

## Supporting Information

### Theoretical Analysis of the Sequential Proton-Coupled Electron Transfer Mechanisms for H<sub>2</sub> Oxidation and Production Pathways Catalyzed by Nickel Molecular Electrocatalysts

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## **I. Computational methods for calculation of solvent reorganization energies**

### **A. Charge distributions**

To obtain the charge distributions of the (Me,Me)<sub>bb</sub> system, we performed the following steps. First, we calculated the electrostatic potential of the solute in solution (acetonitrile) and then fit that potential to a sum of atomic charges that also reproduced the molecular dipole moment using electronic structure methods.<sup>1</sup> To obtain the initial charge distribution, we used the Merz-Singh-Kollman scheme.<sup>2,3</sup> Then we performed a Natural Bond Orbital (NBO) analysis<sup>4-9</sup> on the species of interest to obtain a more realistic value for the atomic charge on the Ni center, as atomic charge models in standard electronic structure methods tend to assign a surplus of electron density on Ni. Using the NBO charge on Ni and the calculated electrostatic potential, we fixed the Ni charge and recalculated the partial charges on the other atoms using the Restrained ElectroStatic Potential fit (RESP) program,<sup>10-12</sup> which is part of the AMBER molecular dynamics package.<sup>13</sup>

### **B. Calculation of $\lambda_s$ for proton transfer**

To obtain an estimate of the reorganization energies for the proton transfer steps, we utilized the charge distributions described above and the Frequency Resolved Cavity Model (FRCM) for advanced solvation calculations, the details of which are discussed elsewhere.<sup>14-16</sup> As reorganization energies can be somewhat geometry dependent, we determined  $\lambda_s$  using all four relevant geometries: (1a), (1b), (2a), and (2b). As can be seen from the results that are presented in Table S1, the solvent reorganization energy for proton transfer is approximately 0.1 eV, which is consistent with other systems previously studied in our group.<sup>17</sup>

### C. Calculation of $\lambda_s$ for electrochemical electron transfer

The calculation of solvent reorganization energies for electrochemical electron transfer using the model of Liu and Newton<sup>18</sup> discussed in the main text involves a number of approximations, such as a single point charge instead of a charge distribution and a spherical cavity instead of a molecule shaped cavity. To determine the effects of these approximations on  $\lambda_s$ , we extended the model of Liu and Newton<sup>18</sup> to include a point charge within a cylindrical cavity as well as the charge distributions described above in a spherical cavity. For the cavity containing the charge distribution, we averaged over the polarization potential on the surface of the spherical cavity. In general, the species studied here are relatively spherical, so we would expect similar results for the two different cavity models. To ensure a relevant comparison, we chose a radius  $r$  of 8.65 Å, which is the approximate radius of a sphere needed to give a cavity that would encompass the atomic charges when the van der Waals radii were included.

The parameters of the cylindrical cavity were chosen such that they reproduced the volume of the sphere with  $r = 8.65$  Å. Note that the results presented in Table S2 are slightly smaller than those given in the main text. This difference arises from the fact that the radius used to generate the data in Table S2 is larger than the radius that reproduces the C-PCM cavity volume used to generate the data in the main text. The value of  $\lambda_s$  is slightly smaller for the cylindrical cavity than for the spherical cavity because the averaged polarization potential is divided by the surface area, and a sphere has a smaller surface area. However, all values for  $\lambda_s$  are on the order of 0.2 eV. These calculations indicate that the representation of the charge distribution by a single point charge does not significantly affect the solvent reorganization energy.

## Tables

Table S1: Solvent reorganization energies<sup>a</sup> ( $\lambda_s$ ) for proton transfer in (Me,Me)<sub>bb</sub>.

<b>Geometry</b>	<b>PT pair</b>	<b><math>\lambda_s</math></b>
(1a)	(1a) ↔ (1b)	0.10
(1a)	(2a) ↔ (2b)	0.08
(1b)	(1a) ↔ (1b)	0.11
(1b)	(2a) ↔ (2b)	0.09
(2a)	(1a) ↔ (1b)	0.10
(2a)	(2a) ↔ (2b)	0.08
(2b)	(1a) ↔ (1b)	0.13
(2b)	(2a) ↔ (2b)	0.10

<sup>a</sup>Units of eV.

Table S2: Solvent reorganization energies<sup>a</sup> ( $\lambda_s$ ) for electrochemical electron transfer in (Me<sub>3</sub>Me)<sub>bb</sub>.

<b>Method</b>	<b><math>\lambda_s</math></b>
point charge in a sphere	0.22
point charge in a cylinder	0.20
charge distribution in a sphere	0.22
model of Liu and Newton <sup>b</sup>	0.22

<sup>a</sup>Units of eV, calculated with  $\epsilon_\infty = 1.806874$  and  $\epsilon_0 = 35.688000$  for acetonitrile. <sup>b</sup>Ref. 16.

Table S3: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{II}}(\text{dmpp})_2]^{2+}$  ( $-2094.87213858 E_h$ ).

atom	x	y	z
C	3.454782	0.434570	1.184639
C	3.215845	1.511535	0.126736
H	2.797094	0.608130	2.045762
H	4.475341	0.549779	1.564238
H	3.197813	2.500260	0.598729
H	4.037525	1.527970	-0.600754
P	1.515400	-1.527776	0.527202
P	1.649418	1.341095	-0.834065
C	1.357230	3.040624	-1.451264
H	2.218838	3.342945	-2.056546
H	0.465420	3.076942	-2.083108
H	1.250391	3.754117	-0.633066
C	2.098651	0.482052	-2.397951
H	2.406380	-0.551699	-2.227378
H	1.242090	0.480667	-3.078937
H	2.921231	1.015948	-2.886695
C	1.685314	-3.185602	-0.235278
H	2.466693	-3.742294	0.293203
H	0.757560	-3.758365	-0.166421
H	1.974090	-3.104510	-1.287131
C	1.083919	-1.879610	2.277260
H	1.772931	-2.628563	2.682927
H	1.165889	-0.971679	2.880992
H	0.063891	-2.259568	2.365248
Ni	0.002381	0.041125	-0.078461
C	-3.153864	1.670313	0.002788
C	-3.893912	0.338403	-0.096463
H	-3.104456	2.152184	-0.982056
H	-3.699169	2.357940	0.661229
C	-3.257113	-0.632431	-1.087783
H	-3.997952	-0.121835	0.893860
H	-4.916112	0.546104	-0.430583
H	-3.940995	-1.463674	-1.299357
H	-3.076157	-0.127189	-2.045385
P	-1.660896	-1.400503	-0.574904
P	-1.438890	1.575188	0.658984
C	-1.199908	-2.273470	-2.120879

H	-0.345050	-2.934879	-1.976101
H	-2.049487	-2.872068	-2.467541
H	-0.951395	-1.546321	-2.899357
C	-2.178789	-2.726016	0.579910
H	-2.951008	-3.342408	0.107511
H	-1.340511	-3.374503	0.845879
H	-2.591839	-2.302667	1.500178
C	-1.638668	1.043946	2.407024
H	-2.380092	1.681917	2.901301
H	-1.967410	0.004715	2.479787
H	-0.689914	1.145618	2.941744
C	-1.008592	3.344606	0.846188
H	-0.029416	3.461601	1.317677
H	-1.012683	3.858195	-0.117546
H	-1.756450	3.819866	1.490078
C	3.279333	-0.991625	0.662811
H	3.770937	-1.118142	-0.309415
H	3.754706	-1.709588	1.342414

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Table S4: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{dmpp})_2]^+$  ( $-2095.20426661 E_h$ ).

atom	x	y	z
C	-3.366086	-0.573209	-1.051027
C	-3.199707	0.946776	-0.929644
H	-2.638756	-0.971154	-1.771790
H	-4.354078	-0.790144	-1.473961
H	-3.716481	1.458016	-1.751181
H	-3.661471	1.309008	-0.001881
P	-1.666158	-1.065172	1.224480
P	-1.423692	1.487152	-0.946763
C	-1.091969	1.591310	-2.757449
H	-1.800655	2.262478	-3.255241
H	-0.076578	1.960588	-2.929394
H	-1.172809	0.598545	-3.209805
C	-1.586965	3.269451	-0.511686
H	-1.829250	3.380931	0.549248
H	-0.641797	3.786726	-0.699783
H	-2.370658	3.752211	-1.105751
C	-2.236003	-0.058710	2.662031
H	-3.086056	-0.527653	3.169785
H	-1.417522	0.053875	3.379535
H	-2.531468	0.940848	2.330085
C	-1.428529	-2.707589	2.023866
H	-2.323546	-3.019985	2.572684
H	-1.203286	-3.464302	1.266436
H	-0.586980	-2.662535	2.721605
Ni	-0.009067	0.041472	0.117317
C	2.829593	-0.788429	-1.654871
C	3.748608	-0.109013	-0.632568
H	2.540681	-0.066589	-2.430307
H	3.376587	-1.593403	-2.162500
C	3.263577	1.246217	-0.106415
H	3.953174	-0.788621	0.205058
H	4.720018	0.053914	-1.114575
H	4.071682	1.730737	0.456883
H	3.030056	1.906672	-0.952149
P	1.759334	1.201367	0.975296
P	1.266410	-1.503846	-0.963369
C	1.628534	2.972107	1.462619



H	0.812265	3.100974	2.179484
H	2.557640	3.330758	1.918887
H	1.410690	3.588177	0.585612
C	2.416336	0.455704	2.527774
H	3.325601	0.966310	2.864113
H	1.661170	0.530800	3.316018
H	2.639435	-0.604540	2.380164
C	1.896618	-2.983715	-0.063397
H	2.545848	-3.592890	-0.701958
H	2.458253	-2.683533	0.825690
H	1.055540	-3.599067	0.268435
C	0.535942	-2.287364	-2.461751
H	-0.358057	-2.854921	-2.186051
H	0.241343	-1.516466	-3.179791
H	1.245836	-2.965894	-2.947563
C	-3.243621	-1.340991	0.268527
H	-4.089814	-1.099046	0.924629
H	-3.310193	-2.415872	0.062260

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Table S5: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{dmpp})_2]$  ( $-2095.38042745 E_h$ ).

atom	x	y	z
C	3.278981	0.788507	0.932807
C	3.019029	1.448467	-0.424315
H	2.574079	1.199053	1.668813
H	4.281584	1.083576	1.271258
H	3.006685	2.537947	-0.292661
H	3.849731	1.226914	-1.108965
P	1.398404	-1.312655	1.105044
P	1.403831	0.964298	-1.260064
C	1.023214	2.573730	-2.105926
H	1.879728	2.969916	-2.666138
H	0.186675	2.428032	-2.797206
H	0.715560	3.314884	-1.361453
C	2.084705	0.058111	-2.733094
H	2.508365	-0.896885	-2.406671
H	1.272166	-0.163994	-3.431655
H	2.859018	0.630885	-3.259486
C	1.643107	-3.120635	0.760558
H	2.485903	-3.545128	1.321024
H	0.733737	-3.667954	1.029717
H	1.818189	-3.266875	-0.310140
C	1.315542	-1.405376	2.959285
H	2.108349	-2.034280	3.384096
H	1.403317	-0.397619	3.378293
H	0.343057	-1.807744	3.259676
Ni	0.006684	-0.095112	-0.009454
C	-2.873537	1.617979	0.420205
C	-3.671932	0.413584	-0.091105
H	-2.665708	2.297286	-0.417825
H	-3.476657	2.181034	1.146772
C	-3.104002	-0.250366	-1.350167
H	-3.776585	-0.332497	0.709748
H	-4.694534	0.748700	-0.312742
H	-3.851418	-0.934303	-1.777242
H	-2.908584	0.521279	-2.107699
P	-1.505037	-1.177187	-1.099997
P	-1.231680	1.191152	1.192832
C	-1.273284	-1.762868	-2.847047

H	-0.420471	-2.447589	-2.892136
H	-2.161056	-2.275421	-3.239224
H	-1.049596	-0.906563	-3.491474
C	-2.179620	-2.750884	-0.379726
H	-3.012375	-3.162308	-0.964816
H	-1.378208	-3.495258	-0.335209
H	-2.517686	-2.569174	0.645259
C	-1.831860	0.629800	2.859453
H	-2.524355	1.346893	3.318787
H	-2.333827	-0.338166	2.763367
H	-0.975004	0.490731	3.525624
C	-0.703921	2.906773	1.666902
H	0.199586	2.850139	2.282784
H	-0.456892	3.474232	0.764046
H	-1.478756	3.448891	2.224218
C	3.169121	-0.741611	0.927068
H	3.565017	-1.144340	-0.015417
H	3.773584	-1.178632	1.733295

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Table S6: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{II}}(\text{dmpp})_2]^+$  ( $-2095.81643501 E_h$ ).

atom	x	y	z
C	-1.687222	2.883192	-0.581116
C	-2.422273	1.614641	-1.032132
H	-1.233246	2.732696	0.407803
H	-2.436515	3.671124	-0.440463
H	-3.356083	1.503176	-0.466020
H	-2.706786	1.711235	-2.088383
P	0.963592	2.485518	-1.635884
P	-1.459779	0.040061	-0.855296
C	-1.718190	-0.339698	0.928804
H	-2.781853	-0.325251	1.190679
H	-1.317930	-1.331562	1.159249
H	-1.187159	0.388097	1.548590
C	-2.634421	-1.170458	-1.604832
H	-2.279346	-2.189832	-1.423754
H	-3.635025	-1.071016	-1.169403
H	-2.714229	-1.025025	-2.685645
C	1.748338	3.296779	-3.093695
H	1.731528	4.386964	-2.985337
H	2.789285	2.973544	-3.186989
H	1.218548	3.035093	-4.014493
C	1.912139	3.270916	-0.269419
H	1.979957	4.356518	-0.398550
H	1.430424	3.053657	0.688228
H	2.922999	2.853658	-0.238036
Ni	0.694599	0.297994	-1.456912
C	1.831395	-2.923805	-1.474161
C	1.839241	-2.983274	-3.004118
H	0.869519	-3.295009	-1.096139
H	2.605748	-3.591880	-1.075709
C	0.688579	-2.239832	-3.693096
H	2.802302	-2.626091	-3.390825
H	1.781459	-4.037430	-3.300023
H	0.678228	-2.489690	-4.761549
H	-0.272653	-2.567913	-3.277839
P	0.758264	-0.399986	-3.540301
P	2.074214	-1.246881	-0.727603
C	-0.567850	0.139123	-4.695087

H	-0.569053	1.230400	-4.774176
H	-0.417291	-0.284233	-5.693898
H	-1.547762	-0.173965	-4.326507
C	2.257397	-0.001288	-4.531493
H	2.233572	-0.519719	-5.496262
H	2.308826	1.074295	-4.715545
H	3.165290	-0.291015	-3.997083
C	3.880813	-0.960154	-0.939258
H	4.466633	-1.795060	-0.538816
H	4.134090	-0.825479	-1.994152
H	4.162781	-0.045419	-0.409480
C	1.997343	-1.652162	1.062237
H	2.268723	-0.768938	1.647564
H	0.979782	-1.941035	1.338048
H	2.679774	-2.470759	1.314693
C	-0.644161	3.407405	-1.574425
H	-1.065076	3.388500	-2.588558
H	-0.412635	4.457470	-1.353833
H	0.810507	0.704565	-0.079823

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Table S7: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{II}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bb}}^{2+}$  ( $-2393.20417104 E_h$ ).

atom	x	y	z
Ni	1.078642	1.562486	-5.386819
N	-0.994028	0.960327	-2.902943
N	3.289956	2.164739	-3.024159
P	0.766327	2.971919	-3.694607
P	1.487900	0.153206	-3.715259
C	0.350844	4.748188	-3.863926
H	1.213749	5.320466	-4.209722
C	-0.730764	2.376101	-2.761127
H	-0.609931	2.692734	-1.704811
H	-1.582770	2.930983	-3.168779
C	-2.249384	0.599007	-2.241321
C	0.114383	0.143751	-2.450864
H	-0.226726	-0.893120	-2.369242
	0.508963	0.428256	-1.456442
C	1.893342	-1.623019	-3.908297
H	1.012139	-2.195302	-4.204315
C	3.035268	0.748975	-2.867695
H	2.974254	0.432318	-1.806253
H	3.862889	0.194145	-3.322827
C	4.580756	2.526384	-2.434868
C	2.208751	2.980658	-2.509561
H	2.553672	4.017521	-2.445870
H	1.870647	2.694525	-1.495068
N	3.151311	0.960327	-7.870695
N	-1.132673	2.164739	-7.749480
P	1.390956	2.971919	-7.079031
P	0.669383	0.153206	-7.058379
C	1.806439	4.748188	-6.909712
H	0.943534	5.320466	-6.563916
C	2.888047	2.376101	-8.012511
H	2.767214	2.692734	-9.068827
H	3.740053	2.930983	-7.604860
C	4.406667	0.599007	-8.532318
C	2.042900	0.143751	-8.322774
H	2.384010	-0.893120	-8.404396
H	1.648321	0.428256	-9.317196
C	0.263941	-1.623019	-6.865341

