
C	2.47822000	-2.53849800	-1.27893300
H	1.68664800	-2.30337400	-2.00386100
C	1.80439600	-3.13415600	-0.03777900

H	1.24538800	-4.04050300	-0.29336600
H	1.10948800	-2.42589200	0.43161400
H	2.55571200	-3.40654900	0.71199700
C	3.40473700	-3.55864000	-1.92893300
H	4.14246200	-3.94800500	-1.22026800
H	3.94805500	-3.13589500	-2.77838800
H	2.83027900	-4.41576200	-2.28947300
C	4.50625500	-1.23419500	-0.48554200
H	5.07924600	-2.15605300	-0.50910800
C	5.10545400	-0.06362200	-0.03903100
H	6.14455100	-0.07525400	0.27564800
C	4.38080600	1.12122100	0.02057200
H	4.85900300	2.02348900	0.38945400
C	3.04562000	1.17088000	-0.38588300
C	2.24373400	2.45636900	-0.32319900
H	1.22393200	2.18244000	-0.01769300
C	2.76522800	3.44605300	0.71171600
H	3.72441000	3.88181100	0.41495300
H	2.89918200	2.98575500	1.69597700
H	2.06166000	4.27708500	0.82083200
C	2.14900600	3.12388500	-1.69705100
H	1.56892700	4.04978500	-1.63632500
H	1.66682700	2.47963700	-2.43889600
H	3.14521000	3.37939200	-2.07247800

3b

G_{THF} = –1526,00824

Ni	-0.19899900	-0.11486000	1.41027600
C	0.69326200	-1.13319300	3.09069200
H	0.91914600	-2.19131800	3.12938000
C	-0.54456700	-0.53051300	3.44535300
H	-1.42773900	-1.05088500	3.79635200
C	-0.41969800	0.88371400	3.27465400
H	-1.18847000	1.61813400	3.48077600
C	0.89010400	1.14811800	2.79658000
H	1.29035000	2.12377700	2.54683300
C	1.57039500	-0.09745800	2.66287100
H	2.57826800	-0.23133600	2.28557700
C	-0.07357200	-0.18470900	-0.44332800
N	-1.15193000	-0.41909500	-1.25946400
C	-2.47580700	-0.20943900	-0.76191000
C	-3.28353000	-1.33399400	-0.51736500
C	-2.75107700	-2.72935700	-0.77409900
H	-2.05620600	-2.66030800	-1.62243300
C	-1.94180800	-3.22715900	0.42599300
H	-1.11726000	-2.54563700	0.66974700
H	-1.52107300	-4.21949300	0.22771000
H	-2.57966300	-3.30416700	1.31462500
C	-3.83987600	-3.73060100	-1.14124100
H	-4.49740200	-3.94483000	-0.29147800
H	-3.39396600	-4.68313700	-1.44272600
H	-4.46699200	-3.37454900	-1.96467900
C	-4.55845800	-1.12023500	0.01027100
H	-5.20341400	-1.97018100	0.21634200
C	-5.01362600	0.16624800	0.27895600

H	-6.00862100	0.31256400	0.69093800
C	-4.20366600	1.26388400	0.01858100
H	-4.56934900	2.26806200	0.22582700
C	-2.91849400	1.10168600	-0.50588600
C	-2.06880500	2.32299500	-0.80320300
H	-1.08705400	1.98515500	-1.15599200
C	-1.81523400	3.16372100	0.44597300
H	-2.75104600	3.53688500	0.87755400
H	-1.19492200	4.03396900	0.20214900
H	-1.29433400	2.57379400	1.20939600
C	-2.69749500	3.16634900	-1.91272100
H	-2.87270300	2.57981600	-2.82107200
H	-2.05020000	4.01043700	-2.17445900
H	-3.66377000	3.57749300	-1.59909900
C	-0.83460200	-0.21253500	-2.67663900
H	-1.17335600	0.78380100	-3.00615400
C	0.68219700	-0.32450200	-2.67320900
H	1.17637700	0.38255200	-3.34736900
N	0.99557200	-0.02968900	-1.27048500
C	2.33819400	0.08249800	-0.80283500
C	3.12340100	-1.07332900	-0.68747300
C	2.55739900	-2.46156300	-0.90876200
H	1.52062000	-2.36531700	-1.25297000
C	2.49926700	-3.21896500	0.41710300
H	2.05461100	-4.21054800	0.28069500
H	1.89216900	-2.66683000	1.14173500
H	3.49966100	-3.35538900	0.84382700
C	3.34013500	-3.23807100	-1.96413000
H	4.37766100	-3.39953700	-1.65101600
H	3.36739100	-2.70802400	-2.92205500
H	2.89509900	-4.22354800	-2.13415500
C	4.45009600	-0.92723700	-0.27034000
H	5.07725100	-1.81180100	-0.17401400
C	4.96284000	0.32216200	0.04534700
H	5.99530100	0.41973000	0.37061600
C	4.14852900	1.44972700	-0.02188800
H	4.55314000	2.41490600	0.26867800
C	2.82039100	1.35690900	-0.43986900
C	1.89753800	2.56237600	-0.44571400
H	0.95752100	2.22540800	0.02037600
C	2.42245000	3.72780100	0.38291500
H	3.29247000	4.20357100	-0.08527900
H	2.71746800	3.41960800	1.39074000
H	1.65011300	4.49700400	0.48106600
C	1.56905600	3.04857000	-1.85823400
H	0.88164500	3.90122800	-1.81847200
H	1.09143900	2.27378900	-2.46391500
H	2.47442400	3.37511900	-2.38284900
H	1.02116800	-1.33611100	-2.94499700
H	-1.32557300	-0.96028600	-3.30795200

3b-oxidized (Ni(II))

G_{THF} = –1525,85854

Ni	-0.71800600	-0.07046200	1.15135300

C	0.29275400	-0.94671700	2.80331100
H	0.80653700	-1.89798100	2.78978500
C	-1.04024300	-0.70691900	3.15397600
H	-1.76916700	-1.45121400	3.45101800
C	-1.31050600	0.71069400	3.02333100
H	-2.27089400	1.17899500	3.20221000
C	-0.14261900	1.35233000	2.59834400
H	-0.01563300	2.40784700	2.40096500
C	0.83320800	0.31447000	2.36193500
H	1.83647100	0.46481400	1.97487300
C	0.00825100	-0.01545500	-0.56208100
N	-1.08923400	0.16876200	-1.31828100
C	-2.25763500	0.10007700	-0.47915200
C	-2.54253600	-1.13746400	0.17923700
C	-1.97936700	-2.46807800	-0.30353600
H	-1.02051200	-2.28472200	-0.80757100
C	-1.71828200	-3.46228300	0.82096000
H	-1.00358700	-3.06697400	1.54913800
H	-1.30660600	-4.39012100	0.41558100
H	-2.63665600	-3.73023500	1.35357400
C	-2.94895800	-3.05838900	-1.33336600
H	-3.90971100	-3.30422700	-0.86899900
H	-2.54260900	-3.97831400	-1.76311600
H	-3.15199400	-2.36088700	-2.15250200
C	-3.65871500	-1.13664400	1.05328200
H	-3.92557200	-2.05994900	1.56073900
C	-4.44784200	-0.01692200	1.20887400
H	-5.31254200	-0.05577600	1.86484400
C	-4.17091800	1.16308000	0.50301900
H	-4.82277200	2.02255800	0.62760800
C	-3.07685600	1.25404200	-0.34270000
C	-2.70331300	2.53721000	-1.05088400
H	-2.24437700	2.26286300	-2.01080700
C	-1.64353000	3.28681400	-0.23742800
H	-2.05570800	3.60095500	0.72901300
H	-1.31071300	4.18474700	-0.76745300
H	-0.76745200	2.66117300	-0.03630500
C	-3.89618700	3.43790400	-1.34268600
H	-4.69304300	2.90818900	-1.87164900
H	-3.58659500	4.28370100	-1.96112900
H	-4.32156900	3.85719200	-0.42524900
C	-0.84430400	-0.16191900	-2.72966800
H	-1.16529800	-1.19076200	-2.94684500
C	0.67820100	-0.00838100	-2.77017100
H	1.17729100	-0.74852000	-3.39978900
N	1.07028500	-0.19043400	-1.34884900
C	2.43725500	-0.09519500	-0.92211200
C	3.18069200	-1.28243200	-0.80205400
C	2.54696400	-2.62725700	-1.09431600
H	1.80455200	-2.47613100	-1.89229300
C	1.78872800	-3.13110500	0.13476800
H	1.30092900	-4.08990800	-0.07019700
H	1.01978300	-2.41773700	0.45447600
H	2.47681300	-3.28103200	0.97498500
C	3.54234000	-3.67412100	-1.57857000
H	4.22761100	-3.97947100	-0.78152300

H	4.14428800	-3.31228900	-2.41666000
H	3.01636400	-4.57514600	-1.90472200
C	4.50017400	-1.17457300	-0.36195200
H	5.10286500	-2.07135800	-0.25193900
C	5.05803300	0.06630500	-0.07125600
H	6.08770700	0.12862200	0.26807200
C	4.30983300	1.22526300	-0.22736200
H	4.76238900	2.19199400	-0.01911900
C	2.98225400	1.17354000	-0.66156400
C	2.20992800	2.46106000	-0.86905900
H	1.16098200	2.20994800	-1.07734900
C	2.22545700	3.34626500	0.37458900
H	3.23619400	3.69644000	0.60613100
H	1.85461900	2.81468500	1.25613800
H	1.60220000	4.23343400	0.22585800
C	2.75425800	3.22850500	-2.07554700
H	2.16783200	4.13422700	-2.25820600
H	2.74103200	2.62488100	-2.98858100
H	3.79188500	3.53461400	-1.90617000
H	-1.36643600	0.51826400	-3.40648300
H	0.98389900	0.99363200	-3.10076700

4a

H_{Bz} = –1677,72778

Ni	-0.259304	-0.086948	-1.233944
C	0.334641	0.536760	-3.129099
H	0.257103	1.575395	-3.433063
C	-0.703411	-0.438148	-3.226571
H	-1.687752	-0.281157	-3.652060
C	-0.160579	-1.702832	-2.798927
C	-0.708691	-2.998008	-2.702974
H	-1.733136	-3.181798	-3.020722
C	0.072036	-4.027484	-2.208434
H	-0.340850	-5.031111	-2.141948
C	1.396069	-3.796401	-1.780661
H	1.982637	-4.625437	-1.391102
C	1.958913	-2.533216	-1.846647
H	2.979035	-2.357801	-1.508177
C	1.199353	-1.468074	-2.367491
C	1.474286	-0.070057	-2.534334
H	2.416850	0.416082	-2.309237
C	-0.141187	0.390669	0.581677
N	0.902597	0.800671	1.371396
C	2.268958	0.776089	0.944500
C	2.720730	1.773780	0.060030
C	1.782983	2.857263	-0.438892
H	0.822617	2.367128	-0.657524
C	2.245521	3.512167	-1.733696
H	1.461480	4.173204	-2.117008
H	2.466546	2.772983	-2.510304
H	3.141323	4.127241	-1.588708
C	1.536330	3.921557	0.632643
H	1.147174	3.491563	1.560538
H	0.807412	4.659960	0.279228
H	2.462593	4.456150	0.873470

C	4.056629	1.715222	-0.337961
H	4.441473	2.463837	-1.024549
C	4.899639	0.703894	0.115356
H	5.935395	0.677791	-0.212072
C	4.419900	-0.273244	0.974563
H	5.079959	-1.068213	1.315728
C	3.089412	-0.262208	1.405843
C	2.591828	-1.349157	2.338828
H	1.521945	-1.189683	2.519872
C	2.735226	-2.735885	1.717005
H	2.155306	-2.818347	0.793381
H	2.382546	-3.504396	2.412636
H	3.780604	-2.964089	1.478537
C	3.310609	-1.276168	3.686139
H	4.385576	-1.455403	3.570371
H	2.923938	-2.033410	4.375420
H	3.194239	-0.296362	4.160479
C	0.478547	1.158325	2.644532
H	1.170669	1.497345	3.399830
C	-0.865293	0.983944	2.671398
H	-1.593285	1.150436	3.449888
N	-1.223101	0.506952	1.420964
C	-2.559294	0.315215	0.944543
C	-3.162562	1.365398	0.235797
C	-2.476594	2.700026	0.023063
H	-1.468746	2.637290	0.449294
C	-2.314731	3.016451	-1.462012
H	-1.774995	2.210585	-1.973197
H	-1.748304	3.945078	-1.596331
H	-3.285113	3.145410	-1.954987
C	-3.218493	3.819074	0.751628
H	-4.234631	3.943128	0.360256
H	-2.698019	4.774357	0.626616
H	-3.300881	3.618079	1.824792
C	-4.446957	1.145064	-0.267722
H	-4.945435	1.940154	-0.818348
C	-5.089404	-0.071384	-0.075280
H	-6.087047	-0.225841	-0.477525
C	-4.463169	-1.094970	0.628741
H	-4.976629	-2.042836	0.767284
C	-3.181592	-0.925782	1.155830
C	-2.458076	-2.050675	1.868714
H	-1.742355	-1.594373	2.565970
C	-1.654276	-2.883746	0.866888
H	-2.322200	-3.371902	0.147744
H	-1.081999	-3.666808	1.376535
H	-0.951214	-2.270950	0.289656
C	-3.392351	-2.939840	2.682040
H	-4.021042	-2.361702	3.366417
H	-2.814235	-3.654335	3.275294
H	-4.054217	-3.527799	2.037189

(IPr)Ni

H_{Bz} = –1330,57218

Ni	0.000510	-0.137862	-1.860193

C	-0.011584	0.051697	-0.113744
N	-1.089473	0.206667	0.738324
C	-2.447100	0.052785	0.316310
C	-3.230281	1.201680	0.115108
C	-2.639600	2.585401	0.300194
H	-1.902495	2.524801	1.112942
C	-1.888525	3.020091	-0.960111
H	-1.119382	2.289648	-1.245157
H	-1.405854	3.991967	-0.809056
H	-2.582219	3.116271	-1.803459
C	-3.673361	3.632392	0.697215
H	-4.368049	3.846495	-0.122497
H	-3.179966	4.576253	0.947300
H	-4.266132	3.320134	1.562706
C	-4.547349	1.014760	-0.306767
H	-5.182600	1.879963	-0.476363
C	-5.058319	-0.262341	-0.515753
H	-6.085793	-0.384802	-0.848446
C	-4.264686	-1.379513	-0.293383
H	-4.674017	-2.375309	-0.451231
C	-2.941265	-1.247492	0.135355
C	-2.097296	-2.479614	0.391820
H	-1.130279	-2.154930	0.794928
C	-1.813706	-3.233122	-0.905266
H	-2.741423	-3.571392	-1.381540
H	-1.194611	-4.116843	-0.712873
H	-1.279334	-2.585644	-1.614560
C	-2.745048	-3.390933	1.432254
H	-2.933279	-2.861067	2.371600
H	-2.097914	-4.246669	1.650818
H	-3.703395	-3.788392	1.079564
C	-0.684982	0.341520	2.058834
H	-1.397110	0.457164	2.861181
C	0.668527	0.283552	2.062733
H	1.384747	0.363575	2.866350
N	1.067323	0.095900	0.747405
C	2.429320	0.027796	0.315000
C	3.146971	1.224635	0.184901
C	2.489615	2.578679	0.359938
H	1.493049	2.428055	0.793190
C	2.289211	3.236480	-1.005769
H	1.780789	4.201216	-0.903692
H	1.684113	2.597356	-1.660368
H	3.250463	3.414273	-1.501822
C	3.273026	3.484264	1.305257
H	4.271865	3.708483	0.914802
H	3.400662	3.026211	2.291388
H	2.757341	4.440114	1.440874
C	4.487059	1.136388	-0.202251
H	5.067563	2.049263	-0.319159
C	5.076207	-0.094109	-0.456166
H	6.119655	-0.143915	-0.755781
C	4.330530	-1.264621	-0.347703
H	4.797195	-2.219201	-0.574437
C	2.989809	-1.232195	0.034883
C	2.154589	-2.493328	0.137092

H	1.148585	-2.231225	-0.223157
C	2.662611	-3.621965	-0.749708
H	3.622983	-4.019866	-0.402224
H	2.787809	-3.294491	-1.786389
H	1.952565	-4.454589	-0.743171
C	2.026430	-2.965875	1.585956
H	1.372866	-3.843542	1.648500
H	1.603192	-2.191740	2.233368
H	3.004360	-3.247537	1.993786

(IPr)Ni(Cp)(Cl)

H_{Bz} = –1984,37313

Ni	-0.083919	-0.237444	1.532269
C	-0.800740	1.562183	2.189457
H	-0.844280	2.450909	1.568852
C	0.274696	1.202214	3.054427
H	1.224717	1.707898	3.156639
C	-0.084233	-0.039496	3.618439
H	0.550530	-0.665766	4.231414
C	-1.444852	-0.356807	3.249455
H	-1.972923	-1.256763	3.536565
C	-1.896734	0.631784	2.381953
H	-2.853658	0.669144	1.875987
C	0.039294	-0.049979	-0.324725
N	1.164472	0.060326	-1.091044
C	2.486983	0.295972	-0.576139
C	2.788310	1.584155	-0.093950
C	1.769932	2.707959	-0.144011
H	0.792589	2.269923	0.090792
C	2.024768	3.810697	0.875695
H	2.166792	3.414404	1.885216
H	1.175305	4.500842	0.901562
H	2.909669	4.405441	0.623246
C	1.681194	3.308613	-1.549686
H	2.645856	3.730214	-1.854208
H	0.941953	4.117580	-1.576470
H	1.386902	2.566414	-2.298548
C	4.080871	1.804382	0.382266
H	4.348540	2.784744	0.766082
C	5.032553	0.790556	0.366387
H	6.034339	0.983692	0.740810
C	4.707559	-0.464379	-0.125767
H	5.457931	-1.250747	-0.133566
C	3.424683	-0.746529	-0.604329
C	3.107554	-2.115446	-1.170042
H	2.016422	-2.228918	-1.175861
C	3.672537	-3.246953	-0.316498
H	4.767404	-3.278948	-0.356573
H	3.310736	-4.210345	-0.688359
H	3.360840	-3.149261	0.725389
C	3.631019	-2.235689	-2.603307
H	3.218641	-1.471670	-3.269892
H	3.383057	-3.214949	-3.025197
H	4.722071	-2.131078	-2.627359
C	0.860660	-0.016871	-2.442464

H	1.625528	0.052545	-3.199451
C	-0.483102	-0.159843	-2.535212
H	-1.136067	-0.266353	-3.387056
N	-0.968351	-0.166039	-1.237247
C	-2.361399	-0.166471	-0.896067
C	-3.032734	1.068219	-0.892936
C	-2.310529	2.365989	-1.196008
H	-1.244826	2.206775	-0.979141
C	-2.784814	3.523148	-0.320550
H	-2.111608	4.380176	-0.429395
H	-2.818601	3.249784	0.738516
H	-3.785511	3.865834	-0.605294
C	-2.434633	2.736890	-2.674897
H	-3.486179	2.879438	-2.949169
H	-2.022397	1.960933	-3.326457
H	-1.904371	3.670258	-2.890765
C	-4.393679	1.057603	-0.582906
H	-4.946555	1.993489	-0.573141
C	-5.044492	-0.131250	-0.269715
H	-6.103869	-0.119670	-0.028234
C	-4.343265	-1.329640	-0.248949
H	-4.857019	-2.251692	0.012810
C	-2.980267	-1.376530	-0.557514
C	-2.228577	-2.689825	-0.537500
H	-1.154312	-2.475030	-0.580222
C	-2.465880	-3.453924	0.760253
H	-3.509823	-3.772797	0.863676
H	-2.197469	-2.838730	1.624011
H	-1.837753	-4.348121	0.791914
C	-2.598617	-3.532103	-1.758292
H	-2.036997	-4.471272	-1.764511
H	-2.383810	-3.005270	-2.694345
H	-3.666183	-3.782211	-1.758221
Cl	0.849816	-2.253777	1.452427

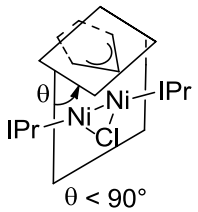
X-ray crystallography

Low-temperature diffraction data (ω-scans) were collected on either a Rigaku R-AXIS RAPID diffractometer coupled to a R-AXIS RAPID imaging plate detector with Mo K_α radiation (λ = 0.71073 Å) for the structure of **1a** or a Rigaku MicroMax-007HF diffractometer coupled to a Saturn994+ CCD detector with Cu K_α (λ = 1.54178 Å) for the structures of **2b**, **3a**, **4a** and **4b**. All structures were solved by direct methods using SHELXS[4] and refined against F^2 on all data by full-matrix least squares with SHELXL-97[5] using established refinement techniques.[6] All hydrogen atoms were included into the model at geometrically calculated positions and refined using a riding model. The isotropic displacement parameters of all hydrogen atoms were fixed to 1.2 times the U value of the atoms they are linked to (1.5 times for methyl groups).

