Supporting Information

Improved Photocatalytic Hydrogen Evolution Driven by Chloro(terpyridine)platinum(II) Derivatives Tethered to a Single Pendant Viologen Acceptor

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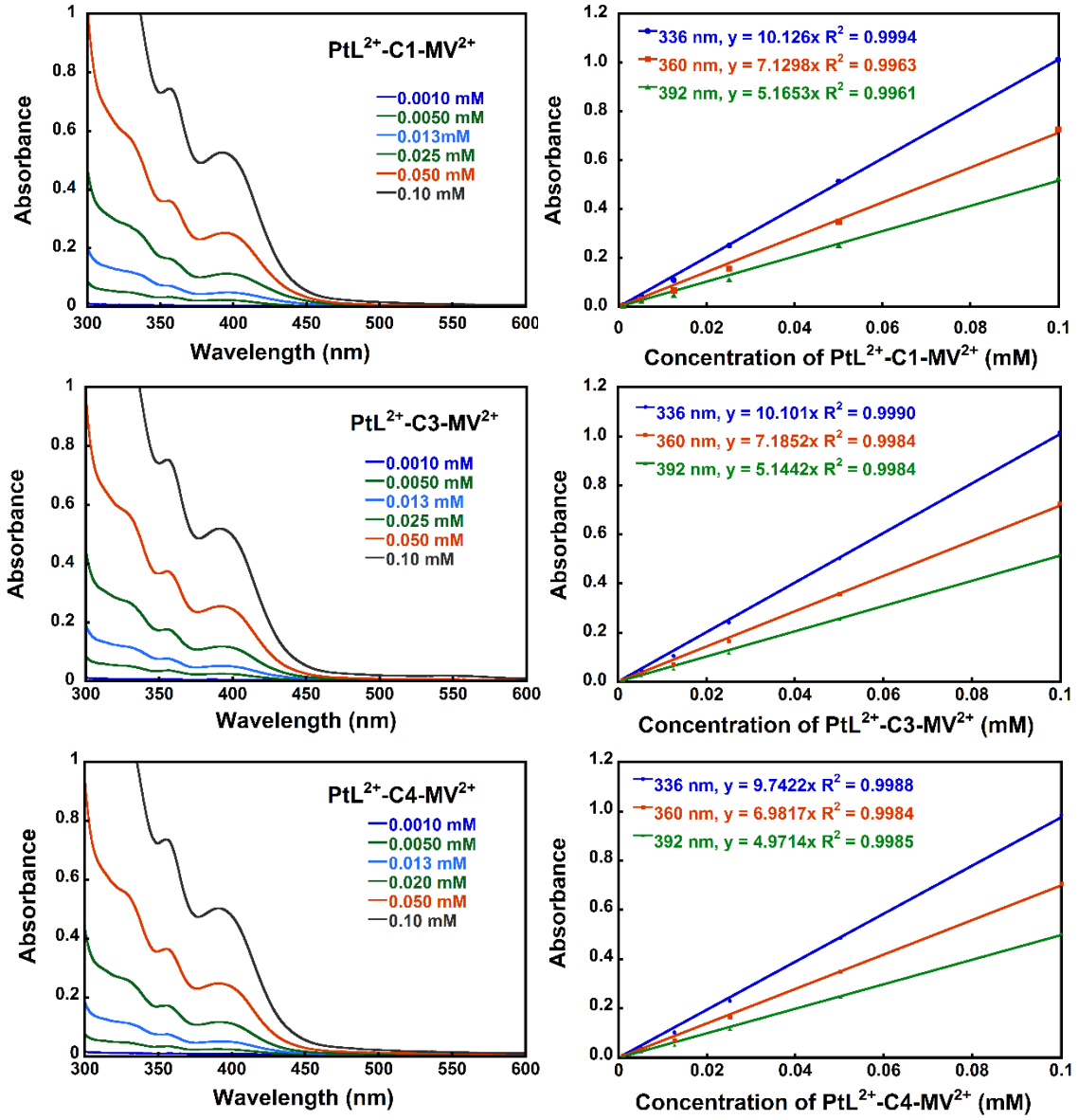


Figure S1. Absorption spectra of various concentration of **PtL**$^{2+}$**-Cn-MV**$^{2+}$ in the 0.1 M NaCl aqueous solution at 20 °C in Air (left), and the concentration dependence of the absorbance at 336, 360 and 392 nm under the concentration range between 0.001 and 0.1 mM.

$$PtLCl + OAc^- \underset{}{\overset{K_{ex}}{\rightleftharpoons}} PtLOAc + Cl^-$$

$$K_{ex} = \frac{[PtLOAc][Cl^-]}{[PtLCl][OAc^-]}, [OAc^-] \gg [Pt]_{total} \ \& \ [Cl^-] \gg [Pt]_{total}$$

$$[PtLOAc] = \frac{K_{ex}[PtLCl][OAc^-]}{[Cl^-]}$$

$$C_0 = [Pt]_{total} = [PtLCl] + [PtLOAc]$$

$$C_0 = \frac{[PtLCl]}{[Cl^-]} ([Cl^-] + K_{ex}[OAc^-])$$

$$ABS_{obs} = [PtLCl]\varepsilon_{PtLCl} + [PtLOAc]\varepsilon_{PtLOAc}$$

$$= [PtLCl]\varepsilon_{PtLCl} + \left[\frac{K_{ex}[PtLCl][OAc^-]}{[Cl^-]}\right]\varepsilon_{PtLOAc}$$

$$= [PtLCl]\left[\frac{\varepsilon_{PtLCl}[Cl^-] + K_{ex}[OAc^-]\varepsilon_{PtLOAc}}{[Cl^-]}\right]$$

$$= \frac{[PtLCl]}{[Cl^-]} (\varepsilon_{PtLCl}[Cl^-] + \varepsilon_{PtLOAc} K_{ex}[OAc^-])$$

$$\boldsymbol{ABS_{obs.} = C_0 \frac{(\varepsilon_{PtLCl} \times [Cl^-] + \varepsilon_{PtLOAc} \times K_{ex}[OAc^-])}{([Cl^-] + K_{ex}[OAc^-])}} \quad \textbf{eqn. S1}$$

Table S1. Relative abundance of **PtLCl** species in acetate buffer solution (0.1 M, pH = 5) containing 0.05 mM **PtL**$^{2+}$**-Cn-MV**$^{2+}$ and 0.1 M NaCl.

Compound	Fitting wavelength (nm)	K_{ex}	R^2	Relative abundance of **PtLCl** species
PtL^{2+}**-C1-MV**$^{2+}$	355	0.09498	0.9926	93.8%
	392	0.06362	0.9972	95.7%
PtL^{2+}**-C3-MV**$^{2+}$	355	0.13054	0.9922	91.6%
	392	0.06333	0.9965	95.8%
PtL^{2+}**-C4-MV**$^{2+}$	355	0.09333	0.9898	93.9%
	392	0.08245	0.9966	94.5%

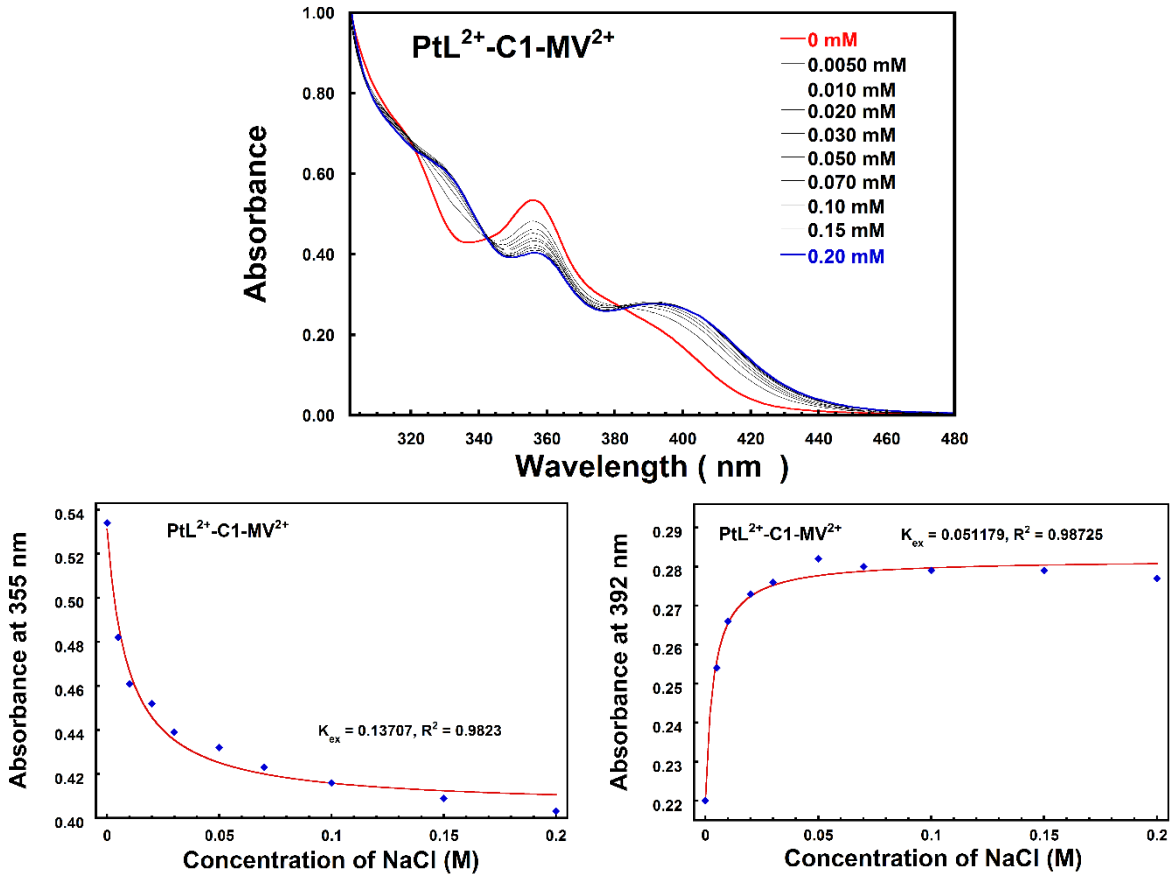


Figure S2. Absorption spectra of **PtL**$^{2+}$**-C1-MV**$^{2+}$ in an aqueous acetate buffer solution (0.1 M, pH = 5.0) in the presence of various concentration of NaCl at 20 °C in Air.

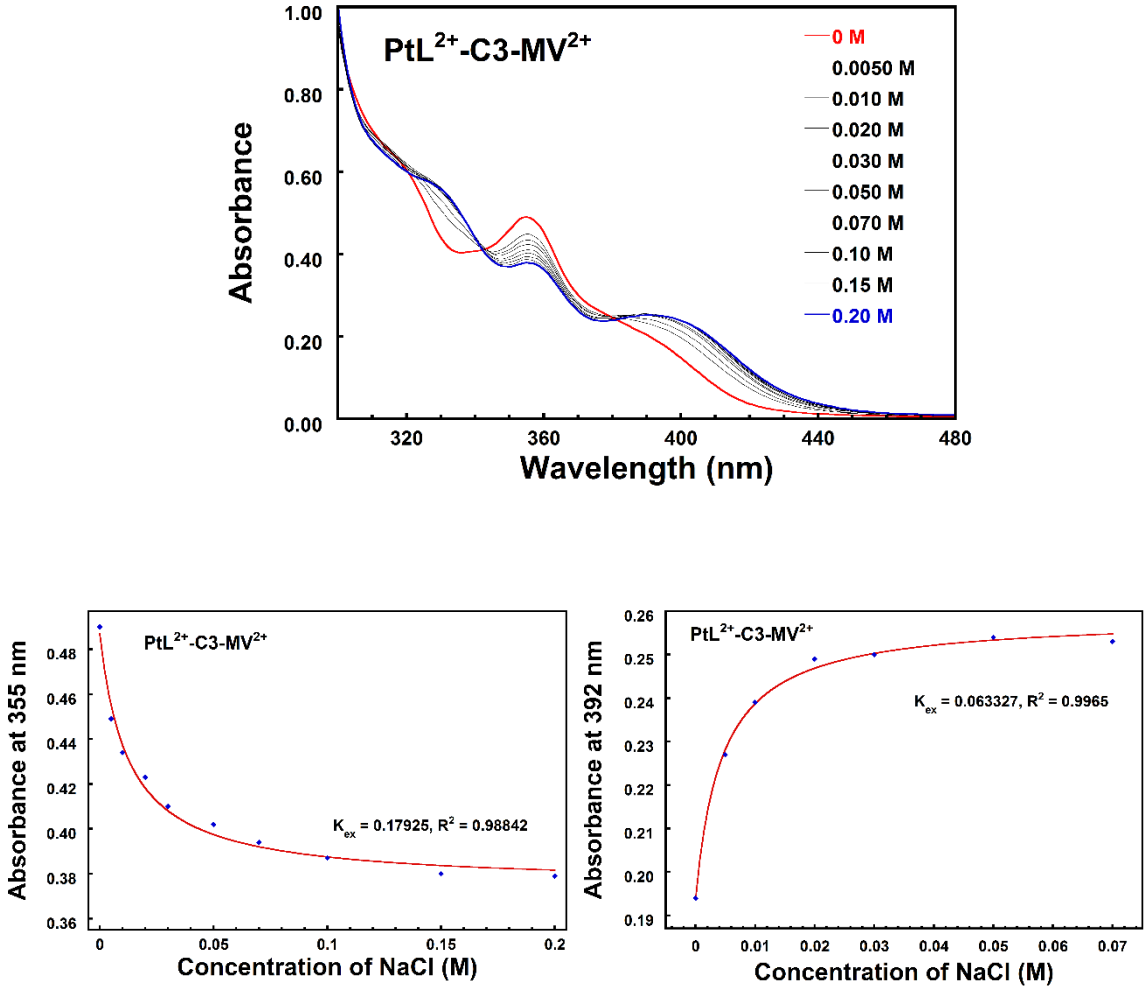


Figure S3. Absorption spectra of **PtL**$^{2+}$**-C3-MV**$^{2+}$ in an aqueous acetate buffer solution (0.1 M, pH = 5.0) in the presence of various concentration of NaCl at 20 °C in Air.

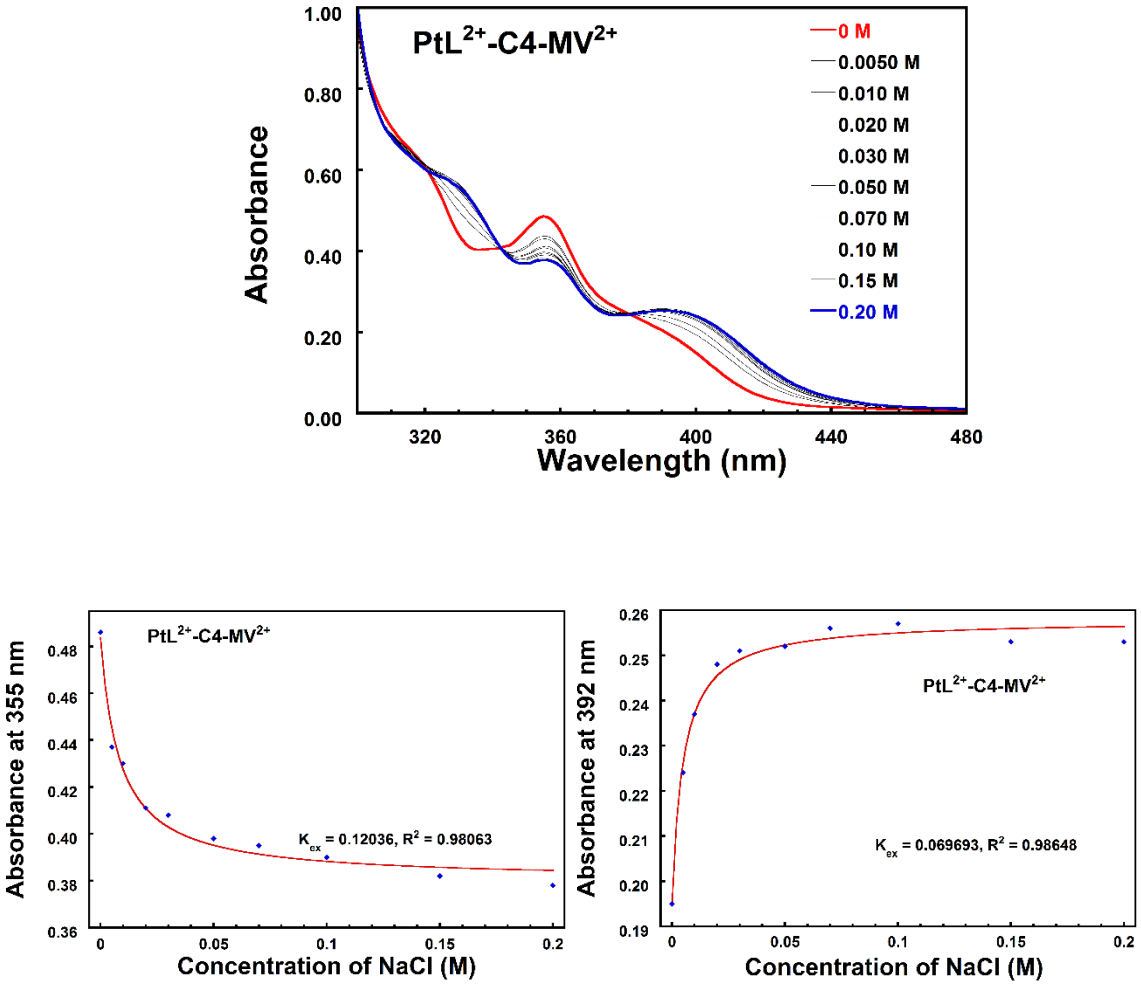


Figure S4. Absorption spectra of **PtL**$^{2+}$**-C4-MV**$^{2+}$ in an aqueous acetate buffer solution (0.1 M, pH = 5.0) in the presence of various concentration of NaCl at 20 °C in Air.

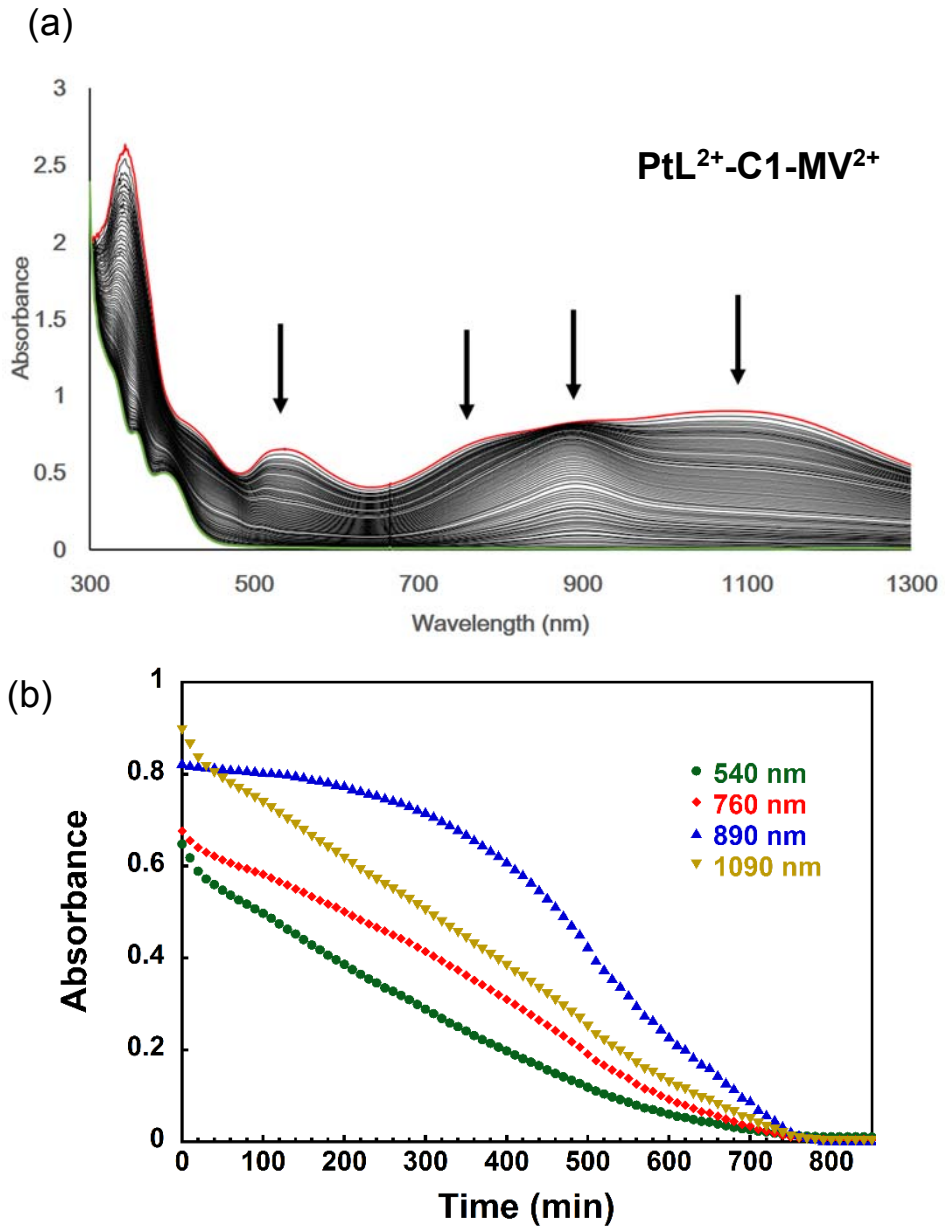


Figure S5. Spectral changes of the 90-min-irradiated aqueous acetate buffer solution containing 30 mM EDTA, 0.05 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$, and 0.1 M NaCl under Ar atmosphere in the dark (a). Time course of the absorbance changes at 540, 760, 890, and 1090 nm (b).

