Electronic Supplementary Material (ESI) for Dalton Transactions.

Supporting Information

Comparison between $DySc_2N@C_{80}$ and $Dy_2ScN@C_{80}$ single-molecule magnetic metallofullerenes encapsulated in single-wall carbon nanotubes

Satoshi Ito,[a] Ryo Nakanishi,[a] Keiichi Katoh,[a] Brian K. Breedlove,[a] Tetsu Sato,[a] Zhao-Yang Li,[c] Yoji Horii,*[b] Masanori Wakizaka,*[a] and Masahiro Yamashita*[a,c]

[a] Department of Chemistry, Graduate School of Science, Tohoku University, 6-3 Aramaki-Aza-Aoba, Aoba-Ku, Sendai 980-8578, Japan

[b] Department of Chemistry, Faculty of Science, Nara Women's University, Nara 6308506, Japan

[c] School of Materials Science and Engineering, Nankai University, Tianjin 300350, China

E-mail:
horiiy20@cc.nara-wu.ac.jp
masanori.wakizaka.a7@tohoku.ac.jp
yamasita@agnus.chem.tohoku.ac.jp

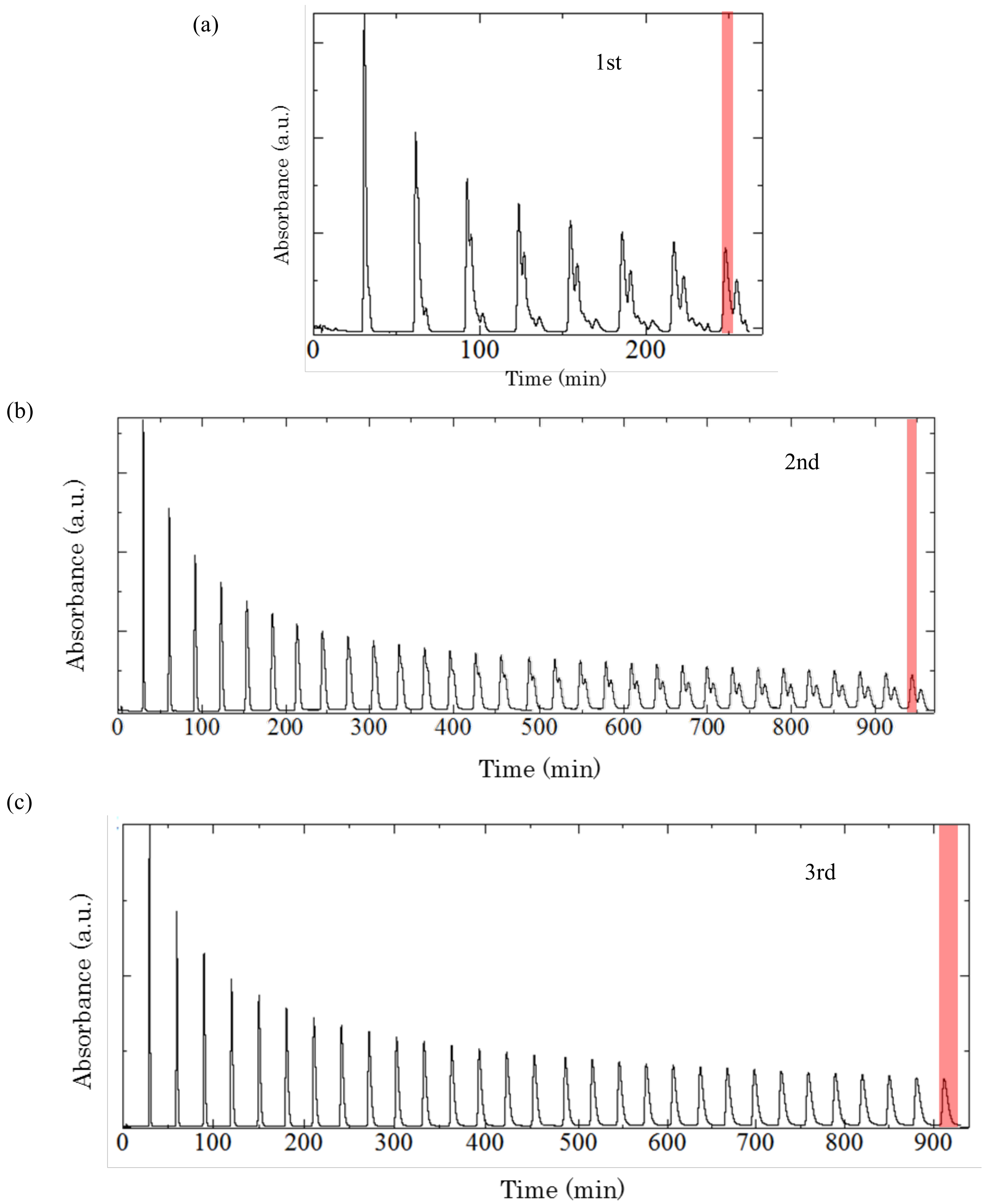

Figure S1. Dy_2 was purified with the three steps (a−c) using recycling HPLC. (a) The extract from the soot was purified and the peak indicated as red was collected. (b) This fraction was further purified, and the first peak indicated as red was collected. (c) Finally, the pure **Dy_2** was obtained by collecting the peak indicated as red.

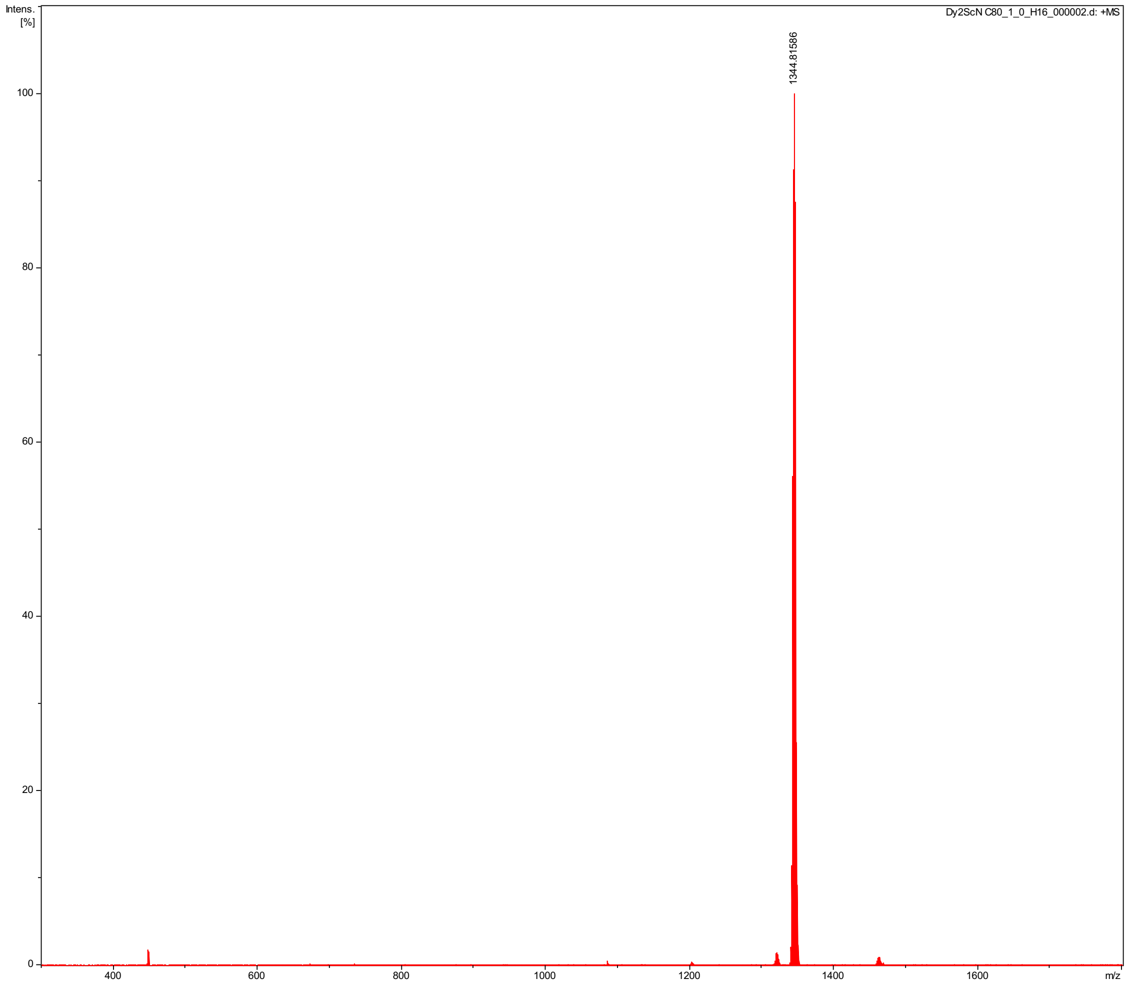

Figure S2. Mass spectrum of **Dy_2** acquired using positive mode.

