

Supporting Information

**Understanding Electrostatics and Covalency Effects in Highly Anisotropic Organometallic Sandwich Dysprosium Complexes [Dy(C<sub>m</sub>R<sub>m</sub>)<sub>2</sub>] (where R = H, SiH<sub>3</sub>, CH<sub>3</sub> and m = 4 to 9): A Computational Perspective**

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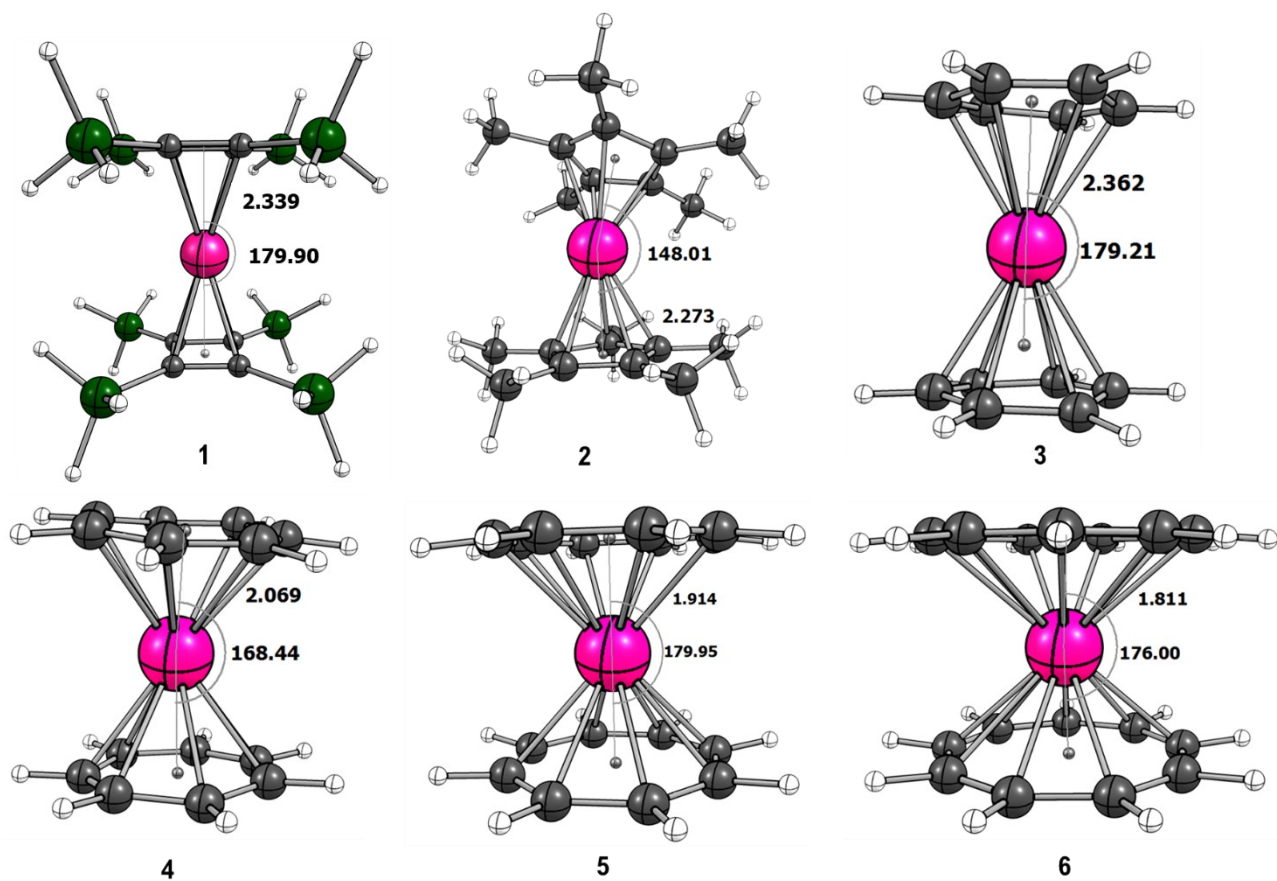
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**Figure S1:** DFT optimized structures of complexes 1-6. (Colour code: Dy (magenta), Si (green), C (grey), H (white)).

**Table S1(a):** Comparison of selected structural parameters of complex 5 at different level of theory with the X-ray crystal structure.

[Dy(C <sub>8</sub> H <sub>8</sub> ) <sub>2</sub> ] <sup>-</sup>	X-ray crystal structure	BP86	B3LYP	TPSSh	M06	wB97X-D3
Avg. Dy-L (Å)	2.641	2.664	2.674	2.651	2.663	2.656
Dy-L <sub>cent</sub> (Å)	1.918	1.912	1.932	1.897	1.921	1.910
∠ L <sub>cent</sub> -M-L <sub>cent</sub> (°)	180	180	180	180	180	180

**Table S1(b):** Comparison of selected structural parameters of 1-6 optimized at BP86 level of theory and relevant X-ray crystal structures.

DFT Optimized geometry			Reported X-ray crystal structure			
Species	Dy-L <sub>cent</sub> (Å)	∠ L <sub>cent</sub> -M-L <sub>cent</sub>	Species	L <sub>cent</sub> -M	∠ L <sub>cent</sub> -M-L <sub>cent</sub>	ref
[Dy(C <sub>4</sub> (SiH <sub>3</sub> ) <sub>4</sub> ) <sub>2</sub> ] <sup>1-</sup>	2.339	179.9°	[Dy(C <sub>4</sub> (Si(CH <sub>3</sub> ) <sub>3</sub> ) <sub>4</sub> (C <sub>5</sub> (CH <sub>3</sub> ) <sub>4</sub> <sup>t</sup> Bu)]	2.273*	141.5°	[1]
[Dy(C <sub>5</sub> (CH <sub>3</sub> ) <sub>5</sub> ) <sub>2</sub> ] <sup>+</sup>	2.273	148.0°	[Dy(C <sub>5</sub> (CH <sub>3</sub> ) <sub>5</sub> ) <sub>2</sub> (NH <sub>3</sub> )] <sup>+</sup>	2.347	143.9°	[2]*
[Dy(C <sub>6</sub> H <sub>6</sub> ) <sub>2</sub> ] <sup>3+</sup>	2.362	179.2°	[Mo(C <sub>6</sub> H <sub>6</sub> )] <sup>3+</sup>	1.779	176.3°	[3]
[Dy(C <sub>7</sub> H <sub>7</sub> ) <sub>2</sub> ] <sup>3-</sup>	2.070	168.4°	[U(C <sub>7</sub> H <sub>7</sub> ) <sub>2</sub> ] <sup>3-</sup>	1.986	180.0°	[4]
[Dy(C <sub>8</sub> H <sub>8</sub> ) <sub>2</sub> ] <sup>-</sup>	1.912	180.0°	[Dy(C <sub>8</sub> H <sub>8</sub> ) <sub>2</sub> ] <sup>-</sup>	1.921	180.0°	[5]
[Dy(C <sub>9</sub> H <sub>9</sub> ) <sub>2</sub> ] <sup>+</sup>	1.809	179.9°	[Dy(C <sub>9</sub> H <sub>9</sub> )(C <sub>8</sub> H <sub>8</sub> )]	1.875*	173.0°	[6]

\*NH<sub>3</sub> is directly bonded with {DyCp<sub>2</sub>};  
 The avg. ∠L<sub>cent</sub>-M-L<sub>cent</sub> bond angle in DFT optimized geometry of various substituted [DyCp<sub>2</sub>]<sup>+</sup> complexes ranges between 144° to 162° (see *J. Am. Chem. Soc.*, 2021, 143, 5943)  
 \*Dy-(C<sub>4</sub>(Si(CH<sub>3</sub>)<sub>3</sub>)<sub>4</sub>(centroid), \*Dy-(C<sub>9</sub>H<sub>9</sub>)(centroid);

**Table S2:** EDA analysis for [Dy(C<sub>m</sub>R<sub>m</sub>)<sub>2</sub>] (where R = H, SiH<sub>3</sub>, CH<sub>3</sub> m = 4 to 9, and n = 1, 3) complexes. All the values provided here are in the kcal/mol.

Parameters	1	2	3	4	5	6
<b>Orbital Interaction</b>	-147.1	-144.5	-123.6	-172.6	-139.4	-134.9
<b>Pauli Repulsion</b>	106.9	109.0	52.9	121.6	101.8	82.9
<b>Electrostatic Energy</b>	-273.9	-267.5	-49.8	-157.9	-281.4	-226.8
<b>Dispersion correction</b>	-8.6	-8.2	-2.9	-4.6	-6.2	-7.9
<b>Total Binding Energy</b>	-322.7	-311.3	-123.4	-213.5	-325.1	-286.7
<b>% of covalency</b>	34.9	35.1	71.3	52.2	33.1	37.3
% of covalency = E <sub>orb</sub> /(E <sub>orb</sub> + E <sub>elec</sub> )						

**Table S3:** Percentage contribution of the decomposed energies to the total bonding energy for complexes **1-6** along with fragmentation scheme.

Full Complex	$f_1$	$f_2$	$E_{orb}$ (%)	$E_{Pauli}$ (%)	$E_{disp}$ (%)	$E_{elec}$ (%)
<b>1</b>	{Dy(C <sub>4</sub> (SiH <sub>3</sub> ) <sub>4</sub> )} <sup>+</sup>	{C <sub>4</sub> (SiH <sub>3</sub> ) <sub>4</sub> } <sup>2-</sup>	45.6	33.1	2.6	84.9
<b>2</b>	{Dy(C <sub>5</sub> (CH <sub>3</sub> ) <sub>5</sub> ) <sub>2</sub> } <sup>2+</sup>	{C <sub>5</sub> (CH <sub>3</sub> ) <sub>5</sub> } <sup>-</sup>	46.4	35.0	2.6	85.9
<b>3</b>	{Dy(C <sub>6</sub> H <sub>6</sub> ) <sub>3</sub> } <sup>3+</sup>	{C <sub>6</sub> H <sub>6</sub> }	100.2	42.9	2.3	40.4
<b>4</b>	{Dy(C <sub>7</sub> H <sub>7</sub> )}	{C <sub>7</sub> H <sub>7</sub> } <sup>3-</sup>	80.8	57.0	2.2	74.0
<b>5</b>	{Dy(C <sub>8</sub> H <sub>8</sub> ) <sub>2</sub> } <sup>+</sup>	{C <sub>8</sub> H <sub>8</sub> } <sup>2-</sup>	42.9	31.3	1.9	86.6
<b>6</b>	{Dy(C <sub>9</sub> H <sub>9</sub> ) <sub>3</sub> } <sup>2+</sup>	{C <sub>9</sub> H <sub>9</sub> } <sup>-</sup>	47.1	28.9	2.7	79.1

% contribution to total binding energy =  $(E/E_{tot}) * 100$

Here  $f_1$  and  $f_2$  represents two different fragments for the EDA calculations.

**Table S4:** AILFT computed the Slater Condon parameters  $F^2$ ,  $F^4$ , and  $F^6$ , the one-electron effective parameters for spin-orbit coupling ( $\zeta$ ) for complexes **1-6** at NEVPT2 level of theory. The values in the parenthesis are the CASSCF computed values. All the values provided here are in the cm<sup>-1</sup>.

	Dy(III) ion	1	2	3	4	5	6
$F^2$	109944.1 (121779.1)	107554.8 (120594.4)	107539.3 (120548.2)	108269.8 (20974.2)	107370.3 (120501.4)	107540.5 (120568.6)	107807.3 (120729.8)
$F^4$	70794.3 (76392.7)	69972.8 (75434.4)	69953.4 (75453.8)	70206.1 (75752.6)	69768.4 (75519.5)	69846.3 (75592.5)	70019.8 (75722.8)
$F^6$	55305.7 (54949.3)	54658.3 (54329.1)	54696.4 (54344.9)	54872.6 (54527.4)	54629.0 (54338.7)	54666.4 (54379)	54763.2 (54460.1)
$\zeta$	1740.2	1730.5	1729.3	1733.9	1728.7	1729.3	1731.0

**Table S5:** AILFT computed the Racah parameters  $E^1$ ,  $E^2$ , and  $E^3$  for complexes **1-6** at NEVPT2 level of theory. The values in the parenthesis are the CASSCF computed values. All the values provided here are in the cm<sup>-1</sup>.

	Dy(III) ion	1	2	3	4	5	6
$E^1$	7140.2 (7670.5)	7018.7 (7588.3)	7018.9 (7587.6)	7055.4 (7614.9)	7006.6 (7587.3)	7015.5 (7592.6)	7031.7 (7603.7)
$E^2$	38.5 (42.6)	37.5 (42.2)	37.5 (42.2)	37.8 (42.3)	37.4 (42.1)	37.5 (42.1)	37.6 (42.2)
$E^3$	716.5 (815.9)	700.0 (808.0)	699.7 (807.6)	704.8 (810.6)	698.4 (807.4)	699.6 (807.9)	701.5 (809.0)

Supporting Information

The Racah parameters  $E^1$ ,  $E^2$  and  $E^3$  (for f-electrons), can be stated in terms of the Slater-Condon parameters  $F^2$ ,  $F^4$ , and  $F^6$ , using the following equation:

$$F_2 = \frac{F^2}{225} \dots\dots\dots(1)$$

$$F_4 = \frac{F^4}{1089} \dots\dots\dots(2)$$

$$F_6 = \frac{F^6}{7361.64} \dots\dots\dots(3)$$

$$E^1 = \frac{70F_2 + 231F_4 + 2002F_6}{9} \dots\dots\dots(4)$$

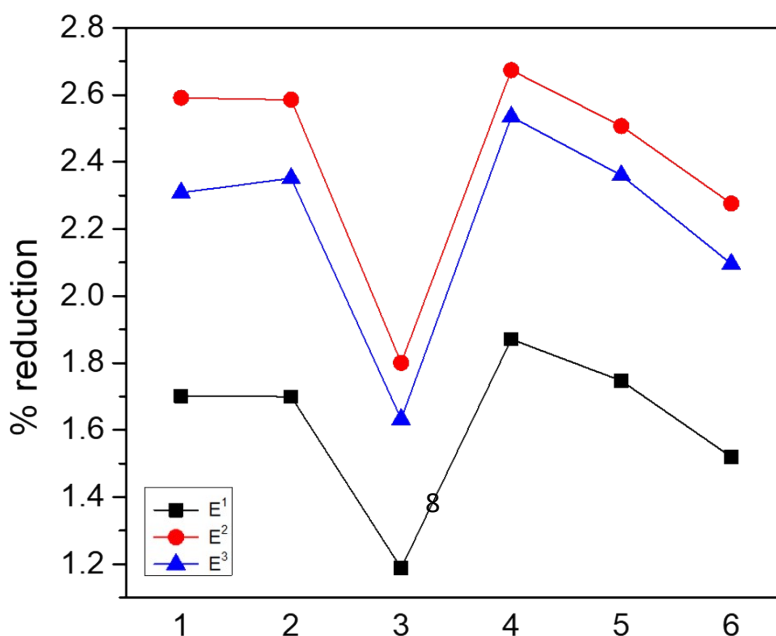
$$E^2 = \frac{F_2 - 3F_4 + 7F_6}{9} \dots\dots\dots(5)$$

$$E^3 = \frac{5F_2 + 6F_4 + 91F_6}{3} \dots\dots\dots(6)$$

**Table S6:** Reduction (%) in Slater Condon parameter  $F^2$ ,  $F^4$ , and  $F^6$  and Racah parameters  $E^1$ ,  $E^2$ , and  $E^3$  for complexes **1-6** at NEVPT2 level of theory. The values in the parenthesis are the CASSCF computed values.

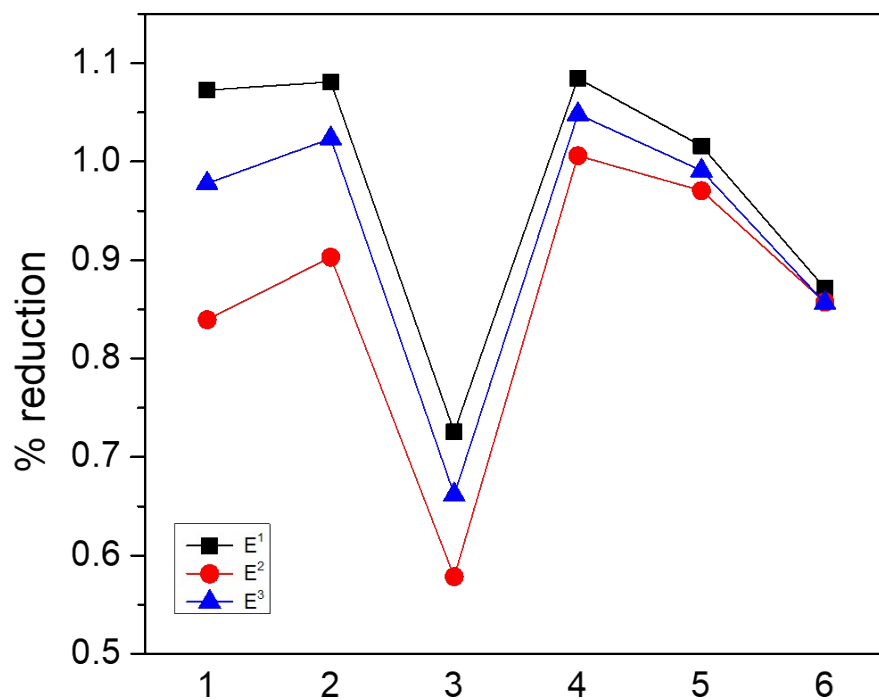
	$F^2(\%)$	$F^4(\%)$	$F^6(\%)$	$E^1(\%)$	$E^2(\%)$	$E^3(\%)$	$\zeta(\%)$
<b>1</b>	2.2(1.0)	1.2(1.3)	1.2(1.1)	1.7(1.1)	2.6(0.8)	2.3(1.0)	0.6
<b>2</b>	2.2(1.0)	1.2(1.2)	1.1(1.1)	1.7(1.1)	2.6(0.9)	2.4(1.0)	0.6
<b>3</b>	1.5(0.7)	0.8(0.8)	0.8(0.8)	1.2(0.7)	1.8(0.6)	1.6(0.7)	0.4
<b>4</b>	2.3(1.0)	1.4(1.1)	1.2(1.1)	1.9(1.1)	2.7(1.0)	2.5(1.0)	0.7
<b>5</b>	2.2(1.0)	1.3(1.0)	1.2(1.0)	1.7(1.0)	2.5(1.0)	2.4(1.0)	0.6
<b>6</b>	1.9(0.9)	1.1(0.9)	1.0(0.9)	1.5(0.9)	2.3(0.9)	2.1(0.9)	0.5

Reduction (%) = [1 - (complex/free-ion)] \* 100

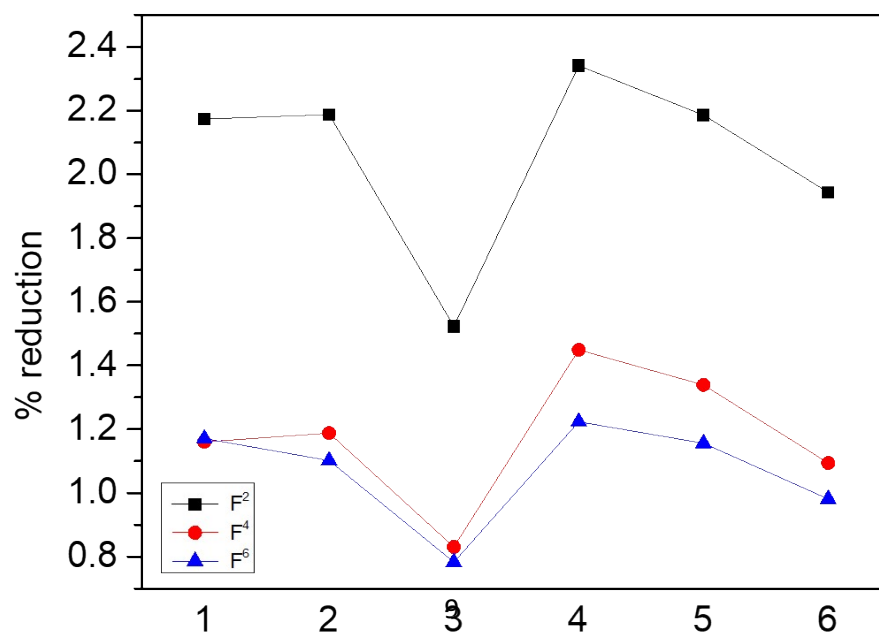


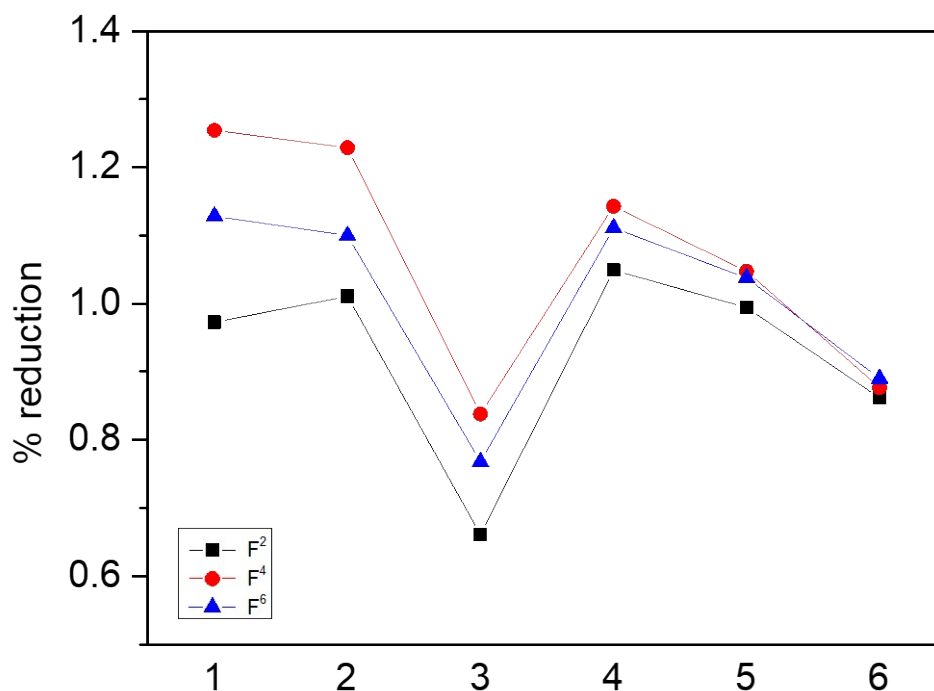


**Figure S2:** AILFT-NEVPT2 computed trends in the reduction (%) in Racah parameters  $E^1$ ,  $E^2$ , and  $E^3$  for complexes 1-6.



**Figure S3:** AILFT-CASSCF computed trends in the reduction (%) in Racah parameters  $E^1$ ,  $E^2$ , and  $E^3$  for complexes 1-6.

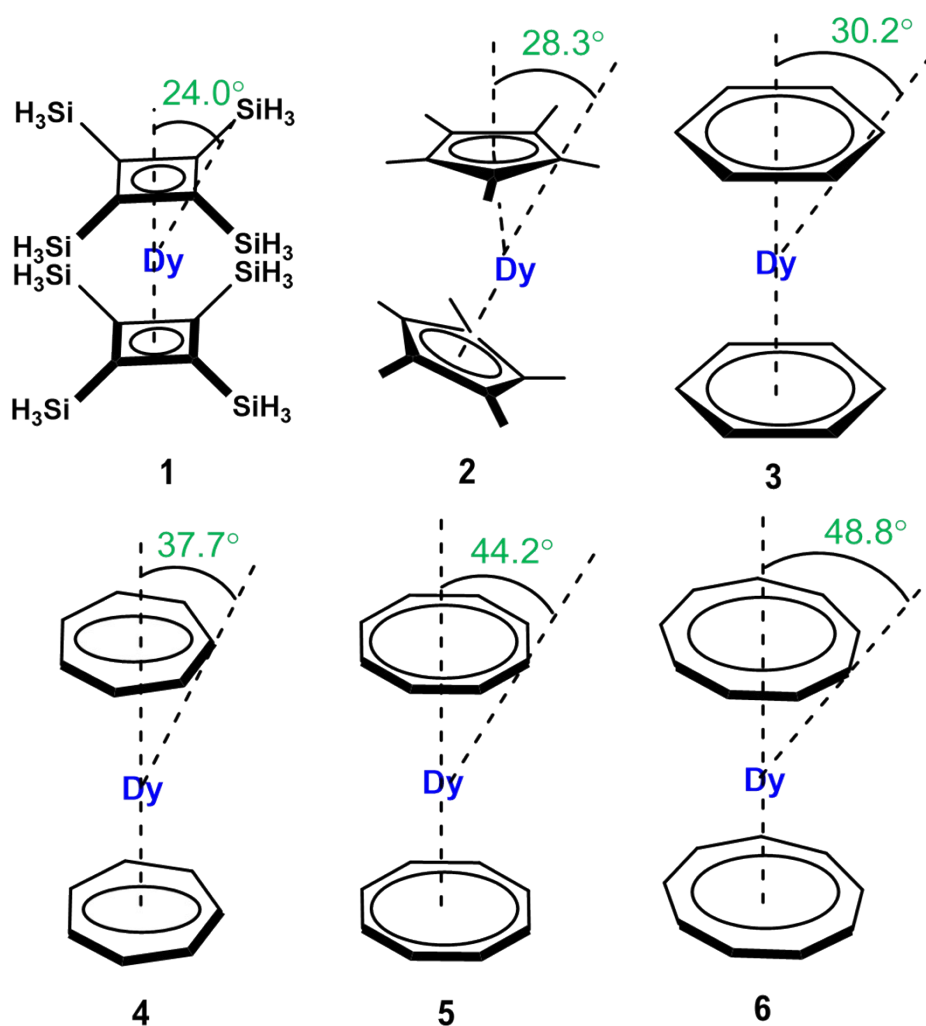


**Figure S4:** AILFT-NEVPT2 computed trends in the reduction (%) in Slater Condon parameters  $F^2$ ,  $F^4$ , and  $F^6$  for complexes 1-6.**Figure S5:** AILFT-CASSCF computed trends in the reduction (%) in Slater Condon parameters  $F^2$ ,  $F^4$ , and  $F^6$  for complexes 1-6.**Table**

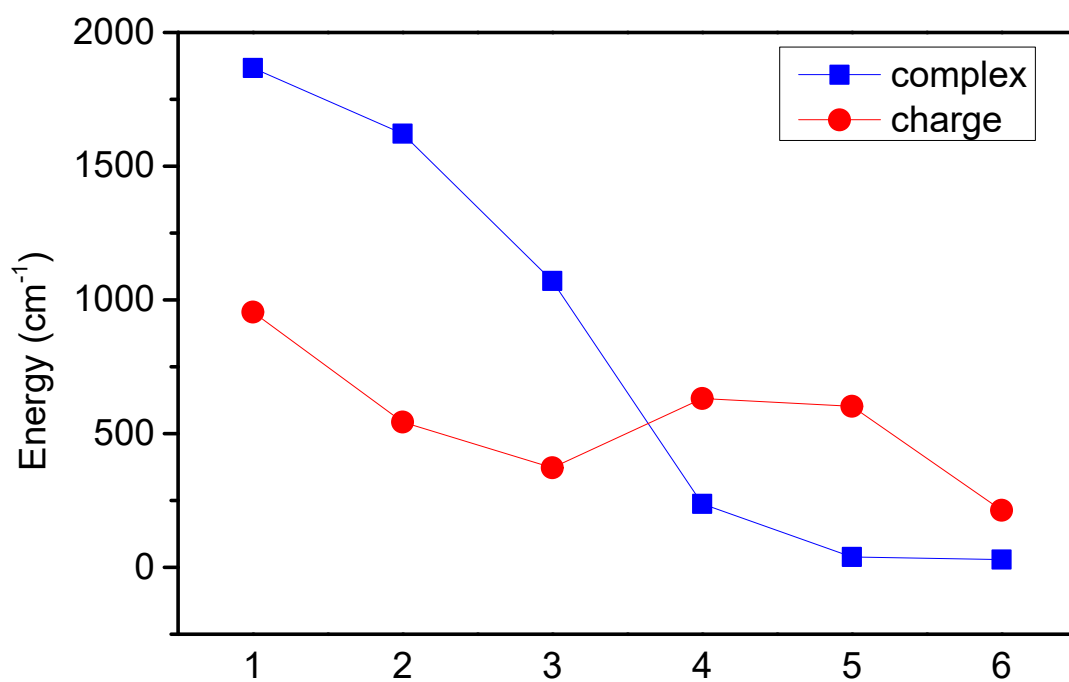
Complex	Total	4f	5d	6s	6p
1	1.24	0.14	0.69	0.35	0.59
2	1.19	0.14	0.95	0.32	0.41
3	0.54	0.27	0.84	0.45	0.90
4	0.71	0.19	1.39	0.20	0.51
5	0.64	0.41	1.29	0.10	0.56
6	0.58	0.53	1.27	0.06	0.56

<sup>a</sup>The total charge of the Dy<sup>III</sup> Ion and the electron populations of 4f, 5d, 6p, and 6s orbitals are presented.

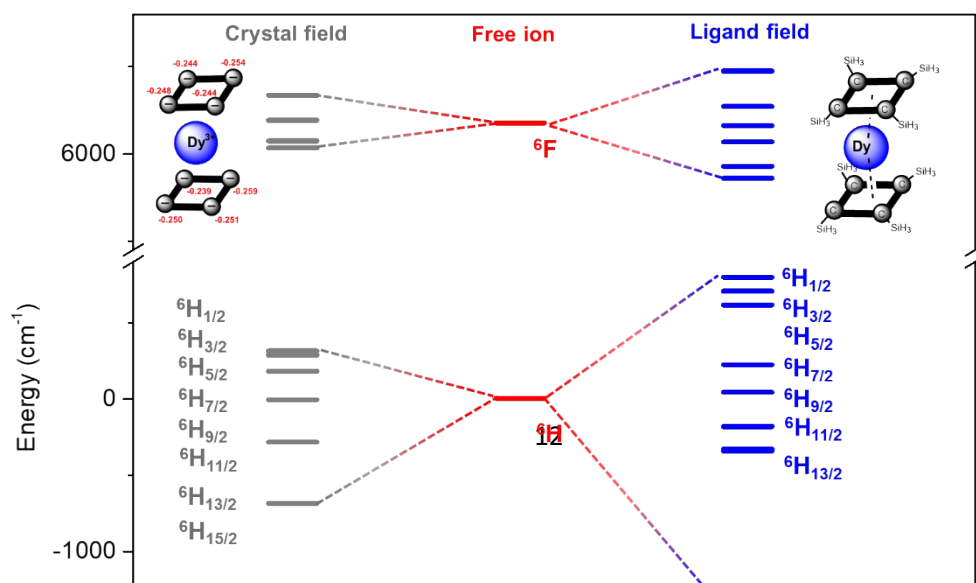
**S7:**Comparison of CASSCF-Derived Mulliken Charges<sup>a</sup> for the Dy<sup>III</sup> Ion in complex 1-6.



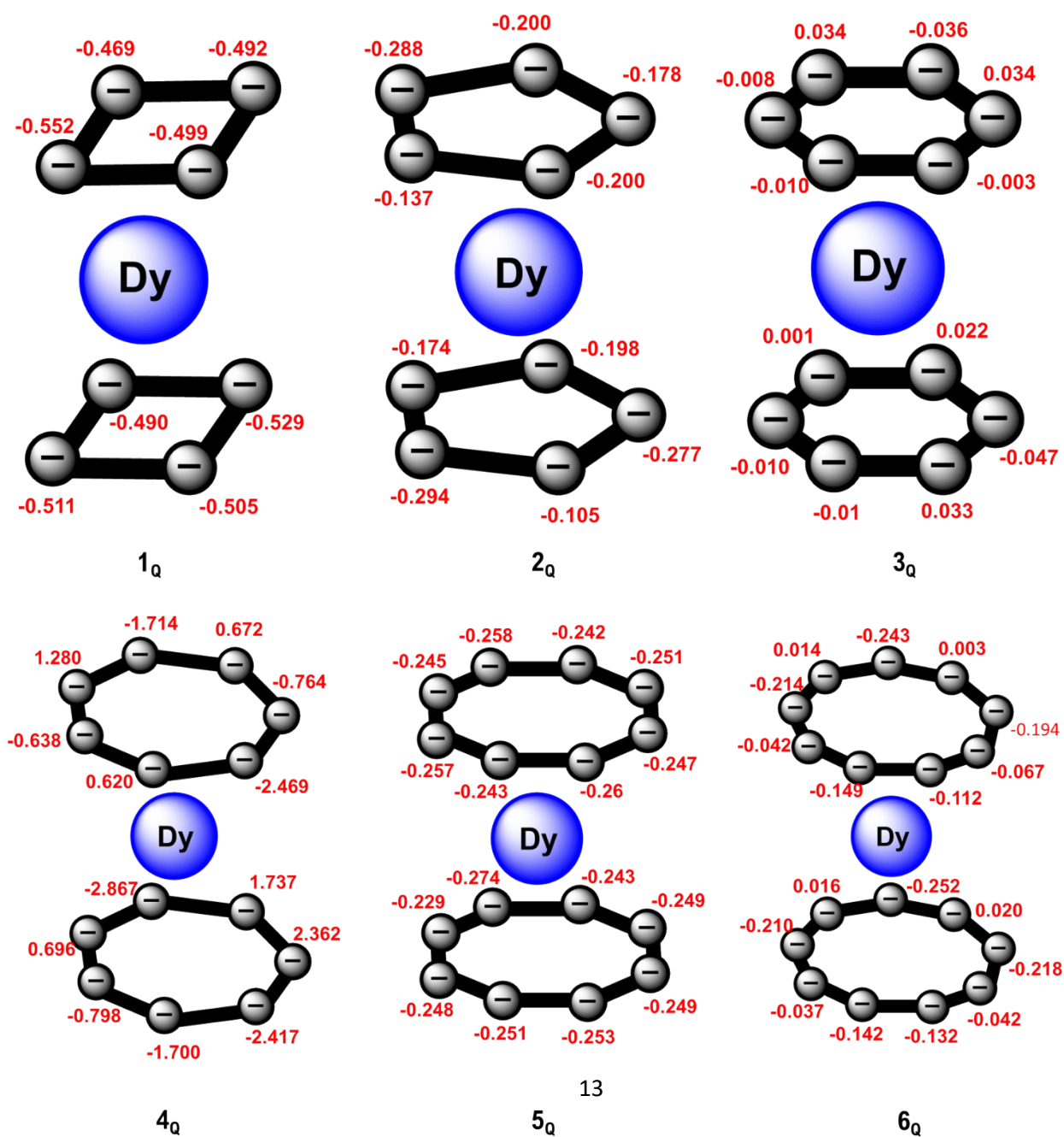
**Scheme S1:** Schematic representation of cone angle ( $\angle L_{\text{cent}}\text{-M-L}$ ) in DFT optimized geometries of **1** – **6** complexes.



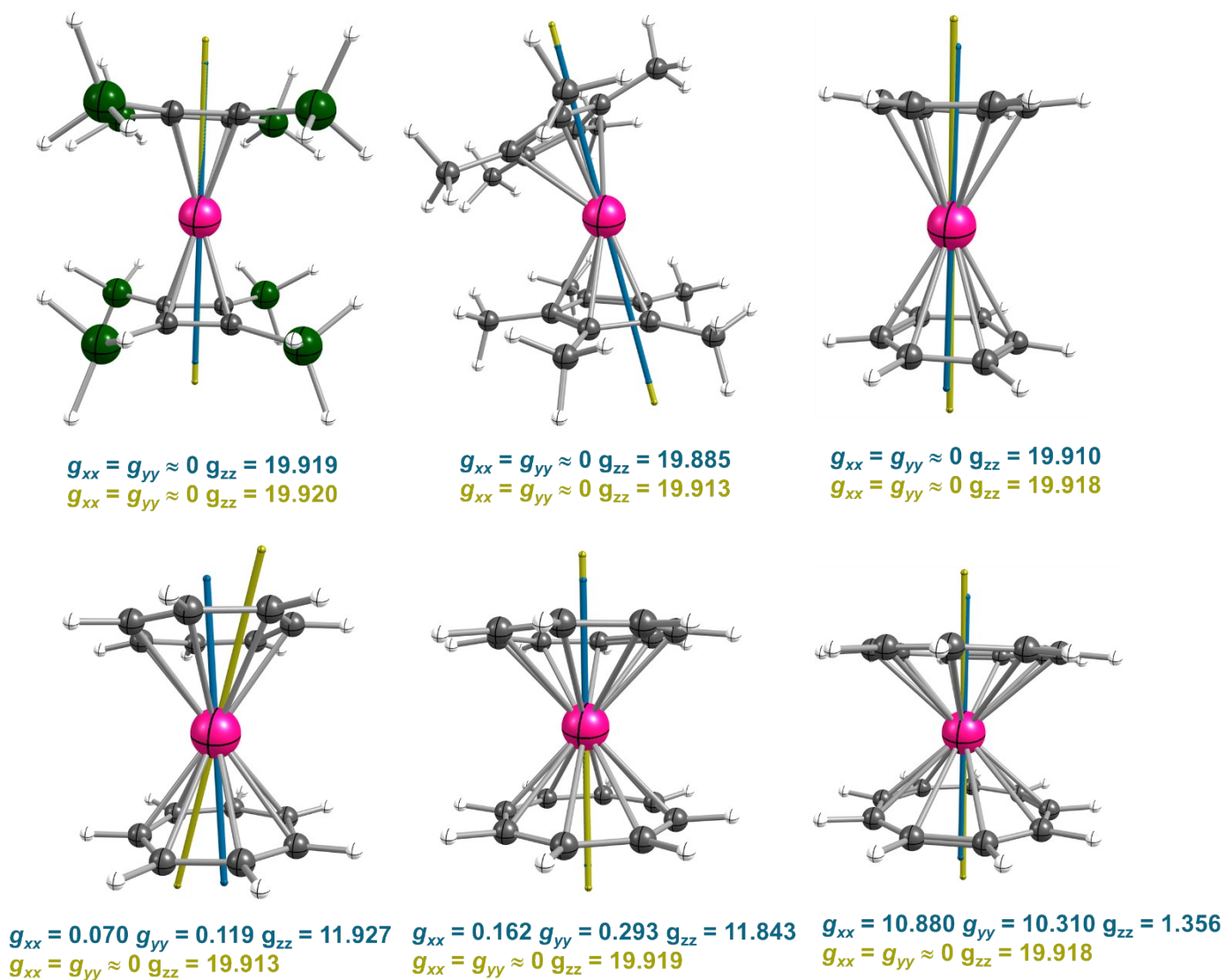
**Figure S6:** CASSCF/NEVPT2 computed the trends of Barrier height ( $U_{cal}$ ) for complexes **1-6** (blue squares) and **1<sub>Q</sub>-6<sub>Q</sub>** (red circles).



**Figure S7:** CASSCF/NEVPT2 computed Splitting of  ${}^6\text{H}$  and  ${}^6\text{F}$  states under Crystal field (grey bars) and Ligand field (blue bars) for the complex 1.



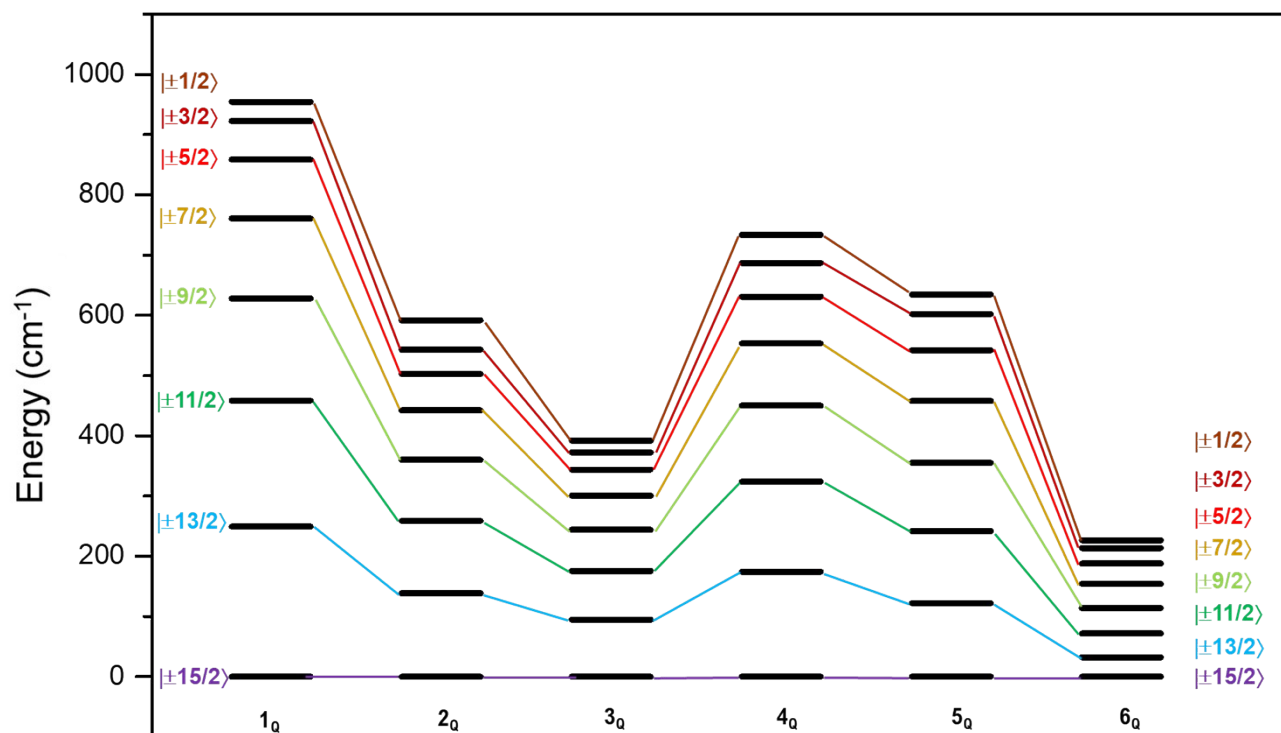
**Figure S8:** DFT computed charges on ligands (by summing up the charges of the atoms attached to the respective carbons of the ring) using CHELPG (CHarges from ELectrostatic Potential Grid) method for complexes 1-6.