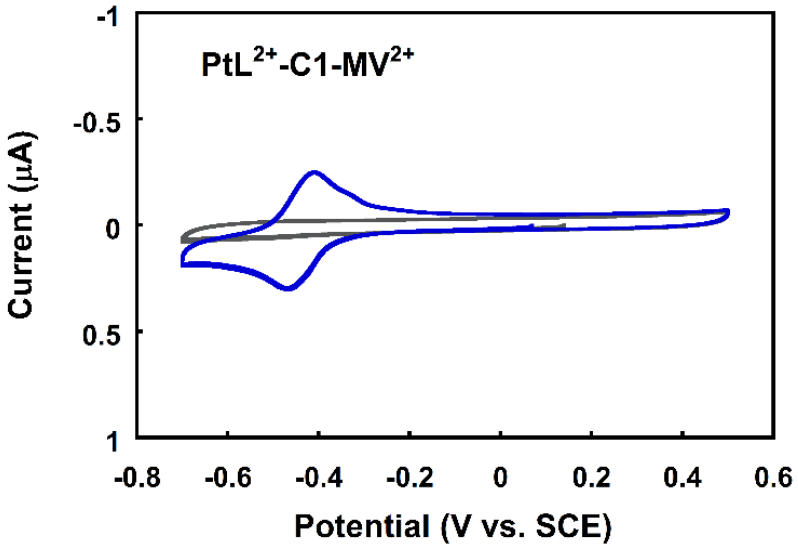


Figure S6. Cyclic voltammogram of 0.1 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$ in an aqueous acetate buffer solution (0.1 M, pH = 5.0) containing 30 mM EDTA and 0.1 M NaCl at 20 °C under Ar atmosphere. Potentials are given vs. SCE. Experiments were carried out using a glassy carbon working electrode, a Pt wire counter electrode, and a SCE reference electrode at the sweep rate of 50 mV/s.

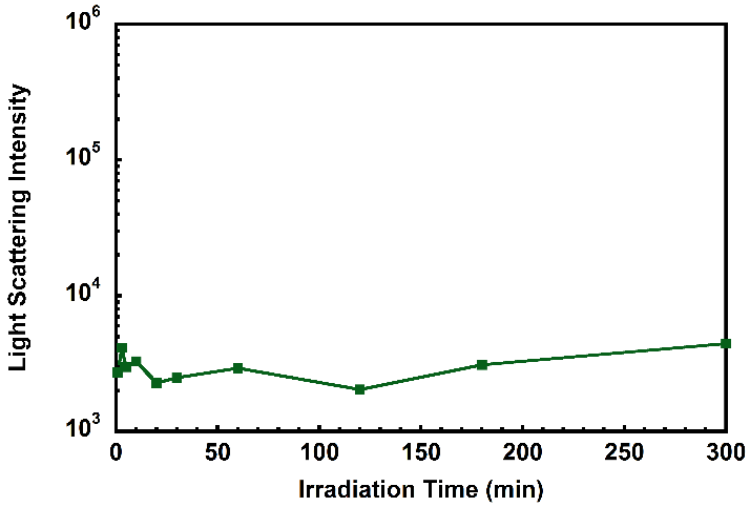


Figure S7. Changes of the light-scattering intensity of the aqueous acetate buffer solution (0.1 M, pH = 5.0) containing 30 mM EDTA, 0.05 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$, and 0.1 M NaCl. Photolysis experiment was carried out under Ar atmosphere at 20 °C.

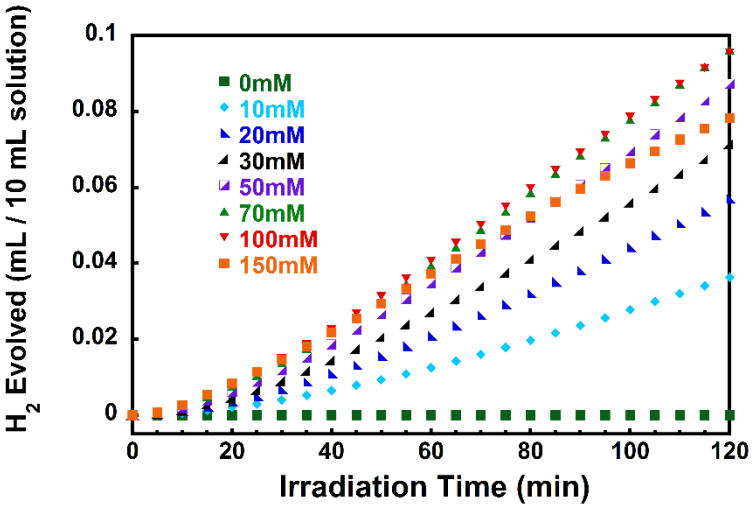


Figure S8. The H_2 production curves from an aqueous acetate buffer solution (10 mL, 0.1 M, pH = 5.0, at 20 °C under Ar atmosphere) containing 0.05 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$, 0.1 M NaCl, and various concentration of EDTA (The raw data of Figure 8a in MS).

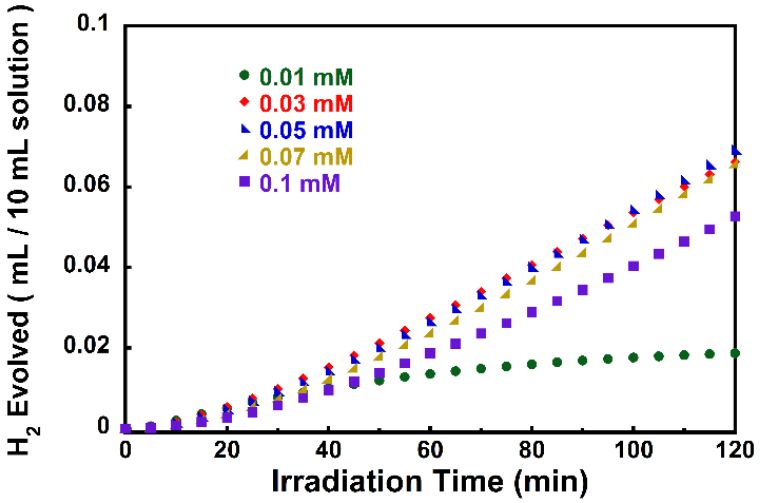


Figure S9. The H_2 production curves from an aqueous acetate buffer solution (10 mL, 0.1 M, pH = 5.0, at 20 °C under Ar atmosphere) containing 30 mM EDTA, 0.1 M NaCl, and various concentration of **PtL**$^{2+}$**-C1-MV**$^{2+}$ (The raw data of Figure 8b in MS).

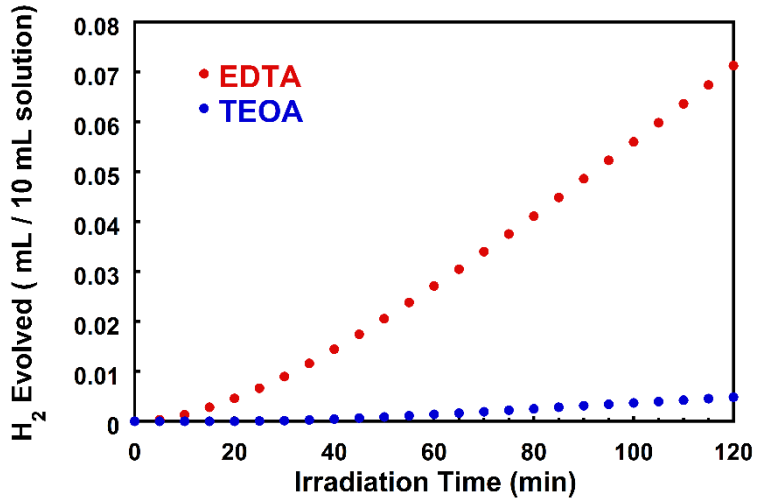
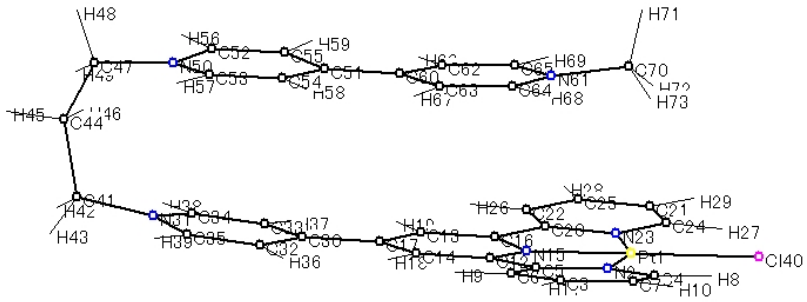


Figure S10. Photochemical H_2 evolution from an aqueous acetate buffer solution (0.1 M, pH = 5.0, 10 mL, at 20 °C under Ar atmosphere) containing 0.05 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$, 0.1 M NaCl and 30 mM electron donor (EDTA or TEOA).

(a)



(b)

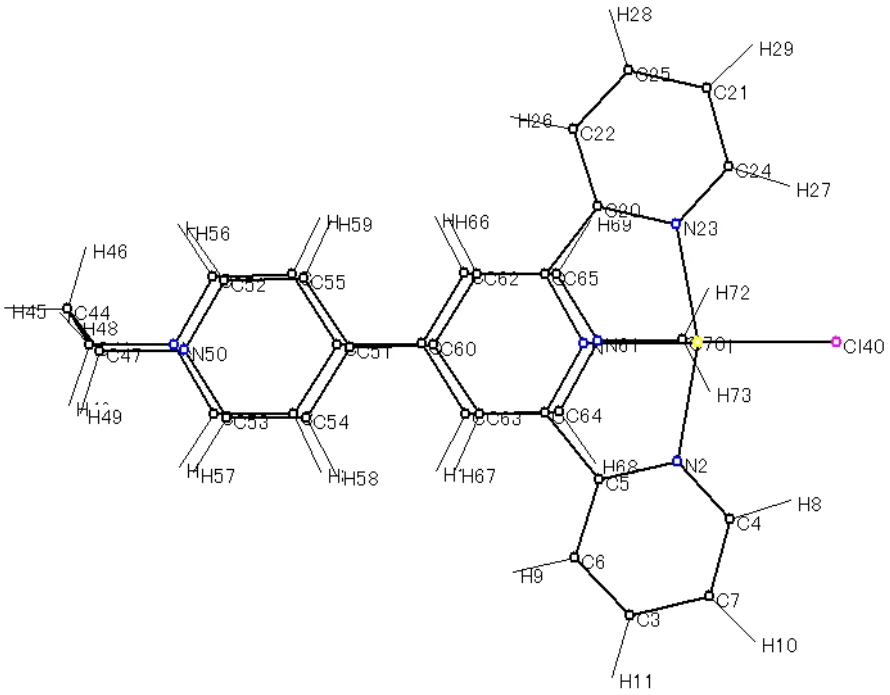


Figure S11. Stereo views showing the geometries for (a) the closed-shell singlet state and (b) the open-shell singlet state of the two-electron-reduced form of **[PtL^{2+}-C1-MV^{2+}]$^{4+}$**. The structures were optimized at the M06 and UM06 level of DFT calculations with the effect of water solvation taken into consideration using the polarizable continuum model (PCM) method, where the SDD basis set was used for Pt and the 6-31G** basis set for H, C, N, O, and Cl.

Table S2. DFT-optimized geometry of the one-electron-reduced form **[PtL**$^{1.5+}$**-C1-MV**$^{1.5+}$**]**$^{3+}$ (doublet) having an intramolecular stacking geometry, computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z	Spin Density
Pt1	3.147621	0.051731	-0.393954	0.018133
N2	2.963529	-2.001237	-0.322334	0.000761
C3	2.386626	-4.701430	-0.272043	0.002030
C4	3.920363	-2.884739	-0.020903	0.001343
C5	1.695351	-2.430953	-0.605118	0.001656
C6	1.389201	-3.781914	-0.581497	-0.000452
C7	3.667117	-4.249591	0.012502	0.000461
H8	4.899654	-2.464721	0.194411	-0.000062
H9	0.380821	-4.118698	-0.800604	-0.000001
H10	4.469121	-4.936198	0.260141	-0.000038
H11	2.157702	-5.762278	-0.252699	-0.000100
C12	0.721444	-1.366187	-0.896742	-0.012347
C13	-0.813551	0.899253	-1.325849	0.028529
C14	-0.626894	-1.511655	-1.167817	0.038482
N15	1.254001	-0.125895	-0.852686	0.075275
C16	0.537756	1.001684	-1.051923	-0.005393
C17	-1.420929	-0.368973	-1.379343	0.006600
H18	-1.055137	-2.506524	-1.229935	-0.001566
H19	-1.404577	1.802674	-1.439038	-0.001535
C20	1.327546	2.233169	-0.890538	-0.001405
C21	2.975744	4.399037	-0.475308	0.003814
C22	0.808314	3.513393	-1.003583	0.002896
N23	2.646324	2.044399	-0.577458	0.003890
C24	3.446023	3.096691	-0.374429	0.000762
C25	1.641381	4.607536	-0.793933	0.001154
H26	-0.239302	3.660208	-1.247670	-0.000137
H27	4.478674	2.864816	-0.125443	-0.000050
H28	1.243775	5.614039	-0.877992	-0.000085
H29	3.654284	5.227071	-0.302315	-0.000197
C30	-2.865584	-0.479782	-1.578704	0.078768
N31	-5.647711	-0.561749	-1.660278	0.040745

C32	-3.578893	-1.624803	-1.184680	-0.007814
C33	-3.618356	0.583739	-2.115283	-0.005775
C34	-4.986721	0.523454	-2.137156	0.027220
C35	-4.950676	-1.642247	-1.236490	0.032701
H36	-3.083682	-2.497737	-0.771906	-0.000353
H37	-3.147094	1.471667	-2.522181	0.000098
H38	-5.598485	1.331477	-2.525550	-0.001303
H39	-5.532534	-2.498958	-0.911898	-0.001786
Cl40	5.429736	0.270242	0.203937	0.001196
C41	-7.126130	-0.527944	-1.574303	-0.002608
H42	-7.456964	-1.534161	-1.301216	0.000172
H43	-7.508144	-0.310757	-2.576275	0.000841
C44	-7.629873	0.511993	-0.584467	0.006550
H45	-8.719250	0.555412	-0.693126	0.001110
H46	-7.273731	1.511180	-0.867179	-0.000230
C47	-7.342955	0.234404	0.883645	-0.008002
H48	-7.838864	0.988805	1.503054	0.004516
H49	-7.735832	-0.743599	1.178350	0.000138
N50	-5.906938	0.241405	1.225220	0.112856
C51	-3.163657	0.260212	1.833458	0.138705
C52	-5.169281	1.378678	1.056223	0.063760
C53	-5.310130	-0.863015	1.753596	0.046845
C54	-3.976751	-0.879084	2.054553	0.016746
C55	-3.835175	1.412471	1.347302	0.009008
H56	-5.702376	2.245828	0.678825	-0.003623
H57	-5.950863	-1.723622	1.918283	-0.002521
H58	-3.576966	-1.792698	2.480354	-0.001430
H59	-3.309007	2.343500	1.162546	-0.001531
C60	-1.744456	0.247330	2.098174	0.087559
N61	1.005266	0.219372	2.658345	0.093610
C62	-0.966445	1.431179	2.107821	0.035447
C63	-1.041171	-0.955470	2.355753	0.032344
C64	0.299297	-0.941411	2.633751	0.027982
C65	0.372143	1.391693	2.394300	0.020470
H66	-1.406671	2.405511	1.921944	-0.002227
H67	-1.533938	-1.921582	2.325477	-0.002305

H68	0.859231	-1.846791	2.847055	-0.001892
H69	0.986472	2.287126	2.428891	-0.001570
C70	2.416398	0.213027	3.061221	-0.006584
H71	2.489461	0.325384	4.146340	0.006980
H72	2.939034	1.032092	2.561367	0.001395
H73	2.871458	-0.730136	2.750627	0.001374

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[a]Part of the Gaussian output file:

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Red. masses --	6.1204	7.2848	5.5882

Zero-point correction=	0.590605 (Hartree/Particle)
Thermal correction to Energy=	0.625193
Thermal correction to Enthalpy=	0.626137
Thermal correction to Gibbs Free Energy=	0.523964
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Sum of electronic and thermal Energies=	-2220.163937
Sum of electronic and thermal Enthalpies=	-2220.162993
Sum of electronic and thermal Free Energies=	-2220.265165

Item	Value	Threshold	Converged?
Maximum Force	0.000151	0.000450	YES
RMS Force	0.000015	0.000300	YES

Table S3. DFT-optimized geometry of the one-electron-reduced form **[PtL^{2+}-C1-MV^{+}•]**$^{3+}$ (doublet) without having a stacking geometry, computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z	Spin Density
Pt1	-5.784572	-0.677936	0.247285	0.000002
N2	-4.890449	-2.356598	-0.554777	0.000000
C3	-3.412921	-4.405418	-1.672708	0.000000
C4	-5.454842	-3.548179	-0.770079	0.000000
C5	-3.579553	-2.148395	-0.886972	0.000000
C6	-2.824368	-3.164227	-1.448602	0.000000
C7	-4.742003	-4.601139	-1.330235	0.000000
C8	-3.066764	-0.797162	-0.612524	-0.000002
C9	-2.469128	1.830121	0.042938	0.000005
C10	-1.787997	-0.315719	-0.852682	0.000006
N11	-3.981685	0.028514	-0.070386	0.000009
C12	-3.736300	1.309684	0.264078	-0.000001
C13	-1.487500	1.009728	-0.522470	-0.000001
C14	-4.900170	2.002714	0.839129	0.000000
C15	-7.190535	3.109229	1.883169	0.000000
C16	-4.884062	3.322082	1.259373	0.000000
N17	-6.038998	1.252228	0.941734	0.000000
C18	-7.151996	1.789954	1.448531	0.000000
C19	-6.043548	3.881938	1.787934	0.000000
C20	-0.134748	1.541564	-0.771283	0.000020
N21	2.395650	2.536284	-1.243447	-0.000030
C22	0.585156	1.161822	-1.908548	-0.000003
C23	0.461844	2.437680	0.121104	-0.000011
C24	1.727194	2.915745	-0.134875	0.000027
C25	1.845277	1.672161	-2.121221	0.000016
Cl26	-7.954187	-1.531697	0.629490	0.000000
N27	7.259468	1.683067	-0.706219	0.154824
C28	9.455091	0.086164	0.094262	0.175975
C29	7.070379	0.580780	0.079077	0.057906
C30	8.536202	1.996825	-1.094910	0.065125
C31	9.606620	1.245053	-0.725157	0.029122

C32	8.111001	-0.205559	0.471313	0.044028
C33	10.559936	-0.716061	0.503341	0.146077
N34	12.746266	-2.307688	1.310942	0.151828
C35	10.403668	-1.881915	1.309822	0.054550
C36	11.905425	-0.412569	0.138765	0.045473
C37	12.943377	-1.195257	0.542717	0.049142
C38	11.475708	-2.632869	1.687322	0.044244
C39	13.891789	-3.088198	1.768615	-0.011967
C40	3.773452	3.025638	-1.473577	-0.000085
C41	4.785159	2.046041	-0.900024	0.000904
C42	6.181639	2.599771	-1.102253	-0.012599
H43	-6.499382	-3.635913	-0.482603	0.000000
H44	-1.785249	-2.994846	-1.711964	0.000000
H45	-5.234207	-5.554244	-1.489881	0.000000
H46	-2.831751	-5.209221	-2.113319	0.000000
H47	-1.023657	-0.966654	-1.265816	0.000000
H48	-2.249775	2.867169	0.278434	0.000000
H49	-3.977062	3.912070	1.178390	0.000000
H50	-8.016460	1.132594	1.495339	0.000000
H51	-6.043641	4.915205	2.120121	0.000000
H52	-8.113113	3.510733	2.288056	0.000000
H53	0.164851	0.490471	-2.650155	0.000000
H54	-0.037121	2.750300	1.032270	0.000000
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H56	2.441923	1.416878	-2.991025	-0.000002
H57	6.052979	0.353831	0.373359	-0.003622
H58	8.635362	2.881390	-1.716164	-0.003813
H59	10.579125	1.563606	-1.084035	-0.002648
H60	7.868066	-1.064776	1.086636	-0.003383
H61	9.427481	-2.212241	1.646945	-0.003672
H62	12.147576	0.451339	-0.470377	-0.003257
H63	13.973904	-0.979760	0.278523	-0.003070
H64	11.374472	-3.521961	2.300858	-0.002859
H65	14.388220	-2.583396	2.602938	0.010285
H66	13.551601	-4.071988	2.094700	0.001105
H67	14.601225	-3.212541	0.947476	0.004657

H68	3.852485	4.011294	-1.005693	0.000019
H69	4.568746	1.902009	0.166554	-0.000178
H70	4.678889	1.073502	-1.399918	-0.000225
H71	6.302068	3.536039	-0.540647	0.010357
H72	6.337446	2.834433	-2.162719	0.005722
H73	3.897712	3.157136	-2.552498	0.000002

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[a]Part of the Gaussian output file:

SCF Done: E(UM06) = -2220.76879091 A.U. after 6 cycles

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Red. masses --	3.3419	6.6854	4.6384

Zero-point correction= 0.590224 (Hartree/Particle)
Thermal correction to Energy= 0.626349
Thermal correction to Enthalpy= 0.627293
Thermal correction to Gibbs Free Energy= 0.515807
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Sum of electronic and thermal Energies= -2220.142442
Sum of electronic and thermal Enthalpies= -2220.141497
Sum of electronic and thermal Free Energies= -2220.252984

Item	Value	Threshold	Converged?
Maximum Force	0.000008	0.000450	YES
RMS Force	0.000001	0.000300	YES

Table S4. DFT-optimized geometry of the one-electron-reduced form **PtL**$^{+}$• (doublet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z	Spin Density
Pt1	2.093212	0.000397	0.000897	0.041820
N2	1.736281	-2.034663	-0.003438	0.012499
C3	0.937566	-4.681214	-0.005809	0.008754
C4	2.650557	-3.015277	-0.001011	0.002374
C5	0.400465	-2.341866	-0.007208	0.002368
C6	-0.010957	-3.669033	-0.008357	0.004513
C7	2.289400	-4.352388	-0.002062	0.013512
C8	-0.511346	-1.193967	-0.006604	0.025208
C9	-1.884562	1.214231	0.001694	0.043565
C10	-1.883835	-1.215643	-0.010917	0.043969
N11	0.143274	-0.000162	-0.000708	0.214590
C12	-0.512030	1.193311	0.002470	0.025563
C13	-2.628585	-0.000902	-0.005826	0.126205
C14	0.399137	2.341687	0.005957	0.002322
C15	2.286954	4.353278	0.007414	0.013619
C16	-0.013029	3.668634	0.007431	0.004593
N17	1.735142	2.035234	0.005058	0.012588
C18	2.648867	3.016381	0.005824	0.002339
C19	0.934918	4.681342	0.008244	0.008729
C20	-4.063410	-0.001554	-0.007917	0.130679
N21	-6.883779	-0.003220	-0.016152	0.121514
C22	-4.827979	-1.200921	0.032292	0.034442
C23	-4.828970	1.197338	-0.052850	0.032023
C24	-6.191681	1.170687	-0.053401	0.043535
C25	-6.191095	-1.175827	0.028956	0.042505
Cl26	4.487858	0.000949	0.001986	0.002481
C27	-8.343766	0.005350	0.043771	-0.009655
H28	-8.725319	0.832222	-0.557948	0.002698
H29	3.687909	-2.689663	0.002044	-0.000229
H30	-1.070332	-3.906326	-0.010277	-0.000259
H31	3.059655	-5.115929	0.000460	-0.000729