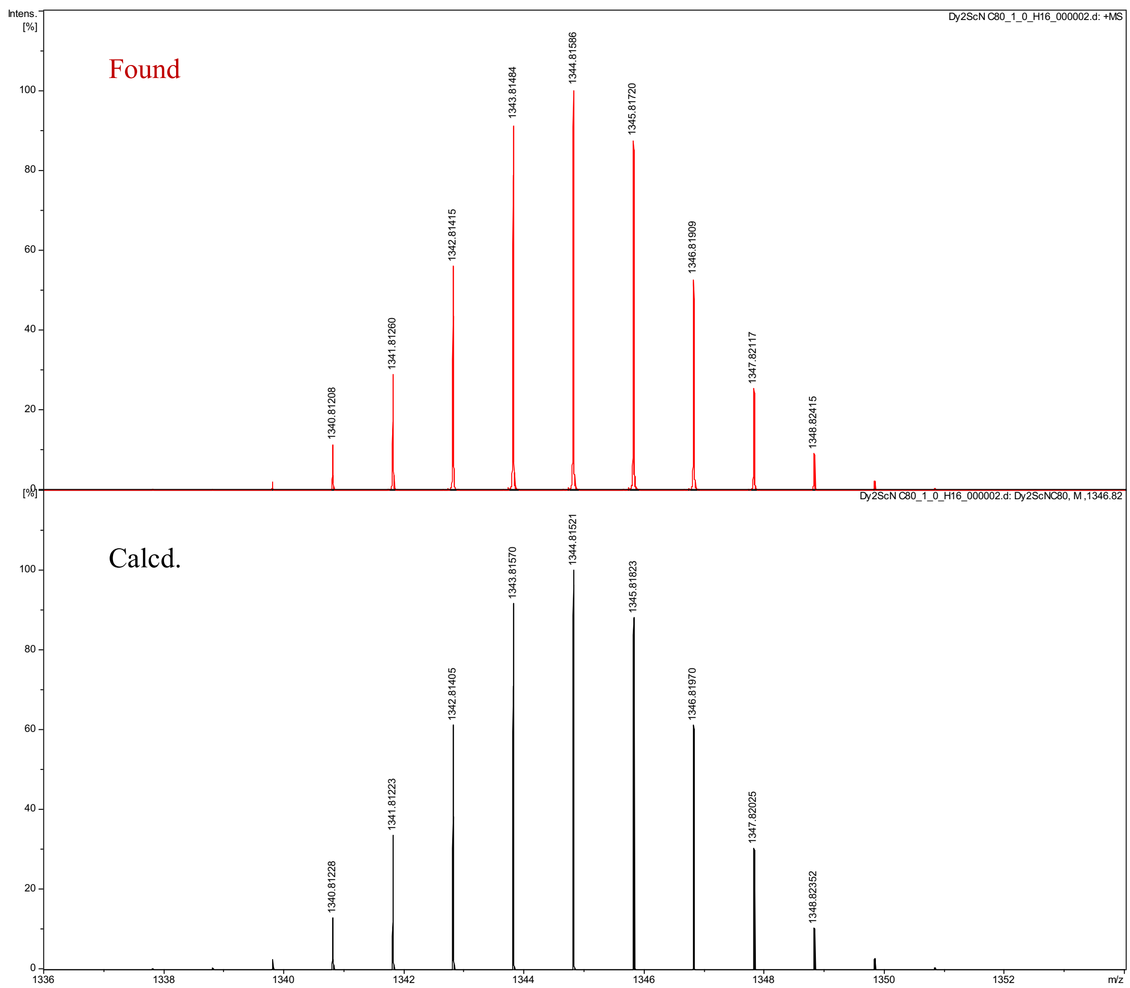

Figure S3. Comparison between experimental and calculated isotope patterns of $\mathbf{Dy_2}$.

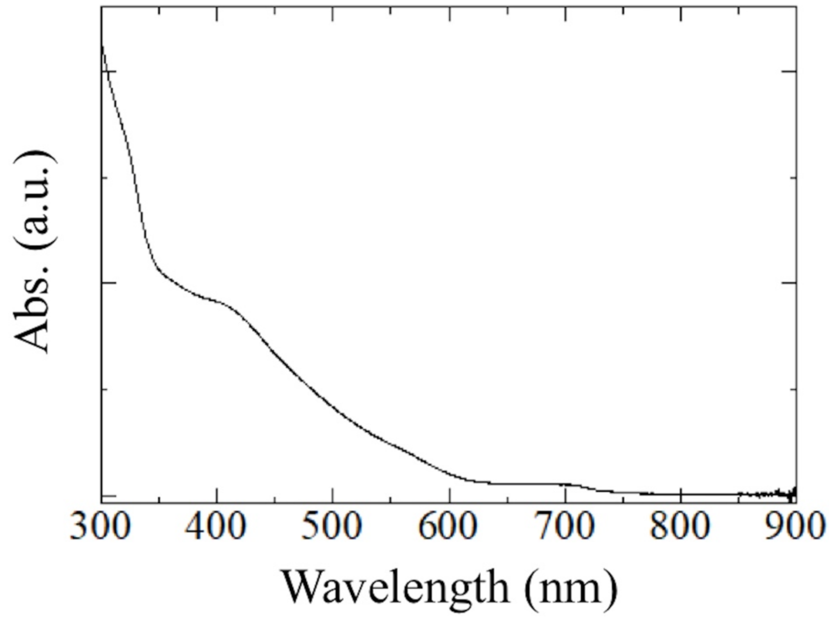

Figure S4. UV-Vis-NIR spectrum of **Dy$_2$** in toluene.

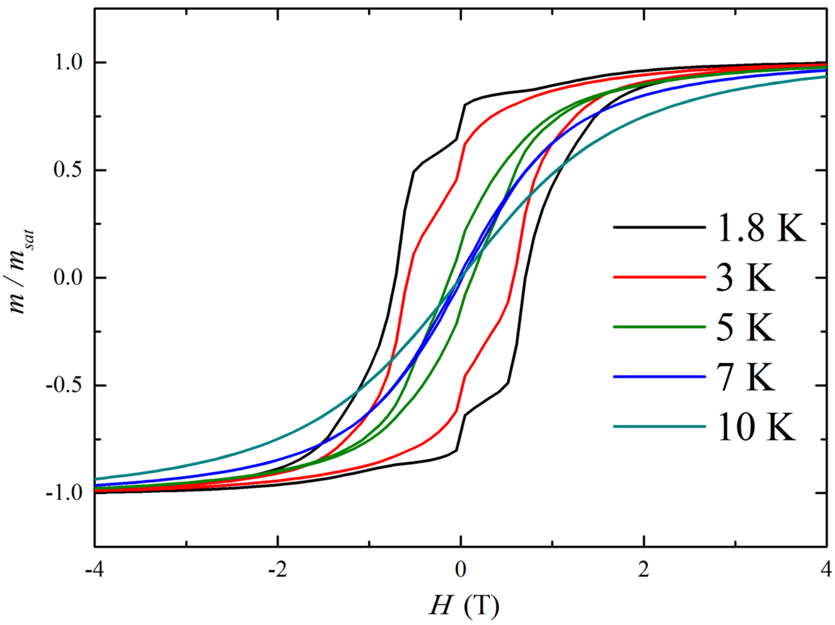

Figure S5. *M* vs. *H* curve for **Dy$_2$**.

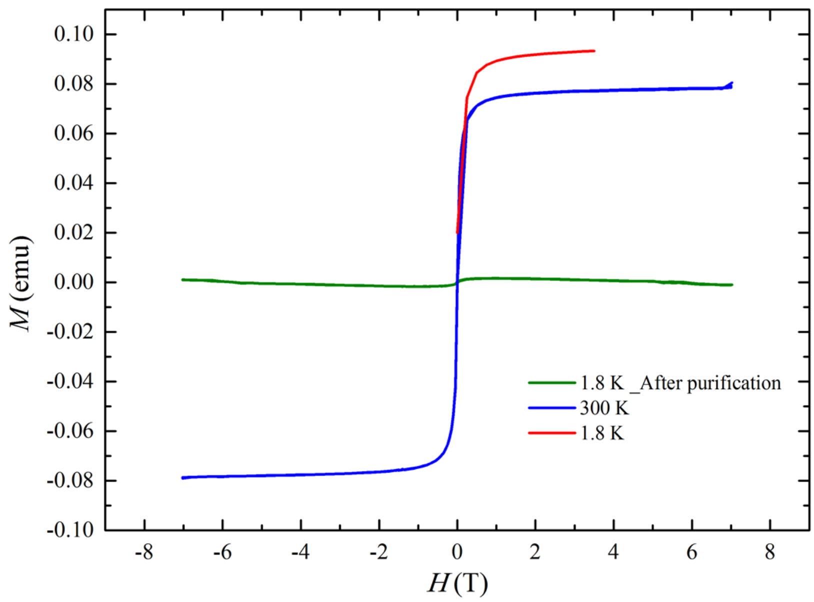

Figure S6. *M* vs. *H* curves of SWCNTs before (red and blue lines) and after (green line) the purification. *M* values become small after purification, indicating the removal of large amount of metal catalysts.

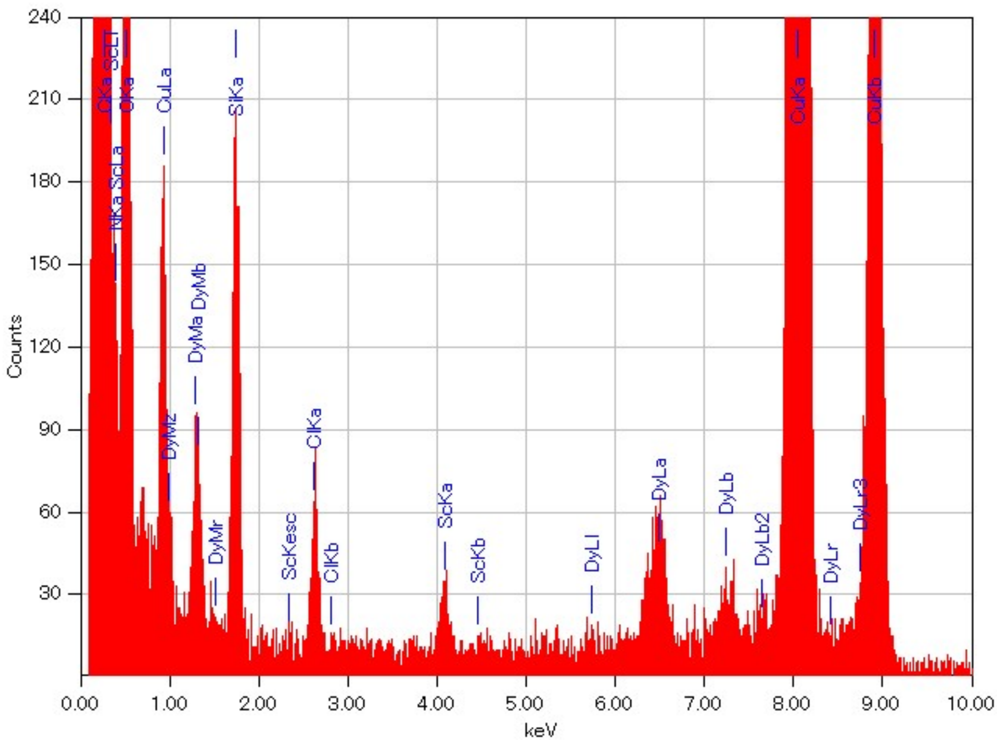

Figure S7. EDX spectra of **Dy_2**@SWCNT.