H	1.145145	-2.195302	-6.569323
C	-0.877984	0.748975	-7.905943
H	-0.816971	0.432318	-8.967385
H	-1.705605	0.194145	-7.450811
C	-2.423473	2.526383	-8.338770
C	-0.051468	2.980658	-8.264077
H	-0.396389	4.017521	-8.327768
H	0.286636	2.694525	-9.278570
H	2.108723	5.138489	-7.888039
H	2.637425	4.882682	-6.212035
H	-0.480142	4.882682	-4.561603
H	0.048560	5.138489	-2.885599
H	-3.213054	1.935839	-7.866872
H	-2.460312	2.354193	-9.426519
H	-2.629310	3.582156	-8.144604
H	4.381831	0.771752	-9.620321
H	5.221686	1.189363	-8.105584
H	4.623261	-0.456851	-8.350720
H	-0.525795	-1.757367	-6.121277
H	-0.093846	-2.013619	-7.824645
H	-3.064403	1.189362	-2.668054
H	-2.465978	-0.456851	-2.422918
H	-2.224548	0.771752	-1.153317
H	2.251129	-2.013619	-2.948993
H	2.683078	-1.757367	-4.652361
H	5.370337	1.935839	-2.906766
H	4.617595	2.354193	-1.347119
H	4.786593	3.582156	-2.629034

Table S8: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bb}}^+$  ( $-2393.51462227 E_h$ ).

atom	x	y	z
Ni	1.078642	1.565335	-5.386819
N	-0.710995	0.378636	-2.813142
N	3.010352	2.745269	-2.920423
P	0.329353	2.826222	-3.674092
P	1.921226	0.300130	-3.721626
C	-0.607173	4.410398	-3.749863
H	0.017565	5.179049	-4.214073
C	-0.826796	1.811392	-2.612763
H	-0.698228	2.104574	-1.550210
H	-1.840801	2.108028	-2.904348
C	-1.768556	-0.320054	-2.093856
C	0.595561	-0.155982	-2.471786
H	0.521557	-1.248650	-2.487776
H	0.924249	0.123832	-1.450824
C	2.850230	-1.284348	-3.857606
H	2.196967	-2.052174	-4.282238
C	3.137641	1.312525	-2.727722
H	3.070922	1.018517	-1.659754
H	4.133033	1.016092	-3.077785
C	4.108786	3.443926	-2.265253
C	1.726375	3.281124	-2.503850
H	1.800388	4.373698	-2.525757
H	1.457683	3.003052	-1.464996
N	2.868278	0.378636	-7.960496
N	-0.853069	2.745269	-7.853215
P	1.827930	2.826223	-7.099546
P	0.236057	0.300130	-7.052012
C	2.764456	4.410398	-7.023775
H	2.139718	5.179049	-6.559566
C	2.984079	1.811392	-8.160875
H	2.855511	2.104574	-9.223428
H	3.998084	2.108029	-7.869290
C	3.925840	-0.320054	-8.679782
C	1.561723	-0.155982	-8.301852
H	1.635726	-1.248650	-8.285863
H	1.233034	0.123832	-9.322814
C	-0.692947	-1.284348	-6.916032



H	-0.039684	-2.052174	-6.491400
C	-0.980358	1.312525	-8.045916
H	-0.913638	1.018516	-9.113884
H	-1.975750	1.016091	-7.695853
C	-1.951503	3.443926	-8.508385
C	0.430908	3.281124	-8.269788
H	0.356895	4.373698	-8.247881
H	0.699600	3.003052	-9.308642
H	3.071038	4.756903	-8.016838
H	3.652716	4.273580	-6.400817
H	-1.495433	4.273580	-4.372821
H	-0.913755	4.756902	-2.756800
H	-2.904263	3.071131	-8.120388
H	-1.952892	3.315947	-9.605931
H	-1.891017	4.513819	-8.287441
H	3.862205	-0.191606	-9.775437
H	4.900005	0.052358	-8.348787
H	3.878177	-1.390008	-8.456040
H	-1.539913	-1.147143	-6.238097
H	-1.061478	-1.632265	-7.887295
H	-2.742721	0.052357	-2.424851
H	-1.720894	-1.390009	-2.317598
H	-1.704922	-0.191606	-0.998201
H	3.218761	-1.632265	-2.886343
H	3.697197	-1.147143	-4.535541
H	5.061546	3.071132	-2.653250
H	4.110175	3.315947	-1.167707
H	4.048300	4.513819	-2.486197

Table S9: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bb}}$  ( $-2393.68103156 E_h$ ).

atom	x	y	z
Ni	1.078641	1.535099	-5.386819
N	-0.406541	0.003300	-2.795329
N	2.708735	3.121547	-2.928485
P	0.094041	2.577650	-3.803496
P	2.150668	0.521049	-3.842300
C	-1.220231	3.888863	-3.808095
H	-0.857704	4.749357	-4.378070
C	-0.731016	1.400714	-2.580063
H	-0.522886	1.725693	-1.537318
H	-1.811722	1.510234	-2.733266
C	-1.247827	-0.853100	-1.984390
C	0.994267	-0.328149	-2.615552
H	1.098292	-1.405259	-2.795472
H	1.340139	-0.144265	-1.574945
C	3.463735	-0.791408	-3.885887
H	3.069139	-1.668116	-4.407990
C	3.047084	1.729074	-2.701420
H	2.904399	1.428832	-1.640500
H	4.116408	1.616157	-2.918818
C	3.596507	3.995282	-2.188639
C	1.321028	3.457753	-2.671271
H	1.205015	4.530192	-2.871209
H	1.039785	3.299896	-1.607281
N	2.563824	0.003300	-7.978309
N	-0.551452	3.121547	-7.845153
P	2.063242	2.577650	-6.970142
P	0.006615	0.521049	-6.931338
C	3.377514	3.888863	-6.965543
H	3.014987	4.749357	-6.395568
C	2.888300	1.400715	-8.193575
H	2.680170	1.725693	-9.236320
H	3.969006	1.510235	-8.040372
C	3.405111	-0.853100	-8.789247
C	1.163016	-0.328149	-8.158087
H	1.058991	-1.405259	-7.978166
H	0.817145	-0.144265	-9.198693
C	-1.306451	-0.791408	-6.887752

H	-0.911855	-1.668116	-6.365649
C	-0.889801	1.729074	-8.072218
H	-0.747115	1.428832	-9.133139
H	-1.959125	1.616157	-7.854820
C	-1.439224	3.995282	-8.584999
C	0.836255	3.457753	-8.102367
H	0.952268	4.530192	-7.902429
H	1.117499	3.299896	-9.166357
H	3.661976	4.217741	-7.973285
H	4.261453	3.499767	-6.451684
H	-2.104170	3.499767	-4.321953
H	-1.504693	4.217741	-2.800353
H	-2.479044	3.784937	-8.310885
H	-1.348040	3.885128	-9.684711
H	-1.225486	5.039960	-8.332779
H	3.248206	-0.718791	-9.878824
H	4.459581	-0.647923	-8.573346
H	3.206484	-1.903223	-8.547631
H	-2.159186	-0.418660	-6.312615
H	-1.649050	-1.091144	-7.886497
H	-2.302297	-0.647923	-2.200292
H	-1.049200	-1.903223	-2.226007
H	-1.090922	-0.718791	-0.894814
H	3.806333	-1.091143	-2.887142
H	4.316470	-0.418660	-4.461024
H	4.636327	3.784937	-2.462754
H	3.505323	3.885128	-1.088928
H	3.382769	5.039960	-2.440859

Table S10: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{II}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bb}}^+$  ( $-2394.12061924 E_h$ ).

atom	x	y	z
Ni	0.020594	-0.375028	0.130323
N	2.769481	-1.613750	-1.248225
N	2.179243	1.777819	1.523091
P	1.740898	-0.964292	1.238253
P	1.458708	0.839212	-0.992201
C	1.652517	-2.027650	2.728360
H	1.052619	-1.525091	3.492267
C	2.931886	-1.907171	0.161893
H	3.965961	-1.723144	0.521309
H	2.719030	-2.971001	0.317443
C	3.651760	-2.447431	-2.052162
C	2.916569	-0.208117	-1.568999
H	2.949954	-0.116345	-2.659772
H	3.861167	0.228947	-1.186683
C	1.133901	1.848662	-2.491307
H	0.859378	1.187841	-3.318924
C	2.267624	2.104607	0.112429
H	3.313681	2.260967	-0.223797
H	1.733573	3.047734	-0.053111
C	2.745376	2.846885	2.333942
C	2.760721	0.492874	1.859090
H	2.785358	0.410778	2.951028
H	3.806774	0.384271	1.508510
N	-2.052025	1.844743	-1.458894
N	-2.870771	-1.566751	1.204682
P	-1.569411	0.907973	1.141841
P	-1.592574	-0.914038	-1.198627
C	-1.469393	2.081199	2.561663
H	-1.039617	1.566402	3.425526
C	-2.525890	1.963963	-0.088942
H	-3.611334	1.761247	0.005190
H	-2.376937	3.005658	0.216345
C	-2.585054	2.921252	-2.285301
C	-2.329236	0.561835	-2.080655
H	-1.886728	0.575008	-3.084196
H	-3.415664	0.383027	-2.216161
C	-1.487082	-2.116564	-2.585867

H	-0.718633	-1.792431	-3.292936
C	-3.056349	-1.594130	-0.228460
H	-3.976064	-1.073265	-0.559243
H	-3.160102	-2.641730	-0.531664
C	-3.784099	-2.469283	1.889969
C	-2.904489	-0.239480	1.786797
H	-2.723726	-0.345215	2.862832
H	-3.893737	0.248910	1.674100
H	-2.448629	2.487181	2.839257
H	-0.798317	2.902555	2.297236
H	1.158729	-2.968406	2.471581
H	2.647870	-2.236961	3.134372
H	-3.663874	-3.482940	1.495862
H	-4.844546	-2.178447	1.784130
H	-3.541607	-2.489999	2.956794
H	-3.685533	2.890273	-2.376704
H	-2.305142	3.888790	-1.857007
H	-2.160199	2.857785	-3.292480
H	-1.189089	-3.090189	-2.187317
H	-2.442193	-2.219036	-3.113106
H	3.452951	-3.501351	-1.835277
H	3.451534	-2.277022	-3.114263
H	4.723209	-2.252858	-1.864754
H	2.014653	2.430967	-2.782742
H	0.289613	2.510157	-2.285972
H	2.235929	3.787908	2.103946
H	3.827327	2.993226	2.164302
H	2.590080	2.627185	3.394547
H	-0.497259	-1.574411	0.764599

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Table S11: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Me}}\text{HN}_2^{\text{Me}})(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})]_{\text{bb}}^+$  ( $-2394.11055848 E_h$ ).

atom	x	y	z
Ni	-0.080653	-0.003392	0.072283
N	2.518775	-1.525393	-1.347629
N	2.274977	1.605433	1.777808
P	1.444240	-1.016475	1.197910
P	1.566573	1.051499	-0.863302
C	1.380224	-2.328408	2.508328
H	0.798737	-1.968289	3.363083
C	2.679011	-1.858266	0.053601
H	3.707859	-1.666796	0.424337
H	2.502583	-2.935151	0.158132
C	3.324073	-2.404428	-2.181838
C	2.786738	-0.133306	-1.657052
H	2.679817	-0.008775	-2.740805
H	3.824016	0.167742	-1.402249
C	1.486958	2.349531	-2.170405
H	0.988614	1.940627	-3.053462
C	2.625636	1.936245	0.408875
H	3.699654	1.762523	0.190866
H	2.445410	3.008685	0.271182
C	2.945343	2.497104	2.712075
C	2.517080	0.219899	2.127651
H	2.267100	0.100250	3.188343
H	3.583256	-0.069136	2.017465
N	-2.360241	1.735631	-1.762333
N	-2.469886	-1.384161	1.302692
P	-1.640252	1.215590	0.888051
P	-1.522515	-0.883881	-1.242425
C	-1.670156	2.508103	2.208414
H	-1.209785	2.106213	3.115314
C	-2.719466	2.064829	-0.398400
H	-3.794412	1.885876	-0.181588
H	-2.552309	3.139698	-0.263944
C	-2.982863	2.655773	-2.702985
C	-2.616950	0.359959	-2.135224
H	-2.374435	0.256703	-3.199138
H	-3.686641	0.078448	-2.029489
C	-1.423687	-2.182528	-2.554439

H	-0.778600	-1.811229	-3.355954
C	-2.794532	-1.739529	-0.113107
H	-3.840181	-1.485974	-0.310720
H	-2.669104	-2.821540	-0.208200
C	-2.908484	-2.404133	2.281016
C	-2.888026	0.005329	1.663976
H	-2.819577	0.087252	2.752187
H	-3.929703	0.150792	1.363113
H	-2.678278	2.871928	2.437559
H	-1.055959	3.349964	1.876228
H	0.894468	-3.224775	2.109752
H	2.380401	-2.605201	2.862448
H	-2.484825	-3.370863	2.001709
H	-3.998539	-2.467852	2.285589
H	-2.552553	-2.121692	3.273798
H	-4.086628	2.592480	-2.706839
H	-2.701181	3.683275	-2.453049
H	-2.625723	2.443723	-3.715430
H	-0.950205	-3.077861	-2.142006
H	-2.399884	-2.445447	-2.977775
H	3.049784	-3.446886	-1.990897
H	3.128917	-2.189705	-3.237296
H	4.410302	-2.297075	-2.004849
H	2.481625	2.712153	-2.453743
H	0.887597	3.188962	-1.808031
H	2.688094	3.535095	2.478365
H	4.046836	2.402994	2.688726
H	2.607531	2.284521	3.731467
H	-1.390259	-1.288892	1.265934

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Table S12: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{III}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bb}}^{2+}$  ( $-2393.79261395 E_h$ ).

atom	x	y	z
Ni	0.050032	-0.421166	-0.000158
N	2.500161	-1.644950	-1.091894
N	2.104126	1.907011	1.484353
P	1.720407	-0.855538	1.404116
P	1.535805	0.927036	-1.060598
C	1.540308	-1.853057	2.923854
H	0.918527	-1.318773	3.647773
C	2.843324	-1.846777	0.309810
H	3.904591	-1.632463	0.535925
H	2.662560	-2.897649	0.558938
C	3.107393	-2.686716	-1.926620
C	2.841219	-0.308432	-1.572573
H	2.832692	-0.328982	-2.666774
H	3.851432	0.020069	-1.268356
C	1.247627	1.917488	-2.571354
H	0.929826	1.263302	-3.388036
C	2.358961	2.147839	0.075023
H	3.439354	2.185262	-0.168288
H	1.942390	3.127782	-0.182391
C	2.594193	3.032701	2.285841
C	2.666049	0.657296	1.962635
H	2.614274	0.656860	3.056333
H	3.732937	0.525127	1.696461
N	-2.024660	1.896929	-1.445510
N	-2.667256	-1.683578	1.079120
P	-1.536277	0.849973	1.095697
P	-1.653170	-0.869707	-1.360316
C	-1.262626	1.910529	2.562947
H	-0.837063	1.316349	3.376167
C	-2.477198	1.976654	-0.067501
H	-3.562820	1.792233	0.046801
H	-2.294584	2.996595	0.286696
C	-2.505568	3.048212	-2.215257
C	-2.401636	0.663442	-2.110474
H	-2.038260	0.707715	-3.143297
H	-3.498534	0.517816	-2.165744
C	-1.467576	-1.999451	-2.788018



H	-0.740359	-1.592713	-3.495587
C	-3.005499	-1.663313	-0.328396
H	-3.963710	-1.158904	-0.539020
H	-3.098513	-2.696342	-0.677911
C	-3.352857	-2.746182	1.818101
C	-2.794142	-0.387573	1.721393
H	-2.597637	-0.517633	2.790667
H	-3.808589	0.038640	1.626593
H	-2.201184	2.358123	2.907107
H	-0.549762	2.697649	2.307602
H	1.056541	-2.803803	2.685279
H	2.517146	-2.049765	3.378138
H	-3.138925	-3.709662	1.348626
H	-4.443156	-2.604684	1.852278
H	-2.974003	-2.773799	2.842932
H	-3.604751	3.090532	-2.280714
H	-2.152027	3.972507	-1.750007
H	-2.103975	3.002261	-3.231631
H	-1.107122	-2.971135	-2.440024
H	-2.424886	-2.135556	-3.302555
H	2.774005	-3.668747	-1.581649
H	2.780582	-2.561147	-2.961899
H	4.208006	-2.661538	-1.901126
H	2.158987	2.442495	-2.876186
H	0.451021	2.638536	-2.375477
H	2.117379	3.956098	1.945483
H	3.686293	3.163121	2.218422
H	2.327749	2.879154	3.335120
H	-0.664209	-1.509292	0.706872