Canting of central carbon atoms of the bridging ligands are canted towards the Ni-Ni-Cl plane

As noted in the main text a feature of the binding of the bridging Cp and indenyl ligands in **1a** and **2b** is that the central carbon atoms of the bridging Cp or indenyl ligand is canted towards the Ni-Ni-Cl plane. As a result, the dihedral angle between the bridging Cp or indenyl plane and the plane containing the two Ni atoms and the two terminal carbon atoms of the bridging Cp or indenyl ligand (the dihedral angle θ in Figure S33 below) is significantly less than 90°

Figure S33: Values of the dihedral angle formed between the Cp or indenyl plane and the plane containing the two Ni centers and the two terminal carbon atoms of the bridging Cp or indenyl ligand in **1a** or **2b**.



Compound	θ
1a	69.9°
2b	67.7°

X-Ray Data for 1a (CCDC 939469)

Compound **1a** crystallizes in the monoclinic space group *P2(1)/n* with one molecule in the asymmetric unit. Two of the iso-propyl groups of the ligand were disordered over two sites. The coordinates for the hydrogen atoms bound to C1, C2, C3, C4, and C5 were taken from the difference Fourier synthesis and the hydrogen atoms were subsequently refined semi-freely with the help of a distance restraint.

Table S10: Crystal data and structure refinement for **1a**

Empirical formula	C59 H77 Cl N4 Ni2	
Formula weight	995.12	
Temperature	150(2) K	
Wavelength	0.71075 Å	
Crystal system	Monoclinic	
Space group	P2(1)/n	
Unit cell dimensions	a = 20.5396(4) Å	α= 90°
	b = 12.1359(2) Å	β= 112.504(8)°
	c = 23.7443(17) Å	γ = 90°
Volume	5468.0(4) $Å^3$	
Z	4	
Density (calculated)	1.209 Mg/m^3	
Absorption coefficient	0.777 mm^{-1}	
F(000)	2128	
Crystal size	0.25 x 0.20 x 0.10 mm^3	
Theta range for data collection	3.07 to 30.51°.	
Index ranges	-29<=h<=29, -17<=k<=17, -33<=l<=33	
Reflections collected	163825	
Independent reflections	16595 [R(int) = 0.0703]	
Completeness to theta = 30.51°	99.4 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.9263 and 0.8294	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	16595 / 520 / 725	
Goodness-of-fit on F^2	1.047	
Final R indices [I>2sigma(I)]	R1 = 0.0435, wR2 = 0.0854	
R indices (all data)	R1 = 0.0639, wR2 = 0.0930	
Largest diff. peak and hole	0.502 and -0.448 $e.Å^{-3}$	

Table S11: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for **1a**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Cl(1)	8076(1)	1507(1)	10245(1)	31(1)
C(1)	6246(1)	773(2)	9501(1)	32(1)
C(2)	6100(1)	1602(2)	9030(1)	32(1)
C(3)	5830(1)	2547(2)	9251(1)	36(1)
C(4)	5802(1)	2314(2)	9801(1)	36(1)
C(5)	6049(1)	1192(2)	9981(1)	33(1)
Ni(1)	7129(1)	1676(1)	9399(1)	22(1)
C(11)	7361(1)	2072(1)	8738(1)	19(1)
N(1)	7251(1)	1445(1)	8228(1)	21(1)

C(21)	6917(1)	381(1)	8121(1)	22(1)
C(22)	7277(1)	-515(1)	8478(1)	25(1)
C(23)	6934(1)	-1532(1)	8360(1)	31(1)
C(27)	8020(1)	-406(1)	8957(1)	30(1)
C(28)	8179(1)	-1186(2)	9495(1)	55(1)
C(29)	8561(1)	-525(3)	8666(1)	62(1)
C(24)	6270(1)	-1652(2)	7908(1)	34(1)
C(25)	5932(1)	-760(2)	7559(1)	30(1)
C(26)	6247(1)	281(1)	7657(1)	26(1)
C(30)	5865(1)	1246(2)	7262(1)	34(1)
C(31)	5096(1)	1325(2)	7199(1)	54(1)
C(32)	5903(1)	1172(2)	6632(1)	53(1)
C(12)	7517(1)	1949(1)	7835(1)	24(1)
C(13)	7798(1)	2910(1)	8093(1)	24(1)
N(2)	7704(1)	2974(1)	8641(1)	19(1)
C(41)	7994(1)	3861(1)	9070(1)	21(1)
C(42)	8708(1)	3802(1)	9453(1)	24(1)
C(47)	9153(1)	2792(2)	9475(1)	30(1)
C(48)	9664(1)	2521(2)	10123(1)	42(1)
C(49)	9567(1)	2927(2)	9062(1)	43(1)
C(43)	8995(1)	4724(2)	9818(1)	30(1)
C(44)	8589(1)	5637(2)	9800(1)	32(1)
C(45)	7878(1)	5647(1)	9437(1)	29(1)
C(46)	7561(1)	4751(1)	9067(1)	23(1)
C(50)	6782(1)	4741(1)	8665(1)	27(1)
C(51)	6334(1)	5413(2)	8928(1)	38(1)
C(52)	6662(1)	5150(2)	8024(1)	39(1)
Ni(2)	7077(1)	1247(1)	10369(1)	23(1)
C(15)	7279(1)	1126(1)	11206(1)	22(1)
N(3)	7665(1)	304(1)	11584(1)	24(1)
C(61)	7887(4)	-672(8)	11365(8)	26(1)
C(62)	8598(4)	-770(7)	11462(6)	29(1)
C(67)	9114(5)	181(8)	11707(5)	32(1)
C(68)	9528(9)	62(15)	12394(6)	50(2)
C(69)	9632(7)	296(11)	11385(7)	47(2)
C(63)	8815(4)	-1768(7)	11295(6)	36(1)
C(64)	8342(5)	-2612(6)	11054(5)	39(1)
C(65)	7646(5)	-2487(6)	10961(5)	40(1)
C(66)	7392(4)	-1513(8)	11111(6)	33(1)
C(70)	6619(4)	-1401(8)	11016(4)	40(1)
C(71)	6457(8)	-1953(14)	11525(5)	58(2)
C(72)	6135(5)	-1886(11)	10406(4)	61(2)
C(61A)	7985(8)	-653(14)	11420(13)	26(2)
C(62A)	8698(8)	-617(13)	11517(12)	29(2)
C(67A)	9162(10)	357(15)	11797(11)	34(2)
C(68A)	9563(15)	240(30)	12483(10)	46(3)
C(69A)	9676(12)	550(20)	11476(12)	44(3)
C(63A)	8978(9)	-1590(13)	11385(10)	37(2)
C(64A)	8567(10)	-2477(13)	11126(9)	43(3)
C(65A)	7856(10)	-2443(12)	10970(9)	42(2)
C(66A)	7533(8)	-1526(14)	11113(11)	36(2)
C(70A)	6744(8)	-1458(14)	10936(8)	44(3)
C(71A)	6522(14)	-1800(30)	11449(11)	63(4)
C(72A)	6323(11)	-2067(18)	10343(8)	68(5)
C(16)	7752(1)	488(2)	12186(1)	28(1)
C(17)	7424(1)	1435(2)	12195(1)	27(1)

N(4)	7135(1)	1815(1)	11596(1)	23(1)
C(81)	6677(1)	2757(1)	11409(1)	25(1)
C(82)	6916(1)	3712(2)	11222(1)	32(1)
C(87)	7643(1)	3809(2)	11210(1)	36(1)
C(88)	8105(1)	4563(2)	11720(1)	52(1)
C(89)	7612(1)	4219(2)	10592(1)	46(1)
C(83)	6452(1)	4603(2)	11050(1)	45(1)
C(84)	5784(1)	4534(2)	11057(1)	51(1)
C(85)	5564(1)	3573(2)	11240(1)	43(1)
C(86)	6001(1)	2661(2)	11421(1)	30(1)
C(90)	5755(1)	1625(2)	11634(1)	35(1)
C(91)	5042(1)	1220(2)	11181(1)	55(1)
C(92)	5725(1)	1804(2)	12261(1)	46(1)

Table S12: Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **1a**. 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}
Cl(1)	26(1)	46(1)	23(1)	2(1)	11(1)	-1(1)
C(1)	31(1)	34(1)	30(1)	-2(1)	10(1)	-12(1)
C(2)	27(1)	45(1)	24(1)	0(1)	10(1)	-8(1)
C(3)	26(1)	45(1)	36(1)	10(1)	11(1)	1(1)
C(4)	29(1)	48(1)	34(1)	2(1)	14(1)	2(1)
C(5)	28(1)	44(1)	30(1)	5(1)	13(1)	-6(1)
Ni(1)	26(1)	23(1)	18(1)	-1(1)	11(1)	-3(1)
C(11)	21(1)	18(1)	20(1)	0(1)	7(1)	0(1)
N(1)	25(1)	19(1)	19(1)	-1(1)	10(1)	-2(1)
C(21)	29(1)	19(1)	20(1)	-3(1)	12(1)	-2(1)
C(22)	30(1)	22(1)	25(1)	-1(1)	14(1)	0(1)
C(23)	40(1)	22(1)	35(1)	2(1)	18(1)	0(1)
C(27)	32(1)	25(1)	29(1)	2(1)	8(1)	3(1)
C(28)	61(2)	44(1)	43(1)	17(1)	0(1)	-6(1)
C(29)	33(1)	100(2)	50(1)	-1(1)	15(1)	11(1)
C(24)	41(1)	25(1)	41(1)	-7(1)	22(1)	-10(1)
C(25)	28(1)	32(1)	32(1)	-9(1)	12(1)	-8(1)
C(26)	28(1)	27(1)	24(1)	-4(1)	12(1)	-1(1)
C(30)	33(1)	30(1)	31(1)	0(1)	2(1)	-1(1)
C(31)	35(1)	54(1)	60(2)	6(1)	2(1)	8(1)
C(32)	66(2)	53(1)	30(1)	8(1)	8(1)	-4(1)
C(12)	32(1)	26(1)	20(1)	1(1)	14(1)	0(1)
C(13)	29(1)	25(1)	23(1)	0(1)	15(1)	-2(1)
N(2)	21(1)	18(1)	18(1)	0(1)	8(1)	-1(1)
C(41)	24(1)	20(1)	20(1)	-1(1)	10(1)	-4(1)
C(42)	23(1)	27(1)	25(1)	-1(1)	11(1)	-4(1)
C(47)	21(1)	32(1)	33(1)	1(1)	8(1)	2(1)
C(48)	28(1)	52(1)	40(1)	12(1)	6(1)	3(1)
C(49)	33(1)	56(1)	44(1)	1(1)	20(1)	13(1)
C(43)	26(1)	38(1)	28(1)	-6(1)	10(1)	-10(1)
C(44)	40(1)	28(1)	34(1)	-12(1)	20(1)	-13(1)
C(45)	36(1)	22(1)	36(1)	-5(1)	20(1)	-4(1)
C(46)	27(1)	20(1)	25(1)	2(1)	14(1)	-2(1)
C(50)	25(1)	23(1)	34(1)	2(1)	13(1)	2(1)
C(51)	33(1)	38(1)	49(1)	2(1)	22(1)	7(1)
C(52)	34(1)	49(1)	35(1)	8(1)	13(1)	10(1)
Ni(2)	27(1)	24(1)	20(1)	1(1)	12(1)	-2(1)

C(15)	22(1)	23(1)	25(1)	1(1)	12(1)	-1(1)
N(3)	27(1)	25(1)	22(1)	2(1)	10(1)	1(1)
C(61)	27(2)	23(2)	24(4)	5(2)	5(2)	6(1)
C(62)	37(2)	26(2)	27(3)	1(2)	16(2)	-1(2)
C(67)	29(2)	29(3)	38(3)	1(2)	13(2)	4(2)
C(68)	43(3)	60(5)	43(3)	-8(3)	13(3)	-14(3)
C(69)	37(3)	49(4)	63(5)	10(3)	29(3)	0(3)
C(63)	41(3)	31(3)	45(4)	-1(2)	26(3)	5(2)
C(64)	54(4)	25(2)	45(3)	-4(2)	26(4)	3(2)
C(65)	45(3)	27(2)	44(2)	-4(2)	13(3)	-4(2)
C(66)	37(2)	24(2)	34(2)	2(2)	11(2)	-1(2)
C(70)	29(2)	31(2)	51(3)	8(2)	8(2)	-4(2)
C(71)	50(4)	72(5)	52(3)	13(3)	21(3)	-2(4)
C(72)	48(3)	76(5)	47(2)	13(2)	4(2)	-14(3)
C(61A)	41(4)	22(3)	13(4)	-1(3)	9(5)	0(3)
C(62A)	39(4)	25(4)	28(5)	5(3)	18(4)	11(3)
C(67A)	31(4)	33(5)	44(6)	-1(4)	19(4)	1(3)
C(68A)	34(6)	62(10)	41(5)	2(5)	11(4)	0(6)
C(69A)	33(5)	55(9)	50(7)	13(6)	21(5)	12(5)
C(63A)	51(5)	29(4)	36(5)	2(3)	22(5)	7(3)
C(64A)	64(6)	29(4)	37(5)	-2(3)	21(6)	7(4)
C(65A)	62(6)	25(3)	34(4)	-2(3)	15(6)	-3(4)
C(66A)	42(4)	27(3)	28(4)	8(3)	3(4)	0(3)
C(70A)	46(4)	24(4)	51(5)	7(3)	8(4)	-11(4)
C(71A)	38(5)	76(8)	64(8)	15(7)	8(5)	-18(5)
C(72A)	61(9)	58(7)	53(5)	16(5)	-14(5)	-33(7)
C(16)	30(1)	35(1)	19(1)	3(1)	8(1)	1(1)
C(17)	29(1)	35(1)	18(1)	-2(1)	9(1)	-1(1)
N(4)	24(1)	25(1)	23(1)	0(1)	12(1)	0(1)
C(81)	28(1)	27(1)	23(1)	-1(1)	12(1)	2(1)
C(82)	40(1)	28(1)	35(1)	0(1)	20(1)	2(1)
C(87)	44(1)	28(1)	46(1)	0(1)	28(1)	-4(1)
C(88)	48(1)	59(2)	53(1)	-6(1)	25(1)	-9(1)
C(89)	66(2)	37(1)	51(1)	0(1)	40(1)	-6(1)
C(83)	61(1)	29(1)	54(1)	8(1)	33(1)	10(1)
C(84)	58(1)	43(1)	58(1)	10(1)	30(1)	25(1)
C(85)	37(1)	52(1)	47(1)	4(1)	22(1)	14(1)
C(86)	27(1)	39(1)	28(1)	-1(1)	14(1)	3(1)
C(90)	26(1)	45(1)	36(1)	0(1)	14(1)	-4(1)
C(91)	37(1)	74(2)	51(1)	-5(1)	14(1)	-18(1)
C(92)	42(1)	63(1)	40(1)	5(1)	24(1)	-4(1)

Table S13: Bond lengths (Å) for **1a**

Cl(1)-Ni(2)	2.2037(5)
Cl(1)-Ni(1)	2.2068(5)
C(1)-C(5)	1.440(3)
C(1)-C(2)	1.447(3)
C(1)-Ni(2)	2.1906(18)
C(1)-Ni(1)	2.2093(17)
C(1)-H(1)	1.014(15)
C(2)-C(3)	1.456(3)
C(2)-Ni(1)	1.9567(18)
C(2)-H(2)	0.978(15)
C(3)-C(4)	1.360(3)
C(3)-H(3)	0.9500

C(4)-C(5)	1.458(3)
C(4)-H(4)	0.9500
C(5)-Ni(2)	1.9551(18)
C(5)-H(5)	0.988(15)
Ni(1)-C(11)	1.8703(15)
Ni(1)-Ni(2)	2.4015(3)
C(11)-N(2)	1.3676(19)
C(11)-N(1)	1.3720(19)
N(1)-C(12)	1.3904(19)
N(1)-C(21)	1.439(2)
C(21)-C(26)	1.400(2)
C(21)-C(22)	1.403(2)
C(22)-C(23)	1.394(2)
C(22)-C(27)	1.521(2)
C(23)-C(24)	1.383(3)
C(23)-H(23)	0.9500
C(27)-C(28)	1.522(3)
C(27)-C(29)	1.524(3)
C(27)-H(27)	1.0000
C(28)-H(28A)	0.9800
C(28)-H(28B)	0.9800
C(28)-H(28C)	0.9800
C(29)-H(29A)	0.9800
C(29)-H(29B)	0.9800
C(29)-H(29C)	0.9800
C(24)-C(25)	1.379(3)
C(24)-H(24)	0.9500
C(25)-C(26)	1.398(2)
C(25)-H(25)	0.9500
C(26)-C(30)	1.518(2)
C(30)-C(31)	1.530(3)
C(30)-C(32)	1.532(3)
C(30)-H(30)	1.0000
C(31)-H(31A)	0.9800
C(31)-H(31B)	0.9800
C(31)-H(31C)	0.9800
C(32)-H(32A)	0.9800
C(32)-H(32B)	0.9800
C(32)-H(32C)	0.9800
C(12)-C(13)	1.340(2)
C(12)-H(12)	0.9500
C(13)-N(2)	1.3901(19)
C(13)-H(13)	0.9500
N(2)-C(41)	1.4440(19)
C(41)-C(46)	1.398(2)
C(41)-C(42)	1.401(2)
C(42)-C(43)	1.399(2)
C(42)-C(47)	1.518(2)
C(47)-C(48)	1.530(3)
C(47)-C(49)	1.534(3)
C(47)-H(47)	1.0000
C(48)-H(48A)	0.9800
C(48)-H(48B)	0.9800
C(48)-H(48C)	0.9800
C(49)-H(49A)	0.9800
C(49)-H(49B)	0.9800

C(49)-H(49C)	0.9800
C(43)-C(44)	1.377(3)
C(43)-H(43)	0.9500
C(44)-C(45)	1.384(3)
C(44)-H(44)	0.9500
C(45)-C(46)	1.393(2)
C(45)-H(45)	0.9500
C(46)-C(50)	1.516(2)
C(50)-C(52)	1.530(2)
C(50)-C(51)	1.531(2)
C(50)-H(50)	1.0000
C(51)-H(51A)	0.9800
C(51)-H(51B)	0.9800
C(51)-H(51C)	0.9800
C(52)-H(52A)	0.9800
C(52)-H(52B)	0.9800
C(52)-H(52C)	0.9800
Ni(2)-C(15)	1.8751(16)
C(15)-N(4)	1.362(2)
C(15)-N(3)	1.373(2)
N(3)-C(16)	1.391(2)
N(3)-C(61)	1.435(6)
N(3)-C(61A)	1.458(10)
C(61)-C(62)	1.393(6)
C(61)-C(66)	1.403(6)
C(62)-C(63)	1.400(6)
C(62)-C(67)	1.524(6)
C(67)-C(68)	1.531(7)
C(67)-C(69)	1.534(7)
C(67)-H(67)	1.0000
C(68)-H(68A)	0.9800
C(68)-H(68B)	0.9800
C(68)-H(68C)	0.9800
C(69)-H(69A)	0.9800
C(69)-H(69B)	0.9800
C(69)-H(69C)	0.9800
C(63)-C(64)	1.377(6)
C(63)-H(63)	0.9500
C(64)-C(65)	1.369(5)
C(64)-H(64)	0.9500
C(65)-C(66)	1.393(6)
C(65)-H(65)	0.9500
C(66)-C(70)	1.522(5)
C(70)-C(71)	1.524(7)
C(70)-C(72)	1.527(7)
C(70)-H(70)	1.0000
C(71)-H(71A)	0.9800
C(71)-H(71B)	0.9800
C(71)-H(71C)	0.9800
C(72)-H(72A)	0.9800
C(72)-H(72B)	0.9800
C(72)-H(72C)	0.9800
C(61A)-C(62A)	1.394(10)
C(61A)-C(66A)	1.414(10)
C(62A)-C(63A)	1.399(10)
C(62A)-C(67A)	1.504(10)

C(67A)-C(68A)	1.524(11)
C(67A)-C(69A)	1.538(12)
C(67A)-H(67A)	1.0000
C(68A)-H(68D)	0.9800
C(68A)-H(68E)	0.9800
C(68A)-H(68F)	0.9800
C(69A)-H(69D)	0.9800
C(69A)-H(69E)	0.9800
C(69A)-H(69F)	0.9800
C(63A)-C(64A)	1.361(9)
C(63A)-H(63A)	0.9500
C(64A)-C(65A)	1.363(10)
C(64A)-H(64A)	0.9500
C(65A)-C(66A)	1.403(10)
C(65A)-H(65A)	0.9500
C(66A)-C(70A)	1.513(10)
C(70A)-C(71A)	1.513(10)
C(70A)-C(72A)	1.530(11)
C(70A)-H(70A)	1.0000
C(71A)-H(71D)	0.9800
C(71A)-H(71E)	0.9800
C(71A)-H(71F)	0.9800
C(72A)-H(72D)	0.9800
C(72A)-H(72E)	0.9800
C(72A)-H(72F)	0.9800
C(16)-C(17)	1.336(2)
C(16)-H(16)	0.9500
C(17)-N(4)	1.393(2)
C(17)-H(17)	0.9500
N(4)-C(81)	1.439(2)
C(81)-C(82)	1.397(2)
C(81)-C(86)	1.404(2)
C(82)-C(83)	1.394(3)
C(82)-C(87)	1.509(3)
C(87)-C(88)	1.526(3)
C(87)-C(89)	1.528(3)
C(87)-H(87)	1.0000
C(88)-H(88A)	0.9800
C(88)-H(88B)	0.9800
C(88)-H(88C)	0.9800
C(89)-H(89A)	0.9800
C(89)-H(89B)	0.9800
C(89)-H(89C)	0.9800
C(83)-C(84)	1.382(3)
C(83)-H(83)	0.9500
C(84)-C(85)	1.380(3)
C(84)-H(84)	0.9500
C(85)-C(86)	1.385(3)
C(85)-H(85)	0.9500
C(86)-C(90)	1.511(3)
C(90)-C(91)	1.529(3)
C(90)-C(92)	1.529(3)
C(90)-H(90)	1.0000
C(91)-H(91A)	0.9800
C(91)-H(91B)	0.9800
C(91)-H(91C)	0.9800