H32	0.622928	-5.720396	-0.006291	-0.000532
H33	-2.391165	-2.174474	-0.025199	-0.002842
H34	-2.392422	2.172808	0.014224	-0.002826
H35	-1.072540	3.905326	0.007269	-0.000262
H36	3.686411	2.691370	0.004959	-0.000228
H37	0.619696	5.720347	0.009156	-0.000532
H38	3.056785	5.117250	0.007711	-0.000734
H39	-4.355776	-2.176184	0.071463	-0.002600
H40	-4.357403	2.172620	-0.099260	-0.002479
H41	-6.791295	2.074529	-0.086636	-0.002609
H42	-6.788167	-2.081317	0.060120	-0.002577
H43	-8.681587	0.121630	1.077727	0.008490
H44	-8.727619	-0.933076	-0.359132	0.001598

---

[a]Part of the Gaussian output file:

SCF Done: E(UM06) = -1608.22289782 A.U. after 6 cycles

	1	2	3
	A	A	A
Frequencies --	40.7681	47.1626	53.4210
Red. masses --	6.0981	5.1882	3.5333

Zero-point correction= 0.338944 (Hartree/Particle)
Thermal correction to Energy= 0.361744
Thermal correction to Enthalpy= 0.362688
Thermal correction to Gibbs Free Energy= 0.285119
Sum of electronic and zero-point Energies= -1607.883954
Sum of electronic and thermal Energies= -1607.861154
Sum of electronic and thermal Enthalpies= -1607.860210
Sum of electronic and thermal Free Energies= -1607.937779

Item	Value	Threshold	Converged?
Maximum Force	0.000005	0.000450	YES
RMS Force	0.000001	0.000300	YES

Table S5. DFT-optimized geometry of the two-electron-reduced form **[PtL$^+$-C1-MV$^+$]**$^{2+}$ (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	3.190226	0.012110	-0.402612
N2	2.838096	-2.026068	-0.436850
C3	2.029195	-4.668795	-0.422991
C4	3.726368	-3.001195	-0.200143
C5	1.524449	-2.335736	-0.669553
C6	1.106508	-3.660392	-0.664450
C7	3.358672	-4.337215	-0.186347
H8	4.746908	-2.672631	-0.019037
H9	0.063016	-3.900332	-0.844748
H10	4.107470	-5.097454	0.007759
H11	1.710249	-5.706581	-0.416498
C12	0.632126	-1.188417	-0.878696
C13	-0.734022	1.208224	-1.102599
C14	-0.721062	-1.219012	-1.114713
N15	1.273995	0.003770	-0.787446
C16	0.619576	1.189940	-0.868775
C17	-1.457923	-0.008793	-1.241462
H18	-1.221849	-2.179634	-1.174874
H19	-1.244753	2.163920	-1.153715
C20	1.500377	2.344673	-0.651433
C21	3.313928	4.360893	-0.152515
C22	1.069363	3.665053	-0.638747
N23	2.816815	2.046401	-0.419519
C24	3.695002	3.028766	-0.174797
C25	1.981630	4.680934	-0.389333
H26	0.023895	3.895958	-0.819342
H27	4.718613	2.709299	0.005365
H28	1.652509	5.715476	-0.376875
H29	4.054924	5.127106	0.047903
C30	-2.874603	-0.016267	-1.481427
N31	-5.679969	-0.033360	-1.725530

C32	-3.619547	-1.218846	-1.617183
C33	-3.635495	1.179848	-1.598628
C34	-4.992695	1.147744	-1.713327
C35	-4.979381	-1.202170	-1.725110
H36	-3.141392	-2.192062	-1.611328
H37	-3.167495	2.158014	-1.584656
H38	-5.591547	2.050411	-1.787079
H39	-5.567061	-2.112674	-1.791081
Cl40	5.527375	0.022574	0.089612
C41	-7.150555	-0.023570	-1.701859
H42	-7.482464	-1.060049	-1.819357
H43	-7.503351	0.536392	-2.574951
C44	-7.721149	0.590544	-0.430994
H45	-8.812402	0.605300	-0.534367
H46	-7.423800	1.644939	-0.354533
C47	-7.392200	-0.145469	0.860970
H48	-7.948479	0.309540	1.688767
H49	-7.700659	-1.194539	0.797258
N50	-5.963324	-0.134078	1.203044
C51	-3.183425	-0.075647	1.715175
C52	-5.317748	1.054598	1.426820
C53	-5.249111	-1.295552	1.317417
C54	-3.911301	-1.293818	1.571568
C55	-3.980950	1.107860	1.671706
H56	-5.936205	1.946764	1.401893
H57	-5.809683	-2.215532	1.181622
H58	-3.419649	-2.259327	1.632495
H59	-3.548895	2.089293	1.835722
C60	-1.776650	-0.042902	1.946716
N61	0.985335	0.019638	2.540397
C62	-1.048740	1.175959	2.087086
C63	-0.990968	-1.227150	2.072827
C64	0.337391	-1.169654	2.376652
C65	0.281519	1.178958	2.387423
H66	-1.528266	2.142265	1.963743
H67	-1.424958	-2.214271	1.947576

H68	0.942508	-2.061898	2.509292
H69	0.844704	2.097529	2.526135
C70	2.359516	0.054108	3.040630
H71	2.360834	0.090415	4.134742
H72	2.868648	0.935683	2.642475
H73	2.890409	-0.838793	2.701938

---

[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -2220.94182487 A.U. after 7 cycles 1

2 3

	A	A	A
Frequencies --	18.8555	26.1787	29.1734
Red. masses --	5.1024	6.1793	5.4375

Zero-point correction= 0.588027 (Hartree/Particle)
Thermal correction to Energy= 0.623109
Thermal correction to Enthalpy= 0.624053
Thermal correction to Gibbs Free Energy= 0.522159
Sum of electronic and zero-point Energies= -2220.353798
Sum of electronic and thermal Energies= -2220.318716
Sum of electronic and thermal Enthalpies= -2220.317772
Sum of electronic and thermal Free Energies= -2220.419666

Item	Value	Threshold	Converged?
Maximum Force	0.000032	0.000450	YES
RMS Force	0.000004	0.000300	YES

Table S6. DFT-optimized geometry of the two-electron-reduced form **[PtL^{+}-C1-MV^{+}]**$^{2+}$ (open-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z	Spin Density
Pt1	3.188958	0.013163	-0.403416	0.000000
N2	2.838824	-2.025348	-0.437088	-0.000001
C3	2.032491	-4.668809	-0.420703	0.000001
C4	3.728035	-2.999365	-0.199375	0.000001
C5	1.525475	-2.336466	-0.669474	0.000001
C6	1.108833	-3.661526	-0.663186	-0.000002
C7	3.361635	-4.335724	-0.184279	-0.000002
H8	4.748229	-2.669604	-0.018461	0.000000
H9	0.065574	-3.902638	-0.843250	0.000000
H10	4.111128	-5.095066	0.010652	0.000000
H11	1.714543	-5.706894	-0.413230	0.000000
C12	0.632076	-1.190125	-0.879426	0.000000
C13	-0.736065	1.205184	-1.105272	0.000000
C14	-0.721063	-1.222080	-1.115423	0.000000
N15	1.272902	0.002713	-0.788964	-0.000002
C16	0.617461	1.188258	-0.870992	0.000000
C17	-1.458862	-0.012512	-1.243253	-0.000001
H18	-1.221126	-2.183121	-1.174998	0.000000
H19	-1.247831	2.160321	-1.156111	0.000000
C20	1.496937	2.343935	-0.653312	0.000001
C21	3.307776	4.362037	-0.152357	-0.000002
C22	1.064516	3.663876	-0.641217	-0.000001
N23	2.813462	2.047052	-0.420202	-0.000001
C24	3.690325	3.030333	-0.174415	0.000001
C25	1.975411	4.680708	-0.390742	0.000001
H26	0.019008	3.893690	-0.823009	0.000000
H27	4.714109	2.711989	0.006735	0.000000
H28	1.645197	5.714905	-0.378653	0.000000
H29	4.047728	5.129009	0.049006	0.000000
C30	-2.875586	-0.021112	-1.483097	0.000000
N31	-5.681117	-0.039371	-1.724015	0.000001

C32	-3.620379	-1.224221	-1.614042	0.000001
C33	-3.636711	1.174511	-1.604080	0.000001
C34	-4.994021	1.141880	-1.717168	-0.000001
C35	-4.980374	-1.208070	-1.720285	-0.000002
H36	-3.142147	-2.197370	-1.604737	0.000000
H37	-3.168692	2.152713	-1.594912	0.000000
H38	-5.592939	2.044255	-1.793856	0.000000
H39	-5.568006	-2.118900	-1.782030	0.000000
Cl40	5.526034	0.026253	0.089109	0.000000
C41	-7.151702	-0.030082	-1.697692	0.000000
H42	-7.483419	-1.067210	-1.809423	0.000000
H43	-7.506372	0.525336	-2.572917	0.000000
C44	-7.719926	0.590428	-0.428852	0.000000
H45	-8.811312	0.605861	-0.530759	0.000000
H46	-7.421431	1.644841	-0.357581	0.000000
C47	-7.389876	-0.140105	0.865979	0.000000
H48	-7.943682	0.319906	1.692667	0.000000
H49	-7.700463	-1.188921	0.808135	0.000000
N50	-5.960314	-0.129428	1.205254	0.000001
C51	-3.179738	-0.072605	1.713852	0.000002
C52	-5.313059	1.059248	1.424464	0.000001
C53	-5.247181	-1.291365	1.321591	0.000001
C54	-3.909053	-1.290362	1.573986	0.000000
C55	-3.975932	1.111736	1.667704	0.000000
H56	-5.930603	1.952009	1.397641	0.000000
H57	-5.808956	-2.211079	1.188946	0.000000
H58	-3.418233	-2.256203	1.636417	0.000000
H59	-3.542482	2.093149	1.828306	0.000000
C60	-1.772893	-0.041041	1.945323	0.000000
N61	0.988558	0.019168	2.541522	0.000000
C62	-1.043409	1.177144	2.083380	0.000000
C63	-0.988759	-1.226061	2.073983	0.000001
C64	0.339308	-1.169689	2.379113	0.000000
C65	0.286589	1.179033	2.385250	0.000000
H66	-1.521488	2.143849	1.957348	0.000000
H67	-1.423954	-2.212832	1.950127	0.000000

H68	0.943276	-2.062363	2.514166	0.000000
H69	0.850849	2.097213	2.522331	0.000000
C70	2.361390	0.051179	3.045608	0.000000
H71	2.359701	0.070827	4.140149	0.000000
H72	2.867478	0.941139	2.662989	0.000000
H73	2.896805	-0.834379	2.694835	0.000000

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[a]Part of the Gaussian output file:

SCF Done: E(UM06) = -2220.94182738 A.U. after 30 cycles

Annihilation of the first spin contaminant:

S**2 before annihilation 0.0000, after 0.0000

	1	2	3
	A	A	A
Frequencies --	20.2220	26.2330	29.3215
Red. masses --	4.9986	6.3129	5.6032

Zero-point correction=	0.587950 (Hartree/Particle)
Thermal correction to Energy=	0.623074
Thermal correction to Enthalpy=	0.624018
Thermal correction to Gibbs Free Energy=	0.522044
Sum of electronic and zero-point Energies=	-2220.353877
Sum of electronic and thermal Energies=	-2220.318753
Sum of electronic and thermal Enthalpies=	-2220.317809
Sum of electronic and thermal Free Energies=	-2220.419783

Item	Value	Threshold	Converged?
Maximum Force	0.000335	0.000450	YES
RMS Force	0.000039	0.000300	YES

Table S7. DFT-optimized geometry of the two-electron-reduced form **[PtL$^+$-C2-MV$^+$]**$^{2+}$ (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	3.361958	-0.014237	-0.411785
N2	3.012714	2.024354	-0.421189
C3	2.208878	4.668384	-0.372617
C4	3.904118	2.995325	-0.177630
C5	1.698139	2.338997	-0.642341
C6	1.283071	3.664716	-0.620414
C7	3.539150	4.331522	-0.146262
C8	0.802469	1.195586	-0.854508
C9	-0.570935	-1.197359	-1.086159
C10	-0.553361	1.230343	-1.069179
N11	1.443757	0.000903	-0.784322
C12	0.785857	-1.183852	-0.869574
C13	-1.295297	0.022140	-1.192647
C14	1.667073	-2.341555	-0.674600
C15	3.484567	-4.366326	-0.225065
C16	1.235346	-3.662034	-0.676465
N17	2.986141	-2.047514	-0.450556
C18	3.866054	-3.034348	-0.230387
C19	2.149278	-4.681916	-0.452299
C20	-2.714852	0.036702	-1.419251
N21	-5.495746	0.077196	-1.882372
C22	-3.453567	1.246635	-1.519697
C23	-3.479760	-1.150903	-1.577852
C24	-4.825425	-1.105857	-1.805391
C25	-4.798184	1.240393	-1.741484
Cl26	5.701363	-0.029947	0.074128
C27	-3.727969	-1.238323	1.608406
C28	-5.139051	1.111504	1.527593
C29	-2.991701	-0.019649	1.710422
C30	-5.082850	-1.240123	1.485196
N31	-5.807991	-0.078921	1.451946

C32	-3.782968	1.166317	1.646355
C33	-7.277431	-0.147912	1.458660
C34	-7.880828	-0.541774	0.121205
C35	-7.779718	0.527671	-0.976420
C36	-6.937568	0.138429	-2.178544
C37	-1.583992	0.005399	1.938257
N38	1.177361	0.049474	2.541055
C39	-0.849403	1.219430	2.087026
C40	-0.807486	-1.184549	2.075587
C41	0.520471	-1.136133	2.380496
C42	0.480223	1.213552	2.390303
C43	2.550241	0.073170	3.044778
H44	4.924862	2.662440	-0.005521
H45	0.238875	3.908479	-0.791468
H46	4.290217	5.088162	0.053089
H47	1.892040	5.706658	-0.352808
H48	-1.051425	2.192873	-1.118008
H49	-1.083788	-2.151785	-1.145039
H50	0.187944	-3.889589	-0.850183
H51	4.891740	-2.718449	-0.056022
H52	1.819593	-5.716371	-0.452153
H53	4.227271	-5.135884	-0.044691
H54	-2.978659	2.216522	-1.421883
H55	-3.024110	-2.133989	-1.531733
H56	-5.417974	-2.004609	-1.946568
H57	-5.374021	2.156865	-1.830303
H58	-3.232418	-2.203444	1.633870
H59	-5.657084	-2.160535	1.430196
H60	-3.332286	2.151964	1.701277
H61	-5.753752	2.006692	1.505855
H62	-7.648554	0.831605	1.778796
H63	-7.565875	-0.868383	2.232352
H64	-7.437640	-1.491277	-0.213305
H65	-8.935016	-0.771696	0.312490
H66	-7.407694	1.476622	-0.562913
H67	-8.779899	0.755603	-1.361507

H68	-7.069889	0.860970	-2.990967
H69	-7.233661	-0.842461	-2.563898
H70	-1.324332	2.189184	1.973273
H71	-1.248237	-2.169340	1.955281
H72	1.118708	-2.032389	2.517035
H73	1.048248	2.128254	2.535118
H74	3.062108	0.959731	2.661689
H75	3.079544	-0.815899	2.693358
H76	2.550075	0.092972	4.139415

---

[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -2260.22426227 A.U. after 7 cycles

	1	2	3
	A	A	A
Frequencies --	23.1773	27.6400	30.1945
Red. masses --	5.0881	5.7576	6.2610

Zero-point correction= 0.616740 (Hartree/Particle)
Thermal correction to Energy= 0.653086
Thermal correction to Enthalpy= 0.654030
Thermal correction to Gibbs Free Energy= 0.549683
Sum of electronic and zero-point Energies= -2259.607522
Sum of electronic and thermal Energies= -2259.571176
Sum of electronic and thermal Enthalpies= -2259.570232
Sum of electronic and thermal Free Energies= -2259.674579

Item	Value	Threshold	Converged?
Maximum Force	0.000182	0.000450	YES
RMS Force	0.000023	0.000300	YES

Table S8. DFT-optimized geometry of the two-electron-reduced form **[PtL^{+}-C3-MV^{+}]**$^{2+}$ (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	3.518644	0.152132	-0.401451
N2	3.368293	-1.908716	-0.429109
C3	2.820525	-4.616831	-0.404651
C4	4.349376	-2.791670	-0.198962
C5	2.088686	-2.344811	-0.649701
C6	1.800995	-3.703319	-0.636807
C7	4.113898	-4.157056	-0.182191
C8	1.088794	-1.288751	-0.857644
C9	-0.497354	0.967654	-1.120303
C10	-0.259098	-1.451126	-1.071407
N11	1.614123	-0.039759	-0.796097
C12	0.853361	1.082415	-0.891982
C13	-1.101779	-0.314111	-1.213642
C14	1.625376	2.315113	-0.693759
C15	3.260456	4.493103	-0.250143
C16	1.085034	3.595128	-0.724334
N17	2.961754	2.138076	-0.446190
C18	3.752185	3.198455	-0.228750
C19	1.906977	4.691219	-0.502929
C20	-2.516468	-0.451123	-1.434934
N21	-5.309651	-0.674383	-1.736201
C22	-3.199464	-1.678923	-1.254963
C23	-3.324245	0.631953	-1.876070
C24	-4.672552	0.493721	-2.033995
C25	-4.555820	-1.755915	-1.385733
Cl26	5.833263	0.374995	0.144824
C27	-6.775765	-0.846138	-1.831106
C28	-7.593892	0.434395	-1.787436
C29	-7.386196	1.407414	-0.626365
C30	-7.856020	0.987820	0.765198
C31	-7.164729	-0.181607	1.446875

N32	-5.691598	-0.117633	1.488806
C33	-2.870447	-0.101150	1.773946
C34	-4.996262	1.063337	1.519790
C35	-4.994035	-1.281156	1.664284
C36	-3.640039	-1.300503	1.806165
C37	-3.640050	1.095415	1.637194
C38	-1.457271	-0.094767	1.952289
N39	1.312529	-0.073770	2.517517
C40	-0.699924	1.112862	2.046362
C41	-0.693357	-1.292326	2.095383
C42	0.639112	-1.255866	2.378839
C43	0.631150	1.095561	2.339031
C44	2.681310	-0.065347	3.029952
H45	5.333545	-2.363424	-0.024908
H46	0.783006	-4.042961	-0.802322
H47	4.934191	-4.840984	0.006519
H48	2.603468	-5.680568	-0.393510
H49	-0.664876	-2.454843	-1.145484
H50	-1.106964	1.865813	-1.157361
H51	0.025415	3.731733	-0.918732
H52	4.798599	2.973329	-0.036945
H53	1.491287	5.693936	-0.526463
H54	3.932459	5.325212	-0.070714
H55	-2.681090	-2.578566	-0.937931
H56	-2.890910	1.589361	-2.144407
H57	-5.283784	1.301082	-2.418329
H58	-5.100385	-2.678057	-1.205744
H59	-7.053154	-1.518119	-1.010469
H60	-6.993144	-1.382720	-2.762796
H61	-7.450011	0.981682	-2.728611
H62	-8.643453	0.110599	-1.806788
H63	-6.337795	1.728033	-0.580981
H64	-7.942875	2.317697	-0.880904
H65	-8.920585	0.719907	0.727640
H66	-7.807072	1.867736	1.420624
H67	-7.417350	-1.124205	0.948200

H68	-7.535009	-0.267868	2.476404
H69	-5.581689	1.973454	1.460468
H70	-5.587516	-2.190942	1.688392
H71	-3.174209	-2.270828	1.944661
H72	-3.170740	2.073853	1.642376
H73	-1.161813	2.085675	1.908218
H74	-1.148391	-2.271679	1.979383
H75	1.230869	-2.156285	2.517347
H76	1.213074	2.006162	2.452284
H77	2.675990	-0.095984	4.124472
H78	3.188843	0.840530	2.689615
H79	3.219366	-0.934152	2.641857

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[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -2299.49883255 A.U. after 7 cycles

	1	2	3
	A	A	A
Frequencies --	14.6111	27.7998	30.8997
Red. masses --	5.6040	6.0367	5.4630

Zero-point correction= 0.645880 (Hartree/Particle)
Thermal correction to Energy= 0.683155
Thermal correction to Enthalpy= 0.684099
Thermal correction to Gibbs Free Energy= 0.577899
Sum of electronic and zero-point Energies= -2298.852952
Sum of electronic and thermal Energies= -2298.815678
Sum of electronic and thermal Enthalpies= -2298.814733
Sum of electronic and thermal Free Energies= -2298.920934

Item	Value	Threshold	Converged?
Maximum Force	0.000096	0.000450	YES
RMS Force	0.000010	0.000300	YES

Table S9. DFT-optimized geometry of the two-electron-reduced form **[PtL^{+}-C4-MV^{+}]**$^{2+}$ (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	3.741605	0.307147	-0.411423
N2	3.067805	2.259041	-0.429580
C3	1.849366	4.739368	-0.427783
C4	3.799075	3.368569	-0.254501
C5	1.712802	2.350385	-0.613761
C6	1.089727	3.592450	-0.609330
C7	3.224580	4.629283	-0.250289
C8	1.010704	1.075910	-0.793774
C9	0.041138	-1.514665	-0.983604
C10	-0.334602	0.886350	-1.009890
N11	1.835105	-0.001705	-0.715412
C12	1.382260	-1.275668	-0.803513
C13	-0.870568	-0.422939	-1.096930
C14	2.453614	-2.271430	-0.655643
C15	4.597433	-3.965088	-0.290885
C16	2.248188	-3.644154	-0.660684
N17	3.711672	-1.761660	-0.472358
C18	4.751653	-2.587176	-0.295043
C19	3.327704	-4.498664	-0.476877
C20	-2.279094	-0.639525	-1.275827
N21	-5.057577	-1.058781	-1.599886
C22	-3.169630	0.397696	-1.673102
C23	-2.881064	-1.910941	-1.097583
C24	-4.227749	-2.080258	-1.230673
C25	-4.506230	0.169174	-1.819423
Cl26	6.068699	0.678885	-0.043980
C27	-3.715831	-1.097424	1.925754
C28	-4.627478	1.462180	1.565180
C29	-2.744129	-0.054321	1.852759
C30	-5.051485	-0.831537	1.845693
N31	-5.527693	0.436370	1.665010
C32	-3.284917	1.252063	1.660206