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Table S13: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Me}}\text{HN}_2^{\text{Me}})(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})]_{\text{bb}}^{2+}$  ( $-2393.81357393 E_h$ ).

atom	x	y	z
Ni	0.027554	-0.229872	-0.138216
N	2.478688	-1.574358	-1.209647
N	2.171789	1.741684	1.659144
P	1.573848	-0.993891	1.348944
P	1.703225	1.057779	-1.005025
C	1.545510	-2.194802	2.748531
H	0.921465	-1.814423	3.562886
C	2.781523	-1.827519	0.192180
H	3.821978	-1.570525	0.466698
H	2.662776	-2.901337	0.370829
C	3.084884	-2.601087	-2.061643
C	2.874945	-0.240476	-1.650001
H	2.790421	-0.209560	-2.741238
H	3.925119	0.001086	-1.401937
C	1.652373	2.276583	-2.377306
H	1.219840	1.809380	-3.265993
C	2.647791	1.992289	0.307634
H	3.732409	1.801142	0.191574
H	2.493880	3.054972	0.091713
C	2.691807	2.760263	2.574741
C	2.504250	0.417345	2.152340
H	2.233746	0.379812	3.213465
H	3.588947	0.195923	2.098047
N	-2.266233	1.805644	-1.651871
N	-2.559240	-1.546819	1.201542
P	-1.536675	1.053005	0.924634
P	-1.711085	-0.912018	-1.382133
C	-1.317565	2.210608	2.330096
H	-0.863009	1.687242	3.175394
C	-2.575428	2.063709	-0.262601
H	-3.650743	1.945194	-0.018742
H	-2.323390	3.108436	-0.049991
C	-2.781254	2.868711	-2.516341
C	-2.694214	0.504420	-2.115764
H	-2.509675	0.453927	-3.194407
H	-3.778194	0.319332	-1.973204
C	-1.680862	-2.120010	-2.764501

H	-1.045880	-1.720202	-3.560294
C	-2.952801	-1.745166	-0.233353
H	-3.981428	-1.393151	-0.354200
H	-2.931052	-2.821887	-0.421956
C	-3.192947	-2.555516	2.102044
C	-2.785523	-0.141514	1.682452
H	-2.637655	-0.152394	2.765633
H	-3.828875	0.119352	1.481384
H	-2.262892	2.663981	2.645765
H	-0.623562	2.995756	2.020075
H	1.137794	-3.152912	2.412920
H	2.553262	-2.367605	3.142317
H	-2.938958	-3.556703	1.750477
H	-4.275589	-2.419511	2.083722
H	-2.818952	-2.412815	3.117014
H	-3.881492	2.936522	-2.506638
H	-2.373227	3.830217	-2.193325
H	-2.454646	2.691684	-3.544519
H	-1.240712	-3.062038	-2.426294
H	-2.679196	-2.308506	-3.172341
H	2.720984	-3.587656	-1.762168
H	2.791781	-2.431822	-3.101100
H	4.185390	-2.604188	-2.007127
H	2.651928	2.651077	-2.621843
H	1.015011	3.118471	-2.094419
H	2.386945	3.752364	2.229888
H	3.791431	2.745636	2.653784
H	2.274424	2.604480	3.573745
H	-1.519022	-1.659955	1.202204

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Table S14: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{II}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bc}}^{2+}$  ( $-2393.20912401 E_h$ ).

atom	x	y	z
Ni	1.985238	14.157571	16.904792
N	3.620791	13.757510	14.094659
N	1.098737	11.080173	14.811960
N	1.395292	14.126669	20.130804
N	2.269258	17.497968	18.745945
P	0.964758	13.811206	14.955793
P	3.065075	12.292187	16.325485
P	3.366856	14.980898	18.452611
P	0.548583	15.550490	17.882667
C	-0.303872	14.937461	14.260877
H	-1.268845	14.793776	14.750993
C	2.255846	13.825916	13.608998
H	2.000347	13.017976	12.897943
H	2.129775	14.781417	13.088980
C	4.565558	14.079144	13.020891
C	3.935908	12.473756	14.699573
H	5.010144	12.444365	14.909210
H	3.688330	11.608574	14.059528
C	4.266463	11.445576	17.420820
H	4.494314	10.456153	17.010822
C	1.803890	10.938734	16.066162
H	2.365625	9.998779	16.039735
H	1.143527	10.897760	16.953225
C	0.500865	9.817855	14.370172
C	0.137283	12.158012	14.805604
H	-0.628440	12.078175	15.602153
H	-0.391432	12.150741	13.845190
C	5.148534	14.558741	18.535772
H	5.649493	15.240932	19.230489
C	2.725293	14.711700	20.170489
H	3.402123	14.000276	20.654815
H	2.774976	15.664452	20.726624
C	1.027529	13.566954	21.434819
C	0.386579	15.064391	19.676568
H	0.353865	15.997685	20.269514
H	-0.590774	14.578190	19.764266
C	-1.208842	15.707529	17.385165

H	-1.755129	16.218784	18.185333
C	1.100749	17.321054	17.915577
H	1.240061	17.661691	16.870711
H	0.276495	17.905895	18.340473
C	2.515758	18.911545	19.041055
C	3.448724	16.834781	18.236708
H	4.311469	17.172657	18.820901
H	3.668217	17.056379	17.174927
H	-0.281568	9.453528	15.054481
H	0.062372	9.954212	13.378212
H	1.279698	9.055293	14.289783
H	5.198868	12.008129	17.483528
H	3.849104	11.317951	18.423719
H	0.056843	13.070627	21.355431
H	0.968131	14.330038	22.227140
H	1.766657	12.817109	21.728418
H	-1.661341	14.724366	17.231300
H	-1.308786	16.300290	16.473775
H	5.614809	14.661378	17.551736
H	5.289864	13.540731	18.901126
H	4.350130	15.079359	12.636129
H	5.582126	14.084081	13.422575
H	4.522490	13.364079	12.183976
H	-0.426202	14.712506	13.195733
H	0.002874	15.981998	14.360331
H	2.761630	19.498722	18.142170
H	3.341595	18.995790	19.752000
H	1.626653	19.342802	19.508333

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Table S15: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bc}}^+$  ( $-2393.52100995 E_h$ ).

atom	x	y	z
Ni	1.936202	14.169294	16.893555
N	3.949023	13.357895	14.335212
N	0.889184	11.338151	14.516122
N	1.960783	14.156121	20.252105
N	1.827255	17.515302	18.775370
P	1.280518	14.078636	14.747357
P	2.705701	12.097628	16.472438
P	3.394591	15.305582	18.173571
P	0.409154	15.202385	18.176117
C	0.381217	15.384164	13.808144
H	-0.615959	15.524251	14.236851
C	2.731279	13.718962	13.624664
H	2.424909	12.958627	12.882365
H	2.932503	14.649924	13.082594
C	5.089435	13.387203	13.426725
C	3.884524	12.076640	15.025574
H	4.879430	11.875520	15.438703
H	3.624272	11.238095	14.354000
C	3.526511	10.995293	17.695116
H	3.829150	10.040057	17.252745
C	1.332725	10.988855	15.853667
H	1.722323	9.964545	15.818772
H	0.514492	11.002841	16.599393
C	0.114830	10.256139	13.919245
C	0.182458	12.601576	14.417088
H	-0.689843	12.662967	15.097157
H	-0.197731	12.691700	13.392521
C	5.224019	15.274154	17.983244
H	5.728999	15.894328	18.731662
C	3.150664	14.954899	19.990290
H	4.018174	14.368136	20.312861
H	3.162404	15.907247	20.551390
C	1.979203	13.651684	21.620202
C	0.705700	14.843229	19.986482
H	0.601972	15.785429	20.556134
H	-0.105270	14.175448	20.298637
C	-1.423563	15.058521	18.051700

H	-1.930886	15.593351	18.862119
C	0.611019	17.060773	18.127632
H	0.536468	17.386215	17.071344
H	-0.237500	17.502140	18.663910
C	1.780520	18.950447	19.029885
C	3.056579	17.143116	18.098565
H	3.883698	17.654942	18.604716
H	3.078754	17.455604	17.036668
H	-0.830774	10.057085	14.453900
H	-0.122973	10.505082	12.880861
H	0.709032	9.337691	13.917625
H	4.410048	11.495267	18.102159
H	2.838611	10.805549	18.524156
H	1.123484	12.988188	21.777846
H	1.939367	14.453549	22.379093
H	2.891914	13.070792	21.784116
H	-1.711593	14.003973	18.085642
H	-1.760784	15.471730	17.096073
H	5.485706	15.632545	16.983413
H	5.582198	14.244568	18.072928
H	5.173668	14.380698	12.975938
H	6.008921	13.190997	13.986560
H	5.015348	12.643581	12.613100
H	0.272889	15.124364	12.749386
H	0.921168	16.331872	13.889059
H	1.749493	19.551547	18.103919
H	2.661678	19.249744	19.604860
H	0.893648	19.188627	19.624443

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Table S16: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bc}}$  ( $-2393.68992724 E_h$ ).

atom	x	y	z
Ni	1.918093	14.181235	16.883186
N	4.135000	13.039412	14.608674
N	0.778243	11.637831	14.340904
N	2.423964	14.196197	20.223339
N	1.480768	17.410138	18.779433
P	1.618131	14.273940	14.773707
P	2.374995	12.115078	16.583353
P	3.288640	15.477125	17.887879
P	0.473362	14.839353	18.307021
C	1.107769	15.679263	13.670091
H	0.099597	16.003325	13.948644
C	3.098200	13.652628	13.791927
H	2.745807	12.982036	12.983197
H	3.544561	14.535280	13.316717
C	5.323002	12.791897	13.815580
C	3.738890	11.832701	15.320471
H	4.616274	11.489053	15.882879
H	3.443247	11.015253	14.633228
C	2.822583	10.811192	17.822115
H	3.089579	9.852896	17.358816
C	0.992186	11.193183	15.709811
H	1.248444	10.125655	15.666423
H	0.079688	11.291009	16.331673
C	-0.124108	10.739266	13.645354
C	0.345897	13.018983	14.189404
H	-0.599021	13.225941	14.730576
H	0.156039	13.181267	13.119543
C	5.069121	15.827670	17.512207
H	5.541931	16.488797	18.249507
C	3.410724	15.148634	19.734716
H	4.399069	14.703937	19.906726
H	3.377922	16.109932	20.284901
C	2.710965	13.829511	21.596194
C	1.035099	14.609745	20.088207
H	0.810405	15.529402	20.664245
H	0.418527	13.802415	20.503153
C	-1.324426	14.417904	18.514446



H	-1.774816	14.882802	19.400827
C	0.295264	16.711150	18.306609
H	0.007376	17.017529	17.281012
H	-0.529801	16.986852	18.977553
C	1.194197	18.815947	18.996298
C	2.672908	17.248985	17.960781
H	3.456240	17.880405	18.402211
H	2.517082	17.591976	16.918219
H	-1.146415	10.734428	14.072357
H	-0.198710	11.030513	12.591935
H	0.265368	9.716295	13.688697
H	3.660002	11.171242	18.426413
H	1.974315	10.657465	18.496503
H	2.019199	13.044705	21.922385
H	2.623581	14.676781	22.305798
H	3.730588	13.434479	21.668255
H	-1.430272	13.331135	18.586566
H	-1.872797	14.747605	17.625856
H	5.136584	16.289516	16.522257
H	5.613715	14.880261	17.468390
H	5.665109	13.727398	13.358764
H	6.125857	12.415167	14.459299
H	5.164315	12.054511	13.003101
H	1.111830	15.412023	12.605562
H	1.784806	16.524060	13.830118
H	0.935488	19.355074	18.063623
H	2.066562	19.307689	19.440373
H	0.354195	18.920387	19.691682

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Table S17: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{II}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bc}}^+$  ( $-2394.12853915 E_h$ ).

atom	x	y	z
Ni	1.612477	13.878626	17.158879
N	3.949555	13.595557	14.665878
N	0.982128	11.512492	14.257980
N	2.160805	14.321722	20.400652
N	1.852287	17.441048	18.456461
P	1.235706	14.197149	14.929117
P	2.509266	12.039635	16.504870
P	3.249572	15.109011	17.964271
P	0.390205	15.090938	18.408980
C	0.389664	15.582441	14.047707
H	-0.646493	15.656743	14.392705
C	2.780268	13.999506	13.893453
H	2.565549	13.310554	13.056677
H	2.999443	14.984907	13.466231
C	5.153496	13.709415	13.849732
C	3.871364	12.261185	15.243134
H	4.816694	12.073767	15.766421
H	3.754806	11.471924	14.477243
C	3.222032	10.837909	17.693407
H	3.677814	9.982805	17.182521
C	1.320337	10.965661	15.557742
H	1.811662	9.998437	15.395136
H	0.438667	10.790070	16.202320
C	0.321975	10.513247	13.426575
C	0.225950	12.752567	14.294515
H	-0.691186	12.679370	14.909468
H	-0.082599	12.980506	13.267361
C	5.023616	14.935798	17.527116
H	5.643504	15.658515	18.068151
C	3.290424	15.039127	19.833758
H	4.198956	14.489802	20.104922
H	3.389777	16.068477	20.226003
C	2.379229	14.038638	21.812846
C	0.879224	14.974117	20.202914
H	0.836074	15.989285	20.641684
H	0.113499	14.363210	20.694165
C	-1.430142	14.888319	18.436835

H	-1.897567	15.560717	19.163814
C	0.558440	16.914813	18.066995
H	0.321782	17.082470	16.997886
H	-0.207347	17.434287	18.655971
C	1.843962	18.898048	18.481775
C	2.973497	16.932428	17.686765
H	3.873134	17.465720	18.016521
H	2.868419	17.104410	16.597855
H	-0.656942	10.197400	13.828132
H	0.167951	10.916472	12.421405
H	0.959894	9.628397	13.340916
H	3.974197	11.334080	18.312421
H	2.426891	10.482431	18.354869
H	1.557443	13.424685	22.193751
H	2.448313	14.949693	22.433999
H	3.307395	13.471554	21.932213
H	-1.670318	13.852562	18.689854
H	-1.837876	15.099621	17.444032
H	5.132289	15.071319	16.448893
H	5.359943	13.924764	17.775479
H	5.249926	14.734366	13.477912
H	6.035258	13.483466	14.458309
H	5.153033	13.027526	12.981010
H	0.384747	15.437452	12.961601
H	0.889888	16.528065	14.276419
H	1.682016	19.344970	17.484828
H	2.798873	19.262832	18.871435
H	1.051208	19.248875	19.149192
H	0.562047	12.916422	17.509664

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Table S18: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Me}}\text{HN}_2^{\text{Me}})(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})]_{\text{bc}}^+$  ( $-2394.12116660 E_h$ ).

atom	x	y	z
Ni	-0.069791	-0.083515	-0.005265
N	2.439944	1.774093	-1.200141
N	3.467234	-1.219159	0.653922
N	-2.387329	0.990458	1.648810
N	-3.565923	-0.785223	-1.266748
P	1.506959	-0.852107	-1.273349
P	1.522872	0.696564	1.215838
P	-1.555155	1.118913	-0.971155
P	-1.603932	-1.438804	0.608110
C	1.313749	-1.849108	-2.814388
H	0.835434	-2.803570	-2.573421
C	2.676773	0.491229	-1.846437
H	3.717987	0.137957	-1.723871
H	2.497322	0.627209	-2.919514
C	3.211392	2.820099	-1.856427
C	2.691015	1.784682	0.232522
H	2.527579	2.810217	0.584618
H	3.731240	1.506105	0.487084
C	1.461656	1.651030	2.803449
H	2.458591	1.963003	3.136131
C	2.697783	-0.662232	1.751773
H	3.410637	-0.232472	2.466470
H	2.106591	-1.427542	2.292818
C	4.575397	-2.023793	1.148207
C	2.693539	-1.948983	-0.334712
H	2.112135	-2.783075	0.105787
H	3.400885	-2.384777	-1.051369
C	-1.395017	2.666666	-1.962548
H	-2.356117	3.150601	-2.169487
C	-2.761508	1.689203	0.375052
H	-2.634087	2.761038	0.549271
H	-3.806099	1.465858	0.147595
C	-2.784758	1.742580	2.861015
C	-2.809860	-0.447911	1.685862
H	-3.846704	-0.517467	1.350056
H	-2.725614	-0.779340	2.724288
C	-1.563149	-3.026393	1.557046