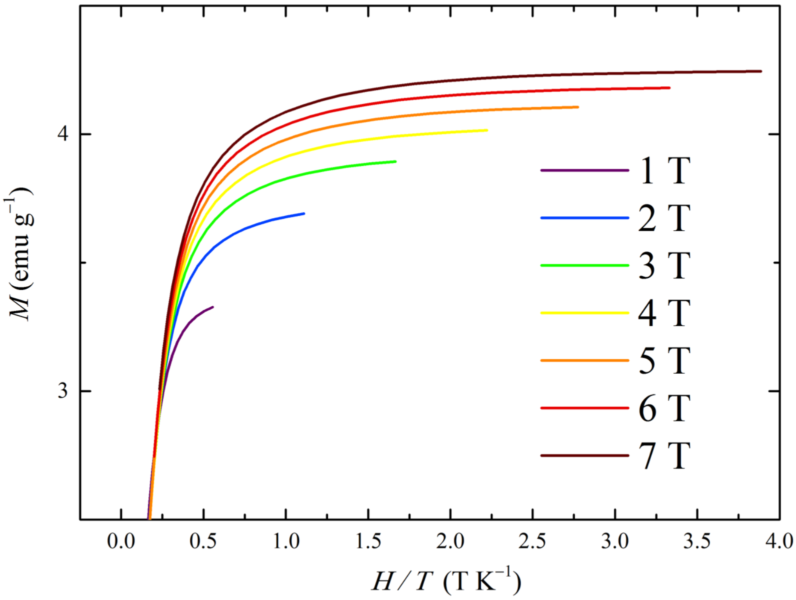

Figure S8. *M* vs. HT^{-1} plots for **Dy_2**@SWCNT.

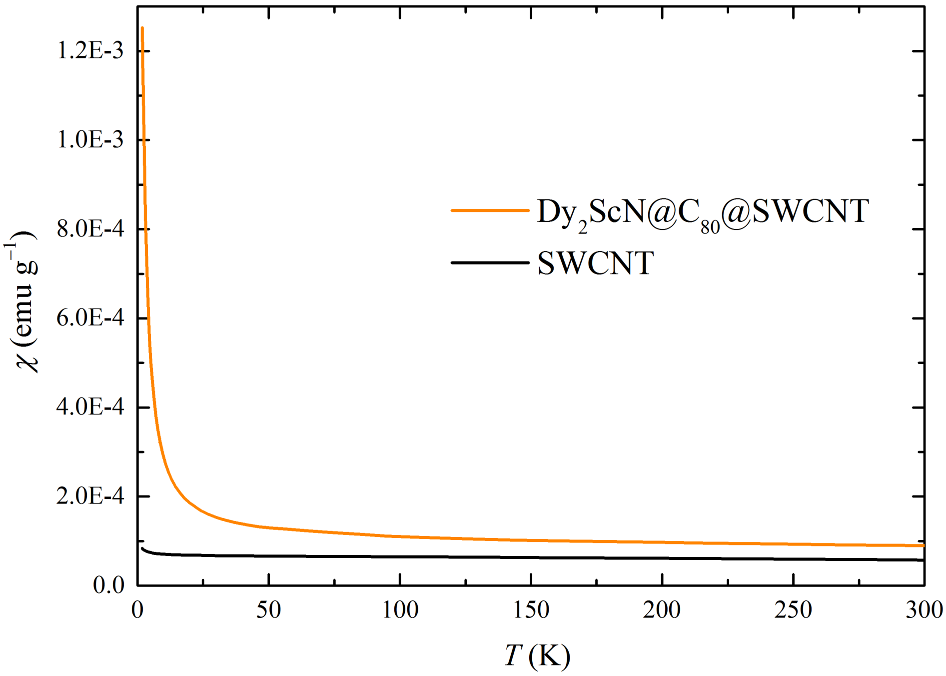

Figure S9. *χ* vs. *T* for empty SWCNTs (black line) and **Dy_2**@SWCNT (orange line).

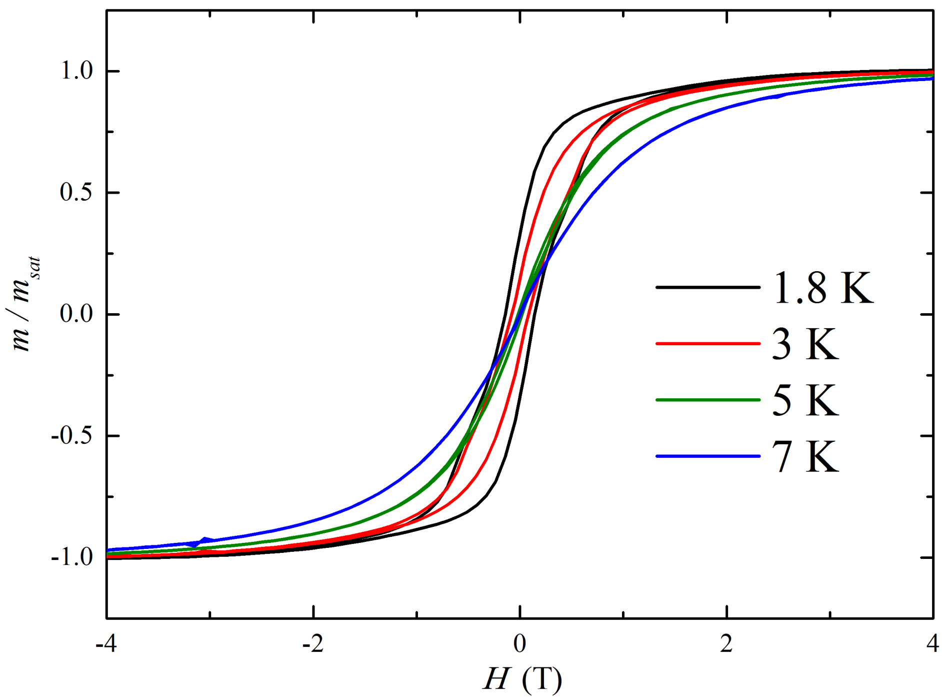

Figure S10. *M* vs. *H* curve for **Dy_2**@SWCNT.

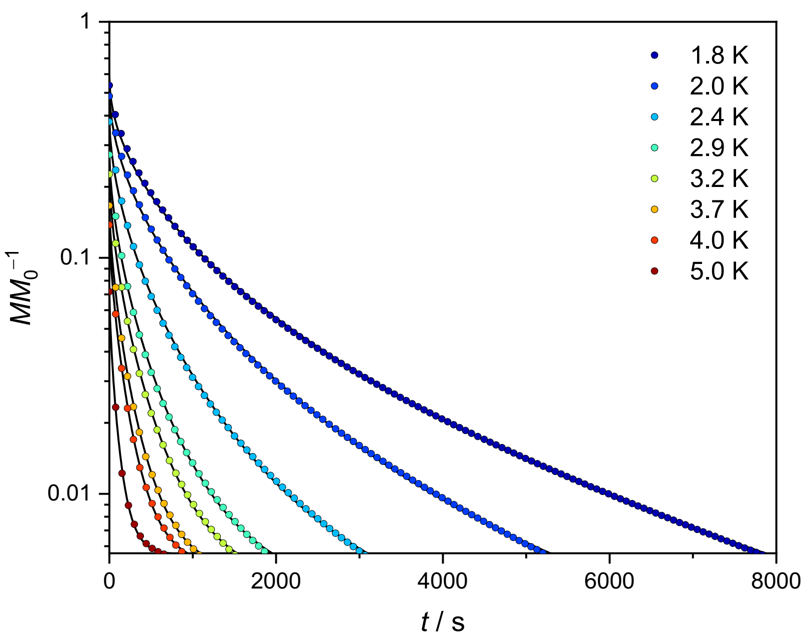

Figure S11. Temperature dependences of the magnetization decay for **Dy_2**@SWCNT.

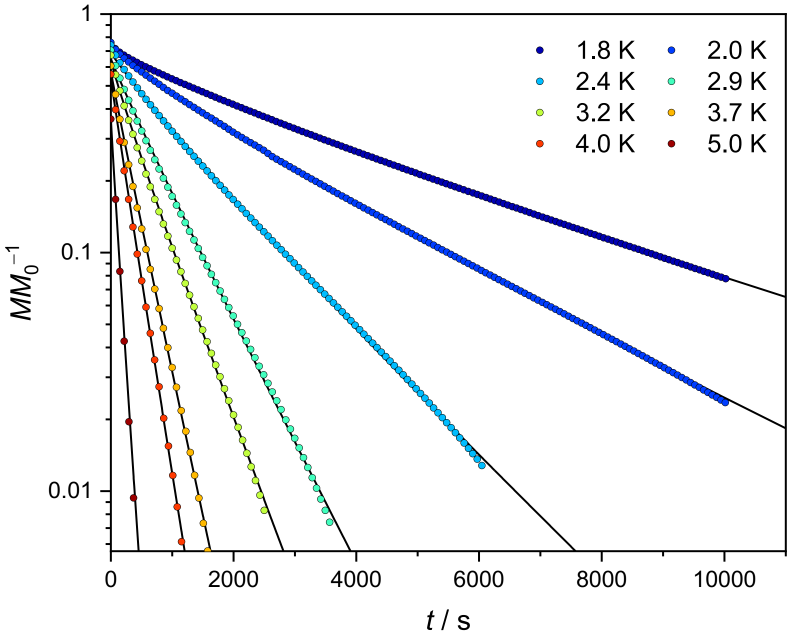

Figure S12. Temperature dependences of the magnetization decay for **Dy_2**.