C33	-1.347679	-0.286679	2.024709
N34	1.402869	-0.739410	2.494631
C35	-0.403269	0.780449	2.161548
C36	-0.792931	-1.594229	2.124040
C37	0.539206	-1.789554	2.340777
C38	0.912772	0.532738	2.408483
C39	2.784868	-0.982525	2.902522
C40	-6.505306	-1.283035	-1.400735
C41	-7.493846	-0.352361	-2.101787
C42	-7.256885	1.560215	0.267750
C43	-6.969251	0.690385	1.492681
C44	-8.694839	-0.047112	-1.192690
C45	-8.587082	1.276756	-0.426080
H46	4.865901	3.209957	-0.116131
H47	0.014460	3.659248	-0.745367
H48	3.850163	5.503293	-0.106191
H49	1.369245	5.713203	-0.423801
H50	-0.987259	1.753322	-1.053587
H51	-0.307616	-2.537999	-1.074384
H52	1.247794	-4.042379	-0.800226
H53	5.715300	-2.104114	-0.151584
H54	3.175362	-5.573526	-0.476141
H55	5.463390	-4.600567	-0.141132
H56	-2.810657	1.394773	-1.904792
H57	-2.303465	-2.777734	-0.793568
H58	-4.707007	-3.036991	-1.047075
H59	-5.174106	0.958017	-2.143444
H60	-3.429013	-2.130895	2.090579
H61	-5.798964	-1.614401	1.939659
H62	-2.642939	2.122733	1.574769
H63	-5.043572	2.453651	1.420727
H64	-0.709167	1.820844	2.109694
H65	-1.408548	-2.481367	2.014615
H66	0.976895	-2.780329	2.420984
H67	1.639716	1.328974	2.544333
H68	3.411897	-0.156750	2.557348
H69	3.143166	-1.908918	2.443439
H70	2.852981	-1.072624	3.991449

H71	-6.700336	-2.318648	-1.696795
H72	-7.827052	-0.829125	-3.028960
H73	-6.439939	1.438446	-0.456166
H74	-7.451640	-0.289822	1.405372
H75	-7.361000	1.151499	2.406939
H76	-7.232903	2.617480	0.556964
H77	-7.032953	0.592264	-2.414529
H78	-8.853130	-0.888531	-0.501850
H79	-9.607723	-0.002459	-1.797428
H80	-9.408022	1.341255	0.301398
H81	-6.661563	-1.250127	-0.313313
H82	-8.753347	2.094761	-1.140162

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[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -2338.77328990 A.U. after 6 cycles

	1	2	3
	A	A	A
Frequencies --	27.3203	32.0528	38.6737
Red. masses --	5.9362	7.4036	6.1691

Zero-point correction=	0.675834 (Hartree/Particle)
Thermal correction to Energy=	0.713981
Thermal correction to Enthalpy=	0.714925
Thermal correction to Gibbs Free Energy=	0.607882
Sum of electronic and zero-point Energies=	-2338.097456
Sum of electronic and thermal Energies=	-2338.059309
Sum of electronic and thermal Enthalpies=	-2338.058365
Sum of electronic and thermal Free Energies=	-2338.165408

Item	Value	Threshold	Converged?
Maximum Force	0.000128	0.000450	YES
RMS Force	0.000016	0.000300	YES

Table S10. DFT-optimized geometry of the two-electron-reduced form **HT-[PtL^{+}·-C1-MV·$^{+}$]$_2^{4+}$** (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	-10.986140	0.884054	-0.383090
N2	-11.229084	-1.033289	0.350854
C3	-11.212150	-3.617880	1.328744
C4	-12.340521	-1.558856	0.883771
C5	-10.080185	-1.777038	0.285187
C6	-10.059040	-3.075633	0.778360
C7	-12.371275	-2.851674	1.381268
C8	-8.923242	-1.102805	-0.316191
C9	-6.975936	0.518313	-1.431132
C10	-7.662838	-1.618798	-0.505260
N11	-9.191708	0.174268	-0.693431
C12	-8.251325	0.987862	-1.231477
C13	-6.633058	-0.815714	-1.063751
C14	-8.756333	2.339793	-1.508656
C15	-9.893165	4.820987	-1.898136
C16	-7.985363	3.370909	-2.028524
N17	-10.071601	2.553706	-1.190786
C18	-10.620569	3.761132	-1.379381
C19	-8.557554	4.620498	-2.226869
C20	-5.303578	-1.322865	-1.261511
N21	-2.730762	-2.340545	-1.829809
C22	-4.952394	-2.683197	-1.033975
C23	-4.244166	-0.503492	-1.736451
C24	-3.011871	-1.018109	-2.013756
C25	-3.704037	-3.150769	-1.320030
Cl26	-13.168603	1.764169	0.008921
N27	2.250224	-2.066679	-1.832511
C28	4.708503	-0.662496	-1.747746
C29	2.298514	-0.767931	-1.407329
C30	3.428961	-2.693759	-2.138132
C31	4.621833	-2.039946	-2.108576

C32	3.468802	-0.072740	-1.353238
C33	5.921806	0.077800	-1.843136
N34	8.291873	1.582634	-2.164852
C35	6.024255	1.441019	-1.441374
C36	7.119391	-0.479500	-2.389664
C37	8.242964	0.273436	-2.548612
C38	7.178755	2.147710	-1.608659
C39	9.451923	2.407210	-2.500282
C40	-1.445700	-2.913993	-2.252776
C41	-0.258226	-2.019896	-1.923021
C42	1.011132	-2.849018	-1.943490
Pt43	10.986248	0.883833	0.381980
N44	11.228363	-1.033671	-0.351773
C45	11.210246	-3.618349	-1.329419
C46	12.339419	-1.559614	-0.885133
C47	10.079291	-1.777103	-0.285482
C48	10.057544	-3.075727	-0.778562
C49	12.369580	-2.852487	-1.382512
C50	8.922791	-1.102469	0.316292
C51	6.976137	0.519602	1.431049
C52	7.662329	-1.618091	0.505969
N53	9.191743	0.174608	0.693202
C54	8.251700	0.988615	1.231217
C55	6.632843	-0.814484	1.064248
C56	8.757329	2.340303	1.508404
C57	9.895541	4.820719	1.898778
C58	7.987086	3.371507	2.029167
N59	10.072559	2.553757	1.190049
C60	10.622177	3.760825	1.379026
C61	8.559990	4.620694	2.228017
C62	5.303265	-1.321188	1.262440
N63	2.730399	-2.338278	1.831652
C64	4.952062	-2.681710	1.036142
C65	4.243804	-0.501338	1.736451
C66	3.011436	-1.015661	2.014194
C67	3.703756	-3.148991	1.322727

Cl68	13.168570	1.763633	-0.011419
N69	-2.250028	-2.065085	1.833924
C70	-4.708640	-0.661517	1.747859
C71	-2.298562	-0.766579	1.407876
C72	-3.428712	-2.692309	2.139479
C73	-4.621739	-2.038798	2.109282
C74	-3.468998	-0.071727	1.353151
C75	-5.922014	0.078661	1.843044
N76	-8.292207	1.583396	2.164404
C77	-6.024549	1.441815	1.441066
C78	-7.119579	-0.478612	2.389648
C79	-8.243191	0.274287	2.548469
C80	-7.179088	2.148462	1.608220
C81	-9.452298	2.407942	2.499744
C82	1.445915	-2.912075	2.255645
C83	0.258336	-2.017071	1.928475
C84	-1.010755	-2.846887	1.945185
H85	-13.209178	-0.905020	0.898448
H86	-9.144330	-3.658556	0.730494
H87	-13.291409	-3.242115	1.802438
H88	-11.204064	-4.632893	1.714035
H89	-7.458875	-2.634978	-0.183070
H90	-6.239768	1.177273	-1.878520
H91	-6.941164	3.197998	-2.270897
H92	-11.667210	3.850131	-1.098523
H93	-7.961519	5.432262	-2.632345
H94	-10.372766	5.784030	-2.035867
H95	-5.665694	-3.402245	-0.645569
H96	-4.375023	0.562128	-1.890767
H97	-2.211610	-0.398481	-2.402111
H98	-3.428497	-4.189313	-1.165682
H99	1.357412	-0.301600	-1.137098
H100	3.348405	-3.738650	-2.422021
H101	5.504383	-2.613955	-2.373174
H102	3.410459	0.960649	-1.028398
H103	5.192674	1.957378	-0.971355

H104	7.168580	-1.512104	-2.720378
H105	9.153717	-0.126448	-2.985577
H106	7.268897	3.190686	-1.318738
H107	9.337615	2.834717	-3.501701
H108	9.545679	3.215203	-1.770638
H109	10.355895	1.793872	-2.457369
H110	-1.484792	-3.121787	-3.329224
H111	-0.990384	-3.591417	1.135219
H112	-0.174897	-1.198703	-2.645948
H113	-0.418394	-1.568868	-0.933257
H114	1.068983	-3.406699	-2.886764
H115	0.993561	-3.595264	-1.135020
H116	13.208252	-0.906023	-0.900277
H117	9.142668	-3.658358	-0.730296
H118	13.289413	-3.243231	-1.804058
H119	11.201683	-4.633380	-1.714653
H120	7.458081	-2.634347	0.184212
H121	6.240209	1.178897	1.878349
H122	6.942944	3.198939	2.272024
H123	11.668739	3.849475	1.097764
H124	7.964572	5.432485	2.634344
H125	10.375685	5.783436	2.036893
H126	5.665364	-3.401109	0.648392
H127	4.374692	0.564442	1.889656
H128	2.211140	-0.395663	2.401918
H129	3.428250	-4.187704	1.169500
H130	-1.357489	-0.300304	1.137438
H131	-3.348005	-3.737056	2.423841
H132	-5.504216	-2.612900	2.373933
H133	-3.410846	0.961474	1.027681
H134	-5.192980	1.958151	0.971015
H135	-7.168660	-1.511135	2.720622
H136	-9.153882	-0.125527	2.985627
H137	-7.269231	3.191417	1.318230
H138	-10.356346	1.794748	2.456177
H139	-9.545647	3.216247	1.770402

H140	-9.338352	2.835014	3.501388
H141	1.349293	-3.875641	1.742025
H142	1.485778	-3.121727	3.331693
H143	0.419218	-1.563512	0.940009
H144	0.174473	-1.197891	2.653604
H145	-1.350553	-3.878476	-1.740624
H146	-1.069761	-3.406775	2.887060

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[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -4441.87339054 A.U. after 6 cycles

	1	2	3
	A	A	A
Frequencies --	10.9912	16.4076	17.6127
Red. masses --	8.3708	6.4531	7.3756

Zero-point correction=	1.182511 (Hartree/Particle)
Thermal correction to Energy=	1.253438
Thermal correction to Enthalpy=	1.254382
Thermal correction to Gibbs Free Energy=	1.076916
Sum of electronic and zero-point Energies=	-4440.690879
Sum of electronic and thermal Energies=	-4440.619953
Sum of electronic and thermal Enthalpies=	-4440.619008
Sum of electronic and thermal Free Energies=	-4440.796475

Item	Value	Threshold	Converged?
Maximum Force	0.000339	0.000450	YES
RMS Force	0.000039	0.000300	YES

Table S11. DFT-optimized geometry of the two-electron-reduced form **HH(σ)-[PtL^{+}·-C1-MV^{+}·]$_2^{4+}$** (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	6.353801	-0.273296	-1.468929
N2	6.398273	-2.279014	-0.960101
C3	6.119433	-4.942650	-0.271455
C4	7.482101	-3.030262	-0.718635
C5	5.146682	-2.830715	-0.868660
C6	4.995038	-4.167631	-0.520220
C7	7.382251	-4.368343	-0.372417
C8	4.033212	-1.926094	-1.175817
C9	2.223302	0.048030	-1.893506
C10	2.694446	-2.236246	-1.209553
N11	4.439641	-0.667388	-1.490175
C12	3.572373	0.310106	-1.855111
C13	1.732125	-1.253763	-1.570044
C14	4.246684	1.576558	-2.170780
C15	5.702256	3.866383	-2.664461
C16	3.587266	2.736851	-2.558588
N17	5.610464	1.567424	-2.044726
C18	6.315166	2.681098	-2.288571
C19	4.318174	3.891534	-2.803810
C20	0.334738	-1.574390	-1.646984
N21	-2.389177	-2.250099	-1.983250
C22	-0.150690	-2.903299	-1.485606
C23	-0.665528	-0.605063	-1.934919
C24	-1.974719	-0.956065	-2.102117
C25	-1.466685	-3.204562	-1.657974
Cl26	8.704261	0.160473	-1.569445
N27	-7.334247	-1.504648	-1.760445
C28	-9.716507	0.001265	-1.525764
C29	-7.286635	-0.161663	-1.515643
C30	-8.560224	-2.114308	-1.820061
C31	-9.719612	-1.412440	-1.714086

C32	-8.421467	0.584404	-1.400388
C33	-10.913446	0.777260	-1.528119
N34	-13.273303	2.321393	-1.619603
C35	-10.906383	2.195313	-1.387745
C36	-12.202154	0.196490	-1.708984
C37	-13.326867	0.963196	-1.752920
C38	-12.059989	2.919556	-1.440423
C39	-14.477659	3.126739	-1.801790
C40	-3.756786	-2.680439	-2.304060
C41	-4.827791	-1.636123	-2.025719
C42	-6.151500	-2.363841	-1.889082
H43	8.438683	-2.520500	-0.811049
H44	4.000652	-4.597623	-0.443058
H45	8.283410	-4.941492	-0.183016
H46	6.009008	-5.987922	0.000756
H47	2.387201	-3.243377	-0.944813
H48	1.543494	0.834855	-2.204834
H49	2.505556	2.738069	-2.657354
H50	7.392401	2.593159	-2.169407
H51	3.808452	4.804705	-3.097349
H52	6.306519	4.748448	-2.847047
H53	0.509152	-3.728084	-1.240634
H54	-0.427730	0.449853	-2.027556
H55	-2.732490	-0.219565	-2.344778
H56	-1.844995	-4.216960	-1.554523
H57	-6.304553	0.289688	-1.429244
H58	-8.545164	-3.189319	-1.972021
H59	-10.644752	-1.975489	-1.781822
H60	-8.291099	1.645883	-1.219141
H61	-9.984096	2.745592	-1.234284
H62	-12.332267	-0.874918	-1.816882
H63	-14.316408	0.538861	-1.894048
H64	-12.072260	4.000816	-1.346319
H65	-14.645114	3.329489	-2.864057
H66	-14.365707	4.074148	-1.270602
H67	-15.341711	2.590687	-1.401922

H68	-3.790239	-2.989119	-3.356458
H69	-3.946298	-3.576980	-1.701346
H70	-4.877076	-0.899218	-2.836980
H71	-4.585005	-1.095396	-1.099909
H72	-6.312445	-3.004809	-2.765524
H73	-6.117622	-3.031795	-1.014225
Pt74	5.610421	1.596438	1.556550
N75	6.156524	-0.315817	2.116414
C76	6.581426	-2.950681	2.830538
C77	7.396695	-0.749668	2.377583
C78	5.094143	-1.177702	2.189707
C79	5.296243	-2.503941	2.553306
C80	7.650022	-2.064594	2.735376
C81	3.788149	-0.601669	1.838252
C82	1.543852	0.854327	1.122922
C83	2.569443	-1.238624	1.811695
N84	3.869994	0.711615	1.505690
C85	2.783113	1.446509	1.152009
C86	1.388839	-0.519196	1.454936
C87	3.103084	2.850101	0.867644
C88	3.920691	5.443357	0.401218
C89	2.169648	3.803597	0.478044
N90	4.422025	3.194830	1.012540
C91	4.814740	4.456373	0.784276
C92	2.579552	5.109079	0.244523
C93	0.090219	-1.131318	1.470238
N94	-2.488732	-2.273217	1.705751
C95	-0.112781	-2.516795	1.719002
C96	-1.104444	-0.380967	1.284571
C97	-2.335817	-0.951135	1.412873
C98	-1.366320	-3.044671	1.825432
Cl99	7.720141	2.708962	1.704186
N100	-7.462873	-1.886197	1.830255
C101	-9.872167	-0.420361	1.583384
C102	-7.464099	-0.522467	1.919099
C103	-8.647845	-2.521864	1.573745

C104	-9.819743	-1.838790	1.449031
C105	-8.609372	0.204154	1.789394
C106	-11.095086	0.315156	1.560377
N107	-13.516443	1.760629	1.624467
C108	-11.137875	1.730100	1.733508
C109	-12.364951	-0.314877	1.418751
C110	-13.520921	0.405439	1.450928
C111	-12.320364	2.404925	1.760934
C112	-14.775223	2.485762	1.762368
C113	-3.798156	-2.928418	1.883002
C114	-4.967930	-1.965798	1.848735
C115	-6.269657	-2.703300	2.105324
H116	8.183653	-0.004569	2.286826
H117	4.452486	-3.185745	2.612265
H118	8.668273	-2.379396	2.937215
H119	6.746895	-3.986658	3.111910
H120	2.518355	-2.285929	2.093571
H121	0.689152	1.455213	0.830642
H122	1.127087	3.524387	0.357180
H123	5.876682	4.649018	0.920040
H124	1.855770	5.858890	-0.060090
H125	4.279083	6.452023	0.226233
H126	0.720448	-3.204289	1.825090
H127	-1.087856	0.681599	1.071210
H128	-3.234369	-0.357297	1.299643
H129	-1.536028	-4.099108	2.021713
H130	-6.512592	-0.038582	2.102202
H131	-8.596453	-3.602942	1.487405
H132	-10.710704	-2.423552	1.245008
H133	-8.516198	1.281727	1.869838
H134	-10.231888	2.317736	1.837169
H135	-12.454158	-1.386553	1.275054
H136	-14.498203	-0.056183	1.347046
H137	-12.372583	3.481409	1.892223
H138	-14.620245	3.533281	1.495600
H139	-15.143566	2.428142	2.791202

H140	-15.521553	2.052641	1.091305
H141	-3.899354	-3.694091	1.100471
H142	-3.771217	-3.454353	2.845546
H143	-5.024536	-1.472693	0.868103
H144	-4.817994	-1.186626	2.606648
H145	-6.337556	-3.590053	1.462054
H146	-6.313736	-3.054705	3.144106

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[a]Part of the Gaussian output file:

SCF Done: E(RM06) = -4441.88394442 A.U. after 7 cycles

	1	2	3
	A	A	A
Frequencies --	14.5135	17.1914	25.4146
Red. masses --	6.5746	6.4069	7.3326

Zero-point correction=	1.180751 (Hartree/Particle)
Thermal correction to Energy=	1.251809
Thermal correction to Enthalpy=	1.252754
Thermal correction to Gibbs Free Energy=	1.077131
Sum of electronic and zero-point Energies=	-4440.703193
Sum of electronic and thermal Energies=	-4440.632135
Sum of electronic and thermal Enthalpies=	-4440.631191
Sum of electronic and thermal Free Energies=	-4440.806814

Item	Value	Threshold	Converged?
Maximum Force	0.000245	0.000450	YES
RMS Force	0.000032	0.000300	YES

Table S12. DFT-optimized geometry of the two-electron-reduced form **HH(C_2)-[PtL^{+}·-C1-MV^{+}·]$_2^{4+}$** (closed-shell singlet), computed at the M06/SDD(Pt)/6-31G**(HCNOCl) level using PCM.[a]

Atom	X	Y	Z
Pt1	-5.890889	-1.639430	1.438801
N2	-6.227569	0.191028	2.334253
C3	-6.362942	2.699883	3.484553
C4	-7.398224	0.665532	2.777010
C5	-5.091003	0.947770	2.446670
C6	-5.147277	2.210069	3.023047
C7	-7.506460	1.919630	3.359541
C8	-3.866059	0.330100	1.916429
C9	-1.789768	-1.195135	0.892373
C10	-2.603394	0.873278	1.875271
N11	-4.077637	-0.911170	1.415844
C12	-3.076650	-1.682512	0.911228
C13	-1.511205	0.115845	1.353382
C14	-3.540540	-2.991769	0.440390
C15	-4.622530	-5.399608	-0.361779
C16	-2.717605	-3.956962	-0.129080
N17	-4.881900	-3.231209	0.591178
C18	-5.403860	-4.401762	0.198269
C19	-3.260373	-5.170228	-0.528986
C20	-0.190809	0.676381	1.286507
N21	2.408113	1.782683	1.274147
C22	0.053470	2.058209	1.524565
C23	0.969231	-0.101084	1.018018
C24	2.211839	0.459182	1.000539
C25	1.315802	2.569797	1.498719
Cl26	-8.086679	-2.568941	1.547814
N27	7.389927	1.511278	1.436448
C28	9.842702	0.122153	1.620150
C29	7.425405	0.148697	1.352448
C30	8.572515	2.189689	1.553451
C31	9.764480	1.543015	1.662954

C32	8.594737	-0.544505	1.445348
C33	11.075346	-0.577086	1.799475
N34	13.506003	-1.949823	2.185271
C35	11.183231	-1.986749	1.658081
C36	12.284564	0.091523	2.155628
C37	13.445575	-0.598415	2.343470
C38	12.373598	-2.627983	1.835006
C39	14.745759	-2.691697	2.397844
C40	3.714469	2.440611	1.154625
C41	4.891322	1.512340	1.370087
C42	6.171742	2.312433	1.271737
Pt43	-6.353533	0.829889	-1.202174
N44	-6.033392	-1.013580	-2.083993
C45	-5.288469	-3.406986	-3.247052
C46	-6.961147	-1.931034	-2.387392
C47	-4.711508	-1.258587	-2.344241
C48	-4.325758	-2.455452	-2.935944
C49	-6.625726	-3.145065	-2.966682
C50	-3.781069	-0.192500	-1.947536
C51	-2.381623	2.068644	-1.161341
C52	-2.411969	-0.189090	-2.070921
N53	-4.410254	0.875216	-1.398770
C54	-3.742981	1.992536	-0.990762
C55	-1.657975	0.959504	-1.677514
C56	-4.628225	3.009824	-0.415348
C57	-6.474447	4.791787	0.602020
C58	-4.193246	4.215724	0.124350
N59	-5.965078	2.702621	-0.425028
C60	-6.858027	3.571923	0.069979
C61	-5.120454	5.113449	0.633431
C62	-0.234051	1.001610	-1.854296
N63	2.492083	1.114213	-2.567252
C64	0.522037	-0.161319	-2.169765
C65	0.522776	2.205832	-1.779004
C66	1.836754	2.237803	-2.150362
C67	1.840078	-0.082413	-2.515230

