Key Role of Frustration in Suppression of Magnetization Blocking in Single-Molecule Magnets

Electronic Supplementary Information

*Veacheslav Vieru,[1] Liviu Ungur,[1] & Liviu F. Chibotaru[1]**

1. *Theory of Nanomaterials Group, Department of Chemistry, and Institute of Nanoscale Physics and Chemistry - INPAC, KU Leuven, Celestijnenlaan 200F, 3001 Leuven, Belgium*

Contents

I. Computational details

Structures of the *c3,csa,csb*-$DySc_2N@C_{80}$, *c3,csa*-$Dy_2ScN@C_{80}$ and of *c3,cs,optimized*-$Dy_3N@C_{80}$ were optimized at the B3LYP level of theory, using SVP basis sets for the carbon and nitrogen atoms and Dy: ECP55MWB (7s6p5d)/[5s4p3d] basis, ECP55MWB effective core potential by Dolg et al.; Sc: ECP10MDF core potential, (8s7p6d2f)/[6s5p3d2f] basis (g function was removed from the basis) for the metal atoms. The geometry optimizations were done by Alexey Popov, and the detailed characterization of these structures will be published in a separated paper. The structure of the X-ray-$Dy_3N@C_{80}$ complex was taken from the following paper: S. Yang, S.I. Troyanov, A. Popov, M. Krause and L. Dunsch, *J. Am. Chem. Soc.,* **2006**, 128,16733-16739.

All calculations were carried out with MOLCAS 7.8 and are of CASSCF/RASSI/SINGLE_ANISO type.
Each magnetic center was calculated keeping the geometry of the entire molecule replacing the neighboring Dy(III) ions by nonmagnetic Lu(III) ions.

Two basis set approximations have been employed: **1** – small, and **2** – large. Table S1 shows the contractions of the employed basis sets for all elements.

Table S1. Contraction of the employed basis sets in computational approximations **1** and **2**.

Basis 1	**Basis 2**
Dy.ANO-RCC...7s6p4d2f1g.	Dy.ANO-RCC... 8s7p5d3f2g1h.
Sc.ANO-RCC...5s4p2d1f.	Sc.ANO-RCC...6s5p3d2f1g.
Lu.ANO-RCC...7s6p4d2f1g.	Lu.ANO-RCC... 8s7p5d3f2g1h.
N.ANO-RCC... 3s2p.	N.ANO-RCC... 4s3p2d1f.
C.ANO-DK3.Tsuchiya.12s8p.2s1p.	C.ANO-RCC... 3s2p1d. (close)
	C.ANO-DK3.Tsuchiya.12s8p.2s1p. (distant)

Active space of the CASSCF method included 9 electrons in 7 orbitals (4*f* orbitals of Dy^{3+} ion).
We have mixed 21 sextets, 128 quartet and 130 doublet states by spin-orbit coupling.
On the basis of the resulting spin-orbital multiplets SINGLE_ANISO program computed local magnetic properties (*g*-tensors, magnetic axes, local magnetic susceptibility, etc.)
The magnetic properties of the polynuclear complexes were calculated using the POLY_ANISO package.

II. The $DySc_2N@C_{80}$ compound

The electronic and magnetic properties of Dy center

Table S2. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy center for different optimized structures.

Spin-orbit energies, cm^{-1}					
Dy_c3_basis1	**Dy_c3_basis2**	**Dy_csa_basis1**	**Dy_csa_basis2**	**Dy_csb_basis1**	**Dy_csb_basis2**
0	0	0	0	0	0
331	372	373	412	377	415
655	729	708	780	674	747
906	1009	966	1067	900	1003
1049	1180	1091	1229	1032	1159
1125	1268	1153	1297	1119	1256
1201	1338	1246	1382	1191	1334
1254	1401	1365	1501	1308	1466

Table S3. The *g* tensors of the low-lying Kramers doublets (KD) of Dy center.