H	-2.558490	-3.390351	1.836296
C	-2.806227	-1.916118	-0.749831
H	-2.230079	-2.435940	-1.538328
H	-3.524822	-2.637066	-0.341225
C	-4.707315	-1.249873	-2.049870
C	-2.782490	0.178790	-2.028195
H	-3.482762	0.895308	-2.475132
H	-2.221186	-0.292038	-2.857531
H	4.243358	-2.920011	1.703517
H	5.194772	-2.351836	0.307850
H	5.200686	-1.420459	1.813464
H	0.842487	2.544758	2.674806
H	1.014719	1.030874	3.587763
H	-2.396417	1.229724	3.743073
H	-3.873580	1.798971	2.917366
H	-2.366593	2.749740	2.809496
H	-0.956795	-2.895581	2.458159
H	-1.075215	-3.787741	0.940008
H	-0.915008	2.416962	-2.913719
H	-0.732135	3.360631	-1.439168
H	2.953044	2.855251	-2.919667
H	2.965146	3.791462	-1.415418
H	4.303888	2.672859	-1.773536
H	2.276038	-2.050856	-3.298572
H	0.667123	-1.315556	-3.516957
H	-4.407735	-1.806658	-2.954755
H	-5.313952	-0.393576	-2.359803
H	-5.333322	-1.904646	-1.436100
H	-1.317133	0.908879	1.556790

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Table S19: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{III}}(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})_2]_{\text{bc}}^{2+}$  ( $-2393.79549860 E_h$ ).

atom	x	y	z
Ni	1.649131	13.889080	17.268582
N	3.938775	13.570971	14.748185
N	0.970559	11.427714	14.403046
N	2.113915	14.230776	20.050729
N	1.874101	17.557135	18.442231
P	1.222559	14.107589	14.984634
P	2.553550	11.979025	16.582992
P	3.337482	15.296956	17.862062
P	0.311272	15.292508	18.326890
C	0.344493	15.532914	14.233689
H	-0.672789	15.601497	14.630812
C	2.767484	13.929669	13.960170
H	2.555446	13.214119	13.146668
H	2.957743	14.907775	13.505141
C	5.154702	13.729025	13.943741
C	3.888707	12.229494	15.307604
H	4.842523	12.037836	15.811988
H	3.752671	11.444731	14.541908
C	3.265288	10.877072	17.855812
H	3.712441	9.992030	17.390885
C	1.324372	10.899701	15.702589
H	1.807901	9.926639	15.556080
H	0.461194	10.744039	16.376249
C	0.346708	10.408391	13.555672
C	0.193600	12.649561	14.443360
H	-0.697208	12.588788	15.096642
H	-0.159485	12.862820	13.428150
C	5.098325	15.143472	17.398908
H	5.712401	15.811163	18.012342
C	3.283295	15.042279	19.702367
H	4.179755	14.471631	19.964959
H	3.323177	16.003452	20.239217
C	2.311654	13.530306	21.325074
C	0.866730	14.994724	20.066833
H	0.938714	15.943640	20.622685
H	0.093954	14.379056	20.538176
C	-1.501957	15.095391	18.276564

H	-1.982333	15.769546	18.993271
C	0.579293	17.097450	17.989202
H	0.383011	17.290868	16.917290
H	-0.194325	17.629903	18.555145
C	1.894253	19.010625	18.630363
C	2.985915	17.106816	17.628147
H	3.884236	17.645566	17.949607
H	2.857631	17.311678	16.548118
H	-0.622990	10.062592	13.946833
H	0.194527	10.813903	12.552133
H	1.015770	9.548033	13.471203
H	4.031368	11.403988	18.430138
H	2.476725	10.557628	18.542925
H	1.459068	12.873921	21.516126
H	2.420611	14.222061	22.174235
H	3.210195	12.911326	21.264535
H	-1.767402	14.062652	18.516112
H	-1.878658	15.326154	17.275749
H	5.221166	15.393037	16.343484
H	5.434873	14.114143	17.546341
H	5.223085	14.759546	13.583535
H	6.033360	13.524482	14.562372
H	5.182498	13.056185	13.071945
H	0.281780	15.419982	13.145984
H	0.870437	16.465068	14.458138
H	1.750373	19.565430	17.689819
H	2.851493	19.305912	19.067435
H	1.105109	19.297608	19.330199
H	0.481850	13.023253	17.427728

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Table S20: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Me}}\text{HN}_2^{\text{Me}})(\text{P}_2^{\text{Me}}\text{N}_2^{\text{Me}})]_{\text{bc}}^{2+}$  ( $-2393.82246145 E_h$ ).

atom	x	y	z
Ni	0.033992	0.162956	-0.164276
N	2.409235	1.706849	-1.025795
N	3.478832	-1.387145	0.628618
N	-2.475398	1.205943	1.560506
N	-3.566297	-0.827679	-1.227805
P	1.637695	-0.907864	-1.385066
P	1.660315	0.578492	1.377769
P	-1.704935	1.212918	-1.116431
P	-1.554731	-1.271421	0.628120
C	1.463728	-1.812790	-2.975183
H	0.948218	-2.763614	-2.808946
C	2.775284	0.508963	-1.783908
H	3.828618	0.214382	-1.644326
H	2.627586	0.730751	-2.846107
C	2.964843	2.902812	-1.666594
C	2.805445	1.656080	0.381050
H	2.702360	2.665842	0.792253
H	3.850324	1.336138	0.527174
C	1.624422	1.407688	3.023423
H	2.637346	1.613810	3.385830
C	2.693132	-0.934638	1.758510
H	3.385901	-0.648395	2.557461
H	2.031418	-1.720265	2.172494
C	4.625291	-2.188509	1.061573
C	2.722768	-2.065736	-0.405675
H	2.097641	-2.893065	-0.016697
H	3.436255	-2.508982	-1.109399
C	-1.563943	2.698983	-2.180965
H	-2.540234	3.131983	-2.420432
C	-2.882005	1.782746	0.231414
H	-2.826209	2.870209	0.327790
H	-3.913128	1.478845	0.036664
C	-3.068393	1.969462	2.699562
C	-2.745780	-0.270125	1.681737
H	-3.786744	-0.442688	1.398034
H	-2.602051	-0.531803	2.733306
C	-1.307006	-2.766135	1.665757



H	-2.252565	-3.185183	2.024884
C	-2.728753	-1.885727	-0.689123
H	-2.128636	-2.402516	-1.459981
H	-3.380445	-2.635982	-0.227643
C	-4.742976	-1.380338	-1.909586
C	-2.857799	0.109117	-2.083620
H	-3.597508	0.749287	-2.577439
H	-2.267311	-0.383985	-2.876758
H	4.329582	-3.131831	1.549231
H	5.250053	-2.426207	0.196651
H	5.232049	-1.609442	1.762724
H	1.080661	2.354925	2.958487
H	1.123863	0.768229	3.757620
H	-2.687217	1.565313	3.638540
H	-4.154429	1.868910	2.666202
H	-2.790382	3.020479	2.608116
H	-0.668672	-2.529166	2.521653
H	-0.796620	-3.528179	1.068511
H	-1.070092	2.412593	-3.114438
H	-0.937608	3.448394	-1.690058
H	2.605900	2.965479	-2.697383
H	2.625990	3.795445	-1.133657
H	4.066400	2.905071	-1.682364
H	2.440061	-2.021828	-3.424997
H	0.869569	-1.221557	-3.677448
H	-4.479028	-1.981397	-2.793427
H	-5.396593	-0.563773	-2.227238
H	-5.304497	-2.010721	-1.215117
H	-1.435110	1.287524	1.573885

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Table S21: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{II}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})_2]_{\text{bc}}^{2+}$  ( $-3931.69013548 E_h$ ).

atom	x	y	z
Ni	5.576892	9.951417	1.459623
P	3.440887	10.239878	0.962719
P	5.016412	10.861412	3.387890
P	7.743958	10.397494	1.437097
P	6.110645	8.406729	-0.020338
N	2.328251	10.126991	3.423746
N	4.176219	12.838720	1.735487
N	7.125891	10.571325	-1.298597
N	8.749291	7.978411	0.766445
C	2.229674	9.469403	2.143345
H	1.223703	9.641520	1.748095
H	2.404848	8.380130	2.171645
C	3.570954	9.928890	4.135588
H	3.864151	8.866289	4.218229
H	3.457208	10.306661	5.155165
C	3.080534	12.073036	1.159902
H	2.874760	12.453172	0.158569
H	2.154346	12.153749	1.752622
C	4.405223	12.593723	3.145059
H	3.519605	12.760850	3.781088
H	5.205714	13.260105	3.476791
C	6.829239	9.174718	-1.545748
H	7.698172	8.581741	-1.877508
H	6.056556	9.126547	-2.317429
C	8.197967	10.799431	-0.340775
H	8.463048	11.857237	-0.356095
H	9.103939	10.217448	-0.577784
C	7.469821	7.312502	0.667438
H	7.114296	6.901163	1.629882
H	7.563575	6.469641	-0.022455
C	8.861884	8.961745	1.816237
H	9.890920	9.334293	1.825619
H	8.624913	8.584453	2.825988
C	2.879167	9.855963	-0.720003
C	3.552174	10.478811	-1.783043
H	4.399788	11.134326	-1.590939
C	3.138919	10.260791	-3.093192
H	3.656424	10.757017	-3.908695
C	2.059327	9.416071	-3.352598
H	1.731052	9.253104	-4.374567
C	1.398178	8.786496	-2.301125
H	0.556846	8.130314	-2.501017
C	1.803507	9.002235	-0.985781
H	1.274826	8.498744	-0.182917
C	6.191743	10.808267	4.770937

C	6.785045	9.568015	5.051401
H	6.578751	8.706487	4.418412
C	7.626890	9.424987	6.149910
H	8.071339	8.459004	6.369999
C	7.887232	10.519693	6.972938
H	8.540459	10.408805	7.832873
C	7.300933	11.752928	6.698339
H	7.497076	12.604451	7.342432
C	6.451309	11.900678	5.604645
H	5.997631	12.868863	5.418527
C	1.147907	10.106221	4.243952
C	0.200492	9.085410	4.146194
H	0.345262	8.258639	3.456798
C	-0.939624	9.113572	4.948142
H	-1.672607	8.317384	4.861113
C	-1.131415	10.142109	5.864580
H	-2.017497	10.157873	6.490775
C	-0.179633	11.155303	5.970524
H	-0.325511	11.968481	6.674897
C	0.948479	11.144610	5.158553
H	1.670639	11.954939	5.221398
C	4.221824	14.211636	1.323931
C	4.092847	15.269453	2.228010
H	3.901810	15.082079	3.279932
C	4.177211	16.588672	1.781286
H	4.070533	17.398747	2.496447
C	4.368748	16.868561	0.433183
H	4.422125	17.896137	0.088665
C	4.485117	15.814555	-0.472588
H	4.634061	16.018140	-1.528829
C	4.423530	14.497772	-0.032878
H	4.542451	13.685221	-0.745176
C	8.352461	11.822028	2.383429
C	7.761670	13.069387	2.126417
H	6.946801	13.157240	1.410064
C	8.215875	14.205580	2.787917
H	7.762378	15.168583	2.573336
C	9.254561	14.105313	3.713878
H	9.615063	14.993636	4.223446
C	9.833873	12.867420	3.980422
H	10.643530	12.787726	4.699087
C	9.387122	11.725096	3.319796
H	9.851986	10.770974	3.545982
C	4.880661	7.152532	-0.480258
C	4.208481	6.509036	0.569984
H	4.391201	6.806828	1.601206
C	3.317126	5.474471	0.303394
H	2.810963	4.970879	1.121495
C	3.086077	5.078318	-1.013128
H	2.394188	4.268433	-1.222564

C	3.750784	5.714234	-2.058968
H	3.577201	5.402158	-3.084099
C	4.649711	6.745643	-1.798152
H	5.163231	7.217916	-2.629464
C	7.179704	11.412803	-2.458828
C	7.053637	12.797935	-2.288186
H	6.916999	13.210200	-1.291671
C	7.089880	13.648403	-3.386526
H	6.998714	14.719864	-3.235091
C	7.230427	13.133689	-4.675007
H	7.253535	13.799019	-5.531890
C	7.347083	11.759509	-4.849167
H	7.471459	11.344793	-5.844954
C	7.333076	10.900237	-3.749848
H	7.467572	9.835733	-3.913663
C	9.892402	7.130823	0.562391
C	10.781404	6.831178	1.596349
H	10.616402	7.216111	2.598530
C	11.887832	6.019236	1.350740
H	12.575786	5.796009	2.160458
C	12.102408	5.484548	0.084802
H	12.961835	4.848734	-0.101313
C	11.208252	5.774212	-0.944881
H	11.373023	5.370768	-1.939246
C	10.115192	6.600666	-0.711879
H	9.439818	6.850749	-1.526384

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Table S22: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})_2]_{\text{bc}}^+$  ( $-3931.98283105 E_h$ ).

atom	x	y	z
Ni	5.588851	10.188802	1.292286
P	3.460944	10.829984	0.811973
P	4.958287	10.625513	3.420481
P	7.767409	10.732163	0.932845
P	6.156734	8.291375	0.199905
N	2.216125	10.072646	3.147553
N	4.160056	13.056906	2.361863
N	7.129737	10.046536	-1.708351
N	8.873221	8.211635	0.915738
C	2.192826	9.796073	1.727921
H	1.202801	10.062282	1.349459
H	2.373007	8.738234	1.473923
C	3.406297	9.654891	3.864359
H	3.636998	8.585559	3.709737
H	3.232732	9.789918	4.934382
C	3.082003	12.520334	1.540619
H	2.919375	13.193889	0.698459
H	2.139072	12.453263	2.106859
C	4.372985	12.392106	3.631416
H	3.486297	12.413085	4.288973
H	5.188607	12.912202	4.142064
C	6.831163	8.642407	-1.511687
H	7.691905	7.981075	-1.713551
H	6.027314	8.377301	-2.204526
C	8.208288	10.583958	-0.887980
H	8.438950	11.591553	-1.235796
H	9.124048	9.977681	-0.978023
C	7.634005	7.465310	1.026642
H	7.352409	7.271169	2.077159
H	7.770905	6.491838	0.550274
C	8.939608	9.456213	1.649821
H	9.957128	9.844978	1.561584
H	8.713927	9.355671	2.724557
C	2.791788	10.929498	-0.882664
C	3.693244	10.799067	-1.944902
H	4.745942	10.624850	-1.733473
C	3.252469	10.914067	-3.261830
H	3.965192	10.819798	-4.075989
C	1.906868	11.156094	-3.524447
H	1.559541	11.243200	-4.549836
C	1.002607	11.294442	-2.471261
H	-0.045801	11.491039	-2.675563
C	1.440583	11.185203	-1.155528
H	0.723025	11.310207	-0.348449
C	5.948314	10.295730	4.924570

C	6.771690	9.163034	4.917504
H	6.829964	8.551357	4.020025
C	7.510001	8.812543	6.044578
H	8.139683	7.927785	6.025987
C	7.443689	9.600846	7.190821
H	8.024779	9.335482	8.068830
C	6.627240	10.729651	7.209885
H	6.569073	11.344243	8.103354
C	5.876395	11.071743	6.088358
H	5.233018	11.945789	6.134276
C	0.989313	9.900722	3.855786
C	-0.031148	9.066776	3.387071
H	0.103785	8.492791	2.475492
C	-1.226370	8.949704	4.095112
H	-2.007765	8.298868	3.713359
C	-1.414673	9.641846	5.286146
H	-2.344824	9.544760	5.837021
C	-0.395802	10.465624	5.763199
H	-0.531986	11.022027	6.686015
C	0.789821	10.601873	5.052475
H	1.563039	11.274331	5.414852
C	4.273212	14.476041	2.362340
C	4.226009	15.235526	3.536899
H	4.038079	14.759510	4.493656
C	4.393920	16.619615	3.491452
H	4.350684	17.188597	4.415803
C	4.592280	17.270961	2.279523
H	4.714811	18.348961	2.247472
C	4.630506	16.519917	1.104268
H	4.794818	17.010638	0.149248
C	4.483042	15.139913	1.143291
H	4.556651	14.563560	0.225012
C	8.508486	12.324309	1.428379
C	7.644017	13.334023	1.865606
H	6.575534	13.136926	1.917050
C	8.142602	14.588573	2.210916
H	7.458459	15.365507	2.539612
C	9.509315	14.839522	2.125322
H	9.901582	15.815271	2.396772
C	10.377479	13.840714	1.684099
H	11.442805	14.038512	1.609901
C	9.881982	12.589081	1.333611
H	10.572969	11.828596	0.978496
C	5.088636	6.822055	-0.026177
C	4.209350	6.500701	1.015320
H	4.158817	7.146339	1.889201
C	3.405815	5.366055	0.944120
H	2.733004	5.126615	1.762433
C	3.462596	4.544431	-0.179029
H	2.831042	3.663435	-0.242002