C(92)-H(92A)	0.9800
C(92)-H(92B)	0.9800
C(92)-H(92C)	0.9800

Table S14: Bond angles (°) for **1a**

Ni(2)-Cl(1)-Ni(1)	65.979(14)
C(5)-C(1)-C(2)	109.14(17)
C(5)-C(1)-Ni(2)	61.09(10)
C(2)-C(1)-Ni(2)	113.66(12)
C(5)-C(1)-Ni(1)	113.70(12)
C(2)-C(1)-Ni(1)	60.52(9)
Ni(2)-C(1)-Ni(1)	66.16(5)
C(5)-C(1)-H(1)	122.7(12)
C(2)-C(1)-H(1)	123.8(12)
Ni(2)-C(1)-H(1)	109.5(12)
Ni(1)-C(1)-H(1)	110.5(12)
C(1)-C(2)-C(3)	105.31(16)
C(1)-C(2)-Ni(1)	79.40(11)
C(3)-C(2)-Ni(1)	107.28(13)
C(1)-C(2)-H(2)	126.6(13)
C(3)-C(2)-H(2)	119.0(12)
Ni(1)-C(2)-H(2)	111.3(12)
C(4)-C(3)-C(2)	110.14(18)
C(4)-C(3)-H(3)	124.9
C(2)-C(3)-H(3)	124.9
C(3)-C(4)-C(5)	109.70(18)
C(3)-C(4)-H(4)	125.1
C(5)-C(4)-H(4)	125.1
C(1)-C(5)-C(4)	105.67(16)
C(1)-C(5)-Ni(2)	78.76(11)
C(4)-C(5)-Ni(2)	107.37(13)
C(1)-C(5)-H(5)	124.5(13)
C(4)-C(5)-H(5)	121.8(13)
Ni(2)-C(5)-H(5)	109.5(12)
C(11)-Ni(1)-C(2)	102.38(7)
C(11)-Ni(1)-Cl(1)	111.63(5)
C(2)-Ni(1)-Cl(1)	145.98(5)
C(11)-Ni(1)-C(1)	134.64(7)
C(2)-Ni(1)-C(1)	40.08(7)
Cl(1)-Ni(1)-C(1)	109.22(5)
C(11)-Ni(1)-Ni(2)	168.38(5)
C(2)-Ni(1)-Ni(2)	89.04(5)
Cl(1)-Ni(1)-Ni(2)	56.948(13)
C(1)-Ni(1)-Ni(2)	56.55(5)
N(2)-C(11)-N(1)	102.58(12)
N(2)-C(11)-Ni(1)	131.21(11)
N(1)-C(11)-Ni(1)	126.11(11)
C(11)-N(1)-C(12)	111.92(13)
C(11)-N(1)-C(21)	124.04(13)
C(12)-N(1)-C(21)	124.03(13)
C(26)-C(21)-C(22)	122.71(15)
C(26)-C(21)-N(1)	118.40(14)
C(22)-C(21)-N(1)	118.89(14)
C(23)-C(22)-C(21)	117.11(16)
C(23)-C(22)-C(27)	120.83(15)

C(21)-C(22)-C(27)	122.02(15)
C(24)-C(23)-C(22)	121.44(17)
C(24)-C(23)-H(23)	119.3
C(22)-C(23)-H(23)	119.3
C(22)-C(27)-C(28)	113.96(16)
C(22)-C(27)-C(29)	110.45(16)
C(28)-C(27)-C(29)	111.17(19)
C(22)-C(27)-H(27)	107.0
C(28)-C(27)-H(27)	107.0
C(29)-C(27)-H(27)	107.0
C(27)-C(28)-H(28A)	109.5
C(27)-C(28)-H(28B)	109.5
H(28A)-C(28)-H(28B)	109.5
C(27)-C(28)-H(28C)	109.5
H(28A)-C(28)-H(28C)	109.5
H(28B)-C(28)-H(28C)	109.5
C(27)-C(29)-H(29A)	109.5
C(27)-C(29)-H(29B)	109.5
H(29A)-C(29)-H(29B)	109.5
C(27)-C(29)-H(29C)	109.5
H(29A)-C(29)-H(29C)	109.5
H(29B)-C(29)-H(29C)	109.5
C(25)-C(24)-C(23)	120.18(17)
C(25)-C(24)-H(24)	119.9
C(23)-C(24)-H(24)	119.9
C(24)-C(25)-C(26)	121.06(17)
C(24)-C(25)-H(25)	119.5
C(26)-C(25)-H(25)	119.5
C(25)-C(26)-C(21)	117.49(16)
C(25)-C(26)-C(30)	119.85(16)
C(21)-C(26)-C(30)	122.66(15)
C(26)-C(30)-C(31)	112.31(17)
C(26)-C(30)-C(32)	110.50(16)
C(31)-C(30)-C(32)	110.16(18)
C(26)-C(30)-H(30)	107.9
C(31)-C(30)-H(30)	107.9
C(32)-C(30)-H(30)	107.9
C(30)-C(31)-H(31A)	109.5
C(30)-C(31)-H(31B)	109.5
H(31A)-C(31)-H(31B)	109.5
C(30)-C(31)-H(31C)	109.5
H(31A)-C(31)-H(31C)	109.5
H(31B)-C(31)-H(31C)	109.5
C(30)-C(32)-H(32A)	109.5
C(30)-C(32)-H(32B)	109.5
H(32A)-C(32)-H(32B)	109.5
C(30)-C(32)-H(32C)	109.5
H(32A)-C(32)-H(32C)	109.5
H(32B)-C(32)-H(32C)	109.5
C(13)-C(12)-N(1)	106.73(13)
C(13)-C(12)-H(12)	126.6
N(1)-C(12)-H(12)	126.6
C(12)-C(13)-N(2)	106.62(14)
C(12)-C(13)-H(13)	126.7
N(2)-C(13)-H(13)	126.7
C(11)-N(2)-C(13)	112.15(13)

C(11)-N(2)-C(41)	125.45(12)
C(13)-N(2)-C(41)	122.21(13)
C(46)-C(41)-C(42)	123.31(15)
C(46)-C(41)-N(2)	118.80(14)
C(42)-C(41)-N(2)	117.85(14)
C(43)-C(42)-C(41)	116.69(15)
C(43)-C(42)-C(47)	121.28(15)
C(41)-C(42)-C(47)	122.04(15)
C(42)-C(47)-C(48)	112.36(16)
C(42)-C(47)-C(49)	111.46(15)
C(48)-C(47)-C(49)	109.32(15)
C(42)-C(47)-H(47)	107.8
C(48)-C(47)-H(47)	107.8
C(49)-C(47)-H(47)	107.8
C(47)-C(48)-H(48A)	109.5
C(47)-C(48)-H(48B)	109.5
H(48A)-C(48)-H(48B)	109.5
C(47)-C(48)-H(48C)	109.5
H(48A)-C(48)-H(48C)	109.5
H(48B)-C(48)-H(48C)	109.5
C(47)-C(49)-H(49A)	109.5
C(47)-C(49)-H(49B)	109.5
H(49A)-C(49)-H(49B)	109.5
C(47)-C(49)-H(49C)	109.5
H(49A)-C(49)-H(49C)	109.5
H(49B)-C(49)-H(49C)	109.5
C(44)-C(43)-C(42)	121.16(16)
C(44)-C(43)-H(43)	119.4
C(42)-C(43)-H(43)	119.4
C(43)-C(44)-C(45)	120.62(16)
C(43)-C(44)-H(44)	119.7
C(45)-C(44)-H(44)	119.7
C(44)-C(45)-C(46)	120.84(16)
C(44)-C(45)-H(45)	119.6
C(46)-C(45)-H(45)	119.6
C(45)-C(46)-C(41)	117.18(15)
C(45)-C(46)-C(50)	121.90(15)
C(41)-C(46)-C(50)	120.90(14)
C(46)-C(50)-C(52)	110.74(14)
C(46)-C(50)-C(51)	113.18(15)
C(52)-C(50)-C(51)	109.88(15)
C(46)-C(50)-H(50)	107.6
C(52)-C(50)-H(50)	107.6
C(51)-C(50)-H(50)	107.6
C(50)-C(51)-H(51A)	109.5
C(50)-C(51)-H(51B)	109.5
H(51A)-C(51)-H(51B)	109.5
C(50)-C(51)-H(51C)	109.5
H(51A)-C(51)-H(51C)	109.5
H(51B)-C(51)-H(51C)	109.5
C(50)-C(52)-H(52A)	109.5
C(50)-C(52)-H(52B)	109.5
H(52A)-C(52)-H(52B)	109.5
C(50)-C(52)-H(52C)	109.5
H(52A)-C(52)-H(52C)	109.5
H(52B)-C(52)-H(52C)	109.5

C(15)-Ni(2)-C(5)	104.90(7)
C(15)-Ni(2)-C(1)	138.86(7)
C(5)-Ni(2)-C(1)	40.15(7)
C(15)-Ni(2)-Cl(1)	108.26(5)
C(5)-Ni(2)-Cl(1)	146.61(5)
C(1)-Ni(2)-Cl(1)	110.03(5)
C(15)-Ni(2)-Ni(1)	163.84(5)
C(5)-Ni(2)-Ni(1)	89.53(5)
C(1)-Ni(2)-Ni(1)	57.29(5)
Cl(1)-Ni(2)-Ni(1)	57.072(15)
N(4)-C(15)-N(3)	102.92(13)
N(4)-C(15)-Ni(2)	130.97(12)
N(3)-C(15)-Ni(2)	125.99(12)
C(15)-N(3)-C(16)	111.67(14)
C(15)-N(3)-C(61)	123.2(7)
C(16)-N(3)-C(61)	124.9(7)
C(15)-N(3)-C(61A)	127.7(13)
C(16)-N(3)-C(61A)	120.6(13)
C(61)-N(3)-C(61A)	7.7(10)
C(62)-C(61)-C(66)	123.4(5)
C(62)-C(61)-N(3)	117.9(5)
C(66)-C(61)-N(3)	118.5(5)
C(61)-C(62)-C(63)	117.0(4)
C(61)-C(62)-C(67)	122.1(6)
C(63)-C(62)-C(67)	120.8(5)
C(62)-C(67)-C(68)	110.9(6)
C(62)-C(67)-C(69)	113.1(6)
C(68)-C(67)-C(69)	109.4(8)
C(62)-C(67)-H(67)	107.7
C(68)-C(67)-H(67)	107.7
C(69)-C(67)-H(67)	107.7
C(67)-C(68)-H(68A)	109.5
C(67)-C(68)-H(68B)	109.5
H(68A)-C(68)-H(68B)	109.5
C(67)-C(68)-H(68C)	109.5
H(68A)-C(68)-H(68C)	109.5
H(68B)-C(68)-H(68C)	109.5
C(67)-C(69)-H(69A)	109.5
C(67)-C(69)-H(69B)	109.5
H(69A)-C(69)-H(69B)	109.5
C(67)-C(69)-H(69C)	109.5
H(69A)-C(69)-H(69C)	109.5
H(69B)-C(69)-H(69C)	109.5
C(64)-C(63)-C(62)	120.7(5)
C(64)-C(63)-H(63)	119.7
C(62)-C(63)-H(63)	119.7
C(65)-C(64)-C(63)	120.8(5)
C(65)-C(64)-H(64)	119.6
C(63)-C(64)-H(64)	119.6
C(64)-C(65)-C(66)	121.5(5)
C(64)-C(65)-H(65)	119.2
C(66)-C(65)-H(65)	119.2
C(65)-C(66)-C(61)	116.5(5)
C(65)-C(66)-C(70)	120.7(5)
C(61)-C(66)-C(70)	122.8(5)
C(66)-C(70)-C(71)	112.0(6)

C(66)-C(70)-C(72)	111.7(6)
C(71)-C(70)-C(72)	108.8(7)
C(66)-C(70)-H(70)	108.1
C(71)-C(70)-H(70)	108.1
C(72)-C(70)-H(70)	108.1
C(70)-C(71)-H(71A)	109.5
C(70)-C(71)-H(71B)	109.5
H(71A)-C(71)-H(71B)	109.5
C(70)-C(71)-H(71C)	109.5
H(71A)-C(71)-H(71C)	109.5
H(71B)-C(71)-H(71C)	109.5
C(70)-C(72)-H(72A)	109.5
C(70)-C(72)-H(72B)	109.5
H(72A)-C(72)-H(72B)	109.5
C(70)-C(72)-H(72C)	109.5
H(72A)-C(72)-H(72C)	109.5
H(72B)-C(72)-H(72C)	109.5
C(62A)-C(61A)-C(66A)	123.4(9)
C(62A)-C(61A)-N(3)	118.9(9)
C(66A)-C(61A)-N(3)	117.4(9)
C(61A)-C(62A)-C(63A)	115.6(9)
C(61A)-C(62A)-C(67A)	123.0(10)
C(63A)-C(62A)-C(67A)	121.2(9)
C(62A)-C(67A)-C(68A)	113.2(12)
C(62A)-C(67A)-C(69A)	109.7(11)
C(68A)-C(67A)-C(69A)	110.6(13)
C(62A)-C(67A)-H(67A)	107.7
C(68A)-C(67A)-H(67A)	107.7
C(69A)-C(67A)-H(67A)	107.7
C(67A)-C(68A)-H(68D)	109.5
C(67A)-C(68A)-H(68E)	109.5
H(68D)-C(68A)-H(68E)	109.5
C(67A)-C(68A)-H(68F)	109.5
H(68D)-C(68A)-H(68F)	109.5
H(68E)-C(68A)-H(68F)	109.5
C(67A)-C(69A)-H(69D)	109.5
C(67A)-C(69A)-H(69E)	109.5
H(69D)-C(69A)-H(69E)	109.5
C(67A)-C(69A)-H(69F)	109.5
H(69D)-C(69A)-H(69F)	109.5
H(69E)-C(69A)-H(69F)	109.5
C(64A)-C(63A)-C(62A)	122.2(10)
C(64A)-C(63A)-H(63A)	118.9
C(62A)-C(63A)-H(63A)	118.9
C(63A)-C(64A)-C(65A)	120.9(9)
C(63A)-C(64A)-H(64A)	119.5
C(65A)-C(64A)-H(64A)	119.5
C(64A)-C(65A)-C(66A)	121.0(9)
C(64A)-C(65A)-H(65A)	119.5
C(66A)-C(65A)-H(65A)	119.5
C(65A)-C(66A)-C(61A)	116.2(9)
C(65A)-C(66A)-C(70A)	122.1(9)
C(61A)-C(66A)-C(70A)	121.6(10)
C(71A)-C(70A)-C(66A)	112.4(11)
C(71A)-C(70A)-C(72A)	111.7(11)
C(66A)-C(70A)-C(72A)	113.4(10)

C(71A)-C(70A)-H(70A)	106.2
C(66A)-C(70A)-H(70A)	106.2
C(72A)-C(70A)-H(70A)	106.2
C(70A)-C(71A)-H(71D)	109.5
C(70A)-C(71A)-H(71E)	109.5
H(71D)-C(71A)-H(71E)	109.5
C(70A)-C(71A)-H(71F)	109.5
H(71D)-C(71A)-H(71F)	109.5
H(71E)-C(71A)-H(71F)	109.5
C(70A)-C(72A)-H(72D)	109.5
C(70A)-C(72A)-H(72E)	109.5
H(72D)-C(72A)-H(72E)	109.5
C(70A)-C(72A)-H(72F)	109.5
H(72D)-C(72A)-H(72F)	109.5
H(72E)-C(72A)-H(72F)	109.5
C(17)-C(16)-N(3)	106.70(15)
C(17)-C(16)-H(16)	126.7
N(3)-C(16)-H(16)	126.7
C(16)-C(17)-N(4)	106.83(14)
C(16)-C(17)-H(17)	126.6
N(4)-C(17)-H(17)	126.6
C(15)-N(4)-C(17)	111.88(14)
C(15)-N(4)-C(81)	124.37(13)
C(17)-N(4)-C(81)	123.42(13)
C(82)-C(81)-C(86)	123.09(16)
C(82)-C(81)-N(4)	119.30(15)
C(86)-C(81)-N(4)	117.60(15)
C(83)-C(82)-C(81)	116.92(17)
C(83)-C(82)-C(87)	120.28(17)
C(81)-C(82)-C(87)	122.80(16)
C(82)-C(87)-C(88)	110.52(16)
C(82)-C(87)-C(89)	111.45(18)
C(88)-C(87)-C(89)	110.65(17)
C(82)-C(87)-H(87)	108.0
C(88)-C(87)-H(87)	108.0
C(89)-C(87)-H(87)	108.0
C(87)-C(88)-H(88A)	109.5
C(87)-C(88)-H(88B)	109.5
H(88A)-C(88)-H(88B)	109.5
C(87)-C(88)-H(88C)	109.5
H(88A)-C(88)-H(88C)	109.5
H(88B)-C(88)-H(88C)	109.5
C(87)-C(89)-H(89A)	109.5
C(87)-C(89)-H(89B)	109.5
H(89A)-C(89)-H(89B)	109.5
C(87)-C(89)-H(89C)	109.5
H(89A)-C(89)-H(89C)	109.5
H(89B)-C(89)-H(89C)	109.5
C(84)-C(83)-C(82)	121.3(2)
C(84)-C(83)-H(83)	119.3
C(82)-C(83)-H(83)	119.3
C(85)-C(84)-C(83)	120.08(19)
C(85)-C(84)-H(84)	120.0
C(83)-C(84)-H(84)	120.0
C(84)-C(85)-C(86)	121.47(19)
C(84)-C(85)-H(85)	119.3

C(86)-C(85)-H(85)	119.3
C(85)-C(86)-C(81)	117.10(18)
C(85)-C(86)-C(90)	120.61(17)
C(81)-C(86)-C(90)	122.28(16)
C(86)-C(90)-C(91)	112.60(17)
C(86)-C(90)-C(92)	110.55(17)
C(91)-C(90)-C(92)	110.18(17)
C(86)-C(90)-H(90)	107.8
C(91)-C(90)-H(90)	107.8
C(92)-C(90)-H(90)	107.8
C(90)-C(91)-H(91A)	109.5
C(90)-C(91)-H(91B)	109.5
H(91A)-C(91)-H(91B)	109.5
C(90)-C(91)-H(91C)	109.5
H(91A)-C(91)-H(91C)	109.5
H(91B)-C(91)-H(91C)	109.5
C(90)-C(92)-H(92A)	109.5
C(90)-C(92)-H(92B)	109.5
H(92A)-C(92)-H(92B)	109.5
C(90)-C(92)-H(92C)	109.5
H(92A)-C(92)-H(92C)	109.5
H(92B)-C(92)-H(92C)	109.5

X-Ray Data for 2b (CCDC 939470)

Compound **2b** crystallizes in the monoclinic space group *P2(1)/n* with one molecule in the asymmetric unit. The coordinates for the hydrogen atoms bound to C1, C2 and C9 were taken from the difference Fourier synthesis and the hydrogen atoms were subsequently refined semi-freely with the help of a distance restraint.