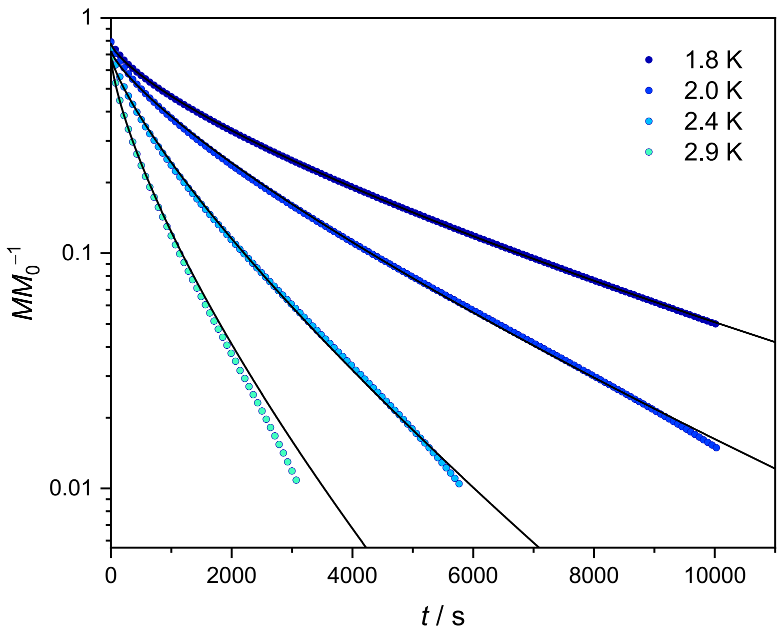

Figure S13. Temperature dependences of the magnetization decay for **Dy_2**/C_{60}.

Table S1. Optimized parameters obtained by the magnetization decay of **Dy_2**@SWCNT using eq. 1.

T / K	τ / s	dev(τ / s)	b	dev(b)	A	dev(A)	y_0	dev(y_0)
1.8	372.95163	8.98659	0.51352	0.00428	0.58585	0.00791	7.20191E-4	1.11835E-4
2.0	271.63047	8.26446	0.53259	0.00597	0.51836	0.00905	0.00159	1.45012E-4
2.4	181.70005	6.89077	0.56199	0.00874	0.39356	0.00877	0.00274	1.74212E-4
2.9	130.80726	5.41162	0.59715	0.01183	0.27785	0.00685	0.00378	1.96873E-4
3.2	111.78967	4.72607	0.61382	0.01331	0.22713	0.00573	0.00404	1.88369E-4
3.7	89.34667	3.61247	0.64572	0.01534	0.16397	0.00395	0.00454	1.70537E-4
4.0	79.37277	2.9907	0.67189	0.01668	0.13418	0.003	0.0049	1.6417E-4
5.0	49.8655	2.45651	0.71528	0.02682	0.06661	0.00164	0.00552	9.92467E-5

Table S2. Optimized parameters obtained by the magnetization decay of **Dy_2** using eq. 1.

T / K	τ / s	dev(τ / s)	b	dev(b)	A	dev(A)	y_0	dev(y_0)
1.8	3947.53535	9.64873	0.86087	0.00366	0.72976	0.00231	0	9.95363E-4
2.0	2520.19471	17.69361	0.88423	0.00641	0.72615	0.00476	0	5.33679E-4
2.4	1381.35499	21.29137	0.92352	0.01252	0.69045	0.00961	0	5.91254E-4
2.9	759.6171	21.87075	0.95218	0.02187	0.65026	0.01704	0	6.26821E-4
3.2	533.02812	14.84504	0.93503	0.02111	0.6397	0.01591	0	6.80563E-4
3.7	332.81459	14.14842	0.96688	0.03262	0.5757	0.02234	0	7.33598E-4
4.0	253.1515	10.00756	0.9742	0.03228	0.53896	0.01939	0	7.76271E-4
5.0	99.74188	45.11588	0.94655	0.39908	0.36611	0.13918	0	0.00824

Table S3. Optimized parameters obtained by the magnetization decay of **Dy_2**/C_{60} using eq. 1.

T / K	τ / s	dev(τ / s)	b	dev(b)	A	dev(A)	y_0	dev(y_0)
1.8	2489.1224	10.01265	0.72233	0.00333	0.78078	0.00299	0	6.31149E-4
2.0	1772.76264	32.33832	0.7718	0.01005	0.72557	0.01035	0	5.87373E-4
2.4	955.72603	26.36164	0.78359	0.01444	0.68595	0.01479	0	6.17424E-4
2.9	470.54035	80.01058	0.71515	0.07886	0.68508	0.08416	0	0.00458

Table S4. Optimized parameters obtained by Arrhenius fit using eq. 2.

	U_{eff} / cm^{-1}	dev(U_{eff} / cm^{-1})	τ_0 / s	dev(τ_0 / s)
Dy_2@SWCNT	3.62606	0.12203	20.60354	1.55231
Dy_2	6.94844	0.56838	20.02469	8.59645
Dy_2@C_{60}	4.70988	0.2108	57.74542	9.73499

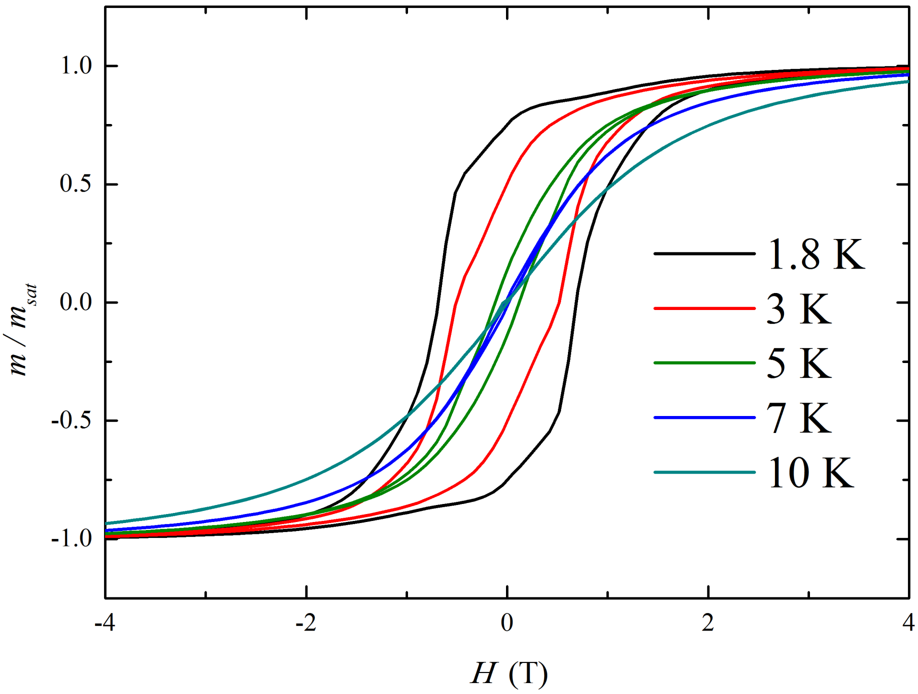

Figure S14. *M* vs. *H* curve for **Dy_2**/C_{60}.

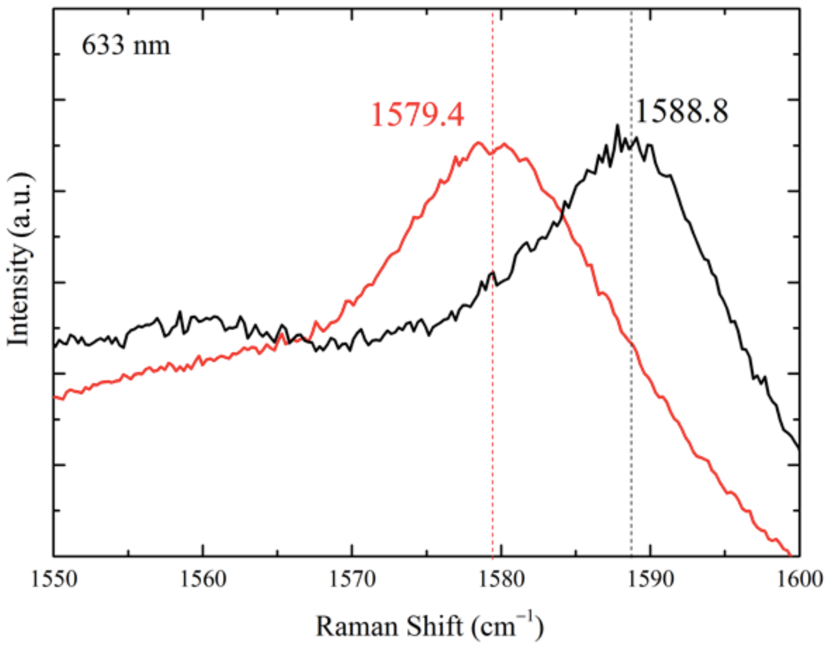

Figure S15. Comparison of Raman spectra between empty SWCNTs (black line) and **Dy_2**@SWCNT (red line) with 633 nm laser excitation.