C	4.334232	4.853574	-1.221038
H	4.384942	4.212839	-2.096413
C	5.149634	5.979530	-1.143380
H	5.834168	6.188911	-1.960407
C	7.099272	10.518063	-3.051603
C	6.972884	11.896879	-3.283072
H	6.899156	12.577273	-2.439038
C	6.907780	12.395032	-4.577403
H	6.807167	13.465605	-4.731081
C	6.947143	11.531476	-5.672593
H	6.889000	11.922423	-6.683489
C	7.062971	10.164049	-5.451089
H	7.105688	9.477060	-6.291475
C	7.148012	9.657459	-4.154108
H	7.272834	8.588793	-4.013856
C	10.059046	7.418489	0.886799
C	11.051951	7.526764	1.866035
H	10.924101	8.203255	2.705480
C	12.209136	6.753562	1.784558
H	12.969645	6.853896	2.553620
C	12.385088	5.849665	0.742881
H	13.285308	5.246057	0.685185
C	11.392914	5.730110	-0.229535
H	11.520565	5.036980	-1.055970
C	10.246434	6.511650	-0.164872
H	9.495963	6.436205	-0.947501

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Table S23: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})_2]_{\text{bc}}$  ( $-3932.15677115 E_h$ ).

atom	x	y	z
Ni	5.586349	10.172812	1.311407
P	3.707939	11.122955	0.817718
P	4.958228	10.401273	3.361023
P	7.539820	10.837705	0.662653
P	6.135388	8.278776	0.442337
N	2.182122	10.208122	2.966805
N	4.372146	13.087343	2.726400
N	6.912130	9.728743	-1.851371
N	8.912438	8.420769	0.867263
C	2.239204	10.165740	1.518407
H	1.323447	10.628114	1.139616
H	2.290890	9.141719	1.111253
C	3.288200	9.589962	3.686498
H	3.394567	8.521703	3.432900
H	3.069624	9.640293	4.755376
C	3.376186	12.750712	1.712805
H	3.387150	13.526427	0.947004
H	2.364506	12.730584	2.152226
C	4.488709	12.169346	3.841691
H	3.573031	12.146642	4.460640
H	5.310555	12.520385	4.472244
C	6.678583	8.382637	-1.367684
H	7.555157	7.727433	-1.524689
H	5.843205	7.966754	-1.938456
C	7.935911	10.497631	-1.150689
H	8.028397	11.470934	-1.632039
H	8.914891	9.994740	-1.223892
C	7.742820	7.604524	1.163783
H	7.587568	7.495324	2.250821
H	7.916833	6.601734	0.768087
C	8.919041	9.772299	1.391257
H	9.873295	10.228666	1.115222
H	8.832556	9.817654	2.489929
C	3.003736	11.511257	-0.842489
C	3.731301	11.120329	-1.969217
H	4.689631	10.623574	-1.827635
C	3.244665	11.378489	-3.250932
H	3.824954	11.070396	-4.115975
C	2.030179	12.038252	-3.415435
H	1.651011	12.242180	-4.413361
C	1.300084	12.441724	-2.297330
H	0.353695	12.961595	-2.421701
C	1.781806	12.176163	-1.019394
H	1.199378	12.495347	-0.157793
C	5.784394	9.893867	4.940563



C	6.949655	9.127983	4.866095
H	7.332945	8.862559	3.885299
C	7.613169	8.713099	6.019368
H	8.516379	8.114352	5.938653
C	7.119528	9.070110	7.270655
H	7.635094	8.752501	8.172751
C	5.958772	9.837268	7.362784
H	5.568342	10.119215	8.337067
C	5.295119	10.240578	6.207968
H	4.388387	10.834073	6.301854
C	0.906102	10.050259	3.559238
C	-0.175168	9.479297	2.873036
H	-0.040395	9.099452	1.865703
C	-1.426453	9.373636	3.477690
H	-2.245880	8.925871	2.921469
C	-1.627466	9.816754	4.779924
H	-2.602947	9.730252	5.248946
C	-0.555729	10.379613	5.473047
H	-0.694803	10.745492	6.486914
C	0.689522	10.501488	4.872142
H	1.502213	10.979115	5.412370
C	4.465916	14.463126	3.040816
C	4.420801	14.941031	4.359413
H	4.254852	14.256488	5.183380
C	4.562995	16.300348	4.633519
H	4.520832	16.636373	5.666365
C	4.738675	17.220889	3.607529
H	4.843375	18.279645	3.824465
C	4.779869	16.757406	2.292065
H	4.928353	17.455460	1.472335
C	4.652562	15.404181	2.010659
H	4.733816	15.066234	0.982129
C	8.345455	12.486396	0.864372
C	7.654177	13.476637	1.566467
H	6.663573	13.249425	1.955611
C	8.217842	14.739595	1.749472
H	7.664035	15.498324	2.295062
C	9.474818	15.024895	1.224251
H	9.913961	16.009168	1.364036
C	10.170536	14.046311	0.514197
H	11.150402	14.266344	0.098479
C	9.611142	12.784560	0.338852
H	10.167308	12.032467	-0.217185
C	5.220389	6.668016	0.364010
C	4.029975	6.538123	1.082422
H	3.672520	7.389514	1.653757
C	3.308067	5.345926	1.069961
H	2.386356	5.265507	1.639840
C	3.767123	4.263645	0.325225
H	3.206167	3.333309	0.309482

C	4.951837	4.377573	-0.401158
H	5.315881	3.536265	-0.984960
C	5.673700	5.567088	-0.376997
H	6.598651	5.632708	-0.945634
C	6.876617	9.905111	-3.253334
C	6.808584	11.203851	-3.792590
H	6.774311	12.060933	-3.127160
C	6.736034	11.406793	-5.163569
H	6.678059	12.423069	-5.544430
C	6.715967	10.326035	-6.045992
H	6.654275	10.487933	-7.117886
C	6.774214	9.039006	-5.525013
H	6.766265	8.180278	-6.191355
C	6.860653	8.825713	-4.150234
H	6.933114	7.808410	-3.782781
C	10.150369	7.738042	0.830440
C	11.252207	8.139716	1.599882
H	11.159717	8.971673	2.290084
C	12.469632	7.467901	1.506824
H	13.305005	7.802666	2.116290
C	12.616781	6.373180	0.662690
H	13.566035	5.850417	0.594998
C	11.524225	5.960450	-0.099763
H	11.620467	5.115480	-0.776463
C	10.312785	6.633629	-0.023914
H	9.486860	6.321185	-0.656839

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Table S24: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{II}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})_2]_{\text{bc}}^+$  ( $-3932.59357588 E_h$ ).

atom	x	y	z
Ni	5.602404	10.554358	1.053181
P	3.533552	10.961760	0.747854
P	5.058888	10.489920	3.218543
P	7.691059	10.932228	0.848326
P	6.091530	8.533864	0.228333
N	2.420618	9.722055	2.942744
N	4.001720	12.978756	2.611524
N	7.242885	10.073332	-1.769176
N	8.715952	8.390795	1.080260
C	2.403183	9.672272	1.494905
H	1.384813	9.891834	1.162349
H	2.690200	8.693083	1.077639
C	3.662894	9.296454	3.571995
H	3.964144	8.287995	3.237320
H	3.500800	9.255381	4.651983
C	2.997759	12.497122	1.678477
H	2.815816	13.268155	0.928201
H	2.041523	12.278514	2.182659
C	4.266630	12.116211	3.741259
H	3.368897	11.890523	4.341466
H	4.990779	12.614611	4.390008
C	6.915803	8.700671	-1.455041
H	7.786594	8.023961	-1.487705
H	6.185763	8.346913	-2.186855
C	8.251768	10.687896	-0.924538
H	8.484682	11.678806	-1.317280
H	9.183937	10.099181	-0.908721
C	7.449715	7.681686	1.193833
H	7.131141	7.586628	2.246939
H	7.586403	6.673705	0.794336
C	8.772095	9.684276	1.729612
H	9.803051	10.045684	1.677406
H	8.473420	9.665629	2.790334
C	2.872359	11.177987	-0.936999
C	3.757115	11.153676	-2.020853
H	4.819075	11.001331	-1.840617
C	3.282295	11.336380	-3.318335
H	3.980489	11.319378	-4.150089
C	1.923887	11.549439	-3.539168
H	1.553037	11.694340	-4.549658
C	1.037480	11.583376	-2.462858
H	-0.020893	11.757329	-2.633104
C	1.506468	11.397151	-1.166442
H	0.801873	11.434838	-0.339128
C	6.236508	10.156977	4.580581

C	7.230640	11.106847	4.861256
H	7.274307	12.037603	4.299467
C	8.177828	10.867211	5.851731
H	8.933210	11.617993	6.064688
C	8.158182	9.670257	6.566558
H	8.897527	9.484091	7.339652
C	7.185555	8.715302	6.285103
H	7.162670	7.780621	6.837762
C	6.229797	8.954565	5.299049
H	5.480306	8.193968	5.101300
C	1.238889	9.265827	3.603955
C	0.335674	8.386034	2.999174
H	0.532006	7.994511	2.005880
C	-0.817122	7.987605	3.674677
H	-1.508791	7.305716	3.188280
C	-1.074896	8.440904	4.963598
H	-1.971419	8.125036	5.487691
C	-0.170896	9.309315	5.574550
H	-0.364376	9.681190	6.576519
C	0.969733	9.725309	4.899744
H	1.650997	10.429176	5.370866
C	4.000309	14.380550	2.849612
C	3.835998	14.923530	4.128976
H	3.646650	14.276578	4.979516
C	3.887272	16.303407	4.322486
H	3.755831	16.703075	5.323980
C	4.081415	17.163731	3.247588
H	4.111868	18.237842	3.401076
C	4.233951	16.628515	1.968219
H	4.393428	17.285684	1.118031
C	4.203745	15.253894	1.769771
H	4.362084	14.847779	0.774088
C	8.391317	12.537740	1.350596
C	9.763625	12.805811	1.247037
H	10.449944	12.052870	0.866855
C	10.261438	14.050603	1.618703
H	11.324768	14.253629	1.532114
C	9.397104	15.034526	2.098674
H	9.790258	16.004532	2.388749
C	8.032295	14.776426	2.199978
H	7.351810	15.541760	2.561567
C	7.528650	13.532259	1.824971
H	6.461612	13.328631	1.888869
C	4.880037	7.188363	-0.046174
C	3.911237	7.342580	-1.049505
H	3.906656	8.233332	-1.674078
C	2.940397	6.366386	-1.250631
H	2.205716	6.494834	-2.040205
C	2.909561	5.230606	-0.442461
H	2.151398	4.469468	-0.600012

C	3.855782	5.076615	0.566657
H	3.839050	4.194564	1.200112
C	4.836050	6.047322	0.764787
H	5.565138	5.901109	1.556187
C	7.270564	10.441621	-3.140860
C	7.147222	11.796753	-3.487053
H	7.028644	12.542221	-2.705147
C	7.144293	12.189121	-4.819680
H	7.047533	13.243344	-5.063437
C	7.243789	11.241372	-5.838592
H	7.235111	11.550066	-6.879176
C	7.357282	9.896973	-5.502626
H	7.446950	9.145931	-6.282314
C	7.381422	9.496403	-4.167446
H	7.509777	8.444446	-3.933835
C	9.875370	7.566227	1.197878
C	10.799266	7.718031	2.236850
H	10.634259	8.456535	3.015434
C	11.931957	6.907167	2.294820
H	12.639512	7.041059	3.108082
C	12.149519	5.924130	1.335771
H	13.030597	5.292277	1.387020
C	11.224741	5.761422	0.304968
H	11.386304	5.005776	-0.458321
C	10.104183	6.578765	0.230423
H	9.408139	6.466448	-0.596891
H	5.636038	11.766621	0.220450

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Table S25: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Ph}}\text{HN}_2^{\text{Ph}})(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})]_{\text{bc}}^+$  ( $-3932.57800144 E_h$ ).

atom	x	y	z
Ni	5.630130	10.157136	1.351128
P	3.759033	11.084743	0.789641
P	4.969871	10.495811	3.400050
P	7.653357	10.770330	0.892198
P	6.095901	8.220653	0.542871
N	2.217235	10.361834	2.996530
N	4.483363	13.130971	2.551562
N	6.740565	9.924297	-1.592818
N	8.877080	8.271462	0.794635
C	2.276535	10.197823	1.562965
H	1.380329	10.654814	1.134010
H	2.305800	9.143534	1.238997
C	3.287730	9.744442	3.762359
H	3.359007	8.657512	3.583291
H	3.073656	9.876770	4.825503
C	3.476209	12.771051	1.557605
H	3.520893	13.517207	0.765027
H	2.459404	12.799339	1.981993
C	4.543708	12.300122	3.740355
H	3.611111	12.338230	4.330125
H	5.355362	12.683506	4.365947
C	6.586577	8.460293	-1.276254
H	7.524901	7.966159	-1.535777
H	5.782657	8.094925	-1.918507
C	7.952126	10.561910	-0.961244
H	8.058185	11.544569	-1.424620
H	8.817380	9.938152	-1.194696
C	7.713765	7.455837	1.121246
H	7.622708	7.278730	2.206587
H	7.833123	6.477850	0.648568
C	8.960326	9.547716	1.481126
H	9.943369	9.977230	1.274689
H	8.856745	9.465688	2.575602
C	3.046005	11.220574	-0.908266
C	2.718076	12.413709	-1.563548
H	2.861960	13.374202	-1.078933
C	2.183152	12.394690	-2.852370
H	1.929705	13.332416	-3.338969
C	1.955241	11.185484	-3.503913
H	1.525324	11.173744	-4.501017
C	2.277108	9.989419	-2.863640
H	2.100899	9.039925	-3.361114
C	2.825782	10.008265	-1.585290
H	3.081158	9.066587	-1.101977
C	5.835404	10.023264	4.951263

C	6.855868	9.071049	4.884842
H	7.123147	8.658709	3.915803
C	7.519126	8.653288	6.036607
H	8.307116	7.908568	5.969319
C	7.172947	9.194420	7.271498
H	7.691427	8.875981	8.171017
C	6.156114	10.144955	7.352319
H	5.880853	10.566985	8.314556
C	5.486844	10.551474	6.202175
H	4.688496	11.284661	6.287745
C	0.925952	10.302139	3.594150
C	-0.160755	9.683679	2.965429
H	-0.032778	9.194677	2.004926
C	-1.416478	9.674590	3.570806
H	-2.246196	9.190615	3.063316
C	-1.607524	10.261611	4.816367
H	-2.585729	10.249168	5.286646
C	-0.526433	10.871066	5.452292
H	-0.660557	11.345665	6.420155
C	0.722861	10.899169	4.846668
H	1.547366	11.409763	5.337038
C	4.603029	14.532272	2.773476
C	4.455490	15.112545	4.039292
H	4.190324	14.502858	4.895948
C	4.625797	16.485422	4.215751
H	4.503536	16.909683	5.208326
C	4.929690	17.308861	3.138203
H	5.054130	18.377962	3.278789
C	5.074728	16.739564	1.872817
H	5.326172	17.363823	1.019907
C	4.923472	15.371163	1.692794
H	5.089196	14.939526	0.709542
C	8.528771	12.357105	1.181272
C	7.832739	13.370371	1.845203
H	6.802067	13.193256	2.143653
C	8.444279	14.598728	2.093795
H	7.889661	15.378869	2.606462
C	9.752207	14.822394	1.674296
H	10.230171	15.778483	1.867423
C	10.451086	13.820379	0.999995
H	11.469714	13.996013	0.666120
C	9.845025	12.592662	0.756791
H	10.406442	11.824102	0.229472
C	5.083767	6.690883	0.375117
C	3.935590	6.566363	1.162454
H	3.670083	7.375916	1.836128
C	3.144042	5.421454	1.092679
H	2.258741	5.336573	1.716077
C	3.489561	4.390450	0.223856
H	2.873070	3.498454	0.164354

C	4.632653	4.501307	-0.567617
H	4.907600	3.696045	-1.242665
C	5.427815	5.639970	-0.487331
H	6.323613	5.700131	-1.102010
C	6.669121	10.205862	-3.036269
C	5.675601	11.053898	-3.506951
H	4.944224	11.486336	-2.829973
C	5.626567	11.337472	-4.870573
H	4.850312	11.996246	-5.245904
C	6.558834	10.779418	-5.739999
H	6.515468	11.004332	-6.801009
C	7.550567	9.931815	-5.248521
H	8.281507	9.496663	-5.922596
C	7.613100	9.640553	-3.889670
H	8.391345	8.986656	-3.508310
C	10.096619	7.535734	0.680105
C	11.183597	7.743906	1.535583
H	11.112554	8.458556	2.349309
C	12.364478	7.022792	1.364571
H	13.196763	7.201001	2.039397
C	12.475727	6.072455	0.355784
H	13.395176	5.509699	0.229367
C	11.391313	5.850832	-0.492133
H	11.463819	5.116514	-1.289287
C	10.218763	6.579374	-0.337616
H	9.389047	6.414233	-1.020991
H	5.935079	10.368307	-1.080122

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Table S26: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{III}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})_2]_{\text{bc}}^{2+}$  ( $-3932.28212730 E_h$ ).