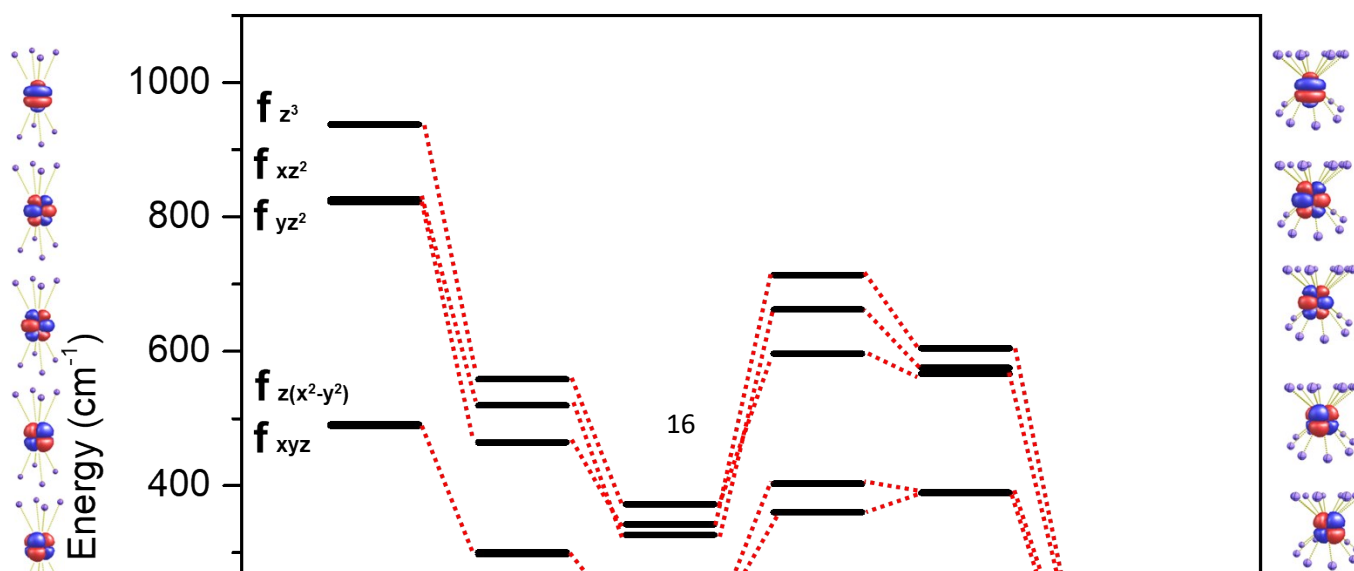
**Figure S9:** SINGLE\_ANISO computed g-tensor orientation along with g-values and wave function decomposition for ground state Kramer doublet for complexes **1-6** (cyan) and **1<sub>Q</sub>-6<sub>Q</sub>** (yellow). The wave function decomposition for **1<sub>Q</sub>-6<sub>Q</sub>** is 100%  $|\pm 15/2\rangle$ . Color code: Dy (magenta); C (grey); Si (green); H (white).

**Table S8:** SINGLE\_ANISO computed the properties of the two lowest Kramers' doublets of the  ${}^6\text{H}_{15/2}$  multiplet in complexes **1<sub>Q</sub>-6<sub>Q</sub>**.

Complex	KD	E / cm <sup>-1</sup>	g <sub>xx</sub>	g <sub>yy</sub>	g <sub>zz</sub>	$\theta$	K <sub>QTM</sub>	$m_J$	U <sub>cal</sub> /cm <sup>-1</sup>
1 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.920	-	0.18E-08	$ \pm 15/2\rangle$	954.5
	KD2	249.4	0.000	0.000	17.140	0.0	0.16E-07	$ \pm 13/2\rangle$	
2 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.913	-	0.61E-07	$ \pm 15/2\rangle$	543.0
	KD2	138.4	0.000	0.000	17.178	0.1	0.71E-06	$ \pm 13/2\rangle$	
3 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.918	-	0.20E-07	$ \pm 15/2\rangle$	372.5
	KD2	94.7	0.000	0.000	17.216	3.8	0.30E-05	$ \pm 13/2\rangle$	
4 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.913	-	0.21E-05	$ \pm 15/2\rangle$	631.2
	KD2	174.2	0.000	0.000	17.161	2.0	0.11E-03	$ \pm 13/2\rangle$	
5 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.919	-	0.32E-09	$ \pm 15/2\rangle$	602.2
	KD2	122.2	0.000	0.000	17.205	0.4	0.80E-07	$ \pm 13/2\rangle$	
6 <sub>Q</sub>	KD1	0.0	0.000	0.000	19.918	-	0.20E-05	$ \pm 15/2\rangle$	213.7
	KD2	32.1	0.000	0.000	17.247	0.7	0.24E-04	$ \pm 13/2\rangle$	



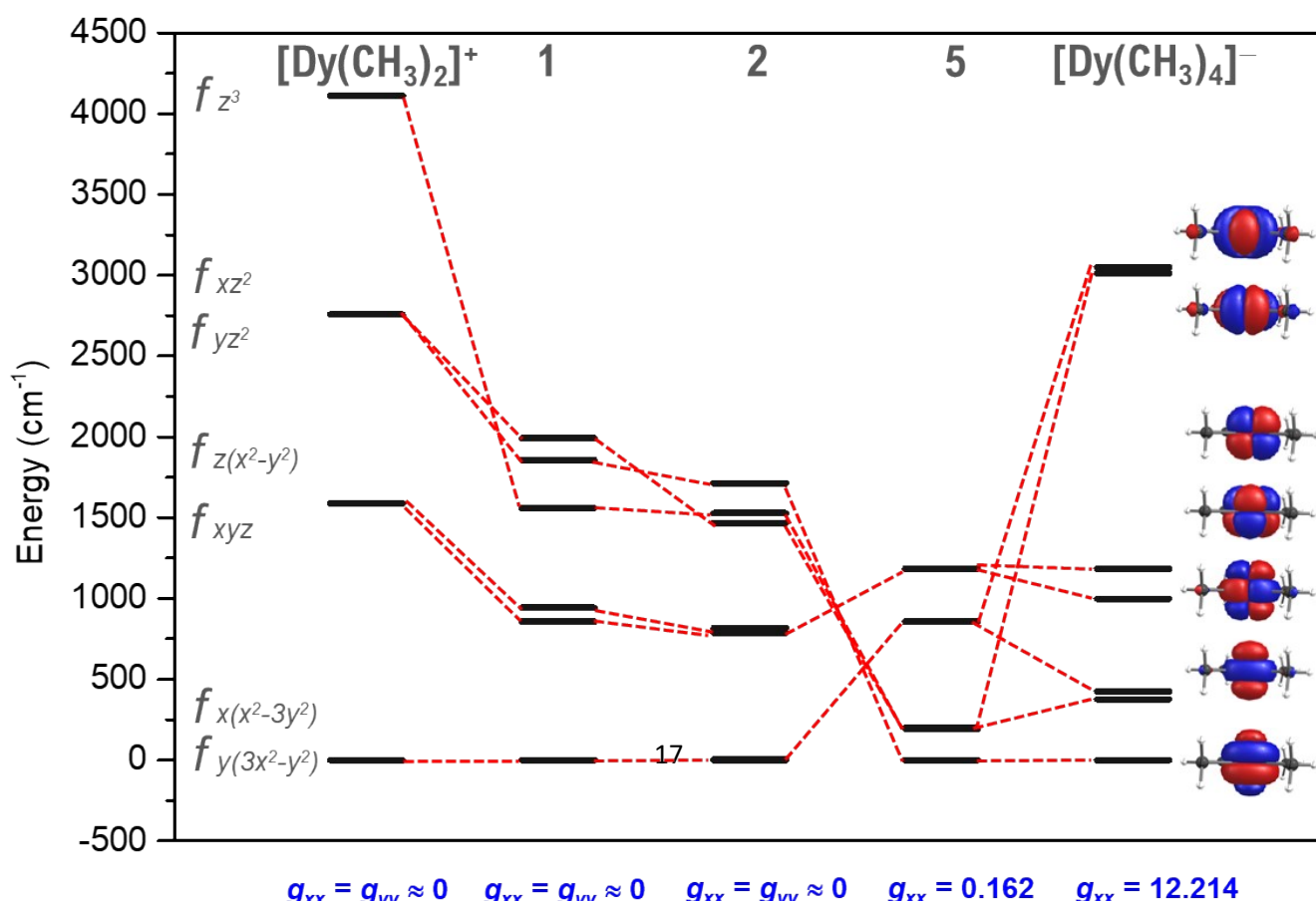
**Figure S10:** CASSCF/NEVPT2 computed the trends of Kramer's Doublets (in  $\text{cm}^{-1}$ ) for  $1_Q$ - $6_Q$ .



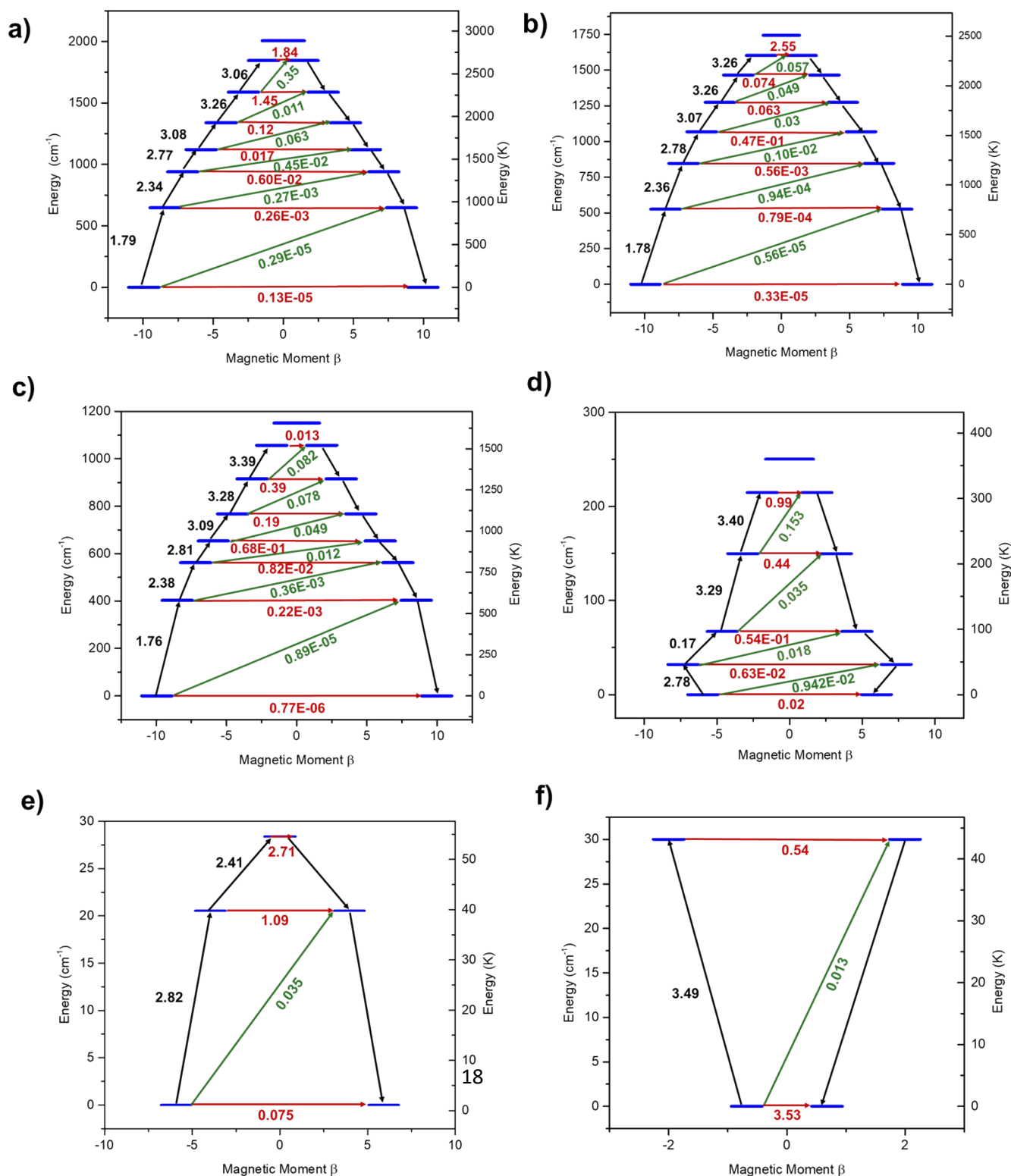


**Figure S11:** AILFT-NEVPT2 splitting pattern of 4*f* orbitals in complexes **1Q-6Q**.**Table S9:** SINGLE\_ANISO computed the properties of the two lowest Kramers' doublets of the  ${}^6\text{H}_{15/2}$  multiplet for  $[\text{Dy}(\text{CH}_3)_2]^+$  and  $[\text{Dy}(\text{CH}_3)_4]^-$ .

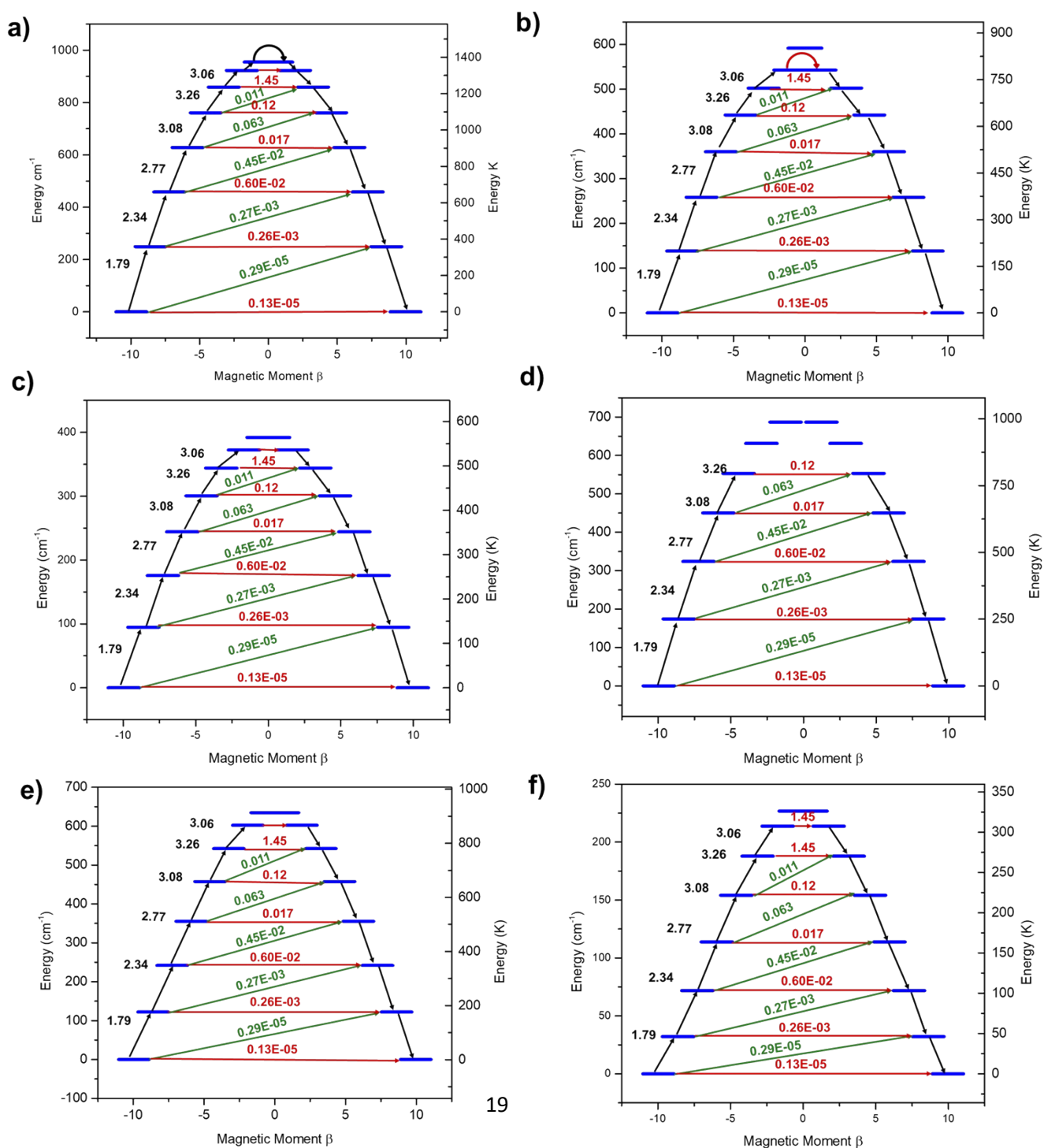
Complex	KD	E / $\text{cm}^{-1}$	$g_{xx}$	$g_{yy}$	$g_{zz}$	$\theta$	$K_{\text{QTM}}$	$m_J$	$U_{\text{cal}}/\text{cm}^{-1}$
$[\text{Dy}(\text{CH}_3)_2]^+$	KD1	0.0	0.0000	0.0000	19.9127	-	0.173E-08	$ \pm 15/2\rangle$	3022.6
	KD2	824.4	0.0000	0.0000	16.7536	0.0	0.16E-05	$ \pm 13/2\rangle$	
$[\text{Dy}(\text{CH}_3)_4]^-$	KD1	0.0	12.214	8.646	1.131	-	0.35E+01	$ \pm 1/2\rangle$	63.5
	KD2	63.5	9.381	6.084	2.258	0.0	0.26E+01	$ \pm 3/2\rangle$	



**Figure S12:** AILFT-NEVPT2 computed splitting pattern of 4*f* orbitals in [Dy(CH<sub>3</sub>)<sub>2</sub>]<sup>+</sup>, [Dy(C<sub>4</sub>(SiH<sub>3</sub>)<sub>4</sub>)<sub>2</sub>]<sup>-</sup> (**1**), [Dy(C<sub>5</sub>(CH<sub>3</sub>)<sub>5</sub>)<sub>2</sub>]<sup>+</sup> (**2**), [Dy(C<sub>8</sub>H<sub>8</sub>)<sub>2</sub>]<sup>-</sup> (**5**) and [Dy(CH<sub>3</sub>)<sub>4</sub>]<sup>-</sup> along with the *g* values for ground state KDs.



**Figure S13:** *Ab-Initio* computed blockade barrier for magnetic relaxation for complexes **1-6**. The blue line indicates the KDs as a function of magnetic moments. The black, red, and green lines represent thermal, QTM, and possible Orbach relaxations.



**Figure S14:** *Ab-Initio* computed blockade barrier for magnetic relaxation for  $1Q-6Q$ . The blue line indicates the KDs as a function of magnetic moments. The black, red, and green lines represent thermal, QTM, and possible Orbach relaxations.

**Table S10:** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[Dy(C_4(SiH_3)_4)_2]^-$  complex. All the values are reported herein  $cm^{-1}$ .

1						
Spin free States						
CASSCF						
0.0	34532.6	44053.5	55693.1	64401.5	86308.5	101789.6
1065.7	34533.5	44088.2	55712.3	66594.9	86357.7	101791.4
1067.0	34645.8	44088.2	55723.1	66766.3	86372.3	101814.7
1128.3	34645.9	44238.9	55760.0	67390.3	86801.4	102689.6
1132.8	34786.6	44239.4	55780.4	67463.8	86805.0	102785.1
1474.1	34885.7	44271.3	55841.4	67515.5	86842.7	102825.0
1549.9	34901.7	44271.3	55848.7	67676.6	86908.5	102980.8
1862.2	34912.4	44691.9	55974.3	67703.5	86967.7	103008.6
1950.0	34920.0	44725.2	55977.0	67927.7	87083.3	103177.8
2031.1	35030.8	44789.1	56135.4	67929.8	87087.8	103181.1
8350.3	35042.0	44929.6	56135.7	68254.2	87335.8	103335.2
8402.0	37071.2	44967.9	56196.8	68254.7	87346.4	103335.3
8542.3	37071.3	45056.9	56287.1	68651.1	87428.8	103804.9
8583.5	37182.9	45057.1	56287.2	68828.2	87460.5	103804.9
8614.1	37183.1	45062.8	56364.0	68828.2	87505.9	105046.2
8837.6	37346.9	45095.5	56364.2	69731.0	87715.2	105351.9
8838.5	37369.4	45157.9	56409.8	69731.3	87715.8	105457.0
33852.6	37381.9	45163.7	56442.8	69745.4	87722.6	105893.7
35810.4	37405.5	45280.2	56449.0	69745.4	87729.6	105941.3
35985.1	37405.5	45293.8	56452.6	69759.4	88284.5	106303.0
24616.7	37441.6	45335.1	56588.4	69760.0	88284.9	106380.1
24616.7	37442.1	45335.6	56588.7	69877.7	88624.0	106437.0
24952.5	37472.8	45340.9	56595.3	69877.7	88624.0	106505.2
24954.1	37525.2	45341.0	56596.4	69880.3	89050.7	106507.0
25009.5	37644.3	45384.7	56652.7	69886.4	89050.8	106551.7
25048.0	37645.1	45385.3	56658.8	69950.1	89230.6	106552.0
25077.8	37654.5	45438.7	56719.6	69950.1	89230.6	106561.1
25114.6	37658.5	45644.8	56719.8	70030.2	89399.0	106578.3
25127.2	37732.7	45645.9	56772.0	70034.3	89399.0	107494.8
25152.4	37765.3	45645.9	56772.0	70074.0	93476.6	107497.5
25154.3	37769.7	45733.1	56885.9	71037.7	93477.2	108327.4
25299.0	37842.3	46212.8	56905.8	71095.2	93676.9	108328.2
25301.7	37875.6	46212.8	56906.3	71188.3	93711.2	110011.0
25306.9	37953.9	46473.8	56912.7	71996.1	93890.5	110012.4
25309.5	38096.8	46548.3	56914.6	72044.7	93976.2	110794.2
25642.7	38345.5	46751.6	56922.0	73337.6	93981.8	110815.4
25645.0	38407.5	47508.3	56930.4	73386.6	95212.0	111010.9
25665.8	38411.9	47551.7	56943.6	73568.3	95212.1	111080.6
25667.6	38550.7	47598.1	56952.9	73644.1	95514.8	111217.7
25673.9	38561.0	47676.4	56953.2	73732.5	95532.0	114011.9
25675.8	38773.0	47679.2	56978.8	73817.5	95535.1	115398.2
25689.0	38773.0	47745.8	56982.0	73834.1	95537.3	115569.6
25699.5	38792.1	47746.9	57021.4	73872.8	95594.9	119563.3
25711.6	38792.2	47747.0	57075.0	76790.3	95595.7	119589.9
25717.9	39064.7	47758.9	57077.0	76795.4	95697.9	119691.6
25748.4	39079.7	47839.8	57078.0	76802.3	95732.7	119823.2
25752.4	39100.3	47867.3	57088.8	76819.2	95870.2	119861.1
25759.6	39132.2	47915.2	57088.9	76839.3	95871.7	120138.3
25773.7	39152.0	47983.3	57597.8	76857.5	95884.2	120143.0
25777.9	39188.7	48001.6	57603.4	76858.9	96087.4	120495.1

## Supporting Information

25789.5	39246.2	48140.8	57653.5	77215.4	96104.4	120495.5
25797.7	39264.3	48143.9	57653.5	77215.9	96144.8	120889.6
25799.3	39268.6	48203.3	58058.8	77293.3	96189.2	120889.6
25856.0	39273.7	48203.6	58067.2	77293.4	96220.3	121393.2
25863.1	39277.4	48380.1	58094.6	77450.3	96253.6	121393.2
25974.4	41743.3	48380.4	58098.4	77965.9	96277.0	122320.5
25974.4	41836.8	48597.7	58277.5	77967.1	96288.2	122320.5
26277.1	41842.4	48598.0	58393.2	78660.4	96348.1	122346.0
26278.5	41842.6	48714.5	58417.3	78676.2	96582.8	122381.8
26509.8	41850.1	48714.5	58465.0	79379.2	96583.1	123629.4
26509.8	41894.1	49112.4	58465.5	79416.9	96612.6	123723.5
26897.4	41899.5	49112.4	59053.2	79452.1	96642.8	123965.9
26900.5	41920.0	49551.1	59053.9	79452.1	96645.9	131562.1
26930.8	41964.5	49551.1	59070.7	79476.0	96759.7	131922.7
26931.2	41975.6	50042.4	59107.6	79783.5	96834.5	132055.5
27119.4	42044.4	50042.4	59167.0	79783.6	96977.3	132667.1
27172.7	42045.8	50282.1	59360.0	79914.8	97004.7	132698.7
27192.7	42289.2	50282.1	59536.0	79918.9	97005.6	134034.5
29501.5	42289.2	51730.1	59536.9	79928.0	97018.7	134035.8
29501.5	42516.1	51751.7	59673.9	79975.4	97104.0	139226.1
29837.0	42561.7	51754.9	59785.8	79980.1	97104.0	139226.3
29837.7	42561.7	51799.4	60185.0	79980.2	97213.6	139739.8
29855.2	42571.7	51807.8	60185.0	79997.2	97306.8	139739.9
29858.1	42704.9	51844.7	60273.5	80001.5	97346.7	140578.7
29875.8	42717.2	51894.4	60303.2	80009.9	97422.6	140585.9
29876.8	42736.1	51903.1	60305.8	80121.0	97480.4	140895.9
29929.5	42736.1	51911.6	60320.7	80123.9	97484.5	140968.3
29929.5	42778.3	51970.2	60682.8	80155.7	97487.0	140968.9
30070.2	42778.3	51971.2	60686.4	80162.5	97493.0	142256.0
30087.2	43122.4	52144.4	60754.7	80174.0	97583.4	142401.9
30262.7	43143.5	52144.5	60830.5	80186.7	97710.5	143839.3
30279.0	43209.9	52301.5	60832.6	80186.9	97721.7	145756.0
30323.2	43218.0	52372.5	61245.8	80293.9	97723.4	145756.1
31388.6	43451.5	52395.1	61265.4	80296.0	98130.1	146560.7
31424.3	43476.6	52523.3	61723.1	80506.6	98130.2	146563.1
31428.7	43489.7	52556.1	61723.2	80506.6	98292.0	146946.4
31468.3	43491.1	52665.4	61785.0	80681.5	98424.1	146975.1
31468.3	43510.1	52669.0	61809.3	80681.6	98750.9	146986.0
31522.7	43514.0	52743.1	61859.6	80812.9	98750.9	147038.6
31540.6	43547.4	52743.3	61947.6	80812.9	99330.9	147040.9
31636.2	43547.4	52753.9	61959.8	81013.7	99330.9	147047.3
31636.8	43549.1	52754.0	62195.7	81013.7	99378.2	147054.9
31650.5	43552.8	52775.8	62196.5	81388.0	99378.2	173538.9
31651.2	43555.8	52776.0	62556.2	81388.0	99465.1	173676.5
31715.1	43580.5	53238.5	62556.3	84290.2	99470.5	173967.8
31715.9	43620.4	53238.5	63022.9	84778.9	100943.0	175049.2
31744.8	43637.3	54707.1	63022.9	84896.0	100943.0	175111.5
31745.0	43668.0	55001.5	63315.3	85849.6	101154.7	177865.7
31759.5	43669.1	55008.8	63315.3	85905.7	101154.8	178183.2
31759.6	43704.7	55134.9	64131.6	86027.9	101486.5	178188.0
33114.6	43759.2	55140.5	64138.4	86048.4	101486.6	178237.4
33139.6	43761.1	55469.2	64192.8	86204.8	101736.6	178367.9
33222.9	43808.7	55490.0	64227.5	86274.4	101742.7	179895.4
33273.2	43865.2	55560.4	64234.7	86276.7	101778.2	180005.6
33380.1	44053.5	55560.5	64257.8	86306.2	101780.4	

**Table S13:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_4(\text{SiH}_3)_2)_2]^-$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

1						
Spin free States						
NEVPT2						
0.1	33248.2	43969.3	52843.2	59885.9	79879.6	94408.9
1031.7	33249.2	43978.3	52857.7	60422.7	79939.8	94425.3
1044.8	33463.8	43996.5	52943.9	60634.5	79956.4	94430.7
1213.8	33464.1	44010.0	52951.6	62643.2	79973.6	94436.5
1217.7	33563.6	44014.5	52965.9	63522.1	79976.2	94709.9
1421.0	33719.6	44105.2	53029.9	63628.1	80048.3	94858.2
1604.6	33749.6	44335.4	53154.3	63684.4	80052.7	94897.5
2008.1	33781.7	44335.5	53309.5	63874.9	80120.5	94898.6
2097.3	33786.0	44574.6	53311.0	63924.7	80315.6	95036.5
2190.5	33934.6	44574.6	53402.9	64148.0	80328.5	95063.0
7117.8	33970.2	44641.1	53416.2	64152.2	81000.4	95121.8
7188.5	34672.3	44641.1	53717.6	64449.7	81002.1	95285.6
7332.7	34893.5	44651.8	53915.2	64451.2	81032.4	95291.8
7427.8	35056.0	44654.4	53921.3	65086.1	81052.5	95442.5

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7548.1	35150.8	44803.1	54002.1	65086.2	81099.5	95443.4
7742.6	35165.9	44803.1	54015.6	65196.8	81205.5	95517.2
7745.7	35174.6	44906.9	54050.1	65263.8	81217.9	95582.1
28478.6	35193.1	44982.6	54070.2	65935.9	81231.3	96078.6
30539.7	35713.6	45021.1	54072.7	66059.5	81280.8	96078.6
30753.7	36108.4	45025.0	54082.3	66119.1	81923.7	96204.5
23974.6	36112.4	45028.5	54347.3	66552.7	81923.8	96205.1
24284.8	36189.2	45059.6	54347.7	66553.4	81941.9	96231.0
24503.4	36207.1	45086.7	54397.5	66575.7	81942.4	96236.4
24571.4	37677.9	45109.7	54404.7	66576.1	82196.3	96874.3
24606.4	37677.9	45120.8	54470.7	66620.4	82196.3	97012.7
24627.5	37735.2	45150.3	54477.9	66620.4	83018.5	97044.3
24648.9	37735.9	45236.5	54488.4	66734.8	83018.5	97054.1
24695.2	37993.0	45262.6	54488.5	66769.7	83587.7	97101.6
24868.5	37994.5	45268.5	54488.5	66795.6	83587.7	98141.8
24985.2	38002.6	45414.3	54488.7	66795.6	85397.3	98147.6
24991.9	38090.0	45425.9	54506.5	66800.2	85398.1	99000.3
25146.1	38092.7	45431.1	54552.0	66931.7	85522.5	99001.2
25177.5	38157.7	45660.2	54552.5	66931.7	85585.0	100615.1
25304.0	38218.4	45667.7	54570.9	66948.0	85696.4	100617.2
25314.3	38275.6	45849.0	54582.7	66955.1	85887.5	101517.5
25366.8	38275.7	45849.0	54635.0	66962.3	85893.2	101557.2
25422.1	38432.8	45863.9	54637.7	67093.3	87625.6	101720.3
25425.3	38434.6	45864.2	54677.1	67263.3	87694.7	101804.8
25442.5	38437.0	45954.2	54678.2	67474.6	87694.8	101975.9
25446.7	38437.3	45982.6	54701.5	67566.8	87835.4	103908.3
25467.5	38453.8	45982.6	54710.5	67673.5	88195.6	105305.8
25568.7	38458.9	46154.9	54729.7	68149.8	88237.9	105520.7
25577.9	38458.9	46154.9	54744.2	68181.4	88245.2	110446.2
25660.5	38461.8	46211.3	54770.1	70268.1	88365.4	110475.8
25722.1	38604.6	46211.6	55006.3	70402.3	88470.5	110586.3
25739.0	38705.8	49420.9	55041.3	70420.6	88584.4	110723.7
25759.6	38780.2	50071.3	55046.9	70452.2	88595.3	110790.2
25878.0	38802.1	50080.6	55052.9	70461.7	88598.1	111066.7
25885.3	38809.5	50091.7	55056.7	70464.2	88614.3	111091.4
26025.6	38818.4	50123.9	55108.3	70468.7	88713.0	111101.2
26251.9	38836.2	50131.1	55137.2	70494.9	88756.5	111116.3
26327.6	38916.6	50137.3	55204.4	70628.2	88792.0	111471.7
26404.0	38923.8	50199.5	55499.3	70628.3	88890.6	111472.5
26465.5	38939.8	50215.5	55499.4	71033.4	88919.2	111853.7
26469.8	38946.7	50249.9	55655.2	71033.5	89111.0	111853.7
26667.3	38969.4	50262.1	55659.7	71036.1	89146.6	112340.0
27034.0	39608.3	50278.5	55662.9	71043.5	89364.2	112340.1
27085.3	39678.1	50358.6	55808.6	71329.5	89414.5	112505.8
27311.4	39986.2	50367.0	55809.3	71342.1	89440.1	112616.2
27369.4	39997.1	50382.4	55850.1	72176.7	89448.0	112884.2
27525.5	40454.1	50385.8	56034.7	72306.1	89453.3	113451.1
27608.1	40558.4	50400.1	56059.9	72374.3	89552.5	113451.1
27622.1	40580.0	50429.8	56096.4	74859.2	89574.6	118598.6
27973.3	40688.3	50474.1	56112.3	75136.5	89728.5	119066.0
27973.3	40791.3	50478.3	56154.8	75136.5	89753.6	119232.0
28714.9	40906.3	50529.7	56170.5	75402.7	89761.2	119741.2
28714.9	41302.7	50538.5	56266.8	75544.9	89817.1	119803.0
29670.3	41384.7	50583.1	56303.4	75568.5	89831.6	121284.4
29670.3	41561.8	50583.5	56303.5	75569.4	89844.4	121287.8
30031.8	41891.2	50790.7	56664.8	75716.0	89949.5	127085.4
30034.0	41891.8	50791.8	56674.3	75733.4	90040.8	127086.7
30035.9	42248.4	51190.5	56891.3	75795.7	90150.6	127552.4
30049.6	42462.3	51190.5	57011.0	75799.9	90223.8	127552.7
30117.7	42597.8	51366.7	57271.1	75830.0	90262.2	128608.7
30126.9	42923.6	51459.7	57271.4	76139.7	90289.3	128621.1
30294.3	43009.3	51485.7	57595.5	76169.7	90301.0	128901.0
30295.4	43021.9	51657.9	57615.6	76204.6	90320.3	128944.6
30310.8	43044.7	51657.9	57641.4	76319.8	90332.5	128987.3
30310.8	43089.6	51671.5	57648.4	76399.5	90377.5	129043.8
30448.4	43089.7	51692.2	57648.8	76546.1	90975.6	129095.9
30481.8	43126.2	51807.4	57680.9	76580.7	90975.7	130743.7
30525.5	43232.9	51807.8	57682.6	76581.2	91057.3	132871.4
30546.1	43282.4	51834.5	57886.1	76589.6	91089.9	132871.6
30596.5	43328.5	51838.0	57927.7	76768.6	91186.7	133825.7
30619.4	43354.7	51851.1	58137.6	77017.3	91194.0	133832.0
30736.1	43457.3	51851.4	58144.8	77074.4	91432.6	134263.5
30817.0	43458.6	51894.5	58196.8	77100.2	91435.4	134287.1
32185.0	43460.8	51894.7	58216.8	77248.7	91896.4	134335.8
32233.3	43484.9	51943.6	58300.8	77266.9	91896.6	134398.5
32237.4	43503.0	51943.8	58362.8	77491.5	92714.5	134402.9
	43530.6	52032.9	58387.3	77491.7	92714.5	134433.8

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32352.5	43531.0	52151.8	58647.5	77729.3	93132.2	134437.4
32397.3	43535.2	52151.9	58649.9	77729.5	93132.2	156513.3
32397.3	43546.5	52453.7	59065.6	77864.6	93380.9	156590.3
32410.1	43723.3	52453.7	59065.8	77864.6	93380.9	156983.9
32535.4	43741.5	52457.2	59569.3	78030.6	93539.6	157663.4
32537.9	43743.9	52463.8	59589.8	78030.6	93539.6	157782.2
32551.2	43778.5	52497.9	59607.9	78489.2	93721.8	160261.9
32551.6	43784.6	52516.2	59608.0	78489.2	93721.9	160320.1
32617.7	43809.6	52673.8	59610.3	79238.1	94114.3	160396.5
32620.4	43810.2	52687.2	59623.9	79309.2	94114.5	160706.7
32658.3	43841.8	52725.7	59706.9	79532.3	94323.4	160931.1
32658.9	43853.6	52725.7	59736.0	79566.7	94343.4	162989.2
32679.8	43874.3	52821.8	59805.7	79838.8	94369.5	163237.2
32680.4	43969.1	52843.2	59805.7	79859.2	94406.4	

**Table S14:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_4(\text{SiH}_3)_4)_2]^-$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

1							
Spin orbit states							
CASSCF							
0.0	31177.8	40563.9	50701.8	61324.7	70842.5	90105.5	105583.3
597.8	31224.2	40817.3	50726.2	61366.2	70932.4	90280.5	105849.5
933.6	31829.9	40847.8	50763.3	61411.0	71001.2	90285.2	105867.2
1153.6	32142.3	40858.5	50777.6	61418.9	71187.9	90379.4	106096.5
1364.3	32286.3	40885.3	50784.3	61563.8	71828.9	90425.1	106325.6
1576.1	32339.2	40900.3	50801.3	61614.9	72254.5	90445.5	106334.6
1768.8	32349.4	40997.3	50852.0	61645.4	72333.4	90708.4	106397.9
1904.8	32358.1	41019.9	50879.0	61707.9	72383.9	90765.0	106430.3
3446.4	32375.9	41082.0	51087.5	61806.4	72465.3	90797.3	106661.0
3584.2	32395.9	41102.0	51112.3	61813.1	72717.5	90987.0	106716.3
4009.5	32415.3	41127.8	51194.5	61860.8	73092.4	90995.4	106791.5
4351.5	32469.5	41144.4	51215.0	61881.4	73339.0	91126.2	106924.1
4523.9	32525.3	41182.6	51276.1	61901.5	73547.9	91203.8	107052.6
4617.4	32605.1	41203.3	51300.4	61941.3	74557.1	91218.4	107746.8
4712.1	32617.6	41222.5	51334.7	61970.1	74582.5	91220.7	107858.7
5839.7	32621.5	41298.5	51339.6	61979.0	74687.9	91243.5	108075.4
5945.1	32638.0	41464.9	51421.8	62015.7	74919.3	91336.5	108198.6
6282.1	32827.8	41654.1	51531.4	62114.7	74992.4	91359.7	108227.0
6616.1	32861.9	42361.6	51535.4	62232.7	75127.9	91527.4	108283.0
6738.2	32939.4	42471.5	51552.5	62237.0	75132.9	91550.5	108290.9
6821.0	33239.6	42570.6	51566.1	62251.8	75194.6	91573.8	109293.3
7693.9	33279.2	42579.6	51652.1	62304.5	75224.1	91657.4	109541.0
7796.4	33381.2	42766.8	51676.9	62337.5	75246.9	91881.6	109883.2
8066.8	33460.2	43054.5	51746.6	62365.1	75263.4	91953.6	109983.0
8384.3	33609.9	43119.5	51772.7	62573.5	75351.0	92016.1	110043.2
8471.0	34088.4	43216.0	51862.7	62628.4	75371.7	92110.2	110156.7
9087.1	34113.3	43286.1	52032.2	62630.0	75405.0	92146.3	110218.4
9190.3	34151.6	43312.4	52070.9	62657.7	75455.2	92266.6	110286.8
9681.3	34190.3	43399.5	52178.6	62693.3	75503.9	92282.9	110413.9
9768.9	34206.7	43431.4	52292.9	62763.4	75732.1	92377.4	110455.6
10069.1	34228.8	43471.3	52306.8	62775.7	75738.7	92493.7	110501.9
10431.8	34251.3	43477.0	52326.3	62779.1	75759.9	92573.4	110700.4
10472.5	34312.6	43512.6	52354.9	62832.3	75868.5	92830.2	110833.2
10524.6	34382.2	43516.8	52436.0	62843.5	75891.2	93047.5	111073.7
10572.5	34422.5	43565.9	52440.2	62861.3	75966.8	93309.9	111375.6
10644.2	34464.4	43599.4	52512.7	63005.1	76008.5	93400.1	111487.1
10666.8	34524.1	43615.9	52585.5	63079.3	76058.9	94031.2	111611.7
10851.8	34540.4	43628.8	52607.2	63235.8	76135.8	94201.8	112217.9
10936.2	34636.7	43710.1	52773.9	63292.5	76222.3	94573.7	113251.3
11812.3	34641.4	43721.8	53026.1	63361.7	76400.4	95210.0	113307.6
11907.9	34682.1	44784.2	53028.8	63470.8	77621.1	95239.3	113339.7
11934.4	34713.9	44863.2	53254.6	63525.2	77646.5	95363.1	113426.6
11971.3	34744.9	44988.4	53357.1	63639.2	78005.3	95520.3	113689.1
12080.9	34758.8	44993.2	53373.7	63767.6	78730.6	95740.5	113909.8
13528.1	34832.5	45014.2	53749.2	63787.4	79219.2	95826.4	114251.5
13594.5	35011.5	45089.6	53928.2	63813.0	80037.3	96347.2	114520.9
13640.7	35156.0	45183.9	54175.4	63831.1	80612.8	96952.8	114587.0
13675.0	35272.4	45260.7	54577.3	63935.0	81122.6	96997.5	114701.2
14856.0	35765.2	45443.1	54693.6	63973.2	81273.2	97096.0	114931.1
14880.3	35828.1	45448.5	54813.0	64091.1	81414.6	97134.1	115117.6