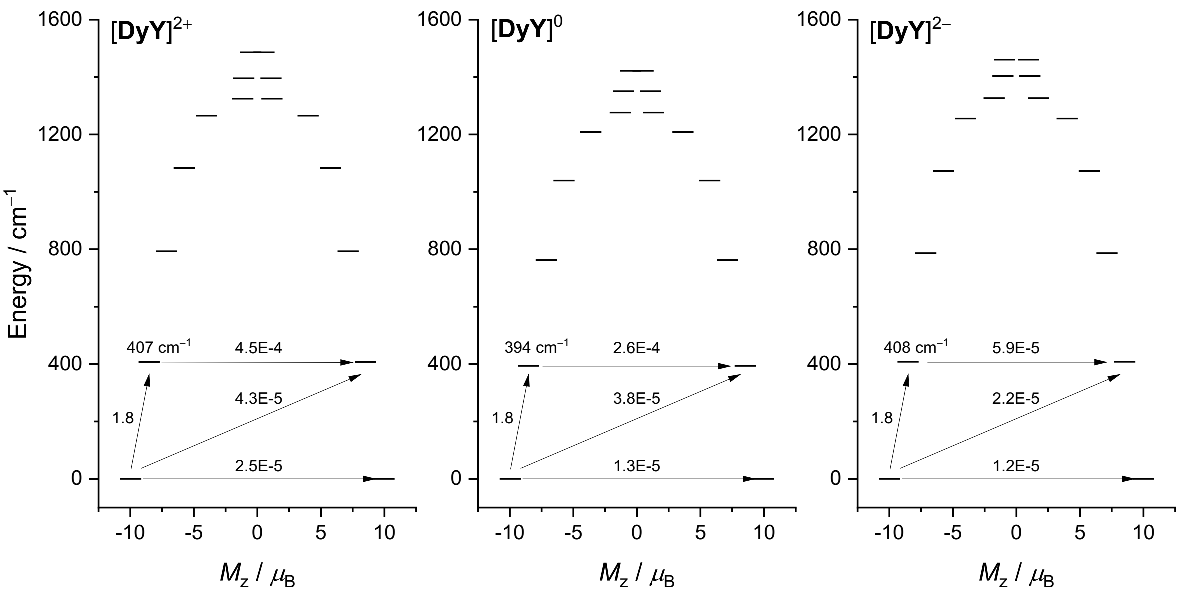

Figure S16. Ligand field splitting of Dy^{A} in [**DyY**]$^{2+}$ (left), [**DyY**]$^{2+}$ (middle) and [**DyY**]$^{2-}$ (right). Energy levels of the first excited Kramers doublets are shown for comparison. The arrows indicate the relaxation pathway with the transition magnetic moment (The unit is μ_B).

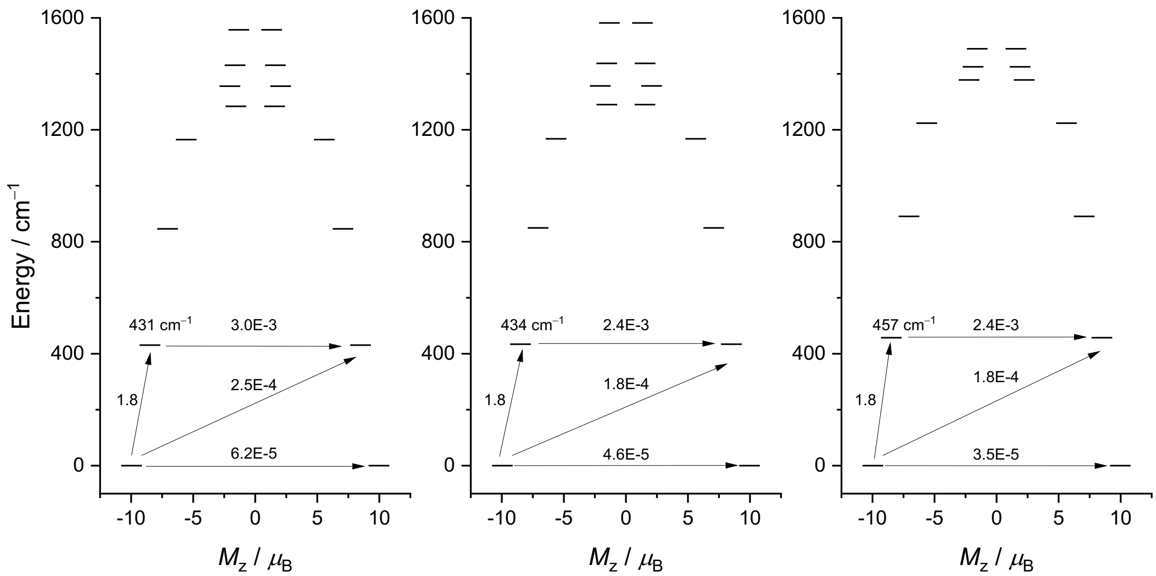

Figure S17. Ligand field splitting of Dy^{B} in [**DyY**]$^{2+}$ (left), [**DyY**]$^{2+}$ (middle) and [**DyY**]$^{2-}$ (right). Energy levels of the first excited Kramers doublets are shown for comparison. The arrows indicate the relaxation pathway with the transition magnetic moment (The unit is μ_B).

Table S5. Energy levels of the CASSCF-SO wavefunctions.

	Dy^A			Dy^B		
	[DyY]$^{2+}$	**[DyY]**^{0}	**[DyY]**$^{2-}$	**[DyY]**$^{2+}$	**[DyY]**^{0}	**[DyY]**$^{2-}$
w.f.1	0	0	0	0	0	0
w.f.2	0	0	0	0	0	0
w.f.3	407.357	393.557	408.127	430.718	433.875	457.125
w.f.4	407.357	393.557	408.127	430.718	433.875	457.125
w.f.5	793.217	762.125	786.257	846.195	849.686	890.782
w.f.6	793.217	762.125	786.257	846.195	849.686	890.782
w.f.7	1083.193	1039.615	1072.772	1165.205	1168.096	1223.879
w.f.8	1083.193	1039.615	1072.772	1165.205	1168.096	1223.879
w.f.9	1265.596	1208.601	1255.573	1284.053	1290.122	1378.109
w.f.10	1265.596	1208.601	1255.573	1284.053	1290.122	1378.109
w.f.11	1324.572	1276.229	1326.623	1355.896	1356.801	1425.045
w.f.12	1324.572	1276.229	1326.623	1355.896	1356.801	1425.045
w.f.13	1395.889	1350.961	1403.669	1431.061	1437.625	1489.736
w.f.14	1395.889	1350.961	1403.669	1431.061	1437.625	1489.736
w.f.15	1486.520	1421.957	1460.543	1557.507	1582.146	1638.485
w.f.16	1486.520	1421.957	1460.543	1557.507	1582.146	1638.485

Table S6. Main values of the *g*-tensor of the Kramers doublets (KDs) for Dy^A in the series of **[DyY]**$^{2+/0/2-}$.

	[DyY]$^{2+}$			**[DyY]**^{0}			**[DyY]**$^{2-}$		
	g_x	g_y	g_z	g_x	g_y	g_z	g_x	g_y	g_z
KD1	6.845E-05	8.293E-05	19.926469	3.611E-05	4.419E-05	19.932659	3.329E-05	3.854E-05	19.946214
KD2	0.0013181	0.0013888	17.095941	0.0007455	0.0008035	17.11013	0.0001691	0.0001834	17.09033
KD3	0.033366	0.0363775	14.308495	0.0251473	0.0297682	14.308567	0.0332485	0.0379379	14.267965
KD4	0.2485752	0.3160432	11.57604	0.2368723	0.2909002	11.469098	0.1804834	0.2521148	11.472527
KD5	1.7543592	4.0386737	8.2093394	2.9021097	4.8365502	8.5477143	2.5024492	3.561124	8.9913681
KD6	1.2691556	4.8978767	12.975945	0.9580333	4.2729712	11.499317	1.7783002	4.8665008	12.694472
KD7	1.8764655	2.517866	15.956154	2.0047122	2.3813417	15.410417	0.6081767	2.2006805	13.538614
KD8	0.1018203	0.4885829	18.899598	0.2633605	1.0110969	18.61543	0.6530152	3.1222028	17.012736

Table S7. Main values of the *g*-tensor of the Kramers doublets (KDs) for Dy^B in the series of **[DyY]**$^{2+/0/2-}$.