Cl68	-8.734269	0.812294	-1.006006
N69	7.495412	0.717949	-2.302881
C70	10.125812	0.132957	-1.453124
C71	8.362080	1.724670	-1.976027
C72	7.887743	-0.575913	-2.095264
C73	9.148416	-0.881402	-1.678575
C74	9.629472	1.469959	-1.543381
C75	11.501146	-0.169612	-1.227228
N76	14.243299	-0.769448	-0.917715
C77	12.474912	0.830904	-0.940581
C78	12.017548	-1.496899	-1.347330
C79	13.346237	-1.757662	-1.201150
C80	13.789591	0.514157	-0.779265
C81	15.672888	-1.037082	-0.804466
C82	3.819779	1.201979	-3.188309
C83	4.983617	0.936537	-2.243965
C84	6.268546	0.996422	-3.057240
H85	-8.251331	0.002668	2.655791
H86	-4.243263	2.804090	3.120159
H87	-8.472441	2.268140	3.708683
H88	-6.414161	3.686249	3.936510
H89	-2.437852	1.859633	2.297143
H90	-1.000568	-1.816811	0.479848
H91	-1.657593	-3.757690	-0.257820
H92	-6.475872	-4.509846	0.346697
H93	-2.622906	-5.930027	-0.970689
H94	-5.082444	-6.333805	-0.665537
H95	-0.754107	2.757995	1.712998
H96	0.911113	-1.168828	0.834863
H97	3.091399	-0.138651	0.793336
H98	1.514503	3.623853	1.666103
H99	6.479668	-0.366509	1.235412
H100	8.496533	3.272511	1.575333
H101	10.650540	2.159782	1.767437
H102	8.529745	-1.626299	1.400959
H103	10.330407	-2.597497	1.379786

H104	12.316988	1.162833	2.319220
H105	14.370321	-0.107523	2.631615
H106	12.482183	-3.701838	1.718721
H107	15.524895	-2.006636	2.733958
H108	14.594559	-3.459388	3.161079
H109	15.061261	-3.172439	1.465854
H110	3.734444	3.249477	1.893393
H111	3.771812	2.914328	0.160868
H112	4.814525	1.021846	2.349439
H113	4.895238	0.729886	0.602295
H114	6.189315	3.094182	2.040709
H115	6.231847	2.820749	0.295885
H116	-7.985966	-1.660477	-2.143709
H117	-3.277408	-2.643083	-3.148948
H118	-7.404712	-3.865188	-3.192908
H119	-4.993574	-4.347750	-3.702937
H120	-1.919948	-1.042522	-2.527475
H121	-1.863617	2.973743	-0.858657
H122	-3.131518	4.444341	0.152801
H123	-7.897074	3.252696	0.028125
H124	-4.786852	6.057150	1.054320
H125	-7.227503	5.468211	0.991852
H126	0.074442	-1.150221	-2.163723
H127	0.063693	3.151499	-1.510479
H128	2.405269	3.163021	-2.184696
H129	2.419912	-0.956031	-2.798423
H130	7.989218	2.735658	-2.114588
H131	7.148140	-1.339245	-2.319015
H132	9.382378	-1.933716	-1.551538
H133	10.260466	2.327215	-1.334304
H134	12.198072	1.873038	-0.814526
H135	11.373383	-2.335448	-1.587842
H136	13.755119	-2.758163	-1.308675
H137	14.545117	1.259250	-0.549238
H138	16.051344	-0.647106	0.145945
H139	16.215005	-0.559108	-1.625564

H140	15.847018	-2.113412	-0.845093
H141	3.834849	0.487401	-4.019240
H142	3.907938	2.200926	-3.629448
H143	4.872949	-0.048263	-1.765523
H144	5.007326	1.700902	-1.455253
H145	6.223789	0.277027	-3.884044
H146	6.382926	1.987070	-3.510195

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[a]Part of the Gaussian output file:

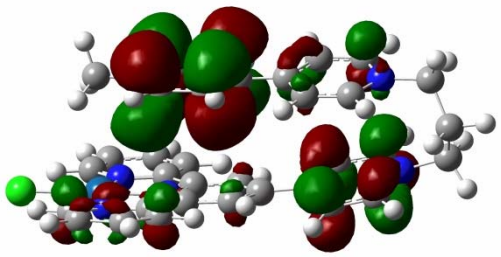
SCF Done: E(RM06) = -4441.88579140 A.U. after 6 cycles

	1	2	3
	A	A	A
Frequencies --	12.0766	15.7519	18.4728
Red. masses --	5.3396	6.1160	7.1426

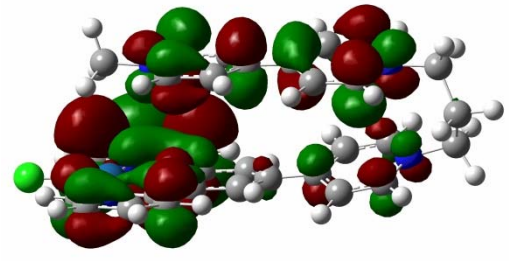
Zero-point correction=	1.180311 (Hartree/Particle)
Thermal correction to Energy=	1.251619
Thermal correction to Enthalpy=	1.252563
Thermal correction to Gibbs Free Energy=	1.075213
Sum of electronic and zero-point Energies=	-4440.705481
Sum of electronic and thermal Energies=	-4440.634172
Sum of electronic and thermal Enthalpies=	-4440.633228
Sum of electronic and thermal Free Energies=	-4440.810578

Item	Value	Threshold	Converged?
Maximum Force	0.000144	0.000450	YES
RMS Force	0.000016	0.000300	YES

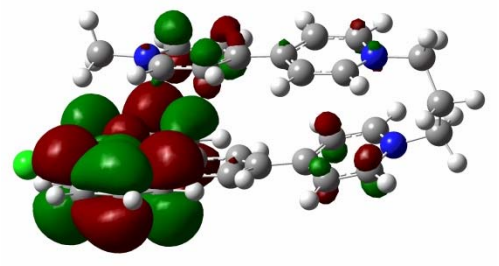
Table S13. Electronic transitions computed by TD-DFT for the one-electron-reduced form with a stacking geometry **[PtL**$^{1.5+}$**-C1-MV**$^{1.5+}$**]**$^{3+}$ (doublet), for which part of the Gaussian output is shown. Relevant MO's are shown below:



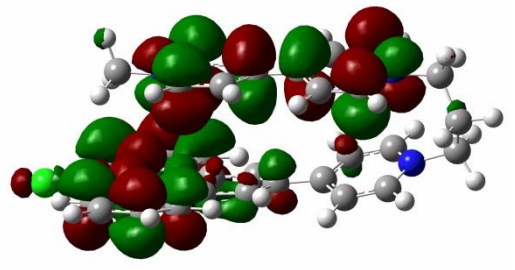
MO164A



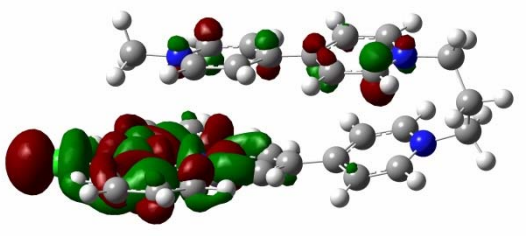
MO163A



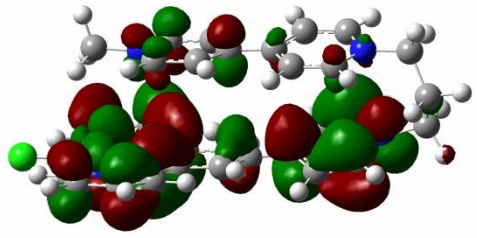
MO162A



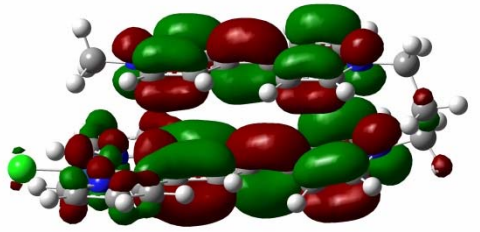
MO161A



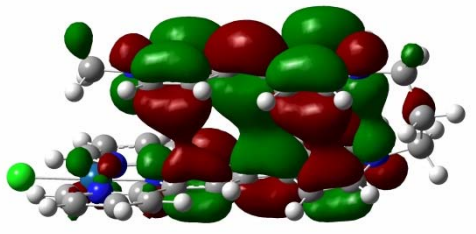
MO160A



MO158A



MO156A



MO155A (SOMO)

Excitation energies and oscillator strengths (λ > 200 nm, f > 0.02 only):

Excited State 1: 2.004-A 0.8059 eV 1538.46 nm f=0.0851 <S**2>=0.754
155A ->156A 1.00474
155A <-156A -0.12151

Excited State 3: 2.013-A 1.5867 eV 781.41 nm f=0.1745 <S**2>=0.763

155A ->158A 0.98475

Excited State 5: 2.144-A 2.3923 eV 518.26 nm f=0.0720 <S**2>=0.899
154A ->156A -0.17131
155A ->160A -0.43652
155A ->161A 0.54001
155A ->162A 0.26195
155A ->163A 0.36671
155A ->164A 0.42917
154B ->155B 0.18836

Excited State 7: 2.113-A 2.4400 eV 508.14 nm f=0.0329 <S**2>=0.866
154A ->160A -0.17557
155A ->160A 0.54012
155A ->161A 0.67022
155A ->163A 0.19948
155A ->164A -0.36422

Excited State 8: 2.091-A 2.4634 eV 503.31 nm f=0.0139 <S**2>=0.843
154A ->160A -0.14978
155A ->160A 0.60262
155A ->162A 0.21430
155A ->164A 0.71731

Excited State 14: 2.548-A 2.9050 eV 426.80 nm f=0.0382 <S**2>=1.373
150A ->157A -0.17420
151A ->157A 0.31898
154A ->156A 0.28130
155A ->161A 0.11981
155A ->162A 0.13373
155A ->163A -0.31749
155A ->165A 0.22807
155A ->166A -0.35796
155A ->167A -0.13677
150B ->157B 0.16375
151B ->157B -0.24594
154B ->155B 0.50355

Excited State 16: 2.746-A 2.9638 eV 418.33 nm f=0.0269 <S**2>=1.635
150A ->157A -0.10931
151A ->157A 0.14686
153A ->156A 0.64708
153A ->158A -0.18235
154A ->156A -0.53771
149B ->155B -0.11066
150B ->157B 0.10633
151B ->157B -0.24283
153B ->155B 0.14573
154B ->155B -0.20250

Excited State 20: 2.455-A 3.1597 eV 392.40 nm f=0.0930 <S**2>=1.256
145A ->156A -0.14306
149A ->156A 0.32016
154A ->156A -0.17448

145B ->156B 0.12918
146B ->163B 0.10297
148B ->159B -0.11760
149B ->155B 0.68189
149B ->156B -0.38750
154B ->155B -0.10512
154B ->156B -0.12310

Excited State 36: 2.558-A 3.5832 eV 346.01 nm f=0.0706 <S**2>=1.385
146A ->156A -0.11904
148A ->156A 0.23774
150A ->156A 0.42500
151A ->158A -0.14816
154A ->157A -0.41841
146B ->155B 0.16870
146B ->156B -0.15243
148B ->156B -0.15716
150B ->155B 0.33396
151B ->158B 0.13306
152B ->160B 0.14066
153B ->156B -0.17163
154B ->157B -0.36323

Excited State 39: 2.972-A 3.6332 eV 341.25 nm f=0.0201 <S**2>=1.958
148A ->156A 0.62107
148A ->158A 0.14260
149A ->156A -0.16952
150A ->156A -0.24110
154A ->157A 0.11663
154A ->160A -0.15311
143B ->155B 0.26256
143B ->158B 0.10408
148B ->155B 0.16810
148B ->156B -0.40101
148B ->158B -0.11324
148B ->164B 0.10092
150B ->155B -0.16331
154B ->157B 0.12399
154B ->160B -0.13119

Excited State 42: 2.882-A 3.7274 eV 332.63 nm f=0.0212 <S**2>=1.826
146A ->156A 0.15353
149A ->156A -0.12451
150A ->156A 0.19880
150A ->157A 0.20705
151A ->157A 0.11717
151A ->158A 0.16263
152A ->160A 0.15682
154A ->156A 0.14509
154A ->158A 0.54074
146B ->155B -0.18764
146B ->156B 0.15718
150B ->155B 0.10713
150B ->156B 0.12401

150B ->157B -0.17656
151B ->156B -0.38085
151B ->157B -0.13604
152B ->160B 0.15642

Excited State 45: 2.176-A 3.8095 eV 325.46 nm f=0.0356 <S**2>=0.934
136A ->160A -0.10079
144A ->160A -0.13455
150A ->156A -0.11804
152A ->159A -0.10685
152A ->160A 0.54136
152A ->161A 0.17328
136B ->160B -0.10951
144B ->160B -0.14835
150B ->155B -0.11659
151B ->156B 0.31456
152B ->159B 0.11529
152B ->160B 0.58962
154B ->157B 0.10392

Excited State 46: 2.530-A 3.8246 eV 324.18 nm f=0.0391 <S**2>=1.350
145A ->156A 0.11512
146A ->156A -0.26379
150A ->156A -0.16559
150A ->157A 0.11189
151A ->157A 0.10851
151A ->158A 0.10835
152A ->160A 0.13104
146B ->155B 0.48060
146B ->156B -0.27259
147B ->155B -0.31179
147B ->156B 0.12856
150B ->156B -0.11002
151B ->155B 0.15991
151B ->156B -0.44680
152B ->160B 0.13270

Excited State 53: 2.807-A 3.9321 eV 315.31 nm f=0.0339 <S**2>=1.720
147A ->156A 0.10604
149A ->156A 0.10598
150A ->157A 0.13716
151A ->157A 0.20613
147B ->155B -0.15282
149B ->155B -0.18691
149B ->156B -0.12074
154B ->156B 0.14506
154B ->158B 0.84485
154B ->161B 0.10514

Excited State 54: 2.107-A 3.9735 eV 312.02 nm f=0.0815 <S**2>=0.860
150A ->157A 0.18269
151A ->157A 0.60426
150B ->156B -0.20622
150B ->157B 0.24450

151B ->157B 0.60517
154B ->158B -0.10747

Excited State 55: 2.938-A 4.0031 eV 309.72 nm f=0.0160 <S**2>=1.908
147A ->162A 0.11862
150A ->156A 0.20518
150A ->158A 0.35485
150A ->161A -0.15661
150A ->163A 0.14912
151A ->157A 0.14739
154A ->162A -0.11994
150B ->155B -0.21533
150B ->156B 0.51224
150B ->157B 0.11813
150B ->158B -0.31839
150B ->161B -0.17358
150B ->162B -0.11451
151B ->157B 0.17944
154B ->162B -0.11313

Excited State 56: 3.105-A 4.0637 eV 305.10 nm f=0.0463 <S**2>=2.161
140A ->156A 0.10481
145A ->156A -0.22641
145A ->164A 0.11656
146A ->157A -0.13389
146A ->164A -0.26912
149A ->156A -0.18598
150A ->157A -0.10307
138B ->155B -0.13323
138B ->156B 0.11286
145B ->155B 0.21842
145B ->156B 0.12473
146B ->157B -0.17126
146B ->163B 0.30089
147B ->155B -0.29429
147B ->163B -0.14630
148B ->166B -0.11667
149B ->155B -0.23581
149B ->156B -0.28029
154B ->158B -0.12588

Excited State 62: 3.101-A 4.1784 eV 296.72 nm f=0.0201 <S**2>=2.154
137A ->156A -0.12850
141A ->159A 0.15638
145A ->156A 0.12140
146A ->156A 0.15617
146A ->164A -0.21403
148A ->166A 0.13488
149A ->156A 0.15252
150A ->162A 0.14037
138B ->156B 0.11269
143B ->159B 0.20834
145B ->155B -0.29139
146B ->155B 0.26080

146B ->163B 0.22476
147B ->155B 0.18118
148B ->166B -0.18989
149B ->156B 0.31737
150B ->162B 0.11721

Excited State 63: 2.432-A 4.2150 eV 294.15 nm f=0.6645 <S**2>=1.229
145A ->156A -0.15689
146A ->156A -0.11760
147A ->156A 0.11556
148A ->156A 0.10696
149A ->156A 0.63673
143B ->159B 0.13519
145B ->155B 0.42764
146B ->155B -0.12621
148B ->156B -0.11368
148B ->166B -0.13332
149B ->156B 0.37682

Excited State 68: 2.555-A 4.3185 eV 287.10 nm f=0.1388 <S**2>=1.383
146A ->156A 0.25079
147A ->156A 0.23964
148A ->156A 0.25790
155A ->169A -0.45238
145B ->155B 0.15804
146B ->155B 0.29389
146B ->156B 0.12845
148B ->155B 0.17434
148B ->156B 0.51780
150B ->156B -0.10108

Excited State 70: 2.774-A 4.3373 eV 285.85 nm f=0.1029 <S**2>=1.674
145A ->156A 0.11317
146A ->156A -0.24761
146A ->164A -0.11532
147A ->156A 0.49283
147A ->158A -0.11743
148A ->166A -0.10404
149A ->156A -0.10442
149A ->158A 0.11394
155A ->169A 0.27182
143B ->155B -0.10017
143B ->159B -0.12211
146B ->156B -0.13101
146B ->163B 0.12748
147B ->155B 0.41727
148B ->166B 0.13634
149B ->156B 0.17536

Excited State 71: 2.050-A 4.3725 eV 283.55 nm f=0.1411 <S**2>=0.800
147A ->156A -0.14974
150A ->157A 0.61189
151A ->157A -0.19661
145B ->155B -0.12482

147B ->155B -0.12981
150B ->157B 0.62981
151B ->157B -0.18597

Excited State 74: 2.917-A 4.4440 eV 278.99 nm f=0.0753 <S**2>=1.877
145A ->156A 0.45795
146A ->156A 0.12424
147A ->156A -0.12779
149A ->157A -0.19105
150A ->158A -0.22179
155A ->169A -0.23037
145B ->155B 0.29286
146B ->155B -0.21263
146B ->157B -0.11708
149B ->157B 0.43957
149B ->159B 0.12641
154B ->159B 0.16577

Excited State 78: 2.769-A 4.5363 eV 273.32 nm f=0.0684 <S**2>=1.667
146A ->156A -0.12551
147A ->157A 0.24207
150A ->158A 0.52759
155A ->169A -0.17113
147B ->157B -0.25043
150B ->158B 0.39633
151B ->158B 0.25403
151B ->162B -0.10991
154B ->159B -0.14714
154B ->161B -0.10187
154B ->162B 0.13603

Excited State 81: 2.791-A 4.5766 eV 270.91 nm f=0.0456 <S**2>=1.698
146A ->156A 0.26566
147A ->157A -0.13187
149A ->157A 0.15436
150A ->158A 0.27859
151A ->158A 0.10732
151A ->161A 0.13944
151A ->163A -0.14341
154A ->159A 0.19724
146B ->156B -0.30599
147B ->156B 0.18939
149B ->159B 0.17760
150B ->158B 0.41033
151B ->158B -0.13115
154B ->159B 0.35122

Excited State 127: 2.176-A 5.1205 eV 242.13 nm f=0.0242 <S**2>=0.934
140A ->156A -0.30346
143A ->156A 0.16470
150A ->160A 0.11869
151A ->159A -0.45194
151A ->160A -0.17858
151A ->162A -0.16334

140B ->155B -0.26934
142B ->155B 0.14010
147B ->157B 0.10551
150B ->160B 0.11964
151B ->159B 0.50467
151B ->160B -0.18235
151B ->161B -0.10217

Excited State 128: 2.416-A 5.1336 eV 241.52 nm f=0.3245 <S**2>=1.209
146A ->157A -0.13579
146A ->158A 0.13448
147A ->157A 0.40390
147A ->158A 0.10384
151A ->160A -0.11323
151A ->161A 0.41871
151A ->163A -0.18210
154A ->164A -0.16863
146B ->157B 0.23749
147B ->157B 0.13369
149B ->159B 0.12378
151B ->161B -0.39857
151B ->162B -0.25449

Excited State 129: 2.843-A 5.1410 eV 241.17 nm f=0.0598 <S**2>=1.770
137A ->156A 0.11060
140A ->156A 0.24452
142A ->156A -0.14784
143A ->156A 0.25124
149A ->157A 0.11941
149A ->159A 0.12253
151A ->159A -0.10448
151A ->161A -0.15387
151A ->162A 0.17275
151A ->163A 0.10050
154A ->164A -0.18042
138B ->155B -0.18389
142B ->155B 0.15565
145B ->156B -0.10175
145B ->157B 0.27989
147B ->157B -0.18095
148B ->158B -0.17303
149B ->158B 0.10753
149B ->159B 0.15326
151B ->159B 0.30808
151B ->161B 0.19484
154B ->163B -0.26770

Excited State 130: 2.779-A 5.1538 eV 240.57 nm f=0.0308 <S**2>=1.681
138A ->156A 0.10538
140A ->156A 0.38349
141A ->156A -0.11245
143A ->156A 0.21414
145A ->157A 0.11442
146A ->158A -0.14155

148A ->157A -0.16607
148A ->158A -0.10830
151A ->159A -0.18195
151A ->162A 0.14692
151A ->163A -0.15210
154A ->163A -0.12174
140B ->155B 0.24640
142B ->155B 0.16494
145B ->156B -0.12349
145B ->157B -0.14305
146B ->157B 0.18433
147B ->157B 0.12603
149B ->159B -0.19148
151B ->159B 0.13850
151B ->162B -0.15225
154B ->163B 0.37090

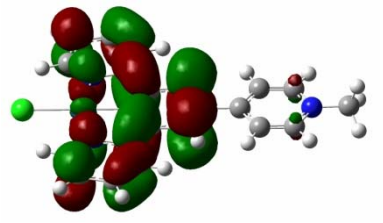
Excited State 153: 2.436-A 5.4201 eV 228.75 nm f=0.0210 <S**2>=1.233
139A ->156A 0.13744
145A ->158A 0.11211
150A ->159A 0.68483
150A ->160A 0.20971
150A ->161A -0.11936
150A ->162A -0.12336
151A ->159A 0.10179
151A ->163A 0.13265
151A ->164A -0.26076
139B ->155B 0.16380
146B ->158B -0.13801
147B ->158B 0.13806
150B ->159B -0.23886
150B ->162B 0.20802

Excited State 154: 2.359-A 5.4242 eV 228.58 nm f=0.0440 <S**2>=1.141
139A ->156A 0.30793
139A ->158A -0.10545
143A ->157A 0.17100
147A ->158A -0.10316
150A ->159A -0.35485
150A ->161A -0.32960
150A ->162A 0.12169
150A ->163A 0.13657
151A ->163A 0.16822
151A ->164A 0.15050
139B ->155B 0.42262
145B ->158B 0.11994
146B ->158B -0.25262
150B ->161B 0.30337
150B ->162B 0.11381