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14919.3	35999.3	45587.7	54835.4	64105.1	81474.3	97614.3	115236.1
15683.5	36007.3	45593.9	54924.8	64140.1	81479.4	97629.6	115851.6
15698.3	36065.4	45617.8	54942.4	64151.1	81520.0	97687.5	116076.6
16213.9	36073.5	45651.3	54965.0	64180.8	81587.2	97763.7	116483.4
24485.6	36188.5	45713.0	54999.2	64271.8	81624.8	97946.5	116496.7
24583.6	36237.6	45719.3	55035.4	64279.4	81823.2	97981.7	116778.4
24643.9	36241.4	45817.9	55048.9	64296.1	81834.3	98520.3	117832.4
24718.6	36246.1	45819.2	55061.4	64313.3	81873.0	98688.7	117874.7
24826.3	36329.3	45845.2	55073.1	64413.4	81919.0	98709.5	118113.7
24976.6	36423.9	45930.1	55180.6	64581.7	81933.9	98887.9	118246.3
24980.9	36528.5	45988.8	55316.8	64625.9	81946.4	99459.0	118584.9
25214.8	36690.9	46044.3	55328.8	64637.4	82100.8	99961.2	120522.7
25628.0	36830.6	46127.0	55346.8	64712.3	82134.0	100024.3	121766.8
25707.3	36845.1	46326.3	55608.8	64727.8	82178.3	100060.5	125050.2
25817.3	37175.2	46673.0	55764.2	65154.3	82206.0	100144.0	125172.9
25850.3	37237.6	46698.3	55785.4	65448.3	82257.1	100332.4	125350.6
25895.5	37338.7	46780.6	55843.2	65493.3	82271.4	100624.2	125605.4
26783.1	37459.4	46790.6	55889.2	65598.2	82295.1	100660.8	125883.4
27167.7	37557.4	46858.4	55941.0	65782.4	82315.6	100765.8	125904.0
27229.3	37561.3	46864.7	55964.8	65809.0	82493.9	100781.8	126077.1
27264.7	37571.7	46960.5	55987.4	65840.6	82512.5	100796.1	126279.9
27308.3	37584.1	47010.7	56108.4	65904.0	82547.3	100809.4	126351.7
27351.6	37627.7	47029.6	56153.5	66021.4	82560.0	101238.4	126714.5
27533.5	37705.7	47045.2	56199.8	66038.7	82656.8	101439.8	126989.2
27556.3	37810.2	47061.6	56239.4	66192.0	82816.7	101660.0	127115.0
27602.2	37875.4	47075.2	56283.2	66301.7	83178.5	101731.5	127563.4
27608.6	37890.3	47121.3	56404.1	66392.1	83554.5	101844.6	127975.0
27622.2	38023.1	47145.9	56441.5	66483.4	83691.3	101850.3	128166.3
27646.6	38026.5	47274.1	56624.0	66654.5	84461.5	101882.7	129025.6
27674.3	38136.6	47293.9	56816.9	66735.7	84578.6	101904.2	129147.5
27679.4	38140.9	47304.3	56905.8	66748.2	84744.5	101928.5	129325.4
27725.7	38180.7	47317.5	57230.6	66782.3	84749.3	101993.9	129391.8
27971.0	38305.0	47509.0	57600.0	67124.4	84794.0	102006.8	130809.8
28096.9	38314.7	47606.3	58010.8	67146.6	84838.3	102120.4	136281.6
28234.5	38340.9	47786.9	58045.0	67193.6	84891.7	102176.2	136839.6
28464.8	38362.3	47875.3	58081.2	67256.8	84946.7	102391.7	138044.1
28563.5	38392.9	47976.3	58122.4	67329.7	84977.5	102522.4	139150.5
28590.1	38425.3	47995.3	58162.6	67342.3	85039.4	102746.7	139581.5
28689.2	38529.1	48079.6	58215.6	67398.0	85178.5	102818.3	140390.3
28732.3	38563.7	48116.9	58257.7	67442.9	85238.8	103171.8	141458.6
28751.7	38683.0	48173.6	58543.4	67474.5	85337.5	103281.6	143699.3
28768.8	38694.7	48174.2	59171.3	67547.7	85378.0	103392.1	144026.2
28825.9	38855.8	48300.8	59190.9	67633.9	85402.1	103395.2	144471.9
28839.8	38984.6	48323.9	59242.1	67649.3	85465.1	103405.8	144959.7
28857.9	39138.0	48503.6	59300.0	67750.0	85508.0	103432.1	145150.4
28931.9	39146.4	48750.5	59433.5	67751.2	85552.6	103465.4	146447.2
28960.0	39162.7	49218.8	59509.1	67784.1	85579.3	103667.1	147139.2
28997.8	39181.9	49267.0	59554.7	68011.2	85604.8	103708.8	147278.2
29061.4	39190.5	49276.5	59593.1	68022.3	85626.7	103771.2	147778.2
29077.0	39343.9	49312.3	59731.2	68053.6	85638.6	103822.0	147931.4
29130.9	39419.9	49353.9	59800.3	68087.5	85676.2	103841.3	148061.0
29154.8	39514.8	49391.3	59811.2	68108.7	85692.5	103859.9	150033.4
29203.3	39558.7	49486.8	60065.7	68127.5	85742.8	104069.6	150331.4
29288.9	39560.6	49496.9	60123.7	68389.4	85777.2	104122.7	150718.8
29304.0	39566.4	49620.1	60219.3	68398.9	85780.0	104243.2	151102.2
29443.4	39572.8	49709.0	60273.5	68425.0	85862.6	104272.9	151169.4
29450.5	39582.3	49722.5	60303.5	68452.9	85930.8	104308.9	151214.3
29534.9	39596.7	49775.5	60356.0	68576.6	85946.6	104457.1	151227.3
29577.5	39615.7	49808.5	60501.5	68746.0	86021.3	104529.7	153319.6
29603.0	39679.6	49828.0	60572.8	68810.2	86207.8	104561.6	154219.0
29631.8	39692.1	49850.5	60630.3	69117.4	86239.6	104591.6	154716.7
29686.9	39767.2	49996.6	60636.0	69307.0	86395.0	104776.8	154844.1
29702.0	39808.4	50052.5	60691.7	69469.4	86553.1	104801.3	154883.4
29718.8	39850.2	50145.7	60765.9	69485.8	86670.5	104814.5	177155.9
30254.0	39934.9	50275.8	60778.2	69549.8	86811.8	104861.5	177967.1
30428.8	40020.1	50366.1	60789.6	69833.4	86814.1	104873.2	179590.1
30675.0	40104.0	50375.4	60824.4	69906.4	86927.4	105059.0	181180.2
30782.3	40162.2	50395.1	60912.9	69983.8	86965.9	105135.1	182355.9
30810.9	40167.8	50477.7	60935.3	70000.7	87076.0	105274.5	182789.2
30846.9	40221.8	50487.8	60944.5	70148.6	87197.7	105304.3	182920.0
30882.2	40235.0	50526.1	60969.5	70222.7	88009.7	105315.1	182949.5
30935.9	40348.1	50545.0	61002.3	70277.1	89396.1	105334.5	184002.7
31055.2	40393.2	50554.7	61104.9	70314.4	89539.8	105367.1	185585.3
31083.3	40421.7	50582.7	61136.6	70570.6	89549.4	105381.7	186068.3
31112.6	40534.6	50635.0	61216.4	70579.5	89783.2	105434.6	187620.7
31176.7							



**Table S15:** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_4(\text{SiH}_3)_4)_2]^-$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

1							
Spin orbit states							
NEVPT2							
0.0	31450.3	40097.1	49671.8	58945.6	67120.6	83611.5	98533.0
662.0	31461.5	40130.0	49979.9	58971.3	67188.0	83647.1	98628.6
958.0	31602.4	40152.5	50195.0	59057.2	67264.0	83789.7	98830.8
1142.2	31673.1	40222.2	50290.7	59208.3	67453.8	83849.7	98864.4
1360.9	31763.3	40229.7	50317.5	59288.5	67518.5	83862.9	98929.1
1609.7	31797.8	40270.8	50447.0	59301.8	67797.1	83932.8	99043.6
1867.5	31893.2	40282.7	50503.9	59333.6	67869.3	84009.0	99113.2
2027.4	31950.6	40405.2	50635.4	59351.4	68019.4	84094.4	99123.5
3518.4	32048.9	40410.2	50660.8	59380.6	68407.8	84108.4	99268.2
3543.6	32080.8	40416.1	50719.9	59426.1	68505.6	84113.8	99276.2
4045.9	32089.7	40535.1	50771.1	59501.3	68595.8	84149.8	99317.3
4427.6	32188.0	40593.9	50815.8	59605.9	68895.1	84204.6	99584.8
4588.3	32231.5	40598.1	50868.9	59609.7	68969.3	84207.1	99757.5
4665.7	32289.6	40622.8	50931.6	59687.9	69006.2	84393.6	99935.6
4785.5	32297.4	40638.6	50939.6	59780.7	69287.9	84456.9	100255.0
5866.7	32319.3	40688.0	50996.6	59871.6	69329.7	84506.6	100384.0
5907.7	32368.1	40719.7	51016.8	59879.9	69356.3	84610.4	100394.7
6294.4	32423.3	40815.1	51436.8	59889.8	69631.9	84693.8	100620.1
6665.2	32569.2	40819.7	51970.8	59932.0	69817.5	84741.4	100641.7
6758.0	32625.7	40975.4	52308.3	59951.9	70040.1	84802.9	100694.2
6845.9	32792.1	41140.9	52326.6	60009.6	70690.0	84838.3	100733.1
7686.2	32827.2	41180.3	52392.0	60028.7	70957.5	84858.4	100778.4
7787.2	32940.9	41226.7	52436.1	60057.1	71198.9	84894.7	100804.0
8054.0	33061.6	41299.7	52504.7	60074.5	71386.4	85044.5	100845.0
8363.1	33201.7	41303.3	52584.7	60104.7	71410.4	85074.4	100877.8
8507.1	33219.4	41317.3	52621.7	60107.1	71436.7	85355.6	101214.2
9031.6	33273.5	41356.2	52673.7	60114.5	71868.3	85392.4	101255.6
9183.5	33298.8	41485.1	52697.8	60290.2	71947.6	85464.3	101340.5
9215.9	33329.5	41499.0	52735.6	60354.3	71953.7	85545.4	101363.3
9283.3	33396.1	41539.1	52751.8	60372.2	72008.9	85673.8	101427.9
9316.6	33558.3	41592.1	52803.5	60380.6	72087.9	85816.8	101632.5
9405.6	33599.5	41614.5	52942.2	60407.4	72145.1	85977.4	101662.1
9499.6	33739.7	41705.9	52952.8	60413.1	72193.7	86036.3	101702.5
9680.6	33823.5	42044.9	52956.1	60450.9	72251.4	86190.4	101746.4
9736.5	33868.4	42343.5	52971.7	60469.9	72300.3	86233.4	101824.8
9793.9	33935.3	42541.9	52979.3	60510.9	72359.2	86729.9	101968.1
10060.2	34071.1	42792.8	53001.0	60557.7	72580.0	86767.0	102345.6
10547.7	34204.0	43074.7	53071.3	60619.2	72633.4	86937.1	103034.3
10659.0	34234.1	44040.6	53086.9	60636.7	72661.7	87360.2	103663.5
10804.1	34388.4	44047.2	53214.4	60661.0	72670.8	88087.0	103724.6
10877.8	34553.7	44059.1	53274.2	60678.0	72724.0	88586.4	103752.1
10898.0	34599.2	44162.2	53371.4	60731.8	72814.2	88690.4	103911.3
10968.7	34626.9	44269.5	53526.7	60830.2	72869.5	88804.6	104753.1
10993.5	34702.6	44317.6	53541.9	61011.1	72879.9	88916.7	104850.4
12369.4	34736.3	44359.8	53597.0	61050.9	72891.3	89191.3	104986.4
12473.5	34748.0	44499.2	53612.6	61068.2	73790.1	89215.3	105186.2
12503.8	34780.9	44511.4	53645.1	61106.6	74051.9	89546.1	105247.5
12579.6	34850.8	44541.7	53660.0	61148.6	74929.9	89692.4	105508.8
13668.7	34891.3	44559.3	53686.4	61158.5	75091.2	89785.7	105705.6
13706.9	34928.0	44637.9	53705.0	61195.0	75142.0	89953.4	105823.6
13769.4	35108.5	44705.3	53889.9	61241.2	75171.2	90566.2	106189.3
14463.1	35176.6	44768.7	53999.3	61289.7	75287.5	90639.7	106331.6
14495.5	35203.1	44852.3	54014.1	61385.8	75318.1	90664.9	106562.6
14998.9	35266.0	45105.0	54057.0	61409.6	75330.5	90789.1	106914.4
23814.7	35302.1	45158.9	54424.1	61438.8	75350.2	90929.8	107114.7
23862.8	35386.7	45197.0	54634.0	61461.3	75443.0	91143.4	107415.5
24123.6	35497.2	45512.1	54734.4	61625.6	75448.2	91156.5	108301.3
24271.3	35667.7	45724.5	54817.9	61675.4	75457.6	91215.5	108501.2
24355.4	35917.5	45759.5	54852.8	61696.6	75534.8	91628.9	108636.4
24837.1	35982.1	45804.9	54924.5	61725.0	75578.9	92178.8	108685.4
24846.6	36068.5	45936.9	54936.8	61764.8	75633.9	92373.5	108876.9
24947.3	36074.6	46271.2	54966.5	62213.9	75793.7	92584.2	110417.0
25583.1	36093.0	46381.5	55014.9	62262.9	75915.1	92836.6	111619.2
25617.6	36269.0	46522.3	55132.4	62310.2	75974.8	93059.7	116016.4
25699.8	36327.3	46580.0	55152.2	62383.2	75981.6	93066.6	116154.2
25770.7	36400.0	46584.7	55275.6	62454.3	75983.9	93185.2	116363.2
25791.0	36450.5	46647.7	55346.1	62478.1	76008.0	93210.4	116649.4
25865.4	36481.5	46651.7	55480.0	62500.6	76010.0	93264.1	116813.2
25967.0	36508.9	46686.3	55527.4	62544.5	76048.2	93293.0	116830.7
26012.0	36513.2	46721.8	55609.1	62588.2	76124.8	93399.2	116950.0
26124.9	36545.8	46759.8	55650.5	62669.1	76156.1	93476.0	117023.2

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26325.9	36698.3	46781.0	55709.4	62711.9	76315.4	93631.4	117312.9
26473.7	36704.2	46808.3	55748.5	62734.9	76341.5	93678.0	117330.5
27648.0	36741.9	46840.9	55785.0	62981.9	76491.4	93755.8	117724.0
27712.5	36800.7	46899.1	55825.4	63022.1	76543.1	93777.4	117881.4
27771.7	36837.2	46923.9	55876.0	63071.7	76571.1	93840.3	118132.1
27799.3	36863.9	46975.7	55926.2	63137.8	76619.3	93888.0	118152.0
27829.3	36940.7	46985.5	55932.8	63251.3	76763.3	94377.4	118249.7
27994.9	36978.0	46996.8	56018.0	63439.9	77927.4	94424.4	118329.4
28153.0	37014.8	47020.1	56053.0	63479.9	78457.7	94453.1	118572.2
28247.4	37219.1	47025.0	56139.0	63494.8	78536.9	94516.0	119204.8
28265.3	37237.5	47088.1	56306.1	63580.8	78806.8	94689.2	119752.0
28358.5	37270.8	47168.6	56453.4	63659.3	78913.4	94837.9	120323.9
28466.6	37322.0	47259.1	56479.2	63854.5	78984.6	95047.5	123523.2
28515.7	37733.4	47417.6	56481.1	63949.0	79056.0	95061.6	124066.0
28719.0	37848.5	47454.3	56635.5	64001.9	79091.6	95111.0	125319.3
28791.7	37861.9	47475.4	56789.9	64063.0	79117.7	95121.7	126398.8
28823.1	37887.2	47508.5	56833.1	64092.3	79412.7	95163.2	126865.2
28922.4	37915.4	47550.2	56923.0	64119.0	79735.1	95250.9	127675.5
29047.8	37965.2	47605.4	56945.8	64154.2	79869.5	95273.7	128850.7
29086.4	38062.4	47674.6	56978.3	64165.1	80030.2	95288.3	131542.6
29131.3	38069.9	47690.5	57052.9	64182.8	80140.5	95441.8	131855.4
29184.2	38081.4	47707.8	57069.7	64304.2	80198.6	95613.9	132397.8
29310.4	38089.8	47713.1	57118.9	64384.7	80379.5	95677.9	133000.8
29357.2	38126.8	47758.9	57142.5	64452.3	80590.7	95695.2	133184.0
29421.6	38274.7	47789.3	57176.7	64496.9	80678.4	95786.1	133983.6
29477.7	38286.6	47819.7	57209.2	64550.9	80818.9	95880.9	134321.9
29519.1	38302.5	47824.3	57216.1	64583.3	80917.2	95997.4	134761.4
29572.4	38333.9	47830.0	57229.5	64588.9	80933.8	96250.6	135089.6
29643.8	38361.2	47912.1	57236.4	64606.1	80941.1	96495.3	135851.9
29670.7	38444.2	47925.1	57255.8	64772.3	80991.1	96613.3	136091.2
29696.3	38487.9	47961.9	57329.8	64826.6	81082.6	96680.2	137179.2
29756.2	38535.6	47979.7	57388.8	64857.3	81095.0	96702.8	137220.1
29780.3	38562.5	47985.2	57404.6	64918.7	81181.1	96772.2	138003.6
29839.8	38564.8	48033.6	57492.1	65113.9	81217.4	96779.1	138454.9
29975.1	38681.2	48051.2	57529.8	65154.4	81276.2	96937.0	138527.1
30045.6	38702.6	48079.9	57540.8	65189.1	81373.4	97034.7	138601.1
30059.6	38725.4	48134.6	57583.0	65200.7	81487.9	97089.2	138633.3
30404.2	38768.4	48170.6	57725.0	65309.7	81626.3	97152.8	140519.6
30550.4	38871.7	48293.5	57818.3	65498.7	81660.2	97182.9	141551.8
30576.3	38894.7	48336.8	57855.7	65507.8	82022.8	97202.2	142107.8
30675.0	38937.7	48466.7	57933.8	65554.7	82068.5	97273.4	142264.1
30759.7	38996.1	48601.2	58103.3	65621.7	82078.8	97296.9	142305.2
30804.1	39164.1	48611.7	58203.6	65667.6	82091.5	97323.2	160126.0
30886.2	39251.2	48658.2	58383.8	65946.5	82266.9	97438.4	160857.4
30913.5	39258.5	48748.7	58417.8	65981.0	82286.0	97463.7	162220.9
31016.0	39281.3	49029.5	58476.5	65988.7	82472.8	97491.1	164109.6
31109.2	39361.6	49033.5	58561.1	66100.8	82630.0	97516.2	165055.5
31125.1	39368.4	49104.7	58705.4	66173.9	82646.6	97579.5	165222.2
31142.4	39536.5	49194.1	58752.8	66222.1	82680.8	97727.0	165326.2
31194.3	39965.1	49244.4	58834.4	66237.8	82926.0	97745.6	165557.4
31235.5	39996.8	49315.6	58839.9	66282.7	82946.2	97861.7	167164.0
31240.9	40039.3	49415.4	58880.7	66436.4	83048.7	97889.7	167891.5
31289.6	40060.8	49457.4	58928.9	66602.3	83162.2	98019.2	168595.4
31342.9	40065.8	49501.2	58937.6	66691.1	83411.5	98400.0	170786.3
31410.1							

**Table S16:** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_5\text{H}_5)_2]^+$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

2						
Spin free states						
CASSCF						
0.1	37261.5	47391.8	57776.6	79875.8	98435.4	
845.1	37286.6	47439.9	57842.1	79878.8	98435.4	
846.0	37286.8	47442.0	57845.6	79884.7	98881.1	
1083.7	37336.2	47618.5	58073.1	79901.6	98881.1	
1086.6	37342.2	47623.9	58097.5	79904.9	99010.0	
1395.3	37346.8	47684.7	58157.1	79912.2	99010.0	
1398.6	37455.1	47692.4	58197.4	79923.0	99253.0	
1584.8	37456.5	47712.7	58198.1	79971.0	99253.5	
1713.5	37459.7	47745.2	58907.2	79976.6	100822.0	
1739.9	37461.1	47753.3	58984.9	80026.1	100822.0	
8181.4	37602.4	47818.5	59006.9	80038.4	101010.2	
8313.3	37624.5	47819.2	59025.9	80048.8	101010.2	
8332.5	37711.5	47919.9	59033.6	80141.4	101263.3	

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8415.4	37712.1	47920.0	59296.4	80141.9	101263.4
8427.7	37726.9	47976.8	59360.1	80295.8	101464.2
8624.9	37839.1	47977.8	59360.7	80295.8	101465.0
8626.4	37841.0	48067.2	59612.2	80453.5	101543.0
33837.9	38141.9	48067.2	59621.6	80453.5	101553.1
35635.4	38150.6	48274.8	59886.9	80607.5	101554.7
35643.5	38153.0	48274.8	59886.9	80607.5	101582.5
24601.8	38376.0	48366.3	60004.7	80815.0	101592.8
24601.9	38376.2	48366.3	60004.7	80815.0	102556.0
24863.3	38643.2	48545.6	60168.8	81108.8	102624.6
24864.5	38643.2	48545.6	60169.9	81108.8	102633.3
25058.5	38747.9	48869.0	60504.0	84235.6	102791.1
25076.3	38747.9	48869.0	60505.7	84704.0	102795.7
25082.8	38932.3	49234.5	60575.5	84708.5	102970.6
25091.6	38933.0	49234.5	60631.6	85820.2	102970.7
25121.2	38981.7	49648.3	60639.9	85831.6	103158.7
25160.3	38982.7	49648.3	60987.9	85850.3	103158.7
25180.9	39028.8	49982.5	60988.7	85928.3	103484.8
25228.9	39030.4	49982.5	61656.4	86002.6	103484.8
25229.4	39051.3	51643.3	61663.0	86052.3	104974.7
25276.8	39092.7	51644.0	61690.2	86053.9	105180.2
25283.3	39095.3	51693.2	61737.4	86088.5	105237.0
25376.9	39095.4	51693.7	61741.0	86096.2	105653.5
25393.8	39096.9	51697.4	61855.6	86131.1	105659.6
25430.2	41693.1	51705.0	61856.2	86132.7	106183.0
25444.7	41818.7	51725.3	62061.1	86704.2	106183.2
25465.3	41824.9	51758.7	62061.2	86745.7	106204.4
25490.7	41829.1	51763.1	62351.2	86796.7	106237.4
25500.3	41834.5	51763.8	62351.3	86801.2	106250.1
25533.4	41836.1	51765.1	62733.9	86822.6	106250.2
25544.7	41847.8	51953.3	62733.9	87045.5	106317.2
25574.3	41853.5	51953.4	63068.5	87047.8	106503.2
25587.3	41889.9	52230.5	63068.5	87270.3	106508.2
25588.1	41890.6	52243.4	64000.4	87280.0	107230.8
25590.5	41990.9	52322.6	64001.0	87329.6	107231.3
25594.9	41991.0	52377.6	64065.3	87387.5	108011.6
25604.3	42145.5	52381.4	64073.2	87399.9	108011.6
25614.3	42145.6	52464.8	64079.2	87545.7	109908.4
25621.6	42341.8	52465.0	64129.2	87546.0	109908.6
25621.7	42341.8	52562.5	64207.3	87630.7	110586.6
25666.9	42413.8	52562.8	66622.1	87634.7	110589.8
25667.0	42418.5	52596.9	66623.6	88076.2	110834.9
25877.7	42549.4	52597.0	67331.0	88076.6	110873.8
25877.7	42549.4	52665.0	67375.4	88258.0	110974.2
26151.5	42612.9	52665.1	67411.1	88258.1	113934.8
26151.6	42614.9	52976.9	67538.0	88683.8	115269.6
26201.3	42735.2	52976.9	67538.8	88683.9	115285.2
26201.3	42735.2	54791.5	67754.4	88903.2	119540.3
26682.7	42999.8	54850.4	67754.7	88903.2	119576.1
26683.3	43007.2	55002.8	68068.8	89115.6	119609.0
26772.4	43056.4	55139.1	68068.8	89115.6	119743.8
26784.0	43274.9	55155.6	68374.3	93377.3	119744.5
26924.0	43284.2	55168.4	68569.7	93377.5	119984.9
26961.8	43298.9	55202.7	68569.7	93576.6	119985.4
26965.2	43301.4	55465.8	69585.9	93584.7	120293.2
29444.0	43316.0	55468.8	69586.0	93750.1	120293.2
29444.1	43316.0	55613.0	69617.0	93750.2	120664.6
29708.2	43356.0	55623.4	69617.9	93785.1	120664.6
29708.2	43361.3	55625.3	69640.4	95137.0	121152.3
29720.8	43366.6	55628.8	69640.7	95137.0	121152.3
29720.9	43385.4	55649.7	69721.0	95414.2	121902.2
29725.4	43395.5	55714.7	69722.6	95414.5	121902.2
29726.2	43398.1	55722.5	69734.3	95488.6	122270.2
29822.5	43417.7	55817.9	69735.0	95489.7	122271.5
29824.2	43454.9	55818.2	69735.4	95531.5	123404.9
29942.8	43483.2	55953.8	69740.9	95533.0	123430.3
29953.7	43491.2	55953.9	69772.8	95625.8	123695.3
30012.3	43519.1	56114.4	69857.3	95642.9	131539.0
30097.3	43519.9	56114.4	69861.5	95694.0	131837.8
30102.3	43615.5	56242.2	70867.5	95857.0	131865.8
31281.5	43615.6	56272.2	71020.1	95857.1	132505.8
31283.2	43637.1	56272.4	71021.5	96012.8	132510.7
31317.3	43670.9	56274.9	71994.3	96014.2	133668.2
31317.3	43716.2	56342.7	71995.7	96047.6	133668.8
31341.7	43716.2	56342.9	73198.8	96055.3	139205.4
31367.4	43891.3	56408.2	73200.0	96126.2	139205.4
31376.9	43891.3	56421.6	73391.2	96143.0	139723.0

## Supporting Information

31430.1	43925.1	56422.0	73493.6	96156.0	139723.9
31430.6	43925.1	56453.8	73535.7	96156.5	140367.3
31451.1	44022.6	56468.9	73557.4	96195.6	140371.7
31451.1	44026.0	56500.3	73577.9	96391.1	140661.5
31473.0	44618.2	56502.7	73582.2	96391.2	140724.2
31473.0	44665.2	56536.0	76663.9	96514.6	140764.5
31530.6	44675.3	56536.0	76668.1	96515.0	142186.2
31530.7	44812.9	56541.3	76672.7	96581.1	142187.6
31532.9	44817.7	56541.4	76678.0	96695.2	143530.1
31532.9	44989.0	56723.0	76685.7	96718.6	145659.5
33012.0	44990.1	56725.9	76738.9	96756.1	145659.5
33039.0	45001.2	56729.6	76739.2	96756.1	146369.0
33097.5	45002.0	56770.6	76997.8	96846.9	146369.0
33098.4	45006.9	56778.5	76997.9	96861.9	146700.1
33191.7	45009.8	56791.2	77116.7	97037.5	146700.8
34458.9	45080.1	56791.5	77116.8	97037.5	146740.5
34459.1	45135.8	56808.5	77140.3	97068.6	146745.1
34470.5	45142.8	56844.3	77853.3	97072.7	146766.4
34471.2	45152.4	56854.0	77856.9	97148.5	146786.1
34669.3	45173.9	56854.9	78493.8	97179.3	146795.7
34687.3	45177.2	56867.5	78510.9	97182.3	173605.2
34698.0	45178.9	56875.2	79060.3	97320.2	173809.0
34747.5	45218.8	56876.3	79144.1	97320.7	173855.6
34793.9	45218.8	56902.2	79152.7	97515.6	175032.6
34798.2	45484.5	56916.6	79391.6	97580.8	175034.8
34800.1	45484.6	56965.8	79391.6	97593.4	177610.9
37078.2	45559.8	56965.9	79639.6	97593.6	177944.2
37078.3	45587.7	57386.4	79639.6	97598.6	177944.6
37099.6	45904.9	57386.5	79767.2	97968.2	177987.9
37099.7	45904.9	57456.0	79770.5	97968.2	178076.8
37248.4	46295.4	57456.3	79823.1	98168.8	179372.4
37249.6	46325.7	57775.5	79856.3	98191.9	179374.9
37258.4	46588.9				

**Table S17:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_5\text{H}_5)_2]^+$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

2					
Spin free states					
NEVPT2					
0.6	35400.1	45467.2	55480.0	75607.8	91779.7
923.3	35648.5	45595.9	55582.0	75668.5	92347.9
926.2	35775.3	45599.3	55619.1	75740.3	92347.9
1071.4	36028.8	45697.9	55619.5	75861.2	92779.9
1080.3	36029.5	45697.9	55702.8	76028.4	92780.0
1427.3	37786.2	45736.7	55805.1	76028.8	92848.0
1441.0	37786.5	45736.7	55952.1	76173.8	92848.0
1652.9	37815.9	45977.6	55962.5	76249.4	93421.6
1830.8	37816.4	45977.6	55966.1	76275.6	93421.6
1848.9	37932.5	50034.3	56068.0	76437.3	93593.2
6931.0	37943.8	50035.9	56078.7	76468.5	93593.3
7122.2	37996.7	50091.7	56082.6	76680.2	93873.9
7185.5	37999.3	50094.4	56089.7	76745.5	93874.1
7210.4	38008.5	50116.8	56094.6	76748.4	94109.5
7239.6	38010.9	50117.2	56094.7	76782.4	94111.0
7477.7	38051.1	50125.8	56464.1	76876.5	94155.3
7482.4	38058.3	50127.4	56465.7	77127.7	94156.9
28453.1	38067.3	50155.9	56777.7	77130.1	94176.6
30337.8	38236.6	50163.7	56781.4	77290.5	94195.7
30367.2	38237.5	50182.6	57097.9	77290.6	94203.9
24050.5	38245.6	50202.4	57098.5	77479.5	94406.4
24186.9	38249.1	50212.9	57127.0	77479.5	94575.5
24210.4	38300.7	50218.5	57127.2	77647.1	94660.9
24210.6	38300.7	50218.6	57395.9	77647.1	94680.9
24498.1	38414.4	50221.4	57416.3	77848.5	94691.0
24500.3	38414.5	50258.1	57421.0	77848.5	94778.0
24534.6	38580.4	50258.9	57461.1	78192.1	94863.6
24569.1	38582.7	50352.5	57474.7	78192.1	94869.8
24721.2	38608.3	50352.6	57615.0	79272.9	95059.2
24722.3	38611.8	50398.7	57616.7	79296.3	95060.4
24905.1	38659.9	50398.8	58081.4	79327.5	95252.5
24929.9	38672.7	50492.8	58085.3	79459.8	95262.5
25117.1	38682.8	50492.8	58133.1	79579.3	95264.9
25118.9	38759.6	50710.8	58164.8	79700.8	95265.0
25129.5	38761.5	50710.8	58172.5	79710.7	95679.1
25183.8	38762.8	51009.8	58280.1	79734.1	95679.1

## Supporting Information

25209.3	38763.9	51009.8	58281.9	79738.1	95866.2
25421.9	38767.8	51308.0	58507.3	79890.6	95866.3
25452.9	39521.1	51323.4	58507.5	79913.9	95875.8
25454.7	39534.2	51369.8	58829.4	79983.1	95878.5
25462.9	39881.3	51369.8	58829.4	80012.5	96734.7
25471.3	39886.5	51425.4	59261.1	80034.8	96851.8
25486.9	40311.0	51492.9	59261.1	80262.9	96929.6
25511.9	40325.4	51500.5	59468.8	80270.9	97013.3
25536.5	40513.4	51595.1	59469.9	80779.6	97026.8
25571.7	40651.6	51596.1	59471.6	80780.2	97841.7
25638.5	40714.0	51695.9	59516.9	80966.8	97843.5
25692.7	40835.8	51696.0	59529.0	81024.6	98656.6
25880.3	40890.0	51697.9	59553.1	81060.3	98656.6
25897.4	41057.7	51699.3	59573.2	81092.0	100523.5
25979.2	41247.8	51756.6	59573.2	81101.7	100524.0
25988.9	41310.2	51756.7	59671.8	81244.1	101291.6
25991.1	41644.1	51774.6	60461.6	81250.6	101297.5
26186.6	41655.6	51774.6	60482.4	81483.6	101543.7
26240.6	42244.4	51930.7	62352.8	81483.9	101601.8
26420.7	42384.0	51931.0	63488.8	81785.8	101717.1
26506.7	42930.6	52150.7	63543.6	81786.8	103827.5
26534.6	42987.5	52150.7	63591.1	81931.1	105196.0
26574.0	42988.8	52150.8	63726.4	81931.1	105235.1
26757.9	42994.2	52199.4	63729.7	82587.5	110462.1
27134.2	43027.2	52254.1	63950.5	82587.6	110499.4
27362.2	43030.0	52254.1	63952.1	83257.6	110538.0
27648.7	43062.0	52329.5	64264.0	83257.6	110675.3
27650.6	43067.0	52559.3	64264.1	85294.8	110678.1
27884.4	43076.3	52579.5	64814.6	85296.4	110933.5
27884.4	43155.2	52579.5	64814.7	85445.3	110935.4
28291.2	43163.3	52621.6	65221.0	85463.7	111024.8
28291.4	43216.2	52630.3	65223.5	85597.0	111025.3
29632.8	43223.0	52645.0	65869.2	85610.1	111253.0
29632.9	43263.7	52651.1	65897.6	85683.2	111253.1
29929.1	43290.9	52696.8	65898.5	87654.8	111625.1
29929.9	43318.6	52765.8	66445.9	87654.9	111625.1
29958.6	43320.0	52799.8	66446.2	87802.6	112130.6
29958.8	43322.2	52820.3	66461.8	87954.9	112130.6
30011.5	43328.7	52925.5	66464.9	88016.9	112277.9
30011.7	43357.1	52945.9	66495.4	88047.8	112320.5
30043.1	43359.5	53030.5	66496.6	88048.7	112600.3
30048.1	43571.1	53050.1	66630.1	88049.8	113012.2
30159.6	43571.2	53267.4	66634.6	88049.8	113012.2
30171.9	43827.5	53269.8	66638.2	88301.0	118616.6
30227.6	43827.6	53559.5	66644.8	88304.2	118974.8
30345.8	43836.2	53561.8	66652.0	88422.1	119006.4
30348.5	43838.2	53713.6	66652.4	88423.0	119620.4
30355.9	43840.8	53759.6	66659.7	88537.1	119626.5
30386.1	43849.7	53802.2	66674.4	88560.6	120896.3
30492.4	43863.6	53803.9	66779.4	88567.5	120897.6
30494.2	43889.9	53812.9	66781.8	88590.7	127109.8
30585.4	43894.2	53826.9	66887.1	88606.1	127109.8
32123.6	43901.4	53988.0	66918.2	88611.7	127628.1
32123.8	43907.2	54081.3	67342.4	88613.5	127630.8
32199.3	43993.4	54130.8	67344.6	89210.3	128394.9
32226.4	43993.8	54131.5	67498.1	89386.3	128402.4
32233.7	44162.9	54159.5	67857.7	89492.6	128709.9
32241.0	44162.9	54160.4	67869.9	89657.4	128806.7
32241.0	44287.4	54161.6	69892.7	89660.0	128815.4
32290.2	44287.5	54167.5	70310.3	89670.9	128887.5
32292.6	44375.4	54207.2	70311.5	89675.2	128905.3
32338.5	44375.4	54212.6	70315.6	89704.5	130428.2
32338.6	44597.9	54264.9	70317.2	89706.4	132823.9
32342.9	44597.9	54278.1	70327.3	89719.8	132824.0
32343.0	44681.0	54296.3	70365.2	89746.2	133677.3
32425.3	44681.0	54323.1	70366.5	89798.5	133677.3
32425.3	44845.7	54327.1	70495.2	89848.3	134072.2
32427.7	44850.4	54350.1	70504.8	89883.6	134073.0
32427.7	44916.5	54354.7	70780.0	89883.8	134075.5
33221.0	44938.6	54360.4	70780.6	90030.7	134086.6
33222.6	44948.0	54363.1	70853.8	90058.1	134121.3
33239.7	44965.4	54494.5	70862.5	90095.4	134136.6
33240.8	44974.0	54495.1	71157.1	90154.6	134150.4
33484.1	44983.1	54581.1	71187.1	90175.5	156716.6
33494.4	44983.2	54581.6	71853.5	90176.2	156875.6
33525.7	45001.6	54928.4	71921.9	90424.2	156891.5
33597.2	45030.1	54981.0	71974.1	90424.8	157795.0

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33655.8	45034.4	54985.3	74813.8	90808.3	157810.4
33661.9	45087.0	54995.4	75081.5	90809.0	159981.2
33668.5	45120.6	55004.5	75081.5	91012.8	160205.5
34443.6	45186.7	55014.6	75316.5	91030.7	160210.1
34640.8	45197.0	55042.9	75321.9	91030.8	160495.4
34938.2	45287.4	55052.0	75379.5	91034.8	160647.0
34969.5	45287.6	55053.0	75379.6	91362.4	162447.4
35055.3	45465.8	55191.9	75519.4	91362.8	162450.7
35197.9	45465.8	55192.2	75581.6	91779.6	
35249.5	45467.2	55479.8			

**Table S18:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_5\text{H}_5)_2]^+$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

2							
Spin Orbit states							
CASSCF							
0.0	31004.2	40565.1	50563.8	61152.1	70680.4	89912.8	105430.1
496.4	31011.7	40596.7	50621.6	61185.1	70708.5	90163.9	105542.6
819.6	31754.0	40746.2	50623.6	61246.8	70885.5	90164.1	105653.9
1047.2	32046.4	40761.1	50639.1	61260.9	70967.9	90212.3	105900.6
1247.3	32180.1	40781.1	50669.6	61428.6	71627.4	90262.3	105904.6
1421.7	32218.6	40806.5	50687.4	61456.3	72082.8	90281.4	105949.6
1544.0	32226.7	40840.9	50713.0	61532.9	72160.0	90532.9	106195.9
1646.5	32241.8	40866.6	50731.6	61583.9	72209.5	90584.4	106259.2
3344.9	32266.0	40889.1	50856.0	61627.1	72325.5	90630.5	106496.2
3559.5	32285.5	40927.0	50896.7	61669.0	72544.7	90774.4	106535.1
3903.5	32293.6	40947.3	51056.4	61688.1	72871.6	90809.8	106598.4
4180.3	32341.0	40983.9	51062.7	61725.8	73184.8	90967.4	106649.3
4350.3	32346.2	40988.1	51133.6	61737.0	73293.9	91003.9	106821.3
4447.0	32426.5	41017.0	51169.0	61835.9	74436.7	91073.9	107557.1
4499.1	32442.1	41023.8	51177.3	61862.3	74541.6	91133.6	107665.6
5732.6	32451.9	41079.3	51201.8	61876.6	74630.5	91169.5	107827.6
5885.4	32471.8	41239.8	51301.0	61896.7	74736.5	91248.2	107933.9
6167.2	32620.9	41470.3	51303.9	62008.0	74833.5	91252.2	107995.0
6439.4	32744.6	42236.8	51373.4	62021.6	75006.2	91328.6	108003.7
6576.4	32804.7	42305.0	51416.4	62069.1	75008.8	91424.6	108023.2
6632.8	33037.4	42380.4	51419.5	62089.4	75020.9	91460.8	109193.4
7583.1	33139.4	42423.1	51473.5	62157.9	75052.4	91472.0	109412.0
7695.9	33253.1	42586.3	51512.3	62167.4	75109.7	91653.8	109731.0
7952.8	33348.4	42891.1	51552.6	62203.9	75146.2	91785.8	109774.1
8224.3	33403.4	42996.7	51637.6	62426.2	75154.9	91851.1	109833.7
8286.1	33982.8	43066.4	51671.4	62448.9	75157.8	91925.2	109939.0
8978.0	34043.4	43123.8	51778.8	62465.6	75232.8	92009.3	110033.2
9063.9	34046.5	43247.5	51781.3	62478.5	75254.5	92060.9	110071.4
9505.6	34054.1	43266.4	51954.3	62517.6	75362.3	92108.0	110150.7
9603.2	34076.5	43290.6	51978.8	62542.0	75552.2	92210.9	110205.1
9956.7	34094.1	43297.2	52194.0	62561.9	75558.7	92211.7	110352.6
10263.0	34102.5	43333.9	52280.0	62567.1	75650.2	92314.4	110532.3
10298.9	34153.5	43344.1	52296.1	62618.5	75727.7	92537.0	110585.1
10346.7	34173.4	43354.3	52321.6	62622.1	75751.0	92684.4	110861.7
10403.4	34319.6	43417.1	52361.9	62677.3	75756.0	93024.2	111027.2
10490.1	34324.8	43431.0	52428.6	62836.0	75831.1	93078.3	111175.8
10506.6	34382.4	43433.7	52467.8	62955.8	75864.5	93958.5	111337.6
10640.8	34386.7	43444.1	52535.7	63117.0	75911.5	94093.0	111883.8
10726.5	34408.5	43535.1	52692.6	63171.1	75927.9	94414.0	113106.4
11657.7	34438.4	43536.8	52811.3	63247.0	76141.2	94940.6	113130.8
11716.3	34479.6	44647.2	52904.4	63328.6	77487.4	95194.4	113179.2
11764.3	34488.5	44696.3	53138.5	63370.8	77521.2	95268.3	113260.7
11775.8	34589.8	44869.0	53152.8	63453.5	77836.2	95423.5	113577.2
11888.6	34596.2	45008.3	53170.7	63592.0	78540.6	95473.0	113736.7
13372.7	34624.2	45010.8	53485.7	63599.4	79009.5	95712.7	114014.7
13407.5	34857.0	45135.6	53760.6	63676.8	79868.5	96140.2	114327.5
13458.8	35012.4	45155.3	53841.6	63695.3	80438.4	96690.8	114390.2
13471.3	35105.5	45220.6	54215.5	63761.9	80998.3	96914.4	114574.8
14683.3	35729.6	45270.9	54536.7	63831.7	81150.2	97020.8	114643.5
14706.7	35754.0	45346.4	54603.2	63965.9	81253.9	97140.1	114801.3
14718.5	35890.6	45410.1	54693.8	63973.1	81349.0	97274.8	115017.5
15506.2	35925.2	45422.6	54756.8	63973.9	81368.2	97472.7	115553.1
15507.1	35949.9	45442.2	54794.6	63991.4	81378.8	97567.3	115962.7
16025.0	35981.0	45472.8	54825.2	64006.2	81406.7	97618.6	116151.8
24411.1	36034.3	45513.8	54877.7	64054.8	81483.0	97803.8	116324.1