atom	x	y	z
Ni	5.593820	10.592118	0.981012
P	3.411434	10.971150	0.659816
P	5.003143	10.530291	3.199587
P	7.797431	10.951342	0.854308
P	6.072449	8.461357	0.362263
N	2.358681	9.820212	2.916359
N	4.039245	13.015057	2.419485
N	7.128596	10.007717	-1.661454
N	8.687755	8.374873	1.205039
C	2.313064	9.701484	1.476282
H	1.290671	9.909055	1.146808
H	2.593613	8.708862	1.089637
C	3.587199	9.358854	3.536075
H	3.872600	8.345429	3.203065
H	3.434675	9.324403	4.618482
C	2.955899	12.543906	1.573063
H	2.744859	13.301827	0.816357
H	2.035978	12.353939	2.145333
C	4.274399	12.215033	3.601260
H	3.366675	12.042756	4.199030
H	5.005928	12.728175	4.229467
C	6.851326	8.620323	-1.326062
H	7.740082	7.970092	-1.364706
H	6.118108	8.235670	-2.039044
C	8.244909	10.595058	-0.920951
H	8.495923	11.555214	-1.375200
H	9.142550	9.956367	-0.944112
C	7.421513	7.693945	1.397930
H	7.105434	7.683234	2.455792
H	7.536236	6.656252	1.072908
C	8.785094	9.684076	1.807237
H	9.829698	10.005125	1.765798
H	8.465181	9.727514	2.860544
C	2.791722	11.137006	-1.034020
C	3.696468	11.066138	-2.100709
H	4.755032	10.902260	-1.909482
C	3.243003	11.216144	-3.409232
H	3.951704	11.165101	-4.230180
C	1.890435	11.440506	-3.655251
H	1.536752	11.559149	-4.674863
C	0.986745	11.519668	-2.595081
H	-0.065924	11.701456	-2.788326
C	1.430800	11.368160	-1.286015
H	0.714415	11.440106	-0.471413
C	6.208031	10.205354	4.520347

C	6.219067	8.985733	5.212942
H	5.474824	8.221345	5.009787
C	7.180413	8.745001	6.191732
H	7.171654	7.803527	6.732244
C	8.138565	9.712153	6.486506
H	8.879685	9.525373	7.257376
C	8.140271	10.923360	5.795057
H	8.882294	11.681788	6.025117
C	7.186507	11.169126	4.812321
H	7.211766	12.114737	4.275517
C	1.164754	9.429428	3.618662
C	0.295679	8.459110	3.115246
H	0.514771	7.953059	2.179359
C	-0.858104	8.123338	3.822160
H	-1.531056	7.372149	3.419930
C	-1.140220	8.731958	5.040944
H	-2.037003	8.464656	5.590486
C	-0.265328	9.690231	5.550728
H	-0.481696	10.177745	6.496379
C	0.875798	10.044890	4.840098
H	1.539323	10.814780	5.226480
C	4.125912	14.439701	2.572526
C	3.929455	15.068276	3.804202
H	3.660565	14.492141	4.684250
C	4.049538	16.453736	3.906895
H	3.892038	16.931133	4.869198
C	4.343665	17.222344	2.785457
H	4.424639	18.301387	2.868020
C	4.526861	16.596285	1.552444
H	4.758067	17.185991	0.670468
C	4.428795	15.213508	1.445734
H	4.598340	14.727999	0.487612
C	8.554796	12.546348	1.272862
C	7.773877	13.623189	1.706840
H	6.697287	13.514553	1.805732
C	8.369338	14.848238	1.997989
H	7.751338	15.677641	2.327883
C	9.745448	15.005740	1.854472
H	10.209635	15.961085	2.079259
C	10.529721	13.939066	1.415205
H	11.601427	14.062946	1.294647
C	9.941628	12.712726	1.126229
H	10.569010	11.896979	0.775459
C	4.806087	7.163818	0.200694
C	4.733881	6.100026	1.110052
H	5.454430	6.003577	1.916818
C	3.737301	5.134631	0.980511
H	3.696767	4.308108	1.683213
C	2.807829	5.221046	-0.052618
H	2.039615	4.461289	-0.157466

C	2.869577	6.281118	-0.956798
H	2.150310	6.349568	-1.767204
C	3.857458	7.252773	-0.829495
H	3.881745	8.078906	-1.536628
C	7.170482	10.304764	-3.069038
C	6.982743	11.633809	-3.469754
H	6.800654	12.401140	-2.721063
C	7.011919	11.972408	-4.817737
H	6.868571	13.007579	-5.112784
C	7.209826	10.988532	-5.786647
H	7.227278	11.252597	-6.839099
C	7.389643	9.667225	-5.391935
H	7.555826	8.893921	-6.135705
C	7.380380	9.322924	-4.040289
H	7.556909	8.289534	-3.759227
C	9.840767	7.526037	1.349956
C	10.692793	7.620654	2.452560
H	10.489712	8.332062	3.247999
C	11.807049	6.787998	2.545329
H	12.467034	6.872742	3.403332
C	12.065368	5.843688	1.556815
H	12.931120	5.194025	1.634805
C	11.206726	5.738926	0.463530
H	11.405151	5.011496	-0.317632
C	10.105890	6.581152	0.354513
H	9.456284	6.516793	-0.514908
H	5.467609	12.038993	1.195780

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Table S27: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Ph}}\text{HN}_2^{\text{Ph}})(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Ph}})]_{\text{bc}}^{2+}$  ( $-3932.29995204 E_h$ ).

atom	x	y	z
Ni	5.640008	10.127109	1.479972
P	3.589949	10.902566	0.865758
P	4.920161	10.618611	3.588262
P	7.809870	10.671755	1.056737
P	6.105048	8.213838	0.366704
N	2.183797	10.297033	3.160106
N	4.395258	13.080809	2.422443
N	6.840035	10.127163	-1.556539
N	8.870892	8.154536	0.675419
C	2.186876	10.012281	1.748430
H	1.258276	10.394421	1.315033
H	2.257242	8.939515	1.500053
C	3.285417	9.753616	3.929756
H	3.431336	8.671941	3.763821
H	3.065338	9.882394	4.992337
C	3.347199	12.641243	1.504969
H	3.365528	13.317323	0.649743
H	2.344591	12.701692	1.958344
C	4.411655	12.414477	3.714549
H	3.447581	12.476722	4.245253
H	5.174021	12.900528	4.330083
C	6.614907	8.644055	-1.386042
H	7.534875	8.141661	-1.690898
H	5.805700	8.387146	-2.072747
C	8.037859	10.669934	-0.811562
H	8.165069	11.698985	-1.153368
H	8.902794	10.072765	-1.106202
C	7.677935	7.379547	0.963921
H	7.551061	7.172544	2.040428
H	7.750868	6.414345	0.456523
C	9.046194	9.332863	1.495632
H	10.049090	9.727839	1.318269
H	8.952440	9.145279	2.577922
C	2.975064	10.878959	-0.860909
C	2.507350	12.002417	-1.555534
H	2.483008	12.981870	-1.089117
C	2.034658	11.877655	-2.861889
H	1.665581	12.756721	-3.381846
C	2.011217	10.633522	-3.487771
H	1.627827	10.539172	-4.499041
C	2.465996	9.507388	-2.803281
H	2.433436	8.531048	-3.277761
C	2.950892	9.629103	-1.504084
H	3.284781	8.737484	-0.976296
C	5.855574	10.277730	5.112747

C	6.898000	9.345709	5.067865
H	7.148495	8.870452	4.121916
C	7.604901	9.020133	6.222559
H	8.408326	8.290826	6.181343
C	7.280773	9.633146	7.430181
H	7.833617	9.384796	8.330847
C	6.243473	10.563489	7.485343
H	5.988248	11.038625	8.427546
C	5.527959	10.880684	6.335399
H	4.713899	11.598497	6.399100
C	0.907965	10.282219	3.814354
C	-0.168000	9.541614	3.318008
H	-0.053349	8.927717	2.429621
C	-1.400009	9.571944	3.969812
H	-2.227844	8.994199	3.569826
C	-1.566848	10.319040	5.130705
H	-2.525648	10.335840	5.638603
C	-0.491319	11.048473	5.635231
H	-0.611074	11.643408	6.535627
C	0.733368	11.038943	4.978479
H	1.556159	11.636261	5.363215
C	4.523670	14.510469	2.473470
C	4.239558	15.254228	3.621824
H	3.868973	14.769230	4.518921
C	4.410469	16.638863	3.622289
H	4.183737	17.201286	4.523028
C	4.846394	17.298934	2.478796
H	4.969599	18.377145	2.481556
C	5.123902	16.561145	1.328011
H	5.475208	17.062697	0.431096
C	4.974623	15.179741	1.327619
H	5.230934	14.606941	0.439538
C	8.630767	12.219122	1.540784
C	7.842355	13.218858	2.122053
H	6.777960	13.049054	2.264480
C	8.414597	14.435229	2.487809
H	7.793727	15.208432	2.929411
C	9.773216	14.655485	2.278736
H	10.221333	15.601018	2.568620
C	10.562667	13.665913	1.691445
H	11.620675	13.841472	1.522866
C	9.997248	12.451369	1.320337
H	10.629765	11.698508	0.857021
C	4.983966	6.796010	0.140702
C	4.010930	6.572500	1.123093
H	3.919187	7.264862	1.956151
C	3.172666	5.463029	1.044748
H	2.427215	5.291981	1.815236
C	3.293489	4.575227	-0.021395
H	2.638258	3.712150	-0.086058

C	4.261622	4.787908	-1.002712
H	4.361875	4.090269	-1.828469
C	5.110489	5.887052	-0.920140
H	5.873899	6.020472	-1.682955
C	6.866264	10.506806	-2.989100
C	5.803640	11.245882	-3.492110
H	4.967796	11.534797	-2.858598
C	5.822592	11.607393	-4.837774
H	5.001057	12.187653	-5.244598
C	6.885608	11.226811	-5.652307
H	6.895867	11.512769	-6.699084
C	7.938103	10.478540	-5.126836
H	8.766744	10.180818	-5.760957
C	7.936625	10.110301	-3.784591
H	8.761422	9.530638	-3.381043
C	10.050386	7.362602	0.444348
C	11.130267	7.362578	1.329922
H	11.096367	7.946045	2.245070
C	12.260957	6.594909	1.053507
H	13.094390	6.606057	1.749230
C	12.317482	5.806508	-0.090856
H	13.196686	5.205411	-0.298537
C	11.234579	5.793150	-0.968508
H	11.268613	5.184244	-1.866839
C	10.113219	6.572463	-0.708323
H	9.280815	6.568517	-1.408836
H	6.013664	10.574290	-1.123841

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Table S28: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{II}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})_2]^{2+}$  ( $-4089.53436066 E_h$ ).

atom	x	y	z
Ni	1.219105	19.451572	9.588450
N	2.772307	21.542952	6.813219
N	2.383418	22.473176	10.349603
N	0.080488	15.958709	10.666339
N	-0.882778	19.250498	11.988067
P	3.268456	20.152017	9.180466
P	0.650863	21.275796	8.466346
P	1.593660	18.117249	11.313537
P	-0.673136	18.323003	9.439447
C	4.584130	17.749356	8.955797
H	3.671406	17.424455	8.459050
C	5.661358	16.874253	9.049045
H	5.587379	15.876780	8.626338
C	6.836967	17.284195	9.675956
H	7.680509	16.604379	9.745539
C	6.932074	18.568450	10.205988
H	7.848051	18.890644	10.691384
C	5.859755	19.452321	10.109031
H	5.960700	20.450807	10.521501
C	4.676957	19.045397	9.484738
C	3.482514	20.400261	7.332119
H	4.556858	20.557724	7.187270
H	3.229225	19.444920	6.833974
C	3.333970	22.020010	5.530884
H	3.160888	21.282316	4.728693
H	4.418381	22.090146	5.673506
C	1.992522	23.490787	3.978645
H	1.779715	22.614589	3.369765
C	1.496198	24.736642	3.596999
H	0.893498	24.826489	2.698280
C	1.783369	25.863647	4.361578
H	1.404024	26.835845	4.062025
C	2.572429	25.744492	5.506033
H	2.812266	26.625599	6.093881
C	3.069841	24.501341	5.882147
H	3.705385	24.413579	6.760516
C	2.782878	23.360815	5.122828
C	1.337247	21.371425	6.749990
H	1.019661	20.482717	6.174976
H	0.914363	22.257642	6.263859
C	-1.773996	21.956445	7.171733
H	-1.276193	21.805358	6.219512
C	-3.104746	22.369029	7.181220
H	-3.618382	22.541079	6.240521
C	-3.768987	22.572886	8.387652

H	-4.801571	22.908264	8.389166
C	-3.105374	22.353294	9.595068
H	-3.619600	22.522378	10.536735
C	-1.779030	21.934422	9.594610
H	-1.264495	21.759951	10.538193
C	-1.102564	21.743791	8.379555
C	1.441257	22.764626	9.278245
H	1.878468	23.366426	8.461445
H	0.612197	23.333011	9.708571
C	2.707447	23.742516	11.041913
H	3.274307	24.409047	10.367899
H	1.752092	24.238194	11.244158
C	2.768079	23.229064	13.507372
H	1.698398	23.036315	13.460431
C	3.430488	23.168523	14.730056
H	2.880832	22.920079	15.633301
C	4.794557	23.456683	14.800372
H	5.307408	23.431408	15.757279
C	5.490117	23.800770	13.643754
H	6.547472	24.042727	13.695021
C	4.820996	23.867130	12.422491
H	5.360300	24.177521	11.529856
C	3.454772	23.583283	12.340163
C	3.607552	21.821739	9.901374
H	4.244434	21.687194	10.781401
H	4.167476	22.416751	9.156367
C	3.993573	17.295606	12.555036
H	3.832625	16.306890	12.139116
C	5.114751	17.519050	13.350611
H	5.800718	16.701668	13.550199
C	5.349696	18.779491	13.892961
H	6.220763	18.945977	14.519520
C	4.467097	19.827914	13.633959
H	4.643410	20.814094	14.052331
C	3.345867	19.612654	12.839936
H	2.667612	20.440844	12.640219
C	3.098449	18.340923	12.302955
C	1.447413	16.299620	10.988141
H	2.174823	16.004321	10.209113
H	1.712405	15.775960	11.913217
C	-0.190383	14.513563	10.817232
H	0.425803	13.923178	10.117657
H	-1.237053	14.362728	10.528467
C	-0.763292	14.524007	13.271840
H	-1.543781	15.251899	13.062315
C	-0.571644	14.072419	14.573207
H	-1.198843	14.455786	15.372326
C	0.409991	13.119550	14.846600
H	0.551793	12.759761	15.861227
C	1.199110	12.621795	13.813744



H	1.957429	11.872186	14.018913
C	1.010055	13.079646	12.510484
H	1.619961	12.679247	11.703446
C	0.027788	14.031282	12.227098
C	-0.371273	16.476670	9.399811
H	-1.340930	16.027619	9.158932
H	0.304592	16.259359	8.550695
C	-1.093839	18.373575	6.716798
H	-0.013223	18.255329	6.657240
C	-1.847288	18.388780	5.547092
H	-1.355875	18.282507	4.584610
C	-3.232674	18.528423	5.614346
H	-3.822401	18.536104	4.702907
C	-3.861291	18.645839	6.851684
H	-4.940901	18.745697	6.906567
C	-3.113084	18.623350	8.026174
H	-3.626779	18.700866	8.979066
C	-1.722024	18.494101	7.965118
C	-1.683547	18.593803	10.965066
H	-2.492280	19.278973	10.690650
H	-2.128931	17.637806	11.284701
C	-1.729076	19.803112	13.072993
H	-2.439848	20.487411	12.594486
H	-1.070732	20.412169	13.702608
C	-1.904824	18.304362	15.103082
H	-0.942664	18.684925	15.439725
C	-2.575744	17.359278	15.875847
H	-2.133080	17.007132	16.803028
C	-3.823323	16.887060	15.472725
H	-4.353443	16.159535	16.079842
C	-4.401553	17.372848	14.301437
H	-5.384617	17.026897	13.996242
C	-3.729810	18.322080	13.535610
H	-4.206631	18.722867	12.643228
C	-2.469046	18.793335	13.920190
C	0.198982	18.436465	12.521864
H	-0.124653	17.454599	12.904548
H	0.653942	18.986742	13.350650

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Table S29: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})_2]_{\text{bc}}^+$  ( $-4089.82226185 E_h$ ).