Table S15: Crystal data and structure refinement for **2b**

Empirical formula	C63 H83 Cl N4 Ni2	
Formula weight	1049.20	
Temperature	93(2) K	
Wavelength	1.54187 Å	
Crystal system	Monoclinic	
Space group	P2(1)/n	
Unit cell dimensions	a = 12.1415(2) Å	α= 90°.
	b = 24.2931(4) Å	β= 102.953(7)°.
	c = 19.8300(14) Å	γ = 90°.
Volume	5700.1(4) $Å^3$	
Z	4	
Density (calculated)	1.223 Mg/m^3	
Absorption coefficient	1.556 mm^{-1}	
F(000)	2248	
Crystal size	0.15 x 0.10 x 0.05 mm^3	
Theta range for data collection	2.92 to 65.09°.	
Index ranges	-14<=h<=14, -28<=k<=28, -23<=l<=23	
Reflections collected	104065	
Independent reflections	9726 [R(int) = 0.1397]	
Completeness to theta = 65.09°	100.0 %	
Absorption correction	Semi-empirical from equivalents	

Max. and min. transmission	0.9263 and 0.8001
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	9726 / 3 / 656
Goodness-of-fit on F^2	1.090
Final R indices [I>2sigma(I)]	R1 = 0.0482, wR2 = 0.1182
R indices (all data)	R1 = 0.0623, wR2 = 0.1339
Largest diff. peak and hole	0.500 and -0.514 e.Å^{-3}

Table S16: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^{2}x 10^3) for **2b**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Cl(1)	-96(1)	1000(1)	3197(1)	43(1)
C(1)	385(2)	1155(1)	1506(1)	35(1)
C(2)	-108(2)	632(1)	1253(1)	33(1)
C(3)	-1263(2)	762(1)	876(1)	33(1)
C(4)	-2105(2)	427(1)	501(1)	43(1)
C(5)	-3101(3)	660(1)	135(2)	51(1)
C(6)	-3268(3)	1224(1)	142(2)	53(1)
C(7)	-2443(2)	1566(1)	519(1)	44(1)
C(8)	-1440(2)	1339(1)	892(1)	34(1)
C(9)	-416(2)	1600(1)	1299(1)	35(1)
Ni(1)	30(1)	560(1)	2248(1)	31(1)
C(11)	-682(2)	2266(1)	2520(1)	28(1)
N(1)	-87(2)	2716(1)	2401(1)	29(1)
C(21)	892(2)	2701(1)	2110(1)	31(1)
C(22)	838(2)	2946(1)	1464(1)	37(1)
C(27)	-203(2)	3246(1)	1071(1)	45(1)
C(28)	27(3)	3860(1)	1032(2)	58(1)
C(29)	-634(4)	3014(2)	339(2)	77(1)
C(23)	1797(3)	2917(1)	1188(2)	53(1)
C(24)	2765(3)	2664(1)	1534(2)	62(1)
C(25)	2817(3)	2442(1)	2180(2)	56(1)
C(26)	1892(2)	2453(1)	2487(2)	39(1)
C(30)	2007(2)	2230(1)	3210(2)	47(1)
C(31)	2521(4)	2666(2)	3740(2)	95(2)
C(32)	2673(3)	1697(2)	3345(2)	66(1)
C(12)	-343(2)	3215(1)	2756(2)	40(1)
C(13)	-1403(2)	3053(1)	2981(2)	41(1)
N(2)	-1444(2)	2453(1)	2868(1)	31(1)
C(41)	-2190(2)	2130(1)	3183(1)	31(1)
C(42)	-3225(2)	1943(1)	2778(1)	35(1)
C(47)	-3508(2)	1987(1)	1993(1)	43(1)
C(48)	-3806(4)	2570(2)	1750(2)	75(1)
C(49)	-4447(3)	1602(1)	1637(2)	59(1)
C(43)	-3978(2)	1701(1)	3125(2)	45(1)
C(44)	-3724(3)	1649(1)	3834(2)	51(1)
C(45)	-2693(3)	1814(1)	4215(2)	46(1)
C(46)	-1895(2)	2054(1)	3898(1)	37(1)
C(50)	-750(3)	2218(1)	4336(2)	48(1)
C(51)	-844(3)	2695(1)	4830(2)	64(1)
C(52)	-174(3)	1733(2)	4772(2)	69(1)
Ni(2)	-371(1)	1543(1)	2289(1)	30(1)
C(16)	284(2)	-187(1)	2470(1)	30(1)
N(3)	1213(2)	-370(1)	2934(1)	37(1)

C(61)	2251(2)	-72(1)	3165(2)	38(1)
C(62)	2949(2)	29(1)	2701(2)	45(1)
C(67)	2636(3)	-142(2)	1948(2)	56(1)
C(68)	3196(3)	-691(2)	1843(2)	72(1)
C(69)	2940(3)	288(2)	1455(2)	76(1)
C(63)	3995(3)	271(1)	2961(2)	60(1)
C(64)	4345(3)	395(1)	3645(2)	68(1)
C(65)	3656(3)	297(1)	4098(2)	62(1)
C(66)	2581(2)	64(1)	3868(2)	46(1)
C(70)	1846(3)	-38(1)	4375(2)	57(1)
C(71)	1674(4)	485(2)	4772(2)	78(1)
C(72)	2337(4)	-493(2)	4894(2)	80(1)
C(17)	1230(2)	-971(1)	3042(2)	48(1)
C(18)	10(2)	-1118(1)	2734(2)	44(1)
N(4)	-383(2)	-638(1)	2295(1)	34(1)
C(81)	-1469(2)	-655(1)	1829(1)	33(1)
C(82)	-1564(2)	-947(1)	1212(1)	39(1)
C(87)	-570(3)	-1219(1)	1001(2)	48(1)
C(88)	-787(4)	-1829(2)	856(3)	88(1)
C(89)	-270(4)	-934(2)	391(2)	95(2)
C(83)	-2639(3)	-975(1)	770(2)	49(1)
C(84)	-3560(3)	-732(1)	942(2)	54(1)
C(85)	-3444(2)	-460(1)	1557(2)	52(1)
C(86)	-2404(2)	-417(1)	2022(2)	41(1)
C(90)	-2325(3)	-132(1)	2706(2)	51(1)
C(91)	-2828(3)	445(1)	2621(2)	75(1)
C(92)	-2894(4)	-477(2)	3177(2)	73(1)

Table S17: Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **2b**. 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}
Cl(1)	63(1)	34(1)	33(1)	1(1)	8(1)	3(1)
C(1)	34(1)	35(2)	38(1)	0(1)	11(1)	0(1)
C(2)	35(1)	26(1)	39(1)	-4(1)	11(1)	2(1)
C(3)	37(1)	30(1)	32(1)	-2(1)	8(1)	3(1)
C(4)	49(2)	35(2)	42(2)	-2(1)	6(1)	-4(1)
C(5)	43(2)	57(2)	45(2)	-5(2)	-3(1)	-3(1)
C(6)	46(2)	58(2)	48(2)	-4(2)	-3(1)	15(2)
C(7)	49(2)	40(2)	40(2)	-2(1)	5(1)	12(1)
C(8)	40(1)	33(2)	30(1)	0(1)	10(1)	4(1)
C(9)	42(2)	26(1)	37(1)	1(1)	9(1)	-1(1)
Ni(1)	32(1)	25(1)	34(1)	0(1)	4(1)	1(1)
C(11)	23(1)	34(1)	25(1)	2(1)	1(1)	0(1)
N(1)	28(1)	25(1)	35(1)	-1(1)	9(1)	1(1)
C(21)	31(1)	21(1)	40(1)	-4(1)	8(1)	-6(1)
C(22)	40(2)	35(2)	36(1)	-2(1)	10(1)	-7(1)
C(27)	47(2)	50(2)	37(2)	7(1)	6(1)	-8(1)
C(28)	62(2)	49(2)	63(2)	10(2)	14(2)	4(2)
C(29)	104(3)	68(3)	47(2)	9(2)	-8(2)	-15(2)
C(23)	63(2)	50(2)	55(2)	2(2)	33(2)	-8(2)
C(24)	52(2)	58(2)	87(3)	4(2)	40(2)	6(2)
C(25)	38(2)	54(2)	78(2)	8(2)	20(2)	8(1)
C(26)	32(1)	35(2)	49(2)	-1(1)	7(1)	-1(1)
C(30)	36(2)	52(2)	47(2)	6(1)	-2(1)	6(1)

C(31)	133(4)	68(3)	59(2)	-7(2)	-29(2)	9(3)
C(32)	58(2)	59(2)	81(3)	19(2)	13(2)	16(2)
C(12)	45(2)	27(1)	52(2)	-6(1)	19(1)	-3(1)
C(13)	49(2)	23(1)	57(2)	-5(1)	22(1)	1(1)
N(2)	35(1)	25(1)	36(1)	0(1)	13(1)	0(1)
C(41)	34(1)	24(1)	39(1)	0(1)	15(1)	-1(1)
C(42)	34(1)	31(1)	41(2)	-2(1)	11(1)	2(1)
C(47)	39(2)	49(2)	40(2)	-3(1)	6(1)	3(1)
C(48)	97(3)	54(2)	59(2)	10(2)	-15(2)	-2(2)
C(49)	55(2)	61(2)	54(2)	-8(2)	-1(2)	-4(2)
C(43)	38(2)	44(2)	56(2)	-8(1)	18(1)	-6(1)
C(44)	58(2)	44(2)	60(2)	-3(2)	34(2)	-10(1)
C(45)	63(2)	43(2)	38(2)	3(1)	21(1)	-2(1)
C(46)	43(2)	34(2)	34(1)	0(1)	12(1)	3(1)
C(50)	49(2)	53(2)	39(2)	2(1)	4(1)	1(1)
C(51)	78(2)	50(2)	54(2)	-9(2)	-5(2)	-1(2)
C(52)	81(3)	66(2)	49(2)	-8(2)	-9(2)	26(2)
Ni(2)	33(1)	25(1)	32(1)	0(1)	6(1)	1(1)
C(16)	25(1)	31(1)	36(1)	-3(1)	6(1)	-1(1)
N(3)	32(1)	27(1)	47(1)	6(1)	-3(1)	-2(1)
C(61)	28(1)	26(1)	53(2)	3(1)	-6(1)	4(1)
C(62)	31(1)	37(2)	67(2)	5(1)	8(1)	4(1)
C(67)	40(2)	68(2)	62(2)	5(2)	14(2)	10(2)
C(68)	80(3)	59(2)	80(3)	-5(2)	27(2)	10(2)
C(69)	72(2)	79(3)	86(3)	24(2)	38(2)	33(2)
C(63)	37(2)	50(2)	88(3)	10(2)	6(2)	0(1)
C(64)	38(2)	45(2)	112(3)	1(2)	-3(2)	-10(2)
C(65)	54(2)	44(2)	72(2)	-9(2)	-20(2)	5(2)
C(66)	42(2)	35(2)	52(2)	1(1)	-7(1)	6(1)
C(70)	60(2)	58(2)	48(2)	1(2)	-2(2)	8(2)
C(71)	93(3)	78(3)	55(2)	0(2)	2(2)	31(2)
C(72)	103(3)	67(3)	66(2)	20(2)	10(2)	15(2)
C(17)	44(2)	26(2)	65(2)	11(1)	-8(1)	2(1)
C(18)	44(2)	28(2)	56(2)	10(1)	0(1)	0(1)
N(4)	29(1)	26(1)	43(1)	5(1)	1(1)	-2(1)
C(81)	29(1)	28(1)	41(1)	4(1)	5(1)	-4(1)
C(82)	40(2)	33(2)	42(2)	2(1)	8(1)	-9(1)
C(87)	51(2)	44(2)	53(2)	-10(1)	19(1)	-6(1)
C(88)	74(3)	49(2)	143(4)	-27(2)	28(3)	0(2)
C(89)	128(4)	86(3)	98(3)	3(3)	80(3)	10(3)
C(83)	52(2)	43(2)	46(2)	4(1)	-1(1)	-14(1)
C(84)	37(2)	49(2)	67(2)	13(2)	-9(2)	-11(1)
C(85)	31(2)	48(2)	74(2)	7(2)	8(1)	2(1)
C(86)	33(1)	36(2)	54(2)	3(1)	11(1)	-1(1)
C(90)	45(2)	48(2)	63(2)	-11(2)	22(2)	-7(1)
C(91)	55(2)	49(2)	127(4)	-19(2)	34(2)	4(2)
C(92)	88(3)	74(3)	67(2)	-12(2)	37(2)	-18(2)

Table S18: Bond lengths (Å) for **2b**

Cl(1)-Ni(2)	2.1967(8)
Cl(1)-Ni(1)	2.1998(8)
C(1)-C(2)	1.444(4)
C(1)-C(9)	1.451(4)
C(1)-Ni(1)	2.174(3)
C(1)-Ni(2)	2.187(3)

C(1)-H(1)	0.990(17)
C(2)-C(3)	1.468(4)
C(2)-Ni(1)	1.950(3)
C(2)-H(2)	0.979(17)
C(3)-C(4)	1.385(4)
C(3)-C(8)	1.421(4)
C(4)-C(5)	1.384(4)
C(4)-H(4)	0.9500
C(5)-C(6)	1.383(4)
C(5)-H(5)	0.9500
C(6)-C(7)	1.384(4)
C(6)-H(6)	0.9500
C(7)-C(8)	1.389(4)
C(7)-H(7)	0.9500
C(8)-C(9)	1.466(4)
C(9)-Ni(2)	1.955(3)
C(9)-H(9)	0.984(17)
Ni(1)-C(16)	1.875(3)
Ni(1)-Ni(2)	2.4425(6)
C(11)-N(2)	1.351(3)
C(11)-N(1)	1.359(3)
C(11)-Ni(2)	1.875(3)
N(1)-C(21)	1.433(3)
N(1)-C(12)	1.469(3)
C(21)-C(22)	1.402(4)
C(21)-C(26)	1.409(4)
C(22)-C(23)	1.394(4)
C(22)-C(27)	1.516(4)
C(27)-C(28)	1.521(4)
C(27)-C(29)	1.536(4)
C(27)-H(27)	1.0000
C(28)-H(28A)	0.9800
C(28)-H(28B)	0.9800
C(28)-H(28C)	0.9800
C(29)-H(29A)	0.9800
C(29)-H(29B)	0.9800
C(29)-H(29C)	0.9800
C(23)-C(24)	1.366(5)
C(23)-H(23)	0.9500
C(24)-C(25)	1.379(5)
C(24)-H(24)	0.9500
C(25)-C(26)	1.392(4)
C(25)-H(25)	0.9500
C(26)-C(30)	1.510(4)
C(30)-C(32)	1.520(4)
C(30)-C(31)	1.523(5)
C(30)-H(30)	1.0000
C(31)-H(31A)	0.9800
C(31)-H(31B)	0.9800
C(31)-H(31C)	0.9800
C(32)-H(32A)	0.9800
C(32)-H(32B)	0.9800
C(32)-H(32C)	0.9800
C(12)-C(13)	1.506(4)
C(12)-H(12A)	0.9900
C(12)-H(12B)	0.9900

C(13)-N(2)	1.474(3)
C(13)-H(13A)	0.9900
C(13)-H(13B)	0.9900
N(2)-C(41)	1.443(3)
C(41)-C(46)	1.395(4)
C(41)-C(42)	1.406(4)
C(42)-C(43)	1.392(4)
C(42)-C(47)	1.522(4)
C(47)-C(48)	1.512(4)
C(47)-C(49)	1.520(4)
C(47)-H(47)	1.0000
C(48)-H(48A)	0.9800
C(48)-H(48B)	0.9800
C(48)-H(48C)	0.9800
C(49)-H(49A)	0.9800
C(49)-H(49B)	0.9800
C(49)-H(49C)	0.9800
C(43)-C(44)	1.376(4)
C(43)-H(43)	0.9500
C(44)-C(45)	1.369(4)
C(44)-H(44)	0.9500
C(45)-C(46)	1.395(4)
C(45)-H(45)	0.9500
C(46)-C(50)	1.518(4)
C(50)-C(52)	1.534(4)
C(50)-C(51)	1.538(4)
C(50)-H(50)	1.0000
C(51)-H(51A)	0.9800
C(51)-H(51B)	0.9800
C(51)-H(51C)	0.9800
C(52)-H(52A)	0.9800
C(52)-H(52B)	0.9800
C(52)-H(52C)	0.9800
C(16)-N(4)	1.360(3)
C(16)-N(3)	1.361(3)
N(3)-C(61)	1.436(3)
N(3)-C(17)	1.476(3)
C(61)-C(66)	1.402(4)
C(61)-C(62)	1.404(4)
C(62)-C(63)	1.389(4)
C(62)-C(67)	1.514(5)
C(67)-C(69)	1.530(5)
C(67)-C(68)	1.533(5)
C(67)-H(67)	1.0000
C(68)-H(68A)	0.9800
C(68)-H(68B)	0.9800
C(68)-H(68C)	0.9800
C(69)-H(69A)	0.9800
C(69)-H(69B)	0.9800
C(69)-H(69C)	0.9800
C(63)-C(64)	1.362(5)
C(63)-H(63)	0.9500
C(64)-C(65)	1.379(5)
C(64)-H(64)	0.9500
C(65)-C(66)	1.402(4)
C(65)-H(65)	0.9500

C(66)-C(70)	1.507(5)
C(70)-C(71)	1.533(5)
C(70)-C(72)	1.538(5)
C(70)-H(70)	1.0000
C(71)-H(71A)	0.9800
C(71)-H(71B)	0.9800
C(71)-H(71C)	0.9800
C(72)-H(72A)	0.9800
C(72)-H(72B)	0.9800
C(72)-H(72C)	0.9800
C(17)-C(18)	1.514(4)
C(17)-H(17A)	0.9900
C(17)-H(17B)	0.9900
C(18)-N(4)	1.470(3)
C(18)-H(18A)	0.9900
C(18)-H(18B)	0.9900
N(4)-C(81)	1.431(3)
C(81)-C(82)	1.397(4)
C(81)-C(86)	1.402(4)
C(82)-C(83)	1.401(4)
C(82)-C(87)	1.515(4)
C(87)-C(89)	1.509(5)
C(87)-C(88)	1.520(5)
C(87)-H(87)	1.0000
C(88)-H(88A)	0.9800
C(88)-H(88B)	0.9800
C(88)-H(88C)	0.9800
C(89)-H(89A)	0.9800
C(89)-H(89B)	0.9800
C(89)-H(89C)	0.9800
C(83)-C(84)	1.373(5)
C(83)-H(83)	0.9500
C(84)-C(85)	1.366(5)
C(84)-H(84)	0.9500
C(85)-C(86)	1.390(4)
C(85)-H(85)	0.9500
C(86)-C(90)	1.506(4)
C(90)-C(91)	1.525(5)
C(90)-C(92)	1.529(5)
C(90)-H(90)	1.0000
C(91)-H(91A)	0.9800
C(91)-H(91B)	0.9800
C(91)-H(91C)	0.9800
C(92)-H(92A)	0.9800
C(92)-H(92B)	0.9800
C(92)-H(92C)	0.9800

Table S19: Bond angles (°) for **2b**

Ni(2)-Cl(1)-Ni(1)	67.50(2)
C(2)-C(1)-C(9)	110.9(2)
C(2)-C(1)-Ni(1)	61.34(14)
C(9)-C(1)-Ni(1)	117.65(18)
C(2)-C(1)-Ni(2)	114.64(18)
C(9)-C(1)-Ni(2)	61.12(14)
Ni(1)-C(1)-Ni(2)	68.12(8)

C(2)-C(1)-H(1)	124.2(17)
C(9)-C(1)-H(1)	120.4(17)
Ni(1)-C(1)-H(1)	107.9(16)
Ni(2)-C(1)-H(1)	108.4(16)
C(1)-C(2)-C(3)	105.3(2)
C(1)-C(2)-Ni(1)	78.12(16)
C(3)-C(2)-Ni(1)	113.02(17)
C(1)-C(2)-H(2)	124.8(16)
C(3)-C(2)-H(2)	118.6(16)
Ni(1)-C(2)-H(2)	110.3(16)
C(4)-C(3)-C(8)	119.6(2)
C(4)-C(3)-C(2)	130.8(2)
C(8)-C(3)-C(2)	109.4(2)
C(5)-C(4)-C(3)	119.6(3)
C(5)-C(4)-H(4)	120.2
C(3)-C(4)-H(4)	120.2
C(6)-C(5)-C(4)	120.8(3)
C(6)-C(5)-H(5)	119.6
C(4)-C(5)-H(5)	119.6
C(5)-C(6)-C(7)	120.6(3)
C(5)-C(6)-H(6)	119.7
C(7)-C(6)-H(6)	119.7
C(6)-C(7)-C(8)	119.5(3)
C(6)-C(7)-H(7)	120.3
C(8)-C(7)-H(7)	120.3
C(7)-C(8)-C(3)	119.9(2)
C(7)-C(8)-C(9)	131.1(3)
C(3)-C(8)-C(9)	109.0(2)
C(1)-C(9)-C(8)	105.4(2)
C(1)-C(9)-Ni(2)	78.36(15)
C(8)-C(9)-Ni(2)	110.59(18)
C(1)-C(9)-H(9)	123.4(16)
C(8)-C(9)-H(9)	120.8(16)
Ni(2)-C(9)-H(9)	110.2(16)
C(16)-Ni(1)-C(2)	107.28(11)
C(16)-Ni(1)-C(1)	138.99(10)
C(2)-Ni(1)-C(1)	40.54(10)
C(16)-Ni(1)-Cl(1)	108.02(8)
C(2)-Ni(1)-Cl(1)	144.55(8)
C(1)-Ni(1)-Cl(1)	108.56(7)
C(16)-Ni(1)-Ni(2)	164.05(8)
C(2)-Ni(1)-Ni(2)	88.40(8)
C(1)-Ni(1)-Ni(2)	56.18(7)
Cl(1)-Ni(1)-Ni(2)	56.19(2)
N(2)-C(11)-N(1)	105.6(2)
N(2)-C(11)-Ni(2)	129.66(18)
N(1)-C(11)-Ni(2)	124.57(17)
C(11)-N(1)-C(21)	124.7(2)
C(11)-N(1)-C(12)	113.73(19)
C(21)-N(1)-C(12)	119.86(19)
C(22)-C(21)-C(26)	121.6(2)
C(22)-C(21)-N(1)	118.9(2)
C(26)-C(21)-N(1)	119.4(2)
C(23)-C(22)-C(21)	117.7(3)
C(23)-C(22)-C(27)	119.8(3)
C(21)-C(22)-C(27)	122.5(2)