## Supporting Information

24476.9	36079.9	45605.8	54881.0	64073.0	81654.8	97820.1	116592.4
24553.0	36097.1	45619.8	54897.0	64099.0	81684.3	98333.2	117646.8
24563.4	36133.9	45647.6	54924.8	64128.5	81706.0	98412.9	117741.3
24681.6	36205.8	45663.4	54938.1	64153.5	81713.3	98532.9	117895.2
24864.3	36254.2	45755.7	55045.0	64326.1	81758.5	98626.7	117982.0
24988.1	36346.6	45836.4	55109.5	64394.2	81781.4	99137.5	118419.2
25220.0	36529.8	45917.3	55155.6	64423.8	81966.6	99963.2	120374.1
25469.0	36641.0	46038.9	55205.1	64443.2	81979.1	99967.5	121505.4
25600.0	36643.6	46220.0	55439.3	64524.2	82007.8	100034.5	124981.0
25629.9	37014.1	46457.1	55545.4	64887.0	82013.2	100060.8	125058.4
25696.4	37053.4	46596.0	55670.4	65318.5	82040.4	100260.0	125205.9
25712.8	37142.8	46630.4	55720.1	65351.5	82074.7	100467.0	125421.3
26695.6	37210.1	46657.5	55800.2	65443.6	82140.2	100511.6	125695.1
27047.2	37410.8	46706.6	55827.8	65623.3	82175.0	100595.1	125897.7
27146.0	37417.8	46724.1	55903.6	65722.1	82316.3	100603.3	126020.8
27175.7	37422.0	46756.1	55922.3	65752.4	82335.9	100633.1	126061.6
27234.2	37439.0	46833.8	55949.3	65803.5	82352.9	100653.0	126236.4
27244.5	37477.9	46869.6	56005.4	65866.1	82354.6	100964.0	126536.8
27376.9	37535.8	46880.6	56029.5	65905.2	82488.7	101263.4	126648.9
27403.6	37610.2	46883.3	56116.3	66047.6	82658.6	101517.0	126898.0
27448.8	37680.7	46907.1	56144.3	66229.4	82975.7	101690.6	127326.1
27454.3	37722.3	46930.0	56269.9	66243.4	83283.5	101700.3	127885.7
27475.7	37873.2	46953.4	56294.3	66340.9	83421.9	101705.5	127886.9
27507.7	37946.3	47024.6	56485.0	66460.9	84400.8	101727.8	128687.1
27519.4	37951.2	47088.0	56554.8	66520.3	84476.8	101762.6	128818.9
27538.1	37971.5	47141.7	56725.0	66557.4	84631.4	101833.3	129135.1
27600.9	38065.8	47188.8	57008.4	66639.3	84701.2	101858.5	129205.6
27842.4	38144.8	47340.3	57340.3	66860.6	84708.9	101923.5	130531.1
28004.1	38175.5	47390.3	57747.5	66896.0	84737.0	102051.0	136182.2
28162.2	38202.7	47713.1	57909.0	67094.2	84755.6	102100.6	136670.6
28196.8	38216.4	47767.9	57918.7	67151.0	84835.9	102282.9	137729.1
28475.6	38220.5	47787.4	57962.2	67187.1	84870.1	102463.1	139080.7
28532.1	38222.8	47870.1	58018.9	67245.9	84887.8	102604.7	139446.3
28561.6	38438.5	47955.9	58022.5	67261.6	85037.7	102681.2	140157.2
28654.5	38443.5	47966.6	58086.4	67300.2	85046.4	102985.1	141107.7
28667.1	38486.5	48016.0	58181.9	67375.0	85113.6	102994.0	143662.5
28705.6	38573.4	48028.3	59066.4	67428.0	85194.6	103235.5	143982.3
28711.0	38678.2	48092.2	59093.2	67515.1	85250.5	103290.4	144378.4
28722.2	38838.2	48162.8	59122.0	67524.4	85289.7	103388.9	144754.5
28734.6	38967.2	48342.9	59207.5	67607.2	85353.2	103410.3	144925.8
28790.0	38993.0	48575.4	59367.3	67612.2	85372.6	103438.6	146400.9
28797.8	38996.2	48920.6	59379.7	67644.6	85391.8	103495.3	147015.2
28838.1	39037.4	49150.3	59464.0	67809.8	85445.4	103593.4	147142.6
28863.5	39049.2	49179.6	59475.6	67841.2	85475.6	103677.6	147552.9
28888.1	39161.1	49193.4	59585.6	67856.3	85484.3	103704.0	147721.0
28961.6	39213.8	49207.8	59631.3	67884.6	85511.6	103734.5	147835.9
28978.6	39368.5	49300.1	59704.7	67907.9	85545.0	103766.6	149924.3
29092.4	39403.7	49355.7	59878.3	67924.1	85565.8	103932.4	150077.1
29116.6	39405.4	49381.3	59960.4	68150.9	85567.4	103989.6	150533.8
29172.7	39408.2	49464.2	60055.8	68174.7	85614.4	104088.8	150845.3
29266.3	39412.2	49552.8	60081.8	68292.3	85716.6	104143.0	150920.5
29270.1	39417.3	49563.8	60153.4	68338.4	85718.2	104149.3	150937.4
29305.3	39444.6	49605.0	60270.4	68445.9	85772.7	104299.7	150962.6
29369.0	39454.0	49649.1	60373.4	68488.1	85846.6	104328.0	153240.8
29379.8	39517.8	49654.5	60413.9	68625.4	86000.3	104355.7	154038.0
29457.9	39545.8	49677.5	60420.3	68859.5	86048.1	104399.6	154447.7
29484.4	39606.5	49840.8	60537.7	69064.3	86166.4	104649.5	154571.0
29521.3	39630.6	49945.6	60551.1	69242.5	86340.0	104655.3	154604.4
29528.6	39655.6	50041.3	60620.8	69387.0	86413.7	104656.7	177217.2
30148.8	39803.0	50072.5	60623.6	69390.6	86562.2	104698.1	177911.7
30354.8	39861.4	50225.1	60647.4	69700.2	86683.8	104705.6	179463.1
30576.6	39917.9	50245.6	60726.6	69808.4	86753.6	104773.7	181209.9
30685.9	39972.8	50286.5	60757.6	69844.3	86791.2	104855.8	182295.8
30722.6	39979.5	50356.8	60827.1	69886.0	86825.6	105045.8	182581.7
30749.3	40020.2	50388.0	60846.1	69960.2	87499.4	105103.3	182679.9
30774.1	40031.3	50400.7	60849.4	70105.2	87818.4	105126.6	182685.9
30785.1	40205.9	50416.6	60884.2	70157.6	89271.5	105130.9	183506.1
30868.5	40231.4	50440.5	60965.3	70194.2	89351.4	105150.8	185354.9
30892.2	40272.9	50465.4	60985.3	70390.8	89396.5	105186.4	185782.1
30932.6	40370.0	50467.8	61058.9	70421.3	89612.6	105273.8	187050.7
30987.2							

**Table S19:** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_5\text{H}_5)_2]^+$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

Supporting Information

Spin Orbit states							
NEVPT2							
0.0	31376.4	39937.8	49537.7	58797.8	66989.6	83404.7	98406.9
535.5	31440.7	40016.6	49766.7	58843.1	67024.9	83453.1	98512.6
859.2	31478.9	40031.1	50058.8	58925.2	67097.3	83629.0	98529.4
1082.2	31524.1	40070.6	50086.9	59080.6	67289.5	83719.8	98590.4
1290.5	31587.1	40089.9	50156.8	59107.2	67297.8	83819.3	98660.0
1482.7	31683.3	40102.5	50264.6	59111.3	67573.6	83845.1	98824.2
1621.5	31812.4	40154.2	50438.8	59140.3	67766.6	83867.7	98862.7
1764.3	31834.3	40185.3	50493.2	59218.1	67802.6	83885.4	98959.5
3383.3	31890.2	40220.8	50498.2	59254.6	68332.9	83909.9	98983.3
3560.9	31933.8	40278.0	50637.5	59279.7	68390.0	83914.4	99084.8
3945.5	31990.4	40342.8	50655.7	59326.7	68466.4	83961.3	99184.4
4244.9	32008.2	40357.3	50671.7	59433.0	68716.0	84044.3	99432.8
4416.3	32067.9	40377.7	50694.0	59458.6	68778.4	84102.1	99445.7
4517.4	32074.5	40415.5	50731.0	59511.5	68864.3	84268.1	99714.6
4572.2	32114.1	40419.4	50793.0	59596.6	69099.6	84312.4	99891.6
5744.2	32126.9	40467.5	50798.9	59633.6	69110.6	84460.1	99897.5
5875.9	32182.8	40557.5	50806.0	59677.8	69113.8	84535.2	100121.1
6177.7	32291.9	40582.4	51171.3	59784.3	69369.9	84568.9	100367.8
6479.1	32381.6	40611.2	52336.4	59791.5	69561.7	84601.3	100437.8
6615.5	32593.2	40809.7	52357.4	59801.0	69848.3	84610.1	100473.0
6655.9	32655.4	40987.2	52368.8	59848.3	70627.0	84675.4	100498.4
7574.2	32737.4	41011.2	52392.2	59869.0	70798.6	84734.2	100577.9
7683.0	32757.4	41116.5	52404.7	59921.2	71014.6	84781.1	100619.2
7947.9	32851.8	41160.0	52424.6	59963.1	71239.9	84807.8	100629.4
8246.5	32968.1	41185.4	52456.4	59982.6	71262.0	84892.0	100649.2
8291.0	32980.1	41220.1	52515.6	59997.7	71315.2	85171.1	100872.7
8934.8	33043.2	41256.1	52543.1	60034.6	71697.5	85217.6	100988.5
9021.9	33086.2	41272.4	52549.2	60129.9	71749.8	85301.2	101122.9
9046.3	33100.2	41316.4	52587.5	60139.5	71883.5	85341.9	101190.5
9096.9	33184.0	41323.4	52610.7	60198.7	71889.3	85473.3	101228.6
9143.7	33391.9	41400.2	52738.0	60220.8	71953.9	85623.8	101358.3
9207.7	33490.4	41419.3	52740.8	60255.0	72012.7	85654.9	101429.5
9314.5	33595.6	41525.6	52780.2	60257.4	72049.1	85692.5	101434.3
9483.3	33691.8	41898.4	52815.6	60295.2	72099.5	85892.5	101495.1
9520.6	33723.8	42117.4	52854.5	60329.3	72101.4	85974.9	101507.1
9590.5	33787.1	42353.5	52869.5	60338.3	72157.3	86413.9	101730.1
9937.2	33860.2	42650.4	52897.5	60346.5	72427.1	86715.8	102091.0
10412.4	33961.8	42905.9	52977.0	60413.6	72497.5	86850.3	102652.7
10498.4	34039.0	44054.8	52987.6	60486.1	72513.4	87195.7	103511.7
10636.5	34207.2	44080.7	53014.8	60510.3	72527.6	87772.4	103560.7
10651.9	34388.0	44089.3	53123.5	60562.7	72547.8	88316.2	103599.6
10658.4	34455.8	44138.5	53171.5	60580.5	72608.7	88660.6	103740.2
10738.5	34481.9	44166.6	53413.0	60730.2	72645.1	88727.5	104639.6
10763.1	34547.5	44189.8	53420.6	60749.8	72664.6	88867.5	104733.4
12207.8	34570.0	44257.1	53424.3	60759.2	72735.2	89127.3	104811.3
12262.9	34585.1	44281.3	53434.5	60774.6	73566.2	89138.9	105002.1
12312.7	34664.8	44317.7	53439.7	60905.5	73821.1	89386.2	105020.0
12328.2	34710.7	44335.8	53443.1	60966.3	74781.4	89531.7	105265.3
13483.3	34724.3	44443.7	53473.3	60981.1	74937.0	89591.3	105459.6
13512.4	34743.0	44512.4	53548.6	61016.2	74973.4	89672.9	105569.1
13535.4	34875.0	44550.6	53669.0	61039.6	75011.7	90216.7	105825.9
14265.5	35024.7	44631.7	53684.2	61066.5	75185.8	90281.9	106001.3
14274.5	35058.3	44661.9	53725.0	61126.2	75195.6	90545.5	106467.3
14778.8	35091.4	44919.7	53784.6	61169.1	75217.7	90683.2	106555.8
23611.9	35142.1	44946.7	54603.0	61214.7	75226.3	90822.3	106940.2
23847.1	35190.3	44992.9	54695.2	61253.5	75286.3	90833.4	107230.9
23881.7	35349.4	45276.6	54722.4	61409.5	75302.3	90988.7	108163.3
24144.8	35526.8	45541.6	54771.9	61611.8	75305.7	91086.6	108346.8
24310.2	35680.4	45620.9	54791.9	61647.9	75416.6	91503.2	108431.8
24683.2	35799.1	45644.9	54819.7	61704.7	75432.3	92107.9	108475.1
24817.6	35819.5	45700.3	54867.1	61778.7	75477.8	92191.7	108593.0
25159.6	35861.7	46203.6	54881.4	62091.7	75547.4	92568.1	110273.9
25420.1	36000.5	46267.7	54969.5	62153.4	75746.8	92824.8	111371.3
25529.7	36125.7	46382.1	55022.0	62195.5	75767.1	92857.3	115995.7
25609.0	36211.4	46499.7	55099.3	62285.6	75808.2	92930.9	116076.1
25622.7	36298.0	46524.5	55154.6	62289.9	75819.8	92999.3	116234.4
25638.3	36318.5	46544.1	55266.1	62410.6	75846.6	93035.4	116463.2
25770.5	36345.7	46560.2	55365.5	62447.4	75848.9	93237.1	116744.6
25797.9	36375.9	46637.6	55435.4	62468.1	75863.3	93318.0	116771.3
25858.0	36402.2	46657.0	55502.5	62494.6	75965.2	93340.7	116875.5
25977.8	36467.0	46678.3	55527.0	62531.5	75989.8	93422.9	117002.1
26120.0	36541.0	46689.3	55546.2	62604.5	76075.9	93448.7	117123.9
26251.7	36585.1	46707.7	55583.8	62676.7	76171.9	93506.5	117227.1
27679.9	36598.9	46735.2	55607.7	62743.7	76268.8	93607.9	117543.3
27714.6	36621.3	46802.0	55622.4	62817.4	76305.4	93619.4	117717.5



## Supporting Information

27732.1	36681.5	46815.3	55664.1	62920.3	76345.0	93715.8	117815.2
27822.1	36719.5	46835.1	55762.1	62976.9	76362.3	93923.0	117916.0
27842.6	36759.6	46861.7	55820.9	63016.2	76450.8	94225.5	118077.2
27874.1	36762.7	46910.9	55875.9	63270.6	77756.6	94321.0	118125.1
28006.4	36906.6	46938.9	55944.3	63286.4	78327.1	94340.4	118349.1
28152.6	37027.9	47046.4	56005.8	63366.7	78335.6	94394.2	118945.0
28182.3	37099.2	47060.6	56041.7	63417.5	78608.5	94580.7	119474.6
28202.7	37104.9	47096.7	56301.7	63522.8	78746.0	94638.6	119850.9
28316.1	37117.5	47178.7	56380.9	63600.8	78819.7	94817.6	123435.3
28352.1	37554.4	47216.6	56401.9	63705.1	78899.6	94832.8	123909.4
28417.4	37665.9	47258.6	56407.0	63880.3	78910.2	94880.9	125006.9
28436.5	37694.7	47387.7	56546.4	63886.8	78916.3	94919.6	126345.3
28712.7	37713.4	47446.6	56718.1	63908.0	79471.8	95004.3	126731.1
28833.3	37734.5	47485.9	56728.2	63911.4	79654.5	95199.2	127457.0
28859.2	37797.3	47529.0	56743.2	63915.8	79731.9	95206.6	128485.9
28980.8	37879.7	47555.5	56819.8	63975.9	79931.2	95301.3	131557.1
29091.1	37908.8	47571.7	56835.2	64077.5	80021.4	95514.1	131894.0
29196.3	37921.2	47600.7	56890.7	64091.1	80103.2	95549.6	132360.0
29249.5	37940.2	47610.5	56921.0	64271.4	80188.8	95564.9	132802.9
29310.2	37978.9	47615.5	56943.0	64305.6	80430.7	95584.1	132982.2
29366.2	38151.9	47637.2	56962.9	64384.3	80481.4	95631.2	133897.9
29440.2	38164.5	47646.6	56985.7	64416.9	80564.3	95708.9	134340.7
29520.3	38182.3	47677.8	57000.7	64434.2	80668.9	95837.0	134596.9
29541.2	38195.7	47717.9	57018.7	64530.1	80672.4	96351.2	135011.0
29614.2	38266.1	47737.7	57032.8	64539.9	80787.9	96449.2	135634.8
29651.8	38286.1	47780.2	57056.3	64640.7	80833.1	96469.9	135877.6
29673.8	38335.1	47810.9	57099.3	64677.6	80881.2	96483.5	136973.6
29692.3	38399.0	47822.2	57148.8	64741.0	80908.8	96537.4	137125.4
29721.7	38403.0	47836.3	57172.0	64747.1	80974.3	96680.8	137867.0
29733.7	38414.1	47868.2	57239.3	64968.4	81008.5	96790.9	138243.6
29791.1	38486.1	47884.6	57316.3	64999.1	81102.6	96874.5	138292.4
29838.0	38555.3	47912.3	57319.9	65023.7	81318.1	96894.3	138323.9
30020.8	38587.2	47932.1	57435.5	65077.6	81370.0	96933.7	138348.8
30127.3	38651.2	48009.2	57496.4	65191.6	81491.8	96978.9	140495.2
30257.7	38675.6	48141.9	57506.1	65283.7	81625.9	97013.3	141420.0
30306.8	38697.2	48178.5	57672.6	65289.5	81795.6	97082.7	141881.3
30457.2	38711.4	48305.2	57928.0	65354.7	81832.5	97126.9	141980.7
30490.1	38799.3	48404.1	58123.7	65403.6	81875.6	97170.1	142020.6
30670.6	38910.6	48417.7	58222.0	65464.9	81925.2	97224.4	160314.8
30724.3	38967.9	48426.6	58273.4	65667.0	82070.2	97239.8	160896.3
30826.5	39065.7	48544.2	58315.1	65728.2	82231.6	97266.8	162213.4
30860.1	39117.6	48659.7	58388.1	65777.6	82245.8	97353.3	164193.8
30944.6	39209.8	48683.8	58498.8	65841.4	82426.9	97414.6	164991.6
31001.2	39220.2	48730.3	58641.9	65961.7	82439.7	97464.6	165095.9
31056.5	39293.8	48859.0	58661.9	66046.0	82586.7	97550.3	165196.2
31084.1	39839.1	48895.2	58668.1	66081.3	82707.8	97571.1	165417.1
31093.3	39855.9	49079.8	58679.7	66118.9	82794.4	97581.2	166611.0
31118.2	39873.7	49261.4	58698.9	66233.3	82825.1	97637.0	167740.9
31159.3	39879.9	49317.8	58717.5	66393.9	82989.4	97965.1	168328.8
31198.5	39906.5	49390.1	58743.7	66434.1	83199.0	98180.6	170136.1
31267.5							

**Table S20:** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_6\text{H}_6)_2]^{3+}$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

3					
Spin free States					
CASSCF					
0.0	37063.8	47289.6	57489.1	79741.4	98302.5
626.1	37063.8	47319.0	57528.0	79745.9	98311.8
629.3	37096.0	47351.7	57537.4	79780.1	98365.5
654.2	37096.2	47439.8	57653.4	79805.7	98365.5
655.1	37101.1	47442.3	57688.4	79835.8	98451.0
832.1	37102.9	47672.4	57688.8	79940.3	98451.0
848.5	37200.3	47673.3	57716.2	79960.7	99018.3
1111.4	37203.5	47753.6	57717.9	79980.9	99019.6
1137.2	37220.0	47754.1	58863.5	80006.8	100902.3
1235.2	37220.1	47800.1	58866.9	80012.4	100902.4
7872.0	37383.3	47805.6	58903.5	80042.3	101013.4
7911.0	37423.6	47828.5	58936.9	80043.4	101013.4
8017.4	37463.3	47849.8	58983.6	80111.7	101205.9
8024.1	37464.2	47855.4	59109.5	80111.8	101206.0
8053.1	37542.6	47901.5	59112.0	80226.7	101335.9

Supporting Information

8178.7	37582.3	47902.6	59389.4	80226.8	101336.7
8180.7	37703.9	47925.0	59492.6	80331.1	101338.7
33992.8	37743.3	47925.0	59492.7	80331.1	101344.6
35100.1	37890.4	47985.2	59583.5	80403.6	101352.9
35190.2	37893.9	47985.3	59599.5	80403.6	101363.6
24497.9	37942.3	48121.1	59599.6	80526.6	101365.8
24497.9	37942.5	48121.1	59641.2	80526.6	102629.8
24713.9	38511.7	48314.1	60041.4	80735.8	102673.6
24713.9	38511.7	48314.1	60048.6	80735.8	102705.7
24815.0	38541.0	48546.4	60362.9	84457.7	102795.2
24848.1	38541.0	48546.4	60363.6	84728.8	102800.9
24858.1	38697.7	48799.0	60396.6	84788.9	102875.3
24866.6	38710.5	48799.0	60438.5	85616.9	102876.8
24869.3	38711.6	49066.0	60443.5	85618.0	102921.0
24876.5	38727.9	49066.0	60560.4	85777.3	102921.1
24899.3	38739.4	49181.5	60564.1	85779.3	103236.9
24920.4	38795.8	49181.5	61698.8	85883.8	103236.9
24922.2	38802.8	51472.0	61699.0	85893.3	105121.7
25077.2	38821.2	51496.5	61732.3	86008.6	105259.7
25080.3	38821.4	51503.2	61747.3	86028.7	105310.3
25105.8	38834.9	51503.9	61757.1	86030.2	105528.2
25168.8	38835.9	51516.7	61814.5	86073.8	105535.8
25170.5	41674.9	51564.9	61815.8	86076.4	105788.2
25183.9	41780.7	51574.9	61952.8	86909.1	105789.0
25194.3	41780.8	51590.2	61952.9	86911.6	105962.1
25266.5	41799.1	51591.3	62162.0	86941.7	105962.1
25299.8	41800.6	51631.3	62162.1	86973.3	106428.6
25302.3	41822.7	51632.6	62420.5	87000.7	106480.2
25310.6	41834.5	51710.7	62420.5	87088.5	106510.1
25314.8	41845.1	51710.7	62559.1	87089.4	106576.3
25316.1	41851.7	52106.0	62559.1	87465.1	106602.1
25317.0	41856.1	52152.6	63881.3	87466.5	107156.5
25319.0	41930.4	52162.1	63882.9	87466.7	107157.5
25319.3	41930.4	52261.8	63945.4	87470.3	107575.4
25367.0	42061.3	52262.8	63956.6	87499.6	107575.5
25367.5	42061.3	52352.4	63981.8	87516.8	110114.5
25377.9	42137.1	52354.3	63985.0	87537.5	110115.0
25379.1	42167.6	52370.8	64085.1	87622.6	110573.2
25389.0	42206.5	52370.8	66715.5	87625.1	110576.4
25389.1	42206.5	52385.8	66805.0	87954.7	110702.5
25416.9	42322.3	52386.0	67331.4	87954.9	110734.4
25416.9	42322.3	52416.4	67374.7	87996.3	110869.5
25767.3	42407.6	52416.6	67399.5	87996.3	114284.4
25767.3	42407.6	52657.7	67508.4	88247.4	115136.4
26022.5	42552.4	52657.7	67511.2	88247.4	115222.6
26022.6	42553.5	54685.0	67652.1	88422.1	119866.5
26368.9	42739.4	54882.2	67652.3	88422.2	119885.9
26370.1	42739.4	54891.0	67830.3	88568.8	119937.7
26376.2	42831.6	54929.5	67830.4	88568.8	120027.7
26377.7	42838.7	54932.3	67883.8	93484.6	120034.5
26545.9	42942.5	55163.4	68152.3	93485.0	120201.4
26551.1	42977.2	55166.9	68152.3	93564.7	120202.0
26559.3	42979.4	55206.2	69474.7	93568.4	120395.7
29329.7	43052.5	55206.8	69474.7	93665.1	120395.8
29329.7	43092.5	55556.1	69487.2	93717.6	120603.5
29504.3	43092.8	55561.7	69487.3	93718.2	120603.6
29504.7	43106.8	55572.2	69494.9	95244.6	120873.2
29518.1	43116.9	55576.6	69496.8	95244.6	120873.2
29520.6	43117.2	55584.8	69569.2	95436.2	121394.4
29553.0	43144.2	55656.7	69574.0	95436.3	121394.4
29553.2	43147.9	55657.0	69582.0	95476.2	122569.4
29608.1	43220.3	55754.4	69582.0	95476.8	122572.1
29608.1	43224.6	55754.4	69652.0	95531.6	123277.0
29653.6	43248.0	55814.2	69652.0	95535.1	123329.0
29656.1	43248.2	55814.2	69660.1	95622.4	123466.8
29770.1	43282.6	55895.8	69668.2	95640.6	131914.5
29773.7	43286.2	55895.8	69700.6	95687.6	132120.6
29807.9	43298.0	55991.5	70820.4	96105.9	132193.8
30996.1	43307.3	55991.5	70959.7	96106.1	132532.6
31008.8	43350.4	56148.1	71000.6	96214.7	132539.3
31019.5	43350.4	56148.1	72229.4	96219.5	133317.3
31025.1	43417.5	56176.1	72232.1	96231.2	133318.2
31025.1	43433.6	56289.5	73083.6	96243.5	139762.9
31060.4	43446.4	56300.6	73090.5	96287.6	139762.9
31064.1	43446.4	56326.9	73191.4	96340.8	140061.6
31078.1	43568.9	56327.6	73217.1	96370.0	140062.7
31078.1	43568.9	56363.4	73267.7	96379.4	140522.6

## Supporting Information

31116.5	43639.5	56363.8	73270.7	96380.7	140529.0
31117.0	43643.2	56382.5	73274.0	96493.6	140642.7
31137.5	44577.3	56383.7	73289.8	96494.7	140664.7
31137.5	44600.5	56441.2	76647.4	96582.7	140693.2
31152.0	44638.1	56441.2	76650.4	96582.7	142409.1
31152.0	44729.1	56504.7	76654.7	96816.7	142484.5
31162.3	44736.5	56516.1	76655.5	96839.9	143307.0
31162.3	44810.3	56524.3	76660.3	96839.9	146052.9
32834.1	44812.8	56544.9	76705.6	96848.3	146052.9
32836.8	44813.2	56544.9	76705.7	96870.9	146433.6
32852.7	44814.0	56563.3	76733.5	96877.8	146433.7
32860.6	44855.1	56563.4	76877.9	96886.4	146648.2
32928.1	44856.6	56625.8	76878.0	96886.4	146648.7
34196.5	44902.6	56628.6	76907.6	96888.0	146708.6
34196.5	44918.7	56650.9	76907.6	96971.2	146717.8
34301.5	44939.0	56651.3	77933.7	97045.4	146757.9
34301.5	44939.0	56693.1	77937.8	97094.3	146764.6
34362.7	44972.3	56693.9	78320.0	97223.8	146784.6
34422.6	44972.5	56800.7	78323.3	97228.2	174362.9
34440.8	44973.7	56806.7	78651.5	97339.0	174544.9
34449.7	44973.8	56836.5	78750.6	97340.0	174676.9
34451.5	44984.4	56839.8	78787.9	97480.3	175575.2
34524.0	45101.9	56849.8	79429.6	97480.4	175583.6
34528.5	45101.9	56885.6	79429.6	97693.3	177920.3
36815.0	45435.4	56928.8	79587.0	97693.3	177971.6
36815.0	45435.4	57005.2	79587.0	97759.5	178041.7
36923.6	45535.1	57005.2	79678.8	97835.9	178083.1
36923.7	45550.4	57189.0	79679.0	97912.3	178085.9
36989.3	46005.5	57189.5	79715.8	98010.6	178712.8
37014.3	46061.7	57488.3	79717.2	98010.6	178730.4
37020.6	46207.3				

**Table S21:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_6\text{H}_6)_2]^{3+}$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

3							
Spin free States							
NEVPT2							
0.2	31900.4	42860.9	50328.0	55351.7	67352.7	82610.7	95074.2
593.8	31905.1	42868.8	50396.0	55351.8	67484.2	82610.7	95074.7
616.4	31905.1	42897.1	50396.4	55416.2	67485.3	85489.4	95276.5
709.9	31918.7	42928.6	50530.7	55416.3	69401.9	85492.5	95285.4
715.8	31918.7	42967.5	50530.8	55512.6	70443.5	85541.9	95470.6
810.8	31956.9	42998.9	50741.7	55512.7	70446.4	85547.4	95470.6
846.4	31960.7	43005.6	50741.7	55687.2	70446.4	85597.3	95540.2
1167.9	31973.1	43005.6	50999.7	55687.3	70446.8	85706.4	95549.5
1190.7	31973.1	43023.4	50999.7	55687.3	70448.0	85707.3	95705.4
1299.7	31998.0	43077.9	51166.8	55895.2	70517.3	87857.9	95705.7
6627.0	31998.1	43082.2	51223.8	55901.9	70519.2	87857.9	97124.2
6673.5	32008.3	43082.4	51233.0	55923.3	70615.7	88100.5	97208.3
6799.1	32008.4	43087.2	51281.9	55961.2	70635.3	88100.8	97215.1
6816.2	32926.7	43129.1	51281.9	55980.8	70736.9	88194.2	97217.5
6876.9	32926.8	43214.3	51359.5	55997.5	70736.9	88196.3	97273.9
6982.7	33073.7	43220.4	51359.7	56057.0	70762.2	88259.2	97885.4
6992.4	33074.0	43327.0	51438.1	56172.8	70762.3	88267.0	97893.6
28712.1	33125.7	43327.5	51438.5	56186.1	71022.6	88272.9	98297.6
29860.3	33210.1	43431.3	51462.2	56555.9	71029.6	88317.4	98297.8
29955.4	33232.5	43431.3	51465.2	56556.1	71413.3	88350.7	100899.2
24143.7	33243.8	43786.4	51467.2	56559.9	71562.0	88359.4	100902.8
24148.2	33254.7	43786.5	51471.9	56570.0	71598.6	88372.5	101409.5
24148.2	33339.1	43801.4	51526.3	56745.2	75174.9	88506.1	101413.9
24220.7	33347.5	43803.5	51527.6	56745.3	75186.0	88508.8	101539.5
24316.8	34027.9	43803.6	51594.8	57203.5	75186.0	88561.0	101571.8
24316.9	34094.3	43811.0	51594.8	57207.3	75351.7	88694.6	101748.0
24437.9	34375.6	43829.4	51693.2	57215.8	75351.7	88699.2	104358.1
24451.4	34936.6	43845.4	51693.2	57235.7	75371.1	88883.0	105231.1
24462.4	34939.6	43855.0	51773.0	57242.1	75371.4	88896.3	105322.8
24462.9	35054.5	43862.7	51773.0	57250.3	75393.2	88997.2	110993.8
24595.4	35081.1	43873.7	52021.1	57255.7	75396.5	89101.9	111019.6
24613.2	35264.9	43934.9	52221.3	58218.2	75464.9	89167.3	111068.0
24622.6	35469.4	43934.9	52221.7	58219.8	75483.7	89336.8	111159.3
24710.3	35487.1	44088.5	52244.3	58250.8	75521.1	89350.4	111165.7
24715.2	35534.3	44089.0	52255.0	58259.1	75526.4	89615.5	111335.1
24722.8	35535.2	44246.8	52282.5	58273.1	75537.6	89623.9	111338.2
24762.1	37452.4	44246.9	52296.2	58317.1	75558.0	90211.7	111523.5
24764.9	37452.5	44355.3	52535.5	58322.1	75644.2	90227.1	111525.9

## Supporting Information

24765.2	37566.7	44355.3	52540.5	58465.7	76387.6	90261.1	111526.6
24765.2	37567.0	44359.7	52668.9	58466.4	76389.0	90310.6	111527.5
24832.0	37629.9	44359.7	52701.2	58697.2	76852.3	90313.4	111718.2
24835.0	37654.4	44600.4	52702.4	58697.2	76964.7	90319.9	111718.2
24967.6	37660.6	44601.6	52796.0	58984.7	76975.3	90453.3	111983.6
24979.7	37765.0	44629.2	52805.4	58984.7	76996.5	90453.8	111983.6
24983.3	37766.6	44630.9	52831.1	59082.2	77003.6	90521.2	112293.5
25098.4	37767.5	44671.5	52832.7	59082.2	77105.2	90551.3	112348.0
25131.8	37768.7	44691.3	52964.0	59347.8	77106.7	90593.7	112490.9
25164.7	37798.2	44766.9	53015.2	59356.4	77205.3	90774.5	112603.7
25236.4	37798.2	44775.7	53019.9	59374.2	77205.4	90799.5	112603.7
25299.0	37908.0	44779.8	53038.4	59385.5	77289.2	90799.6	119214.5
25328.1	37924.9	44805.7	53238.4	59455.1	77289.2	90945.8	119471.7
25614.7	37946.6	44805.8	53239.0	59459.5	77402.8	90958.3	119548.7
26011.7	37947.4	44812.4	53319.2	59560.4	77402.9	91018.5	119838.8
26083.9	38086.3	44813.2	53380.3	60672.4	77497.5	91022.4	119848.3
26269.4	38086.5	44822.6	53383.1	60769.6	77497.5	91064.5	120696.1
26481.4	38136.7	44837.0	53545.7	61906.0	77607.9	91064.7	120701.9
26518.0	38136.7	44839.8	53560.0	63511.4	77607.9	91169.7	127973.3
26763.1	38221.5	44849.5	53789.8	63567.7	77854.4	91178.2	127973.4
26809.0	38284.4	44856.2	53840.2	63590.7	77854.4	91304.5	128252.2
26965.4	38285.2	44961.6	53840.2	63708.5	79313.0	91304.8	128258.1
26992.6	38329.4	44963.4	53922.5	63713.6	79313.0	91535.3	128796.2
27166.3	38339.9	45020.9	53931.1	63852.4	79469.5	91535.3	128805.4
27247.1	38375.7	45020.9	53966.7	63853.2	79488.7	91937.0	128919.0
27247.6	38386.9	45071.3	53966.7	64019.8	79654.5	91937.0	128924.7
27376.2	38425.8	45071.3	53988.3	64019.9	79679.0	92263.7	128977.4
27376.2	38433.6	45213.5	53988.5	64376.5	79683.3	92263.7	129361.5
27802.9	38437.5	45219.1	53990.8	64376.5	79733.7	92353.4	129447.0
27803.0	38438.5	45240.7	53994.3	65580.0	79749.7	92353.4	130365.6
29487.6	38705.7	45240.7	54007.5	65583.8	80250.1	93601.8	133502.8
29487.7	39183.5	45460.8	54010.9	65839.4	80254.9	93601.8	133502.9
29655.0	39215.1	45461.1	54013.2	65921.3	80267.5	93708.8	133976.0
29657.2	39795.1	49840.1	54045.0	65971.0	80302.2	93708.8	133976.3
29659.4	39801.7	49875.2	54045.1	66365.9	80329.1	93949.3	134231.7
29668.6	40089.1	49876.4	54060.8	66366.0	80374.8	93949.3	134232.8
29751.1	40100.3	49878.8	54066.7	66370.4	80387.0	94054.5	134278.5
29751.6	40199.7	49905.1	54213.8	66371.1	80749.4	94063.0	134296.2
29816.6	40239.5	49961.5	54220.7	66372.1	80750.3	94077.2	134336.1
29824.2	40443.1	49981.9	54248.9	66380.9	81259.7	94099.9	134344.3
29854.9	40489.9	49997.8	54249.1	66465.9	81271.4	94104.8	134370.5
29855.0	40578.8	49998.1	54314.1	66476.4	81273.5	94109.5	157899.8
29957.9	40924.7	50044.3	54320.3	66508.8	81273.6	94112.2	158072.2
29962.2	41022.2	50051.7	54756.4	66509.0	81280.8	94739.8	158219.0
30004.0	41053.7	50114.2	54756.5	66582.5	81341.0	94766.3	158905.0
30173.4	41218.0	50114.2	54991.5	66593.8	81371.8	94827.4	158917.1
30175.6	41240.9	50213.5	55006.5	66596.9	81372.1	94860.4	160618.4
30205.7	42161.9	50214.3	55006.6	66611.5	81377.9	94957.3	160687.8
30220.6	42190.9	50250.6	55006.8	66611.7	81431.8	94967.4	160697.0
30282.7	42616.8	50255.5	55007.9	66621.0	81431.9	94974.9	160740.0
31813.6	42675.8	50283.3	55028.2	66635.5	81781.4	95010.4	160810.1
31830.5	42820.4	50292.2	55032.2	66674.4	81782.3	95044.8	161816.8
31843.8	42825.2	50301.7	55032.7	67242.2	82382.4	95051.6	161847.6
31892.6	42832.0	50324.2	55038.3	67253.4	82382.5		

**Table S22:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_6\text{H}_6)_2]^{3+}$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

3							
Spin Orbit States							
CASSCF							
0.0	30728.4	40309.3	50329.5	61017.2	70471.4	89759.6	105156.4
388.8	30743.9	40481.9	50481.5	61018.7	70603.8	90147.6	105394.6
557.8	31649.7	40497.1	50501.8	61039.4	70672.6	90152.2	105424.3
661.9	31841.3	40554.1	50509.2	61073.7	70911.3	90189.9	105625.8
779.0	31966.5	40555.9	50515.7	61174.8	71405.6	90226.5	105710.8
912.5	31983.1	40581.4	50553.8	61320.6	71595.3	90249.6	105917.3
1034.8	32008.1	40600.2	50557.7	61340.7	72152.3	90440.4	106159.6
1117.9	32011.1	40653.3	50597.4	61407.2	72200.6	90470.7	106228.7
3263.4	32014.2	40667.0	50640.7	61487.2	72286.9	90538.5	106439.4
3356.8	32018.9	40691.0	50777.1	61488.6	72437.3	90620.1	106458.0
3618.8	32031.7	40692.8	50797.6	61546.1	72646.2	90754.8	106528.4
3816.8	32042.9	40712.3	50902.6	61565.5	72880.7	90772.8	106561.0
3914.8	32108.1	40748.9	50915.3	61608.6	73092.2	90958.3	106670.5
3954.1	32130.6	40764.3	50939.0	61760.7	74393.9	91188.3	107557.4

Supporting Information

3999.2	32139.1	40779.0	51009.5	61770.8	74559.3	91262.6	107595.2
5575.1	32159.5	40796.7	51027.6	61773.8	74696.5	91284.7	107702.2
5637.1	32297.9	40967.3	51044.7	61847.8	74775.9	91333.0	107777.6
5894.7	32403.7	41058.3	51103.6	61851.6	74817.4	91354.1	107779.9
6074.0	32485.1	42024.5	51157.1	61945.8	74909.1	91382.0	107804.7
6115.7	32605.4	42097.0	51162.7	61983.8	74918.3	91427.4	107814.8
6157.0	32837.4	42135.0	51187.8	62003.8	74943.8	91464.9	109394.9
7360.4	32844.0	42162.0	51221.3	62034.2	74944.4	91471.2	109549.9
7427.9	32907.2	42260.7	51269.8	62046.2	74985.1	91544.3	109723.8
7682.6	33052.8	42662.4	51282.9	62059.1	75016.8	91691.3	109787.2
7786.2	33238.3	42784.3	51295.6	62199.4	75044.2	91693.0	109803.1
7845.9	33694.1	42842.9	51334.8	62228.3	75054.4	91802.4	109857.1
8709.7	33790.2	42916.1	51422.8	62262.1	75099.4	91979.9	109891.3
8819.0	33837.1	42994.2	51458.9	62266.3	75108.5	91997.1	109993.5
9107.6	33866.1	43005.2	51492.9	62272.1	75300.4	92015.6	110031.8
9176.5	33881.7	43011.1	51646.0	62288.0	75354.2	92087.0	110130.8
9660.4	33901.9	43039.2	51239.6	62294.3	75382.1	92126.8	110407.6
9953.8	33914.9	43082.1	52199.0	62352.1	75495.9	92252.1	110513.3
9975.3	33945.3	43092.0	52287.1	62379.3	75579.7	92278.3	110575.1
10007.6	33998.3	43123.5	52347.7	62387.0	75601.0	92353.9	110631.2
10037.1	34014.5	43168.4	52390.5	62411.4	75622.8	92465.7	110806.0
10059.8	34030.7	43211.1	52422.8	62487.5	75665.2	92555.4	110861.1
10095.4	34061.9	43229.2	52451.0	62934.5	75684.5	94134.1	111120.2
10203.7	34083.9	43238.4	52505.6	63089.5	75685.9	94205.3	111509.8
10257.6	34129.9	43298.9	52535.6	63149.3	75758.2	94392.3	113161.8
11308.5	34141.6	43305.7	52600.9	63169.6	75823.3	94763.1	113189.1
11372.8	34155.2	44449.1	52733.5	63264.8	77460.3	95068.7	113221.9
11383.8	34167.6	44501.1	52744.6	63314.7	77526.3	95415.2	113293.1
11416.1	34203.8	44740.3	52936.4	63335.4	77717.6	95481.7	113721.8
11471.9	34247.3	45009.4	53067.2	63398.9	78410.6	95567.7	113834.8
13007.1	34470.8	45019.3	53165.1	63484.0	78725.7	95743.2	113993.2
13052.3	34638.3	45075.8	53409.9	63534.7	79819.8	96040.9	114149.0
13069.5	34720.5	45096.8	53486.4	63559.7	80264.6	96412.9	114318.4
13097.5	34797.2	45113.4	53631.3	63570.6	81001.0	96722.9	114525.8
14320.8	35617.9	45190.0	53755.8	63656.2	81116.5	97198.0	114602.8
14325.6	35681.1	45219.5	54496.8	63691.0	81199.3	97275.2	114693.9
14350.3	35714.5	45221.4	54591.0	63813.5	81344.8	97312.4	114970.9
15129.9	35746.6	45254.0	54651.4	63822.1	81350.1	97596.2	115349.9
15142.1	35828.4	45267.3	54655.8	63835.0	81356.3	97645.4	115713.6
15641.8	35847.0	45317.6	54669.4	63845.7	81363.3	97716.1	116154.4
24175.1	35861.7	45323.7	54696.5	63855.1	81452.9	97764.0	116402.2
24225.3	35881.0	45433.2	54717.5	63920.7	81581.2	97833.9	116558.2
24294.1	35910.0	45440.6	54726.2	63929.2	81584.6	98188.2	117682.3
24343.3	35919.9	45488.8	54734.5	63938.1	81597.0	98216.5	117768.2
24413.4	35998.1	45535.8	54750.7	63986.6	81600.8	98348.9	117805.7
24816.5	36013.8	45584.8	54860.5	64000.1	81624.0	98452.7	117917.7
24832.1	36065.5	45613.0	54887.3	64015.2	81660.5	98606.4	118507.1
24994.3	36254.5	45637.1	54959.0	64120.4	81774.2	100190.4	120527.5
25227.3	36314.1	45684.5	55015.8	64174.7	81902.5	100214.5	121311.7
25245.7	36327.1	45798.1	55229.0	64187.3	81926.2	100238.7	125235.5
25306.3	36661.8	45991.3	55286.8	64488.9	81937.7	100275.5	125307.6
25341.8	36761.3	46222.5	55457.3	65229.5	81956.6	100369.4	125415.3
25355.1	36765.9	46254.0	55592.4	65258.9	81999.7	100498.0	125558.1
26604.3	36807.1	46391.2	55644.6	65324.5	82004.1	100533.0	125715.9
26802.1	37186.0	46494.5	55714.5	65451.8	82153.2	100597.8	125915.2
26878.6	37190.9	46498.7	55799.6	65575.0	82186.8	100625.0	126290.1
26971.4	37196.9	46534.2	55864.2	65771.3	82198.2	100631.0	126308.0
27002.6	37201.2	46540.4	55904.6	65806.4	82219.3	100640.8	126398.2
27032.3	37226.3	46601.3	55977.1	65837.1	82221.7	100887.3	126555.0
27146.6	37272.9	46622.0	55979.1	65899.8	82418.4	100992.6	126758.8
27166.9	37331.4	46648.2	56034.2	65992.8	82581.5	101557.7	126976.6
27184.1	37358.1	46664.1	56086.8	66125.8	82796.3	101684.1	127213.3
27228.2	37372.1	46695.3	56160.8	66151.6	82981.6	101691.3	127534.3
27254.4	37644.3	46713.1	56192.1	66253.2	83026.7	101719.8	128106.7
27268.1	37705.2	46862.5	56256.2	66321.4	84518.9	101770.4	128208.1
27273.8	37742.6	46871.7	56380.4	66349.5	84579.9	101811.3	128796.7
27284.2	37797.7	46952.9	56537.0	66389.4	84673.6	102011.9	129010.2
27298.5	37812.9	47026.5	56717.6	66395.6	84737.5	102083.2	129489.2
27428.0	37858.0	47074.5	56922.9	66537.7	84747.3	102126.3	130353.2
27736.3	37885.6	47101.5	57187.4	66609.9	84775.6	102160.7	136480.7
27959.1	37919.0	47480.5	57454.8	66679.7	84799.5	102296.0	136804.0
28040.3	37921.5	47664.1	57752.9	67109.1	84807.7	102328.9	137490.9
28261.2	37958.9	47717.9	57759.0	67141.3	84845.6	102557.7	139458.2
28374.4	38109.8	47768.2	57826.4	67174.3	84859.5	102653.5	139693.4
28421.4	38220.8	47780.0	57858.1	67282.4	84971.0	102856.0	140135.1
28445.5	38242.9	47800.7	57890.2	67312.2	85065.8	102877.6	140790.6
28454.0	38332.8	47824.5	57898.6	67319.7	85071.7	102995.1	144170.2