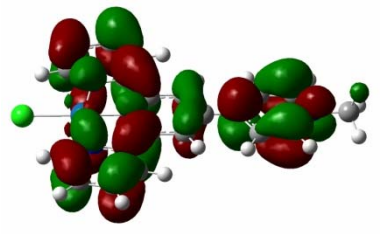
Excited State 174: 2.306-A 5.5722 eV 222.50 nm f=0.0202 <S**2>=1.079
138A ->156A 0.28319
148A ->159A -0.13612
150A ->159A 0.14164

150A ->162A	0.40576
137B ->155B	0.17567
138B ->155B	0.18656
139B ->157B	0.12454
140B ->156B	-0.30054
142B ->156B	-0.27921
142B ->157B	0.11502
145B ->158B	-0.10217
146B ->158B	0.10166
148B ->159B	0.14856
150B ->159B	-0.17138
150B ->161B	0.30270
150B ->162B	-0.23992
154B ->164B	0.14073

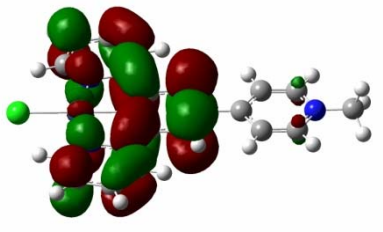
Table S14. Electronic transitions computed by TD-DFT for the one-electron-reduced form **PtL**$^{2+}$ (i.e., **PtL**$^{+}$•; doublet), for which part of the Gaussian output is shown. Relevant MO's are shown below:



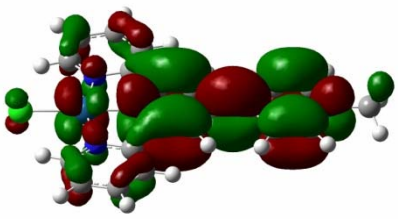
MO105A



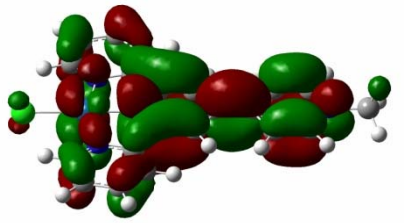
MO104A



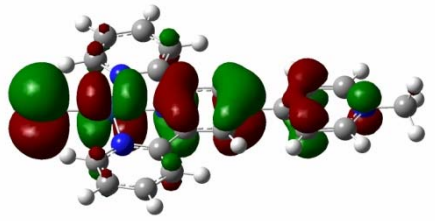
MO104B



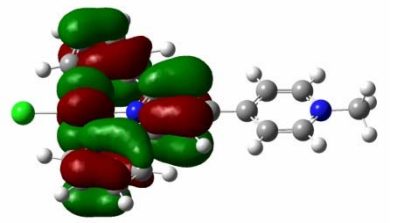
MO103A (SOMO)



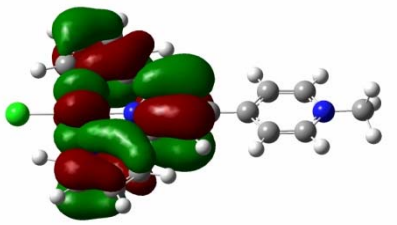
MO103B



MO102B



MO100A



MO100B

Excitation energies and oscillator strengths (λ > 200 nm, f > 0.02 only):

Excitation energies and oscillator strengths:

Excited State 2:	2.013-A	1.3963 eV	887.96 nm	f=0.3245	<S**2>=0.763
103A ->104A	0.98588				
102B ->103B	0.11235				

Excited State 7:	2.477-A	2.7066 eV	458.09 nm	f=0.1412	<S**2>=1.284
98A ->105A	0.15968				
100A ->105A	-0.17912				

103A ->111A	-0.17379				
97B ->103B	0.14058				
98B ->104B	0.13358				
100B ->104B	-0.32651				
102B ->103B	0.82149				

Excited State 10:	2.967-A	3.0475 eV	406.84 nm	f=0.1524	<S**2>=1.951
96A ->104A	-0.10693				
98A ->105A	-0.18589				
98A ->107A	0.10908				
100A ->105A	0.44637				
103A ->108A	0.10383				
103A ->111A	0.35991				
96B ->103B	0.11852				
96B ->108B	0.10300				
98B ->104B	-0.17193				
100B ->104B	0.44020				
102B ->103B	0.47586				
102B ->105B	-0.13335				

Excited State 13:	2.399-A	3.1796 eV	389.94 nm	f=0.0607	<S**2>=1.189
98A ->105A	0.13661				
100A ->105A	-0.29720				
103A ->108A	-0.11064				
103A ->111A	0.85072				
100B ->104B	-0.22245				

Excited State 18:	2.497-A	3.5084 eV	353.40 nm	f=0.0213	<S**2>=1.309
98A ->108A	0.10319				
100A ->104A	0.32563				
102A ->105A	0.22440				
90B ->103B	0.10089				
98B ->103B	0.69026				
98B ->105B	-0.12243				
100B ->103B	-0.24940				
100B ->105B	-0.18881				
102B ->104B	-0.36962				

Excited State 20:	2.867-A	3.5599 eV	348.28 nm	f=0.0565	<S**2>=1.805
97A ->104A	0.10742				
101A ->106A	-0.32736				
102A ->104A	0.62829				
103A ->111A	-0.10777				
103A ->112A	-0.18462				
96B ->103B	-0.10224				
97B ->103B	-0.37423				
97B ->105B	-0.11591				
101B ->106B	-0.31580				
102B ->105B	-0.34655				

Excited State 21:	2.213-A	3.5973 eV	344.66 nm	f=0.0844	<S**2>=0.975
101A ->106A	0.60319				
102A ->104A	0.22629				
103A ->112A	-0.21554				

97B ->103B	-0.20733				
101B ->106B	0.62203				
102B ->105B	-0.20517				

Excited State 25:	2.387-A	3.7088 eV	334.29 nm	f=0.0641	<S**2>=1.174
102A ->105A	0.78523				
102A ->106A	-0.20241				
103A ->115A	-0.14505				
98B ->103B	-0.36165				
98B ->105B	0.10710				
99B ->106B	0.17264				
102B ->104B	-0.24971				
102B ->106B	-0.12136				

Excited State 26:	2.257-A	3.7676 eV	329.08 nm	f=0.2484	<S**2>=1.023
98A ->105A	0.12435				
102A ->104A	0.66452				
95B ->110B	-0.11951				
97B ->103B	0.42324				
97B ->105B	0.11128				
100B ->104B	0.11902				
102B ->105B	0.45610				

Excited State 31:	3.074-A	3.9069 eV	317.35 nm	f=0.0444	<S**2>=2.113
90A ->105A	-0.10633				
91A ->104A	-0.13755				
98A ->104A	0.14349				
98A ->105A	0.41257				
98A ->107A	0.13641				
99A ->106A	0.16856				
100A ->105A	0.20111				
100A ->107A	0.15045				
102A ->104A	-0.14265				
103A ->115A	-0.37949				
90B ->104B	0.10394				
91B ->103B	-0.12757				
91B ->105B	0.13115				
98B ->104B	0.40876				
98B ->107B	-0.14939				
99B ->106B	0.20043				
100B ->107B	-0.12400				
102B ->105B	-0.13242				

Excited State 32:	2.802-A	3.9072 eV	317.32 nm	f=0.0425	<S**2>=1.713
98A ->104A	-0.19535				
98A ->105A	0.29488				
98A ->108A	-0.14632				
99A ->106A	-0.23582				
100A ->105A	0.14272				
100A ->107A	0.10784				
102A ->104A	-0.10075				
102A ->105A	0.10228				
103A ->115A	0.53556				

96B ->107B	-0.10949				
97B ->104B	0.10414				
98B ->104B	0.28968				
98B ->105B	0.10852				
98B ->107B	-0.10511				
98B ->108B	0.11206				
99B ->106B	-0.28054				
100B ->105B	-0.12698				
102B ->104B	-0.11203				
102B ->107B	0.10578				

Excited State 37:	2.879-A	4.0622 eV	305.22 nm	f=0.0604	<S**2>=1.821
91A ->107A	-0.10292				
96A ->107A	0.15986				
98A ->104A	0.40636				
98A ->108A	0.24154				
102A ->107A	0.18121				
103A ->115A	0.46153				
91B ->107B	0.11933				
95B ->103B	-0.30795				
95B ->105B	-0.13835				
96B ->107B	0.14557				
98B ->103B	-0.22829				
98B ->105B	-0.24034				
98B ->108B	-0.22848				
100B ->105B	0.10485				
102B ->107B	-0.19728				

Excited State 41:	2.573-A	4.1245 eV	300.60 nm	f=0.1075	<S**2>=1.405
95A ->104A	-0.22125				
96A ->105A	0.10747				
97A ->105A	-0.15141				
98A ->104A	0.34426				
100A ->104A	0.31258				
95B ->103B	0.62679				
95B ->105B	0.29271				
96B ->104B	-0.15340				
98B ->103B	-0.16839				
98B ->105B	-0.10282				
100B ->105B	0.16688				
100B ->108B	-0.12021				

Excited State 42:	2.572-A	4.1438 eV	299.21 nm	f=0.0542	<S**2>=1.404
96A ->104A	0.15247				
98A ->107A	0.14824				
100A ->105A	-0.40904				
96B ->103B	-0.33076				
96B ->105B	0.16607				
97B ->103B	-0.29382				
98B ->104B	0.24178				
98B ->107B	-0.18748				
100B ->104B	0.43238				
100B ->107B	0.14915				
102B ->103B	0.12502				

102B ->105B	0.36315				
102B ->108B	-0.10404				

Excited State 45:	2.743-A	4.1762 eV	296.88 nm	f=0.0683	<S**2>=1.631
90A ->105A	0.12515				
96A ->104A	0.24239				
96A ->108A	0.14213				
97A ->104A	-0.15381				
98A ->105A	0.12273				
98A ->107A	0.23003				
100A ->105A	0.43375				
100A ->107A	-0.19228				
102A ->108A	0.16077				
90B ->104B	-0.12898				
96B ->103B	-0.41931				
96B ->105B	0.20934				
98B ->104B	-0.23637				
98B ->107B	-0.15219				
100B ->104B	-0.34982				
100B ->107B	0.17751				

Excited State 48:	3.190-A	4.4110 eV	281.08 nm	f=0.0332	<S**2>=2.294
95A ->105A	-0.14953				
95A ->110A	-0.50538				
97A ->104A	0.16425				
103A ->111A	0.13314				
88B ->103B	-0.15594				
94B ->103B	0.12361				
95B ->104B	-0.17284				
95B ->110B	0.55658				
96B ->103B	-0.14635				
97B ->103B	0.36648				
97B ->105B	-0.15688				

Excited State 49:	2.726-A	4.4753 eV	277.04 nm	f=0.1648	<S**2>=1.608
95A ->110A	-0.20286				
96A ->104A	0.14065				
97A ->104A	-0.24837				
98A ->105A	0.44920				
100A ->107A	-0.22761				
88B ->103B	0.13590				
94B ->103B	-0.15636				
95B ->110B	0.23991				
96B ->103B	0.27710				
97B ->105B	0.23323				
98B ->104B	-0.30768				
100B ->104B	0.21192				
102B ->108B	-0.25288				
102B ->112B	-0.12334				

Excited State 51:	2.713-A	4.5444 eV	272.83 nm	f=0.0836	<S**2>=1.590
91A ->104A	0.13034				
97A ->104A	-0.24453				
98A ->105A	-0.37362				

98A ->107A	-0.11185				
100A ->105A	0.24227				
100A ->107A	-0.12973				
102A ->108A	0.17549				
103A ->118A	0.11261				
88B ->103B	0.10407				
91B ->108B	-0.12365				
96B ->103B	0.13495				
97B ->105B	0.12724				
98B ->104B	0.53862				
98B ->107B	0.17656				
100B ->107B	0.34506				

Excited State 54:	2.671-A	4.6090 eV	269.01 nm	f=0.0424	<S**2>=1.533
90A ->104A	-0.10713				
91A ->107A	-0.11133				
96A ->105A	-0.16548				
97A ->105A	-0.18796				
98A ->104A	-0.37887				
100A ->108A	0.26537				
102A ->107A	0.18759				
91B ->104B	-0.10228				
97B ->104B	-0.32603				
98B ->105B	-0.40711				
102B ->107B	0.50922				

Excited State 55:	3.000-A	4.6491 eV	266.68 nm	f=0.0457	<S**2>=1.999
89A ->104A	-0.10077				
90A ->104A	0.10451				
93A ->105A	-0.11394				
96A ->105A	0.29527				
97A ->105A	0.11971				
98A ->104A	-0.30558				
98A ->108A	0.11357				
100A ->108A	0.27264				
103A ->115A	-0.15834				
90B ->103B	-0.16061				
90B ->105B	0.10135				
94B ->104B	-0.11270				
96B ->104B	-0.30980				
97B ->104B	0.42907				
98B ->105B	-0.40676				
98B ->108B	-0.11873				
100B ->105B	-0.14516				
102B ->107B	-0.15582				

Excited State 71:	2.160-A	5.0131 eV	247.32 nm	f=0.0328	<S**2>=0.916
97A ->104A	0.35448				
98A ->105A	0.14679				
100A ->107A	0.52581				
89B ->104B	-0.11911				
90B ->104B	-0.11800				
91B ->103B	-0.19760				
96B ->103B	0.17715				

96B ->105B	0.15914				
97B ->105B	0.16280				
98B ->104B	-0.10896				
100B ->107B	0.56410				

Excited State 73:	2.277-A	5.0324 eV	246.37 nm	f=0.2486	<S**2>=1.046
96A ->105A	-0.12644				
98A ->104A	0.10099				
98A ->108A	0.13740				
100A ->108A	0.51838				
102A ->110A	0.11476				
90B ->103B	-0.10614				
96B ->104B	-0.30805				
97B ->104B	-0.13637				
98B ->105B	0.31209				
98B ->108B	0.10985				
100B ->105B	-0.10981				
100B ->108B	0.56825				
102B ->110B	-0.13839				

Excited State 85:	2.048-A	5.2490 eV	236.20 nm	f=0.3522	<S**2>=0.798
96A ->105A	0.56607				
97A ->105A	-0.17402				
98A ->108A	-0.10230				
100A ->108A	0.28864				
96B ->104B	0.65017				
100B ->108B	0.25068				

Excited State 95:	2.420-A	5.4569 eV	227.21 nm	f=0.0290	<S**2>=1.215
91A ->104A	0.13225				
96A ->104A	0.13642				
97A ->104A	-0.20488				
98A ->107A	-0.41803				
100A ->107A	0.11967				
101A ->109A	0.17127				
102A ->111A	-0.11954				
88B ->103B	-0.10730				
95B ->104B	0.61630				
95B ->110B	0.11843				
96B ->105B	-0.14115				
98B ->107B	-0.37963				
100B ->107B	0.10989				
101B ->109B	0.13828				

Excited State 98:	2.017-A	5.4963 eV	225.58 nm	f=0.0271	<S**2>=0.767
98A ->108A	-0.18734				
99A ->109A	0.68664				
98B ->108B	-0.20127				
99B ->109B	0.63270				

Excited State 105:	2.056-A	5.5980 eV	221.48 nm	f=0.0420	<S**2>=0.806
95A ->104A	0.44236				
102A ->110A	-0.44933				
95B ->103B	-0.10883				

95B ->105B	0.59835				
97B ->110B	0.11201				
102B ->110B	-0.42310				

Excited State 125:	2.100-A	5.8596 eV	211.59 nm	f=0.0318	<S**2>=0.853
88A ->106A	0.10715				
89A ->104A	0.10178				
91A ->105A	0.21748				
94A ->106A	0.50279				
96A ->107A	-0.10159				
99A ->106A	-0.15328				
87B ->106B	0.12436				
89B ->103B	-0.20852				
90B ->103B	0.12677				
93B ->106B	0.64118				
96B ->107B	0.17046				
97B ->107B	-0.10472				
98B ->108B	0.10775				
99B ->106B	-0.19959				

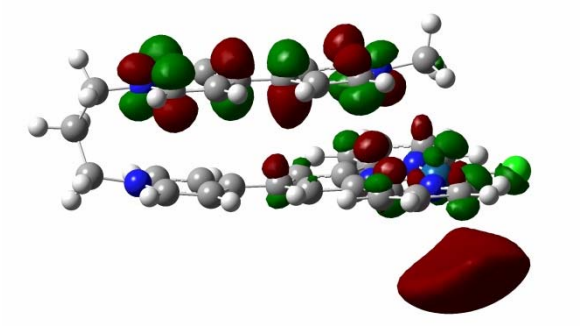
Excited State 140:	2.893-A	5.9794 eV	207.35 nm	f=0.0310	<S**2>=1.843
89A ->104A	0.16229				
89A ->108A	0.17405				
91A ->107A	0.22552				
96A ->107A	-0.18449				
97A ->107A	0.16736				
98A ->108A	0.32405				
89B ->103B	-0.35589				
89B ->108B	-0.17135				
90B ->103B	0.11792				
91B ->107B	-0.21375				
96B ->107B	-0.19951				
97B ->107B	0.59088				
98B ->108B	-0.11649				
100B ->111B	0.16180				

Excited State 151:	2.642-A	6.0898 eV	203.59 nm	f=0.0211	<S**2>=1.495
90A ->105A	-0.19238				
93A ->104A	-0.12916				
95A ->110A	-0.12709				
97A ->108A	0.14813				
98A ->107A	0.13970				
102A ->111A	-0.25197				
89B ->107B	-0.10924				
90B ->104B	0.13924				
91B ->108B	-0.12070				
94B ->105B	-0.17386				
96B ->105B	0.12559				
96B ->108B	-0.21871				
97B ->108B	0.54189				
102B ->112B	-0.48800				

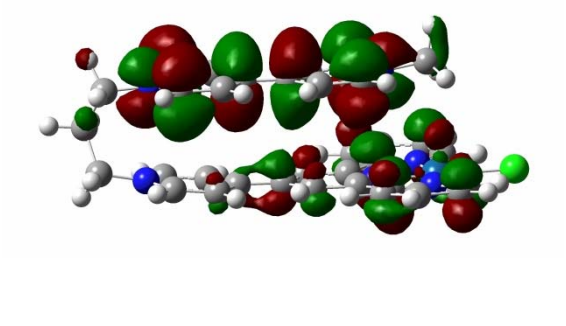
Excited State 152:	2.352-A	6.0966 eV	203.37 nm	f=0.0224	<S**2>=1.133
98A ->112A	0.24454				

100A ->112A	0.88271				
98B ->111B	0.11177				
100B ->111B	0.31469				
Excited State 157:	2.577-A	6.1627 eV	201.18 nm	f=0.0260	<S**2>=1.410
96A ->109A	-0.11877				
97A ->109A	-0.17166				
101A ->112A	0.45514				
102A ->113A	0.67451				
101B ->111B	0.49814				

Table S15. Electronic transitions computed by TD-DFT for the closed-shell singlet state of **[PtL^{+}·-C1-MV^{+}·]$^{2+}$**, for which part of the Gaussian output is shown. Relevant MO's are shown below:



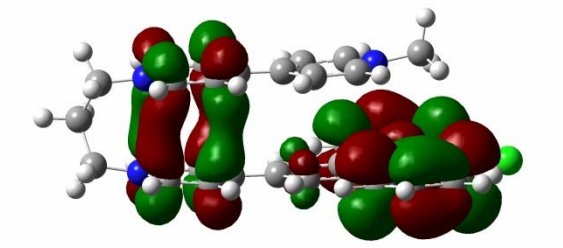
MO164 (LUMO+8)



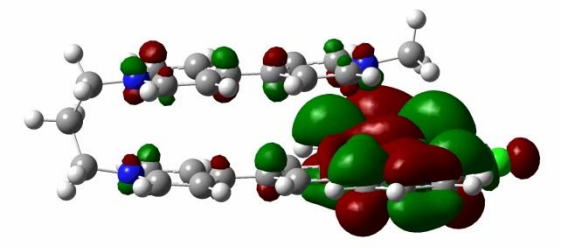
MO163 (LUMO+7)

MO162(LUMO+6)

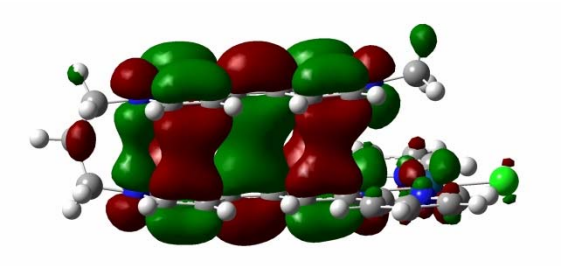
MO161 (LUMO+5)



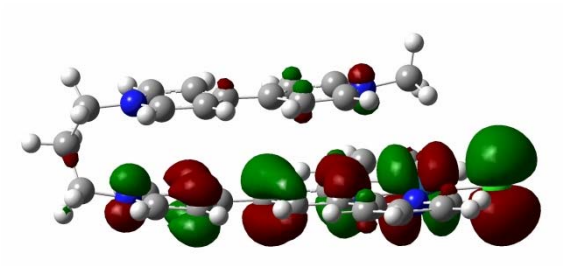
MO160 (LUMO+4)



MO156 (LUMO)



MO155 (HOMO)



MO154 (HOMO-1)

Excitation energies and oscillator strengths (λ > 200 nm, f > 0.02 only):

Excited State 2:	Singlet-A	1.4195 eV	873.44 nm	f=0.2739	<S**2>=0.000
155 ->156	0.72189				
155 <-156	-0.18223				
Excited State 3:	Singlet-A	1.8338 eV	676.09 nm	f=0.3020	<S**2>=0.000
155 ->156	0.10378				
155 ->158	0.69569				
Excited State 7:	Singlet-A	2.5962 eV	477.56 nm	f=0.0418	<S**2>=0.000
155 ->160	-0.28938				
155 ->161	0.62131				
155 ->162	-0.11720				