C(22)-C(27)-C(28)	110.9(2)
C(22)-C(27)-C(29)	112.1(3)
C(28)-C(27)-C(29)	109.8(3)
C(22)-C(27)-H(27)	108.0
C(28)-C(27)-H(27)	108.0
C(29)-C(27)-H(27)	108.0
C(27)-C(28)-H(28A)	109.5
C(27)-C(28)-H(28B)	109.5
H(28A)-C(28)-H(28B)	109.5
C(27)-C(28)-H(28C)	109.5
H(28A)-C(28)-H(28C)	109.5
H(28B)-C(28)-H(28C)	109.5
C(27)-C(29)-H(29A)	109.5
C(27)-C(29)-H(29B)	109.5
H(29A)-C(29)-H(29B)	109.5
C(27)-C(29)-H(29C)	109.5
H(29A)-C(29)-H(29C)	109.5
H(29B)-C(29)-H(29C)	109.5
C(24)-C(23)-C(22)	121.8(3)
C(24)-C(23)-H(23)	119.1
C(22)-C(23)-H(23)	119.1
C(23)-C(24)-C(25)	119.8(3)
C(23)-C(24)-H(24)	120.1
C(25)-C(24)-H(24)	120.1
C(24)-C(25)-C(26)	121.7(3)
C(24)-C(25)-H(25)	119.1
C(26)-C(25)-H(25)	119.1
C(25)-C(26)-C(21)	117.4(3)
C(25)-C(26)-C(30)	119.8(3)
C(21)-C(26)-C(30)	122.8(2)
C(26)-C(30)-C(32)	113.9(3)
C(26)-C(30)-C(31)	110.1(3)
C(32)-C(30)-C(31)	110.6(3)
C(26)-C(30)-H(30)	107.3
C(32)-C(30)-H(30)	107.3
C(31)-C(30)-H(30)	107.3
C(30)-C(31)-H(31A)	109.5
C(30)-C(31)-H(31B)	109.5
H(31A)-C(31)-H(31B)	109.5
C(30)-C(31)-H(31C)	109.5
H(31A)-C(31)-H(31C)	109.5
H(31B)-C(31)-H(31C)	109.5
C(30)-C(32)-H(32A)	109.5
C(30)-C(32)-H(32B)	109.5
H(32A)-C(32)-H(32B)	109.5
C(30)-C(32)-H(32C)	109.5
H(32A)-C(32)-H(32C)	109.5
H(32B)-C(32)-H(32C)	109.5
N(1)-C(12)-C(13)	102.4(2)
N(1)-C(12)-H(12A)	111.3
C(13)-C(12)-H(12A)	111.3
N(1)-C(12)-H(12B)	111.3
C(13)-C(12)-H(12B)	111.3
H(12A)-C(12)-H(12B)	109.2
N(2)-C(13)-C(12)	102.4(2)
N(2)-C(13)-H(13A)	111.3

C(12)-C(13)-H(13A)	111.3
N(2)-C(13)-H(13B)	111.3
C(12)-C(13)-H(13B)	111.3
H(13A)-C(13)-H(13B)	109.2
C(11)-N(2)-C(41)	127.4(2)
C(11)-N(2)-C(13)	114.0(2)
C(41)-N(2)-C(13)	118.32(19)
C(46)-C(41)-C(42)	122.1(2)
C(46)-C(41)-N(2)	117.9(2)
C(42)-C(41)-N(2)	119.7(2)
C(43)-C(42)-C(41)	117.1(2)
C(43)-C(42)-C(47)	120.9(2)
C(41)-C(42)-C(47)	122.0(2)
C(48)-C(47)-C(49)	109.1(3)
C(48)-C(47)-C(42)	112.0(3)
C(49)-C(47)-C(42)	113.9(3)
C(48)-C(47)-H(47)	107.2
C(49)-C(47)-H(47)	107.2
C(42)-C(47)-H(47)	107.2
C(47)-C(48)-H(48A)	109.5
C(47)-C(48)-H(48B)	109.5
H(48A)-C(48)-H(48B)	109.5
C(47)-C(48)-H(48C)	109.5
H(48A)-C(48)-H(48C)	109.5
H(48B)-C(48)-H(48C)	109.5
C(47)-C(49)-H(49A)	109.5
C(47)-C(49)-H(49B)	109.5
H(49A)-C(49)-H(49B)	109.5
C(47)-C(49)-H(49C)	109.5
H(49A)-C(49)-H(49C)	109.5
H(49B)-C(49)-H(49C)	109.5
C(44)-C(43)-C(42)	121.5(3)
C(44)-C(43)-H(43)	119.2
C(42)-C(43)-H(43)	119.2
C(45)-C(44)-C(43)	120.2(3)
C(45)-C(44)-H(44)	119.9
C(43)-C(44)-H(44)	119.9
C(44)-C(45)-C(46)	121.1(3)
C(44)-C(45)-H(45)	119.4
C(46)-C(45)-H(45)	119.4
C(45)-C(46)-C(41)	117.8(2)
C(45)-C(46)-C(50)	119.5(2)
C(41)-C(46)-C(50)	122.8(2)
C(46)-C(50)-C(52)	111.4(3)
C(46)-C(50)-C(51)	111.9(3)
C(52)-C(50)-C(51)	108.1(3)
C(46)-C(50)-H(50)	108.5
C(52)-C(50)-H(50)	108.5
C(51)-C(50)-H(50)	108.5
C(50)-C(51)-H(51A)	109.5
C(50)-C(51)-H(51B)	109.5
H(51A)-C(51)-H(51B)	109.5
C(50)-C(51)-H(51C)	109.5
H(51A)-C(51)-H(51C)	109.5
H(51B)-C(51)-H(51C)	109.5
C(50)-C(52)-H(52A)	109.5

C(50)-C(52)-H(52B)	109.5
H(52A)-C(52)-H(52B)	109.5
C(50)-C(52)-H(52C)	109.5
H(52A)-C(52)-H(52C)	109.5
H(52B)-C(52)-H(52C)	109.5
C(11)-Ni(2)-C(9)	102.53(11)
C(11)-Ni(2)-C(1)	135.80(10)
C(9)-Ni(2)-C(1)	40.52(10)
C(11)-Ni(2)-Cl(1)	111.34(7)
C(9)-Ni(2)-Cl(1)	146.13(8)
C(1)-Ni(2)-Cl(1)	108.22(7)
C(11)-Ni(2)-Ni(1)	167.62(7)
C(9)-Ni(2)-Ni(1)	89.83(8)
C(1)-Ni(2)-Ni(1)	55.70(7)
Cl(1)-Ni(2)-Ni(1)	56.31(2)
N(4)-C(16)-N(3)	105.4(2)
N(4)-C(16)-Ni(1)	131.45(18)
N(3)-C(16)-Ni(1)	122.79(18)
C(16)-N(3)-C(61)	125.9(2)
C(16)-N(3)-C(17)	113.8(2)
C(61)-N(3)-C(17)	118.0(2)
C(66)-C(61)-C(62)	122.0(3)
C(66)-C(61)-N(3)	118.1(3)
C(62)-C(61)-N(3)	119.7(3)
C(63)-C(62)-C(61)	117.8(3)
C(63)-C(62)-C(67)	119.2(3)
C(61)-C(62)-C(67)	122.9(3)
C(62)-C(67)-C(69)	113.6(3)
C(62)-C(67)-C(68)	110.5(3)
C(69)-C(67)-C(68)	109.2(3)
C(62)-C(67)-H(67)	107.8
C(69)-C(67)-H(67)	107.8
C(68)-C(67)-H(67)	107.8
C(67)-C(68)-H(68A)	109.5
C(67)-C(68)-H(68B)	109.5
H(68A)-C(68)-H(68B)	109.5
C(67)-C(68)-H(68C)	109.5
H(68A)-C(68)-H(68C)	109.5
H(68B)-C(68)-H(68C)	109.5
C(67)-C(69)-H(69A)	109.5
C(67)-C(69)-H(69B)	109.5
H(69A)-C(69)-H(69B)	109.5
C(67)-C(69)-H(69C)	109.5
H(69A)-C(69)-H(69C)	109.5
H(69B)-C(69)-H(69C)	109.5
C(64)-C(63)-C(62)	121.3(3)
C(64)-C(63)-H(63)	119.4
C(62)-C(63)-H(63)	119.4
C(63)-C(64)-C(65)	120.7(3)
C(63)-C(64)-H(64)	119.7
C(65)-C(64)-H(64)	119.7
C(64)-C(65)-C(66)	121.0(3)
C(64)-C(65)-H(65)	119.5
C(66)-C(65)-H(65)	119.5
C(61)-C(66)-C(65)	117.2(3)
C(61)-C(66)-C(70)	123.1(3)

C(65)-C(66)-C(70)	119.8(3)
C(66)-C(70)-C(71)	112.1(3)
C(66)-C(70)-C(72)	111.4(3)
C(71)-C(70)-C(72)	109.3(3)
C(66)-C(70)-H(70)	108.0
C(71)-C(70)-H(70)	108.0
C(72)-C(70)-H(70)	108.0
C(70)-C(71)-H(71A)	109.5
C(70)-C(71)-H(71B)	109.5
H(71A)-C(71)-H(71B)	109.5
C(70)-C(71)-H(71C)	109.5
H(71A)-C(71)-H(71C)	109.5
H(71B)-C(71)-H(71C)	109.5
C(70)-C(72)-H(72A)	109.5
C(70)-C(72)-H(72B)	109.5
H(72A)-C(72)-H(72B)	109.5
C(70)-C(72)-H(72C)	109.5
H(72A)-C(72)-H(72C)	109.5
H(72B)-C(72)-H(72C)	109.5
N(3)-C(17)-C(18)	101.3(2)
N(3)-C(17)-H(17A)	111.5
C(18)-C(17)-H(17A)	111.5
N(3)-C(17)-H(17B)	111.5
C(18)-C(17)-H(17B)	111.5
H(17A)-C(17)-H(17B)	109.3
N(4)-C(18)-C(17)	102.1(2)
N(4)-C(18)-H(18A)	111.4
C(17)-C(18)-H(18A)	111.4
N(4)-C(18)-H(18B)	111.4
C(17)-C(18)-H(18B)	111.4
H(18A)-C(18)-H(18B)	109.2
C(16)-N(4)-C(81)	126.7(2)
C(16)-N(4)-C(18)	113.1(2)
C(81)-N(4)-C(18)	119.3(2)
C(82)-C(81)-C(86)	122.4(2)
C(82)-C(81)-N(4)	117.6(2)
C(86)-C(81)-N(4)	119.8(2)
C(81)-C(82)-C(83)	117.1(3)
C(81)-C(82)-C(87)	123.3(2)
C(83)-C(82)-C(87)	119.7(3)
C(89)-C(87)-C(82)	111.7(3)
C(89)-C(87)-C(88)	110.9(3)
C(82)-C(87)-C(88)	111.3(3)
C(89)-C(87)-H(87)	107.5
C(82)-C(87)-H(87)	107.5
C(88)-C(87)-H(87)	107.5
C(87)-C(88)-H(88A)	109.5
C(87)-C(88)-H(88B)	109.5
H(88A)-C(88)-H(88B)	109.5
C(87)-C(88)-H(88C)	109.5
H(88A)-C(88)-H(88C)	109.5
H(88B)-C(88)-H(88C)	109.5
C(87)-C(89)-H(89A)	109.5
C(87)-C(89)-H(89B)	109.5
H(89A)-C(89)-H(89B)	109.5
C(87)-C(89)-H(89C)	109.5

H(89A)-C(89)-H(89C)	109.5
H(89B)-C(89)-H(89C)	109.5
C(84)-C(83)-C(82)	121.4(3)
C(84)-C(83)-H(83)	119.3
C(82)-C(83)-H(83)	119.3
C(85)-C(84)-C(83)	120.2(3)
C(85)-C(84)-H(84)	119.9
C(83)-C(84)-H(84)	119.9
C(84)-C(85)-C(86)	121.7(3)
C(84)-C(85)-H(85)	119.2
C(86)-C(85)-H(85)	119.2
C(85)-C(86)-C(81)	117.3(3)
C(85)-C(86)-C(90)	119.8(3)
C(81)-C(86)-C(90)	122.9(2)
C(86)-C(90)-C(91)	112.3(3)
C(86)-C(90)-C(92)	110.6(3)
C(91)-C(90)-C(92)	110.3(3)
C(86)-C(90)-H(90)	107.8
C(91)-C(90)-H(90)	107.8
C(92)-C(90)-H(90)	107.8
C(90)-C(91)-H(91A)	109.5
C(90)-C(91)-H(91B)	109.5
H(91A)-C(91)-H(91B)	109.5
C(90)-C(91)-H(91C)	109.5
H(91A)-C(91)-H(91C)	109.5
H(91B)-C(91)-H(91C)	109.5
C(90)-C(92)-H(92A)	109.5
C(90)-C(92)-H(92B)	109.5
H(92A)-C(92)-H(92B)	109.5
C(90)-C(92)-H(92C)	109.5
H(92A)-C(92)-H(92C)	109.5
H(92B)-C(92)-H(92C)	109.5

X-Ray Data for 3a (CCDC 939471)

The compound **3a** crystallizes in the triclinic space group *P-1* with one molecule in the asymmetric unit.

Table S20: Crystal data and structure refinement for **3a**

Empirical formula	C32 H41 N2 Ni	
Formula weight	512.38	
Temperature	93(2) K	
Wavelength	1.54187 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 9.1674(2) Å	α= 92.759(7)°.
	b = 9.3136(2) Å	β= 90.240(6)°.
	c = 18.2167(13) Å	γ = 116.310(8)°.
Volume	1392.06(15) $Å^3$	
Z	2	
Density (calculated)	1.222 Mg/m^3	
Absorption coefficient	1.151 mm^{-1}	
F(000)	550	
Crystal size	0.15 x 0.15 x 0.07 mm^3	

Theta range for data collection	2.43 to 68.29°.
Index ranges	-11<=h<=11, -10<=k<=10, -21<=l<=21
Reflections collected	48798
Independent reflections	4964 [R(int) = 0.0533]
Completeness to theta = 68.29°	97.7 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.9238 and 0.8463
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	4964 / 0 / 324
Goodness-of-fit on F^2	1.033
Final R indices [I>2sigma(I)]	R1 = 0.0288, wR2 = 0.0733
R indices (all data)	R1 = 0.0291, wR2 = 0.0735
Largest diff. peak and hole	0.265 and -0.258 e.Å^{-3}

Table S21: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^{2}x 10^3) for **3a**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Ni(1)	4779(1)	4464(1)	2632(1)	17(1)
C(1)	6066(2)	6594(2)	2016(1)	22(1)
C(2)	5716(2)	7002(2)	2727(1)	23(1)
C(3)	3990(2)	6290(2)	2788(1)	23(1)
C(4)	3289(2)	5445(2)	2114(1)	23(1)
C(5)	4571(2)	5604(2)	1643(1)	23(1)
C(11)	4911(1)	2509(2)	2572(1)	15(1)
N(1)	5347(1)	1849(1)	3138(1)	15(1)
C(21)	5770(2)	2628(2)	3862(1)	16(1)
C(22)	7323(2)	3913(2)	3980(1)	19(1)
C(27)	8601(2)	4347(2)	3393(1)	22(1)
C(28)	9789(2)	6131(2)	3422(1)	34(1)
C(29)	9533(2)	3343(2)	3461(1)	27(1)
C(23)	7679(2)	4716(2)	4672(1)	23(1)
C(24)	6540(2)	4251(2)	5217(1)	25(1)
C(25)	5036(2)	2945(2)	5094(1)	23(1)
C(26)	4615(2)	2091(2)	4412(1)	18(1)
C(30)	2949(2)	677(2)	4269(1)	21(1)
C(31)	1708(2)	1269(2)	4045(1)	30(1)
C(32)	2365(2)	-378(2)	4927(1)	30(1)
C(12)	5390(2)	425(2)	2912(1)	18(1)
C(13)	4974(2)	158(2)	2192(1)	18(1)
N(2)	4688(1)	1436(1)	1988(1)	15(1)
C(41)	4150(2)	1629(2)	1271(1)	16(1)
C(42)	5290(2)	2198(2)	716(1)	18(1)
C(47)	7074(2)	2583(2)	839(1)	20(1)
C(48)	8045(2)	4316(2)	1141(1)	29(1)
C(49)	7852(2)	2289(2)	143(1)	28(1)
C(43)	4738(2)	2457(2)	43(1)	23(1)
C(44)	3138(2)	2170(2)	-66(1)	25(1)
C(45)	2028(2)	1574(2)	487(1)	23(1)
C(46)	2505(2)	1274(2)	1168(1)	18(1)
C(50)	1278(2)	516(2)	1760(1)	24(1)
C(51)	-206(2)	855(2)	1716(1)	30(1)
C(52)	734(2)	-1298(2)	1723(1)	34(1)

Table S22: Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **3a**. 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}
Ni(1)	22(1)	14(1)	15(1)	2(1)	0(1)	10(1)
C(1)	23(1)	17(1)	29(1)	8(1)	7(1)	10(1)
C(2)	27(1)	15(1)	27(1)	0(1)	-4(1)	11(1)
C(3)	30(1)	21(1)	25(1)	6(1)	7(1)	17(1)
C(4)	21(1)	20(1)	31(1)	7(1)	-2(1)	10(1)
C(5)	33(1)	19(1)	18(1)	6(1)	1(1)	14(1)
C(11)	13(1)	16(1)	14(1)	2(1)	-1(1)	5(1)
N(1)	18(1)	14(1)	14(1)	1(1)	-1(1)	8(1)
C(21)	20(1)	16(1)	13(1)	1(1)	-2(1)	11(1)
C(22)	20(1)	19(1)	17(1)	4(1)	-2(1)	9(1)
C(27)	18(1)	27(1)	18(1)	4(1)	-1(1)	6(1)
C(28)	28(1)	28(1)	44(1)	13(1)	10(1)	10(1)
C(29)	25(1)	27(1)	28(1)	0(1)	4(1)	10(1)
C(23)	23(1)	22(1)	20(1)	0(1)	-5(1)	6(1)
C(24)	31(1)	26(1)	17(1)	-3(1)	-4(1)	12(1)
C(25)	26(1)	26(1)	16(1)	2(1)	3(1)	13(1)
C(26)	20(1)	18(1)	18(1)	3(1)	0(1)	11(1)
C(30)	20(1)	20(1)	21(1)	1(1)	2(1)	8(1)
C(31)	21(1)	31(1)	36(1)	5(1)	-2(1)	9(1)
C(32)	28(1)	27(1)	32(1)	10(1)	4(1)	7(1)
C(12)	21(1)	15(1)	20(1)	2(1)	-1(1)	10(1)
C(13)	21(1)	15(1)	20(1)	0(1)	0(1)	10(1)
N(2)	17(1)	15(1)	14(1)	1(1)	-1(1)	7(1)
C(41)	21(1)	14(1)	14(1)	-1(1)	-3(1)	8(1)
C(42)	21(1)	15(1)	18(1)	0(1)	0(1)	8(1)
C(47)	20(1)	22(1)	20(1)	3(1)	2(1)	11(1)
C(48)	21(1)	29(1)	33(1)	-6(1)	-1(1)	9(1)
C(49)	28(1)	31(1)	28(1)	-1(1)	6(1)	16(1)
C(43)	28(1)	23(1)	17(1)	3(1)	2(1)	12(1)
C(44)	32(1)	28(1)	16(1)	2(1)	-5(1)	14(1)
C(45)	22(1)	24(1)	22(1)	-3(1)	-6(1)	11(1)
C(46)	20(1)	16(1)	17(1)	-3(1)	-2(1)	7(1)
C(50)	18(1)	32(1)	18(1)	-1(1)	-2(1)	7(1)
C(51)	21(1)	30(1)	34(1)	-6(1)	2(1)	9(1)
C(52)	22(1)	35(1)	45(1)	18(1)	6(1)	10(1)

Table S23: Bond lengths (Å) for **3a**

Ni(1)-C(11)	1.8753(13)
Ni(1)-C(2)	2.1246(14)
Ni(1)-C(3)	2.1315(13)
Ni(1)-C(1)	2.1755(14)
Ni(1)-C(4)	2.1835(13)
Ni(1)-C(5)	2.1835(13)
C(1)-C(2)	1.410(2)
C(1)-C(5)	1.415(2)
C(1)-H(1)	0.9500
C(2)-C(3)	1.426(2)
C(2)-H(2)	0.9500
C(3)-C(4)	1.412(2)
C(3)-H(3)	0.9500
C(4)-C(5)	1.416(2)

C(4)-H(4)	0.9500
C(5)-H(5)	0.9500
C(11)-N(1)	1.3701(16)
C(11)-N(2)	1.3726(16)
N(1)-C(12)	1.3870(17)
N(1)-C(21)	1.4392(16)
C(21)-C(26)	1.4006(18)
C(21)-C(22)	1.4028(18)
C(22)-C(23)	1.3944(19)
C(22)-C(27)	1.5206(18)
C(27)-C(28)	1.529(2)
C(27)-C(29)	1.529(2)
C(27)-H(27)	1.0000
C(28)-H(28A)	0.9800
C(28)-H(28B)	0.9800
C(28)-H(28C)	0.9800
C(29)-H(29A)	0.9800
C(29)-H(29B)	0.9800
C(29)-H(29C)	0.9800
C(23)-C(24)	1.384(2)
C(23)-H(23)	0.9500
C(24)-C(25)	1.384(2)
C(24)-H(24)	0.9500
C(25)-C(26)	1.3973(19)
C(25)-H(25)	0.9500
C(26)-C(30)	1.5218(18)
C(30)-C(31)	1.529(2)
C(30)-C(32)	1.5297(19)
C(30)-H(30)	1.0000
C(31)-H(31A)	0.9800
C(31)-H(31B)	0.9800
C(31)-H(31C)	0.9800
C(32)-H(32A)	0.9800
C(32)-H(32B)	0.9800
C(32)-H(32C)	0.9800
C(12)-C(13)	1.3440(19)
C(12)-H(12)	0.9500
C(13)-N(2)	1.3942(17)
C(13)-H(13)	0.9500
N(2)-C(41)	1.4423(16)
C(41)-C(42)	1.4013(18)
C(41)-C(46)	1.4035(18)
C(42)-C(43)	1.3953(19)
C(42)-C(47)	1.5241(18)
C(47)-C(49)	1.5288(19)
C(47)-C(48)	1.529(2)
C(47)-H(47)	1.0000
C(48)-H(48A)	0.9800
C(48)-H(48B)	0.9800
C(48)-H(48C)	0.9800
C(49)-H(49A)	0.9800
C(49)-H(49B)	0.9800
C(49)-H(49C)	0.9800
C(43)-C(44)	1.381(2)
C(43)-H(43)	0.9500
C(44)-C(45)	1.385(2)