## Supporting Information

28476.2	38349.2	47886.5	59031.0	67365.4	85101.9	103071.7	144382.7
28483.4	38437.5	47895.2	59052.0	67445.0	85138.4	103386.9	144635.2
28494.5	38658.7	47974.5	59068.2	67464.7	85142.0	103558.1	144845.3
28506.3	38745.9	48068.1	59117.2	67526.0	85203.5	103620.9	144922.0
28532.4	38758.0	48198.7	59258.0	67529.4	85238.5	103625.8	146876.8
28575.0	38772.3	48471.5	59335.7	67554.6	85275.1	103774.6	147268.7
28584.4	38781.1	49016.4	59423.7	67647.7	85303.6	103781.8	147414.5
28633.2	38787.8	49022.2	59426.8	67656.6	85312.5	103822.9	147627.1
28637.6	38895.3	49038.3	59480.6	67671.3	85322.5	103830.2	147720.8
28749.5	38914.2	49074.8	59521.2	67699.8	85403.3	103937.4	147885.1
28763.0	39158.9	49225.0	59581.3	67728.8	85409.7	104020.0	150059.4
28789.9	39161.1	49231.4	59648.3	67738.9	85489.8	104067.7	150309.0
28844.6	39162.4	49248.4	59705.7	67792.1	85581.4	104130.3	150623.9
28849.9	39166.6	49307.1	59740.0	67878.3	85622.9	104136.5	150825.1
28928.4	39171.0	49354.2	59823.0	68052.7	85661.9	104158.1	150904.2
28931.9	39185.5	49393.0	59986.7	68274.7	85723.1	104172.4	150945.2
28939.8	39204.8	49393.7	60144.0	68306.9	85747.3	104195.0	150970.7
28955.1	39236.8	49396.8	60286.4	68367.5	85805.3	104303.8	153704.8
29017.9	39276.0	49431.9	60291.0	68481.2	85869.5	104371.9	154137.4
29033.4	39278.0	49472.2	60351.1	68633.6	85904.3	104407.1	154431.3
29052.5	39301.3	49566.0	60414.2	68752.2	86008.9	104569.1	154538.3
29096.3	39324.9	49728.2	60416.8	68857.3	86089.8	104608.6	154581.4
29105.9	39378.5	49906.1	60452.7	69293.4	86105.9	104639.1	178018.3
30068.7	39552.8	49931.5	60469.0	69379.2	86251.0	104687.2	178561.3
30191.8	39591.1	49960.3	60476.3	69597.4	86420.4	104698.4	179729.7
30350.3	39636.8	50100.7	60575.7	69765.8	86687.0	104723.3	181999.6
30448.5	39645.9	50107.3	60596.9	69794.8	86694.8	104776.7	182590.7
30463.4	39665.1	50118.9	60733.7	69830.7	86799.0	104790.5	182806.8
30475.6	39684.6	50203.3	60756.9	69869.8	87368.3	104798.7	182819.1
30514.1	39685.1	50251.5	60779.7	70068.6	87636.7	104947.9	183094.8
30553.9	39977.0	50276.1	60791.5	70089.6	89257.0	104997.1	183110.0
30562.9	40024.2	50292.7	60907.2	70110.1	89312.5	105070.7	185582.2
30579.6	40035.7	50292.8	60913.8	70257.3	89384.2	105095.0	185833.5
30661.3	40130.4	50308.2	60953.7	70394.2	89545.2	105117.4	186430.4
30694.2							

**Table S23:** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_6\text{H}_6)_2]^{3+}$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

3							
Spin Orbit States							
NEVPT2							
0.0	31129.4	39697.1	49323.3	58722.8	66949.2	83223.6	98475.6
408.9	31162.7	39733.5	49476.2	58728.1	66976.9	83362.5	98480.8
572.1	31212.1	39818.4	49645.5	58786.0	67018.7	83428.1	98572.2
665.4	31332.9	39840.5	49957.6	58837.0	67160.1	83569.0	98584.1
779.1	31470.5	39844.4	50056.8	58894.8	67262.2	83591.2	98653.5
927.6	31551.7	39848.7	50148.0	58897.4	67349.9	83696.7	98697.5
1070.8	31587.3	39895.9	50230.1	59024.2	67444.8	83814.6	98748.9
1165.9	31632.4	39939.0	50245.3	59055.0	68077.6	83977.3	98758.6
3280.1	31685.2	39954.0	50324.5	59104.6	68355.3	84066.2	98803.9
3354.9	31692.1	39962.8	50335.4	59171.0	68406.9	84192.5	98828.4
3629.0	31725.5	40019.9	50348.9	59182.8	68470.8	84211.9	99061.1
3845.5	31758.3	40044.5	50385.2	59269.7	68657.2	84239.6	99381.0
3946.8	31818.3	40051.1	50398.8	59285.9	68660.9	84276.7	99447.9
3975.6	31845.3	40061.4	50482.2	59297.5	68831.3	84292.2	99522.5
4024.6	31906.7	40072.0	50482.5	59325.9	68878.2	84371.0	99687.6
5559.5	31924.6	40108.8	50489.3	59331.9	68932.0	84521.2	99950.5
5604.8	31951.5	40138.5	50524.1	59378.8	68981.0	84547.8	100115.9
5885.8	31973.4	40180.3	50713.0	59442.1	69120.4	84558.7	100269.2
6068.7	32022.1	40189.7	52270.3	59539.1	69120.7	84617.1	100462.0
6103.8	32031.5	40625.1	52272.4	59621.9	69748.8	84628.4	100484.0
6150.9	32258.8	40689.3	52289.4	59671.4	70667.6	84697.5	100537.3
7325.1	32328.8	40803.4	52294.7	59792.6	70822.2	84735.8	100577.4
7380.2	32511.9	40818.8	52309.1	59814.8	70974.1	84757.8	100591.1
7661.2	32573.3	40823.2	52312.0	59872.7	71059.4	84823.2	100611.0
7749.0	32591.8	40836.5	52369.3	59897.6	71277.4	84935.6	100627.1
7823.6	32615.1	40916.3	52374.6	59913.0	71384.8	85053.8	100683.9
8634.3	32641.4	40943.0	52391.9	59976.2	71411.4	85187.2	100703.1
8705.8	32675.5	40989.8	52396.4	59988.2	71612.5	85350.5	101203.4
8720.2	32934.7	41025.3	52445.7	59989.3	71814.0	85370.2	101273.6
8745.3	32975.6	41058.3	52452.1	60023.5	71840.1	85392.1	101375.7
8809.3	33167.9	41090.1	52529.3	60038.5	71882.1	85460.7	101392.7
8827.5	33298.6	41103.0	52531.0	60086.6	71885.5	85525.5	101402.4
8871.7	33337.1	41134.3	52601.8	60100.7	71976.9	85573.5	101416.7
8994.4	33351.4	41625.2	52623.0	60107.0	71981.6	85713.6	101443.9

Supporting Information

9057.6	33368.2	41789.3	52626.9	60149.9	72033.7	85722.4	101566.5
9158.2	33465.6	41936.1	52644.5	60159.8	72092.9	85950.9	101739.8
9605.2	33481.6	42450.0	52704.5	60197.1	72339.2	86991.1	101961.0
9960.8	33639.1	42635.9	52708.2	60218.7	72399.3	87058.7	102373.4
10110.5	33698.8	43750.4	52718.7	60255.6	72406.6	87249.9	103726.9
10215.7	33791.0	43762.4	52746.4	60337.3	72432.9	87647.8	103747.2
10235.9	34019.2	43808.1	52780.5	60349.6	72433.6	87974.9	103793.7
10262.0	34207.3	43815.7	52819.1	60351.8	72509.5	88985.2	103899.4
10275.9	34268.6	43934.3	52985.5	60374.4	72522.5	89040.2	104872.1
10328.1	34279.4	43947.9	52994.7	60469.8	72556.2	89100.2	104964.1
11810.2	34313.1	44041.4	53090.1	60483.2	72586.0	89254.7	105122.4
11878.7	34322.2	44064.3	53115.7	60495.4	73370.1	89377.7	105142.2
11892.0	34340.3	44070.4	53206.9	60552.6	73579.2	89511.5	105219.1
11934.6	34393.8	44093.5	53218.3	60665.2	74759.9	89578.0	105243.5
13092.8	34407.8	44295.7	53228.6	60695.2	74864.9	89586.8	105364.7
13104.8	34448.0	44370.2	53234.2	60785.2	74915.1	90003.4	105499.5
13133.0	34515.1	44409.0	53251.6	60822.3	75111.2	90016.8	105648.6
13861.7	34720.0	44428.3	53269.7	60860.0	75284.9	90240.7	105897.3
13872.0	34745.7	44457.5	53307.9	60901.7	75288.4	90295.7	106216.3
14356.7	34833.4	44594.6	53449.8	60927.1	75334.7	90434.8	106834.5
23591.2	34884.6	44645.7	54550.4	60979.7	75343.5	90598.1	107144.0
23642.5	34952.0	44703.9	54635.0	61013.1	75347.1	91108.4	107319.3
23699.2	35071.5	44868.9	54705.0	61080.1	75371.0	91184.1	108420.8
23713.1	35141.0	45331.2	54746.5	61684.3	75400.6	91265.8	108484.1
23902.3	35310.9	45386.1	54815.2	61706.8	75445.4	91557.5	108505.8
24629.9	35400.2	45428.2	54844.9	61749.1	75452.7	92058.1	108606.2
24668.4	35410.6	45462.0	54855.3	61752.9	75473.4	92419.1	108648.2
24854.7	35757.6	46153.9	54881.0	61987.3	75522.4	92649.2	110573.3
25106.8	35853.4	46216.0	54891.9	62112.9	75645.2	92973.9	111336.6
25132.1	35913.1	46234.5	54903.9	62132.3	75679.3	93051.2	116453.5
25169.3	36006.5	46278.9	54940.3	62156.5	75717.0	93069.3	116524.7
25204.4	36043.1	46283.5	54978.5	62189.6	75829.5	93156.9	116631.7
25241.4	36068.7	46301.3	55030.7	62257.5	75868.0	93188.7	116770.5
25259.3	36083.5	46343.6	55174.0	62388.6	75888.6	93261.1	116920.0
25317.6	36093.0	46395.4	55227.5	62432.7	75913.8	93324.7	117111.5
25417.0	36101.2	46395.8	55365.4	62524.0	75919.1	93531.3	117276.4
25501.1	36219.5	46414.6	55382.2	62542.3	75930.5	93740.9	117471.7
25611.6	36249.8	46444.3	55404.5	62570.3	76083.4	93901.1	117579.7
25707.9	36259.6	46489.1	55429.3	62586.8	76085.2	93994.2	117581.8
27136.3	36293.2	46576.2	55432.0	62684.8	76164.4	94016.0	117738.0
27274.6	36309.3	46586.1	55474.0	62716.8	76174.0	94050.0	117878.8
27486.3	36341.5	46598.2	55480.4	62813.7	76234.6	94110.5	117941.1
27670.2	36412.6	46645.2	55631.2	62904.0	76267.1	94141.9	118098.4
27678.4	36501.7	46669.0	55756.7	62948.9	76278.5	94352.8	118152.8
27766.1	36587.7	46699.8	55801.0	63093.3	77789.1	94390.4	118380.0
27806.8	36615.8	46806.2	55842.0	63191.4	78217.2	94408.3	118547.4
27877.3	36745.1	46821.3	55864.3	63283.1	78382.2	94441.1	118711.2
27910.3	36748.6	46903.3	55906.4	63308.1	78615.1	94593.5	119365.4
27988.2	36766.4	46911.0	55986.5	63320.1	78677.3	94671.6	119428.3
28125.1	36789.4	46987.5	56168.6	63371.8	78785.5	94693.8	123926.1
28266.8	37339.4	46993.8	56189.8	63429.1	78939.3	94708.0	124230.1
28294.2	37413.2	47033.6	56278.1	63506.0	78942.5	94723.6	124934.8
28382.2	37418.1	47109.1	56389.7	63604.6	78953.2	94789.6	126931.4
28415.5	37423.3	47207.2	56452.9	63731.6	79544.5	95017.3	127178.0
28469.4	37473.8	47282.5	56460.5	63747.0	79689.7	95034.5	127608.2
28535.3	37496.5	47337.2	56483.8	63777.3	79725.8	95500.5	128316.2
28689.2	37637.0	47357.9	56526.0	63806.7	79821.3	95515.6	132364.5
28854.3	37646.5	47392.2	56574.8	63823.6	80045.2	95623.4	132572.8
28892.7	37681.0	47408.1	56650.0	64146.0	80048.3	95629.5	132875.3
28930.1	37687.3	47445.7	56656.2	64268.4	80198.9	95662.6	133114.9
29012.0	37706.7	47457.1	56686.2	64291.7	80368.0	95934.6	133184.7
29119.2	37894.5	47484.6	56714.3	64370.3	80411.1	95951.9	134410.7
29197.3	37906.4	47502.1	56730.9	64469.0	80442.5	96098.9	134915.8
29238.0	37932.7	47523.8	56735.9	64477.6	80507.4	96205.1	135114.2
29270.9	37951.1	47526.6	56761.7	64536.1	80508.2	96513.5	135523.3
29296.4	38019.0	47530.2	56825.4	64559.3	80542.8	96665.1	135935.9
29350.9	38067.0	47580.8	56864.4	64577.0	80599.4	96728.7	136037.6
29378.6	38158.2	47604.4	56903.9	64605.5	80684.0	96767.3	137133.0
29389.0	38170.9	47614.9	56925.1	64654.7	80761.8	96775.4	137789.1
29409.3	38194.8	47627.0	56944.6	64675.0	80917.5	96808.4	138188.5
29480.9	38218.4	47651.0	57057.7	64822.5	80999.2	96869.9	138431.2
29604.4	38233.8	47735.1	57123.1	64833.2	81261.9	96918.7	138508.4
29651.8	38265.4	47763.8	57210.2	64974.8	81559.9	96976.5	138551.2
29718.8	38277.9	47767.3	57242.8	65015.7	81620.6	96996.4	138579.7
29784.3	38293.3	47771.6	57328.9	65046.0	81699.8	97147.9	141235.8
29888.8	38393.1	47818.4	57360.7	65089.6	81745.0	97219.1	141749.8
30104.8	38485.5	47904.7	57386.1	65103.6	81821.0	97247.9	142075.2

## Supporting Information

30259.8	38494.7	47969.5	57698.3	65131.3	81927.3	97250.8	142176.6
30276.0	38516.6	48041.4	57745.8	65222.4	82052.3	97344.1	142225.3
30291.4	38658.4	48051.1	57925.1	65274.8	82149.8	97379.9	161547.1
30308.9	38704.9	48073.9	58359.3	65316.9	82269.1	97478.5	162031.9
30338.1	38750.0	48170.1	58409.3	65444.2	82377.1	97528.4	162986.4
30371.9	38840.2	48188.5	58459.8	65527.6	82429.8	97624.0	165174.4
30427.4	38877.9	48257.4	58513.1	65591.4	82451.2	97691.7	165456.9
30457.0	38913.5	48271.9	58517.4	65750.8	82580.3	97766.7	165501.8
30519.3	39002.7	48334.3	58525.3	65955.3	82615.4	97839.0	165985.4
30650.9	39494.7	48392.6	58530.0	65985.3	82643.2	97905.8	166237.1
30685.1	39530.3	48443.9	58552.6	66002.2	82804.8	97928.8	166494.7
30732.6	39586.1	49161.4	58575.4	66080.7	82887.3	98023.3	168352.7
30904.0	39633.3	49191.3	58603.0	66126.8	82941.3	98156.6	168642.7
30949.0	39666.9	49234.0	58655.1	66193.3	83067.2	98321.2	169558.4
31062.4							

**Table S24:** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_7\text{H}_7)_2]^{3-}$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

4					
Spin free States					
CASSCF					
0.7	36523.1	46708.6	56720.8	79067.6	97205.8
122.2	36536.0	46708.6	56720.9	79082.9	97216.6
122.2	36536.1	46912.6	56764.2	79082.9	97216.7
213.0	36699.9	46912.6	56766.2	79092.5	97531.3
223.3	36702.2	46915.3	56919.4	79092.5	97856.7
364.6	36783.8	46915.4	56919.7	79232.3	97886.2
384.4	36790.0	47092.8	57014.4	79232.3	98113.7
410.1	36824.0	47095.0	57014.4	79403.6	98114.8
1081.8	36829.0	47189.1	58342.8	79403.6	100408.4
1081.8	36873.3	47200.0	58342.8	79541.3	100408.4
7403.6	36918.1	47234.5	58554.1	79541.3	100427.5
7409.6	36918.2	47387.8	58609.9	79692.9	100427.6
7653.7	36950.2	47387.8	58619.9	79692.9	100467.0
7653.9	36953.5	47698.9	58634.6	79750.8	100467.1
7778.9	36953.5	47705.7	58634.8	79752.0	100498.5
7781.8	37128.7	47728.2	58701.6	79764.8	100500.7
8002.7	37383.4	47764.3	58701.7	79779.3	100514.6
34094.4	37383.6	47765.1	58703.9	79779.6	100522.2
34124.4	37452.8	47838.7	58707.1	79803.9	100524.7
34687.2	37456.4	47839.2	59109.7	79804.1	100570.8
24125.9	37582.4	47839.8	59110.3	79814.0	100570.8
24126.1	37595.4	47839.8	59366.1	79814.3	101855.6
24130.4	37882.1	47939.0	59376.0	79835.2	101855.6
24134.2	37882.1	47939.0	59646.1	79835.2	101911.0
24183.5	37883.1	48050.0	59708.3	84132.8	101911.1
24183.7	37883.1	48050.0	59803.3	84134.7	102188.5
24249.8	38049.3	48091.8	59803.6	84547.4	102188.7
24249.8	38049.5	48091.8	59821.0	84553.6	102229.1
24376.8	38130.5	48144.8	59827.0	84575.0	102229.2
24376.8	38130.5	48144.8	60052.3	85062.5	102445.2
24394.8	38268.2	48176.5	60056.3	85143.5	102451.3
24402.3	38268.7	48176.5	60943.8	85143.9	102547.4
24451.8	38431.5	50565.0	60943.8	85212.2	104639.0
24451.8	38440.9	50565.0	61433.1	85218.2	104639.2
24617.9	38496.9	50702.0	61433.1	85517.3	104665.9
24645.7	38707.0	50785.5	61458.4	85520.2	104666.0
24645.7	38707.0	50791.3	61487.7	85553.9	104766.6
24687.4	41341.0	50996.2	61503.9	85554.1	104767.1
24693.5	41341.0	51003.3	61591.3	86276.7	105009.0
24736.7	41414.0	51249.9	61591.4	86276.7	105014.4
24736.7	41414.0	51250.1	61666.8	86548.9	105139.5
24799.4	41438.6	51282.6	61666.8	86548.9	105775.9
24799.5	41448.8	51282.6	61699.4	86600.2	105776.1
24826.2	41488.1	51439.5	61699.5	86600.2	105966.0
24827.0	41530.6	51439.5	61747.5	86788.4	105966.0
24966.8	41530.6	51720.5	61747.5	86788.4	106168.2
24992.3	41628.4	51720.6	63188.5	86923.2	106176.5
24992.7	41628.4	51724.7	63378.1	86923.8	106525.8
24996.3	41680.3	51724.7	63378.6	86966.5	106545.9
24997.6	41680.3	51779.0	63408.2	86973.2	106681.9
25011.9	41700.7	51779.0	63408.2	86977.0	109677.2
25012.3	41700.7	51830.8	63537.7	86998.8	109689.6
25180.4	41712.5	51830.8	63539.9	87000.1	109691.9
25182.1	41712.5	51837.5	66224.3	87243.3	109708.4



## Supporting Information

25202.1	41733.6	51837.6	66758.8	87243.4	109715.8
25202.1	41733.8	51935.8	66758.8	87247.4	110068.2
25296.6	41773.8	51936.7	66771.2	87294.1	110068.5
25302.2	41775.4	52009.1	66800.0	87311.5	113964.2
25366.4	41822.2	52013.8	67118.1	87416.7	113989.0
25660.2	41839.1	52039.9	67119.6	87418.3	114505.5
25843.5	41862.5	54435.6	67130.6	87479.6	119074.2
25855.6	41979.9	54441.2	67152.5	87479.7	119074.2
25895.7	42144.9	54464.8	67152.6	87504.3	119498.8
25896.3	42149.4	54464.9	67163.0	87504.7	119498.8
25985.7	42198.8	54474.9	67163.9	92803.6	119699.9
25985.7	42198.8	54475.1	67241.5	92805.1	119699.9
26069.0	42277.1	54645.0	67241.7	92940.9	119803.5
26069.1	42277.5	54648.0	68954.4	92946.0	119803.7
29157.1	42306.2	54751.8	68954.4	92989.3	119845.7
29159.5	42365.5	54938.8	68989.6	92989.5	119850.8
29161.3	42369.3	54938.8	68989.6	93055.1	119859.2
29161.5	42378.4	55178.5	69002.8	94669.5	119873.5
29165.1	42381.8	55198.9	69002.8	94673.5	119874.3
29173.7	42544.5	55208.7	69006.3	94678.3	119890.1
29175.3	42545.5	55217.4	69006.8	94778.2	119890.3
29181.3	42655.2	55217.4	69018.5	94779.2	122066.0
29181.5	42655.2	55226.9	69021.9	94825.8	122070.8
29190.6	42693.1	55226.9	69030.7	94825.8	122085.9
29190.6	42695.6	55256.3	69030.7	94960.2	122516.6
29194.8	42766.2	55257.7	69037.6	94960.2	122518.2
29194.9	42766.3	55307.5	69038.2	94997.5	131484.0
29319.2	42869.3	55307.7	69044.6	94997.5	131484.3
29319.2	42869.5	55313.3	70304.2	95522.0	131488.3
30338.2	42964.8	55313.3	70309.5	95522.0	131488.6
30338.2	42964.8	55346.9	70555.8	95829.6	131979.9
30489.7	42975.3	55346.9	71980.1	95844.3	131999.6
30497.4	42975.3	55735.1	72045.2	95845.7	132307.7
30498.8	43102.1	55735.1	72071.5	95857.4	139298.2
30529.0	43103.9	55742.0	72252.4	95862.6	139321.9
30532.4	43104.0	55742.0	72581.8	95870.1	139337.4
30601.5	43112.5	55785.4	72585.0	95870.1	139481.7
30601.7	43114.2	55816.3	72604.8	95958.2	139484.7
30708.4	43114.2	55819.3	72605.0	95961.8	139662.6
30708.5	43354.6	55938.3	72692.0	95965.0	139662.9
30735.7	44156.7	55941.4	72695.9	95966.5	139739.8
30735.7	44156.8	55971.3	75976.6	95975.8	139739.9
30812.3	44164.3	56035.4	76035.4	95997.1	141472.2
30812.3	44164.3	56044.3	76035.4	96002.2	141997.6
30841.5	44308.1	56106.9	76144.9	96057.3	142020.9
30841.5	44308.1	56107.2	76145.2	96057.3	145305.5
32241.4	44326.1	56126.9	76157.9	96061.5	145305.7
32243.0	44326.3	56136.1	76158.0	96061.7	145344.4
32454.5	44377.9	56158.8	76198.4	96084.4	145344.4
32458.3	44378.0	56158.9	76203.9	96084.4	145545.3
32759.1	44412.9	56182.5	76206.4	96123.4	145550.0
33812.7	44418.2	56182.5	76216.2	96123.4	145855.2
33857.8	44461.1	56196.7	76222.6	96197.2	145867.8
33861.8	44461.1	56196.7	77536.0	96197.3	145993.7
33902.4	44492.7	56209.9	77538.3	96723.7	146093.7
33907.9	44494.5	56210.4	77579.6	96723.7	146093.7
33926.8	44525.9	56290.8	77593.0	96789.8	173814.5
33927.1	44525.9	56290.8	77733.7	96834.2	173821.5
33958.1	44554.7	56471.1	78100.5	96852.5	174364.3
33958.1	44555.5	56493.5	78100.8	96971.9	174644.6
33988.8	44556.8	56510.0	78891.2	96974.8	174664.6
33988.8	44710.8	56574.1	78927.6	97050.5	176846.3
36263.7	44727.8	56592.0	78929.2	97050.5	176849.6
36263.7	44736.0	56594.6	78972.0	97117.3	176953.0
36292.6	44793.7	56625.7	78972.0	97117.5	176953.6
36292.6	45423.0	56626.6	79004.9	97172.2	177182.9
36370.8	45430.3	56627.0	79007.3	97172.4	177255.9
36370.8	45605.0	56643.6	79067.5	97205.8	177277.0
36523.1	45613.1				

**Table S25:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_7\text{H}_7)_2]^{3-}$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

4
Spin free States
NEVPT2

Supporting Information

1.9	35026.7	44774.7	54677.3	74698.3	90858.2
71.4	35036.4	44774.8	54680.9	74858.9	90858.3
71.5	35053.8	44858.8	54775.5	74859.0	90968.3
310.6	35231.1	44872.3	54778.8	74884.2	91076.0
330.0	35237.4	44910.8	54814.7	74884.2	91102.5
396.3	36837.1	44947.6	54814.7	74925.5	91102.6
435.8	36837.1	44947.6	54831.4	74925.6	91135.9
449.9	36946.3	44997.5	54831.4	75247.5	91136.0
1575.2	36946.3	44997.5	55400.9	75315.1	93015.9
1575.3	36989.5	48796.8	55401.0	75326.7	93016.2
6180.8	36989.6	48796.8	55463.9	76178.3	93047.0
6192.1	37259.7	48800.2	55468.1	76178.3	93047.9
6538.7	37259.7	48800.2	55595.2	76395.7	93084.7
6539.3	37346.0	49013.3	55708.6	76395.7	93104.5
6738.8	37346.5	49133.5	55720.6	76589.7	93138.6
6743.5	37355.3	49138.7	55742.6	76589.7	93151.1
7023.5	37355.9	49421.6	55742.6	76735.3	93154.3
28765.3	37387.9	49434.3	55809.7	76735.3	93166.9
28797.8	37388.4	49747.0	55820.7	76789.9	93171.6
29621.8	37509.8	49747.0	55832.3	76790.0	93308.2
22835.7	37514.9	49780.4	55833.1	76808.1	93308.3
22871.4	37592.4	49780.9	55850.8	76808.4	93841.0
23294.0	37592.4	49841.9	55852.5	76813.2	93841.2
23310.9	37730.4	49841.9	56216.6	76814.5	93876.1
23564.1	37740.3	50060.2	56648.4	76828.6	93876.6
23582.0	37799.7	50060.2	56708.4	76835.2	94107.6
23582.4	37799.7	50157.5	56711.0	76845.3	94113.3
23607.4	37822.4	50172.1	56712.0	76854.9	94174.2
23794.0	37901.4	50194.7	56715.1	76857.5	94175.4
23802.2	37901.4	50245.4	57042.0	78418.6	94235.9
23819.4	37942.3	50250.3	57047.8	78573.1	94237.5
23826.9	37943.7	50364.5	57269.1	78574.7	94268.8
23844.4	38200.3	50365.5	57269.2	78746.6	94285.4
23975.9	38213.7	50375.9	57913.0	78763.6	94343.5
24259.4	38297.8	50377.6	57913.0	78960.7	94343.6
24264.6	38507.0	50522.8	57917.7	78960.7	94604.8
24264.8	38519.0	50522.8	57970.2	79135.2	94609.4
24386.0	38542.9	50697.7	57988.2	79135.3	94620.5
24388.1	38584.1	50697.7	58133.0	79223.7	94623.5
24389.4	38584.1	50756.7	58133.5	79240.2	94743.6
24403.2	39214.5	50756.7	58248.5	79802.0	94804.3
24496.1	39384.0	50761.0	58248.5	79893.5	96093.5
24496.4	39388.1	50761.0	58314.5	79894.5	96096.1
24547.9	39625.5	50836.7	58315.2	79983.5	96494.3
24635.8	39625.8	50836.7	58391.2	80017.6	96494.5
24685.3	39725.4	50844.0	58391.3	80195.4	96725.5
24685.4	39754.5	50844.0	58501.2	80203.3	96755.6
24751.1	40245.9	50890.8	58871.9	80241.0	97123.1
25047.9	40246.7	50890.8	58873.9	80241.8	97248.0
25981.1	40557.1	50935.3	58885.8	80310.8	97440.3
25986.4	40558.1	50935.3	58886.5	80313.1	100297.8
25986.6	40891.3	50951.2	59100.7	80351.7	100298.0
26043.5	40908.9	50952.1	59105.4	80357.2	100309.1
26043.6	41055.1	50956.8	59989.3	80665.2	100353.2
26073.0	41142.1	50957.4	60805.0	80665.5	100365.4
26361.3	42134.6	51133.1	60836.6	81058.2	100894.8
26361.3	42146.4	51133.3	62811.8	81058.2	100895.2
26640.9	42392.6	51283.0	62811.9	81259.9	103814.9
26643.2	42400.8	51286.6	63332.9	81277.0	103840.7
26848.4	42419.4	51339.7	63344.8	81343.9	104539.7
26848.4	42419.4	51662.2	63349.2	81348.6	109755.2
27318.3	42426.3	51672.9	63372.4	81352.6	109755.4
27326.9	42426.9	51792.2	63373.0	81386.5	110363.8
27327.7	42452.3	51792.4	63394.8	81386.8	110363.8
27430.8	42452.3	51853.7	63395.1	84637.9	110597.5
27430.8	42571.4	51854.5	63512.5	84644.2	110597.6
27439.8	42571.7	51980.9	63513.2	84812.7	110734.7
27439.8	42627.9	52014.1	65205.8	84826.7	110741.6
29401.5	42632.3	52036.2	65261.6	84885.0	110749.5
29404.1	42632.3	52036.4	65273.0	84885.4	110750.9
29405.7	42640.8	52036.4	65277.4	84973.6	110770.5
29408.3	42641.7	52057.0	65286.7	87097.5	110818.8
29410.4	42642.7	52059.4	65678.7	87104.8	110826.7
29431.9	42726.6	52157.5	65832.6	87144.1	110870.0
29433.9	42730.3	52174.7	65832.6	87267.4	110877.3
29445.7	42816.9	52298.1	65933.6	87290.3	110877.8
29452.1	42981.9	52604.7	65933.6	87294.4	110902.1

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29492.7	42982.0	52614.3	65949.4	87298.2	110903.1
29492.8	42996.9	52614.5	65951.4	87343.7	111392.3
29521.8	42999.2	52640.1	65952.1	87344.1	111417.8
29521.9	43267.0	52657.9	65953.0	87601.3	118427.7
29583.2	43274.0	52693.4	65957.5	87601.9	118431.3
29585.3	43341.5	52740.3	65967.1	87662.3	118431.6
29690.0	43342.3	52836.1	65969.1	87663.6	118445.7
29690.0	43361.6	52836.4	65975.4	87833.7	119179.9
29897.2	43456.1	52952.3	65975.5	87868.8	119202.4
29905.6	43456.3	52953.8	66007.3	87871.3	119714.1
30372.8	43538.0	52978.4	66007.4	87881.0	127110.1
31161.3	43627.5	52979.3	66067.2	87885.3	127119.8
31161.4	43627.5	53518.1	66067.7	87923.7	127138.2
31368.6	43634.9	53523.6	66242.1	88264.7	127329.1
31371.3	43635.0	53570.4	66750.5	88422.3	127331.3
31372.9	43726.2	53570.4	66756.8	88466.3	127615.7
31403.6	43742.8	53695.0	66974.4	88724.5	127617.8
31408.7	43776.9	53712.9	66981.0	88724.5	127736.0
31507.9	43776.9	53761.7	68559.5	88728.2	127740.1
31508.1	43793.8	53761.7	69798.7	88738.8	127907.6
31685.0	43793.9	53776.4	69799.5	88739.3	128637.8
31685.0	43801.7	53777.2	69836.6	89457.8	128699.3
31758.6	43801.7	53877.5	69837.0	89457.9	132334.6
31758.7	43827.5	53882.1	69925.8	89483.7	132340.4
31862.6	43827.5	53937.4	69925.9	89483.7	132389.9
31862.6	43835.1	53937.4	69937.0	89548.3	132390.7
31910.1	43835.1	53956.7	69948.5	89662.0	132595.3
31910.5	43841.7	53956.7	69952.9	89875.0	132653.0
32549.1	43842.3	53976.2	69991.6	89875.1	133059.3
32661.2	43950.1	53976.3	69999.8	89886.7	133108.4
32669.0	43971.2	53987.7	70099.9	89956.0	133268.5
32717.8	44029.6	54002.4	70102.2	90009.8	133378.4
32728.1	44035.3	54036.8	70105.2	90052.8	133444.1
32772.2	44057.2	54038.1	70120.5	90054.7	156464.4
32772.9	44177.4	54091.9	70450.6	90211.8	156503.3
32859.3	44177.4	54091.9	70994.6	90212.5	157379.2
32859.3	44449.4	54186.9	70995.6	90262.7	157591.3
32933.3	44449.6	54187.0	74420.9	90276.0	157598.7
32933.3	44482.4	54269.9	74509.6	90370.6	158989.3
33741.1	44482.7	54416.2	74511.6	90378.6	159018.7
33747.3	44505.4	54416.9	74613.4	90378.7	159062.3
33749.0	44505.7	54416.9	74613.4	90491.0	159070.1
34385.2	44713.3	54424.8	74648.0	90498.1	159211.9
34392.2	44713.4	54455.3	74651.8	90736.3	159719.2
34645.0	44736.7	54459.1	74697.5	90737.5	159830.3
35025.7	44740.4				

**Table S26:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_7\text{H}_7)_2]^{3-}$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

4							
Spin Orbit States							
CASSCF							
0.0	30318.5	39766.7	49776.1	60362.0	69812.1	88860.5	104255.4
34.0	30362.9	39771.7	49817.9	60407.2	69832.7	89241.5	104266.5
59.6	31215.7	39843.6	49833.5	60417.6	70117.1	89450.6	104273.6
148.1	31220.9	39891.3	49963.6	60423.1	70406.0	89514.2	104847.7
225.4	31369.1	39935.5	50036.4	60458.5	70545.6	89519.5	105199.3
270.9	31381.1	39984.5	50118.5	60465.1	70625.1	89610.4	105295.1
272.6	31392.5	40010.6	50183.7	60839.3	71356.9	89643.4	105313.8
965.2	31459.9	40040.5	50198.9	60880.0	71774.0	89663.7	105381.5
3132.2	31489.8	40097.2	50238.3	60902.7	71801.3	89808.9	105632.6
3141.5	31491.3	40149.6	50266.0	60911.5	71834.9	89848.3	105647.1
3163.2	31534.2	40154.0	50274.2	60940.3	71921.3	89942.3	105683.0
3192.6	31550.0	40189.8	50284.9	60959.2	71948.4	90106.5	105735.7
3201.9	31567.2	40264.5	50308.1	60991.5	72429.8	90231.2	105823.9
3286.8	31570.3	40268.0	50310.8	61072.2	73728.7	90268.8	106776.8
3724.0	31713.0	40314.9	50334.2	61135.0	73800.9	90352.2	106809.5
5318.2	31767.1	40340.2	50339.3	61248.4	74087.6	90558.0	106858.0
5339.9	31793.7	40381.9	50392.9	61276.9	74283.5	90624.4	106863.4
5386.9	31815.3	40440.3	50445.5	61298.1	74291.4	90679.2	106896.0
5464.8	31914.4	41411.7	50481.8	61315.9	74311.3	90788.5	106928.5
5532.7	32209.3	41495.8	50502.8	61323.7	74315.3	90842.4	106951.8
5774.3	32228.7	41575.2	50513.0	61366.9	74323.1	90937.4	108770.7
7007.7	32337.7	41639.0	50563.0	61425.2	74331.6	91071.9	108910.7
7065.6	32433.3	41654.3	50607.0	61459.2	74342.4	91098.0	108917.6

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7138.2	32481.1	42179.9	50616.6	61465.8	74343.9	91120.8	108923.0
7224.9	32862.9	42231.0	50642.2	61490.6	74356.7	91152.0	108945.9
7430.5	33015.5	42245.1	50691.0	61491.7	74363.8	91172.4	109016.4
8337.4	33120.6	42291.9	50861.1	61505.8	74370.6	91188.0	109060.2
8393.9	33132.0	42315.6	50869.1	61508.0	74412.8	91217.4	109124.0
8478.8	33222.3	42343.5	50980.8	61534.2	74522.6	91219.2	109214.9
8782.2	33299.6	42374.8	50992.8	61592.1	74543.9	91238.9	109357.3
9288.4	33309.0	42409.6	51182.2	61611.3	74632.3	91254.3	109408.8
9412.4	33356.9	42447.7	51541.1	61626.6	74682.5	91260.9	109582.1
9420.8	33361.1	42455.7	51701.5	61658.8	74937.7	91281.1	109776.5
9452.3	33368.2	42528.0	51747.2	61730.7	74953.5	91375.4	109793.5
9556.3	33414.4	42550.5	51835.7	61749.6	74958.4	91516.4	109899.5
9577.3	33422.1	42610.2	51898.2	61770.4	74959.2	91518.3	110000.6
9682.0	33549.6	42655.0	52022.7	62494.7	74978.6	93341.3	110266.3
9765.3	33559.0	42754.8	52210.0	62496.2	74982.7	93820.3	110419.4
9910.7	33644.8	42808.8	52240.6	62577.3	75008.5	93839.3	112363.7
10814.9	33649.1	42867.7	52278.3	62728.0	75066.8	93888.0	112367.1
10919.0	33673.7	43860.8	52284.2	62748.0	76781.7	93932.6	112426.4
10959.7	33744.6	43952.7	52358.5	62754.7	76927.7	94665.1	112486.4
10965.2	33818.4	44215.1	52456.1	62755.5	77006.8	94975.7	112984.2
10990.2	33855.9	44369.6	52535.7	62776.2	77759.6	95181.3	113040.0
12500.1	33910.4	44521.9	52558.2	62787.4	77843.5	95295.5	113141.6
12510.8	34090.0	44568.0	52613.8	62844.6	79219.1	95341.0	113224.0
12626.6	34129.7	44593.3	52633.2	62914.7	79437.7	95355.6	113418.8
12707.7	34321.3	44618.8	52640.9	62944.5	80290.9	95374.7	113543.1
13778.3	35086.1	44635.5	52762.2	62987.5	80445.0	96580.0	113928.8
13850.9	35165.8	44644.8	53713.2	62991.5	80527.3	96795.4	113949.6
13933.2	35214.1	44667.5	53777.6	63068.0	80596.5	96808.3	114191.2
14654.4	35273.6	44696.5	53876.7	63072.2	80737.9	96920.0	114434.8
14702.4	35292.3	44715.4	53996.1	63096.8	80760.5	96923.8	114544.4
15175.4	35302.7	44739.1	54014.1	63209.6	80774.3	96999.3	115512.5
23413.7	35315.4	44826.1	54032.3	63216.8	80777.3	97154.8	115522.1
23456.7	35321.0	44827.3	54072.0	63272.5	80780.3	97211.4	115716.7
23731.3	35349.0	44892.6	54178.1	63279.5	80781.4	97249.7	116635.1
23854.9	35521.4	44907.1	54194.0	63287.0	80802.4	97328.1	116899.1
24009.6	35586.0	44946.4	54271.6	63363.7	80820.4	97413.3	117085.6
24417.9	35591.1	44989.2	54328.0	63378.0	80984.7	97464.7	117111.9
24492.9	35605.7	45141.7	54454.2	63493.7	80999.1	97509.3	117709.1
24549.6	35663.6	45265.5	54482.6	63514.9	81030.6	99311.9	119820.0
24648.7	35702.0	45266.7	54490.1	63523.0	81099.0	99675.3	120258.4
24654.2	35715.1	45287.1	54580.1	63524.3	81185.5	99686.7	124282.7
24717.2	35978.4	45314.5	54630.4	63577.5	81214.7	99691.2	124666.1
24732.5	36005.5	45330.6	54739.6	64548.9	81249.8	99731.1	124829.8
25065.6	36182.9	45342.8	54956.9	64717.2	81311.3	99763.7	124893.1
26165.8	36254.6	45350.2	55104.1	64729.2	81329.1	99767.2	124902.2
26267.0	36556.9	45764.4	55158.9	64750.4	81363.8	99821.7	124916.7
26301.0	36648.2	45812.3	55163.3	64760.6	81397.0	99846.3	124922.0
26357.6	36709.5	45855.4	55176.0	64906.4	81480.7	99908.2	125394.6
26369.3	36710.6	45863.3	55241.4	65293.8	81595.6	99942.4	125868.3
26381.4	36718.0	45943.8	55304.1	65426.8	81607.8	99998.0	126105.6
26451.0	36729.7	45978.8	55336.6	65480.9	81908.2	100034.3	126229.4
26489.3	36760.5	46135.8	55350.8	65508.9	81920.7	100811.0	126285.3
26547.3	36764.4	46194.4	55571.2	65547.9	81950.8	100854.3	126304.3
26550.4	36785.7	46224.5	55880.4	65556.4	81975.0	100918.9	126323.1
26647.8	36947.9	46235.8	55914.3	65584.0	82333.9	100939.3	126339.0
26660.6	37148.5	46306.8	55961.5	65608.1	83825.0	101022.3	127636.5
26823.1	37270.5	46349.4	55965.9	65642.2	83942.1	101028.5	127661.2
26961.1	37274.2	46365.4	56033.7	65687.5	83966.3	101213.7	128047.5
27036.8	37288.0	46496.6	56121.6	65692.2	84057.8	101333.4	128814.8
27060.3	37303.4	46526.7	56176.9	65786.9	84079.3	101635.8	129251.3
27109.9	37319.9	46605.0	56205.4	65836.3	84140.5	101657.7	135798.5
27496.6	37365.0	46753.0	56240.8	65838.6	84176.7	101719.7	135856.2
27502.9	37375.4	47087.0	56932.7	66388.5	84198.9	101803.6	136362.5
27533.4	37441.1	47103.9	57028.3	66609.8	84199.6	101855.6	138805.7
27606.0	37612.0	47201.5	57129.1	66712.5	84219.3	101856.4	138854.9
27618.4	37653.7	47216.1	57304.8	66723.5	84224.3	101885.4	139222.6
27746.4	37735.3	47262.9	57399.8	66778.0	84237.4	101924.3	139511.9
27779.7	37791.6	47270.1	57462.6	66784.1	84267.3	102175.1	143549.5
27963.0	37825.0	47363.0	58240.1	66790.4	84269.3	102415.7	143614.8
28098.1	37870.1	47382.8	58467.9	66825.5	84284.6	102624.6	143732.8
28102.7	38086.6	47388.9	58579.2	66915.6	84291.7	102830.4	143844.5
28111.4	38107.9	47482.2	58610.1	66938.8	84457.1	102836.6	143892.2
28131.0	38210.3	47570.4	58672.7	66946.9	84469.2	102867.3	146233.1
28208.1	38214.0	47623.3	58680.2	67003.2	84501.1	102987.4	146365.2
28212.6	38214.9	48317.2	58730.0	67004.8	84567.6	103109.0	146495.3
28277.1	38292.0	48374.6	58742.7	67012.5	84591.3	103165.6	146563.6
28296.7	38311.4	48469.0	58827.5	67014.9	84638.7	103169.1	146681.2