KD		Dy_c3_basis1	Dy_c3_basis2	Dy_csa_basis1	Dy_csa_basis2	Dy_csb_basis1	Dy_csb_basis2
		g	***g***	***g***	***g***	***g***	***g***
1	g_X	2.6E-05	3.3E-05	1.9E-05	5.2E-05	8.3E-05	8.3E-05
	g_Y	3.7E-05	4.4E-05	1.9E-05	5.6E-05	1.1E-04	1.1E-04
	g_Z	19.86	19.86	19.84	19.85	19.86	19.86
2	g_X	7.9E-04	5.8E-04	1.5E-04	4.3E-04	0.0025	0.0027
	g_Y	9.6E-04	6.9E-04	2.4E-04	5.2E-04	0.0027	0.0029
	g_Z	17.08	17.06	17.13	17.09	17.14	17.10
3	g_X	0.027	0.024	0.016	0.0086	0.011	0.020
	g_Y	0.032	0.028	0.021	0.012	0.013	0.023
	g_Z	14.29	14.28	14.33	14.31	14.30	14.28
4	g_X	0.35	0.22	0.68	0.42	0.46	0.36
	g_Y	0.46	0.29	0.77	0.44	0.57	0.46
	g_Z	11.41	11.51	11.39	11.51	11.73	11.66
5	g_X	3.36	2.38	4.50	7.11	0.48	0.84
	g_Y	5.08	3.20	5.52	6.43	1.46	1.59
	g_Z	7.67	8.33	10.86	4.71	10.72	10.34
6	g_X	1.96	8.26	0.48	1.10	7.62	7.61
	g_Y	4.52	6.11	1.42	4.88	5.28	5.25
	g_Z	9.69	2.91	11.25	10.30	1.92	1.86
7	g_X	1.07	1.30	1.01	0.87	2.17	2.10
	g_Y	1.38	1.96	2.25	2.24	5.73	5.81
	g_Z	16.03	15.59	14.95	14.47	12.59	12.50
8	g_X	0.33	0.31	0.17	0.17	0.18	0.16
	g_Y	0.57	0.64	0.47	0.49	0.52	0.44
	g_Z	19.15	18.98	19.11	19.02	19.12	19.13

Using the SINGLE_ANISO module we calculated the molar magnetization of the $DySc_2N@C_{80}$ compound and compared it with the experimental data. The calculated magnetization curve fits very well with the experimental one.

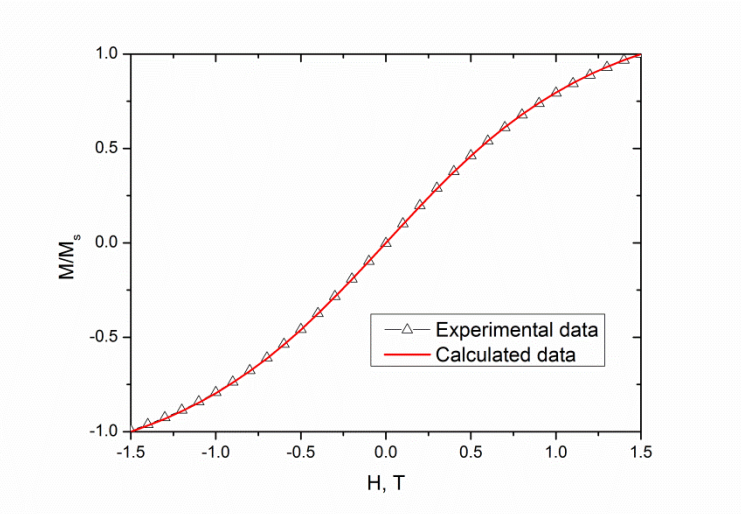

Figure S1: The calculated *vs*. experimental magnetization curves of the $DySc_2N@C_{80}$ at 6 K. The experimental data have been provided by Thomas Greber.

III. The $Dy_2ScN@C_{80}$ compounds

The electronic and magnetic properties of individual Dy centers

Table S4. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for *c3*-$Dy_2ScN@C_{80}$ optimized structure.

Spin-orbit energies, cm^{-1}			
Dy1_c3_basis1	**Dy1_c3_basis2**	**Dy2_c3_basis1**	**Dy2_c3_basis2**
0	0	0	0
431	470	398	435
716	787	701	776
927	1022	929	1036
1062	1173	1064	1197
1159	1284	1151	1294
1232	1364	1232	1379
1343	1465	1322	1455

Table S5. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for *csa*-$Dy_2ScN@C_{80}$ optimized structure.

Spin-orbit energies, cm^{-1}			
Dy1_csa_basis1	**Dy1_csa_basis2**	**Dy2_csa_basis1**	**Dy2_csa_basis2**
0	0	0	0
474	507	347	393
747	812	697	777
932	1018	964	1074
1061	1161	1085	1230
1169	1282	1153	1303
1266	1380	1232	1382
1391	1518	1351	1514

Table S6. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for *c3*-$Dy_2ScN@C_{80}$ optimized structure.