C(44)-H(44)	0.9500
C(45)-C(46)	1.3941(19)
C(45)-H(45)	0.9500
C(46)-C(50)	1.5185(18)
C(50)-C(51)	1.530(2)
C(50)-C(52)	1.533(2)
C(50)-H(50)	1.0000
C(51)-H(51A)	0.9800
C(51)-H(51B)	0.9800
C(51)-H(51C)	0.9800
C(52)-H(52A)	0.9800
C(52)-H(52B)	0.9800
C(52)-H(52C)	0.9800

Table S24: Bond angles (°) for **3a**

C(11)-Ni(1)-C(2)	155.44(5)
C(11)-Ni(1)-C(3)	164.72(5)
C(2)-Ni(1)-C(3)	39.16(5)
C(11)-Ni(1)-C(1)	129.38(5)
C(2)-Ni(1)-C(1)	38.25(5)
C(3)-Ni(1)-C(1)	64.36(5)
C(11)-Ni(1)-C(4)	136.17(5)
C(2)-Ni(1)-C(4)	64.34(5)
C(3)-Ni(1)-C(4)	38.18(5)
C(1)-Ni(1)-C(4)	63.51(5)
C(11)-Ni(1)-C(5)	121.21(5)
C(2)-Ni(1)-C(5)	64.04(5)
C(3)-Ni(1)-C(5)	63.99(5)
C(1)-Ni(1)-C(5)	37.87(5)
C(4)-Ni(1)-C(5)	37.85(5)
C(2)-C(1)-C(5)	107.99(12)
C(2)-C(1)-Ni(1)	68.92(8)
C(5)-C(1)-Ni(1)	71.37(8)
C(2)-C(1)-H(1)	126.0
C(5)-C(1)-H(1)	126.0
Ni(1)-C(1)-H(1)	125.3
C(1)-C(2)-C(3)	107.97(12)
C(1)-C(2)-Ni(1)	72.83(8)
C(3)-C(2)-Ni(1)	70.68(8)
C(1)-C(2)-H(2)	126.0
C(3)-C(2)-H(2)	126.0
Ni(1)-C(2)-H(2)	122.2
C(4)-C(3)-C(2)	107.85(12)
C(4)-C(3)-Ni(1)	72.90(8)
C(2)-C(3)-Ni(1)	70.16(8)
C(4)-C(3)-H(3)	126.1
C(2)-C(3)-H(3)	126.1
Ni(1)-C(3)-H(3)	122.5
C(3)-C(4)-C(5)	107.89(12)
C(3)-C(4)-Ni(1)	68.91(8)
C(5)-C(4)-Ni(1)	71.07(8)
C(3)-C(4)-H(4)	126.1
C(5)-C(4)-H(4)	126.1
Ni(1)-C(4)-H(4)	125.5
C(1)-C(5)-C(4)	108.26(12)

C(1)-C(5)-Ni(1)	70.76(8)
C(4)-C(5)-Ni(1)	71.07(8)
C(1)-C(5)-H(5)	125.9
C(4)-C(5)-H(5)	125.9
Ni(1)-C(5)-H(5)	123.9
N(1)-C(11)-N(2)	102.92(10)
N(1)-C(11)-Ni(1)	125.66(9)
N(2)-C(11)-Ni(1)	131.37(9)
C(11)-N(1)-C(12)	111.87(10)
C(11)-N(1)-C(21)	122.02(10)
C(12)-N(1)-C(21)	126.06(10)
C(26)-C(21)-C(22)	123.04(12)
C(26)-C(21)-N(1)	118.95(11)
C(22)-C(21)-N(1)	118.00(11)
C(23)-C(22)-C(21)	117.28(12)
C(23)-C(22)-C(27)	121.30(12)
C(21)-C(22)-C(27)	121.31(12)
C(22)-C(27)-C(28)	113.32(12)
C(22)-C(27)-C(29)	109.48(11)
C(28)-C(27)-C(29)	110.07(12)
C(22)-C(27)-H(27)	107.9
C(28)-C(27)-H(27)	107.9
C(29)-C(27)-H(27)	107.9
C(27)-C(28)-H(28A)	109.5
C(27)-C(28)-H(28B)	109.5
H(28A)-C(28)-H(28B)	109.5
C(27)-C(28)-H(28C)	109.5
H(28A)-C(28)-H(28C)	109.5
H(28B)-C(28)-H(28C)	109.5
C(27)-C(29)-H(29A)	109.5
C(27)-C(29)-H(29B)	109.5
H(29A)-C(29)-H(29B)	109.5
C(27)-C(29)-H(29C)	109.5
H(29A)-C(29)-H(29C)	109.5
H(29B)-C(29)-H(29C)	109.5
C(24)-C(23)-C(22)	120.95(13)
C(24)-C(23)-H(23)	119.5
C(22)-C(23)-H(23)	119.5
C(23)-C(24)-C(25)	120.51(13)
C(23)-C(24)-H(24)	119.7
C(25)-C(24)-H(24)	119.7
C(24)-C(25)-C(26)	121.03(13)
C(24)-C(25)-H(25)	119.5
C(26)-C(25)-H(25)	119.5
C(25)-C(26)-C(21)	117.11(12)
C(25)-C(26)-C(30)	121.12(12)
C(21)-C(26)-C(30)	121.74(12)
C(26)-C(30)-C(31)	110.21(11)
C(26)-C(30)-C(32)	112.58(11)
C(31)-C(30)-C(32)	110.86(12)
C(26)-C(30)-H(30)	107.7
C(31)-C(30)-H(30)	107.7
C(32)-C(30)-H(30)	107.7
C(30)-C(31)-H(31A)	109.5
C(30)-C(31)-H(31B)	109.5
H(31A)-C(31)-H(31B)	109.5

C(30)-C(31)-H(31C)	109.5
H(31A)-C(31)-H(31C)	109.5
H(31B)-C(31)-H(31C)	109.5
C(30)-C(32)-H(32A)	109.5
C(30)-C(32)-H(32B)	109.5
H(32A)-C(32)-H(32B)	109.5
C(30)-C(32)-H(32C)	109.5
H(32A)-C(32)-H(32C)	109.5
H(32B)-C(32)-H(32C)	109.5
C(13)-C(12)-N(1)	106.99(11)
C(13)-C(12)-H(12)	126.5
N(1)-C(12)-H(12)	126.5
C(12)-C(13)-N(2)	106.50(11)
C(12)-C(13)-H(13)	126.8
N(2)-C(13)-H(13)	126.8
C(11)-N(2)-C(13)	111.72(10)
C(11)-N(2)-C(41)	122.08(10)
C(13)-N(2)-C(41)	126.14(11)
C(42)-C(41)-C(46)	122.99(12)
C(42)-C(41)-N(2)	119.00(11)
C(46)-C(41)-N(2)	117.99(11)
C(43)-C(42)-C(41)	117.11(12)
C(43)-C(42)-C(47)	120.58(12)
C(41)-C(42)-C(47)	122.27(11)
C(42)-C(47)-C(49)	112.81(11)
C(42)-C(47)-C(48)	110.44(11)
C(49)-C(47)-C(48)	110.00(12)
C(42)-C(47)-H(47)	107.8
C(49)-C(47)-H(47)	107.8
C(48)-C(47)-H(47)	107.8
C(47)-C(48)-H(48A)	109.5
C(47)-C(48)-H(48B)	109.5
H(48A)-C(48)-H(48B)	109.5
C(47)-C(48)-H(48C)	109.5
H(48A)-C(48)-H(48C)	109.5
H(48B)-C(48)-H(48C)	109.5
C(47)-C(49)-H(49A)	109.5
C(47)-C(49)-H(49B)	109.5
H(49A)-C(49)-H(49B)	109.5
C(47)-C(49)-H(49C)	109.5
H(49A)-C(49)-H(49C)	109.5
H(49B)-C(49)-H(49C)	109.5
C(44)-C(43)-C(42)	121.13(13)
C(44)-C(43)-H(43)	119.4
C(42)-C(43)-H(43)	119.4
C(43)-C(44)-C(45)	120.56(12)
C(43)-C(44)-H(44)	119.7
C(45)-C(44)-H(44)	119.7
C(44)-C(45)-C(46)	120.85(13)
C(44)-C(45)-H(45)	119.6
C(46)-C(45)-H(45)	119.6
C(45)-C(46)-C(41)	117.30(12)
C(45)-C(46)-C(50)	121.35(12)
C(41)-C(46)-C(50)	121.28(12)
C(46)-C(50)-C(51)	113.85(12)
C(46)-C(50)-C(52)	109.38(12)

C(51)-C(50)-C(52)	110.07(12)
C(46)-C(50)-H(50)	107.8
C(51)-C(50)-H(50)	107.8
C(52)-C(50)-H(50)	107.8
C(50)-C(51)-H(51A)	109.5
C(50)-C(51)-H(51B)	109.5
H(51A)-C(51)-H(51B)	109.5
C(50)-C(51)-H(51C)	109.5
H(51A)-C(51)-H(51C)	109.5
H(51B)-C(51)-H(51C)	109.5
C(50)-C(52)-H(52A)	109.5
C(50)-C(52)-H(52B)	109.5
H(52A)-C(52)-H(52B)	109.5
C(50)-C(52)-H(52C)	109.5
H(52A)-C(52)-H(52C)	109.5
H(52B)-C(52)-H(52C)	109.5

X-Ray Data for 4a (CCDC 939472)

Compound **4a** crystallizes in the monoclinic space group *Cc* with one molecule in the asymmetric unit.

Table S25: Crystal data and structure refinement for **4a**

Empirical formula	C36 H43 N2 Ni	
Formula weight	562.43	
Temperature	93(2) K	
Wavelength	1.54187 Å	
Crystal system	Monoclinic	
Space group	Cc	
Unit cell dimensions	a = 16.3437(4) Å	α= 90°.
	b = 13.7905(3) Å	β= 107.721(8)°.
	c = 14.5977(10) Å	γ = 90°.
Volume	3134.0(2) $Å^3$	
Z	4	
Density (calculated)	1.192 Mg/m^3	
Absorption coefficient	1.068 mm^{-1}	
F(000)	1204	
Crystal size	0.15 x 0.03 x 0.03 mm^3	
Theta range for data collection	4.28 to 68.27°.	
Index ranges	-19<=h<=19, -16<=k<=16, -17<=l<=17	
Reflections collected	23494	
Independent reflections	5142 [R(int) = 0.0899]	
Completeness to theta = 68.27°	98.2 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.9687 and 0.8562	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	5142 / 2 / 360	
Goodness-of-fit on F^2	1.100	
Final R indices [I>2sigma(I)]	R1 = 0.0515, wR2 = 0.1232	
R indices (all data)	R1 = 0.0623, wR2 = 0.1394	
Absolute structure parameter	0.07(3)	
Largest diff. peak and hole	0.515 and -0.575 e.$Å^{-3}$	

Table S26: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for **4a**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Ni(1)	10091(1)	2772(1)	9990(1)	34(1)
C(1)	10675(3)	2168(4)	11350(3)	43(1)
C(2)	10876(3)	3147(4)	11392(3)	44(1)
C(3)	11429(3)	3340(3)	10836(3)	38(1)
C(4)	11860(3)	4194(3)	10672(3)	41(1)
C(5)	12374(3)	4114(3)	10098(3)	41(1)
C(6)	12473(3)	3240(4)	9642(3)	42(1)
C(7)	12050(3)	2426(4)	9755(3)	41(1)
C(8)	11527(3)	2456(3)	10385(3)	29(1)
C(9)	11047(3)	1707(3)	10664(3)	39(1)
C(11)	9287(3)	2676(3)	8755(3)	26(1)
N(1)	9237(2)	2115(2)	7950(2)	24(1)
C(21)	9934(3)	1520(3)	7865(3)	28(1)
C(22)	10032(3)	584(3)	8252(3)	36(1)
C(27)	9409(3)	175(3)	8737(3)	41(1)
C(28)	9783(4)	-633(3)	9453(4)	53(1)
C(29)	8594(4)	-172(4)	7995(4)	57(1)
C(23)	10711(3)	36(3)	8125(3)	44(1)
C(24)	11245(3)	410(4)	7639(4)	49(1)
C(25)	11125(3)	1327(4)	7267(3)	43(1)
C(26)	10472(3)	1920(3)	7380(3)	34(1)
C(30)	10341(3)	2931(3)	6945(3)	37(1)
C(31)	11140(4)	3561(4)	7295(4)	58(1)
C(32)	10033(4)	2875(4)	5861(4)	55(1)
C(12)	8484(3)	2275(3)	7232(3)	28(1)
C(13)	8035(3)	2936(3)	7554(3)	28(1)
N(2)	8535(2)	3180(2)	8475(2)	26(1)
C(41)	8311(3)	3840(3)	9123(3)	28(1)
C(42)	7909(3)	3456(3)	9768(3)	33(1)
C(47)	7676(3)	2397(3)	9767(3)	34(1)
C(48)	7979(4)	1974(4)	10786(4)	50(1)
C(49)	6721(4)	2247(4)	9294(4)	55(1)
C(43)	7713(3)	4113(3)	10394(3)	40(1)
C(44)	7923(3)	5079(3)	10397(3)	41(1)
C(45)	8316(3)	5434(3)	9746(3)	36(1)
C(46)	8514(3)	4819(3)	9084(3)	30(1)
C(50)	8944(3)	5207(3)	8362(3)	34(1)
C(51)	9914(3)	5092(3)	8731(4)	42(1)
C(52)	8709(3)	6263(3)	8078(3)	46(1)

Table S27: Anisotropic displacement parameters ($Å^2$x 10^3) for **4a**. 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}
Ni(1)	30(1)	48(1)	24(1)	-1(1)	6(1)	6(1)
C(1)	27(3)	67(3)	34(2)	20(2)	7(2)	0(2)
C(2)	38(3)	55(3)	35(2)	-2(2)	3(2)	-1(2)
C(3)	33(2)	45(3)	32(2)	-3(2)	3(2)	6(2)
C(4)	37(3)	39(3)	42(3)	1(2)	3(2)	-1(2)
C(5)	44(3)	38(3)	38(2)	2(2)	7(2)	1(2)

C(6)	36(3)	54(3)	38(2)	6(2)	12(2)	2(2)
C(7)	40(3)	46(3)	38(3)	3(2)	13(2)	3(2)
C(8)	24(2)	37(2)	26(2)	4(2)	6(2)	-1(2)
C(9)	31(2)	33(2)	43(2)	6(2)	-2(2)	4(2)
C(11)	25(2)	27(2)	29(2)	2(2)	14(2)	1(2)
N(1)	24(2)	29(2)	18(2)	0(1)	5(1)	2(1)
C(21)	28(2)	30(2)	28(2)	-2(2)	11(2)	6(2)
C(22)	45(3)	32(2)	31(2)	-4(2)	11(2)	8(2)
C(27)	47(3)	35(2)	40(2)	2(2)	14(2)	-2(2)
C(28)	63(4)	42(3)	47(3)	9(2)	7(3)	-5(3)
C(29)	55(3)	56(3)	54(3)	11(3)	6(3)	-7(3)
C(23)	49(3)	35(3)	40(3)	-4(2)	4(2)	13(2)
C(24)	40(3)	63(3)	43(3)	-14(2)	12(2)	14(2)
C(25)	35(3)	61(3)	34(2)	-4(2)	14(2)	8(2)
C(26)	28(2)	49(3)	25(2)	-5(2)	9(2)	2(2)
C(30)	34(3)	46(3)	35(2)	2(2)	16(2)	0(2)
C(31)	53(4)	69(4)	51(3)	14(3)	15(3)	-9(3)
C(32)	60(4)	63(3)	37(3)	4(2)	7(3)	12(3)
C(12)	31(2)	28(2)	27(2)	1(2)	11(2)	0(2)
C(13)	27(2)	33(2)	23(2)	1(2)	7(2)	0(2)
N(2)	31(2)	26(2)	23(2)	-2(1)	13(1)	0(1)
C(41)	29(2)	28(2)	28(2)	0(2)	11(2)	4(2)
C(42)	41(3)	31(2)	33(2)	2(2)	20(2)	2(2)
C(47)	38(3)	35(2)	37(2)	0(2)	21(2)	-3(2)
C(48)	64(4)	42(3)	47(3)	9(2)	21(3)	-9(2)
C(49)	50(3)	53(3)	58(3)	-5(2)	11(3)	-14(3)
C(43)	46(3)	44(3)	34(2)	1(2)	19(2)	5(2)
C(44)	50(3)	42(3)	35(2)	-3(2)	19(2)	10(2)
C(45)	39(3)	31(2)	38(2)	-2(2)	10(2)	2(2)
C(46)	32(2)	28(2)	30(2)	2(2)	10(2)	2(2)
C(50)	38(3)	34(2)	30(2)	3(2)	9(2)	-5(2)
C(51)	41(3)	39(3)	51(3)	-3(2)	22(2)	-6(2)
C(52)	45(3)	45(3)	45(3)	17(2)	12(2)	-2(2)

Table S28: Bond lengths (Å) for **4a**

Ni(1)-C(11)	1.884(4)
Ni(1)-C(1)	2.097(5)
Ni(1)-C(2)	2.123(5)
Ni(1)-C(9)	2.152(4)
Ni(1)-C(8)	2.282(4)
Ni(1)-C(3)	2.298(5)
C(1)-C(2)	1.386(7)
C(1)-C(9)	1.464(7)
C(1)-H(1)	0.9500
C(2)-C(3)	1.412(7)
C(2)-H(2)	0.9500
C(3)-C(8)	1.418(6)
C(3)-C(4)	1.430(7)
C(4)-C(5)	1.358(7)
C(4)-H(4)	0.9500
C(5)-C(6)	1.409(7)
C(5)-H(5)	0.9500
C(6)-C(7)	1.355(7)
C(6)-H(6)	0.9500
C(7)-C(8)	1.434(6)

C(7)-H(7)	0.9500
C(8)-C(9)	1.429(6)
C(9)-H(9)	0.9500
C(11)-N(2)	1.361(5)
C(11)-N(1)	1.389(5)
N(1)-C(12)	1.369(5)
N(1)-C(21)	1.441(5)
C(21)-C(22)	1.397(6)
C(21)-C(26)	1.399(5)
C(22)-C(23)	1.401(6)
C(22)-C(27)	1.516(6)
C(27)-C(29)	1.516(7)
C(27)-C(28)	1.523(6)
C(27)-H(27)	1.0000
C(28)-H(28A)	0.9800
C(28)-H(28B)	0.9800
C(28)-H(28C)	0.9800
C(29)-H(29A)	0.9800
C(29)-H(29B)	0.9800
C(29)-H(29C)	0.9800
C(23)-C(24)	1.382(7)
C(23)-H(23)	0.9500
C(24)-C(25)	1.366(7)
C(24)-H(24)	0.9500
C(25)-C(26)	1.395(6)
C(25)-H(25)	0.9500
C(26)-C(30)	1.520(6)
C(30)-C(32)	1.509(7)
C(30)-C(31)	1.521(7)
C(30)-H(30)	1.0000
C(31)-H(31A)	0.9800
C(31)-H(31B)	0.9800
C(31)-H(31C)	0.9800
C(32)-H(32A)	0.9800
C(32)-H(32B)	0.9800
C(32)-H(32C)	0.9800
C(12)-C(13)	1.341(6)
C(12)-H(12)	0.9500
C(13)-N(2)	1.386(5)
C(13)-H(13)	0.9500
N(2)-C(41)	1.438(5)
C(41)-C(46)	1.396(5)
C(41)-C(42)	1.405(5)
C(42)-C(43)	1.392(6)
C(42)-C(47)	1.509(6)
C(47)-C(49)	1.517(7)
C(47)-C(48)	1.533(7)
C(47)-H(47)	1.0000
C(48)-H(48A)	0.9800
C(48)-H(48B)	0.9800
C(48)-H(48C)	0.9800
C(49)-H(49A)	0.9800
C(49)-H(49B)	0.9800
C(49)-H(49C)	0.9800
C(43)-C(44)	1.374(6)
C(43)-H(43)	0.9500

C(44)-C(45)	1.389(6)
C(44)-H(44)	0.9500
C(45)-C(46)	1.395(5)
C(45)-H(45)	0.9500
C(46)-C(50)	1.532(5)
C(50)-C(51)	1.519(6)
C(50)-C(52)	1.531(6)
C(50)-H(50)	1.0000
C(51)-H(51A)	0.9800
C(51)-H(51B)	0.9800
C(51)-H(51C)	0.9800
C(52)-H(52A)	0.9800
C(52)-H(52B)	0.9800
C(52)-H(52C)	0.9800