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28306.1	38368.8	48572.3	58990.2	67021.6	84696.4	103170.6	146857.8
28308.3	38566.6	48640.0	58991.5	67029.4	84734.4	103172.8	148992.8
28312.2	38581.0	48684.1	59004.0	67068.4	84772.5	103274.1	149422.9
28315.3	38592.9	48756.2	59033.8	67087.0	84866.6	103479.4	149510.4
28322.9	38593.9	48765.1	59066.9	67101.8	84980.8	103511.8	149588.7
28366.6	38619.1	48792.5	59076.3	67125.5	85051.7	103554.2	149870.6
28372.9	38649.3	48794.6	59341.2	67557.6	85105.7	103572.3	150080.4
28396.0	38665.2	48797.2	59606.9	67848.1	85139.2	103591.7	150183.3
28400.3	38690.5	48833.0	59635.3	67876.1	85147.5	103652.3	153011.5
28407.2	38701.2	48851.1	59679.9	67887.4	85206.1	103678.2	153034.4
28450.4	38719.2	48904.3	59724.0	67941.3	85281.5	103705.4	153250.2
28475.8	38745.6	48994.2	59760.8	67993.8	85304.1	103729.8	153491.3
28503.6	38757.5	49044.6	59815.2	68005.7	85321.1	103762.7	153705.7
28572.4	38840.5	49072.3	59841.8	68176.9	85324.0	103784.6	177352.9
29663.9	38951.4	49336.2	59863.6	68767.2	85339.1	103795.2	178001.6
29695.7	39037.5	49391.3	59885.5	68906.0	85343.6	103830.5	178255.6
29722.7	39061.0	49427.5	59893.1	69055.9	85949.2	103924.6	181398.6
29736.2	39097.0	49469.6	59970.0	69184.2	86047.7	103936.9	181512.0
29765.3	39100.2	49500.5	60035.3	69187.0	86081.3	103952.5	181531.7
29813.6	39135.8	49541.5	60222.7	69219.7	86660.6	103996.1	181561.9
29927.9	39141.0	49582.6	60264.1	69362.0	86803.7	104052.3	181645.9
29961.7	39422.3	49617.9	60275.0	69529.3	88511.3	104065.6	182476.6
29969.3	39457.0	49636.2	60284.8	69627.0	88596.2	104120.3	184555.7
30090.3	39513.4	49665.6	60306.5	69691.2	88761.8	104241.5	184589.1
30192.1	39559.6	49716.7	60356.3	69761.2	88837.7	104248.0	184776.0
30249.7							

**Table S27.** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_7\text{H}_7)_2]^{3-}$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

4							
Spin Orbit States							
NEVPT2							
0.0	30634.6	39257.0	48835.2	58260.8	66269.6	82266.3	97566.7
26.2	30653.8	39298.8	48925.3	58302.2	66488.1	82362.3	97593.4
76.0	30671.3	39331.8	48979.3	58330.8	66554.4	82414.5	97639.9
166.8	30785.4	39341.3	49440.3	58380.5	66560.7	82656.1	97667.2
237.1	30822.5	39356.6	49446.0	58403.3	66687.8	82658.8	97701.9
275.1	30938.8	39371.3	49506.8	58418.5	66729.7	82859.5	97761.4
332.1	30956.3	39398.1	49513.9	58429.5	66853.5	82893.9	97774.4
1426.8	31012.8	39420.8	49630.9	58505.1	67475.3	82951.7	97904.5
3119.7	31118.7	39448.3	49641.4	58529.3	67664.3	83017.3	97988.5
3152.8	31317.6	39462.6	49700.1	58537.5	67716.6	83327.1	98016.2
3175.7	31330.8	39492.4	49760.0	58575.9	67886.1	83336.3	98050.2
3243.6	31372.0	39535.3	49824.4	58656.2	68007.4	83384.4	98091.3
3252.1	31428.8	39569.6	49853.7	58744.0	68031.1	83544.9	98116.1
3374.0	31431.7	39570.8	49918.5	58760.1	68049.2	83614.1	98678.5
4091.9	31493.5	39579.8	50010.0	58786.4	68143.8	83711.2	98819.3
5316.7	31520.0	39593.4	50072.8	58814.7	68177.4	83735.4	99000.5
5330.8	31532.1	39645.9	50117.6	58823.5	68220.5	83860.9	99047.7
5354.9	31542.0	39668.6	50309.4	58834.5	68243.8	83876.7	99237.9
5488.6	31606.2	39684.2	51335.3	58901.8	68412.7	83907.8	99307.9
5613.9	31619.8	40134.3	51582.0	58973.4	69061.7	83919.0	99496.8
6060.5	31797.6	40151.1	51621.8	59059.2	70001.1	83974.4	99526.2
6988.7	31874.6	40156.4	51686.8	59130.5	70279.1	84037.7	99558.0
7029.0	31992.0	40287.7	51701.4	59160.7	70339.4	84079.7	99579.3
7104.2	32200.7	40304.6	51804.5	59163.4	70345.1	84082.9	99653.0
7271.8	32314.7	40363.6	51810.6	59189.5	70376.5	84124.4	99662.3
7672.5	32328.8	40387.5	51842.7	59217.4	70566.4	84131.3	99673.1
8224.5	32397.4	40416.1	51848.5	59279.4	70708.0	84173.3	99680.9
8250.3	32444.0	40563.4	51865.6	59305.9	70897.4	84473.9	100284.8
8303.6	32487.2	40590.5	51923.3	59317.5	71193.6	84513.9	100288.8
8317.1	32514.8	40603.2	51931.4	59389.8	71261.8	84525.2	100316.2
8384.4	32692.2	40699.7	51960.7	59426.2	71274.2	84557.6	100370.5
8433.5	32779.4	40765.2	52011.8	59467.1	71285.7	84572.3	100433.3
8489.0	32881.5	40981.8	52013.1	59495.5	71297.1	84592.1	100463.0
8567.5	32893.1	40987.8	52073.0	59506.2	71316.0	84798.1	100472.8
8694.7	32912.5	41370.7	52086.5	59555.5	71336.3	84896.0	100660.1
9052.4	32967.9	41616.5	52096.7	59564.4	71341.5	84988.0	100702.4
9230.7	33158.2	41892.2	52142.6	59584.7	71606.1	85901.1	101019.1
9363.5	33209.1	42034.3	52163.6	59597.9	71674.6	86563.5	101216.4
9705.9	33216.8	43012.0	52182.5	59640.5	71782.2	86591.3	102662.2
9799.2	33252.0	43103.6	52184.2	59661.0	71788.3	86686.5	102770.4
9857.2	33595.8	43145.1	52277.9	59661.8	71809.2	86756.1	102847.0
9894.1	33694.7	43276.5	52290.6	59702.8	71817.9	87908.9	102974.0
9898.4	33720.0	43463.6	52308.8	59721.6	71853.9	88313.6	103938.5

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10218.1	33768.8	43492.6	52342.2	59757.0	71880.9	88523.2	103961.2
11344.4	33833.0	43536.6	52486.9	59786.6	71899.2	88621.6	104125.7
11369.4	33850.7	43607.1	52490.9	59864.0	72618.3	88673.7	104182.0
11537.3	33851.9	43608.0	52542.4	59876.2	72721.0	88695.5	104195.8
11633.6	33892.2	43714.2	52571.6	59925.1	73944.6	88799.8	104284.7
12596.6	33982.0	43738.6	52650.7	59939.1	74119.6	88856.1	104454.5
12719.1	34006.0	43800.9	52724.4	59954.6	74223.7	88871.3	104527.0
12806.5	34293.5	43899.4	52741.3	60277.2	74443.7	88896.5	104627.3
13469.4	34500.8	43963.9	52901.3	60334.2	74504.5	89218.0	104847.2
13559.7	34550.2	44028.3	52962.8	60352.5	74532.0	89295.3	105020.6
14009.1	34637.2	44070.3	53133.3	60389.9	74548.1	89558.4	106018.8
22374.1	34644.3	44080.2	53273.6	60480.5	74551.2	89732.9	106041.1
22431.8	34644.6	44228.4	54036.8	60528.3	74599.1	90180.5	106382.1
22555.1	34696.1	44443.0	54067.0	60612.1	74633.7	90490.1	107183.2
22822.1	34742.1	44707.1	54150.2	61010.7	74687.0	90799.6	107434.5
23295.3	34797.8	44715.3	54183.7	61058.1	74704.3	90974.9	107551.5
24112.6	34807.4	44901.5	54201.1	61142.7	74720.3	91051.1	107692.7
24137.8	34839.5	45075.7	54226.7	61198.2	74727.1	91095.9	107752.5
24176.5	34855.8	45470.3	54339.0	61273.0	74737.7	91574.4	109614.2
24183.9	34930.7	45485.4	54392.2	61299.7	74775.5	92027.8	110154.3
24275.4	34963.8	45624.9	54403.6	61396.2	74789.0	92091.9	115063.5
24283.1	35339.9	45641.0	54643.8	61416.3	74803.2	92199.7	115616.7
24703.2	35447.4	45686.2	54671.3	61521.0	74823.1	92265.6	115813.8
24824.0	35514.0	45695.9	54710.0	61541.4	75007.4	92291.4	115914.0
24889.4	35557.1	45701.6	54763.6	61639.4	75049.4	92373.1	115958.6
24934.5	35568.6	45844.7	54779.9	61649.3	75159.8	92483.9	115984.1
25025.1	35599.8	45890.2	54797.1	61664.6	75190.1	92684.0	115998.3
25081.4	35631.8	45906.7	54798.5	61688.9	75198.5	92975.3	116120.9
25135.3	35668.6	45971.3	54900.3	62007.9	75279.6	93044.6	116441.5
25200.2	35685.2	45976.8	54919.3	62123.5	75311.3	93060.7	116458.9
26665.0	35757.5	45984.4	55013.2	62182.5	75403.9	93184.8	116768.5
26722.3	35802.1	46033.1	55033.8	62199.8	75406.2	93406.3	116997.3
26838.4	35852.9	46042.6	55055.9	62260.2	75431.1	93436.3	117045.9
26902.9	35933.1	46082.8	55098.0	62302.9	75470.1	93493.5	117199.4
27143.4	36048.9	46162.1	55180.6	62316.9	75528.8	93533.9	117295.5
27269.5	36123.1	46197.4	55214.4	62408.2	77041.9	93538.9	117341.2
27336.4	36193.3	46201.4	55331.9	62431.8	77350.1	93544.0	117366.7
27395.8	36243.3	46227.0	55343.7	62490.0	77573.3	93607.5	117390.3
27439.7	36270.9	46255.8	55419.3	62558.3	77809.8	93626.2	117576.5
27496.3	36301.3	46314.7	55454.4	62668.5	77903.5	93635.8	118154.7
27620.9	36322.6	46370.9	55496.5	62733.4	78005.5	93673.7	122859.0
27961.6	36797.5	46564.6	55523.1	62876.4	78120.6	93743.8	122949.6
27987.5	36824.2	46652.5	55588.7	62890.0	78138.2	93830.8	123708.7
28051.1	36850.6	46744.2	55604.9	62926.6	78185.6	93879.6	125903.5
28146.9	36921.2	46760.0	55626.2	62947.1	78686.4	93901.2	125943.3
28321.9	36946.7	46794.2	55709.7	63078.9	79021.2	93916.4	126504.9
28345.4	36999.0	46880.7	55719.8	63193.8	79215.0	94464.2	126971.6
28439.6	37004.0	46923.4	55768.3	63202.1	79232.7	94601.6	131337.7
28540.7	37102.3	46928.4	55907.8	63358.0	79256.8	94623.8	131408.1
28622.8	37114.5	46970.5	55929.6	63509.3	79260.8	94641.4	131569.0
28761.3	37121.6	46988.3	55997.9	63598.3	79295.5	94650.8	131769.9
28763.2	37226.1	46995.2	56001.6	63799.0	79297.5	95054.2	131872.3
28817.5	37440.9	46999.7	56071.3	63908.1	79471.8	95086.0	132997.9
28851.0	37525.9	47037.6	56091.4	63910.1	79484.6	95389.2	133536.4
28962.7	37594.2	47046.3	56171.1	63942.9	79647.9	95558.5	134082.8
29013.2	37600.6	47092.5	56213.4	63960.4	79660.3	95593.3	134234.5
29155.1	37618.7	47113.6	56241.5	63960.9	79719.8	95685.9	134525.0
29163.1	37623.5	47133.2	56323.9	64075.7	79842.5	95720.9	134700.4
29204.5	37645.2	47179.2	56368.0	64091.4	79906.7	95861.5	135661.5
29254.8	37673.0	47227.0	56461.0	64112.9	79927.8	95898.0	136477.4
29262.0	37687.0	47258.8	56472.4	64119.6	80046.8	95931.2	136595.0
29283.0	37716.7	47275.7	56509.7	64218.3	80358.0	96006.3	136669.4
29327.7	37734.7	47295.2	56685.0	64245.3	80436.0	96050.8	137050.0
29381.2	37745.4	47307.0	56789.8	64257.6	80439.0	96116.1	137348.3
29400.3	37775.8	47341.0	56799.2	64290.7	80752.6	96331.0	137518.6
29445.0	37847.8	47401.5	56835.2	64302.6	80959.9	96364.2	140124.3
29506.8	37868.4	47410.0	56867.8	64323.7	81133.2	96392.8	140146.4
29572.7	37981.2	47419.9	56903.5	64384.9	81285.7	96406.9	140446.4
29631.9	38004.0	47421.0	56934.1	64398.4	81361.7	96410.4	140788.1
29765.5	38025.0	47432.7	57040.9	64655.7	81372.1	96423.6	141057.2
29788.2	38034.1	47434.5	57046.6	64686.8	81374.8	96431.1	159999.7
29946.4	38215.2	47439.9	57274.0	64716.2	81383.7	96472.8	160849.3
29948.5	38228.5	47460.9	57776.3	64744.8	81396.8	96519.4	161165.6
29961.7	38271.0	47521.9	57790.2	64791.3	81636.1	96544.9	163580.5
30039.3	38282.7	47563.0	57890.2	64828.5	81676.2	96713.0	163658.2
30047.7	38358.5	47618.6	57890.7	65126.9	81738.0	97119.7	163960.5
30092.6	38473.6	47645.5	57943.7	65286.9	81985.5	97212.8	164012.7

## Supporting Information

30111.4	39081.0	47806.5	58002.7	65306.9	82047.7	97269.7	164350.7
30142.9	39132.2	47925.0	58047.6	65362.1	82078.8	97293.6	165306.8
30221.8	39138.3	48632.9	58113.4	65398.9	82086.9	97315.4	166776.5
30266.2	39223.9	48727.1	58241.7	65456.5	82159.6	97369.8	166984.6
30422.0	39232.4	48758.1	58249.8	65570.2	82169.9	97400.7	167170.6
30473.7							

**Table S28:** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_8\text{H}_8)_2]^-$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

5						
Spin free States						
CASSCF						
1.0	36444.9	46510.5	56693.2	79076.4	97229.2	
74.9	36573.7	46510.5	56696.9	79083.6	97286.8	
75.0	36573.7	46776.5	56818.1	79083.6	97286.9	
115.3	36621.0	46776.5	56818.2	79109.9	97708.3	
136.5	36621.0	46880.2	56953.7	79110.0	98005.8	
137.0	36765.6	46880.3	56954.3	79115.5	98011.4	
156.2	36766.5	47088.9	57160.9	79115.5	98040.6	
162.7	36816.9	47089.0	57161.1	79313.8	98040.7	
1157.3	36819.8	47179.6	58248.3	79313.8	100448.2	
1157.3	36822.6	47179.6	58248.3	79488.6	100448.2	
7345.4	36842.4	47205.5	58552.8	79488.6	100458.3	
7345.4	36842.4	47209.8	58552.8	79695.6	100458.3	
7537.7	36845.6	47255.0	58557.2	79695.6	100480.9	
7538.1	36876.0	47640.2	58635.2	79802.2	100481.1	
7752.5	36876.0	47640.2	58636.8	79803.2	100502.9	
7754.8	37102.3	47806.6	58732.7	79805.0	100503.2	
7949.3	37265.3	47817.0	58733.5	79806.8	100512.3	
33966.9	37266.0	47823.3	58749.5	79807.4	100514.8	
33977.5	37464.4	47859.1	58749.6	79808.7	100515.4	
34829.5	37464.4	47859.5	58959.8	79810.6	100631.5	
24085.3	37548.5	47920.3	58959.8	79811.9	100631.5	
24085.4	37549.6	47920.8	59366.0	79812.4	101890.4	
24091.9	37845.5	47943.6	59368.9	79825.3	101890.4	
24091.9	37845.6	47943.6	59702.0	79825.4	101950.9	
24172.1	37897.5	47996.6	59722.5	83996.8	101951.1	
24172.3	37897.6	47996.6	59772.4	83996.8	102177.8	
24178.2	37982.3	48071.2	59772.6	84618.7	102177.8	
24178.2	37982.6	48071.2	59815.2	84626.2	102220.1	
24277.0	38162.7	48091.7	59817.5	84697.6	102220.2	
24277.4	38162.7	48091.7	60021.5	85061.8	102485.6	
24311.3	38190.9	48117.5	60021.5	85114.7	102490.4	
24311.3	38190.9	48117.5	60816.1	85114.7	102593.6	
24440.5	38352.0	50558.9	60816.1	85241.0	104565.0	
24440.5	38352.5	50558.9	61313.6	85242.0	104565.1	
24486.9	38408.7	50724.9	61313.6	85468.8	104696.9	
24616.2	38642.7	50795.8	61505.5	85469.3	104697.1	
24616.2	38642.7	50795.9	61535.1	85546.7	104834.7	
24641.3	41146.5	50967.6	61539.1	85546.7	104834.7	
24641.3	41146.5	50967.7	61587.5	86089.3	105093.7	
24688.6	41271.7	51191.1	61587.5	86089.3	105098.2	
24688.6	41271.7	51191.2	61617.4	86390.4	105219.7	
24697.4	41373.2	51220.0	61617.5	86390.4	105760.1	
24700.0	41377.1	51220.0	61705.5	86463.0	105760.4	
24729.6	41407.1	51376.7	61705.7	86463.0	105806.2	
24729.7	41407.1	51376.7	61722.0	86757.8	105806.2	
24892.2	41442.6	51661.5	61722.1	86757.8	106261.4	
24892.6	41543.0	51661.6	63121.0	86991.2	106261.5	
24899.9	41543.0	51706.0	63348.2	86991.6	106630.5	
24955.9	41621.6	51706.0	63349.7	87077.0	106637.5	
24956.4	41621.6	51707.6	63416.5	87079.6	106758.9	
24992.1	41659.3	51707.6	63416.7	87087.5	109606.9	
24992.1	41659.3	51740.7	63489.6	87089.8	109634.6	
25025.2	41684.6	51740.7	63489.6	87091.0	109639.0	
25025.2	41684.6	51778.2	66010.3	87091.0	109745.5	
25155.5	41721.8	51778.3	66650.1	87094.4	109745.5	
25155.5	41722.0	51922.8	66650.1	87330.2	110169.7	
25294.7	41783.0	51922.8	66832.3	87386.8	110170.0	
25299.1	41783.1	52024.3	66842.8	87391.8	113913.3	
25393.6	41856.1	52027.4	67084.9	87449.9	113922.0	
25527.7	41863.0	52061.6	67085.0	87450.0	114655.4	
25805.4	41904.4	54348.3	67169.6	87509.2	118906.8	
25810.5	41990.4	54348.4	67171.0	87509.3	118906.8	
25847.3	42073.0	54363.4	67172.0	87561.7	119416.8	

## Supporting Information

25847.3	42076.8	54363.5	67173.6	87562.6	119416.8
25928.2	42238.5	54447.9	67179.8	92799.1	119669.3
25928.5	42238.5	54448.7	67201.4	92799.1	119669.3
26087.9	42255.9	54624.0	67201.5	92915.0	119822.9
26087.9	42302.9	54627.7	68899.9	92915.7	119822.9
29041.0	42303.3	54792.2	68899.9	93003.5	119920.5
29055.3	42340.5	54819.9	68951.9	93003.5	119920.7
29055.6	42341.2	54819.9	68951.9	93034.5	119985.4
29055.6	42359.0	55116.4	68994.5	94629.4	119985.9
29057.6	42359.1	55116.4	68994.5	94635.6	120025.1
29094.4	42540.8	55176.3	68997.5	94638.2	120033.2
29094.5	42540.8	55176.3	68998.2	94744.3	120045.6
29100.6	42553.2	55192.4	68999.2	94744.3	121981.2
29100.6	42553.2	55212.3	69003.6	94893.5	122056.3
29136.4	42624.9	55215.5	69003.6	94893.5	122063.5
29136.8	42625.0	55261.9	69007.9	94929.5	122644.5
29156.8	42748.1	55262.0	69008.3	94929.7	122644.5
29156.9	42748.8	55265.5	69045.7	94998.9	131329.4
29280.7	42791.7	55265.5	69045.7	94999.0	131329.6
29280.7	42792.2	55318.1	70239.8	95342.0	131544.2
30313.3	42862.7	55318.1	70241.1	95342.0	131544.3
30313.3	42862.7	55343.9	70584.9	95704.9	132120.0
30445.4	42999.6	55343.9	71760.3	95704.9	132129.3
30449.6	42999.6	55603.9	71983.5	95763.9	132487.0
30449.9	43060.3	55603.9	71993.2	95768.5	139269.6
30476.6	43060.3	55687.3	72249.6	95787.6	139288.2
30476.6	43101.2	55687.3	72580.1	95814.5	139293.8
30548.5	43101.2	55815.8	72580.1	95815.0	139438.4
30548.5	43136.9	55828.9	72686.0	95943.4	139438.5
30665.7	43139.4	55829.4	72688.6	95943.4	139691.0
30665.7	43270.3	55901.1	72730.2	95958.3	139691.4
30730.2	44097.2	55901.1	72730.2	95958.3	139864.1
30730.2	44097.2	56006.7	75999.9	96019.1	139864.2
30778.7	44124.5	56007.0	76003.7	96019.4	141409.2
30778.7	44124.5	56027.2	76003.7	96028.5	142111.0
30817.7	44243.9	56038.3	76118.1	96029.9	142119.1
30817.7	44244.0	56038.3	76118.2	96033.2	145418.9
32188.4	44303.5	56075.4	76122.7	96037.7	145419.0
32189.8	44303.6	56079.6	76122.8	96038.0	145482.4
32399.0	44334.0	56088.3	76188.4	96059.8	145482.5
32399.0	44334.4	56088.4	76188.6	96059.8	145573.5
32695.3	44343.0	56109.7	76200.2	96067.6	145573.6
33776.8	44343.0	56109.8	76208.2	96068.0	145807.8
33809.4	44384.2	56134.8	76209.6	96291.9	145811.2
33809.7	44384.2	56134.8	77502.5	96291.9	145899.4
33813.2	44414.6	56190.7	77506.7	96637.0	146221.8
33813.2	44414.6	56191.2	77577.1	96637.0	146221.8
33847.9	44447.6	56251.0	77577.3	96911.0	173526.2
33848.4	44448.2	56251.0	77694.2	96951.9	173526.3
33939.8	44508.6	56357.9	78181.7	96952.1	174708.4
33939.8	44558.0	56373.5	78182.5	96966.5	174710.8
33988.2	44558.1	56379.7	78836.3	96971.2	174713.8
33988.2	44626.0	56483.2	78882.0	97005.8	176989.4
36206.1	44745.3	56483.3	78884.2	97005.8	176990.3
36206.1	44750.2	56489.5	78987.6	97099.5	177024.9
36287.7	44813.5	56489.6	78987.7	97099.6	177025.0
36287.7	45478.6	56629.1	78994.8	97219.4	177244.5
36298.2	45478.7	56679.3	78994.8	97219.5	177487.9
36298.2	45503.0	56679.4	79076.3	97229.1	177495.8
36444.2	45507.9				

**Table S29:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_8\text{H}_8)_2]^-$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

5					
Spin free States					
NEVPT2					
0.6	34841.7	44785.0	54609.4	74656.4	90848.1
13.9	34913.2	44785.2	54609.6	74801.9	90848.1
15.7	34913.4	44792.3	54669.2	74802.1	91062.1
60.7	35171.6	44792.3	54669.2	74864.0	91062.5
190.9	35171.8	44803.7	54766.0	74864.0	91068.3
199.4	36753.0	44808.6	54766.0	74895.8	91068.3
223.1	36753.0	44858.4	54790.2	74895.9	91149.7
224.7	36883.3	44885.4	54790.3	75339.7	91150.0
1507.9	36883.3	44885.4	55299.6	75348.1	93039.8



## Supporting Information

1507.9	36932.9	48704.2	55299.6	75352.9	93039.8
6104.7	36932.9	48704.3	55339.1	76077.9	93056.1
6105.2	37107.2	48822.8	55339.3	76077.9	93056.4
6371.3	37108.6	48822.8	55547.7	76293.2	93078.7
6372.0	37309.3	49029.8	55649.0	76293.2	93078.8
6656.3	37309.3	49129.2	55649.0	76523.8	93117.6
6658.8	37369.1	49129.4	55677.7	76523.8	93117.7
6896.6	37369.3	49354.5	55678.7	76640.7	93137.1
28640.8	37374.4	49354.6	55780.7	76640.7	93139.9
28652.4	37374.4	49553.3	55785.7	76731.2	93141.6
29654.7	37433.4	49553.3	55787.5	76749.2	93321.7
22850.7	37433.4	49648.1	55787.7	76753.5	93321.7
22853.9	37499.5	49648.5	55829.7	76753.5	93892.6
23177.1	37500.1	49731.2	55831.3	76784.1	93893.0
23182.1	37602.3	49731.2	56167.9	76789.8	93931.1
23611.5	37604.0	49895.9	56611.0	76827.3	93931.5
23614.9	37698.5	49895.9	56611.4	76828.8	94031.8
23629.8	37712.8	50118.3	56627.5	76829.6	94032.3
23633.3	37712.8	50118.3	56627.6	76835.8	94251.7
23635.2	37809.9	50207.2	56650.7	76866.3	94251.8
23635.3	37809.9	50227.9	56932.8	78485.3	94272.5
23650.2	37816.4	50233.9	56932.9	78543.4	94272.9
23653.9	37816.4	50285.9	57182.8	78543.4	94279.0
23654.0	38051.0	50286.4	57182.8	78772.7	94279.0
23808.2	38051.3	50385.5	57774.2	78773.9	94340.7
23871.0	38125.6	50386.4	57774.2	78957.6	94342.2
23873.9	38398.7	50508.3	57925.9	78957.6	94599.1
24085.0	38398.7	50508.7	57977.6	78995.8	94604.9
24323.6	38415.8	50512.8	57982.0	78996.9	94684.4
24323.6	38420.3	50512.8	58106.1	79219.3	94689.9
24378.9	38488.8	50630.5	58106.2	79219.4	94739.1
24381.7	39207.9	50630.5	58115.5	79698.9	94846.2
24382.0	39305.4	50681.3	58115.6	79698.9	96129.8
24383.2	39310.7	50681.5	58256.1	79957.6	96130.7
24389.2	39619.6	50697.8	58256.5	80064.2	96363.0
24389.8	39620.3	50697.8	58299.1	80069.2	96363.2
24635.8	39746.6	50712.7	58299.4	80183.8	96814.0
24635.8	39747.1	50712.7	58425.4	80184.4	96814.0
24975.1	40298.5	50779.8	58812.5	80259.8	97298.1
24975.1	40298.6	50779.9	58814.2	80260.1	97305.9
25743.5	40385.7	50805.1	58849.2	80302.0	97436.9
25743.5	40387.4	50805.1	58849.9	80302.0	100211.5
25933.0	40857.8	50817.4	58998.7	80327.2	100261.3
25933.0	40860.2	50817.7	58998.7	80329.0	100266.7
26377.0	40943.7	50870.5	59808.7	80525.2	100401.7
26377.0	41017.8	50870.5	60808.6	80525.2	100401.9
26378.8	42132.8	51056.3	60821.1	81007.6	100945.3
26383.3	42133.0	51056.4	62728.1	81007.6	100945.9
26528.3	42228.0	51227.4	62728.1	81310.2	103789.2
26528.7	42232.8	51231.7	63292.2	81311.1	103798.9
26773.5	42326.3	51288.5	63292.4	81396.6	104638.5
26773.5	42326.5	51560.7	63330.7	81397.7	109695.9
27132.7	42380.7	51561.1	63334.0	81405.6	109695.9
27132.7	42380.9	51641.1	63341.8	81406.4	110319.7
27163.0	42385.6	51642.2	63345.7	81406.9	110319.7
27169.0	42385.6	51817.5	63362.8	84632.2	110582.7
27169.6	42449.5	51819.1	63411.0	84632.3	110582.8
27238.5	42450.1	51890.2	63411.2	84775.7	110701.2
27249.9	42537.9	51908.0	64992.9	84776.4	110750.9
29207.9	42538.0	51914.4	65170.1	84873.6	110751.5
29231.8	42561.4	51956.0	65170.8	84873.9	110785.0
29231.8	42561.4	51956.0	65208.5	84943.3	110793.8
29237.4	42583.0	51967.7	65219.6	87086.4	110866.9
29239.9	42583.4	51972.4	65649.1	87090.6	110867.4
29306.8	42615.8	52075.2	65737.4	87093.7	110954.9
29306.8	42616.5	52080.1	65737.4	87161.7	110955.8
29345.8	42705.3	52263.5	65845.3	87162.5	111025.7
29345.8	42753.9	52467.1	65852.4	87259.6	111035.9
29388.0	42753.9	52472.8	65852.4	87259.6	111053.7
29388.7	42933.9	52588.1	65855.1	87418.8	111500.2
29446.9	42934.1	52589.5	65856.2	87418.8	111500.9
29447.1	43137.3	52644.9	65894.2	87554.7	118364.2
29508.6	43137.4	52749.3	65894.5	87555.1	118364.9
29510.3	43193.2	52756.9	65929.4	87637.1	118548.0
29582.4	43198.7	52880.0	65929.5	87637.2	118548.3
29582.4	43283.2	52880.7	65935.3	87757.4	119291.8
29793.1	43299.4	52881.5	65936.1	87763.5	119302.9

## Supporting Information

29793.2	43299.4	52882.8	65979.0	87791.4	119833.4
30230.4	43461.5	52931.8	65979.0	87796.8	127167.9
31110.1	43461.5	52931.9	66122.0	87798.1	127168.1
31110.1	43472.6	53454.0	66122.0	87976.2	127174.8
31283.3	43633.6	53454.1	66253.3	87977.1	127337.4
31283.6	43633.6	53588.3	66705.6	88027.5	127338.2
31285.5	43697.0	53589.9	66705.8	88487.0	127655.0
31312.4	43697.0	53638.3	66923.3	88487.0	127655.9
31312.4	43702.7	53638.7	66927.1	88544.4	127862.2
31408.1	43706.4	53686.3	68568.0	88549.2	127862.8
31408.1	43710.2	53686.4	69755.6	88587.6	127957.8
31581.2	43710.6	53731.0	69755.6	88587.9	128824.6
31581.6	43712.7	53743.6	69795.0	89336.9	128834.7
31679.4	43713.2	53749.9	69795.2	89336.9	132570.0
31679.4	43714.5	53749.9	69871.4	89402.0	132570.2
31751.0	43714.5	53757.0	69871.8	89402.0	132632.1
31751.0	43724.3	53757.0	69890.3	89806.9	132632.7
31805.4	43724.3	53847.2	69890.4	89807.0	132746.9
31805.4	43771.1	53847.5	69927.3	90034.0	132747.4
32491.4	43771.1	53876.8	69940.7	90043.0	133063.0
32576.7	43821.2	53881.3	69942.1	90051.2	133067.6
32576.7	43821.4	53903.8	70042.7	90051.4	133183.0
32588.5	43892.9	53904.6	70047.7	90091.9	133628.3
32588.8	43903.7	53915.7	70164.8	90093.9	133628.3
32639.1	44022.6	53915.9	70164.8	90113.2	156432.6
32639.9	44034.0	53948.1	70398.6	90113.2	156437.4
32792.4	44034.0	53948.6	71011.7	90148.1	157705.9
32792.4	44282.6	54158.9	71013.0	90149.2	157708.4
32877.3	44282.7	54158.9	74376.6	90157.3	157836.8
32877.4	44446.1	54197.2	74453.4	90338.0	159185.9
33593.3	44446.1	54229.0	74455.8	90338.0	159187.4
33750.3	44470.7	54229.0	74565.4	90492.8	159305.9
33754.2	44470.7	54297.6	74565.5	90608.4	159314.1
34308.4	44481.0	54297.6	74634.8	90610.6	159387.0
34308.5	44481.5	54343.1	74634.9	90783.0	160042.4
34593.8	44666.0	54345.8	74656.3	90783.0	160058.2
34840.4	44666.1				

**Table S30:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_8\text{H}_8)_2]^-$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

5							
Spin Orbit States							
CASSCF							
0.0	30241.5	39740.4	49742.9	60402.4	69795.4	88903.4	104352.2
21.3	30351.7	39787.1	49790.0	60412.5	69824.1	89274.5	104360.9
48.7	31199.5	39789.0	49819.4	60439.3	70132.8	89493.9	104380.6
54.4	31227.8	39860.6	49900.7	60457.1	70428.6	89499.6	104717.7
74.4	31303.8	39896.2	50002.9	60462.6	70610.2	89570.1	105260.7
83.4	31344.8	39994.5	50118.6	60492.4	70702.9	89642.4	105299.2
283.9	31378.1	40012.3	50195.9	60747.4	71293.1	89661.4	105372.9
1069.5	31398.9	40056.6	50224.9	60871.9	71754.3	89697.0	105424.6
3030.0	31440.0	40097.3	50228.0	60897.5	71859.7	89828.1	105662.6
3049.0	31453.1	40155.7	50231.6	60901.7	71897.7	89904.5	105722.1
3072.8	31513.1	40159.9	50273.1	60918.5	71932.7	89911.9	105746.7
3086.0	31537.0	40175.7	50278.9	60985.3	71947.6	90209.2	105777.9
3172.5	31546.9	40205.3	50297.4	60994.8	72428.5	90234.0	105887.5
3329.4	31564.5	40292.8	50299.5	61075.1	73710.9	90250.2	106852.5
3805.9	31658.1	40298.8	50335.9	61098.7	73855.2	90310.9	106879.3
5233.7	31738.0	40298.9	50339.7	61218.4	74036.4	90491.6	106919.8
5240.8	31761.6	40401.5	50407.4	61251.8	74260.0	90546.2	106938.8
5276.6	31834.9	40406.3	50408.1	61282.6	74300.7	90721.9	106972.3
5399.0	31892.7	41382.3	50419.2	61312.0	74319.5	90768.4	107007.1
5557.6	32199.2	41523.3	50489.5	61321.7	74330.4	90848.4	107033.4
5851.7	32201.1	41564.1	50527.9	61330.0	74335.2	90876.7	108680.2
6938.4	32379.4	41611.7	50538.0	61434.2	74349.8	91060.0	108948.7
6961.4	32403.2	41653.4	50577.2	61451.7	74358.3	91161.3	108957.7
7037.4	32442.3	42153.2	50642.3	61457.5	74363.1	91190.6	108966.0
7224.1	32783.4	42214.2	50675.7	61485.9	74372.2	91196.4	108978.1
7504.9	33046.9	42253.1	50741.6	61513.4	74392.6	91240.4	109083.3
8266.2	33118.9	42291.3	50810.0	61543.5	74393.0	91266.0	109124.7
8275.5	33119.2	42311.7	50931.3	61545.9	74423.4	91279.4	109189.0
8446.9	33221.8	42345.8	50998.4	61563.3	74451.5	91293.8	109340.2
8862.0	33294.3	42377.0	51020.0	61606.1	74492.3	91308.4	109389.0
9235.8	33299.9	42419.5	51033.1	61638.2	74678.5	91329.1	109510.3
9266.5	33320.3	42420.2	51516.3	61671.4	74710.0	91378.2	109678.4

## Supporting Information

9374.4	33354.7	42450.2	51647.7	61720.6	74958.2	91409.7	109802.0
9421.2	33361.9	42483.1	51659.3	61754.6	74959.1	91440.8	109813.6
9491.2	33373.2	42611.2	51724.2	61781.1	74967.8	91493.9	109928.2
9537.8	33417.4	42615.9	51938.5	61802.9	74999.4	91566.6	110099.5
9671.6	33541.7	42618.2	52057.5	62392.7	75005.9	93256.3	110391.9
9758.8	33573.5	42729.5	52109.4	62396.5	75008.4	93805.5	110551.5
10003.0	33593.8	42792.2	52357.3	62545.9	75015.5	93935.9	112375.0
10751.3	33609.0	42833.3	52373.4	62699.4	75219.2	93985.8	112402.9
10912.7	33660.8	43823.0	52407.4	62732.6	76763.0	93986.9	112475.6
10929.0	33734.1	43968.9	52411.8	62755.7	76880.1	94510.5	112548.9
10938.5	33777.8	44158.9	52464.2	62779.6	77069.6	94879.1	113034.3
10956.5	33808.9	44347.9	52535.6	62814.5	77796.7	95166.4	113070.5
12465.8	33889.8	44537.3	52548.2	62831.3	77894.6	95360.3	113122.6
12486.9	34053.4	44569.1	52575.3	62873.8	79189.1	95455.8	113321.1
12606.0	34163.5	44572.7	52600.9	62906.5	79512.1	95490.4	113524.9
12661.8	34292.9	44594.2	52626.5	62951.8	80274.9	95505.8	113652.4
13752.5	35050.7	44604.2	52815.4	62959.9	80486.0	96400.5	113839.1
13830.3	35155.6	44627.0	53715.7	63045.5	80524.8	96722.1	113938.4
13890.0	35173.5	44629.0	53815.0	63046.3	80608.6	96779.6	114309.7
14607.9	35239.8	44650.4	53895.2	63113.2	80731.3	96922.9	114567.1
14687.0	35256.6	44669.5	53974.4	63130.9	80764.1	96951.5	114677.8
15157.2	35261.5	44730.2	53996.6	63234.6	80778.3	97024.1	115525.3
23424.4	35281.4	44730.4	54063.7	63252.7	80786.0	97220.1	115528.0
23440.3	35286.1	44766.5	54074.1	63259.9	80790.4	97270.6	115770.3
23698.0	35337.3	44802.4	54161.7	63308.1	80793.4	97305.3	116735.0
23821.2	35519.1	44859.5	54178.6	63310.7	80840.8	97450.1	117011.1
24008.1	35527.6	44862.2	54273.8	63343.3	80867.8	97474.8	117125.8
24428.0	35544.4	44929.2	54318.6	63426.4	80993.9	97572.0	117128.2
24431.0	35562.6	45124.3	54383.2	63487.6	81019.5	97618.1	117738.9
24482.4	35636.0	45289.4	54426.4	63501.7	81038.3	99244.7	119780.2
24571.7	35702.5	45336.3	54498.6	63515.0	81131.7	99562.9	120423.4
24636.5	35706.4	45361.0	54584.6	63594.1	81216.4	99720.9	124214.3
24654.1	35930.3	45396.4	54672.6	63623.7	81251.4	99727.8	124649.9
24707.6	36033.3	45401.7	54769.9	64517.3	81256.8	99783.4	124849.8
25084.0	36242.6	45437.8	54775.2	64703.5	81340.2	99804.9	124955.8
26091.9	36273.6	45441.6	55006.4	64772.6	81361.8	99809.6	125012.1
26220.3	36526.3	45745.7	55122.7	64775.0	81366.2	99845.4	125044.9
26316.0	36608.7	45821.9	55152.1	64782.7	81424.5	99866.6	125064.5
26338.5	36697.4	45850.4	55168.1	64803.6	81509.2	99889.1	125216.5
26351.6	36702.8	45865.4	55238.8	65198.5	81617.9	99994.1	125795.8
26397.8	36710.5	45959.0	55325.9	65391.7	81629.6	100014.7	126093.4
26432.4	36732.6	45973.6	55378.3	65475.8	81938.2	100082.3	126272.1
26456.2	36735.9	46093.6	55402.7	65564.9	81942.4	100815.2	126387.6
26532.2	36763.9	46098.4	55424.3	65586.8	81943.9	100864.8	126466.1
26615.7	36771.2	46196.0	55836.4	65622.1	82048.2	100972.0	126519.8
26617.3	36940.2	46250.0	56029.2	65638.9	82428.3	101005.5	126550.5
26641.5	37177.9	46317.3	56045.7	65653.1	83713.3	101060.0	127624.8
26807.3	37240.8	46346.3	56074.7	65653.2	83864.2	101062.9	127696.4
26862.8	37244.0	46353.3	56089.9	65658.3	83939.9	101122.3	128202.5
26966.2	37254.9	46476.3	56118.6	65721.5	84021.3	101163.0	128771.2
27087.1	37272.8	46536.4	56171.1	65782.6	84089.6	101551.3	129411.3
27147.0	37321.1	46563.8	56202.1	65841.6	84181.7	101756.7	135727.6
27464.2	37340.4	46775.8	56212.5	65847.6	84190.0	101775.7	135968.7
27481.9	37397.9	47080.0	56934.5	66307.8	84200.4	101832.7	136534.6
27531.9	37398.4	47123.6	57067.1	66520.3	84202.9	101854.1	138669.4
27574.7	37573.3	47164.4	57137.0	66630.0	84236.0	101913.0	138909.8
27613.8	37646.7	47182.2	57277.7	66760.2	84255.9	101949.5	139381.2
27687.0	37732.8	47238.3	57392.2	66761.4	84269.4	102002.7	139714.1
27742.6	37746.4	47247.8	57427.4	66770.0	84337.9	102310.2	143561.4
27920.7	37754.5	47259.4	58172.3	66777.5	84365.1	102473.0	143625.0
28009.8	37886.5	47376.4	58416.2	66854.8	84371.4	102502.3	143743.9
28046.1	38082.2	47377.2	58639.2	66938.9	84379.0	102684.6	143896.3
28059.1	38102.7	47511.7	58661.6	66943.1	84399.0	102892.6	144028.8
28077.7	38195.9	47615.5	58672.9	66972.2	84531.4	102934.9	146233.7
28157.8	38197.2	47677.1	58689.1	66979.0	84536.2	102940.1	146365.4
28206.0	38205.2	48331.4	58715.0	67021.7	84578.6	103080.8	146506.3
28220.5	38264.0	48385.0	58729.1	67046.4	84653.7	103197.0	146625.0
28246.8	38330.3	48444.7	58752.6	67052.2	84683.1	103217.5	146844.5
28270.2	38334.3	48552.9	58952.8	67069.1	84684.6	103240.5	146990.6
28283.2	38554.9	48607.4	59033.2	67070.8	84700.6	103246.8	149094.1
28298.1	38559.8	48659.9	59058.8	67085.5	84788.6	103306.7	149569.9
28321.0	38564.4	48712.6	59062.4	67092.8	84873.4	103410.7	149675.8
28328.8	38577.9	48739.3	59083.2	67114.3	84930.3	103423.4	149684.0
28338.5	38579.2	48765.8	59097.3	67140.4	85087.0	103646.1	149888.6
28342.1	38605.0	48779.5	59308.9	67504.9	85113.3	103653.2	150049.1
28393.1	38639.0	48790.5	59579.1	67852.4	85160.0	103660.8	150337.3
28396.1	38668.7	48804.0	59612.6	67901.5	85212.1	103689.3	153113.5