KD		Dy1_c3_basis1	Dy1_c3_basis2	Dy2_c3_basis1	Dy2_c3_basis2
		g	*g*	*g*	*g*
1	g_X	8.0E-06	1.0E-06	4.9E-05	3.5E-05
	g_Y	1.8E-05	3.2E-05	5.6E-05	4.3E-05
	g_Z	19.88	19.88	19.87	19.87
2	g_X	0.0035	0.0041	0.0027	0.0014
	g_Y	0.0040	0.0044	0.0031	0.0016
	g_Z	17.17	17.11	17.14	17.10
3	g_X	0.083	0.056	0.09	0.065
	g_Y	0.098	0.062	0.10	0.074
	g_Z	14.31	14.32	14.26	14.27
4	g_X	1.24	0.98	0.72	0.42
	g_Y	1.47	1.14	0.86	0.50
	g_Z	11.25	11.43	11.30	11.48
5	g_X	3.47	3.49	3.63	2.66
	g_Y	4.28	4.14	4.10	2.88
	g_Z	8.93	9.13	8.31	8.93
6	g_X	1.10	0.61	2.44	3.17
	g_Y	3.74	4.01	3.79	4.50
	g_Z	9.98	9.35	10.26	9.15
7	g_X	1.64	1.94	1.13	1.50
	g_Y	3.70	4.23	2.03	2.38
	g_Z	13.61	12.61	14.81	13.50
8	g_X	0.19	0.27	0.21	0.36
	g_Y	0.55	0.86	0.65	1.29
	g_Z	18.77	18.43	18.63	18.07

Table S7. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for *csa*-$Dy_2ScN@C_{80}$ optimized structure.

KD		Dy1_csa_basis1	Dy1_csa_basis2	Dy2_csa_basis1	Dy2_csa_basis2
		g	*g*	*g*	*g*
1	g_X	2.7E-04	3.3E-04	6.3E-05	8.3E-05
	g_Y	3.5E-04	4.3E-04	8.6E-05	1.1E-04
	g_Z	19.87	19.88	19.82	19.84
2	g_X	0.014	0.018	0.0013	0.0015
	g_Y	0.018	0.022	0.0015	0.0017
	g_Z	17.06	17.03	17.04	17.03
3	g_X	0.17	0.18	0.036	0.034
	g_Y	0.19	0.20	0.042	0.038
	g_Z	14.26	14.25	14.27	14.25
4	g_X	1.33	1.38	0.46	0.38
	g_Y	1.50	1.56	0.58	0.45
	g_Z	11.70	11.60	11.23	11.37
5	g_X	0.50	1.22	2.43	2.99
	g_Y	3.12	3.93	3.88	3.84
	g_Z	9.26	9.05	10.81	9.36
6	g_X	8.43	8.64	3.05	3.06
	g_Y	5.80	5.48	4.52	4.03
	g_Z	2.94	1.97	10.18	9.78
7	g_X	1.82	1.90	0.88	1.00
	g_Y	3.63	4.25	2.11	2.58
	g_Z	14.06	13.74	15.56	15.12
8	g_X	0.20	0.18	0.12	0.095
	g_Y	0.55	0.47	0.32	0.24
	g_Z	19.06	19.13	19.30	19.30

IV. Account of the total magnetic interaction in *csa*-$Dy_2ScN@C_{80}$ complex

Using the exchange parameters obtained from BS-DFT (see the part VI) we simulated the magnetic properties of the complex.

Table S8. Exchange interactions between Dy ions in the complex *csa*-**$Dy_2N@C_{80}$**.
Ising parameters (cm^{-1}):

Molecule/approximation		J_{dip}*	J_{exch}	$J_{total} = J_{dip}$*$+ J_{exch}$
***csa*-Dy_2Sc/basis2**	Dy- -Dy	6.43	10.78	17.21

* -- contribution coming only from the Ising terms $\sim \hat{\tilde{s}}_{1,z}\hat{\tilde{s}}_{2,z}$ to the dipolar coupling. In the calculation of the exchange spectrum (Table S9) the dipolar interaction included all terms.

Table S9. Energies (cm^{-1}) and the corresponding tunneling gaps and g_Z values of the lowest 2 exchange doublet states of the complex *csa*-**$Dy_2ScN@C_{80}$** considering the total magnetic interaction.

***csa*-Dy_2Sc/basis2**		
energy	Δ_{tun}	g_Z
0.00 3.25E-10	3.25E-10	34.13
8.37 8.37	2.21E-09	20.43

V. The $Dy_3N@C_{80}$ compounds

The electronic and magnetic properties of individual Dy centers

Table S10. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for *c3*-$Dy_3N@C_{80}$ optimized structure.

Spin-orbit energies, cm^{-1}	
basis1	**basis2**
0	0
488	527
775	845
971	1061
1100	1206
1202	1325
1281	1412
1407	1524

Dy1=Dy2=Dy3 by symmetry

Table S11. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for *cs*-$Dy_3N@C_{80}$ optimized structure.

Spin-orbit energies, cm^{-1}			
Dy1_cs_basis1	**Dy1_cs_basis2**	**Dy2_cs_basis1**	**Dy2_cs_basis2**
0	0	0	0
356	403	383	432
679	768	733	806
922	1049	980	1073
1076	1224	1089	1213
1164	1320	1140	1267
1205	1364	1240	1366
1320	1478	1385	1522

Dy2=Dy3 by symmetry

Table S12. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for $Dy_3N@C_{80}$ optimized structure.