Table S29: Bond angles (°) for **4a**

C(11)-Ni(1)-C(1)	149.55(19)
C(11)-Ni(1)-C(2)	168.6(2)
C(1)-Ni(1)-C(2)	38.35(19)
C(11)-Ni(1)-C(9)	126.02(17)
C(1)-Ni(1)-C(9)	40.29(18)
C(2)-Ni(1)-C(9)	65.35(19)
C(11)-Ni(1)-C(8)	126.37(15)
C(1)-Ni(1)-C(8)	63.09(16)
C(2)-Ni(1)-C(8)	61.92(17)
C(9)-Ni(1)-C(8)	37.46(15)
C(11)-Ni(1)-C(3)	144.21(16)
C(1)-Ni(1)-C(3)	62.69(18)
C(2)-Ni(1)-C(3)	36.99(17)
C(9)-Ni(1)-C(3)	63.04(17)
C(8)-Ni(1)-C(3)	36.06(15)
C(2)-C(1)-C(9)	108.1(4)
C(2)-C(1)-Ni(1)	71.9(3)
C(9)-C(1)-Ni(1)	71.9(2)
C(2)-C(1)-H(1)	125.9
C(9)-C(1)-H(1)	125.9
Ni(1)-C(1)-H(1)	122.0
C(1)-C(2)-C(3)	110.0(4)
C(1)-C(2)-Ni(1)	69.8(3)
C(3)-C(2)-Ni(1)	78.2(3)
C(1)-C(2)-H(2)	125.0
C(3)-C(2)-H(2)	125.0
Ni(1)-C(2)-H(2)	118.6
C(2)-C(3)-C(8)	106.7(4)
C(2)-C(3)-C(4)	133.4(4)
C(8)-C(3)-C(4)	119.9(4)
C(2)-C(3)-Ni(1)	64.8(3)
C(8)-C(3)-Ni(1)	71.4(2)
C(4)-C(3)-Ni(1)	128.9(3)
C(5)-C(4)-C(3)	117.7(4)
C(5)-C(4)-H(4)	121.1
C(3)-C(4)-H(4)	121.1
C(4)-C(5)-C(6)	122.9(5)
C(4)-C(5)-H(5)	118.6
C(6)-C(5)-H(5)	118.6
C(7)-C(6)-C(5)	120.9(4)

C(7)-C(6)-H(6)	119.5
C(5)-C(6)-H(6)	119.5
C(6)-C(7)-C(8)	118.7(4)
C(6)-C(7)-H(7)	120.6
C(8)-C(7)-H(7)	120.6
C(3)-C(8)-C(9)	109.8(4)
C(3)-C(8)-C(7)	119.7(4)
C(9)-C(8)-C(7)	130.4(4)
C(3)-C(8)-Ni(1)	72.6(2)
C(9)-C(8)-Ni(1)	66.3(2)
C(7)-C(8)-Ni(1)	127.7(3)
C(8)-C(9)-C(1)	105.1(4)
C(8)-C(9)-Ni(1)	76.2(2)
C(1)-C(9)-Ni(1)	67.8(2)
C(8)-C(9)-H(9)	127.5
C(1)-C(9)-H(9)	127.5
Ni(1)-C(9)-H(9)	120.4
N(2)-C(11)-N(1)	102.4(3)
N(2)-C(11)-Ni(1)	123.7(3)
N(1)-C(11)-Ni(1)	133.9(3)
C(12)-N(1)-C(11)	111.5(3)
C(12)-N(1)-C(21)	125.2(3)
C(11)-N(1)-C(21)	123.1(3)
C(22)-C(21)-C(26)	123.8(4)
C(22)-C(21)-N(1)	119.2(3)
C(26)-C(21)-N(1)	117.0(3)
C(21)-C(22)-C(23)	116.3(4)
C(21)-C(22)-C(27)	121.4(4)
C(23)-C(22)-C(27)	122.2(4)
C(22)-C(27)-C(29)	110.7(4)
C(22)-C(27)-C(28)	114.0(4)
C(29)-C(27)-C(28)	110.0(4)
C(22)-C(27)-H(27)	107.3
C(29)-C(27)-H(27)	107.3
C(28)-C(27)-H(27)	107.3
C(27)-C(28)-H(28A)	109.5
C(27)-C(28)-H(28B)	109.5
H(28A)-C(28)-H(28B)	109.5
C(27)-C(28)-H(28C)	109.5
H(28A)-C(28)-H(28C)	109.5
H(28B)-C(28)-H(28C)	109.5
C(27)-C(29)-H(29A)	109.5
C(27)-C(29)-H(29B)	109.5
H(29A)-C(29)-H(29B)	109.5
C(27)-C(29)-H(29C)	109.5
H(29A)-C(29)-H(29C)	109.5
H(29B)-C(29)-H(29C)	109.5
C(24)-C(23)-C(22)	121.0(4)
C(24)-C(23)-H(23)	119.5
C(22)-C(23)-H(23)	119.5
C(25)-C(24)-C(23)	120.8(4)
C(25)-C(24)-H(24)	119.6
C(23)-C(24)-H(24)	119.6
C(24)-C(25)-C(26)	121.2(4)
C(24)-C(25)-H(25)	119.4
C(26)-C(25)-H(25)	119.4

C(25)-C(26)-C(21)	116.7(4)
C(25)-C(26)-C(30)	120.3(4)
C(21)-C(26)-C(30)	122.9(4)
C(32)-C(30)-C(26)	110.6(4)
C(32)-C(30)-C(31)	111.1(4)
C(26)-C(30)-C(31)	112.7(4)
C(32)-C(30)-H(30)	107.4
C(26)-C(30)-H(30)	107.4
C(31)-C(30)-H(30)	107.4
C(30)-C(31)-H(31A)	109.5
C(30)-C(31)-H(31B)	109.5
H(31A)-C(31)-H(31B)	109.5
C(30)-C(31)-H(31C)	109.5
H(31A)-C(31)-H(31C)	109.5
H(31B)-C(31)-H(31C)	109.5
C(30)-C(32)-H(32A)	109.5
C(30)-C(32)-H(32B)	109.5
H(32A)-C(32)-H(32B)	109.5
C(30)-C(32)-H(32C)	109.5
H(32A)-C(32)-H(32C)	109.5
H(32B)-C(32)-H(32C)	109.5
C(13)-C(12)-N(1)	107.5(4)
C(13)-C(12)-H(12)	126.3
N(1)-C(12)-H(12)	126.3
C(12)-C(13)-N(2)	106.3(4)
C(12)-C(13)-H(13)	126.8
N(2)-C(13)-H(13)	126.8
C(11)-N(2)-C(13)	112.3(3)
C(11)-N(2)-C(41)	120.8(3)
C(13)-N(2)-C(41)	126.8(3)
C(46)-C(41)-C(42)	123.8(4)
C(46)-C(41)-N(2)	118.3(3)
C(42)-C(41)-N(2)	117.9(3)
C(43)-C(42)-C(41)	116.3(4)
C(43)-C(42)-C(47)	121.5(4)
C(41)-C(42)-C(47)	122.2(3)
C(42)-C(47)-C(49)	111.0(4)
C(42)-C(47)-C(48)	111.1(4)
C(49)-C(47)-C(48)	111.9(4)
C(42)-C(47)-H(47)	107.6
C(49)-C(47)-H(47)	107.6
C(48)-C(47)-H(47)	107.6
C(47)-C(48)-H(48A)	109.5
C(47)-C(48)-H(48B)	109.5
H(48A)-C(48)-H(48B)	109.5
C(47)-C(48)-H(48C)	109.5
H(48A)-C(48)-H(48C)	109.5
H(48B)-C(48)-H(48C)	109.5
C(47)-C(49)-H(49A)	109.5
C(47)-C(49)-H(49B)	109.5
H(49A)-C(49)-H(49B)	109.5
C(47)-C(49)-H(49C)	109.5
H(49A)-C(49)-H(49C)	109.5
H(49B)-C(49)-H(49C)	109.5
C(44)-C(43)-C(42)	121.7(4)
C(44)-C(43)-H(43)	119.2

C(42)-C(43)-H(43)	119.2
C(43)-C(44)-C(45)	120.5(4)
C(43)-C(44)-H(44)	119.8
C(45)-C(44)-H(44)	119.8
C(44)-C(45)-C(46)	120.8(4)
C(44)-C(45)-H(45)	119.6
C(46)-C(45)-H(45)	119.6
C(45)-C(46)-C(41)	116.9(4)
C(45)-C(46)-C(50)	121.1(4)
C(41)-C(46)-C(50)	122.0(3)
C(51)-C(50)-C(52)	110.1(4)
C(51)-C(50)-C(46)	111.6(3)
C(52)-C(50)-C(46)	112.8(4)
C(51)-C(50)-H(50)	107.3
C(52)-C(50)-H(50)	107.3
C(46)-C(50)-H(50)	107.3
C(50)-C(51)-H(51A)	109.5
C(50)-C(51)-H(51B)	109.5
H(51A)-C(51)-H(51B)	109.5
C(50)-C(51)-H(51C)	109.5
H(51A)-C(51)-H(51C)	109.5
H(51B)-C(51)-H(51C)	109.5
C(50)-C(52)-H(52A)	109.5
C(50)-C(52)-H(52B)	109.5
H(52A)-C(52)-H(52B)	109.5
C(50)-C(52)-H(52C)	109.5
H(52A)-C(52)-H(52C)	109.5
H(52B)-C(52)-H(52C)	109.5

X-Ray Data for 4b (CCDC 939473)

Compound **4b** crystallizes in the triclinic space group *P-1* with one molecule in the asymmetric unit. The indenyl ligand and the solvent of crystallization (benzene) are both disordered over two sites. An anti-bumping restraint was included to keep the hydrogen atoms (H1T and H6T) on the minor component of the benzene molecule from colliding with the main molecule (H12b and H32C).

Table S30: Crystal data and structure refinement for **4b**

Empirical formula	C42 H51 N2 Ni	
Formula weight	642.56	
Temperature	93(2) K	
Wavelength	1.54187 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 9.2789(2) Å	α= 79.717(6)°.
	b = 12.4450(2) Å	β= 85.508(6)°.
	c = 16.1333(11) Å	γ = 81.988(6)°.
Volume	1812.46(13) $Å^3$	
Z	2	
Density (calculated)	1.177 Mg/m^3	
Absorption coefficient	0.984 mm^{-1}	

F(000)	690
Crystal size	0.05 x 0.05 x 0.03 mm^3
Theta range for data collection	2.79 to 62.39°.
Index ranges	-10<=h<=10, -13<=k<=14, -18<=l<=18
Reflections collected	59772
Independent reflections	5709 [R(int) = 0.1217]
Completeness to theta = 62.39°	99.0 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.9711 and 0.9525
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	5709 / 779 / 551
Goodness-of-fit on F^2	1.119
Final R indices [I>2sigma(I)]	R1 = 0.0670, wR2 = 0.1913
R indices (all data)	R1 = 0.0821, wR2 = 0.2048
Largest diff. peak and hole	1.056 and -0.684 e.$Å^{-3}$

Table S31: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for **4b**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Ni(1)	3666(1)	7289(1)	2052(1)	33(1)
C(1)	5560(16)	7236(17)	1256(10)	37(2)
C(2)	5712(12)	7877(14)	1882(11)	37(2)
C(3)	5795(15)	7153(13)	2680(10)	37(2)
C(4)	5970(20)	7321(18)	3510(10)	47(3)
C(5)	6070(20)	6420(20)	4143(9)	54(3)
C(6)	5990(20)	5349(18)	3989(10)	52(4)
C(7)	5760(20)	5164(13)	3204(12)	48(3)
C(8)	5672(17)	6061(11)	2536(10)	38(2)
C(9)	5480(20)	6134(14)	1661(11)	41(2)
C(1A)	5530(20)	7330(20)	1236(12)	38(3)
C(2A)	5738(15)	7821(18)	1942(15)	38(2)
C(3A)	5838(19)	6966(16)	2668(12)	38(2)
C(4A)	6110(30)	6966(19)	3518(12)	45(3)
C(5A)	6200(20)	5990(20)	4080(10)	48(4)
C(6A)	6010(30)	5011(19)	3809(13)	48(4)
C(7A)	5700(30)	4981(14)	2995(14)	41(3)
C(8A)	5630(20)	5954(13)	2400(12)	34(2)
C(9A)	5380(20)	6206(17)	1512(11)	34(2)
C(11)	1658(3)	7280(3)	1955(2)	27(1)
N(1)	661(3)	8092(2)	2159(2)	29(1)
C(21)	1020(3)	8988(3)	2528(2)	32(1)
C(22)	1470(4)	9908(3)	2002(2)	36(1)
C(27)	1564(4)	10032(3)	1046(2)	37(1)
C(28)	3089(4)	10194(3)	662(3)	48(1)
C(29)	456(5)	10982(3)	668(3)	50(1)
C(23)	1811(5)	10751(3)	2383(3)	50(1)
C(24)	1695(5)	10673(4)	3249(3)	57(1)
C(25)	1254(5)	9755(3)	3757(3)	47(1)
C(26)	901(4)	8890(3)	3407(2)	35(1)
C(30)	435(4)	7871(3)	3968(2)	41(1)
C(31)	1756(5)	7138(4)	4352(3)	54(1)
C(32)	-726(5)	8160(4)	4651(3)	57(1)
C(12)	-824(3)	8029(3)	1939(2)	32(1)
C(13)	-661(3)	6913(3)	1655(2)	31(1)

N(2)	938(3)	6605(2)	1617(2)	28(1)
C(41)	1596(3)	5547(3)	1460(2)	28(1)
C(42)	2046(3)	5424(3)	622(2)	31(1)
C(47)	1770(4)	6368(3)	-100(2)	33(1)
C(48)	3063(5)	6997(4)	-307(3)	54(1)
C(49)	1334(4)	6004(4)	-896(2)	44(1)
C(43)	2760(4)	4404(3)	494(2)	40(1)
C(44)	3022(4)	3549(3)	1154(3)	42(1)
C(45)	2523(4)	3675(3)	1966(2)	40(1)
C(46)	1767(3)	4673(3)	2135(2)	33(1)
C(50)	1099(4)	4749(3)	3012(2)	34(1)
C(51)	2168(4)	4360(3)	3711(3)	45(1)
C(52)	-217(4)	4107(3)	3185(2)	44(1)
C(1S)	4548(9)	9517(8)	6922(6)	118(3)
C(2S)	3330(16)	9352(8)	7478(6)	102(2)
C(3S)	2412(9)	8717(8)	7175(5)	96(2)
C(4S)	2665(9)	8338(9)	6432(6)	104(3)
C(5S)	4033(11)	8381(10)	5972(7)	134(3)
C(6S)	4864(10)	9086(10)	6169(6)	131(3)
C(1T)	2980(50)	10140(20)	6740(20)	127(5)
C(2T)	3250(80)	9320(30)	7440(20)	111(5)
C(3T)	3210(40)	8250(30)	7290(20)	104(4)
C(4T)	3070(60)	8190(30)	6440(20)	117(5)
C(5T)	3750(50)	8910(30)	5800(20)	113(5)
C(6T)	3540(30)	10000(20)	5928(18)	135(6)

Table S32: Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **4b**. 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}
Ni(1)	22(1)	41(1)	36(1)	-9(1)	-2(1)	-8(1)
C(1)	26(4)	48(5)	37(3)	-2(3)	-1(3)	-9(5)
C(2)	26(4)	46(4)	40(4)	-3(3)	-4(5)	-10(4)
C(3)	20(3)	52(4)	39(3)	-3(3)	-4(3)	-10(4)
C(4)	34(5)	69(7)	38(3)	-3(5)	-9(3)	-15(6)
C(5)	39(5)	76(9)	41(4)	3(5)	-15(4)	-4(7)
C(6)	38(5)	68(7)	42(5)	11(6)	-5(5)	2(7)
C(7)	34(4)	53(5)	52(7)	5(4)	-6(5)	-1(4)
C(8)	23(4)	47(3)	39(4)	0(3)	-2(3)	-1(3)
C(9)	36(5)	44(4)	43(4)	-10(4)	7(4)	-3(3)
C(1A)	29(6)	48(5)	36(3)	-3(4)	0(4)	-10(6)
C(2A)	29(4)	47(4)	42(5)	-8(3)	0(6)	-14(5)
C(3A)	23(4)	56(5)	38(3)	-8(3)	-6(4)	-12(5)
C(4A)	33(5)	61(7)	42(4)	-11(5)	-11(4)	-7(6)
C(5A)	31(5)	70(9)	40(5)	-2(6)	-12(4)	1(8)
C(6A)	31(5)	62(7)	45(6)	11(6)	-9(6)	-7(6)
C(7A)	22(5)	54(5)	43(7)	1(4)	-3(5)	-3(4)
C(8A)	17(4)	47(4)	36(5)	-3(3)	-1(4)	-5(3)
C(9A)	22(4)	46(4)	35(4)	-8(4)	-2(4)	-6(3)
C(11)	31(2)	24(2)	26(2)	-6(1)	3(1)	-5(1)
N(1)	24(1)	30(2)	37(2)	-12(1)	-2(1)	-8(1)
C(21)	27(2)	33(2)	37(2)	-11(2)	0(1)	-4(1)
C(22)	41(2)	35(2)	34(2)	-8(2)	1(2)	-12(2)
C(27)	46(2)	34(2)	32(2)	-5(2)	1(2)	-13(2)
C(28)	51(2)	49(3)	42(2)	-6(2)	6(2)	-12(2)

C(29)	54(2)	52(3)	44(2)	-7(2)	-6(2)	-3(2)
C(23)	78(3)	40(2)	38(2)	-8(2)	3(2)	-24(2)
C(24)	87(3)	47(3)	46(3)	-15(2)	3(2)	-34(2)
C(25)	64(2)	46(2)	35(2)	-9(2)	2(2)	-22(2)
C(26)	40(2)	37(2)	31(2)	-8(2)	-1(2)	-10(2)
C(30)	54(2)	40(2)	31(2)	-7(2)	3(2)	-19(2)
C(31)	67(3)	49(3)	44(2)	-2(2)	-3(2)	-7(2)
C(32)	61(3)	56(3)	52(3)	-6(2)	17(2)	-21(2)
C(12)	25(2)	36(2)	38(2)	-10(2)	-4(1)	-8(1)
C(13)	19(2)	35(2)	41(2)	-11(2)	-3(1)	-3(1)
N(2)	21(1)	34(2)	31(2)	-9(1)	0(1)	-4(1)
C(41)	24(2)	32(2)	32(2)	-11(2)	0(1)	-8(1)
C(42)	27(2)	33(2)	36(2)	-10(2)	3(1)	-9(1)
C(47)	33(2)	36(2)	32(2)	-9(2)	-1(1)	-9(2)
C(48)	55(2)	59(3)	49(2)	13(2)	-17(2)	-31(2)
C(49)	43(2)	53(3)	41(2)	-11(2)	-7(2)	-12(2)
C(43)	44(2)	36(2)	41(2)	-10(2)	11(2)	-10(2)
C(44)	43(2)	32(2)	51(2)	-13(2)	10(2)	-6(2)
C(45)	34(2)	37(2)	45(2)	-1(2)	3(2)	-5(2)
C(46)	26(2)	39(2)	34(2)	-7(2)	3(1)	-9(1)
C(50)	34(2)	33(2)	35(2)	-5(2)	0(2)	-8(2)
C(51)	40(2)	50(3)	45(2)	-4(2)	-6(2)	-11(2)
C(52)	37(2)	55(3)	41(2)	-7(2)	5(2)	-13(2)
C(1S)	88(5)	131(6)	150(6)	-57(5)	15(4)	-37(4)
C(2S)	107(5)	69(4)	134(5)	-30(4)	35(4)	-33(4)
C(3S)	87(5)	107(6)	98(5)	-30(4)	26(4)	-28(4)
C(4S)	80(5)	152(6)	87(4)	-27(4)	7(4)	-37(5)
C(5S)	108(6)	204(8)	112(6)	-73(6)	29(5)	-61(6)
C(6S)	85(5)	196(8)	129(6)	-56(6)	24(4)	-53(5)
C(1T)	112(11)	127(8)	143(9)	-33(7)	30(10)	-29(10)
C(2T)	94(9)	109(8)	134(8)	-39(7)	40(9)	-32(9)
C(3T)	96(10)	121(7)	107(7)	-45(8)	25(9)	-42(9)
C(4T)	97(10)	161(9)	104(8)	-41(7)	18(9)	-39(9)
C(5T)	81(9)	176(10)	99(7)	-46(8)	7(8)	-55(9)
C(6T)	113(12)	169(9)	132(8)	-34(10)	20(11)	-47(11)

Table S33: Bond lengths (Å) for **4b**

Ni(1)-C(11)	1.883(3)
Ni(1)-C(1A)	2.094(12)
Ni(1)-C(1)	2.095(10)
Ni(1)-C(2A)	2.104(10)
Ni(1)-C(2)	2.110(8)
Ni(1)-C(9A)	2.168(11)
Ni(1)-C(9)	2.186(10)
Ni(1)-C(3)	2.263(10)
Ni(1)-C(3A)	2.271(12)
Ni(1)-C(8A)	2.321(12)
Ni(1)-C(8)	2.328(10)
C(1)-C(9)	1.420(10)
C(1)-C(2)	1.420(10)
C(1)-H(1)	0.9500
C(2)-C(3)	1.435(9)
C(2)-H(2)	0.9500
C(3)-C(4)	1.416(9)
C(3)-C(8)	1.441(10)