## Supporting Information

28425.6	38679.8	48837.0	59665.4	67932.4	85238.2	103704.0	153218.1
28467.0	38700.8	48918.4	59749.0	67970.0	85304.4	103737.4	153258.3
28468.5	38713.2	49030.7	59772.8	68004.5	85360.9	103768.9	153441.5
28551.3	38730.1	49090.9	59803.4	68006.0	85388.5	103811.7	153895.3
28569.4	38850.9	49121.4	59837.5	68673.9	85396.4	103813.9	177091.1
29645.7	38938.1	49307.7	59848.2	68739.2	85399.2	103832.4	178079.6
29665.3	39021.8	49427.5	59875.6	68950.3	85403.1	103857.3	178585.3
29673.8	39049.9	49431.3	59906.7	69037.3	85955.7	103992.0	181374.6
29708.3	39106.6	49485.0	59962.1	69163.1	86033.1	104030.0	181554.3
29752.4	39136.9	49487.7	59985.0	69172.3	86136.0	104047.0	181571.4
29775.9	39156.3	49522.6	60191.2	69212.7	86673.6	104049.6	181623.2
29921.3	39172.0	49546.5	60231.9	69378.9	86859.7	104090.4	181820.4
29946.9	39371.7	49588.6	60236.4	69596.7	88546.9	104169.8	182743.1
29992.7	39444.9	49597.6	60273.9	69631.9	88556.3	104182.0	184698.9
30063.8	39510.6	49636.2	60346.1	69673.8	88844.1	104280.9	184729.8
30203.7	39573.9	49722.5	60351.6	69729.1	88862.4	104336.3	185017.9
30230.4							

**Table S31:** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_8\text{H}_8)_2]^-$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

5							
Spin Orbit States							
NEVPT2							
0.0	30530.2	39188.6	48797.0	58246.2	66194.5	82363.9	97597.7
26.0	30540.4	39239.3	48890.6	58272.7	66527.7	82416.4	97600.3
38.6	30559.7	39242.8	48946.5	58340.9	66543.8	82448.8	97632.4
43.9	30685.1	39254.4	49406.0	58361.8	66569.6	82642.8	97684.2
60.1	30791.1	39277.9	49410.4	58381.2	66574.8	82723.1	97700.1
71.3	30946.1	39288.0	49417.6	58422.9	66711.7	82873.1	97720.1
307.1	30981.4	39346.7	49561.7	58434.8	66771.7	82922.0	97752.9
1407.5	31033.2	39369.0	49582.8	58507.0	67427.3	82941.0	97880.7
3003.0	31060.7	39376.0	49613.5	58517.0	67584.3	83103.2	97981.5
3043.5	31149.3	39425.2	49630.4	58523.7	67677.8	83319.0	97986.9
3066.7	31178.5	39432.2	49684.3	58567.3	67914.1	83336.6	98071.2
3110.3	31179.4	39489.6	49740.4	58572.7	67957.5	83384.0	98134.5
3204.8	31298.2	39491.3	49859.9	58628.4	68051.7	83576.1	98140.2
3388.4	31412.5	39535.3	49899.4	58699.9	68074.8	83621.9	98603.5
4067.3	31426.0	39536.9	49942.2	58706.5	68123.9	83723.2	98810.4
5209.8	31463.4	39586.3	49989.0	58716.3	68138.9	83750.9	99043.8
5215.4	31467.5	39622.2	50065.8	58744.6	68156.3	83795.7	99180.6
5233.5	31475.1	39671.3	50229.3	58834.1	68245.5	83866.6	99358.8
5392.1	31556.7	39690.8	51280.8	58844.5	68505.7	83878.8	99407.9
5607.3	31707.3	40068.1	51543.0	58896.8	69057.6	83985.7	99557.3
6040.5	31728.4	40099.7	51556.9	58986.3	69964.7	83991.5	99601.2
6898.7	31846.4	40103.6	51592.4	59097.5	70292.3	84072.1	99643.7
6915.6	31979.4	40228.6	51655.8	59108.6	70307.8	84084.3	99650.5
6978.9	32230.8	40232.7	51730.1	59118.5	70336.5	84132.9	99693.2
7242.0	32231.4	40329.3	51775.6	59124.2	70342.7	84159.4	99700.6
7655.2	32326.9	40356.0	51783.8	59215.0	70556.3	84195.0	99732.0
8148.2	32375.6	40356.0	51796.6	59239.3	70646.0	84197.6	99767.6
8172.4	32435.0	40497.9	51797.6	59249.5	70920.8	84458.0	100309.9
8191.1	32436.2	40551.9	51815.9	59285.6	71131.8	84486.2	100350.1
8228.9	32522.8	40606.7	51846.9	59331.0	71196.6	84593.1	100388.3
8299.1	32623.0	40641.2	51912.9	59360.2	71221.2	84633.3	100429.9
8315.7	32689.5	40660.0	51917.4	59431.1	71241.0	84650.1	100481.0
8447.1	32705.1	40878.3	51981.1	59469.7	71305.7	84676.0	100514.4
8507.8	32742.5	40915.7	52002.3	59523.7	71306.5	84819.1	100521.0
8634.9	32745.4	41379.6	52003.0	59532.9	71313.8	84840.4	100622.9
9029.8	32978.1	41526.4	52057.4	59559.1	71342.6	84997.5	100797.0
9162.4	33002.9	41859.8	52082.5	59560.0	71597.2	85893.9	101114.7
9202.3	33168.3	42003.2	52098.6	59566.9	71687.4	86549.5	101311.7
9572.8	33213.4	42974.0	52133.8	59571.0	71734.1	86662.3	102719.4
9753.1	33227.5	43045.8	52163.7	59578.2	71752.9	86759.9	102788.2
9789.6	33541.2	43160.5	52171.3	59620.7	71774.3	86780.3	102896.3
9795.3	33677.9	43188.2	52242.5	59697.4	71797.6	87877.7	103028.2
9813.0	33680.3	43410.1	52306.4	59709.3	71861.4	88245.4	103984.0
10194.2	33717.6	43501.3	52309.1	59718.5	71863.4	88518.8	104032.3
11283.5	33762.6	43505.5	52312.0	59767.8	71877.9	88593.8	104142.3
11287.3	33792.6	43540.0	52517.2	59775.4	72621.8	88670.0	104179.4
11465.4	33809.3	43583.8	52568.7	59840.7	72755.3	88708.8	104229.3

## Supporting Information

11521.8	33831.2	43588.1	52574.5	59877.6	73942.7	88843.4	104256.6
12536.1	33831.9	43681.9	52598.9	59933.3	74132.9	88942.9	104520.0
12650.0	33980.7	43764.4	52683.6	59937.5	74175.1	88958.0	104651.2
12707.2	34235.9	43774.2	52694.7	60195.3	74432.3	88968.3	104693.1
13373.8	34448.4	43962.7	52811.1	60365.7	74535.2	89181.9	104980.0
13493.9	34472.5	43963.8	52890.5	60378.9	74535.5	89259.6	105113.1
13944.7	34541.0	43968.9	53006.7	60384.7	74557.0	89362.4	106030.3
22297.7	34548.8	43997.9	53216.5	60396.8	74568.8	89604.3	106061.4
22366.6	34594.2	44207.4	53998.7	60477.8	74599.6	90109.4	106398.9
22474.1	34600.3	44376.2	54015.5	60507.7	74613.4	90413.5	107288.2
22787.1	34681.4	44642.1	54030.0	60935.2	74680.4	90802.7	107551.6
22943.9	34691.9	44691.0	54033.0	61019.0	74688.4	91042.6	107599.0
23985.4	34721.4	44823.1	54149.7	61096.1	74702.0	91134.5	107684.4
23993.1	34730.7	45032.0	54171.0	61133.5	74709.3	91176.6	107770.8
24031.6	34738.1	45460.6	54306.9	61198.2	74714.8	91482.5	109604.5
24106.3	34853.1	45496.1	54367.2	61278.0	74778.0	92041.4	110295.1
24149.8	34935.2	45580.3	54376.0	61371.9	74800.5	92088.3	115099.6
24370.9	35194.9	45599.3	54580.6	61399.2	74822.2	92103.2	115638.3
24654.6	35384.0	45633.9	54620.7	61454.9	74875.7	92230.8	115848.5
24802.7	35448.6	45674.0	54743.7	61511.8	75041.0	92246.8	115968.1
24905.6	35463.5	45701.7	54759.5	61606.6	75068.8	92307.7	116042.3
24967.9	35468.9	45810.6	54782.4	61620.9	75152.0	92521.8	116053.3
25053.4	35541.3	45819.4	54811.0	61634.5	75185.3	92826.5	116096.7
25072.9	35594.5	45831.1	54837.8	61671.8	75187.9	92969.0	116135.7
25221.4	35598.0	45902.1	54906.2	61911.4	75269.2	92997.5	116465.0
25234.3	35609.7	45941.8	54940.6	62131.1	75288.5	93046.5	116540.4
26606.5	35617.1	45942.8	54947.4	62166.3	75392.4	93414.7	116741.1
26668.8	35718.3	45988.3	54949.7	62166.7	75414.3	93438.1	117054.0
26700.4	35870.5	45991.8	54969.6	62204.5	75425.4	93479.6	117119.0
26852.2	35891.2	45996.9	55020.9	62237.9	75456.7	93536.9	117244.2
27065.6	35998.0	46016.7	55060.6	62317.1	75547.8	93596.1	117376.9
27197.5	36056.6	46096.8	55161.8	62329.7	77011.3	93629.1	117474.2
27233.0	36112.7	46126.9	55181.6	62430.6	77395.6	93632.2	117552.2
27251.3	36155.3	46131.1	55224.1	62440.6	77560.3	93671.1	117561.2
27294.6	36194.7	46192.6	55316.4	62563.4	77826.0	93696.0	117615.8
27431.8	36293.8	46263.5	55497.4	62633.7	77906.4	93718.3	118297.0
27578.6	36295.4	46285.2	55519.9	62752.6	78051.4	93733.1	122874.2
27964.9	36752.2	46570.3	55535.8	62849.7	78124.7	93740.2	123102.5
27980.3	36790.3	46612.7	55537.4	62851.7	78150.3	93836.6	123842.6
28210.8	36791.3	46625.2	55573.1	62877.6	78207.4	93951.0	125851.0
28267.0	36876.7	46704.5	55592.9	62898.9	78635.3	93952.8	126060.9
28326.9	36884.7	46762.7	55676.2	63068.5	78981.7	94032.1	126658.4
28362.3	36951.2	46788.4	55680.9	63142.7	79198.1	94438.9	127131.5
28387.5	36967.6	46798.3	55739.6	63158.2	79285.1	94574.2	131434.3
28504.9	37043.6	46818.8	55833.5	63306.8	79300.5	94647.5	131493.2
28582.1	37074.4	46841.5	55888.3	63472.4	79308.4	94672.1	131631.2
28649.5	37078.8	46939.7	55960.1	63543.0	79319.4	94690.4	131841.1
28705.2	37197.5	46943.4	56026.4	63807.8	79327.4	95046.0	132016.9
28732.7	37358.8	46949.5	56033.6	63815.2	79459.8	95523.2	133107.7
28830.9	37475.5	46972.8	56064.5	63815.4	79469.7	95525.5	133732.1
28877.2	37501.2	46980.8	56172.8	63895.5	79627.3	95548.1	134166.4
28887.4	37540.6	46994.3	56202.5	63936.6	79740.7	95666.6	134291.3
28999.1	37566.5	47040.5	56226.5	63980.9	79784.6	95677.9	134606.9
29071.3	37569.0	47077.9	56291.7	64014.5	79830.0	95714.1	134868.1
29100.0	37591.6	47119.4	56337.9	64049.2	79924.7	95786.6	135835.6
29128.7	37605.0	47125.3	56424.8	64059.9	79934.1	95833.8	136742.7
29152.0	37614.1	47188.7	56462.8	64075.8	80017.0	95912.1	136864.2
29167.5	37629.8	47197.6	56513.3	64190.4	80446.2	95968.6	136875.8
29187.1	37659.2	47228.8	56636.0	64233.3	80450.2	95970.8	137131.6
29242.6	37697.9	47272.8	56693.9	64240.2	80495.8	96194.0	137339.6
29332.1	37701.6	47273.7	56775.1	64259.8	80697.8	96324.1	137764.0
29368.1	37817.8	47328.7	56782.6	64269.0	80894.9	96345.4	140337.1
29400.6	37827.0	47382.5	56812.9	64315.3	81092.5	96369.8	140431.7
29585.1	37869.8	47395.8	56838.9	64343.8	81273.8	96390.2	140525.9
29611.0	37929.6	47429.6	56913.4	64398.2	81367.4	96423.0	140765.9
29792.5	37950.6	47435.1	56937.7	64633.0	81369.4	96453.2	141331.5
29823.5	37979.6	47439.2	57042.5	64671.9	81389.8	96481.4	159982.4
29853.9	38139.0	47443.7	57304.6	64711.0	81411.3	96503.0	161028.5
29862.3	38199.6	47460.6	57750.4	64711.3	81418.0	96526.7	161577.9
29863.1	38218.3	47489.0	57768.7	64751.0	81625.1	96544.7	163803.0
29881.1	38221.4	47536.5	57841.4	64782.0	81669.4	96668.7	163858.9
29953.7	38350.5	47544.0	57890.0	65086.9	81736.6	97255.9	164182.6
30056.9	38440.4	47683.6	57921.9	65251.1	81924.6	97284.1	164221.3
30120.2	39028.5	47781.9	57944.1	65282.8	81977.7	97311.8	164413.2
30218.5	39044.8	47877.8	57979.0	65340.1	82007.7	97333.3	165607.4
30225.4	39050.7	48574.1	58035.6	65343.5	82104.9	97348.6	167013.5
30244.7	39166.2	48652.5	58171.3	65486.6	82138.7	97443.3	167233.4

## Supporting Information

30249.4 30504.4	39169.5	48672.3	58200.4	65530.4	82245.1	97475.6	167502.1
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**Table S32.** CASSCF computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_9\text{H}_9)_2]^+$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

6						
Spin free States						
CASSCF						
87.7	36521.0	46664.6	56874.0	79231.1	97431.8	
88.7	36672.7	46664.6	56874.5	79236.1	97444.5	
135.6	36672.9	46825.1	56967.5	79236.1	97444.5	
135.7	36730.9	46825.1	56967.8	79260.1	98040.2	
168.3	36730.9	47000.7	57083.6	79260.1	98095.5	
178.4	36805.1	47000.7	57083.7	79283.7	98097.1	
186.7	36805.3	47196.7	57360.9	79283.7	98245.4	
186.7	36858.2	47198.3	57360.9	79435.0	98246.1	
1177.9	36874.2	47215.2	58356.9	79435.0	100652.3	
1177.9	36885.1	47215.2	58357.0	79613.8	100652.4	
7436.8	36885.1	47320.2	58617.1	79613.8	100654.6	
7441.2	36948.2	47320.7	58617.1	79822.2	100654.6	
7539.2	36948.5	47367.3	58720.8	79822.2	100667.2	
7539.2	37017.5	47608.7	58795.1	79915.6	100667.4	
7796.3	37017.9	47608.7	58795.3	79915.6	100668.8	
7796.5	37231.4	47903.6	58868.2	79935.3	100669.3	
7924.8	37278.1	47903.6	58868.2	79935.4	100676.8	
34006.8	37278.1	48031.2	58900.9	79944.3	100677.1	
34008.3	37543.0	48040.5	58903.3	79944.4	100678.8	
34990.8	37544.9	48041.4	59004.1	79964.2	100830.4	
24240.1	37566.6	48065.7	59004.4	79965.9	100830.4	
24241.4	37566.7	48070.1	59493.4	79984.5	102131.3	
24255.3	38005.0	48081.1	59493.8	79985.1	102131.3	
24255.3	38005.0	48081.1	59831.3	79992.5	102193.3	
24308.6	38088.3	48105.8	59867.4	84086.9	102193.3	
24308.7	38088.4	48105.9	59888.6	84088.9	102280.3	
24323.3	38088.9	48143.5	59888.6	84806.1	102280.3	
24323.4	38089.0	48143.6	59938.8	84807.2	102412.3	
24348.8	38248.5	48157.6	59939.2	84941.5	102412.6	
24348.9	38250.5	48157.6	60095.7	85270.8	102641.0	
24461.0	38324.0	48167.9	60100.2	85291.7	102641.7	
24461.0	38324.0	48167.9	60949.0	85293.0	102734.4	
24474.5	38379.4	50768.9	60949.0	85438.9	104675.8	
24603.9	38379.5	50768.9	61364.7	85439.1	104675.8	
24603.9	38424.5	50939.7	61364.7	85542.9	104936.1	
24667.4	38652.3	50983.8	61630.3	85542.9	104936.1	
24672.8	38652.3	50983.9	61630.3	85686.5	105069.5	
24708.9	41172.9	51089.1	61694.7	85687.9	105069.7	
24709.2	41172.9	51096.4	61715.4	86193.5	105299.0	
24766.7	41311.4	51254.7	61716.0	86193.5	105299.6	
24766.8	41311.4	51254.7	61764.7	86470.7	105394.3	
24772.4	41452.6	51286.9	61765.2	86470.7	105894.4	
24772.4	41452.6	51287.0	61787.5	86626.9	105894.4	
24813.8	41457.8	51393.4	61787.5	86626.9	105986.6	
24813.9	41458.4	51393.4	61810.3	86887.6	105986.6	
24887.5	41592.5	51751.6	61810.3	86887.6	106499.4	
24887.6	41599.5	51751.6	63260.6	87146.2	106508.0	
24894.5	41599.5	51784.6	63470.9	87146.2	106834.0	
24894.5	41694.7	51784.6	63471.1	87169.1	106835.0	
24937.5	41694.7	51825.4	63569.1	87169.2	106927.0	
25014.3	41750.0	51825.5	63569.1	87281.7	109737.5	
25014.3	41750.0	51846.7	63578.3	87285.9	109799.4	
25084.6	41792.9	51846.7	63580.1	87300.9	109800.1	
25084.6	41792.9	51877.8	66052.8	87302.7	109961.7	
25205.3	41845.1	51877.8	66760.7	87303.7	109965.6	
25209.0	41845.1	52026.8	66760.7	87544.5	110388.3	
25339.2	41914.2	52026.9	66993.9	87544.7	110388.3	
25339.8	41914.9	52133.6	66995.4	87564.8	114083.2	
25431.9	41989.0	52134.1	67161.8	87611.7	114084.4	
25597.4	41989.9	52169.5	67161.8	87612.4	114913.0	
25860.9	42028.8	54414.2	67296.3	87703.2	119064.8	
25861.6	42151.2	54414.2	67296.4	87703.3	119064.8	
25909.5	42193.4	54477.2	67324.9	87712.5	119564.9	
25909.5	42193.9	54481.2	67327.1	87712.6	119564.9	
25998.6	42428.6	54578.0	67354.4	92998.3	119831.1	
25998.7	42475.5	54578.0	67354.9	93000.3	119831.1	
26173.1	42475.5	54725.1	67367.6	93072.0	120007.5	

## Supporting Information

26173.1	42484.2	54725.7	69017.7	93072.1	120007.5
29057.6	42484.2	54895.0	69017.7	93175.8	120137.6
29085.3	42507.2	54926.8	69073.9	93175.8	120137.6
29085.6	42507.2	54926.8	69074.5	93176.7	120235.1
29096.7	42548.7	55180.0	69099.9	94787.0	120238.8
29096.7	42563.1	55180.0	69105.2	94802.6	120305.9
29149.0	42624.9	55288.3	69105.4	94803.0	120307.0
29152.0	42627.6	55288.3	69126.4	94904.9	120332.4
29152.3	42639.3	55370.6	69128.4	94906.9	122136.8
29153.3	42639.3	55376.7	69131.6	95066.6	122268.3
29209.2	42698.0	55376.8	69131.6	95066.6	122269.2
29209.2	42698.3	55380.7	69147.5	95115.1	122893.9
29229.5	42775.3	55381.1	69147.6	95115.1	122895.9
29229.5	42775.3	55406.7	69187.6	95151.0	131450.3
29349.4	42890.2	55407.9	69187.6	95151.0	131450.3
29349.4	42890.2	55448.9	70355.8	95434.0	131833.2
30450.5	42912.4	55448.9	70355.9	95434.0	131834.2
30450.5	42912.4	55456.0	70727.4	95771.8	132386.5
30536.4	43082.8	55456.0	71802.0	95771.8	132387.8
30540.1	43082.8	55664.6	72109.7	95898.5	132720.6
30540.1	43085.2	55664.7	72111.1	95899.1	139492.8
30562.1	43085.2	55801.0	72440.6	95915.9	139506.3
30564.8	43149.3	55801.0	72719.7	95973.6	139507.1
30625.4	43149.3	55990.1	72723.6	95976.2	139625.4
30625.4	43238.7	55990.2	72802.4	96044.0	139628.2
30726.6	43239.0	55997.5	72802.8	96044.1	139902.0
30726.6	43267.8	56001.4	72930.5	96090.5	139902.0
30802.0	44213.5	56001.7	72933.2	96090.5	140140.1
30802.0	44213.5	56041.0	76144.5	96187.2	140140.1
30822.8	44271.8	56041.0	76144.5	96187.2	141633.2
30822.8	44271.8	56102.6	76160.4	96228.7	142392.0
30864.3	44307.2	56102.6	76247.9	96228.7	142393.1
30864.3	44307.2	56148.0	76247.9	96236.0	145772.0
32297.1	44349.3	56148.0	76267.4	96237.3	145772.0
32297.3	44349.4	56182.6	76267.4	96239.5	145820.0
32474.2	44430.8	56182.8	76330.8	96239.5	145825.1
32477.8	44430.8	56221.9	76331.5	96251.7	145851.3
32692.9	44435.5	56224.6	76339.3	96251.9	145851.4
33863.6	44435.5	56230.2	76341.4	96263.8	145944.4
33868.5	44485.3	56241.2	76341.6	96516.4	145944.9
33886.8	44489.9	56241.8	77631.4	96516.4	145987.6
33893.1	44497.3	56282.6	77632.0	96777.5	146445.2
33893.2	44497.4	56282.6	77773.7	96777.5	146445.2
33911.1	44527.9	56314.8	77774.0	97054.8	173637.7
33911.1	44527.9	56314.8	77818.3	97054.8	173646.0
34030.7	44569.5	56433.9	78325.9	97118.2	174986.0
34030.8	44663.0	56444.5	78325.9	97118.2	174986.4
34087.5	44664.6	56445.4	78984.5	97194.3	175210.9
34087.5	44798.4	56519.2	79026.8	97241.3	177247.4
36353.7	44832.9	56519.2	79027.1	97242.0	177247.4
36353.7	44833.6	56571.0	79128.9	97342.9	177435.1
36397.5	44893.4	56574.7	79130.8	97345.3	177436.1
36397.5	45510.6	56774.1	79199.4	97345.3	177495.7
36477.9	45511.3	56774.1	79199.4	97346.7	177805.6
36477.9	45639.5	56816.1	79231.1	97431.8	177806.8
36521.0	45647.0				

**Table S33:** NEVPT2 computed 21 roots of sextet (red) along with 224 roots of quartet and 490 roots of doublet states for  $[\text{Dy}(\text{C}_9\text{H}_9)_2]^+$  complex. All the values are reported herein  $\text{cm}^{-1}$ .

6					
Spin free States					
NEVPT2					
160.1	34866.8	44794.3	54765.7	74937.0	91136.1
160.8	34943.2	44794.4	54765.7	74962.6	91142.1
164.3	34943.3	44830.6	54798.9	74962.6	91214.5
164.6	35231.9	44830.7	54799.8	75058.3	91214.5
199.2	35235.2	44888.9	54929.6	75058.3	91296.7
199.4	36968.4	44889.0	54929.7	75074.0	91296.7
280.0	36968.4	44914.6	54964.6	75074.0	91330.7
298.9	37051.3	44915.2	54964.7	75563.8	91330.8
1447.5	37051.3	44966.8	55428.8	75564.8	93313.4
1447.5	37163.9	49009.7	55442.5	75634.6	93313.4
6233.8	37163.9	49009.7	55453.5	76289.4	93315.7
6242.7	37224.6	49108.5	55453.8	76289.5	93315.8
6390.3	37224.6	49108.5	55747.2	76503.2	93335.3

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6390.3	37435.2	49303.9	55747.3	76503.2	93335.5
6695.7	37436.1	49366.9	55751.8	76713.1	93337.9
6696.3	37536.3	49366.9	55864.4	76713.1	93339.7
6847.9	37536.3	49502.6	55864.6	76783.5	93359.2
28754.8	37537.9	49515.6	55931.1	76783.6	93359.6
28756.3	37538.0	49667.4	55931.7	76871.1	93363.0
29795.4	37633.7	49667.4	55933.2	76871.1	93565.4
23273.8	37634.0	49715.9	55944.6	76945.3	93565.4
23286.2	37652.8	49716.0	55996.1	76945.4	94218.9
23499.8	37653.0	49805.1	55996.2	77003.0	94218.9
23500.6	37682.4	49805.1	56264.8	77003.1	94252.2
23526.5	37682.7	49903.5	56728.0	77042.0	94252.3
23527.4	37696.3	49903.5	56728.0	77044.4	94258.3
23551.6	37696.4	50115.7	56750.6	77072.1	94258.4
23566.9	37716.7	50115.7	56750.8	77072.8	94443.8
23567.0	37889.5	50461.7	56808.0	77082.1	94443.8
23568.3	37892.3	50465.4	56998.1	78783.2	94508.1
23689.2	37994.0	50465.4	57004.2	78800.2	94508.3
23828.9	37994.1	50482.2	57425.0	78803.3	94603.9
23829.1	38073.0	50483.0	57425.0	79038.4	94604.1
23835.7	38073.0	50518.9	57886.5	79038.6	94658.2
23835.8	38129.1	50528.1	57886.5	79116.7	94658.9
23937.5	38373.5	50592.3	58161.9	79116.9	94796.2
24083.4	38373.6	50592.4	58190.7	79268.5	94797.0
24402.5	38535.7	50661.7	58191.0	79268.5	94913.4
24406.9	38536.3	50661.8	58202.0	79384.6	94953.7
24433.2	38700.3	50662.7	58202.4	79387.4	94954.4
24433.4	39362.2	50663.2	58284.2	79851.8	95069.3
24467.5	39480.0	50716.4	58285.9	79851.9	96491.8
24467.6	39480.7	50716.4	58375.0	80270.7	96491.8
24603.4	39815.0	50730.9	58375.1	80326.9	96559.2
24603.5	39815.0	50730.9	58375.5	80328.2	96559.2
24701.8	40020.6	50849.7	58375.6	80352.1	97122.0
24701.8	40045.7	50849.7	58644.8	80352.7	97137.0
25086.4	40285.2	50888.6	58971.5	80489.0	97530.0
25086.4	40303.4	50888.6	58971.7	80490.3	97530.9
25909.0	40436.2	50920.9	59036.0	80507.1	97616.9
25909.8	40436.2	50921.0	59036.2	80508.3	10040.1
26083.1	40920.5	50955.8	59102.3	80528.0	100521.7
26083.2	40946.3	50955.9	59105.3	80528.0	100522.2
26602.5	40946.6	51010.0	59957.3	80709.4	100699.4
26604.4	41292.6	51010.0	60997.7	80709.7	100707.0
26677.4	42226.6	51168.9	60999.3	81195.6	101202.0
26679.6	42227.2	51169.4	62921.3	81195.7	101202.0
26722.8	42295.1	51328.7	62921.4	81517.0	104070.7
26722.9	42306.9	51329.2	63399.1	81517.0	104071.9
26870.8	42474.8	51382.6	63399.2	81606.8	104937.3
26870.8	42475.1	51731.7	63522.0	81624.2	110032.5
27022.8	42549.2	51739.3	63525.7	81630.2	110032.8
27022.8	42549.3	51765.7	63525.8	81631.5	110587.4
27168.9	42561.4	51765.7	63526.9	81632.5	110587.4
27178.0	42561.4	51981.6	63561.9	84917.6	110843.2
27185.7	42586.5	51981.6	63562.6	84921.2	110843.3
27255.4	42586.5	52047.8	63581.8	85007.2	110977.5
27257.1	42630.1	52061.0	65129.1	85007.4	111017.4
29238.6	42641.2	52061.9	65332.1	85112.1	111017.4
29282.4	42677.8	52081.6	65332.3	85112.2	111118.7
29282.7	42678.6	52082.3	65428.2	85155.3	111119.8
29296.8	42685.3	52167.3	65429.7	87324.8	111155.1
29296.9	42685.4	52167.3	65800.3	87345.9	111155.4
29380.4	42738.7	52223.0	65902.8	87346.3	111268.2
29387.5	42738.7	52230.7	65902.8	87443.3	111274.6
29409.2	42752.5	52344.5	65979.5	87443.4	111367.6
29409.5	42757.9	52578.9	65990.9	87499.1	111369.1
29479.2	42757.9	52578.9	65991.4	87501.9	111398.2
29479.2	43047.9	52760.0	66005.7	87717.9	111807.9
29527.1	43050.7	52760.0	66008.5	87718.0	111810.4
29527.2	43233.9	52886.4	66047.0	87744.9	118638.4
29645.1	43233.9	52960.2	66050.9	87745.0	118638.4
29645.1	43267.9	52960.2	66094.6	87845.6	118967.9
29656.2	43268.7	52970.5	66094.6	87845.7	118969.3
29656.5	43344.1	52971.0	66103.9	87950.8	119622.5
29883.3	43386.9	53073.3	66103.9	87951.5	119624.1
29890.2	43386.9	53073.3	66147.3	87992.6	120080.5
30199.5	43538.7	53084.4	66147.4	88004.5	127539.3
31288.1	43538.7	53085.9	66339.2	88014.1	127542.9
31288.1	43702.1	53533.5	66343.7	88458.7	127543.7



## Supporting Information

31403.8	43702.1	53533.5	66525.2	88681.4	127661.4
31403.8	43786.5	53610.6	66870.7	88682.0	127667.5
31403.8	43796.7	53613.6	66878.1	88683.7	127980.2
31428.3	43796.7	53668.0	67053.6	88683.7	127980.3
31433.2	43830.9	53668.0	67054.2	88831.8	128231.1
31507.4	43831.0	53682.5	68773.6	88832.6	128231.1
31507.4	43845.0	53682.5	69963.7	89092.2	128362.9
31642.9	43845.0	53741.6	69965.6	89101.1	129217.1
31642.9	43871.9	53741.6	70028.8	89559.3	129218.5
31739.7	43871.9	53762.2	70028.8	89559.3	133083.6
31739.7	43938.7	53762.6	70061.2	89717.3	133083.9
31775.9	43941.5	53854.7	70061.2	89717.3	133133.4
31775.9	43945.2	53862.8	70099.9	89828.3	133143.1
31830.6	43945.6	53862.9	70100.4	89905.0	133158.2
31830.6	44004.5	53930.3	70111.9	89968.3	133158.2
32650.6	44015.3	53930.4	70114.1	89968.3	133304.5
32658.5	44015.3	53960.9	70114.3	89972.4	133305.9
32661.3	44017.3	53981.2	70272.1	90043.9	133362.1
32707.2	44056.1	53981.3	70272.4	90251.1	133919.3
32707.4	44058.0	54003.0	70432.6	90251.2	133919.7
32722.2	44146.1	54013.4	70433.5	90276.5	156816.0
32722.2	44168.5	54039.9	70576.4	90276.5	156831.7
32894.3	44168.5	54040.6	71146.9	90404.5	158153.9
32894.5	44370.1	54471.5	71146.9	90404.9	158154.1
32977.5	44370.3	54471.5	74612.3	90468.4	158499.2
32977.5	44510.5	54521.0	74672.8	90583.0	159626.4
33617.0	44510.5	54596.0	74673.2	90583.0	159626.6
33952.2	44588.5	54596.0	74758.5	90851.6	159795.7
33952.7	44588.5	54607.2	74761.6	90946.3	159885.4
34465.8	44608.6	54607.2	74817.0	90946.7	159886.3
34466.7	44608.6	54614.3	74819.7	90948.1	160432.6
34771.5	44772.9	54614.6	74937.0	90948.4	160435.0
34866.7	44776.1				

**Table S34:** CASSCF computed the spin-orbit states for  $[\text{Dy}(\text{C}_9\text{H}_9)_2]^+$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

6							
Spin Orbit States							
CASSCF							
0.0	30266.0	39858.0	49823.5	60511.9	69887.1	89072.3	104520.6
20.1	30369.6	39872.3	49880.1	60542.9	69904.2	89473.3	104556.3
48.9	31299.9	39926.2	49912.6	60546.6	70244.5	89614.5	104586.3
71.0	31347.7	39952.4	49955.5	60549.7	70566.7	89685.8	104829.2
92.5	31348.5	40003.4	50096.4	60588.0	70763.4	89743.2	105351.3
156.7	31403.6	40076.8	50191.1	60606.2	70857.4	89751.7	105514.2
369.7	31432.4	40086.5	50209.5	60800.4	71419.4	89813.8	105562.7
1081.6	31476.6	40175.1	50292.3	60974.6	71819.3	89828.1	105579.1
3037.1	31488.1	40191.6	50315.4	60994.7	72002.6	89973.7	105833.0
3039.2	31539.5	40200.2	50320.1	60996.6	72040.3	90039.1	105915.8
3063.5	31598.5	40228.5	50321.6	61002.9	72052.4	90093.3	105936.0
3102.3	31616.7	40244.4	50349.8	61100.3	72058.1	90353.2	105940.7
3233.8	31619.6	40250.7	50408.7	61101.2	72547.1	90381.3	106053.0
3426.8	31635.0	40309.9	50453.8	61191.6	73838.6	90423.9	107064.1
3840.1	31662.5	40368.6	50458.7	61197.5	74019.9	90488.6	107072.0
5248.5	31806.2	40412.8	50473.7	61316.4	74136.1	90594.8	107100.6
5249.0	31824.0	40434.0	50494.8	61367.4	74371.8	90684.5	107121.3
5278.6	31876.1	40475.8	50509.1	61387.9	74417.2	90792.6	107154.1
5421.6	31952.4	41466.5	50527.0	61399.2	74429.2	90967.0	107189.2
5633.2	32259.7	41643.7	50582.2	61414.8	74444.8	90983.9	107214.9
5903.6	32264.2	41647.1	50619.7	61446.3	74457.3	90999.1	108794.3
6968.1	32441.3	41671.6	50668.2	61520.2	74468.2	91144.8	109113.5
6969.9	32474.3	41720.7	50678.1	61571.8	74495.9	91282.3	109124.5
7044.8	32488.2	42201.4	50743.4	61587.5	74501.9	91337.9	109155.8
7288.9	32815.0	42298.3	50777.4	61610.6	74505.6	91404.9	109178.3
7560.0	33138.5	42369.2	50851.3	61638.8	74519.5	91410.6	109272.0
8277.4	33247.9	42384.0	50861.4	61682.2	74523.7	91445.0	109307.0
8295.2	33258.2	42390.7	51028.0	61690.8	74539.4	91454.8	109360.7
8507.6	33344.4	42489.8	51046.8	61697.7	74544.5	91465.2	109547.5
8913.5	33392.6	42492.1	51104.0	61722.7	74608.1	91477.8	109558.2
9248.2	33394.3	42497.9	51171.7	61763.7	74840.1	91495.2	109730.6
9305.7	33400.6	42528.0	51637.2	61798.6	74844.5	91573.5	109890.9
9426.4	33432.7	42565.3	51669.7	61851.4	75076.6	91589.2	109915.1
9480.3	33437.6	42585.2	51692.9	61854.9	75083.9	91608.3	110014.2
9482.9	33451.4	42669.4	51849.6	61891.0	75087.0	91609.2	110134.5
9587.2	33486.4	42679.6	52045.5	61902.6	75119.2	91707.1	110327.0

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9702.1	33618.9	42753.2	52079.9	62455.2	75122.7	93390.9	110596.4
9773.5	33623.8	42784.0	52139.9	62464.9	75123.3	93900.4	110741.9
10042.3	33630.3	42843.8	52369.0	62644.6	75165.9	94128.1	112533.8
10783.3	33664.3	42860.0	52555.5	62767.1	75396.7	94147.7	112593.1
10947.0	33726.9	43896.2	52569.6	62814.6	76893.4	94173.0	112668.1
10962.5	33763.7	44056.8	52578.7	62853.5	76974.5	94613.0	112737.3
10966.7	33775.2	44243.9	52599.2	62897.2	77226.1	94974.3	113167.6
10969.5	33882.8	44422.3	52606.7	62926.9	77941.5	95285.3	113249.7
12514.4	33903.9	44580.7	52629.4	62961.1	78044.6	95514.9	113363.5
12539.1	34116.3	44581.4	52650.0	62968.8	79303.6	95640.8	113544.4
12635.8	34273.0	44617.6	52660.8	63038.8	79675.6	95694.4	113721.0
12660.6	34324.7	44714.7	52675.9	63081.9	80414.1	95716.2	113828.2
13804.7	35156.3	44724.6	52945.9	63087.0	80631.6	96473.0	113943.7
13868.7	35228.2	44741.3	53876.8	63125.2	80653.6	96825.1	114141.9
13906.1	35244.7	44753.6	53992.6	63184.4	80759.8	96941.6	114540.7
14633.7	35269.0	44763.2	54042.1	63229.6	80849.7	97074.2	114774.2
14716.4	35311.1	44769.8	54074.7	63290.3	80893.0	97104.8	114880.0
15189.8	35349.3	44799.6	54117.6	63325.2	80904.5	97178.8	115693.1
23559.5	35369.5	44840.7	54188.0	63355.0	80918.0	97393.4	115705.5
23613.5	35369.6	44862.4	54192.5	63366.6	80926.1	97409.9	115948.9
23782.8	35397.9	44879.7	54227.9	63415.7	80930.9	97480.9	117001.4
23856.0	35551.3	44886.4	54279.6	63416.2	80987.2	97651.0	117242.4
24049.5	35582.5	44911.1	54315.4	63485.5	81044.3	97657.9	117264.1
24495.3	35586.1	45017.1	54379.3	63524.7	81115.1	97772.0	117286.0
24519.6	35646.3	45199.5	54428.2	63551.7	81145.0	97819.1	117923.5
24565.5	35702.6	45369.2	54485.0	63576.5	81158.6	99401.3	119935.5
24579.8	35781.5	45455.9	54602.7	63614.2	81299.4	99637.3	120673.7
24632.2	35784.8	45482.8	54691.7	63746.1	81375.8	99900.4	124397.0
24653.3	35971.1	45493.6	54785.9	63746.6	81388.8	99908.6	124808.0
24836.4	36128.7	45525.6	54868.1	64632.1	81408.1	99924.8	125015.3
25119.9	36299.9	45556.5	54897.7	64801.4	81471.9	99949.0	125141.8
26167.5	36403.4	45560.0	55059.6	64883.2	81476.6	99983.5	125227.3
26283.1	36610.4	45886.0	55199.0	64913.8	81531.6	100023.1	125289.2
26397.4	36676.1	45953.4	55248.3	64920.3	81586.0	100028.5	125325.3
26472.1	36772.9	45968.7	55272.2	64930.2	81651.3	100088.1	125356.8
26488.7	36779.1	45996.6	55338.0	65267.2	81732.1	100140.7	125930.9
26494.7	36784.6	46082.6	55428.1	65517.3	81738.7	100161.2	126242.0
26536.7	36796.9	46103.6	55466.1	65520.1	82066.3	100220.1	126445.1
26565.8	36817.7	46108.4	55490.2	65708.0	82088.8	100981.1	126595.8
26593.0	36823.7	46144.5	55521.0	65743.7	82096.6	101030.4	126713.5
26671.8	36831.1	46231.2	55828.2	65747.1	82215.0	101140.7	126801.0
26719.8	37054.7	46356.3	56074.8	65758.1	82553.3	101209.1	126849.2
26729.5	37292.8	46377.0	56211.0	65774.3	83814.6	101220.6	127785.1
26766.2	37303.1	46381.0	56244.0	65779.5	83985.2	101224.1	127926.2
26880.0	37307.2	46473.9	56248.4	65818.7	84073.7	101260.3	128444.3
27029.5	37325.7	46539.1	56258.2	65858.5	84140.4	101264.3	128940.4
27144.3	37328.2	46589.0	56265.6	65878.6	84251.2	101645.5	129663.9
27201.8	37400.9	46647.0	56267.6	65920.2	84324.4	101853.2	135875.9
27577.9	37406.9	46887.9	56274.9	65932.2	84325.3	101969.9	136249.9
27597.7	37414.5	47193.2	57088.5	66416.1	84354.5	102013.9	136775.6
27654.0	37486.3	47232.3	57229.2	66608.2	84369.3	102057.3	138787.3
27670.0	37674.2	47241.2	57262.9	66686.2	84380.2	102105.2	139167.3
27728.2	37752.3	47260.1	57349.7	66852.6	84417.8	102106.5	139650.4
27758.8	37771.9	47289.7	57463.6	66857.1	84441.6	102196.3	139963.8
27857.3	37793.6	47311.8	57464.5	66871.6	84510.6	102514.5	143780.4
28007.2	37816.0	47373.8	58287.0	66915.9	84527.5	102601.3	143835.1
28033.0	37968.7	47441.7	58526.0	67005.2	84548.1	102637.9	143936.0
28080.5	38177.1	47490.6	58752.0	67031.9	84550.4	102838.4	144100.9
28100.2	38195.4	47615.4	58771.6	67044.4	84567.8	103020.2	144290.2
28139.6	38268.8	47715.0	58789.8	67084.0	84674.4	103057.8	146455.6
28189.9	38279.6	47773.0	58827.2	67087.7	84677.0	103137.5	146562.6
28197.7	38283.6	48484.1	58832.6	67135.7	84691.0	103269.0	146737.1
28241.7	38326.0	48491.3	58845.7	67137.9	84784.4	103360.7	146836.7
28279.6	38374.5	48528.1	58848.4	67158.7	84788.4	103398.9	147121.6
28293.3	38417.9	48635.4	59024.6	67177.3	84801.6	103447.2	147261.5
28310.0	38630.6	48702.9	59106.2	67191.9	84863.8	103458.3	149358.2
28342.6	38632.3	48723.4	59190.2	67202.7	84896.4	103465.4	149919.6
28351.5	38641.9	48753.6	59216.3	67231.4	85030.5	103492.5	149955.1
28409.2	38647.6	48809.9	59219.0	67235.1	85042.3	103569.8	150038.1
28409.8	38648.1	48873.8	59238.2	67239.1	85205.0	103752.4	150064.8
28417.9	38680.4	48883.4	59348.7	67614.1	85268.1	103850.9	150154.0
28448.5	38724.1	48886.4	59679.7	67927.5	85280.2	103857.7	150553.7
28449.9	38749.9	48895.6	59714.7	68060.2	85333.6	103860.6	153421.0
28504.7	38752.4	48913.3	59772.4	68076.2	85421.2	103888.7	153457.0
28523.8	38772.7	49017.0	59856.6	68077.7	85428.2	103925.4	153553.9
28582.9	38774.7	49130.9	59888.9	68091.6	85490.5	103951.2	153579.3
28599.3	38789.3	49197.2	59912.3	68102.6	85528.9	103964.0	154145.0

## Supporting Information

28610.6	38925.6	49227.9	59927.9	68765.7	85553.5	104006.6	177202.8
29734.0	39033.4	49420.4	59963.2	68828.7	85570.4	104048.8	178351.6
29741.6	39090.5	49479.8	59974.8	69083.1	85579.7	104053.8	178996.5
29764.1	39119.2	49592.8	60021.1	69163.7	86099.4	104150.5	181492.1
29795.9	39179.7	49602.3	60073.6	69283.3	86155.3	104184.6	181819.0
29804.6	39225.0	49607.2	60098.9	69293.7	86299.9	104227.4	181914.0
29927.8	39228.1	49629.2	60290.3	69352.7	86819.4	104273.7	181922.9
30018.4	39251.3	49682.2	60309.6	69471.6	87015.2	104310.9	182187.5
30030.3	39433.5	49685.9	60319.4	69683.1	88670.9	104319.4	183057.3
30098.8	39525.8	49715.3	60389.1	69776.2	88715.4	104348.4	185008.0
30125.9	39591.6	49725.1	60436.0	69785.8	88981.2	104421.3	185042.1
30224.5	39656.9	49805.8	60503.7	69851.5	89040.6	104513.4	185362.1
30256.6							

**Table S35:** NEVPT2 computed the spin-orbit states for  $[\text{Dy}(\text{C}_9\text{H}_9)_2]^+$  complex. All the values are reported here in  $\text{cm}^{-1}$ .