Excited State 9:	Singlet-A	3.0348 eV	408.54 nm	f=0.0925	<S**2>=0.000
150 ->156	-0.13373				
154 ->156	-0.25892				
155 ->163	0.62175				
155 ->164	-0.12756				
Excited State 11:	Singlet-A	3.2550 eV	380.91 nm	f=0.2107	<S**2>=0.000
150 ->156	-0.11574				
153 ->160	0.10356				
154 ->156	0.57663				
154 ->158	-0.16100				
155 ->163	0.16760				
155 ->164	-0.25134				
Excited State 21:	Singlet-A	3.8146 eV	325.02 nm	f=0.0784	<S**2>=0.000
149 ->156	0.19190				
150 ->156	-0.10877				
151 ->160	-0.35829				
151 ->161	-0.18859				
152 ->160	0.18172				
154 ->158	0.40588				
155 ->167	-0.15813				
Excited State 22:	Singlet-A	3.8157 eV	324.93 nm	f=0.1040	<S**2>=0.000
149 ->156	-0.18337				
151 ->160	0.32912				
151 ->161	0.17191				
152 ->160	-0.16477				
154 ->156	0.10891				
154 ->158	0.44647				
155 ->167	-0.17353				
Excited State 23:	Singlet-A	3.8997 eV	317.93 nm	f=0.1498	<S**2>=0.000
149 ->156	0.55303				
149 ->160	-0.16158				
152 ->160	-0.22894				
152 ->161	-0.17536				
154 ->157	0.13157				
Excited State 24:	Singlet-A	3.9027 eV	317.69 nm	f=0.0352	<S**2>=0.000
149 ->156	0.24215				
149 ->160	0.31787				
149 ->161	0.15702				
151 ->160	0.30346				
151 ->161	0.15386				
152 ->160	0.36445				
152 ->161	0.16059				
Excited State 25:	Singlet-A	3.9620 eV	312.94 nm	f=0.8606	<S**2>=0.000
150 ->156	0.46687				
151 ->157	-0.16038				
152 ->157	0.42242				
155 ->168	0.13684				

Excited State 26:	Singlet-A	3.9747 eV	311.93 nm	f=0.1289	<S**2>=0.000
149 ->157	0.11250				
150 ->156	-0.35748				
151 ->157	-0.35723				
152 ->157	0.36220				
153 ->158	-0.11568				
155 ->167	0.10743				
155 ->168	-0.17755				
Excited State 27:	Singlet-A	3.9901 eV	310.73 nm	f=0.0996	<S**2>=0.000
150 ->156	-0.18585				
155 ->168	0.66071				
Excited State 29:	Singlet-A	4.0226 eV	308.22 nm	f=0.0284	<S**2>=0.000
149 ->157	-0.10237				
151 ->157	-0.44722				
152 ->157	-0.20524				
153 ->156	0.12267				
153 ->158	0.45907				
Excited State 30:	Singlet-A	4.0755 eV	304.22 nm	f=0.0870	<S**2>=0.000
148 ->156	0.61272				
152 ->158	-0.28649				
Excited State 33:	Singlet-A	4.2839 eV	289.42 nm	f=0.0238	<S**2>=0.000
150 ->157	0.33071				
155 ->169	0.34796				
155 ->170	0.48299				
Excited State 35:	Singlet-A	4.3212 eV	286.92 nm	f=0.0778	<S**2>=0.000
149 ->157	0.62925				
150 ->158	-0.11276				
152 ->157	-0.17175				
Excited State 38:	Singlet-A	4.4907 eV	276.09 nm	f=0.0474	<S**2>=0.000
148 ->157	0.10784				
149 ->158	-0.10212				
150 ->158	0.62361				
155 ->171	-0.19360				
Excited State 39:	Singlet-A	4.5009 eV	275.46 nm	f=0.0853	<S**2>=0.000
147 ->156	-0.14410				
149 ->156	0.10026				
149 ->158	0.64922				
Excited State 50:	Singlet-A	4.9144 eV	252.29 nm	f=0.1167	<S**2>=0.000
146 ->157	-0.24924				
147 ->158	0.14881				
149 ->158	-0.10349				
149 ->161	0.12893				
152 ->160	-0.29263				
152 ->161	0.48413				
Excited State 55:	Singlet-A	5.0576 eV	245.14 nm	f=0.0610	<S**2>=0.000

147 ->157 0.58804
152 ->159 0.29388
152 ->162 -0.12482

Excited State 56: Singlet-A 5.1329 eV 241.55 nm f=0.1608 <S**2>=0.000
143 ->156 0.27318
146 ->157 0.49506
150 ->159 0.21144
150 ->162 0.10099
152 ->161 0.17516
154 ->162 -0.15082
154 ->165 0.14159

Excited State 57: Singlet-A 5.1526 eV 240.62 nm f=0.1415 <S**2>=0.000
143 ->156 -0.30195
146 ->157 0.31840
150 ->159 -0.16096
150 ->162 -0.14405
152 ->161 0.14955
154 ->159 0.14009
154 ->162 0.10316
154 ->163 0.33085
154 ->164 0.16749
154 ->165 -0.12904

Excited State 58: Singlet-A 5.1563 eV 240.45 nm f=0.0648 <S**2>=0.000
143 ->156 0.21874
146 ->157 -0.19414
150 ->159 0.11647
154 ->163 0.49057
154 ->164 0.25625
154 ->165 0.10558

Excited State 59: Singlet-A 5.2020 eV 238.34 nm f=0.0299 <S**2>=0.000
145 ->157 0.50829
147 ->158 0.37708
152 ->161 -0.12571

Excited State 61: Singlet-A 5.2119 eV 237.89 nm f=0.0216 <S**2>=0.000
142 ->156 -0.17697
145 ->157 0.13299
145 ->158 0.20617
152 ->159 0.31849
152 ->162 0.36632
154 ->164 0.30875

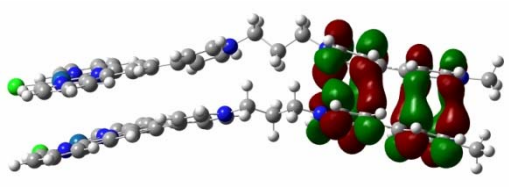
Excited State 64: Singlet-A 5.2849 eV 234.60 nm f=0.0262 <S**2>=0.000
145 ->157 -0.40199
147 ->158 0.51192
149 ->161 -0.10708
154 ->165 -0.16604

Excited State 81: Singlet-A 5.5495 eV 223.42 nm f=0.0456 <S**2>=0.000
150 ->161 -0.18725

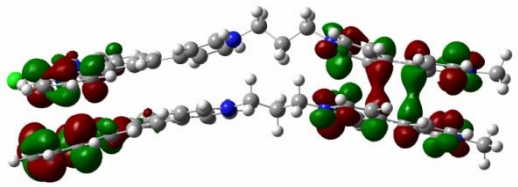
151 ->163	0.37428				
151 ->164	0.39576				
152 ->163	0.29336				
152 ->164	-0.11792				
Excited State 87:	Singlet-A	5.6129 eV	220.89 nm	f=0.0634	<S**2>=0.000
140 ->156	0.31444				
142 ->156	-0.11102				
149 ->162	-0.12830				
152 ->164	0.14063				
152 ->165	0.52808				
Excited State 99:	Singlet-A	5.8204 eV	213.02 nm	f=0.0201	<S**2>=0.000
140 ->157	-0.11841				
144 ->158	0.27699				
144 ->160	0.32122				
144 ->161	0.12445				
148 ->160	-0.20945				
148 ->161	0.21594				
148 ->163	0.19578				
148 ->164	-0.10248				
150 ->163	-0.11765				
151 ->160	-0.10365				
Excited State 120:	Singlet-A	6.0646 eV	204.44 nm	f=0.0314	<S**2>=0.000
135 ->156	0.12430				
138 ->156	0.43609				
140 ->157	-0.31026				
147 ->160	0.11111				
148 ->161	-0.10824				
148 ->163	0.21428				
148 ->164	-0.10445				
149 ->163	-0.15092				
149 ->164	0.17724				
149 ->165	0.10742				
Excited State 129:	Singlet-A	6.1854 eV	200.45 nm	f=0.0260	<S**2>=0.000
137 ->156	-0.12069				
139 ->157	-0.27103				
140 ->158	-0.18911				
147 ->159	0.42431				
147 ->161	0.10558				
152 ->166	0.10657				
154 ->166	0.14565				
154 ->167	-0.10943				
155 ->180	-0.17508				
Excited State 130:	Singlet-A	6.1881 eV	200.36 nm	f=0.0290	<S**2>=0.000
137 ->156	0.15303				
139 ->157	0.37970				
140 ->157	-0.10390				
140 ->158	-0.13050				
147 ->159	0.33551				
152 ->167	-0.14909				

154 ->167	0.12224
155 ->180	-0.16483

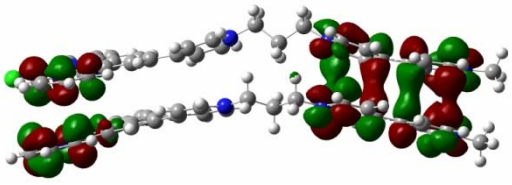
Table S16. Electronic transitions computed by TD-DFT for the closed-shell singlet state of **HH(C_2)-[PtL^{+}·-C1-MV^{+}·]$_2^{4+}$**, for which part of the Gaussian output is shown. Relevant MO's are shown below:



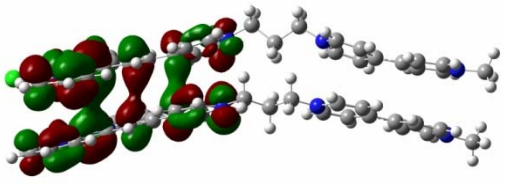
MO326(LUMO+15)



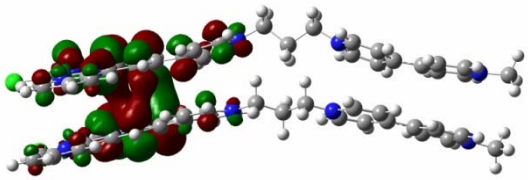
MO320 (LUMO+9)



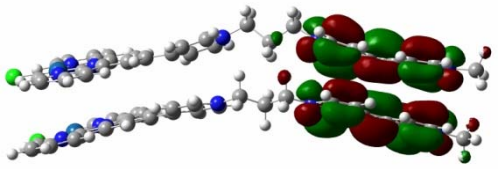
MO319(LUMO+8)



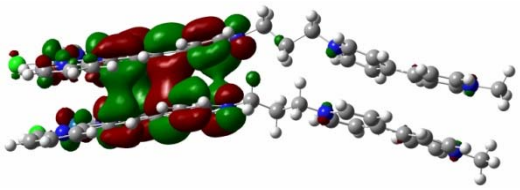
MO314 (LUMO+3)



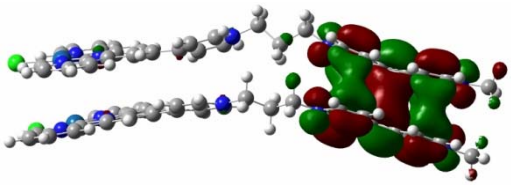
MO313 (LUMO+2)



MO312 (LUMO+1)



MO310 (HOMO)



MO309 (HOMO-1)

Excitation energies and oscillator strengths (λ > 200 nm, f > 0.02 only):

Excited State	1:	Singlet-A	0.9079 eV	1365.67 nm	f=0.0438 <S**2>=0.000
310 -> 311		0.58277			
310 -> 313		-0.36875			
310 -> 314		-0.15936			
310 <- 311		-0.13605			
Excited State	3:	Singlet-A	1.2201 eV	1016.19 nm	f=0.0830 <S**2>=0.000
309 -> 311		0.30225			
310 -> 311		0.35440			
310 -> 312		0.18332			
310 -> 313		0.47335			
310 -> 314		0.16228			
310 <- 311		-0.11801			
Excited State	4:	Singlet-A	1.2779 eV	970.23 nm	f=0.0221 <S**2>=0.000

310 -> 312 0.67618
310 -> 313 -0.15600

Excited State 5: Singlet-A 1.4925 eV 830.70 nm f=0.2249 <S**2>=0.000
309 -> 312 0.72475
309 <- 312 -0.20980

Excited State 7: Singlet-A 1.6137 eV 768.34 nm f=0.5689 <S**2>=0.000
310 -> 313 -0.24772
310 -> 314 0.64873

Excited State 10: Singlet-A 1.8786 eV 659.99 nm f=0.0474 <S**2>=0.000
310 -> 315 0.33840
310 -> 316 0.59921

Excited State 17: Singlet-A 2.4267 eV 510.91 nm f=0.0239 <S**2>=0.000
309 -> 318 0.10464
310 -> 318 0.61584
310 -> 320 -0.15446
310 -> 325 0.25361

Excited State 21: Singlet-A 2.6558 eV 466.85 nm f=0.2041 <S**2>=0.000
301 -> 312 -0.11844
308 -> 311 0.21560
309 -> 319 -0.29578
309 -> 320 -0.23004
309 -> 326 0.48927
310 -> 319 0.11423
310 -> 326 -0.12328

Excited State 24: Singlet-A 2.7367 eV 453.05 nm f=0.3470 <S**2>=0.000
301 -> 312 0.15997
309 -> 319 0.33002
309 -> 320 0.24553
309 -> 326 0.42934
309 -> 331 -0.11225
310 -> 319 -0.21735
310 -> 320 -0.17803
310 -> 326 -0.10233

Excited State 26: Singlet-A 2.7923 eV 444.02 nm f=0.0230 <S**2>=0.000
309 -> 319 0.24217
309 -> 320 0.18578
310 -> 319 0.48138
310 -> 320 0.38402

Excited State 27: Singlet-A 2.9356 eV 422.35 nm f=0.0933 <S**2>=0.000
307 -> 311 -0.44046
308 -> 311 -0.11852
309 -> 318 0.45620
309 -> 321 -0.10439
309 -> 325 -0.13242
310 -> 318 -0.10480

Excited State 28:	Singlet-A	2.9377 eV	422.05 nm	f=0.0994	<S**2>=0.000
307 -> 311	0.46224				
308 -> 311	0.12452				
309 -> 318	0.43091				
309 -> 321	-0.12637				
309 -> 325	-0.14329				
Excited State 31:	Singlet-A	3.0019 eV	413.01 nm	f=0.0362	<S**2>=0.000
309 -> 320	0.11253				
309 -> 330	-0.30167				
309 -> 331	0.58980				
310 -> 331	-0.14975				
Excited State 41:	Singlet-A	3.3097 eV	374.61 nm	f=0.0274	<S**2>=0.000
302 -> 311	0.11435				
307 -> 313	0.55717				
308 -> 313	0.11718				
308 -> 314	0.14365				
310 -> 332	0.29792				
Excited State 45:	Singlet-A	3.3469 eV	370.44 nm	f=0.1832	<S**2>=0.000
302 -> 311	0.18104				
307 -> 313	-0.30398				
308 -> 320	-0.11308				
308 -> 321	-0.20123				
310 -> 332	0.43706				
Excited State 51:	Singlet-A	3.4265 eV	361.84 nm	f=0.1416	<S**2>=0.000
299 -> 322	-0.14196				
300 -> 322	0.13848				
307 -> 311	0.10586				
307 -> 314	-0.11032				
307 -> 321	0.12091				
307 -> 322	-0.13298				
308 -> 320	-0.12763				
308 -> 321	-0.28169				
308 -> 322	0.30131				
308 -> 325	0.10563				
310 -> 332	-0.27926				
Excited State 56:	Singlet-A	3.5774 eV	346.57 nm	f=0.0421	<S**2>=0.000
299 -> 311	-0.23350				
300 -> 311	0.38004				
306 -> 313	-0.35893				
307 -> 315	0.15403				
308 -> 312	-0.10206				
308 -> 314	-0.22281				
308 -> 315	-0.10060				
Excited State 62:	Singlet-A	3.6339 eV	341.18 nm	f=0.0272	<S**2>=0.000
307 -> 312	0.65455				
310 -> 331	0.16304				
Excited State 64:	Singlet-A	3.6534 eV	339.37 nm	f=0.0364	<S**2>=0.000

299 -> 311 0.12595
300 -> 311 0.10817
305 -> 313 0.46372
306 -> 313 -0.20357
307 -> 315 -0.14102
310 -> 330 -0.12917
310 -> 331 -0.20315
310 -> 334 0.18823

Excited State 66: Singlet-A 3.7058 eV 334.57 nm f=0.1147 <S**2>=0.000
299 -> 311 0.41012
305 -> 313 -0.15402
306 -> 313 -0.21998
306 -> 314 0.31499
307 -> 315 -0.10749
310 -> 334 -0.23849

Excited State 67: Singlet-A 3.7293 eV 332.46 nm f=0.0823 <S**2>=0.000
299 -> 311 -0.39176
300 -> 311 -0.11048
302 -> 311 -0.10538
304 -> 322 -0.10565
305 -> 313 0.11502
305 -> 314 -0.22490
306 -> 314 0.40725

Excited State 68: Singlet-A 3.7682 eV 329.03 nm f=0.1307 <S**2>=0.000
305 -> 313 -0.25598
305 -> 314 0.29644
306 -> 314 0.19989
307 -> 315 -0.19509
308 -> 315 -0.12485
308 -> 316 -0.13133
310 -> 334 0.37764

Excited State 69: Singlet-A 3.7928 eV 326.89 nm f=0.0403 <S**2>=0.000
299 -> 311 0.11118
303 -> 321 0.12611
305 -> 313 -0.19183
305 -> 314 -0.39489
307 -> 315 0.15030
310 -> 334 0.40093

Excited State 75: Singlet-A 3.8653 eV 320.76 nm f=0.0872 <S**2>=0.000
303 -> 321 -0.16713
304 -> 313 0.14206
304 -> 322 -0.23704
305 -> 314 0.14798
306 -> 314 0.19703
306 -> 315 -0.10179
307 -> 315 0.37595
307 -> 316 -0.15043
308 -> 315 0.14367

Excited State 77:	Singlet-A	3.8951 eV	318.31 nm	f=1.5320	<S**2>=0.000
296 -> 311	-0.16490				
297 -> 311	0.14345				
301 -> 311	0.20280				
301 -> 312	0.44756				
302 -> 313	0.28227				
307 -> 316	-0.17832				
309 -> 319	-0.11487				
Excited State 79:	Singlet-A	3.9037 eV	317.60 nm	f=0.2031	<S**2>=0.000
296 -> 311	0.13761				
300 -> 321	-0.14226				
300 -> 322	-0.10792				
301 -> 312	-0.20521				
302 -> 313	0.29081				
302 -> 320	0.11772				
302 -> 321	0.21961				
302 -> 322	-0.15848				
303 -> 313	-0.20133				
305 -> 314	0.14446				
305 -> 322	0.12807				
307 -> 316	0.10511				
Excited State 81:	Singlet-A	3.9147 eV	316.72 nm	f=0.0279	<S**2>=0.000
296 -> 311	0.10628				
300 -> 321	0.12346				
302 -> 313	0.46435				
302 -> 321	-0.18451				
303 -> 313	0.27890				
305 -> 321	-0.10463				
Excited State 82:	Singlet-A	3.9409 eV	314.61 nm	f=0.0491	<S**2>=0.000
296 -> 311	0.19664				
297 -> 311	0.58890				
302 -> 313	-0.14456				
307 -> 316	-0.13237				
Excited State 84:	Singlet-A	3.9770 eV	311.76 nm	f=0.1210	<S**2>=0.000
296 -> 311	0.25274				
301 -> 312	0.31329				
307 -> 315	0.18505				
307 -> 316	0.42760				
308 -> 316	0.15100				
Excited State 85:	Singlet-A	4.0041 eV	309.64 nm	f=0.0200	<S**2>=0.000
302 -> 314	-0.25983				
304 -> 311	-0.10676				
304 -> 313	-0.24949				
304 -> 314	0.46095				
304 -> 316	0.11366				
310 -> 336	-0.17393				
Excited State 88:	Singlet-A	4.0492 eV	306.20 nm	f=0.0299	<S**2>=0.000
296 -> 311	-0.18746				

298 -> 312 -0.15954
301 -> 311 0.48783
301 -> 312 -0.20856
304 -> 314 0.10344
306 -> 315 0.21260
307 -> 315 0.10149
307 -> 316 0.19272

Excited State 89: Singlet-A 4.0592 eV 305.44 nm f=0.0328 <S**2>=0.000
298 -> 311 0.11608
298 -> 312 0.66208
301 -> 311 0.13126

Excited State 92: Singlet-A 4.0869 eV 303.37 nm f=0.1982 <S**2>=0.000
296 -> 311 0.42587
297 -> 311 -0.19375
301 -> 311 0.39262
307 -> 315 -0.10698
307 -> 316 -0.17910
310 -> 336 0.13623

Excited State 93: Singlet-A 4.0975 eV 302.59 nm f=0.1247 <S**2>=0.000
296 -> 311 -0.10648
302 -> 314 -0.31802
307 -> 316 0.11167
310 -> 336 0.52564
310 -> 337 0.13381

Excited State 95: Singlet-A 4.1680 eV 297.47 nm f=0.0248 <S**2>=0.000
300 -> 314 -0.20283
302 -> 314 -0.16250
305 -> 315 -0.38405
306 -> 315 0.20476
306 -> 316 0.39664

Excited State 97: Singlet-A 4.1891 eV 295.97 nm f=0.0314 <S**2>=0.000
299 -> 313 -0.25807
300 -> 314 0.14492
305 -> 315 0.38108
306 -> 316 0.40037
310 -> 335 0.16354

Excited State 102: Singlet-A 4.2368 eV 292.63 nm f=0.0413 <S**2>=0.000
295 -> 311 0.13644
295 -> 312 0.67444

Excited State 109: Singlet-A 4.3518 eV 284.90 nm f=0.0237 <S**2>=0.000
294 -> 311 0.65913

Excited State 112: Singlet-A 4.3680 eV 283.85 nm f=0.0256 <S**2>=0.000
293 -> 311 0.64892
310 -> 337 0.16856

Excited State 120: Singlet-A 4.4903 eV 276.11 nm f=0.2218 <S**2>=0.000

297 -> 313	0.57254				
307 -> 318	-0.23842				
Excited State 122:	Singlet-A	4.5246 eV	274.02 nm	f=0.0387	<S**2>=0.000
295 -> 311	0.15290				
296 -> 313	0.33781				
299 -> 312	-0.27164				
300 -> 315	-0.17646				
301 -> 313	-0.10349				
304 -> 316	-0.14587				
307 -> 318	0.34344				
Excited State 133:	Singlet-A	4.5811 eV	270.65 nm	f=0.0501	<S**2>=0.000
291 -> 311	0.25949				
291 -> 312	0.15351				
292 -> 312	0.57952				
296 -> 312	-0.10356				
307 -> 323	0.10824				
Excited State 134:	Singlet-A	4.5961 eV	269.76 nm	f=0.1039	<S**2>=0.000
296 -> 314	0.19911				
297 -> 314	0.55247				
299 -> 315	-0.12195				
300 -> 316	-0.10122				
307 -> 323	-0.12077				
Excited State 144:	Singlet-A	4.6978 eV	263.92 nm	f=0.0359	<S**2>=0.000
296 -> 314	-0.13116				
297 -> 312	-0.10686				
299 -> 315	0.12727				
300 -> 316	-0.28767				
306 -> 318	-0.15883				
307 -> 324	-0.25930				
310 -> 340	0.21849				
310 -> 342	0.32011				
Excited State 152:	Singlet-A	4.7917 eV	258.75 nm	f=0.0252	<S**2>=0.000
290 -> 311	-0.26795				
294 -> 313	0.45137				
306 -> 319	-0.16572				
306 -> 320	0.20985				
306 -> 321	-0.16872				
306 -> 322	-0.10220				
306 -> 323	-0.16234				
307 -> 324	0.14278				
Excited State 165:	Singlet-A	4.9309 eV	251.44 nm	f=0.0262	<S**2>=0.000
288 -> 311	-0.26136				
303 -> 318	0.41352				
303 -> 319	-0.13314				
303 -> 323	-0.20668				
304 -> 318	0.15060				
305 -> 323	-0.14971				
306 -> 323	0.12607				