C(4)-C(5)	1.377(10)
C(4)-H(4)	0.9500
C(5)-C(6)	1.406(12)
C(5)-H(5)	0.9500
C(6)-C(7)	1.366(11)
C(6)-H(6)	0.9500
C(7)-C(8)	1.404(9)
C(7)-H(7)	0.9500
C(8)-C(9)	1.422(10)
C(9)-H(9)	0.9500
C(1A)-C(9A)	1.418(12)
C(1A)-C(2A)	1.418(12)
C(1A)-H(1A)	0.9500
C(2A)-C(3A)	1.434(11)
C(2A)-H(2A)	0.9500
C(3A)-C(4A)	1.413(11)
C(3A)-C(8A)	1.443(11)
C(4A)-C(5A)	1.372(12)
C(4A)-H(4A)	0.9500
C(5A)-C(6A)	1.410(14)
C(5A)-H(5A)	0.9500
C(6A)-C(7A)	1.374(12)
C(6A)-H(6A)	0.9500
C(7A)-C(8A)	1.401(10)
C(7A)-H(7A)	0.9500
C(8A)-C(9A)	1.442(10)
C(9A)-H(9A)	0.9500
C(11)-N(1)	1.340(4)
C(11)-N(2)	1.352(4)
N(1)-C(21)	1.443(4)
N(1)-C(12)	1.466(4)
C(21)-C(22)	1.392(5)
C(21)-C(26)	1.398(5)
C(22)-C(23)	1.390(5)
C(22)-C(27)	1.520(5)
C(27)-C(28)	1.524(5)
C(27)-C(29)	1.526(5)
C(27)-H(27)	1.0000
C(28)-H(28A)	0.9800
C(28)-H(28B)	0.9800
C(28)-H(28C)	0.9800
C(29)-H(29A)	0.9800
C(29)-H(29B)	0.9800
C(29)-H(29C)	0.9800
C(23)-C(24)	1.381(6)
C(23)-H(23)	0.9500
C(24)-C(25)	1.375(6)
C(24)-H(24)	0.9500
C(25)-C(26)	1.388(5)
C(25)-H(25)	0.9500
C(26)-C(30)	1.515(5)
C(30)-C(31)	1.524(6)
C(30)-C(32)	1.534(6)
C(30)-H(30)	1.0000
C(31)-H(31A)	0.9800
C(31)-H(31B)	0.9800

C(31)-H(31C)	0.9800
C(32)-H(32A)	0.9800
C(32)-H(32B)	0.9800
C(32)-H(32C)	0.9800
C(12)-C(13)	1.522(5)
C(12)-H(12A)	0.9900
C(12)-H(12B)	0.9900
C(13)-N(2)	1.479(4)
C(13)-H(13A)	0.9900
C(13)-H(13B)	0.9900
N(2)-C(41)	1.428(4)
C(41)-C(46)	1.396(5)
C(41)-C(42)	1.413(5)
C(42)-C(43)	1.389(5)
C(42)-C(47)	1.509(5)
C(47)-C(48)	1.507(5)
C(47)-C(49)	1.534(5)
C(47)-H(47)	1.0000
C(48)-H(48A)	0.9800
C(48)-H(48B)	0.9800
C(48)-H(48C)	0.9800
C(49)-H(49A)	0.9800
C(49)-H(49B)	0.9800
C(49)-H(49C)	0.9800
C(43)-C(44)	1.374(5)
C(43)-H(43)	0.9500
C(44)-C(45)	1.382(5)
C(44)-H(44)	0.9500
C(45)-C(46)	1.400(5)
C(45)-H(45)	0.9500
C(46)-C(50)	1.513(5)
C(50)-C(51)	1.528(5)
C(50)-C(52)	1.532(5)
C(50)-H(50)	1.0000
C(51)-H(51A)	0.9800
C(51)-H(51B)	0.9800
C(51)-H(51C)	0.9800
C(52)-H(52A)	0.9800
C(52)-H(52B)	0.9800
C(52)-H(52C)	0.9800
C(1S)-C(2S)	1.402(12)
C(1S)-C(6S)	1.408(9)
C(1S)-H(1S)	0.9500
C(2S)-C(3S)	1.408(10)
C(2S)-H(2S)	0.9500
C(3S)-C(4S)	1.356(9)
C(3S)-H(3S)	0.9500
C(4S)-C(5S)	1.422(10)
C(4S)-H(4S)	0.9500
C(5S)-C(6S)	1.338(10)
C(5S)-H(5S)	0.9500
C(6S)-H(6S)	0.9500
C(1T)-C(2T)	1.392(15)
C(1T)-C(6T)	1.399(14)
C(1T)-H(1T)	0.9500
C(2T)-C(3T)	1.405(15)

C(2T)-H(2T)	0.9500
C(3T)-C(4T)	1.405(15)
C(3T)-H(3T)	0.9500
C(4T)-C(5T)	1.413(15)
C(4T)-H(4T)	0.9500
C(5T)-C(6T)	1.390(15)
C(5T)-H(5T)	0.9500
C(6T)-H(6T)	0.9500

Table S34: Bond angles (°) for **4b**

C(11)-Ni(1)-C(1A)	136.7(7)
C(11)-Ni(1)-C(1)	137.0(6)
C(1A)-Ni(1)-C(1)	3.2(12)
C(11)-Ni(1)-C(2A)	161.2(5)
C(1A)-Ni(1)-C(2A)	39.5(4)
C(1)-Ni(1)-C(2A)	40.7(13)
C(11)-Ni(1)-C(2)	158.5(4)
C(1A)-Ni(1)-C(2)	38.1(13)
C(1)-Ni(1)-C(2)	39.5(3)
C(2A)-Ni(1)-C(2)	3.0(9)
C(11)-Ni(1)-C(9A)	124.5(6)
C(1A)-Ni(1)-C(9A)	38.8(4)
C(1)-Ni(1)-C(9A)	35.6(11)
C(2A)-Ni(1)-C(9A)	65.5(5)
C(2)-Ni(1)-C(9A)	66.0(12)
C(11)-Ni(1)-C(9)	128.0(6)
C(1A)-Ni(1)-C(9)	41.9(11)
C(1)-Ni(1)-C(9)	38.7(3)
C(2A)-Ni(1)-C(9)	64.0(12)
C(2)-Ni(1)-C(9)	64.8(5)
C(9A)-Ni(1)-C(9)	6.9(7)
C(11)-Ni(1)-C(3)	158.6(5)
C(1A)-Ni(1)-C(3)	64.5(10)
C(1)-Ni(1)-C(3)	63.8(5)
C(2A)-Ni(1)-C(3)	35.1(8)
C(2)-Ni(1)-C(3)	38.1(3)
C(9A)-Ni(1)-C(3)	67.9(9)
C(9)-Ni(1)-C(3)	62.4(4)
C(11)-Ni(1)-C(3A)	158.0(6)
C(1A)-Ni(1)-C(3A)	63.6(6)
C(1)-Ni(1)-C(3A)	62.6(10)
C(2A)-Ni(1)-C(3A)	38.0(3)
C(2)-Ni(1)-C(3A)	41.0(8)
C(9A)-Ni(1)-C(3A)	63.2(5)
C(9)-Ni(1)-C(3A)	57.5(9)
C(3)-Ni(1)-C(3A)	5.9(8)
C(11)-Ni(1)-C(8A)	135.2(5)
C(1A)-Ni(1)-C(8A)	62.7(5)
C(1)-Ni(1)-C(8A)	59.9(9)
C(2A)-Ni(1)-C(8A)	63.1(5)
C(2)-Ni(1)-C(8A)	65.4(9)
C(9A)-Ni(1)-C(8A)	37.3(3)
C(9)-Ni(1)-C(8A)	30.4(7)
C(3)-Ni(1)-C(8A)	42.5(7)
C(3A)-Ni(1)-C(8A)	36.6(3)

C(11)-Ni(1)-C(8)	138.4(5)
C(1A)-Ni(1)-C(8)	65.2(9)
C(1)-Ni(1)-C(8)	62.7(4)
C(2A)-Ni(1)-C(8)	60.3(9)
C(2)-Ni(1)-C(8)	62.9(4)
C(9A)-Ni(1)-C(8)	43.4(7)
C(9)-Ni(1)-C(8)	36.5(3)
C(3)-Ni(1)-C(8)	36.5(3)
C(3A)-Ni(1)-C(8)	30.6(8)
C(8A)-Ni(1)-C(8)	7.0(7)
C(9)-C(1)-C(2)	108.3(9)
C(9)-C(1)-Ni(1)	74.1(6)
C(2)-C(1)-Ni(1)	70.8(5)
C(9)-C(1)-H(1)	125.9
C(2)-C(1)-H(1)	125.9
Ni(1)-C(1)-H(1)	120.9
C(1)-C(2)-C(3)	107.9(9)
C(1)-C(2)-Ni(1)	69.7(5)
C(3)-C(2)-Ni(1)	76.7(5)
C(1)-C(2)-H(2)	126.1
C(3)-C(2)-H(2)	126.1
Ni(1)-C(2)-H(2)	119.3
C(4)-C(3)-C(2)	133.1(9)
C(4)-C(3)-C(8)	119.2(8)
C(2)-C(3)-C(8)	107.7(8)
C(4)-C(3)-Ni(1)	126.6(11)
C(2)-C(3)-Ni(1)	65.2(5)
C(8)-C(3)-Ni(1)	74.2(5)
C(5)-C(4)-C(3)	118.0(9)
C(5)-C(4)-H(4)	121.0
C(3)-C(4)-H(4)	121.0
C(4)-C(5)-C(6)	122.2(9)
C(4)-C(5)-H(5)	118.9
C(6)-C(5)-H(5)	118.9
C(7)-C(6)-C(5)	121.3(8)
C(7)-C(6)-H(6)	119.3
C(5)-C(6)-H(6)	119.3
C(6)-C(7)-C(8)	118.4(9)
C(6)-C(7)-H(7)	120.8
C(8)-C(7)-H(7)	120.8
C(7)-C(8)-C(9)	131.9(9)
C(7)-C(8)-C(3)	120.7(8)
C(9)-C(8)-C(3)	107.4(7)
C(7)-C(8)-Ni(1)	129.9(12)
C(9)-C(8)-Ni(1)	66.2(5)
C(3)-C(8)-Ni(1)	69.3(5)
C(1)-C(9)-C(8)	108.7(9)
C(1)-C(9)-Ni(1)	67.2(5)
C(8)-C(9)-Ni(1)	77.2(6)
C(1)-C(9)-H(9)	125.7
C(8)-C(9)-H(9)	125.7
Ni(1)-C(9)-H(9)	121.5
C(9A)-C(1A)-C(2A)	109.3(11)
C(9A)-C(1A)-Ni(1)	73.4(7)
C(2A)-C(1A)-Ni(1)	70.6(6)
C(9A)-C(1A)-H(1A)	125.3

C(2A)-C(1A)-H(1A)	125.3
Ni(1)-C(1A)-H(1A)	122.2
C(1A)-C(2A)-C(3A)	107.8(11)
C(1A)-C(2A)-Ni(1)	69.9(6)
C(3A)-C(2A)-Ni(1)	77.3(7)
C(1A)-C(2A)-H(2A)	126.1
C(3A)-C(2A)-H(2A)	126.1
Ni(1)-C(2A)-H(2A)	118.6
C(4A)-C(3A)-C(2A)	132.4(11)
C(4A)-C(3A)-C(8A)	120.0(10)
C(2A)-C(3A)-C(8A)	107.6(9)
C(4A)-C(3A)-Ni(1)	128.3(14)
C(2A)-C(3A)-Ni(1)	64.7(6)
C(8A)-C(3A)-Ni(1)	73.6(6)
C(5A)-C(4A)-C(3A)	119.6(11)
C(5A)-C(4A)-H(4A)	120.2
C(3A)-C(4A)-H(4A)	120.2
C(4A)-C(5A)-C(6A)	119.8(10)
C(4A)-C(5A)-H(5A)	120.1
C(6A)-C(5A)-H(5A)	120.1
C(7A)-C(6A)-C(5A)	122.5(10)
C(7A)-C(6A)-H(6A)	118.8
C(5A)-C(6A)-H(6A)	118.8
C(6A)-C(7A)-C(8A)	119.1(11)
C(6A)-C(7A)-H(7A)	120.5
C(8A)-C(7A)-H(7A)	120.5
C(7A)-C(8A)-C(9A)	133.4(10)
C(7A)-C(8A)-C(3A)	119.0(9)
C(9A)-C(8A)-C(3A)	107.6(8)
C(7A)-C(8A)-Ni(1)	129.8(14)
C(9A)-C(8A)-Ni(1)	65.6(6)
C(3A)-C(8A)-Ni(1)	69.8(6)
C(1A)-C(9A)-C(8A)	107.4(10)
C(1A)-C(9A)-Ni(1)	67.8(6)
C(8A)-C(9A)-Ni(1)	77.1(7)
C(1A)-C(9A)-H(9A)	126.3
C(8A)-C(9A)-H(9A)	126.3
Ni(1)-C(9A)-H(9A)	120.5
N(1)-C(11)-N(2)	106.7(3)
N(1)-C(11)-Ni(1)	121.9(2)
N(2)-C(11)-Ni(1)	131.0(2)
C(11)-N(1)-C(21)	123.1(3)
C(11)-N(1)-C(12)	114.6(3)
C(21)-N(1)-C(12)	122.3(3)
C(22)-C(21)-C(26)	122.8(3)
C(22)-C(21)-N(1)	119.1(3)
C(26)-C(21)-N(1)	118.1(3)
C(23)-C(22)-C(21)	117.3(3)
C(23)-C(22)-C(27)	119.6(3)
C(21)-C(22)-C(27)	123.0(3)
C(22)-C(27)-C(28)	112.7(3)
C(22)-C(27)-C(29)	110.2(3)
C(28)-C(27)-C(29)	110.3(3)
C(22)-C(27)-H(27)	107.8
C(28)-C(27)-H(27)	107.8
C(29)-C(27)-H(27)	107.8

C(27)-C(28)-H(28A)	109.5
C(27)-C(28)-H(28B)	109.5
H(28A)-C(28)-H(28B)	109.5
C(27)-C(28)-H(28C)	109.5
H(28A)-C(28)-H(28C)	109.5
H(28B)-C(28)-H(28C)	109.5
C(27)-C(29)-H(29A)	109.5
C(27)-C(29)-H(29B)	109.5
H(29A)-C(29)-H(29B)	109.5
C(27)-C(29)-H(29C)	109.5
H(29A)-C(29)-H(29C)	109.5
H(29B)-C(29)-H(29C)	109.5
C(24)-C(23)-C(22)	120.9(4)
C(24)-C(23)-H(23)	119.6
C(22)-C(23)-H(23)	119.6
C(25)-C(24)-C(23)	120.7(4)
C(25)-C(24)-H(24)	119.6
C(23)-C(24)-H(24)	119.6
C(24)-C(25)-C(26)	120.6(4)
C(24)-C(25)-H(25)	119.7
C(26)-C(25)-H(25)	119.7
C(25)-C(26)-C(21)	117.7(3)
C(25)-C(26)-C(30)	120.5(3)
C(21)-C(26)-C(30)	121.8(3)
C(26)-C(30)-C(31)	110.4(3)
C(26)-C(30)-C(32)	112.0(3)
C(31)-C(30)-C(32)	111.4(3)
C(26)-C(30)-H(30)	107.6
C(31)-C(30)-H(30)	107.6
C(32)-C(30)-H(30)	107.6
C(30)-C(31)-H(31A)	109.5
C(30)-C(31)-H(31B)	109.5
H(31A)-C(31)-H(31B)	109.5
C(30)-C(31)-H(31C)	109.5
H(31A)-C(31)-H(31C)	109.5
H(31B)-C(31)-H(31C)	109.5
C(30)-C(32)-H(32A)	109.5
C(30)-C(32)-H(32B)	109.5
H(32A)-C(32)-H(32B)	109.5
C(30)-C(32)-H(32C)	109.5
H(32A)-C(32)-H(32C)	109.5
H(32B)-C(32)-H(32C)	109.5
N(1)-C(12)-C(13)	102.1(2)
N(1)-C(12)-H(12A)	111.3
C(13)-C(12)-H(12A)	111.3
N(1)-C(12)-H(12B)	111.3
C(13)-C(12)-H(12B)	111.3
H(12A)-C(12)-H(12B)	109.2
N(2)-C(13)-C(12)	102.4(2)
N(2)-C(13)-H(13A)	111.3
C(12)-C(13)-H(13A)	111.3
N(2)-C(13)-H(13B)	111.3
C(12)-C(13)-H(13B)	111.3
H(13A)-C(13)-H(13B)	109.2
C(11)-N(2)-C(41)	122.6(3)
C(11)-N(2)-C(13)	113.2(3)

C(41)-N(2)-C(13)	122.0(3)
C(46)-C(41)-C(42)	122.3(3)
C(46)-C(41)-N(2)	119.2(3)
C(42)-C(41)-N(2)	118.4(3)
C(43)-C(42)-C(41)	117.2(3)
C(43)-C(42)-C(47)	121.7(3)
C(41)-C(42)-C(47)	121.1(3)
C(48)-C(47)-C(42)	111.5(3)
C(48)-C(47)-C(49)	110.1(3)
C(42)-C(47)-C(49)	113.0(3)
C(48)-C(47)-H(47)	107.3
C(42)-C(47)-H(47)	107.3
C(49)-C(47)-H(47)	107.3
C(47)-C(48)-H(48A)	109.5
C(47)-C(48)-H(48B)	109.5
H(48A)-C(48)-H(48B)	109.5
C(47)-C(48)-H(48C)	109.5
H(48A)-C(48)-H(48C)	109.5
H(48B)-C(48)-H(48C)	109.5
C(47)-C(49)-H(49A)	109.5
C(47)-C(49)-H(49B)	109.5
H(49A)-C(49)-H(49B)	109.5
C(47)-C(49)-H(49C)	109.5
H(49A)-C(49)-H(49C)	109.5
H(49B)-C(49)-H(49C)	109.5
C(44)-C(43)-C(42)	121.6(3)
C(44)-C(43)-H(43)	119.2
C(42)-C(43)-H(43)	119.2
C(43)-C(44)-C(45)	120.2(3)
C(43)-C(44)-H(44)	119.9
C(45)-C(44)-H(44)	119.9
C(44)-C(45)-C(46)	121.0(3)
C(44)-C(45)-H(45)	119.5
C(46)-C(45)-H(45)	119.5
C(41)-C(46)-C(45)	117.4(3)
C(41)-C(46)-C(50)	122.8(3)
C(45)-C(46)-C(50)	119.7(3)
C(46)-C(50)-C(51)	114.0(3)
C(46)-C(50)-C(52)	109.7(3)
C(51)-C(50)-C(52)	110.2(3)
C(46)-C(50)-H(50)	107.5
C(51)-C(50)-H(50)	107.5
C(52)-C(50)-H(50)	107.5
C(50)-C(51)-H(51A)	109.5
C(50)-C(51)-H(51B)	109.5
H(51A)-C(51)-H(51B)	109.5
C(50)-C(51)-H(51C)	109.5
H(51A)-C(51)-H(51C)	109.5
H(51B)-C(51)-H(51C)	109.5
C(50)-C(52)-H(52A)	109.5
C(50)-C(52)-H(52B)	109.5
H(52A)-C(52)-H(52B)	109.5
C(50)-C(52)-H(52C)	109.5
H(52A)-C(52)-H(52C)	109.5
H(52B)-C(52)-H(52C)	109.5
C(2S)-C(1S)-C(6S)	125.5(8)

C(2S)-C(1S)-H(1S)	117.2
C(6S)-C(1S)-H(1S)	117.2
C(1S)-C(2S)-C(3S)	111.1(8)
C(1S)-C(2S)-H(2S)	124.4
C(3S)-C(2S)-H(2S)	124.4
C(4S)-C(3S)-C(2S)	124.1(8)
C(4S)-C(3S)-H(3S)	118.0
C(2S)-C(3S)-H(3S)	118.0
C(3S)-C(4S)-C(5S)	121.2(8)
C(3S)-C(4S)-H(4S)	119.4
C(5S)-C(4S)-H(4S)	119.4
C(6S)-C(5S)-C(4S)	115.7(8)
C(6S)-C(5S)-H(5S)	122.2
C(4S)-C(5S)-H(5S)	122.2
C(5S)-C(6S)-C(1S)	119.9(8)
C(5S)-C(6S)-H(6S)	120.0
C(1S)-C(6S)-H(6S)	120.0
C(2T)-C(1T)-C(6T)	122(2)
C(2T)-C(1T)-H(1T)	119.1
C(6T)-C(1T)-H(1T)	119.1
C(1T)-C(2T)-C(3T)	114.4(19)
C(1T)-C(2T)-H(2T)	122.8
C(3T)-C(2T)-H(2T)	122.8
C(4T)-C(3T)-C(2T)	113.6(18)
C(4T)-C(3T)-H(3T)	123.2
C(2T)-C(3T)-H(3T)	123.2
C(3T)-C(4T)-C(5T)	119(2)
C(3T)-C(4T)-H(4T)	120.3
C(5T)-C(4T)-H(4T)	120.3
C(6T)-C(5T)-C(4T)	114(2)
C(6T)-C(5T)-H(5T)	123.1
C(4T)-C(5T)-H(5T)	123.1
C(5T)-C(6T)-C(1T)	114.0(17)
C(5T)-C(6T)-H(6T)	123.0
C(1T)-C(6T)-H(6T)	123.0

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