6							
Spin Orbit States							
NEVPT2							
0.0	30597.6	39288.9	48895.5	58336.2	66267.4	82607.4	97765.7
29.0	30622.8	39295.8	48981.6	58381.1	66624.8	82651.6	97836.4
73.0	30705.0	39310.9	49033.3	58525.1	66681.4	82669.8	97857.7
105.5	30827.7	39350.2	49474.2	58540.4	66722.1	82811.2	97861.4
120.4	30904.3	39354.0	49565.9	58567.1	66732.6	82954.9	97865.5
171.8	31060.8	39372.0	49573.8	58571.7	66809.3	83083.9	97940.4
408.0	31072.7	39404.8	49690.0	58593.1	66809.8	83147.5	98102.8
1329.5	31108.9	39442.6	49714.3	58613.8	67600.7	83158.7	98109.9
3041.6	31111.8	39478.9	49749.8	58644.1	67731.8	83356.2	98127.8
3063.1	31162.4	39509.0	49771.1	58645.2	67851.7	83456.7	98180.6
3106.8	31237.7	39512.6	49782.7	58671.0	68038.6	83530.4	98281.5
3118.7	31255.5	39540.5	49837.6	58688.0	68115.3	83588.3	98285.4
3280.7	31330.8	39555.9	49943.4	58712.6	68230.6	83806.7	98343.5
3497.0	31469.1	39608.1	50015.8	58717.9	68243.6	83831.7	98809.0
4028.6	31483.2	39611.4	50040.5	58731.6	68253.4	83907.3	99043.8
5247.2	31545.6	39674.8	50063.3	58827.9	68259.9	83923.1	99330.0
5250.8	31549.1	39709.3	50129.1	58830.5	68288.2	83974.1	99478.3
5259.7	31585.2	39757.7	50268.2	58902.5	68399.0	84003.1	99631.6
5430.9	31643.0	39768.9	51580.4	58909.8	68685.3	84016.6	99659.8
5688.2	31700.9	40139.7	51676.5	58960.9	69211.8	84081.0	99820.4
6028.5	31840.7	40185.1	51697.9	59059.9	70115.0	84213.0	99836.2
6931.6	31941.8	40201.3	51711.2	59228.5	70434.6	84258.7	99856.7
6969.1	32064.3	40325.3	51809.0	59290.3	70474.2	84286.7	99887.1
7010.9	32331.4	40332.8	51842.0	59329.6	70486.9	84307.8	99893.8
7316.8	32343.1	40381.0	51851.5	59355.4	70513.1	84350.9	99906.5
7654.4	32446.5	40432.8	51883.1	59382.8	70737.5	84361.7	99945.3
8207.5	32510.8	40493.1	51887.7	59389.4	70791.7	84367.3	99994.3
8208.9	32535.4	40561.8	51921.5	59407.1	71094.2	84614.2	100541.3
8264.3	32563.2	40611.6	51962.3	59464.0	71273.1	84678.0	100593.5
8276.1	32638.3	40680.1	51975.4	59473.9	71320.3	84818.8	100632.8
8290.0	32689.1	40705.4	52000.1	59487.2	71354.0	84835.3	100662.5
8367.0	32695.8	40736.8	52010.3	59592.6	71369.9	84870.6	100703.1
8523.4	32727.5	40868.6	52066.8	59613.3	71445.4	84899.6	100730.0
8528.2	32753.7	41012.2	52111.2	59634.9	71451.2	84941.2	100809.6
8628.5	32784.4	41493.6	52141.6	59666.5	71463.2	85039.5	100812.6
9009.9	32962.2	41543.3	52151.7	59674.5	71486.8	85155.9	101080.9
9206.7	32992.9	41972.3	52190.2	59680.2	71773.0	86125.2	101354.4
9254.2	33216.1	42112.5	52209.1	59682.8	71853.4	86699.9	101529.8
9582.1	33290.1	43159.5	52215.5	59706.9	71857.3	86919.0	102998.3
9791.1	33328.9	43196.6	52217.2	59730.2	71886.3	86948.7	103018.0
9797.2	33609.1	43302.3	52280.0	59770.6	71911.4	86973.9	103148.8
9806.0	33784.7	43346.4	52350.5	59823.7	71935.8	88092.0	103256.1
9809.9	33797.4	43477.1	52392.4	59829.7	71985.7	88416.9	104237.0
10151.8	33818.3	43605.9	52422.3	59846.9	71991.7	88751.5	104305.6
11322.5	33824.4	43635.7	52425.0	59867.2	72034.8	88761.6	104324.4
11343.3	33832.2	43652.1	52608.1	59917.5	72784.7	88875.4	104450.1
11479.9	33891.3	43693.2	52646.6	59935.4	72932.9	88916.8	104498.1
11497.7	33909.3	43726.8	52696.0	59980.9	74128.1	89023.2	104522.1
12591.1	33916.8	43750.9	52702.6	60046.6	74311.1	89146.1	104744.6
12673.7	34057.2	43760.3	52726.7	60051.0	74319.4	89178.0	104890.5
12712.9	34288.9	43904.0	52806.6	60241.5	74631.8	89190.3	104949.0
13395.8	34483.0	43992.1	52861.6	60430.9	74733.7	89362.3	105214.9
13502.7	34573.7	44042.1	52941.2	60524.6	74750.1	89437.7	105324.8
13961.3	34599.6	44063.7	52999.5	60526.8	74759.8	89880.5	106273.5
22443.9	34646.1	44085.0	53502.1	60528.9	74762.6	89955.8	106309.8
22486.7	34667.2	44291.5	54113.5	60536.0	74784.8	90283.7	106618.1

## Supporting Information

22567.7	34697.2	44405.0	54127.3	60544.5	74793.7	90582.8	107635.2
22654.9	34716.2	44752.6	54156.0	61089.2	74834.1	90988.0	107842.3
22816.7	34718.3	44809.4	54167.5	61247.8	74875.0	91250.1	107854.2
24014.4	34738.1	44873.6	54259.2	61287.4	74877.2	91350.5	107880.8
24055.3	34758.3	45065.4	54299.7	61335.3	74881.4	91388.8	107972.2
24075.4	34767.2	45627.7	54419.1	61348.7	74893.9	91730.4	109852.6
24180.5	34798.4	45673.5	54483.7	61444.6	74956.5	92268.0	110589.1
24237.7	35077.1	45740.4	54500.6	61504.4	74983.5	92304.0	115437.6
24534.8	35190.2	45749.8	54567.4	61537.2	75009.2	92334.0	115895.0
24743.5	35415.5	45750.7	54746.4	61659.6	75069.6	92459.4	116091.5
24867.3	35451.3	45801.4	54903.9	61679.9	75231.2	92494.2	116213.2
24958.8	35532.9	45825.7	54907.8	61739.5	75279.5	92781.1	116301.6
25028.2	35540.4	45847.4	54909.2	61774.7	75333.4	92794.2	116354.3
25128.4	35552.6	45912.0	54985.7	61777.6	75362.9	93096.9	116386.8
25195.7	35634.2	45956.6	54994.5	61778.8	75368.4	93217.1	116443.0
25290.5	35688.4	45963.0	55015.5	62029.3	75427.4	93234.5	116721.6
245308.7	35720.6	45976.3	55039.2	62258.4	75442.6	93244.1	116860.5
26776.0	35726.1	46015.2	55045.2	62259.9	75561.1	93615.7	116980.6
26793.5	35823.5	46034.6	55057.1	62322.9	75592.0	93641.6	117285.1
26860.4	35958.9	46062.3	55077.6	62384.2	75605.2	93649.4	117400.4
26977.4	35961.9	46105.2	55113.3	62405.0	75619.1	93653.3	117483.8
27093.7	36096.0	46121.4	55120.9	62429.1	75672.8	93760.2	117635.4
27153.2	36126.2	46187.0	55170.7	62490.4	77183.9	93800.6	117767.2
27163.2	36163.9	46263.9	55209.2	62528.8	77578.0	93809.8	117838.3
27264.6	36201.6	46332.6	55213.7	62551.2	77769.4	93885.2	117878.6
27413.7	36252.9	46347.7	55402.6	62700.5	78002.2	93894.8	117947.7
27554.2	36371.6	46371.2	55566.8	62760.8	78081.5	93918.2	118600.6
27722.9	36381.1	46465.2	55662.3	62945.4	78237.5	93956.6	123154.2
28104.3	36860.3	46593.7	55670.1	62950.6	78311.1	93961.0	123481.1
28135.5	36871.4	46720.9	55685.8	62977.8	78346.1	93974.6	124113.3
28344.8	36894.2	46728.1	55687.5	62990.9	78396.6	93997.1	126101.2
28454.1	36957.6	46742.0	55746.7	63016.6	78818.0	94109.2	126433.2
28526.0	36965.8	46807.7	55820.7	63208.4	79131.3	94209.8	126988.1
28539.5	36965.8	46831.7	55827.2	63249.2	79347.6	94680.7	127396.6
28563.5	37049.7	46838.1	55887.7	63282.2	79454.7	94869.7	131785.9
28609.8	37063.9	46883.4	55957.2	63395.7	79483.5	94913.6	131835.3
28639.1	37141.7	46910.8	56031.2	63610.2	79498.5	94925.7	131942.2
28752.3	37167.2	47013.4	56082.0	63676.7	79523.1	94968.1	132144.5
28801.7	37167.5	47043.9	56151.7	63901.0	79556.2	95276.6	132353.3
28836.7	37282.2	47049.4	56193.4	63922.8	79641.3	95630.3	133490.2
28876.2	37444.8	47052.7	56196.2	64001.8	79663.9	95692.4	134098.2
28913.6	37555.7	47056.0	56291.8	64043.0	79798.8	95699.4	134517.1
28962.8	37579.6	47102.7	56317.0	64084.0	79973.2	95772.0	134610.4
29020.5	37629.8	47149.9	56353.5	64122.7	79985.5	95898.7	134918.0
29115.0	37638.6	47164.0	56381.3	64138.1	80004.5	95955.5	135222.1
29128.6	37647.5	47179.3	56447.6	64143.3	80100.5	95982.2	136206.1
29152.5	37651.7	47210.0	56501.3	64166.1	80111.5	96047.7	137234.2
29160.6	37660.4	47233.4	56541.2	64180.3	80187.5	96079.4	137274.2
29204.6	37661.8	47320.1	56625.2	64292.7	80611.0	96113.8	137363.6
29213.3	37691.6	47362.3	56649.8	64375.5	80697.9	96131.6	137411.6
29409.3	37721.3	47371.7	56770.3	64382.9	80706.2	96480.8	137524.1
29425.1	37779.5	47385.3	56785.0	64401.2	80896.4	96495.4	138030.9
29454.1	37793.1	47428.3	56878.1	64411.5	81084.7	96602.0	140779.3
29512.9	37898.6	47476.3	56878.3	64466.6	81260.1	96639.9	140834.4
29693.2	37917.2	47504.8	56989.0	64468.9	81418.9	96644.5	140930.0
29720.6	37937.7	47507.0	57048.5	64537.0	81500.1	96648.8	140962.5
29793.3	37985.5	47528.7	57152.9	64727.8	81536.2	96705.3	141636.9
29868.8	38019.7	47539.8	57177.2	64830.9	81579.7	96706.0	160348.5
29921.0	38057.0	47547.6	57450.5	64834.1	81613.7	96738.2	161474.9
29924.3	38173.0	47557.7	57842.6	64844.7	81629.8	96739.5	162117.0
29942.3	38261.9	47581.8	57945.1	64855.1	81789.4	96842.7	164215.2
29948.8	38313.1	47601.0	58028.9	64859.8	81849.8	96910.0	164370.0
29978.8	38322.1	47625.3	58038.8	65225.6	81951.3	97462.6	164574.6
30141.3	38449.9	47778.4	58044.6	65380.6	82108.0	97471.3	164581.8
30219.3	38521.7	47867.2	58061.2	65416.8	82125.2	97605.8	164879.2
30249.7	39108.4	47939.6	58065.2	65457.8	82179.5	97614.8	166075.2
30316.4	39130.4	48690.5	58148.7	65486.0	82304.0	97654.9	167487.7
30360.3	39154.2	48704.4	58250.7	65616.8	82351.7	97687.2	167650.2
30398.4	39230.2	48777.7	58276.3	65651.8	82512.9	97736.8	167946.1
30523.2	39264.9						

**Table S36:** NEVPT2 computed the eight low lying Kramer Doublets for  $1_Q - 6_Q$ . All the values are reported here in  $\text{cm}^{-1}$ .

Spin Orbit States

NEVPT2						
	1 <sub>Q</sub>	2 <sub>Q</sub>	3 <sub>Q</sub>	4 <sub>Q</sub>	5 <sub>Q</sub>	6 <sub>Q</sub>
<b>KD1</b>	0.00	0.0	0.00	0.00	0.00	0.00
<b>KD2</b>	249.35	138.4	94.70	174.24	122.23	32.11
<b>KD3</b>	458.34	258.6	175.69	323.82	241.85	71.80
<b>KD4</b>	628.11	360.2	244.15	450.10	355.30	113.84
<b>KD5</b>	760.97	442.4	300.66	553.21	457.62	154.10
<b>KD6</b>	858.70	502.6	344.09	631.17	542.19	187.85
<b>KD7</b>	922.61	543.0	372.46	686.84	602.22	213.69
<b>KD8</b>	954.46	591.8	392.03	733.54	634.60	226.74

Table S37: DFT-optimized geometry coordinates

1		
Dy	0.000000000000	0.000000000000
Si	2.078484935773	-2.003997946204
Si	-2.079447509189	2.003544441301
Si	2.079137940498	-2.007359280089
Si	-1.975262034051	-2.102628647759
Si	1.983851639157	2.102186784756
Si	1.976745847057	2.105022645633
Si	-1.975803382377	-2.105952410060
Si	-2.071575468433	2.007637098145
C	-0.739970772721	0.728853817074
C	0.703554911154	0.765727946139
C	-0.703649633882	-0.765519450303
C	0.739970772721	-0.728853817074
C	0.709153077271	0.764146764519
C	0.742297035908	-0.730526546358
C	-0.734418115847	0.730526546358
C	-0.701345066962	-0.763976369445
H	1.581377415594	-3.310560386961
H	3.271561082064	-1.557833261432
H	2.600924170861	-2.292117495435
H	-2.496205285057	-2.405658295158
H	-3.182791598544	-1.729126878578
H	-1.407217774467	-3.386653958658
H	1.413977484645	3.385808862591
H	2.512452614133	2.406444123372
H	3.186331265163	1.725702572600
H	1.410864334890	3.388340975728
H	3.183967430392	1.727227661432
H	2.497250410124	2.412136483040
H	-3.180635201255	-1.732585051901
H	-2.501126104295	-2.410383847991
H	-1.404228452187	-3.388753204805
H	2.603518197736	-2.295176669111
H	3.271125040011	-1.562025403598
H	1.580782091189	-3.314037610664
H	-3.270316986352	1.552063641886
H	-1.582710413084	3.308509295944
H	-2.604407215514	2.296963171504
H	-2.595657798879	2.300642540916
H	-1.573043403100	3.312084417438
H	-3.263715500170	1.558737096173

2		
Dy	0.000000000000	0.000000000000
C	1.157919000000	0.404148000000
C	0.747885000000	-0.967427000000
C	-0.696901000000	-1.001636000000
C	-1.176609000000	0.346457000000
C	-0.032294000000	1.218458000000
C	1.896266000000	0.917978000000
C	0.662944000000	1.043876000000
C	0.183298000000	-0.271222000000
C	1.105514000000	-1.213948000000

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C	2.172199000000	-0.476684000000	1.344925000000
C	-2.617103000000	0.786715000000	-2.271302000000
H	-2.783443000000	1.712436000000	-1.689377000000
H	-2.955269000000	1.004736000000	-3.302661000000
H	-3.295835000000	0.011070000000	-1.874795000000
C	-1.531373000000	-2.254130000000	-2.273175000000
H	-1.081418000000	-3.068107000000	-1.674450000000
H	-2.555805000000	-2.080543000000	-1.899742000000
H	-1.633186000000	-2.649281000000	-3.302750000000
C	1.643244000000	-2.170482000000	-2.389288000000
H	1.665690000000	-2.551156000000	-3.429030000000
H	2.682641000000	-1.934846000000	-2.108404000000
H	1.302013000000	-3.011128000000	-1.757142000000
C	2.561060000000	0.915460000000	-2.461810000000
H	2.796225000000	1.051306000000	-3.535155000000
H	2.711606000000	1.894306000000	-1.975592000000
H	3.310217000000	0.215276000000	-2.055784000000
C	-0.064808000000	2.721842000000	-2.302410000000
H	-0.069919000000	3.088050000000	-3.347899000000
H	-0.969300000000	3.136995000000	-1.822558000000
H	0.819200000000	3.171518000000	-1.816014000000
C	3.416730000000	-1.079621000000	0.756225000000
H	3.222435000000	-2.050901000000	0.269752000000
H	3.888663000000	-0.415355000000	0.013125000000
H	4.168787000000	-1.263200000000	1.547787000000
C	-1.053206000000	-0.616762000000	3.339556000000
H	-0.806267000000	-0.793591000000	4.404134000000
H	-1.802832000000	0.194081000000	3.320985000000
H	-1.542895000000	-1.539944000000	2.977849000000
C	0.032120000000	2.346987000000	2.663593000000
H	0.140358000000	3.131345000000	1.891307000000
H	-1.042116000000	2.240120000000	2.895142000000
H	0.514028000000	2.745659000000	3.577643000000
C	2.774731000000	2.063313000000	1.090185000000
H	3.509378000000	1.760349000000	0.326677000000
H	2.198569000000	2.917110000000	0.687850000000
H	3.346182000000	2.452701000000	1.955172000000
C	1.018577000000	-2.712386000000	2.078048000000
H	1.569017000000	-3.079100000000	2.966700000000
H	-0.021632000000	-3.068016000000	2.186928000000
H	1.464688000000	-3.217611000000	1.202572000000

3

Dy	0.000000000000	0.000000000000	0.000000000000
C	1.435209000000	0.017739000000	-2.301847000000
H	2.535421000000	0.014424000000	-2.319288000000
C	0.712687000000	-1.224133000000	-2.407184000000
H	1.268678000000	-2.168207000000	-2.525468000000
C	-0.715809000000	-1.244391000000	-2.355559000000
C	-1.436900000000	-0.017457000000	-2.335965000000
H	-2.536333000000	-0.015020000000	-2.377457000000
C	0.715809000000	1.244391000000	-2.366047000000
H	1.253691000000	2.202688000000	-2.427420000000
C	-0.712404000000	1.223636000000	-2.437184000000
H	-1.266326000000	2.166869000000	-2.569872000000
C	1.469076000000	0.005703000000	2.309936000000
H	2.569011000000	-0.006132000000	2.332174000000
C	0.758634000000	1.238623000000	2.345215000000
H	1.304174000000	2.193635000000	2.390558000000
C	-0.668857000000	1.230511000000	2.417806000000
C	-1.403374000000	-0.005977000000	2.324928000000
H	-2.503148000000	0.006889000000	2.359674000000
C	0.735562000000	-1.229850000000	2.425739000000
H	1.284141000000	-2.176944000000	2.552744000000
C	-0.693257000000	-1.238623000000	2.379105000000
H	-1.238299000000	-2.192192000000	2.451158000000
H	-1.215302000000	2.179098000000	2.543882000000
H	-1.252564000000	-2.203965000000	-2.408387000000

4

Dy	0.000000000000	0.000000000000	0.000000000000
C	1.167466000000	1.437299000000	1.850307000000
C	-0.211822000000	1.491501000000	2.187822000000
C	-1.133357000000	0.446522000000	2.296666000000

## Supporting Information

C	-0.866773000000	-0.971332000000	2.282371000000
C	-0.797778000000	-1.415621000000	-2.112860000000
C	0.602633000000	-1.515388000000	-2.051939000000
C	-1.612420000000	-0.263394000000	-2.002659000000
C	1.995264000000	0.285421000000	1.762404000000
C	1.563474000000	-0.455175000000	-2.100195000000
C	-1.193547000000	1.106116000000	-2.091085000000
C	1.622036000000	-1.066016000000	1.760000000000
C	1.346085000000	0.918179000000	-2.062210000000
C	0.091553000000	1.625283000000	-2.064384000000
C	0.330820000000	-1.623395000000	2.054938000000
H	0.162449000000	2.697021000000	-1.994600000000
H	2.223465000000	1.539353000000	-1.979562000000
H	2.592753000000	-0.769268000000	-2.019916000000
H	0.999528000000	-2.514132000000	-1.993504000000
H	-1.321355000000	-2.355258000000	-2.050622000000
H	-2.674786000000	-0.434351000000	-1.956774000000
H	-1.984295000000	1.835110000000	-2.022147000000
H	1.648014000000	2.387608000000	1.692008000000
H	3.020300000000	0.481765000000	1.486188000000
H	2.396561000000	-1.780297000000	1.534790000000
H	0.270296000000	-2.696393000000	1.989338000000
H	-1.731115000000	-1.607405000000	2.379291000000
H	-2.161039000000	0.733954000000	2.443029000000
H	-0.634406000000	2.482215000000	2.202953000000

5

Dy	0.000000000000	0.000000000000	0.000000000000
C	-0.779857000000	-1.686901000000	-1.914212000000
H	-1.238816000000	-2.678957000000	-1.798256000000
C	-1.744006000000	-0.641330000000	-1.909224000000
H	-2.769418000000	-1.018862000000	-1.790152000000
C	-1.686555000000	0.780105000000	-1.908189000000
H	-2.678110000000	1.239061000000	-1.788431000000
C	-0.641125000000	1.744480000000	-1.913398000000
H	-1.018244000000	2.770377000000	-1.796925000000
C	0.779826000000	1.686934000000	-1.914889000000
H	1.238960000000	2.678997000000	-1.799736000000
C	1.743879000000	0.641306000000	-1.910521000000
H	2.769387000000	1.018966000000	-1.792607000000
C	1.686555000000	-0.780105000000	-1.909665000000
H	2.678209000000	-1.239020000000	-1.790564000000
C	0.641119000000	-1.744439000000	-1.913880000000
H	1.018289000000	-2.770334000000	-1.797482000000
C	0.739310000000	1.705771000000	1.914009000000
H	1.173493000000	2.708871000000	1.797710000000
C	1.729047000000	0.684333000000	1.908877000000
H	2.744793000000	1.087280000000	1.790183000000
C	1.706992000000	-0.738068000000	1.908681000000
H	2.709614000000	-1.172354000000	1.789076000000
C	0.685798000000	-1.727969000000	1.913646000000
H	1.088194000000	-2.744229000000	1.797258000000
C	-0.736141000000	-1.705735000000	1.914583000000
H	-1.170399000000	-2.708900000000	1.799144000000
C	-1.725933000000	-0.684338000000	1.910615000000
H	-2.741777000000	-1.087205000000	1.792570000000
C	-1.703759000000	0.738068000000	1.910427000000
H	-2.706570000000	1.172219000000	1.791844000000
C	-0.682644000000	1.728047000000	1.914507000000
H	-1.085343000000	2.744184000000	1.798008000000

6

Dy	0.000000000000	0.000000000000	0.000000000000
C	-0.036037000000	2.066538000000	-1.806997000000
H	-0.054019000000	3.160044000000	-1.712554000000
C	-1.357550000000	1.562392000000	-1.810024000000
H	-2.074476000000	2.388197000000	-1.717621000000
C	-2.041239000000	0.325371000000	-1.804740000000
H	-3.121393000000	0.495688000000	-1.708689000000
C	-1.773027000000	-1.066882000000	-1.810272000000
H	-2.711611000000	-1.628293000000	-1.718492000000
C	-0.676345000000	-1.953957000000	-1.808778000000
H	-1.030608000000	-2.988741000000	-1.716283000000
C	0.743287000000	-1.930327000000	-1.809896000000

## Supporting Information

H	1.132486000000	-2.952461000000	-1.717877000000
C	1.807565000000	-1.006022000000	-1.809978000000
H	2.765141000000	-1.534359000000	-1.717414000000
C	2.029119000000	0.396086000000	-1.808287000000
H	3.103173000000	0.602376000000	-1.714831000000
C	1.304227000000	1.606800000000	-1.812462000000
H	1.991729000000	2.457507000000	-1.722131000000
C	0.039603000000	-2.066538000000	1.805657000000
H	0.057103000000	-3.159923000000	1.709737000000
C	1.361257000000	-1.562790000000	1.808276000000
H	2.077852000000	-2.388766000000	1.714844000000
C	2.045247000000	-0.325988000000	1.803414000000
H	3.125349000000	-0.496571000000	1.707252000000
C	1.777380000000	1.066305000000	1.808471000000
H	2.716063000000	1.627414000000	1.715825000000
C	0.681041000000	1.953749000000	1.807108000000
H	1.035512000000	2.988334000000	1.713214000000
C	-0.738599000000	1.930600000000	1.810260000000
H	-1.127657000000	2.952766000000	1.718009000000
C	-1.803159000000	1.006584000000	1.813157000000
H	-2.760791000000	1.535154000000	1.722518000000
C	-2.025110000000	-0.395487000000	1.812124000000
H	-3.099441000000	-0.601464000000	1.721187000000
C	-1.300539000000	-1.606434000000	1.813859000000
H	-1.988453000000	-2.456861000000	1.724047000000

## EDA Input File

```
#!/bin/sh
```

```
# dependency: D:/ADF_DATA/Homoleptic/Dy55/Dy55.Region_1 Dy55.Region_1.results/adf.rkf Region_1.rkf
```

```
# dependency: D:/ADF_DATA/Homoleptic/Dy55/Dy55.Region_2 Dy55.Region_2.results/adf.rkf Region_2.rkf
```

```
"$AMSBIN/ams" << eor
```

```
Task SinglePoint
```

```
System
```

```
Atoms
```

```
Dy 0.0 0.0 0.0 region=Region_1 adf.f=Region_1
C 1.157919 0.404148 -2.293779 region=Region_2 adf.f=Region_2
C 0.747885 -0.967427 -2.285179 region=Region_2 adf.f=Region_2
C -0.696901 -1.001636 -2.261709 region=Region_2 adf.f=Region_2
C -1.176609 0.346457 -2.260421 region=Region_2 adf.f=Region_2
C -0.032294 1.218458 -2.264046 region=Region_2 adf.f=Region_2
C 1.896266 0.917978 1.51 region=Region_1 adf.f=Region_1
C 0.662944 1.043876 2.252933 region=Region_1 adf.f=Region_1
C 0.183298 -0.271222 2.551573 region=Region_1 adf.f=Region_1
C 1.105514 -1.213948 1.97816 region=Region_1 adf.f=Region_1
C 2.172199 -0.476684 1.344925 region=Region_1 adf.f=Region_1
C -2.617103 0.7867150000000001 -2.271302 region=Region_2 adf.f=Region_2
H -2.783443 1.712436 -1.689377 region=Region_2 adf.f=Region_2
H -2.955269 1.004736 -3.302661 region=Region_2 adf.f=Region_2
H -3.295835 0.01107 -1.874795 region=Region_2 adf.f=Region_2
C -1.531373 -2.25413 -2.273175 region=Region_2 adf.f=Region_2
H -1.081418 -3.068107 -1.67445 region=Region_2 adf.f=Region_2
H -2.555805 -2.080543 -1.899742 region=Region_2 adf.f=Region_2
H -1.633186 -2.649281 -3.30275 region=Region_2 adf.f=Region_2
C 1.643244 -2.170482 -2.389288 region=Region_2 adf.f=Region_2
H 1.66569 -2.551156 -3.42903 region=Region_2 adf.f=Region_2
H 2.682641 -1.934846 -2.108404 region=Region_2 adf.f=Region_2
H 1.302013 -3.011128 -1.757142 region=Region_2 adf.f=Region_2
C 2.56106 0.9154600000000001 -2.46181 region=Region_2 adf.f=Region_2
H 2.796225 1.051306 -3.535155 region=Region_2 adf.f=Region_2
H 2.711606 1.894306 -1.975592 region=Region_2 adf.f=Region_2
H 3.310217 0.215276 -2.055784 region=Region_2 adf.f=Region_2
C -0.064808 2.721842 -2.30241 region=Region_2 adf.f=Region_2
H -0.069919 3.08805 -3.347899 region=Region_2 adf.f=Region_2
```



## Supporting Information

H -0.969300000000001 3.136995 -1.822558 region=Region\_2 adf.f=Region\_2  
H 0.8192 3.171518 -1.816014 region=Region\_2 adf.f=Region\_2  
C 3.41673 -1.079621 0.756225 region=Region\_1 adf.f=Region\_1  
H 3.222435 -2.050901 0.269752 region=Region\_1 adf.f=Region\_1  
H 3.888663 -0.415355 0.013125 region=Region\_1 adf.f=Region\_1  
H 4.168787 -1.2632 1.547787 region=Region\_1 adf.f=Region\_1  
C -1.053206 -0.616762 3.339556 region=Region\_1 adf.f=Region\_1  
H -0.806267 -0.793591 4.404134 region=Region\_1 adf.f=Region\_1  
H -1.802832 0.194081 3.320985 region=Region\_1 adf.f=Region\_1  
H -1.542895 -1.539944 2.977849 region=Region\_1 adf.f=Region\_1  
C 0.03212 2.346987 2.663593 region=Region\_1 adf.f=Region\_1  
H 0.140358 3.131345 1.891307 region=Region\_1 adf.f=Region\_1  
H -1.042116 2.24012 2.895142 region=Region\_1 adf.f=Region\_1  
H 0.514028 2.745659 3.577643 region=Region\_1 adf.f=Region\_1  
C 2.774731 2.063313 1.090185 region=Region\_1 adf.f=Region\_1  
H 3.509378 1.760349 0.326677 region=Region\_1 adf.f=Region\_1  
H 2.198569 2.91711 0.68785 region=Region\_1 adf.f=Region\_1  
H 3.346182 2.452701 1.955172 region=Region\_1 adf.f=Region\_1  
C 1.018577 -2.712386 2.078048 region=Region\_1 adf.f=Region\_1  
H 1.569017 -3.0791 2.9667 region=Region\_1 adf.f=Region\_1  
H -0.021632 -3.068016 2.186928 region=Region\_1 adf.f=Region\_1  
H 1.464688 -3.217611 1.202572 region=Region\_1 adf.f=Region\_1

End

Charge 1.0

BondOrders

2 3 1.5  
2 6 1.5  
2 24 1.0  
3 4 1.5  
3 20 1.0  
4 5 1.5  
4 16 1.0  
5 6 1.5  
5 12 1.0  
6 28 1.0  
7 8 1.5  
7 11 1.5  
7 44 1.0  
8 9 1.5  
8 40 1.0  
9 10 1.5  
9 36 1.0  
10 11 1.5  
10 48 1.0  
11 32 1.0  
12 13 1.0  
12 14 1.0  
12 15 1.0  
16 17 1.0  
16 18 1.0  
16 19 1.0  
20 21 1.0  
20 22 1.0  
20 23 1.0  
24 25 1.0  
24 26 1.0  
24 27 1.0  
28 29 1.0  
28 30 1.0  
28 31 1.0  
32 33 1.0  
32 34 1.0

## Supporting Information

```
32 35 1.0
36 37 1.0
36 38 1.0
36 39 1.0
40 41 1.0
40 42 1.0
40 43 1.0
44 45 1.0
44 46 1.0
44 47 1.0
48 49 1.0
48 50 1.0
48 51 1.0
End
End

Engine ADF
Basis
  Type TZ2P
  Core None
End
SpinPolarization 5.0
Fragments
  Region_1 Region_1.rkf
  Region_2 Region_2.rkf
End

Print ETSLOWDIN-Unrestricted
Print NOCVHIRSHFELD
XC
  Hybrid PBE0
  DISPERSION GRIMME3 BJDAMP
End
Symmetry NOSYM
Unrestricted Yes
BeckeGrid
  Quality Good
End
NumericalQuality Good
SCF
  Iterations 2000
  DIIS
  N 15
  End
  AccelerationMethod MESA
End
UnrestrictedFragments Yes
ETSNOCV
  Enabled Yes
End
EndEngine
eor
```

## CASSCF Input File

```
!DKH2 DKH-def2-svp slowconv tightscf autoaux
!moread
%moinp "/home/mshruti.iith/ibtesham/homoleptic/Dy55/Dy55-n.gbw"

%pal nprocs 12
end
```

## Supporting Information

%Maxcore 6000

%basis newgto Dy

0 6  
1 1205955.486947000027 0.008165729207  
2 535980.216420999961 -0.004019299653  
3 238213.429520000005 0.016042861724  
4 105872.635341999994 0.006534642408  
5 47054.504596999999 0.032501357805  
6 20913.113153999999 0.044343203836  
0 1  
7 9294.716957000001 1.000000000000  
0 1  
8 4130.985314000000 1.000000000000  
0 1  
9 1835.993473000000 1.000000000000  
0 1  
10 815.997099000000 1.000000000000  
0 1  
11 362.665377000000 1.000000000000  
0 1  
12 161.184612000000 1.000000000000  
0 1  
13 71.637605000000 1.000000000000  
0 1  
14 31.838936000000 1.000000000000  
0 1  
15 14.150638000000 1.000000000000  
0 1  
16 6.289172000000 1.000000000000  
0 1  
17 2.795188000000 1.000000000000  
0 1  
18 1.242306000000 1.000000000000  
0 1  
19 0.552136000000 1.000000000000  
0 1  
20 0.245394000000 1.000000000000  
0 1  
21 0.109064000000 1.000000000000  
0 1  
22 0.048473000000 1.000000000000  
0 1  
23 0.021543000000 1.000000000000  
1 5  
1 16500.520027999999 0.004760264705  
2 6600.208011000000 0.005897483480  
3 2640.083204000000 0.029326527847  
4 1056.033282000000 0.089161858001  
5 422.413313000000 0.265122289728  
1 1  
6 168.965325000000 1.000000000000  
1 1  
7 67.586130000000 1.000000000000  
1 1  
8 27.034452000000 1.000000000000  
1 1  
9 10.813781000000 1.000000000000  
1 1  
10 4.325512000000 1.000000000000  
1 1  
11 1.730205000000 1.000000000000

## Supporting Information

```
1 1
12 0.692082000000 1.000000000000
1 1
13 0.276833000000 1.000000000000
1 1
14 0.110733000000 1.000000000000
1 1
15 0.044293000000 1.000000000000
1 1
16 0.017717000000 1.000000000000
2 4
1 1185.048662000000 0.004734300415
2 430.926786000000 0.026490774838
3 156.700650000000 0.145474353692
4 56.982054000000 0.463452784138
2 1
5 20.720747000000 1.000000000000
2 1
6 7.534817000000 1.000000000000
2 1
7 2.739933000000 1.000000000000
2 1
8 0.996339000000 1.000000000000
2 1
9 0.362305000000 1.000000000000
2 1
10 0.131747000000 1.000000000000
2 1
11 0.047908000000 1.000000000000
2 1
12 0.017421000000 1.000000000000
3 4
1 33.836944000000 0.101241740066
2 11.278981000000 0.330021330584
3 3.759660000000 0.494377985697
4 1.253220000000 0.347706969111
3 1
5 0.417740000000 0.145206846483
3 1
6 0.139247000000 1.000000000000
end
end
```

```
%method
SpecialGridAtoms 66
SpecialGridIntAcc 9
end
```

```
%casscf
nel 9
norb 7
mult 6,4,2
nroots 21,224,120
actorbs forbs
nevpt2 true
maxiter 100
ci
nguessmat 8000
maxiter 100
end
```

```
rel
```

## Supporting Information

dosoc true  
gtensor true  
printlevel 3  
NDoubGTensor 8  
domagnetization true  
dosusceptibility true

SUSTempMIN 2.0  
SUSTempMAX 300.0  
SUSNPoints 100

MAGTemperatureMIN 2.0  
MAGTemperatureMAX 5.0  
MAGTemperatureNPoints 4

MAGFieldMIN 0.0  
MAGFieldMAX 70000.0  
MAGNpoints 15

end

ANISO  
doaniso true  
MLTP 2,2,2,2,2,2,2,2  
TINT 0, 300, 100  
HINT 0, 7.0, 10  
TMAG 2.0, 3.0, 5.0  
CRYS\_element "Dy"  
CRYS\_charge 3  
PLOT true  
UBAR true  
end  
end

\*xyz 1 6

Dy	0.000000000000	0.000000000000	0.000000000000
C	1.157918712184	0.404148230124	-2.293778758050
C	0.747884502475	-0.967427218409	-2.285178865700
C	-0.696900842632	-1.001636036929	-2.261708727721
C	-1.176608525320	0.346457484487	-2.260420583506
C	-0.032293846672	1.218457540662	-2.264045730813
C	1.896265882687	0.917978302092	1.510000355064
C	0.662943989271	1.043876413575	2.252932513009
C	0.183297780820	-0.271222473181	2.551572897517
C	1.105514012837	-1.213948170343	1.978160146375
C	2.172198785582	-0.476684072208	1.344924504544
C	-2.617102872079	0.786715070251	-2.271301782725
H	-2.783443023321	1.712436405819	-1.689377024691
H	-2.955269481754	1.004735576285	-3.302661122935
H	-3.295834672753	0.011070137836	-1.874794835371
C	-1.531372699577	-2.254130280088	-2.273174943648
H	-1.081417727741	-3.068107099222	-1.674450454046
H	-2.555805013620	-2.080543440205	-1.899741630380
H	-1.633186375053	-2.649281432905	-3.302749691041
C	1.643244092855	-2.170481780600	-2.389287899682
H	1.665689964282	-2.551155598827	-3.429030034480
H	2.682641049214	-1.934845684983	-2.108403672315
H	1.302013105727	-3.011128081554	-1.757142326961
C	2.561059797030	0.915460395816	-2.461810204421
H	2.796224768917	1.051306422457	-3.535154978248
H	2.711606183204	1.894306089143	-1.975591647029
H	3.310216952019	0.215276139540	-2.055783720193

## Supporting Information

C	-0.064808108766	2.721841806701	-2.302409749379
H	-0.069918640385	3.088049783819	-3.347899016859
H	-0.969299965123	3.136994632776	-1.822557951200
H	0.819200232511	3.171518137984	-1.816014247935
C	3.416729696373	-1.079620621612	0.756224660955
H	3.222434852091	-2.050900704856	0.269752030260
H	3.888662502108	-0.415355075462	0.013124767693
H	4.168786594178	-1.263200203999	1.547787424005
C	-1.053205703625	-0.616761844125	3.339556171619
H	-0.806266546253	-0.793591094400	4.404134480890
H	-1.802832230215	0.194081478199	3.320984914742
H	-1.542894690355	-1.539944214904	2.977848513048
C	0.032119949540	2.346986650164	2.663592507768
H	0.140358337304	3.131345453470	1.891307139167
H	-1.042116468064	2.240119802668	2.895142136441
H	0.514028002145	2.745658820205	3.577642525900
C	2.774731350726	2.063313039789	1.090184648141
H	3.509377520640	1.760348719377	0.326677025841
H	2.198568995049	2.917110156156	0.687850414681
H	3.346181589227	2.452700793817	1.955171920150
C	1.018576928426	-2.712386204176	2.078048467505
H	1.569016618081	-3.079100126918	2.966699600844
H	-0.021632118398	-3.068015964206	2.186927773380
H	1.464688187219	-3.217611153764	1.202572027910

\*

## References

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