atom	x	y	z
C	9.383923	5.957045	8.474362
H	9.266597	5.025688	9.038604
H	10.191307	5.794986	7.737056
C	10.056755	8.284594	8.819163
H	10.887014	8.170409	8.097376
H	10.398649	8.937694	9.628929
C	10.639701	6.590393	10.437848
H	11.645640	6.457887	9.999175
H	10.315863	5.601783	10.785330
C	10.718604	7.530013	11.614562
C	11.925126	8.140097	11.962537
H	12.815557	7.942390	11.369483
C	12.001354	8.989215	13.065721
H	12.948497	9.452306	13.327034
C	10.865253	9.241008	13.829102
H	10.922178	9.901392	14.689541
C	9.654648	8.637974	13.487903
H	8.763484	8.823596	14.080237
C	9.582426	7.787028	12.390220
H	8.638035	7.313119	12.134657
C	7.310751	9.048372	9.288497
H	7.794515	8.838881	10.255943
H	6.869213	10.048949	9.347960
C	6.628976	6.712093	8.925424
H	5.723134	6.125772	8.738540
H	7.068260	6.364249	9.874979
C	5.033719	8.342993	9.757912
H	4.248371	7.700526	9.342103
H	4.726642	9.381709	9.583387
C	5.164107	8.098551	11.246542
C	4.946187	6.826256	11.787132
H	4.635784	6.012361	11.135341
C	5.094932	6.593889	13.152115
H	4.913037	5.601706	13.555225
C	5.456931	7.637174	14.002026
H	5.560713	7.460417	15.068590
C	5.663646	8.912313	13.479548
H	5.922278	9.736466	14.138566
C	5.518980	9.137762	12.112540
H	5.665640	10.140131	11.715984
C	7.254635	4.620679	7.004973
C	6.816229	4.462500	5.683915
H	6.833958	5.316573	5.005914
C	6.350170	3.223706	5.245820
H	6.006101	3.106945	4.222007

C	6.318067	2.138860	6.118307
H	5.955220	1.174503	5.774907
C	6.747217	2.291564	7.436206
H	6.717823	1.448245	8.120003
C	7.211776	3.526113	7.878940
H	7.533698	3.630953	8.912190
C	9.118336	10.899488	7.961799
C	10.430558	11.324257	8.201854
H	11.207691	10.609473	8.454122
C	10.760238	12.676400	8.125268
H	11.781303	12.990730	8.321226
C	9.787217	13.620034	7.809791
H	10.045535	14.673453	7.757341
C	8.479369	13.206242	7.559856
H	7.714219	13.935582	7.310021
C	8.148829	11.856788	7.625502
H	7.128574	11.546983	7.408722
C	8.532520	6.867931	2.900538
H	8.920285	7.393704	2.013153
H	8.790723	5.808351	2.800722
C	10.210614	9.059513	3.721005
H	10.706775	9.607289	4.543402
H	10.983565	8.760822	3.003822
C	6.371833	6.101531	2.087656
H	5.320225	6.085120	2.398249
H	6.762684	5.084665	2.217777
C	6.463654	6.502273	0.630307
C	7.536867	6.085275	-0.165499
H	8.294364	5.427594	0.255292
C	7.635008	6.479645	-1.497428
H	8.470860	6.139725	-2.102188
C	6.651851	7.292460	-2.058779
H	6.719965	7.589192	-3.101340
C	5.570510	7.703611	-1.282111
H	4.787532	8.317443	-1.718677
C	5.480880	7.311472	0.051621
H	4.624820	7.622859	0.646485
C	8.253309	10.517291	3.860453
H	7.700016	11.230678	3.242674
H	8.707859	11.081332	4.695264
C	6.545912	8.311493	3.073181
H	5.455420	8.225953	3.127469
H	6.793278	8.898843	2.173881
C	9.887401	10.825849	2.104450
H	10.649380	10.267831	1.546779
H	10.418506	11.614413	2.668109
C	8.922916	11.459304	1.133324
C	8.197755	10.664157	0.238363
H	8.329952	9.585156	0.247771
C	7.315711	11.244676	-0.667184

H	6.764275	10.612059	-1.356508
C	7.149618	12.629578	-0.694623
H	6.465810	13.082822	-1.406534
C	7.868944	13.428894	0.189081
H	7.748349	14.508240	0.171343
C	8.747871	12.844036	1.099921
H	9.309310	13.471222	1.788984
C	10.869921	6.354728	4.555545
C	12.196240	6.803553	4.577575
H	12.427114	7.857538	4.455401
C	13.244529	5.902696	4.757028
H	14.268119	6.266020	4.764722
C	12.983929	4.545003	4.917424
H	13.802625	3.844324	5.051022
C	11.666280	4.089238	4.908431
H	11.453730	3.031822	5.036781
C	10.617066	4.986514	4.738258
H	9.593598	4.617814	4.753477
C	5.562828	10.299151	4.941723
C	5.027508	11.203623	4.014508
H	5.503915	11.353459	3.048756
C	3.871134	11.915258	4.319064
H	3.462278	12.615093	3.596079
C	3.238004	11.728719	5.547364
H	2.335244	12.285267	5.781768
C	3.760417	10.825996	6.470159
H	3.261267	10.673951	7.423049
C	4.919678	10.111392	6.171911
H	5.328208	9.397343	6.887726
N	9.667477	7.020629	9.421296
N	6.244586	8.115992	8.948168
N	7.087716	6.958897	3.049950
N	9.236479	9.867516	3.010655
Ni	8.061364	7.997053	6.109929
P	7.805741	6.282232	7.529053
P	8.627787	9.135830	7.961570
P	9.444159	7.494332	4.412106
P	7.046899	9.294439	4.588550

Table S30: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})_2]_{\text{bc}}$  ( $-4089.98792395 E_h$ ).

atom	x	y	z
C	8.983311	5.857517	8.396502
H	8.696064	4.958130	8.954487
H	9.759542	5.563220	7.665626
C	10.041283	8.045605	8.793026
H	10.870750	7.817563	8.096905
H	10.443924	8.634437	9.625040
C	10.406061	6.213623	10.300863
H	11.369964	5.975203	9.810308
H	9.968726	5.254049	10.605957
C	10.663539	7.043922	11.535623
C	11.965833	7.345644	11.937176
H	12.802613	7.004160	11.331399
C	12.203482	8.075981	13.100683
H	13.223979	8.301992	13.398160
C	11.134664	8.519337	13.874172
H	11.316301	9.091362	14.780038
C	9.829453	8.227305	13.478654
H	8.986307	8.568511	14.072730
C	9.595979	7.494056	12.319999
H	8.577896	7.270535	12.013001
C	7.475379	9.206533	9.236645
H	7.947752	8.943704	10.198014
H	7.189788	10.263348	9.288222
C	6.412815	7.002699	8.865165
H	5.417793	6.587642	8.665689
H	6.789046	6.546511	9.797449
C	5.145533	8.844635	9.817638
H	4.249459	8.341762	9.431419
H	4.997750	9.923492	9.676947
C	5.287414	8.550968	11.298651
C	4.889555	7.318462	11.828584
H	4.436841	6.578077	11.172722
C	5.049480	7.034333	13.182532
H	4.730122	6.072618	13.575050
C	5.605738	7.986255	14.034718
H	5.723224	7.769910	15.092945
C	5.997156	9.222065	13.523400
H	6.416298	9.977420	14.183000
C	5.839549	9.497983	12.167014
H	6.138925	10.468278	11.776994
C	6.729456	4.852269	6.980451
C	6.490077	4.597899	5.625897
H	6.762480	5.359781	4.892991
C	5.905760	3.395056	5.228699
H	5.723897	3.204974	4.173851

C	5.548953	2.440904	6.177923
H	5.091126	1.505464	5.867098
C	5.778306	2.688821	7.531071
H	5.498643	1.948092	8.275899
C	6.367621	3.885581	7.928291
H	6.541811	4.066134	8.987156
C	9.516047	10.749286	7.952850
C	10.875117	10.981587	8.197384
H	11.541165	10.154420	8.424352
C	11.397545	12.273840	8.153422
H	12.454990	12.431265	8.349932
C	10.572254	13.357652	7.869595
H	10.979892	14.364312	7.840826
C	9.217660	13.139964	7.616851
H	8.563808	13.977687	7.388546
C	8.699999	11.849984	7.646981
H	7.647600	11.689819	7.419476
C	8.782900	6.966924	2.994678
H	9.136121	7.536682	2.118532
H	9.207626	5.958903	2.928568
C	10.055777	9.380943	3.791175
H	10.459227	10.021407	4.597853
H	10.865073	9.182432	3.077063
C	6.827198	5.882128	2.041748
H	5.774318	5.689948	2.286405
H	7.369784	4.937796	2.180777
C	6.929036	6.311129	0.590752
C	8.092051	6.075226	-0.151012
H	8.920482	5.542762	0.310743
C	8.197137	6.498144	-1.473688
H	9.107663	6.302054	-2.033453
C	7.132205	7.159784	-2.082204
H	7.208982	7.482082	-3.116945
C	5.963809	7.392616	-1.359407
H	5.121538	7.892776	-1.830368
C	5.867415	6.972478	-0.034610
H	4.949043	7.148938	0.521134
C	7.858183	10.482797	3.872692
H	7.195500	11.063601	3.222699
H	8.186338	11.143911	4.696735
C	6.562395	8.056261	3.089316
H	5.504289	7.771776	3.132420
H	6.703869	8.688595	2.196248
C	9.498106	11.124106	2.234952
H	10.398605	10.749037	1.731043
H	9.821344	11.981458	2.856923
C	8.519994	11.604146	1.189201
C	7.946555	10.698871	0.289365
H	8.189226	9.642691	0.368500
C	7.068688	11.140109	-0.695534

H	6.634223	10.420268	-1.383546
C	6.753487	12.494955	-0.799641
H	6.069223	12.839380	-1.570381
C	7.319634	13.403381	0.090049
H	7.077180	14.460373	0.019913
C	8.194461	12.956985	1.079589
H	8.629862	13.667925	1.778527
C	11.101362	6.852706	4.678367
C	12.344467	7.496551	4.726992
H	12.407785	8.575282	4.616485
C	13.518446	6.770208	4.923272
H	14.472144	7.290907	4.954647
C	13.473935	5.387167	5.071291
H	14.389719	4.821820	5.219576
C	12.242078	4.733638	5.034278
H	12.193545	3.654552	5.155240
C	11.070540	5.459806	4.851258
H	10.114009	4.940455	4.853410
C	5.256362	9.900006	4.863765
C	4.562391	10.524586	3.818487
H	4.985747	10.547340	2.816515
C	3.325893	11.121003	4.048370
H	2.795357	11.600585	3.229783
C	2.770985	11.105550	5.327809
H	1.806521	11.573518	5.507266
C	3.454859	10.489327	6.372541
H	3.022295	10.474185	7.369848
C	4.691404	9.885705	6.143455
H	5.233424	9.390398	6.951185
N	9.467359	6.833215	9.365349
N	6.264778	8.447561	8.959555
N	7.336251	6.822981	3.042765
N	8.978545	10.030493	3.055933
Ni	8.077396	8.064331	6.113749
P	7.499597	6.472175	7.417948
P	8.768193	9.066209	7.869574
P	9.499444	7.757415	4.552666
P	6.889282	9.069896	4.644255

Table S31: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{II}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})_2]_{\text{bc}}^+$  ( $-4090.42875865 E_h$ ).

atom	x	y	z
C	9.178843	5.938173	8.307908
H	9.049431	5.018250	8.889009
H	9.921374	5.726817	7.518063
C	9.967632	8.235086	8.570064
H	10.746411	8.080185	7.799581
H	10.376828	8.896371	9.341160
C	10.624038	6.528782	10.148412
H	11.579375	6.352270	9.621044
H	10.292873	5.556035	10.532576
C	10.847848	7.470863	11.304360
C	12.106564	8.026935	11.539412
H	12.931006	7.786693	10.871416
C	12.318586	8.875904	12.624775
H	13.305004	9.296540	12.797622
C	11.267142	9.181920	13.483667
H	11.429748	9.842084	14.330697
C	10.005147	8.633584	13.255163
H	9.179446	8.861769	13.922707
C	9.797366	7.782724	12.174816
H	8.813424	7.352087	12.006891
C	7.305842	9.083834	9.232461
H	7.872486	8.863704	10.151444
H	6.917729	10.104272	9.317712
C	6.487185	6.783022	8.948222
H	5.546203	6.238678	8.812528
H	6.975711	6.399581	9.859747
C	5.055294	8.479465	9.932397
H	4.205777	7.873666	9.594392
H	4.780620	9.531402	9.784880
C	5.313888	8.225926	11.402641
C	5.082782	6.965380	11.964622
H	4.669831	6.170454	11.347049
C	5.347980	6.722346	13.309994
H	5.153979	5.740024	13.731205
C	5.842302	7.743605	14.118951
H	6.037360	7.559368	15.171355
C	6.064476	9.007493	13.575786
H	6.426254	9.815719	14.205346
C	5.803113	9.243371	12.228024
H	5.963721	10.237558	11.816497
C	6.941551	4.686616	6.961883
C	6.519476	4.507398	5.639418
H	6.566237	5.345947	4.944335
C	6.034240	3.267572	5.226203
H	5.704144	3.132260	4.200125



C	5.962399	2.206592	6.125342
H	5.581247	1.242761	5.800805
C	6.375327	2.382246	7.445487
H	6.314827	1.558231	8.150320
C	6.864927	3.615659	7.862939
H	7.177269	3.739201	8.897196
C	9.035057	10.833696	7.699093
C	10.383234	11.208958	7.744882
H	11.160145	10.464776	7.891742
C	10.749508	12.547212	7.611087
H	11.799182	12.822701	7.657347
C	9.777604	13.526965	7.430945
H	10.064881	14.569588	7.333707
C	8.432811	13.162846	7.376675
H	7.667567	13.920621	7.235318
C	8.064336	11.827108	7.500863
H	7.012482	11.556879	7.436102
C	8.417592	6.876302	2.938027
H	8.862533	7.417160	2.087426
H	8.695015	5.821238	2.843016
C	10.017263	9.063746	3.887861
H	10.466337	9.604773	4.741996
H	10.821401	8.784788	3.198776
C	6.327945	6.116348	1.949794
H	5.251904	6.113707	2.162076
H	6.689014	5.091149	2.099274
C	6.566730	6.532168	0.513524
C	7.643232	6.011701	-0.211733
H	8.293348	5.271542	0.249823
C	7.879853	6.409869	-1.525490
H	8.710017	5.979861	-2.079017
C	7.041067	7.341368	-2.134229
H	7.218579	7.647263	-3.161094
C	5.960179	7.862694	-1.426001
H	5.290806	8.574437	-1.900733
C	5.724757	7.456471	-0.114909
H	4.861859	7.846516	0.420906
C	8.035074	10.488826	3.963917
H	7.474747	11.190487	3.335893
H	8.421878	11.057594	4.828096
C	6.405656	8.285722	3.037808
H	5.314333	8.189948	3.040153
H	6.691880	8.914563	2.177986
C	9.768664	10.897738	2.335212
H	10.379613	11.568602	2.966550
H	8.991002	11.520418	1.875863
C	10.628574	10.295131	1.252955
C	11.997181	10.564168	1.194819
H	12.448732	11.206942	1.947529
C	12.785977	10.026791	0.178579

H	13.848516	10.249570	0.143744
C	12.210881	9.207230	-0.788106
H	12.822872	8.788035	-1.581489
C	10.844759	8.930469	-0.736734
H	10.385291	8.296611	-1.489467
C	10.058929	9.472144	0.274852
H	8.993277	9.259299	0.304563
C	10.634573	6.364776	4.736033
C	11.949413	6.826654	4.871410
H	12.174550	7.885987	4.793165
C	12.993016	5.931434	5.100440
H	14.008608	6.304996	5.194660
C	12.739227	4.566371	5.196553
H	13.554681	3.870275	5.368406
C	11.432419	4.097012	5.070169
H	11.225002	3.033421	5.144690
C	10.387294	4.988503	4.850323
H	9.370620	4.608823	4.773982
C	5.341888	10.189381	4.982546
C	4.778687	11.058528	4.037998
H	5.228907	11.174337	3.054947
C	3.626664	11.775533	4.344599
H	3.193501	12.443745	3.606146
C	3.030132	11.634275	5.596924
H	2.131283	12.195460	5.835062
C	3.583948	10.770680	6.538597
H	3.113507	10.654583	7.510871
C	4.735538	10.045713	6.235883
H	5.166697	9.357776	6.963402
N	9.582457	6.993129	9.220487
N	6.171750	8.199406	9.013892
N	6.965448	6.946308	2.985862
N	9.075226	9.887871	3.148182
Ni	7.665407	7.916706	6.073309
P	7.553226	6.325477	7.479642
P	8.500039	9.084324	7.786413
P	9.213942	7.499568	4.517448
P	6.835902	9.212570	4.605570
H	6.264843	7.480474	5.985145

Table S32: Cartesian coordinates of the optimized structure of  $[\text{Ni}^0(\text{P}_2^{\text{Ph}}\text{HN}_2^{\text{Bz}})(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})]_{\text{bc}}^+$  ( $-4090.42295786 E_h$ ).