	[DyY]$^{2+}$			**[DyY]**^{0}			**[DyY]**$^{2-}$		
	g_x	g_y	g_z	g_x	g_y	g_z	g_x	g_y	g_z
KD1	0.0001763	0.000197	19.925174	0.0001289	0.0001483	19.921822	9.165E-05	0.0001187	19.927141
KD2	0.0086971	0.0090359	17.017433	0.0071543	0.007312	17.002509	0.0069889	0.0071963	16.99143
KD3	0.1010042	0.1087714	14.167513	0.0812747	0.0893273	14.179722	0.0982988	0.1087306	14.149688
KD4	0.6257483	0.9261182	11.222221	0.3761152	0.55314	11.326055	0.598152	0.7703649	11.282266
KD5	2.0100559	3.5819115	15.20295	2.4464126	3.7205067	15.069545	2.5201553	4.7657592	11.795063
KD6	1.8919385	4.5704295	10.680621	1.5514779	4.0948667	10.731896	0.5962332	2.2197962	10.561054
KD7	1.1180533	2.1428579	16.041707	0.9812141	1.9837221	15.930153	1.1144005	2.6166358	15.440749
KD8	0.1806188	0.6162918	19.015572	0.1495314	0.4979378	19.089629	0.0650801	0.3867962	18.987874

Table S8. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^A in [**DyY**]$^{2+}$.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	86.5	12.6	0.0	0.0	0.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	92.3	6.2	0.9	0.0	0.4	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0
-5.5	0.7	0.1	0.7	0.0	95.8	0.0	2.2	0.0	0.1	0.0	0.0	0.1	0.1	0.0	0.0	0.0
-4.5	0.0	0.0	0.5	0.0	2.2	0.0	92.3	0.1	1.9	0.3	0.6	0.9	0.3	0.5	0.1	0.2
-3.5	0.0	0.0	0.0	0.0	0.0	0.0	3.1	0.0	71.3	10.3	5.2	4.2	4.6	0.1	0.2	0.9
-2.5	0.0	0.0	0.1	0.0	0.0	0.0	1.4	0.0	0.5	0.7	23.2	19.1	13.3	30.9	6.2	4.5
-1.5	0.0	0.0	0.0	0.0	0.1	0.0	0.3	0.1	7.1	1.5	11.8	7.5	32.4	0.9	34.3	3.8
-0.5	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.1	4.3	1.8	16.2	11.1	1.5	15.2	15.4	34.3
0.5	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	1.8	4.3	11.1	16.2	15.2	1.5	34.3	15.4
1.5	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.3	1.5	7.1	7.5	11.8	0.9	32.4	3.8	34.3
2.5	0.0	0.0	0.0	0.1	0.0	0.0	0.0	1.4	0.7	0.5	19.1	23.2	30.9	13.3	4.5	6.2
3.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	3.1	10.3	71.3	4.2	5.2	0.1	4.6	0.9	0.2
4.5	0.0	0.0	0.0	0.5	0.0	2.2	0.1	92.3	0.3	1.9	0.9	0.6	0.5	0.3	0.2	0.1
5.5	0.1	0.7	0.0	0.7	0.0	95.8	0.0	2.2	0.0	0.1	0.1	0.0	0.0	0.1	0.0	0.0
6.5	0.0	0.0	6.2	92.3	0.0	0.9	0.0	0.4	0.0	0.0	0.1	0.0	0.0	0.1	0.0	0.0
7.5	12.6	86.5	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S9. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^A in [**DyY**]^{0}.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	57.0	42.2	0.0	0.0	0.6	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	93.2	5.5	0.7	0.1	0.3	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0
-5.5	0.4	0.3	0.7	0.0	84.7	11.0	2.1	0.0	0.3	0.1	0.0	0.1	0.0	0.1	0.0	0.1
-4.5	0.0	0.0	0.3	0.0	2.1	0.3	90.9	0.6	1.3	0.6	0.6	1.8	0.4	0.6	0.3	0.1
-3.5	0.0	0.0	0.1	0.0	0.2	0.0	2.8	0.1	69.6	3.1	13.6	4.4	0.6	4.3	0.8	0.5
-2.5	0.0	0.0	0.1	0.0	0.0	0.0	2.7	0.0	0.6	2.1	22.4	17.9	35.0	9.9	6.0	3.3
-1.5	0.0	0.0	0.0	0.0	0.1	0.0	0.1	0.0	14.2	0.3	7.0	6.8	5.1	26.9	1.0	38.5
-0.5	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.3	1.8	5.9	19.2	6.1	9.1	8.0	40.2	9.2
0.5	0.0	0.0	0.0	0.0	0.0	0.1	0.3	0.0	5.9	1.8	6.1	19.2	8.0	9.1	9.2	40.2
1.5	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.1	0.3	14.2	6.8	7.0	26.9	5.1	38.5	1.0
2.5	0.0	0.0	0.0	0.1	0.0	0.0	0.0	2.7	2.1	0.6	17.9	22.4	9.9	35.0	3.3	6.0
3.5	0.0	0.0	0.0	0.1	0.0	0.2	0.1	2.8	3.1	69.6	4.4	13.6	4.3	0.6	0.5	0.8
4.5	0.0	0.0	0.0	0.3	0.3	2.1	0.6	90.9	0.6	1.3	1.8	0.6	0.6	0.4	0.1	0.3
5.5	0.3	0.4	0.0	0.7	11.0	84.7	0.0	2.1	0.1	0.3	0.1	0.0	0.1	0.0	0.1	0.0
6.5	0.0	0.0	5.5	93.2	0.1	0.7	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0
7.5	42.2	57.0	0.0	0.0	0.1	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S10. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^A in [**DyY**]$^{2-}$.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	99.5	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	99.2	0.1	0.4	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0
-5.5	0.0	0.0	0.4	0.0	97.5	0.0	0.8	0.1	0.3	0.1	0.0	0.0	0.0	0.0	0.1	0.1
-4.5	0.0	0.0	0.2	0.0	1.0	0.0	85.5	9.1	1.0	0.2	1.1	0.5	0.6	0.1	0.0	0.5
-3.5	0.0	0.0	0.1	0.0	0.4	0.0	0.9	0.1	59.1	23.0	8.1	2.8	1.8	2.1	1.1	0.5
-2.5	0.0	0.0	0.1	0.0	0.0	0.0	2.8	0.3	2.3	0.4	33.3	8.5	35.8	6.3	0.0	10.1
-1.5	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	6.3	3.6	18.5	4.8	0.3	21.0	16.8	28.5
-0.5	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.2	0.8	2.7	18.6	3.7	10.6	21.1	26.1	16.1
0.5	0.0	0.0	0.0	0.0	0.0	0.1	0.2	0.0	2.7	0.8	3.7	18.6	21.1	10.6	16.1	26.1
1.5	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	3.6	6.3	4.8	18.5	21.0	0.3	28.5	16.8
2.5	0.0	0.0	0.0	0.1	0.0	0.0	0.3	2.8	0.4	2.3	8.5	33.3	6.3	35.8	10.1	0.0
3.5	0.0	0.0	0.0	0.1	0.0	0.4	0.1	0.9	23.0	59.1	2.8	8.1	2.1	1.8	0.5	1.1
4.5	0.0	0.0	0.0	0.2	0.0	1.0	9.1	85.5	0.2	1.0	0.5	1.1	0.1	0.6	0.5	0.0
5.5	0.0	0.0	0.0	0.4	0.0	97.5	0.1	0.8	0.1	0.3	0.0	0.0	0.0	0.0	0.1	0.1
6.5	0.0	0.0	0.1	99.2	0.0	0.4	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0
7.5	0.0	99.5	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S11. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^B in [**DyY**]$^{2+}$.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	99.2	0.1	0.0	0.0	0.2	0.1	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	98.1	0.1	0.4	0.3	0.5	0.0	0.0	0.1	0.3	0.1	0.0	0.0	0.1	0.0
-5.5	0.4	0.0	0.6	0.0	54.2	41.6	1.5	0.0	0.1	0.4	0.6	0.3	0.1	0.0	0.0	0.1
-4.5	0.3	0.0	0.6	0.0	1.1	0.7	87.6	0.9	0.7	2.4	1.4	2.0	1.5	0.2	0.5	0.1
-3.5	0.0	0.0	0.5	0.0	0.4	0.3	2.5	0.1	7.6	15.5	35.7	19.3	2.0	9.0	4.8	2.2
-2.5	0.0	0.0	0.1	0.0	0.1	0.1	3.5	0.1	8.2	3.1	7.6	10.6	44.9	0.0	9.7	11.9
-1.5	0.0	0.0	0.0	0.0	0.1	0.1	1.3	0.1	0.4	26.7	2.6	4.0	17.1	15.3	30.2	2.0
-0.5	0.0	0.0	0.0	0.0	0.2	0.1	0.6	1.1	31.1	3.5	8.5	6.8	6.5	3.1	4.7	33.8
0.5	0.0	0.0	0.0	0.0	0.1	0.2	1.1	0.6	3.5	31.1	6.8	8.5	3.1	6.5	33.8	4.7
1.5	0.0	0.0	0.0	0.0	0.1	0.1	0.1	1.3	26.7	0.4	4.0	2.6	15.3	17.1	2.0	30.2
2.5	0.0	0.0	0.0	0.1	0.1	0.1	0.1	3.5	3.1	8.2	10.6	7.6	0.0	44.9	11.9	9.7
3.5	0.0	0.0	0.0	0.5	0.3	0.4	0.1	2.5	15.5	7.6	19.3	35.7	9.0	2.0	2.2	4.8
4.5	0.0	0.3	0.0	0.6	0.7	1.1	0.9	87.6	2.4	0.7	2.0	1.4	0.2	1.5	0.1	0.5
5.5	0.0	0.4	0.0	0.6	41.6	54.2	0.0	1.5	0.4	0.1	0.3	0.6	0.0	0.1	0.1	0.0
6.5	0.0	0.0	0.1	98.1	0.3	0.4	0.0	0.5	0.1	0.0	0.1	0.3	0.0	0.0	0.0	0.1
7.5	0.1	99.2	0.0	0.0	0.1	0.2	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S12. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^B in [**DyY**]^{0}.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	99.3	0.0	0.0	0.0	0.3	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	64.0	34.5	0.3	0.0	0.5	0.0	0.1	0.1	0.4	0.0	0.1	0.0	0.0	0.0
-5.5	0.4	0.0	0.2	0.1	97.0	0.1	0.7	0.0	0.2	0.2	0.9	0.0	0.1	0.0	0.1	0.0
-4.5	0.3	0.0	0.4	0.2	1.1	0.0	89.4	2.2	0.0	2.0	1.8	0.9	0.6	0.7	0.3	0.3
-3.5	0.0	0.0	0.3	0.2	0.7	0.0	1.5	0.1	15.3	9.9	49.0	5.3	9.3	1.9	3.4	3.2
-2.5	0.0	0.0	0.1	0.0	0.2	0.0	2.8	0.1	6.0	5.0	8.7	12.1	22.0	22.3	17.8	2.8
-1.5	0.0	0.0	0.0	0.0	0.2	0.0	1.2	0.2	5.0	20.3	2.1	4.4	29.9	5.2	14.2	17.2
-0.5	0.0	0.0	0.0	0.0	0.1	0.0	0.5	0.6	26.9	9.0	8.6	5.7	4.8	3.2	29.8	10.8
0.5	0.0	0.0	0.0	0.0	0.0	0.1	0.6	0.5	9.0	26.9	5.7	8.6	3.2	4.8	10.8	29.8
1.5	0.0	0.0	0.0	0.0	0.0	0.2	0.2	1.2	20.3	5.0	4.4	2.1	5.2	29.9	17.2	14.2
2.5	0.0	0.0	0.0	0.1	0.0	0.2	0.1	2.8	5.0	6.0	12.1	8.7	22.3	22.0	2.8	17.8
3.5	0.0	0.0	0.2	0.3	0.0	0.7	0.1	1.5	9.9	15.3	5.3	49.0	1.9	9.3	3.2	3.4
4.5	0.0	0.3	0.2	0.4	0.0	1.1	2.2	89.4	2.0	0.0	0.9	1.8	0.7	0.6	0.3	0.3
5.5	0.0	0.4	0.1	0.2	0.1	97.0	0.0	0.7	0.2	0.2	0.0	0.9	0.0	0.1	0.0	0.1
6.5	0.0	0.0	34.5	64.0	0.0	0.3	0.0	0.5	0.1	0.1	0.0	0.4	0.0	0.1	0.0	0.0
7.5	0.0	99.3	0.0	0.0	0.0	0.3	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S13. Weights of M_J components (in %) in the wavefunction of the J = 15/2 ground state for Dy^B in [**DyY**]$^{2-}$.