Spin-orbit energies, cm^{-1}					
Dy1_basis1	**Dy1_basis2**	**Dy2_basis1**	**Dy2_basis2**	**Dy3_basis1**	**Dy3_basis2**
0	0	0	0	0	0
434	481	438	487	439	485
740	818	741	825	743	820
967	1067	966	1073	967	1066
1103	1221	1100	1229	1101	1217
1178	1315	1177	1327	1178	1313
1246	1390	1246	1403	1247	1390
1366	1499	1367	1513	1369	1500

Table S13. Energies of the low-lying Kramers doublets (cm^{-1}) of Dy centers for the X-ray $Dy_3N@C_{80}$ structure.

Spin-orbit energies, cm^{-1}					
Dy1A_basis1	**Dy1A_basis2**	**Dy2A_basis1**	**Dy2A_basis2**	**Dy3A_basis1**	**Dy3A_basis2**
0	0	0	0	0	0
533	555	539	569	571	610
808	852	820	880	843	924
955	1019	1013	1084	976	1088
1069	1145	1123	1207	1091	1222
1195	1287	1234	1332	1209	1364
1283	1373	1353	1459	1320	1468
1440	1511	1433	1533	1479	1614

Table S14. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for *c3*-$Dy_3N@C_{80}$ optimized structure.

KD		basis1	basis2
		g	***g***
1	g_X	1.1E-04	6.6E-05
	g_Y	1.6E-04	1.1E-04
	g_Z	19.87	19.87
2	g_X	0.0056	0.0054
	g_Y	0.0065	0.0059
	g_Z	17.09	17.04
3	g_X	0.038	0.035
	g_Y	0.054	0.043
	g_Z	14.35	14.34
4	g_X	1.15	0.95
	g_Y	1.41	1.17
	g_Z	11.37	11.49
5	g_X	3.59	3.49
	g_Y	4.31	4.14
	g_Z	9.07	9.08
6	g_X	0.60	8.36
	g_Y	4.39	4.93
	g_Z	9.33	0.24
7	g_X	1.79	2.09
	g_Y	4.13	4.42
	g_Z	13.10	12.34
8	g_X	0.21	0.29
	g_Y	0.58	0.95
	g_Z	18.66	18.29

Table S15. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for *cs*-$Dy_3N@C_{80}$ optimized structure.

KD		**Dy1_cs_basis1**	**Dy1_cs_basis2**	**Dy2_cs_basis1**	**Dy2_cs_basis2**
		g	***g***	***g***	***g***
1	g_X	3.8E-04	3.3E-04	2.1E-04	2.1E-04
	g_Y	4.9E-04	4.2E-04	2.8E-04	2.9E-04
	g_Z	19.85	19.86	19.80	19.82
2	g_X	0.0087	0.0067	0.0026	0.0036
	g_Y	0.011	0.0078	0.0027	0.0038
	g_Z	16.99	16.99	17.07	17.05
3	g_X	0.16	0.13	0.012	0.015
	g_Y	0.19	0.14	0.015	0.019

	g_Z	14.04	14.11	14.35	14.32
4	g_X	0.79	0.66	0.42	0.47
	g_Y	0.91	0.76	0.54	0.58
	g_Z	11.48	11.46	11.55	11.60
5	g_X	0.60	0.31	1.85	0.34
	g_Y	1.20	1.12	2.29	1.45
	g_Z	9.10	9.18	16.42	13.56
6	g_X	6.96	6.18	3.97	8.17
	g_Y	5.50	4.74	5.41	7.07
	g_Z	3.10	2.96	9.72	3.38
7	g_X	1.34	1.48	1.22	1.57
	g_Y	5.26	5.74	1.98	3.00
	g_Z	14.09	14.10	15.73	15.08
8	g_X	0.013	0.037	0.12	0.10
	g_Y	0.11	0.084	0.33	0.30
	g_Z	19.58	19.62	19.27	19.23

Table S16. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for $Dy_3N@C_{80}$ optimized structure.