atom	x	y	z
C	9.208109	5.724307	8.491540
H	8.879683	4.843641	9.054008
H	10.027153	5.403627	7.822934
C	10.324239	7.874108	8.861482
H	11.162641	7.552939	8.217161
H	10.731994	8.470882	9.684234
C	10.396777	6.160361	10.556773
H	11.403034	5.850154	10.222643
H	9.874210	5.247750	10.867977
C	10.515280	7.092112	11.737260
C	11.761443	7.536247	12.182225
H	12.661240	7.208662	11.666155
C	11.863835	8.385619	13.283361
H	12.841454	8.718550	13.619614
C	10.715843	8.804335	13.949230
H	10.793760	9.464469	14.808111
C	9.465183	8.366364	13.513438
H	8.564767	8.680871	14.033467
C	9.367866	7.515416	12.417688
H	8.391987	7.162358	12.091986
C	7.807978	9.175810	9.120505
H	8.073731	8.788475	10.105314
H	7.560201	10.236855	9.192892
C	6.658483	6.978124	8.758564
H	5.652661	6.589491	8.584654
H	6.987067	6.735912	9.769903
C	5.294857	9.061612	9.136950
H	4.496792	8.564033	8.579403
H	5.306667	10.110576	8.826934
C	5.107164	8.925524	10.621064
C	4.462580	7.804819	11.156514
H	4.078946	7.033315	10.493207
C	4.287376	7.677334	12.531233
H	3.779536	6.805510	12.932496
C	4.747251	8.675350	13.388016
H	4.601842	8.581146	14.459973
C	5.374597	9.804022	12.864657
H	5.714899	10.595457	13.525642
C	5.550262	9.928652	11.489452
H	6.022450	10.822191	11.088585
C	6.961584	4.738184	6.979601
C	6.666902	4.508335	5.631024
H	6.943921	5.262569	4.891670
C	6.020652	3.332832	5.248800
H	5.796910	3.155483	4.200475

C	5.660463	2.387442	6.205241
H	5.157244	1.472868	5.904927
C	5.946030	2.613583	7.551828
H	5.664809	1.877520	8.299588
C	6.596242	3.781537	7.937039
H	6.815006	3.942038	8.990849
C	10.001088	10.593948	7.932801
C	11.364351	10.744681	8.212544
H	11.978673	9.881245	8.449260
C	11.956828	12.006398	8.188888
H	13.015565	12.105313	8.410527
C	11.197695	13.134448	7.892811
H	11.660309	14.116723	7.881564
C	9.840203	12.995835	7.605215
H	9.241344	13.870870	7.368714
C	9.252034	11.735882	7.612318
H	8.197863	11.638236	7.357474
C	8.809346	6.942603	2.963426
H	9.081586	7.531608	2.072339
H	9.246646	5.944716	2.848924
C	10.084277	9.381057	3.723976
H	10.533641	10.008807	4.515588
H	10.841607	9.223188	2.946984
C	6.804148	5.855685	2.124775
H	5.772898	5.644258	2.435758
H	7.364213	4.915032	2.202016
C	6.805451	6.330345	0.686404
C	7.918104	6.125852	-0.137507
H	8.779491	5.585356	0.248660
C	7.928153	6.583853	-1.452558
H	8.798197	6.409964	-2.079307
C	6.816693	7.247213	-1.969084
H	6.818098	7.593212	-2.998631
C	5.696919	7.446243	-1.164065
H	4.817590	7.942379	-1.565594
C	5.695382	6.992441	0.153016
H	4.811634	7.139073	0.770356
C	7.883490	10.446843	3.995517
H	7.159883	11.015613	3.401329
H	8.269213	11.105530	4.796105
C	6.597153	7.999461	3.243012
H	5.542939	7.712596	3.323780
H	6.693043	8.665908	2.370477
C	9.364161	11.111842	2.212598
H	10.197651	10.729984	1.610209
H	9.761878	11.960492	2.800324
C	8.273355	11.602103	1.293465
C	7.658193	10.723960	0.394124
H	7.959320	9.679719	0.370897
C	6.669586	11.179229	-0.471929

H	6.205566	10.483837	-1.165147
C	6.284394	12.519623	-0.455443
H	5.516063	12.874985	-1.136106
C	6.892290	13.401155	0.433575
H	6.600410	14.447388	0.450803
C	7.878860	12.941251	1.304568
H	8.354589	13.632796	1.996822
C	11.244585	6.844107	4.516362
C	12.477429	7.503205	4.447521
H	12.522021	8.578423	4.301715
C	13.669711	6.789936	4.566637
H	14.617930	7.316643	4.505688
C	13.648237	5.410978	4.752009
H	14.578151	4.856619	4.837053
C	12.425283	4.744738	4.829162
H	12.399144	3.668400	4.974577
C	11.235320	5.456347	4.723146
H	10.286812	4.928551	4.804277
C	5.365011	9.702891	5.222107
C	5.149241	11.077008	5.388893
H	5.940315	11.788059	5.168592
C	3.914145	11.556030	5.826238
H	3.763924	12.625982	5.940568
C	2.872323	10.673211	6.096351
H	1.907877	11.049093	6.424540
C	3.073404	9.302063	5.933280
H	2.262114	8.605765	6.128334
C	4.310167	8.822242	5.512756
H	4.458322	7.749058	5.403834
N	9.620605	6.740890	9.449401
N	6.583693	8.469112	8.623033
N	7.372585	6.772907	3.125689
N	8.936567	10.013025	3.089010
Ni	8.310200	7.974302	6.147450
P	7.802847	6.314801	7.397666
P	9.183791	8.950572	7.839183
P	9.633706	7.721629	4.461112
P	7.007495	8.993730	4.784953
H	6.644248	8.608989	7.567969

Table S33: Cartesian coordinates of the optimized structure of  $[\text{HNi}^{\text{III}}(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})_2]_{\text{bc}}^{2+}$  ( $-4090.12410979 E_h$ ).

atom	x	y	z
C	9.332350	5.829006	8.239984
H	9.234069	4.890910	8.797167
H	10.066787	5.656530	7.435528
C	10.104036	8.115068	8.532125
H	10.859789	7.966306	7.739639
H	10.538517	8.761313	9.303213
C	10.674621	6.423741	10.180333
H	11.657388	6.224270	9.720440
H	10.301215	5.466853	10.563478
C	10.825642	7.402044	11.315404
C	12.046890	8.036838	11.552622
H	12.900056	7.816897	10.914328
C	12.188122	8.931385	12.612880
H	13.146439	9.408968	12.793340
C	11.104080	9.203305	13.442284
H	11.213926	9.894958	14.272046
C	9.879920	8.574507	13.213157
H	9.033395	8.771071	13.864106
C	9.742454	7.678210	12.158616
H	8.791450	7.173951	12.001727
C	7.395780	8.949302	9.227192
H	7.947609	8.700732	10.145772
H	6.997240	9.961907	9.339083
C	6.629352	6.643386	8.948203
H	5.703775	6.066552	8.859252
H	7.156506	6.321794	9.857548
C	5.090297	8.352363	9.776608
H	4.279859	7.723097	9.392015
H	4.819782	9.394353	9.568777
C	5.256236	8.148983	11.264800
C	5.012198	6.899936	11.846927
H	4.652141	6.077628	11.231967
C	5.189138	6.705369	13.214259
H	4.982473	5.734333	13.654190
C	5.605510	7.762985	14.020847
H	5.727589	7.616342	15.089718
C	5.837156	9.015408	13.455537
H	6.132530	9.850839	14.083550
C	5.663764	9.204702	12.086460
H	5.822325	10.192726	11.659006
C	7.046888	4.579708	6.877752
C	6.519477	4.489139	5.582961
H	6.515349	5.365365	4.935359
C	5.996530	3.279202	5.131300
H	5.581572	3.209053	4.130286

C	5.996094	2.162346	5.963587
H	5.586299	1.221508	5.609262
C	6.516324	2.250428	7.254671
H	6.511424	1.381185	7.904967
C	7.040593	3.453662	7.714358
H	7.433247	3.507444	8.726593
C	9.142474	10.754728	7.706807
C	10.495124	11.122406	7.685071
H	11.276849	10.375735	7.787717
C	10.854522	12.461826	7.553148
H	11.904475	12.738152	7.552960
C	9.873392	13.443653	7.438580
H	10.156964	14.487347	7.345489
C	8.525778	13.084800	7.450011
H	7.757758	13.847584	7.365614
C	8.159633	11.748577	7.576124
H	7.104149	11.485611	7.572583
C	8.450312	7.005958	2.985813
H	8.828291	7.556563	2.110761
H	8.744624	5.958160	2.873484
C	10.052884	9.245871	3.937889
H	10.473523	9.788626	4.803822
H	10.873597	9.002682	3.254190
C	6.324191	6.140859	2.176815
H	5.260773	6.139473	2.442359
H	6.709582	5.134348	2.378582
C	6.498783	6.475567	0.713498
C	7.548926	5.919597	-0.024801
H	8.223365	5.207727	0.446632
C	7.722167	6.238695	-1.369483
H	8.528691	5.780043	-1.934128
C	6.845223	7.122859	-1.994975
H	6.970910	7.363127	-3.046340
C	5.788516	7.675646	-1.273884
H	5.086735	8.344220	-1.763496
C	5.615680	7.349689	0.068776
H	4.765329	7.756510	0.612284
C	8.059811	10.634824	4.026906
H	7.507691	11.359483	3.419275
H	8.451357	11.178188	4.903899
C	6.425866	8.389613	3.093183
H	5.336160	8.288732	3.081845
H	6.726737	8.968331	2.205634
C	9.712769	10.991230	2.281471
H	10.323791	11.721629	2.838645
H	8.896907	11.551146	1.809681
C	10.548799	10.322680	1.222278
C	11.927815	10.535917	1.162727
H	12.401432	11.196250	1.886186
C	12.698155	9.929261	0.171370

H	13.767482	10.113065	0.129201
C	12.093924	9.097483	-0.766714
H	12.691085	8.629065	-1.543210
C	10.717131	8.877974	-0.714995
H	10.236711	8.241257	-1.451712
C	9.949413	9.488289	0.271065
H	8.873448	9.330096	0.290529
C	10.739730	6.523435	4.765518
C	12.035607	7.022841	4.947499
H	12.233146	8.089765	4.904641
C	13.098197	6.148510	5.168579
H	14.101061	6.544481	5.296004
C	12.878897	4.774275	5.209393
H	13.710320	4.095417	5.371462
C	11.590120	4.270623	5.035092
H	11.415244	3.199356	5.061039
C	10.523655	5.138366	4.821627
H	9.522763	4.729967	4.699536
C	5.321369	10.345972	4.990963
C	4.802938	11.216590	4.019206
H	5.291142	11.329725	3.054463
C	3.642165	11.937947	4.275476
H	3.244680	12.605782	3.517557
C	2.990531	11.801405	5.500695
H	2.084685	12.366220	5.698113
C	3.497418	10.936828	6.467401
H	2.983691	10.823467	7.417230
C	4.657927	10.208433	6.215379
H	5.042834	9.521170	6.965082
N	9.698159	6.878273	9.167982
N	6.284356	8.053700	8.951518
N	6.998821	7.049627	3.137653
N	9.077466	10.022895	3.200905
Ni	7.684660	7.908803	6.056687
P	7.672788	6.176764	7.466745
P	8.634049	9.014421	7.825696
P	9.320276	7.633436	4.507428
P	6.815900	9.380801	4.622591
H	6.464230	8.116909	6.847625

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Table S34: Cartesian coordinates of the optimized structure of  $[\text{Ni}^{\text{I}}(\text{P}_2^{\text{Ph}}\text{HN}_2^{\text{Bz}})(\text{P}_2^{\text{Ph}}\text{N}_2^{\text{Bz}})]_{\text{bc}}^{2+}$  ( $-4090.14971533 E_h$ ).

atom	x	y	z
C	8.706020	5.572918	8.632405
H	8.156907	4.792836	9.170533
H	9.478583	5.067279	8.026480
C	10.229529	7.428101	9.050226
H	11.022109	6.914960	8.477461
H	10.698007	7.951360	9.890115
C	9.747459	5.829125	10.809135
H	10.644871	5.231930	10.576488
H	8.966902	5.124242	11.117961
C	10.043421	6.784615	11.934999
C	11.347833	6.950015	12.406028
H	12.157270	6.378206	11.957539
C	11.618324	7.823093	13.458959
H	12.635579	7.932489	13.822441
C	10.583998	8.544198	14.048060
H	10.791642	9.218667	14.873191
C	9.277553	8.385620	13.585447
H	8.463759	8.931451	14.053392
C	9.009321	7.510785	12.537813
H	7.983273	7.369574	12.203354
C	8.030920	9.257243	9.064439
H	8.152694	8.878035	10.081608
H	8.018874	10.349478	9.081517
C	6.444072	7.338558	8.666839
H	5.395849	7.174975	8.406799
H	6.624921	7.050188	9.703348
C	5.539268	9.636231	9.141559
H	4.652883	9.325288	8.580758
H	5.761980	10.672723	8.874124
C	5.362586	9.473629	10.620381
C	4.522862	8.475789	11.131012
H	3.984663	7.817960	10.452474
C	4.340611	8.341098	12.503905
H	3.678900	7.571586	12.889048
C	4.985419	9.212002	13.380867
H	4.829029	9.117778	14.451040
C	5.807505	10.220888	12.882012
H	6.286380	10.920232	13.560369
C	5.994585	10.351148	11.508946
H	6.615510	11.158585	11.128020
C	6.435870	5.017392	6.915359
C	6.415087	4.694564	5.552327
H	7.006272	5.281570	4.847811
C	5.641683	3.622481	5.108742
H	5.632226	3.361962	4.054584

C	4.885906	2.881150	6.013480
H	4.285205	2.047120	5.663685
C	4.902958	3.202183	7.371388
H	4.317225	2.620019	8.076169
C	5.680039	4.262373	7.824551
H	5.696397	4.491693	8.888078
C	10.555761	10.137284	7.988202
C	11.868843	10.050767	8.464588
H	12.260191	9.118646	8.859112
C	12.698411	11.170343	8.435783
H	13.714180	11.093210	8.811049
C	12.227387	12.382324	7.939321
H	12.874596	13.253714	7.926901
C	10.922703	12.474987	7.455147
H	10.551971	13.418112	7.065031
C	10.096376	11.356614	7.467905
H	9.086124	11.435586	7.069359
C	9.161172	7.167080	2.844581
H	9.291394	7.817050	1.965105
H	9.777895	6.275342	2.693491
C	9.992120	9.802306	3.690021
H	10.312259	10.467536	4.512586
H	10.768854	9.828586	2.917920
C	7.395331	5.711501	2.031005
H	6.421825	5.315770	2.346320
H	8.118805	4.891789	2.112654
C	7.306797	6.185397	0.597565
C	8.435888	6.186623	-0.229895
H	9.382127	5.804391	0.147090
C	8.356201	6.638910	-1.544510
H	9.238881	6.622392	-2.176879
C	7.139493	7.087379	-2.055401
H	7.072717	7.423504	-3.085734
C	6.004366	7.076837	-1.247209
H	5.046666	7.396352	-1.648048
C	6.090707	6.631010	0.069712
H	5.194081	6.604990	0.685753
C	7.647730	10.419747	4.000839
H	6.835335	10.895713	3.440696
H	7.919669	11.087742	4.840238
C	6.795899	7.788882	3.131910
H	5.807064	7.321264	3.191012
H	6.791071	8.472997	2.268739
C	8.925880	11.349158	2.161748
H	9.782174	11.101709	1.523769
H	9.196981	12.261306	2.722251
C	7.717494	11.618168	1.303808
C	7.279078	10.662496	0.379480
H	7.815405	9.721993	0.276721
C	6.173303	10.916707	-0.425048

H	5.851306	10.169242	-1.143653
C	5.493963	12.130604	-0.320745
H	4.637507	12.331863	-0.957229
C	5.925041	13.088536	0.592309
H	5.407605	14.040065	0.670463
C	7.029972	12.830030	1.402301
H	7.372687	13.586205	2.105628
C	11.614750	7.467168	4.408027
C	12.712820	8.330488	4.319792
H	12.574129	9.396673	4.169562
C	14.008659	7.827641	4.422382
H	14.853912	8.504577	4.343273
C	14.220444	6.465075	4.612202
H	15.231599	6.076085	4.681488
C	13.131074	5.599719	4.710783
H	13.291546	4.535680	4.856655
C	11.836033	6.097658	4.619212
H	10.992963	5.414572	4.704315
C	5.292004	9.205869	5.231240
C	4.849330	10.523072	5.420533
H	5.486542	11.364606	5.165805
C	3.569302	10.774562	5.914587
H	3.234335	11.800192	6.038032
C	2.715201	9.719067	6.224551
H	1.713131	9.918497	6.591748
C	3.147404	8.403920	6.044903
H	2.479865	7.576091	6.266392
C	4.427933	8.147608	5.561982
H	4.748192	7.116721	5.423298
N	9.241800	6.513583	9.597223
N	6.688058	8.816433	8.552995
N	7.786134	6.723260	3.040113
N	8.740789	10.203747	3.076109
Ni	8.486390	7.878665	6.083043
P	7.505684	6.385652	7.448808
P	9.434501	8.699072	7.940875
P	9.889719	8.051721	4.325348
P	6.994407	8.807968	4.683980
H	6.696189	9.005679	7.533684

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