M_J	w.f. 1	w.f. 2	w.f. 3	w.f. 4	w.f. 5	w.f. 6	w.f. 7	w.f. 8	w.f. 9	w.f. 10	w.f. 11	w.f. 12	w.f. 13	w.f. 14	w.f. 15	w.f. 16
-7.5	99.3	0.1	0.0	0.0	0.3	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
-6.5	0.0	0.0	2.1	96.4	0.2	0.0	0.5	0.0	0.1	0.1	0.3	0.0	0.0	0.0	0.0	0.0
-5.5	0.4	0.0	0.0	0.3	96.7	0.2	0.9	0.0	0.5	0.1	0.7	0.0	0.1	0.0	0.0	0.0
-4.5	0.2	0.0	0.0	0.5	1.2	0.0	92.0	0.1	1.2	0.3	1.9	0.6	0.1	1.0	0.0	0.7
-3.5	0.0	0.0	0.0	0.4	0.7	0.0	1.7	0.0	26.3	13.1	38.1	3.9	5.9	2.1	6.4	1.3
-2.5	0.0	0.0	0.0	0.1	0.2	0.0	2.2	0.0	6.6	0.4	15.4	8.6	17.1	26.5	11.5	11.3
-1.5	0.0	0.0	0.0	0.0	0.2	0.0	1.1	0.2	9.2	11.9	1.8	7.2	34.8	2.1	8.1	23.4
-0.5	0.0	0.0	0.0	0.0	0.2	0.0	0.3	0.7	26.7	3.3	15.6	5.7	9.1	1.2	35.4	1.7
0.5	0.0	0.0	0.0	0.0	0.0	0.2	0.7	0.3	3.3	26.7	5.7	15.6	1.2	9.1	1.7	35.4
1.5	0.0	0.0	0.0	0.0	0.0	0.2	0.2	1.1	11.9	9.2	7.2	1.8	2.1	34.8	23.4	8.1
2.5	0.0	0.0	0.1	0.0	0.0	0.2	0.0	2.2	0.4	6.6	8.6	15.4	26.5	17.1	11.3	11.5
3.5	0.0	0.0	0.4	0.0	0.0	0.7	0.0	1.7	13.1	26.3	3.9	38.1	2.1	5.9	1.3	6.4
4.5	0.0	0.2	0.5	0.0	0.0	1.2	0.1	92.0	0.3	1.2	0.6	1.9	1.0	0.1	0.7	0.0
5.5	0.0	0.4	0.3	0.0	0.2	96.7	0.0	0.9	0.1	0.5	0.0	0.7	0.0	0.1	0.0	0.0
6.5	0.0	0.0	96.4	2.1	0.0	0.2	0.0	0.5	0.1	0.1	0.0	0.3	0.0	0.0	0.0	0.0
7.5	0.1	99.3	0.0	0.0	0.0	0.3	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table S14. Mulliken atomic charges of **[DyY]**$^{2+/0/2-}$ with Dy^{A} atom.

	Coordinate / Å			Mulliken atomic charges / e		
	x	*y*	*z*	cation	neutral	anion
Dy^{A}	0	0	2.07902	1.685841	1.646197	1.55747
Y	-0.85262	1.34723	-1.14697	1.362022	1.390006	1.394089
S	0.5705	-1.57826	-1.01142	1.036028	1.0112	0.954502
N	0	0	0	-2.29299	-2.26614	-2.25072
C	0.483	-0.24695	-3.88475	-0.07499	-0.06973	-0.08933
C	-0.98334	-0.36067	-3.82554	0.009524	-0.0249	-0.04223
C	-1.35453	-1.67069	-3.37713	0.051869	0.038555	-0.00062
C	-0.1167	-2.43156	-3.18612	-0.05049	-0.07053	-0.06381
C	0.9817	-1.59206	-3.50784	-0.04136	-0.07935	-0.09935
C	2.27425	-1.70251	-2.87012	0.009201	0.003774	-0.1339
C	2.41253	-2.81591	-1.91509	0.025704	-0.00023	0.007998
C	1.29996	-3.62713	-1.54303	0.076813	0.056196	0.028953
C	-0.02641	-3.43874	-2.16083	0.015077	-0.0032	-0.14812
C	-1.18543	-3.63066	-1.35873	-0.02856	-0.07475	-0.0042
C	-1.09516	-3.927	0.04888	-0.05685	-0.07176	-0.15139
C	0.1628	-3.97712	0.71792	0.062503	0.060686	0.06166
C	1.32615	-3.83804	-0.11057	-0.05429	-0.08223	-0.09787
C	2.46817	-3.16953	0.44435	-0.0866	-0.08811	-0.11996
C	2.50401	-2.63013	1.78235	0.104976	0.076107	0.06361
C	1.37749	-2.86556	2.60918	0.010021	-0.00111	-0.03014
C	0.20855	-3.53206	2.04458	-0.00748	-0.06211	-0.0982
C	-0.9555	-3.0021	2.6835	0.048299	0.033662	0.03244
C	-0.51928	-2.02266	3.67194	-0.12473	-0.14256	-0.18313
C	0.9576	-1.91047	3.61146	-0.02131	-0.05979	-0.05666
C	1.65416	-0.73282	3.80408	-0.05368	-0.05221	-0.09641
C	0.88401	0.42789	4.22427	0.029761	-0.00329	-0.04583
C	-0.61809	0.30965	4.33966	-0.07774	-0.09854	-0.10116
C	-1.30756	-0.8836	4.02457	0.097847	0.060603	0.028803
C	-2.56326	-0.77727	3.29206	-0.09018	-0.1093	-0.116
C	-3.07825	0.53726	2.86987	-0.05635	-0.07785	-0.09966
C	-2.37534	1.68603	3.07342	0.079575	0.033955	0.024712
C	-1.12652	1.61745	3.82434	-0.00752	-0.02109	-0.0491
C	-0.03015	2.43739	3.41912	-0.08989	-0.12712	-0.14544
C	-0.0807	3.41992	2.33322	0.102381	0.075121	0.05632
C	-1.36862	3.54831	1.69194	-0.01057	-0.02666	-0.04798
C	-2.43212	2.71485	2.05138	-0.0105	-0.02071	-0.04731
C	-3.20502	2.49835	0.84103	0.049707	0.018654	0.020203
C	-2.61937	3.21948	-0.26082	-0.07383	-0.08763	-0.14919
C	-1.44621	3.88926	0.29324	-0.01559	-0.04122	-0.04878
C	-0.30712	4.0688	-0.48624	0.017118	-0.00284	-0.02071
C	-0.35822	3.683	-1.88585	-0.04119	-0.07635	-0.12467
C	-1.56671	3.07583	-2.50314	0.025573	0.00406	-0.02393
C	-2.70272	2.79275	-1.63542	0.042673	0.01028	-0.00365
C	-3.40832	1.55745	-1.81741	-0.09982	-0.11329	-0.13352
C	-2.98777	0.54609	-2.78904	-0.00026	-0.04626	-0.05998
C	-1.78822	0.79949	-3.56522	-0.01965	-0.02225	-0.05237
C	-1.11607	2.04375	-3.45022	0.06456	0.014096	-0.00934
C	0.35224	2.07352	-3.41391	-0.15896	-0.16652	-0.19735
C	0.82211	3.07868	-2.47179	0.002458	-0.02399	-0.02428
C	2.05991	2.90542	-1.76274	0.014513	0.020501	-0.02738
C	2.87051	1.76748	-2.13022	0.075072	0.016903	0.029335
C	2.43519	0.81865	-3.00906	-0.01445	-0.01639	-0.10915
C	1.14294	0.89434	-3.65519	0.161429	0.112562	0.117115
C	2.9338	-0.51507	-2.64234	-0.02361	-0.07993	-0.00879
C	3.71986	-0.2795	-1.47052	-0.00212	-0.0139	-0.08261
C	3.67925	1.14062	-1.12307	-0.0015	-0.04924	-0.08212
C	3.67672	1.58735	0.19365	0.024395	0.028184	0.019176
C	3.81867	0.60319	1.20632	0.00578	-0.0175	-0.01991
C	3.86317	-0.79339	0.89964	0.05209	0.00037	-0.05798
C	3.78683	-1.29274	-0.44262	0.032485	0.024449	0.042099
C	3.15478	-2.53264	-0.67192	-0.00726	-0.04505	-0.08091
C	3.23093	-1.47635	1.98748	-0.04376	-0.05108	-0.06497
C	2.8151	-0.48607	2.97897	0.019494	-0.02959	-0.02762
C	3.16872	0.77749	2.47224	-0.05419	-0.06151	-0.10662
C	2.3454	1.9118	2.77372	0.097359	0.04684	0.005267
C	1.22443	1.70622	3.64523	-0.0925	-0.0908	-0.08622
C	1.0479	3.67092	1.58031	0.004303	-0.03527	-0.05919
C	2.2631	2.92205	1.78371	-0.0252	-0.03212	-0.03262
C	2.92515	2.7786	0.56204	0.059323	-0.00521	-0.05398
C	2.12796	3.42237	-0.46021	0.00123	-0.04116	-0.04505
C	0.98455	3.97777	0.17312	0.019721	0.000243	-0.02406
C	-2.46072	-1.87254	-2.55109	0.003641	-0.02496	-0.01195
C	-2.38908	-2.88496	-1.566	0.046089	0.038185	-0.02905
C	-3.03502	-2.71876	-0.33258	-0.04612	-0.10326	-0.11906
C	-3.7849	-1.57091	0.02094	0.139221	0.13855	0.111154
C	-3.92765	-0.56776	-0.99125	-0.07422	-0.09906	-0.11041
C	-3.30165	-0.72843	-2.27431	0.049655	0.033728	-0.00744
C	-3.9867	0.83131	-0.70457	0.083054	0.046204	0.01634
C	-3.87141	1.29299	0.63697	-0.0232	-0.03424	-0.06691
C	-3.84025	0.28363	1.68446	0.0881	0.05941	0.038268
C	-3.77602	-1.07877	1.35165	-0.07454	-0.10782	-0.12461
C	-2.9872	-1.75021	2.34613	0.077448	0.03377	0.012787
C	-2.18829	-2.87714	1.98392	-0.0169	-0.01987	-0.04307
C	-2.25561	-3.39257	0.69084	0.080981	0.038506	0.02563