KD		**Dy1_basis1**	**Dy1_basis2**	**Dy2_basis1**	**Dy2_basis2**	**Dy3_basis1**	**Dy3_basis2**
		g	***g***	***g***	***g***	***g***	***g***
1	g_X	5.3E-05	4.0E-05	4.6E-05	2.8E-05	4.8E-05	3.6E-05
	g_Y	6.8E-05	5.9E-05	6.4E-05	4.8E-05	6.6E-05	5.9E-05
	g_Z	19.87	19.87	19.87	19.87	19.87	19.87
2	g_X	0.0030	0.0033	0.0033	0.0033	0.0034	0.0037
	g_Y	0.0033	0.0036	0.0037	0.0035	0.0037	0.0040
	g_Z	17.19	17.12	17.19	17.12	17.19	17.12
3	g_X	0.049	0.044	0.058	0.047	0.056	0.049
	g_Y	0.057	0.050	0.068	0.054	0.065	0.056
	g_Z	14.32	14.31	14.31	14.30	14.32	14.31
4	g_X	0.88	0.67	0.93	0.69	0.91	0.69
	g_Y	0.98	0.72	1.04	0.72	1.01	0.74
	g_Z	11.33	11.47	11.32	11.46	11.34	11.47
5	g_X	4.08	3.73	4.02	3.69	3.98	3.72
	g_Y	4.61	4.16	4.65	4.10	4.63	4.21
	g_Z	9.68	9.74	9.76	9.66	9.86	9.85
6	g_X	0.27	0.43	0.23	0.36	0.19	0.29
	g_Y	4.89	4.64	4.80	4.84	4.89	4.63
	g_Z	10.72	10.21	10.59	9.77	10.49	10.00
7	g_X	1.64	1.87	1.65	1.91	1.68	1.91
	g_Y	4.14	4.51	4.15	4.68	4.26	4.59
	g_Z	13.24	12.39	13.19	12.19	13.09	12.28
8	g_X	0.21	0.29	0.21	0.29	0.21	0.29
	g_Y	0.62	0.96	0.61	0.95	0.61	0.96
	g_Z	18.77	18.42	18.76	18.39	18.75	18.38

Table S17. The *g* tensors of the low-lying Kramers doublets (KD) of Dy centers for the X-ray $Dy_3N@C_{80}$ structure.

KD		**Dy1A_basis1**	**Dy1A_basis2**	**Dy2A_basis1**	**Dy2A_basis2**	**Dy3A_basis1**	**Dy3A_basis2**
		g	***g***	***g***	***g***	***g***	***g***
1	g_X	2.9E-04	2.3E-04	1.0E-04	1.4E-04	1.9E-04	1.7E-04
	g_Y	3.9E-04	3.3E-04	1.9E-04	2.5E-04	2.7E-04	2.4E-04
	g_Z	19.87	19.88	19.87	19.87	19.87	19.87
2	g_X	0.020	0.024	0.0091	0.011	0.010	0.014
	g_Y	0.024	0.026	0.0095	0.012	0.012	0.016
	g_Z	16.95	16.96	17.11	17.04	16.90	16.90

3	g_X	0.15	0.019	0.087	0.11	0.21	0.10
	g_Y	0.18	0.039	0.098	0.12	0.24	0.11
	g_Z	14.25	14.28	14.51	14.45	14.29	14.30
4	g_X	2.07	1.47	0.51	0.38	1.40	1.11
	g_Y	2.26	1.75	0.74	0.65	1.65	1.28
	g_Z	11.24	11.41	11.77	11.81	11.44	11.51
5	g_X	7.58	7.87	3.04	3.26	2.65	3.56
	g_Y	6.74	6.44	4.68	4.88	5.25	5.54
	g_Z	3.59	4.16	10.89	10.11	8.09	8.00
6	g_X	0.11	1.07	1.16	1.38	1.94	0.096
	g_Y	3.92	4.30	4.40	4.46	4.52	4.76
	g_Z	9.59	8.71	10.27	10.30	11.00	9.81
7	g_X	1.53	1.89	1.02	0.42	1.52	1.98
	g_Y	3.89	4.85	2.57	2.56	3.07	4.51
	g_Z	13.00	11.95	11.59	10.96	13.84	12.30
8	g_X	0.11	0.17	0.67	0.75	0.14	0.21
	g_Y	0.28	0.51	3.14	3.70	0.41	0.66
	g_Z	18.90	18.53	16.46	15.97	18.65	18.36

VI. Account of the total magnetic interaction in $Dy_3N@C_{80}$ complex

BS-DFT calculations on Gd_3N^{6+} complex with the geometry of $Dy_3N@C_{80}$ complex.

The Broken Symmetry DFT calculations were carried out within ORCA, using the B3LYP functional and SVP basis set. The exchange interaction is considered by the phenomenological Heisenberg spin Hamiltonian:

$$\hat{H} = -\sum_{<i,j>} J_{ij} S_i S_j$$

The Dysprosium ions were replaced by gadolinium ions. The C_{80} cage was omitted from the calculation. The exchange coupling constants were calculated using the generalized spin projection method (M. Shoji, K. Koizumi, Y. Kitagawa, T. Kawakami, S. Yamanaka, M. Okumura, K. Yamaguchi, *Chem. Phys. Lett.,* **2006***,* 432, 343–347).