306 -> 324 0.12386

Excited State 166: Singlet-A 4.9384 eV 251.06 nm f=0.0325 <S**2>=0.000
288 -> 311 -0.17928
293 -> 312 -0.13882
293 -> 313 -0.16188
297 -> 315 0.41066
299 -> 316 -0.14728
305 -> 321 0.10849
305 -> 323 0.18822
306 -> 324 0.13560

Excited State 169: Singlet-A 4.9549 eV 250.23 nm f=0.0873 <S**2>=0.000
287 -> 311 0.11684
291 -> 311 0.11709
292 -> 311 0.30783
294 -> 314 0.10003
296 -> 315 -0.19760
302 -> 318 -0.10551
304 -> 318 0.10631
305 -> 324 -0.11601
306 -> 324 0.22790
308 -> 317 -0.16179
308 -> 325 0.14974

Excited State 170: Singlet-A 4.9563 eV 250.15 nm f=0.0301 <S**2>=0.000
287 -> 311 -0.11868
288 -> 311 -0.15326
291 -> 311 0.17037
292 -> 311 0.39216
293 -> 313 -0.10528
296 -> 315 -0.13503
297 -> 315 -0.14368
305 -> 323 0.13929
308 -> 317 0.24293
308 -> 325 -0.20927

Excited State 172: Singlet-A 4.9626 eV 249.84 nm f=0.0805 <S**2>=0.000
287 -> 311 -0.14904
288 -> 311 0.14908
292 -> 311 -0.13188
305 -> 323 -0.10642
305 -> 324 -0.11512
306 -> 324 0.18826
308 -> 317 0.40140
308 -> 325 -0.19685

Excited State 176: Singlet-A 4.9895 eV 248.49 nm f=0.0308 <S**2>=0.000
288 -> 311 0.13899
293 -> 313 0.16100
296 -> 315 -0.24514
297 -> 315 0.21604
301 -> 315 0.12895
305 -> 323 0.17855

306 -> 323 0.29810
306 -> 324 -0.20657
307 -> 318 0.11499
307 -> 324 0.10630

Excited State 177: Singlet-A 4.9956 eV 248.19 nm f=0.0088 <S**2>=0.000
293 -> 313 -0.35665
296 -> 315 -0.12694
297 -> 316 -0.10795
298 -> 313 0.23493
299 -> 322 0.10847
300 -> 322 -0.11691
305 -> 323 -0.10797
308 -> 317 0.19673
308 -> 325 0.36767

Excited State 179: Singlet-A 5.0016 eV 247.89 nm f=0.0418 <S**2>=0.000
284 -> 311 0.19514
293 -> 313 -0.11332
296 -> 315 0.14113
302 -> 318 -0.14226
303 -> 318 -0.20642
303 -> 319 -0.20200
303 -> 320 0.27636
303 -> 321 -0.20963
303 -> 322 -0.13493
303 -> 323 -0.14330
303 -> 324 -0.13573
304 -> 324 -0.13248
305 -> 323 0.10581
305 -> 324 -0.15272

Excited State 181: Singlet-A 5.0125 eV 247.35 nm f=0.0528 <S**2>=0.000
296 -> 315 0.16581
296 -> 316 -0.11497
297 -> 316 0.13293
302 -> 318 -0.17182
303 -> 318 0.16618
303 -> 319 0.11770
303 -> 320 -0.16869
303 -> 321 0.13094
305 -> 323 0.14881
305 -> 324 -0.19095
306 -> 324 -0.10559
308 -> 325 0.23992

Excited State 198: Singlet-A 5.1074 eV 242.75 nm f=0.0399 <S**2>=0.000
294 -> 314 -0.13124
294 -> 315 -0.10784
300 -> 318 0.16254
302 -> 318 -0.26323
302 -> 320 0.15928
302 -> 321 -0.13731
305 -> 318 0.10304

305 -> 324 0.47527

Excited State 212: Singlet-A 5.1775 eV 239.47 nm f=0.0244 <S**2>=0.000
290 -> 313 0.14525
296 -> 316 0.17663
297 -> 316 -0.12238
300 -> 318 0.31052
301 -> 316 0.34967
302 -> 323 0.23967
307 -> 327 -0.19383
308 -> 327 -0.13106

Excited State 221: Singlet-A 5.2283 eV 237.14 nm f=0.0615 <S**2>=0.000
290 -> 313 0.10955
294 -> 315 -0.13756
300 -> 318 -0.16169
300 -> 319 0.12945
300 -> 320 -0.12337
300 -> 321 0.10904
300 -> 323 0.20302
302 -> 323 0.18675
302 -> 324 0.25127
303 -> 324 -0.18002
306 -> 327 0.10607
307 -> 328 -0.12107
308 -> 328 -0.16930

Excited State 222: Singlet-A 5.2331 eV 236.92 nm f=0.0292 <S**2>=0.000
300 -> 318 -0.11947
300 -> 319 0.10436
300 -> 324 0.11582
302 -> 323 0.13839
303 -> 318 -0.20112
303 -> 323 -0.28144
303 -> 324 0.44046
304 -> 324 0.13873

Excited State 223: Singlet-A 5.2376 eV 236.72 nm f=0.1918 <S**2>=0.000
282 -> 311 -0.12059
290 -> 313 0.30343
291 -> 312 0.33714
291 -> 314 -0.13571
292 -> 312 -0.10356
294 -> 315 -0.24949
300 -> 318 -0.12099
302 -> 324 -0.10074

Excited State 224: Singlet-A 5.2406 eV 236.58 nm f=0.0527 <S**2>=0.000
280 -> 311 0.14748
290 -> 313 -0.17607
291 -> 312 0.49148
292 -> 312 -0.15106
294 -> 315 0.13022
300 -> 323 0.10013

302 -> 324	0.16985				
303 -> 324	0.11203				
Excited State 225:	Singlet-A	5.2477 eV	236.26 nm	f=0.0201	<S**2>=0.000
280 -> 311	0.13672				
299 -> 318	-0.11885				
300 -> 318	0.12598				
302 -> 324	0.18605				
303 -> 324	0.13449				
307 -> 327	0.36278				
308 -> 327	-0.13208				
309 -> 342	0.19366				
309 -> 344	-0.16551				
Excited State 227:	Singlet-A	5.2619 eV	235.63 nm	f=0.0292	<S**2>=0.000
280 -> 311	-0.15904				
281 -> 311	-0.10290				
290 -> 313	0.18490				
291 -> 312	0.15082				
291 -> 313	-0.11323				
291 -> 314	0.44635				
292 -> 314	-0.19522				
302 -> 323	-0.14470				
305 -> 325	-0.13671				
Excited State 228:	Singlet-A	5.2654 eV	235.47 nm	f=0.0329	<S**2>=0.000
280 -> 311	0.11858				
291 -> 314	0.15082				
294 -> 315	0.16540				
300 -> 324	0.15278				
302 -> 323	0.29817				
302 -> 324	-0.29343				
307 -> 327	0.25696				
308 -> 327	-0.13590				
Excited State 252:	Singlet-A	5.4361 eV	228.08 nm	f=0.0208	<S**2>=0.000
294 -> 315	0.19109				
294 -> 316	0.44554				
299 -> 319	0.20506				
299 -> 320	-0.23995				
299 -> 321	0.21375				
300 -> 323	0.13456				
308 -> 332	-0.11122				
Excited State 272:	Singlet-A	5.5499 eV	223.40 nm	f=0.0323	<S**2>=0.000
278 -> 312	0.30628				
287 -> 313	0.25962				
293 -> 316	0.30913				
295 -> 315	0.11066				
307 -> 329	-0.19617				
307 -> 332	-0.13027				
309 -> 346	0.11551				
309 -> 347	-0.11005				

Excited State 290:	Singlet-A	5.6173 eV	220.72 nm	f=0.0280	<S**2>=0.000
278 -> 312	-0.11628				
295 -> 317	0.59242				
304 -> 327	-0.10617				
Excited State 310:	Singlet-A	5.6808 eV	218.25 nm	f=0.0351	<S**2>=0.000
282 -> 313	0.24882				
283 -> 313	0.38934				
283 -> 314	0.16840				
287 -> 314	-0.22639				
290 -> 315	-0.10870				
305 -> 329	0.12358				
306 -> 329	-0.18086				
Excited State 317:	Singlet-A	5.7231 eV	216.64 nm	f=0.0283	<S**2>=0.000
276 -> 311	0.40307				
279 -> 311	-0.11667				
290 -> 316	0.14080				
291 -> 316	-0.20227				
297 -> 318	-0.19158				
297 -> 319	-0.14373				
297 -> 320	0.19384				
297 -> 321	-0.16780				
Excited State 406:	Singlet-A	6.0611 eV	204.56 nm	f=0.0252	<S**2>=0.000
273 -> 311	-0.17911				
277 -> 313	0.10261				
279 -> 314	-0.14796				
281 -> 314	0.17913				
287 -> 315	-0.15965				
288 -> 315	-0.11093				
288 -> 316	-0.10207				
291 -> 316	-0.11268				
292 -> 316	-0.20044				
294 -> 319	0.10397				
294 -> 320	-0.13764				
294 -> 321	0.10865				
294 -> 324	0.17217				
296 -> 325	-0.10059				
297 -> 325	-0.13330				
308 -> 335	0.22693				
Excited State 415:	Singlet-A	6.0928 eV	203.49 nm	f=0.0287	<S**2>=0.000
272 -> 311	-0.10401				
273 -> 311	0.15086				
279 -> 312	-0.13245				
279 -> 314	-0.15004				
281 -> 314	-0.13889				
285 -> 313	0.13341				
288 -> 316	-0.12279				
294 -> 318	-0.11996				
294 -> 324	-0.10716				
302 -> 329	0.10601				
302 -> 332	-0.10786				

304 -> 329	0.12345
307 -> 335	0.15032
308 -> 335	0.35639

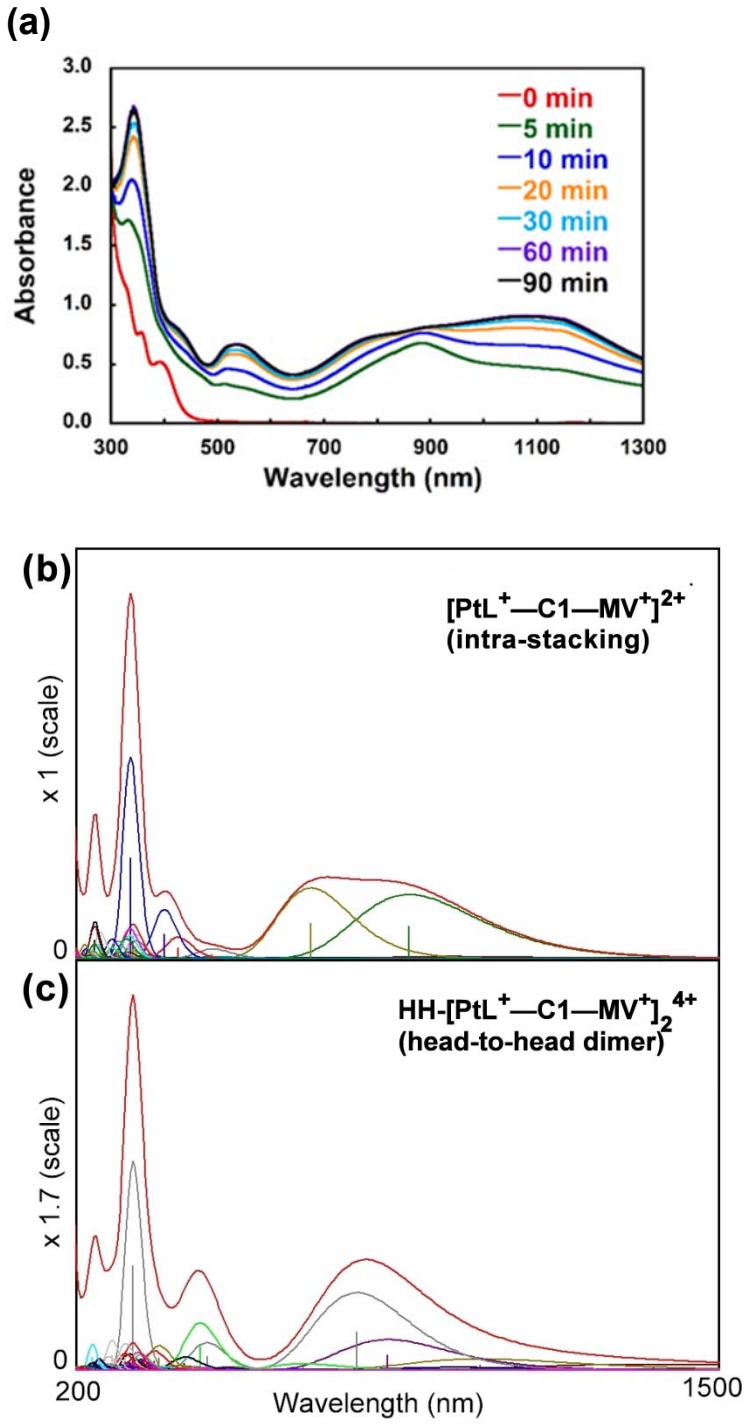
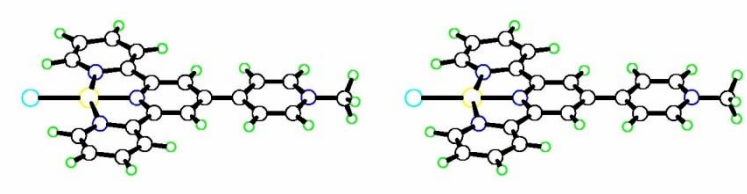
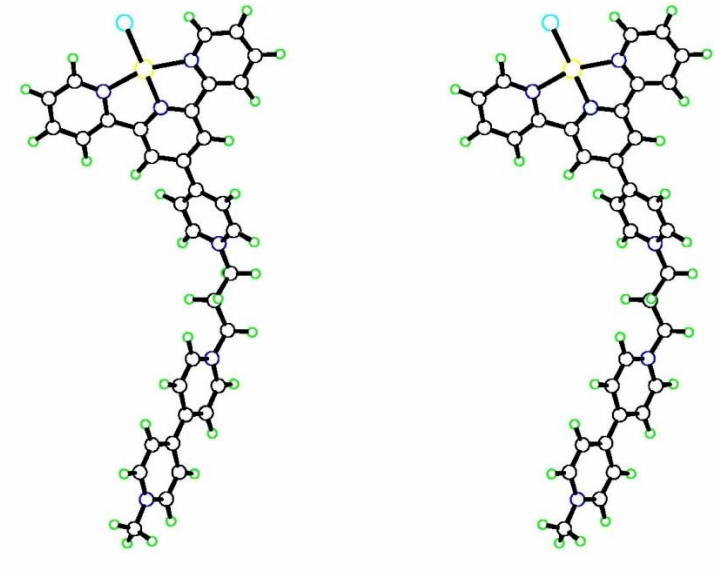


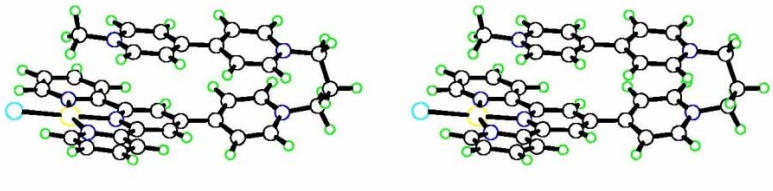
Figure S12. Spectral changes during 90-min irradiation of an aqueous acetate buffer solution (0.1 M, pH = 5.0, 20 °C under Ar atmosphere) containing 30 mM EDTA, 0.1 mM **PtL**$^{2+}$**-C1-MV**$^{2+}$, and 0.1 M NaCl (a, Figure 3 in MS). Spectral features simulated for two relevant reduced species (b) and (c).



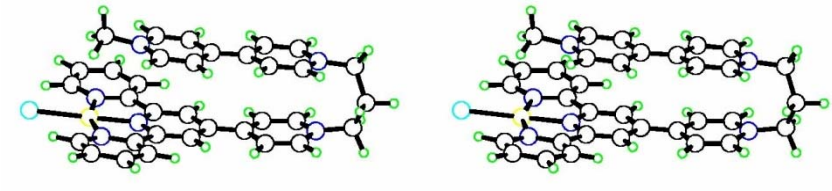
$PtL^{+}\bullet$



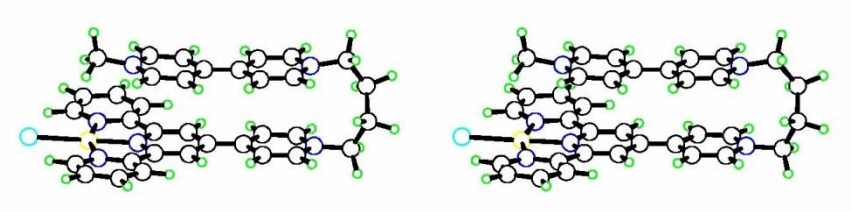
$[PtL^{2+}—C1—MV^{+}\bullet]^{3+}$



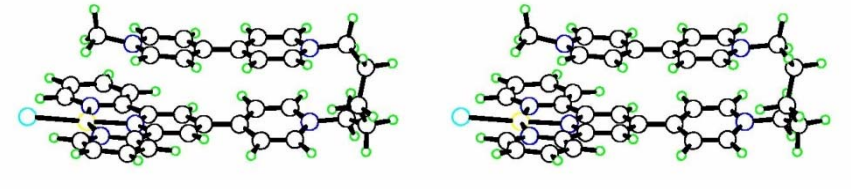
$[PtL^{1.5+}—C1—MV^{1.5+}]^{3+}$



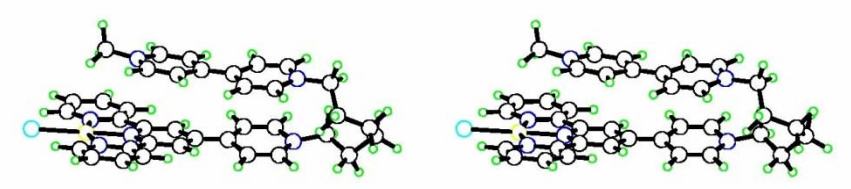
$[PtL^{+}\bullet—C1—MV^{+}\bullet]^{2+}$



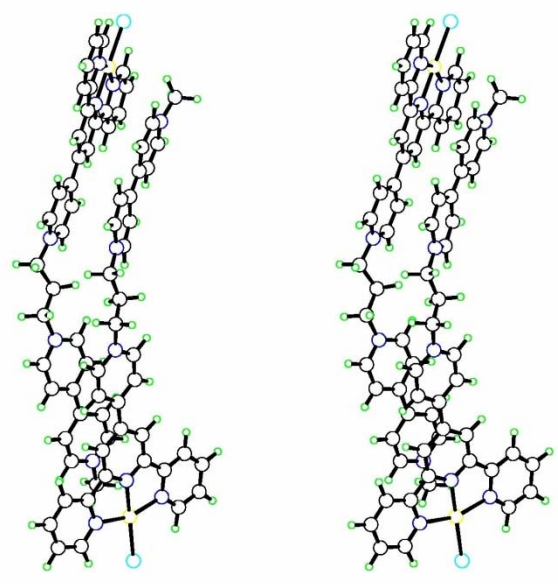
$[PtL^{+}\bullet—C2—MV^{+}\bullet]^{2+}$



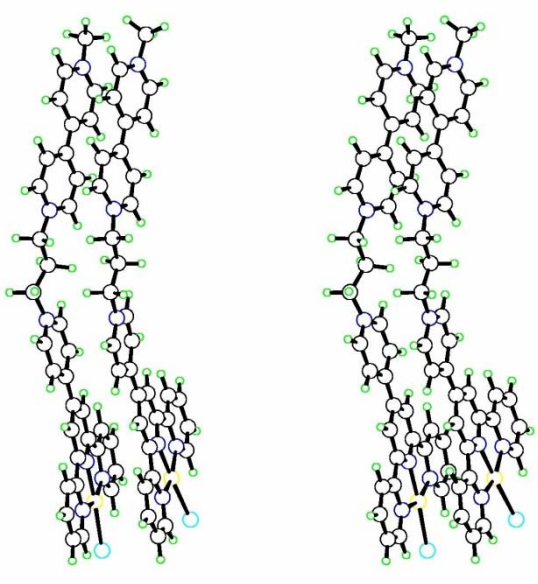
$[PtL^{+}\bullet—C3—MV^{+}\bullet]^{2+}$



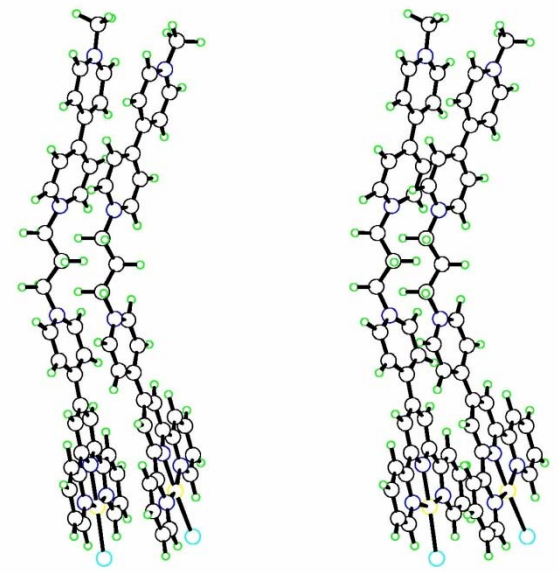
$[PtL^{+}\bullet—C4—MV^{+}\bullet]^{2+}$



HT-[PtL^{+}•—C1—MV^{+}•]$_2^{4+}$



HH(m)-[PtL^{+}•—C1—MV^{+}•]$_2^{4+}$



HH(c2)-[PtL^{+}•—C1—MV^{+}•]$_2^{4+}$

Figure S13. Electronic transitions computed by TD-DFT for the closed-shell singlet state of **[PtL^{+}-C1-MV^{+}]$^{2+}$**, for which part of the Gaussian output is shown. Relevant MO's are shown below