Table S15. Mulliken atomic charges of **[DyY]$^{2+/0/2-}$** with Dy^{B} atom.

	Coordinate / Å			Mulliken atomic charges / e		
	x	*y*	*z*	cation	neutral	anion
Y	0	0	2.07902	1.366826	1.415176	1.441272
Dy^{B}	-0.85262	1.34723	-1.14697	1.696511	1.638486	1.532189
S	0.5705	-1.57826	-1.01142	1.044402	1.017553	0.974069
N	0	0	0	-2.31004	-2.28597	-2.27612
C	0.483	-0.24695	-3.88475	-0.06877	-0.0641	-0.08253
C	-0.98334	-0.36067	-3.82554	0.00037	-0.03264	-0.04574
C	-1.35453	-1.67069	-3.37713	0.050469	0.036537	-0.00404
C	-0.1167	-2.43156	-3.18612	-0.05169	-0.07099	-0.06133
C	0.9817	-1.59206	-3.50784	-0.03703	-0.07481	-0.09944
C	2.27425	-1.70251	-2.87012	0.004718	-0.00014	-0.13428
C	2.41253	-2.81591	-1.91509	0.026398	0.000261	0.008583
C	1.29996	-3.62713	-1.54303	0.077248	0.057267	0.028552
C	-0.02641	-3.43874	-2.16083	0.016879	-0.00218	-0.14626
C	-1.18543	-3.63066	-1.35873	-0.02876	-0.074	-0.00427
C	-1.09516	-3.927	0.04888	-0.05758	-0.0723	-0.15101
C	0.1628	-3.97712	0.71792	0.055723	0.055108	0.054926
C	1.32615	-3.83804	-0.11057	-0.05024	-0.07832	-0.09211
C	2.46817	-3.16953	0.44435	-0.09181	-0.09287	-0.12517
C	2.50401	-2.63013	1.78235	0.101368	0.072746	0.061158
C	1.37749	-2.86556	2.60918	0.022525	0.009419	-0.02489
C	0.20855	-3.53206	2.04458	-0.00402	-0.05964	-0.09489
C	-0.9555	-3.0021	2.6835	0.058559	0.042214	0.037032
C	-0.51928	-2.02266	3.67194	-0.10987	-0.13171	-0.17688
C	0.9576	-1.91047	3.61146	-0.01052	-0.05293	-0.05498
C	1.65416	-0.73282	3.80408	-0.0742	-0.07573	-0.12529
C	0.88401	0.42789	4.22427	0.055184	0.012749	-0.03911
C	-0.61809	0.30965	4.33966	-0.04356	-0.07065	-0.08164
C	-1.30756	-0.8836	4.02457	0.081462	0.038882	0.001342
C	-2.56326	-0.77727	3.29206	-0.06848	-0.08817	-0.09789
C	-3.07825	0.53726	2.86987	-0.05764	-0.0813	-0.10581
C	-2.37534	1.68603	3.07342	0.086054	0.039865	0.029223
C	-1.12652	1.61745	3.82434	-0.00015	-0.01855	-0.05242
C	-0.03015	2.43739	3.41912	-0.08316	-0.1213	-0.14318
C	-0.0807	3.41992	2.33322	0.107639	0.078033	0.056491
C	-1.36862	3.54831	1.69194	-0.0188	-0.03328	-0.05273
C	-2.43212	2.71485	2.05138	-0.00394	-0.01588	-0.04454
C	-3.20502	2.49835	0.84103	0.032053	0.003574	0.008159
C	-2.61937	3.21948	-0.26082	-0.09227	-0.10397	-0.16336
C	-1.44621	3.88926	0.29324	-0.0067	-0.03014	-0.03498
C	-0.30712	4.0688	-0.48624	0.009157	-0.01045	-0.02539
C	-0.35822	3.683	-1.88585	-0.04353	-0.07226	-0.1142
C	-1.56671	3.07583	-2.50314	-0.02073	-0.03675	-0.05994
C	-2.70272	2.79275	-1.63542	0.069414	0.042875	0.035894
C	-3.40832	1.55745	-1.81741	-0.13205	-0.14101	-0.15726
C	-2.98777	0.54609	-2.78904	-0.00299	-0.04474	-0.05442
C	-1.78822	0.79949	-3.56522	-0.0165	-0.01711	-0.04665
C	-1.11607	2.04375	-3.45022	0.050804	0.006438	-0.0097
C	0.35224	2.07352	-3.41391	-0.16419	-0.16755	-0.1941
C	0.82211	3.07868	-2.47179	-0.02756	-0.05115	-0.04527
C	2.05991	2.90542	-1.76274	0.008783	0.017238	-0.03169
C	2.87051	1.76748	-2.13022	0.071353	0.014407	0.030842
C	2.43519	0.81865	-3.00906	-0.01506	-0.01548	-0.10997
C	1.14294	0.89434	-3.65519	0.157881	0.109644	0.11683
C	2.9338	-0.51507	-2.64234	-0.01715	-0.07538	-0.00591
C	3.71986	-0.2795	-1.47052	-0.00211	-0.01361	-0.08011
C	3.67925	1.14062	-1.12307	-0.00017	-0.04777	-0.08204
C	3.67672	1.58735	0.19365	0.02782	0.031202	0.023305
C	3.81867	0.60319	1.20632	0.006458	-0.01797	-0.0219
C	3.86317	-0.79339	0.89964	0.051874	-0.00021	-0.05811
C	3.78683	-1.29274	-0.44262	0.027447	0.019764	0.036186
C	3.15478	-2.53264	-0.67192	-0.00457	-0.04161	-0.0757
C	3.23093	-1.47635	1.98748	-0.03617	-0.04603	-0.06401
C	2.8151	-0.48607	2.97897	0.044261	-0.00621	-0.00721
C	3.16872	0.77749	2.47224	-0.05544	-0.06431	-0.11143
C	2.3454	1.9118	2.77372	0.104154	0.054313	0.010228
C	1.22443	1.70622	3.64523	-0.07916	-0.08088	-0.08077
C	1.0479	3.67092	1.58031	0.00199	-0.03737	-0.06127
C	2.2631	2.92205	1.78371	-0.01416	-0.02483	-0.02715
C	2.92515	2.7786	0.56204	0.058957	-0.00469	-0.05528
C	2.12796	3.42237	-0.46021	0.000335	-0.04176	-0.04309
C	0.98455	3.97777	0.17312	0.005121	-0.01116	-0.03281
C	-2.46072	-1.87254	-2.55109	0.005253	-0.02281	-0.00874
C	-2.38908	-2.88496	-1.566	0.050805	0.042373	-0.025
C	-3.03502	-2.71876	-0.33258	-0.04748	-0.10415	-0.12206
C	-3.7849	-1.57091	0.02094	0.13855	0.138388	0.113709
C	-3.92765	-0.56776	-0.99125	-0.08361	-0.10665	-0.11783
C	-3.30165	-0.72843	-2.27431	0.040054	0.025062	-0.01359
C	-3.9867	0.83131	-0.70457	0.072771	0.037842	0.011862
C	-3.87141	1.29299	0.63697	-0.02726	-0.03751	-0.0714
C	-3.84025	0.28363	1.68446	0.096052	0.065987	0.04505
C	-3.77602	-1.07877	1.35165	-0.07131	-0.1049	-0.12355
C	-2.9872	-1.75021	2.34613	0.084296	0.038211	0.016125
C	-2.18829	-2.87714	1.98392	-0.01479	-0.01806	-0.04294
C	-2.25561	-3.39257	0.69084	0.07722	0.034328	0.022347