Table S18. Exchange interactions between Dy ions in the complex **$Dy_3N@C_{80}$**.
Ising parameters (cm^{-1}):

Molecule/approximation		**J_{dip}***	**J_{exch}**	**$J_{total} = J_{dip}$*+ J_{exch}**
Dy_3A/basis1	Dy1- -Dy2	6.69	12.26	18.95
	Dy1- -Dy3	6.76	12.08	18.84
	Dy2- -Dy3	6.98	13.89	20.87
Dy_3A/basis2	Dy1- -Dy2	6.69	12.22	18.91
	Dy1- -Dy3	6.76	12.08	18.84
	Dy2- -Dy3	6.98	13.97	20.95

Table S19. Energies (cm^{-1}) and g values of the lowest 4 exchange doublet states of the complex **$Dy_3N@C_{80}$** considering the total magnetic interaction.

$Dy_3N@C_{80}$					
Basis 1			Basis 2		
g		Energy	**g**		Energy
g_x	1.0E-07	0	g_x	1.0E-07	0
g_y	1.0E-07		g_y	3.3E-08	
g_z	39.46		g_z	39.47	
g_x	1.0E-07	0.057	g_x	1.0E-07	0.034
g_y	4.8E-07		g_y	3.4E-07	
g_z	38.85		g_z	38.89	
g_x	1.0E-07	0.97	g_x	1.0E-07	1.00
g_y	2.7E-07		g_y	2.2E-07	
g_z	41.11		g_z	41.08	
g_x	1.0E-07	19.40	g_x	1.0E-07	19.42
g_y	3.3E-08		g_y	8.0E-09	
g_z	2.87		g_z	2.77	

Table S20. The spin-scale relationship for Lines parameters for a series of dinuclear Ln2:*

	distance Ln-Ln (Angstrom)	Fitted Lines parameters (in cm^{-1}) (J. Am. Chem. Soc. 2011, 133, 5319-5328)	Calculated Lines parameters with spin-scaled relationship (in cm^{-1})
Gd_2	3.811(1)	-0.08	-0.08
Tb_2	3.788(1)	-0.10	=(49/36) × (-0.08)=-0.1088
Dy_2	3.768(3)	-0.21	=(49/25) × (-0.08)=-0.1568
Ho_2	3.754(3)	-0.44	=(49/16) × (-0.08)=-0.2450

*The rescaling of the exchange interaction for Gd_2, having spins 7/2 on Gd sites, to the exchange interaction for Ln_2 complexes, with spin S on Ln sites, is done with the formula:

$$J_{exch}(Ln_2)=(7/2)^2/S^2\ J_{exch}(Gd_2)$$

We can see from the Table S20 that the simple rescaling of the exchange parameter for Gd_2 gives reasonable values for the Lines parameters of other lanthanide dimers (last column), in particular, it catches the tendency of the increase of this parameter when moving towards the end of the lanthanide series. We should note that in the real Ln_2 compounds the Ln-Ln distance decrease thus additionally enhancing the Lines exchange parameter when moving towards Gd. If this effect would be taken into account, the agreement between fitted and rescaled (from Gd_2) exchange parameters would be almost perfect.

We may conclude from this example that the rescaling procedure performs satisfactorily well for the isostructural series of lanthanide compounds.

VII. The xyz coordinates of the investigated complexes

All geometry optimizations were carried out by Alexey Popov.

Table S21. The xyz coordinates (Å) of *c3*- $DySc_2N@C_{80}$ compound.

C	0.826954587	2.555466482	3.110890065
C	0.442863883	1.424487856	3.908838584
C	-0.983990648	1.108642818	4.017088624
C	1.368907757	0.315491464	3.890897578
C	2.081655648	2.615762773	2.386524958
C	3.726837147	1.261813820	1.159410762
C	2.966689873	1.504722756	2.369073282
C	2.613572503	0.368221547	3.151674752
C	0.914008591	-1.065003014	3.875785844
C	-0.488503643	-1.403179106	3.886458884
C	-1.445422256	-0.303085277	4.003767227
C	-0.856318132	-2.547994842	3.101230777
C	1.881282383	-1.841022748	3.131516274
C	2.231395230	-3.241069098	1.160837882
C	1.489721975	-2.973198595	2.353594365
C	0.111858423	-3.338415356	2.377730328
C	4.176163929	-0.051975269	0.731551566
C	3.662315106	-1.195335645	1.472561250
C	2.932313684	-0.959452547	2.694395855
C	3.285682225	-2.356033520	0.723841014
C	-2.103905109	-2.621526481	2.365805005
C	-2.979324894	-1.505069746	2.330575473
C	-2.613135777	-0.359137469	3.138485224
C	-3.725303773	-1.273569963	1.129739692
C	-1.904303329	-3.450023072	1.209381582
C	-1.893937190	-3.428462210	-1.242527830
C	-2.581628296	-3.164589767	-0.019347228
C	-3.531613221	-2.096944861	-0.034034862
C	1.588113917	-3.773775170	-0.005939630
C	0.185310035	-4.067757711	-0.010731747
C	-0.532006931	-3.882299494	1.211516980
C	-0.529546798	-3.879172754	-1.238381036
C	-4.043453615	0.056544674	0.685086423
C	-3.610887238	1.199484133	1.421354001
C	-2.902566372	0.962633531	2.655536791
C	-3.283259004	2.374178497	0.695990579
C	-4.064254429	0.060939786	-0.760237573
C	-3.041909815	0.999275506	-2.811414646
C	-3.672019012	1.221482209	-1.515144365
C	-3.286106818	2.375676654	-0.749363306
C	-2.101833625	-2.601423942	-2.415188160
C	-3.064673679	-1.528079042	-2.465662848
C	-3.753510928	-1.284905527	-1.217907494
C	-2.746566042	-0.363944209	-3.306438565
C	-2.240067502	3.259040458	1.150452477
C	-1.503555339	2.993499353	2.344524962
C	-1.890591924	1.853311286	3.151229036
C	-0.128126055	3.361923642	2.377224491
C	-1.590054651	3.807712458	-0.011510611
C	0.527420866	3.904452029	-1.228918268
C	-0.188842118	4.090473642	-0.007194993
C	0.525453656	3.907991444	1.222872978
C	-1.917984113	1.852967606	-3.161465574
C	-1.497831092	2.993337682	-2.370801913
C	-2.231458403	3.264742506	-1.178788390
C	-0.118354345	3.356654874	-2.395055219
C	1.893378357	3.453218758	-1.223123605
C	2.564309174	3.169492466	0.000125711
C	1.886865792	3.444169428	1.225329486
C	3.502326362	2.073004851	-0.002447029
C	2.100544838	2.630374271	-2.388776757

C	2.635716285	0.374031950	-3.146569215
C	2.987063590	1.514365405	-2.365163775
C	3.776465563	1.274834685	-1.170457515
C	-0.938838044	1.061300889	-3.856443308
C	1.401634572	0.313264649	-3.875727798
C	0.468101879	1.394236817	-3.818819351
C	0.851489937	2.567486188	-3.111965390
C	-0.835449984	-2.549154440	-3.123022334
C	0.945732918	-1.054308879	-3.868325089
C	-0.450942111	-1.378770987	-3.842786399
C	-1.422137795	-0.300494387	-3.927874058
C	2.239248848	-3.232326097	-1.167449224
C	1.902236289	-1.837894978	-3.132432597
C	1.502297217	-2.968927839	-2.363542545
C	0.122678721	-3.345732795	-2.403263731
C	4.240545109	-0.049173090	-0.747656949
C	2.960862160	-0.960603298	-2.678807303
C	3.738064100	-1.210634809	-1.492388288
C	3.307665299	-2.359028918	-0.729667723
N	0.189541453	0.015137363	0.207318238
Sc	2.097632469	-0.180729105	-0.186272576
Sc	-0.373716161	0.173840114	2.078985310
Dy	-1.336802063	-0.073029664	-1.340156781

Table S22. The xyz coordinates (Å) of *csa*- $DySc_2N@C_{80}$ compound.

C	-0.717476736	3.180689696	-2.374831154
C	-0.717476736	3.180689696	2.374831154
C	-1.495710755	3.517072605	-1.216547748
C	-1.495710755	3.517072605	1.216547748
C	-2.689828711	2.693421170	-1.211174266
C	-2.689828711	2.693421170	1.211174266
C	-2.622266363	1.837584515	-2.380711797
C	-2.622266363	1.837584515	2.380711797
C	-1.408727082	2.147886424	-3.098976558
C	-1.408727082	2.147886424	3.098976558
C	-0.699389437	1.132166742	-3.806850622
C	-0.699389437	1.132166742	3.806850622
C	0.744813527	1.210017346	-3.850455789
C	0.744813527	1.210017346	3.850455789
C	1.420293668	2.225266296	-3.093907329
C	1.420293668	2.225266296	3.093907329
C	0.711226745	3.223741969	-2.336009417
C	0.711226745	3.223741969	2.336009417
C	1.344546840	3.711970044	-1.162020698
C	1.344546840	3.711970044	1.162020698
C	0.567885559	4.055751908	0.000000000
C	-0.855222572	3.911133828	0.000000000
C	-3.357254167	2.280793256	0.000000000
C	-4.048197574	0.981406576	0.000000000
C	-3.917297241	0.108710230	-1.179630447
C	-3.917297241	0.108710230	1.179630447
C	-3.181434520	0.520049080	-2.368666366
C	-3.181434520	0.520049080	2.368666366
C	-2.490872013	-0.474761869	-3.140736620
C	-2.490872013	-0.474761869	3.140736620
C	-1.274969080	-0.163982330	-3.849581634
C	-1.274969080	-0.163982330	3.849581634
C	-0.429114915	-1.337210602	-3.850710319
C	-0.429114915	-1.337210602	3.850710319
C	1.006378781	-1.256829919	-3.885791793
C	1.006378781	-1.256829919	3.885791793
C	1.624168026	0.061087520	-3.989030457
C	1.624168026	0.061087520	3.989030457
C	2.878480120	0.378975585	-3.270047056
C	2.878480120	0.378975585	3.270047056
C	2.703662985	1.713708011	-2.702672684
C	2.703662985	1.713708011	2.702672684

C	3.282388475	2.153241887	-1.449305675
C	3.282388475	2.153241887	1.449305675
C	2.610515071	3.175487291	-0.721846752
C	2.610515071	3.175487291	0.721846752
C	-3.739319620	-1.257653396	-0.720327164
C	-3.739319620	-1.257653396	0.720327164
C	-2.977899709	-2.249069396	-1.458829164
C	-2.977899709	-2.249069396	1.458829164
C	-2.396902891	-1.844572754	-2.699893601
C	-2.396902891	-1.844572754	2.699893601
C	-1.134060401	-2.375242993	-3.136506707
C	-1.134060401	-2.375242993	3.136506707
C	-2.312620549	-3.270332852	-0.721453799
C	-2.312620549	-3.270332852	0.721453799
C	-1.052478512	-3.812697832	-1.165133236
C	-1.052478512	-3.812697832	1.165133236
C	-0.426783906	-3.336203873	-2.354292596
C	-0.426783906	-3.336203873	2.354292596
C	1.005417174	-3.279012719	-2.384574570
C	1.005417174	-3.279012719	2.384574570
C	1.709017854	-2.256910406	-3.126086077
C	1.709017854	-2.256910406	3.126086077
C	2.938810587	-1.949585061	-2.408019522
C	2.938810587	-1.949585061	2.408019522
C	3.522371338	-0.629364803	-2.426261334
C	3.522371338	-0.629364803	2.426261334
C	4.087612320	-0.191422449	-1.172635063
C	4.087612320	-0.191422449	1.172635063
C	4.030635392	1.184305588	-0.715348630
C	4.030635392	1.184305588	0.715348630
C	-0.271424699	-4.135361758	0.000000000
C	1.148777801	-3.999011434	0.000000000
C	1.782338333	-3.604989986	-1.223837105
C	1.782338333	-3.604989986	1.223837105
C	2.965993279	-2.790321532	-1.226759251
C	2.965993279	-2.790321532	1.226759251
C	3.541916747	-2.334659008	0.000000000
C	4.124099331	-1.033840712	0.000000000
N	0.444831348	-0.028460508	0.000000000
Dy	-1.703123196	0.352143753	0.000000000
Sc	1.336910161	-0.167858337	-1.743094049
Sc	1.336910161	-0.167858337	1.743094049

Table S23. The xyz coordinates (Å) of *csb*- $DySc_2N@C_{80}$ compound.

C	-0.430866895	-0.987713995	1.220353725
C	0.765998747	-0.423931460	1.776191210
C	1.605996883	-1.507307110	2.236020734
C	0.917220967	-2.751424420	1.948294869
C	-0.347830541	-2.419876201	1.318409287
C	-0.917721699	-3.271289419	0.325020821
C	-1.686359765	-2.672690311	-0.750862514
C	-1.699351604	-1.236882844	-0.860596176
C	-1.081163541	-0.365609814	0.109382266
C	-0.574551981	0.876532253	-0.360720745
C	0.619573050	1.451077985	0.203943727
C	1.330721320	0.779683777	1.242151902
C	2.755520344	0.892228598	1.257513169
C	3.585461718	-0.201222368	1.706741733
C	3.044859254	-1.422333874	2.237646083
C	3.800142856	-2.660452986	2.042641166
C	3.092230959	-3.930651839	1.748069944
C	1.633930311	-3.961581278	1.654064728
C	1.029412679	-4.809449287	0.657238217
C	-0.225902137	-4.477910150	0.028149219
C	-0.231196162	-5.023771724	-1.313197913
C	-0.983379603	-4.429486495	-2.392993998
C	-1.815678733	-3.257536555	-2.088657324
C	-1.852308738	-2.117558735	-3.032997813
C	-1.775338020	-0.894000623	-2.252150146
C	-1.177291613	0.337859688	-2.726087774

C	-0.619729047	1.221662575	-1.760970531
C	0.556686870	2.001535124	-2.063944795
C	1.323452035	2.134424703	-0.850606596
C	2.746676629	2.164802938	-0.881051105
C	3.456507809	1.587738625	0.215071761
C	4.730008004	0.943299358	0.012381040
C	4.806757497	-0.162479826	0.928399731
C	5.479460304	-1.367626814	0.590154378
C	4.944002581	-2.598058623	1.141948064
C	4.976307848	-3.793088769	0.340583350
C	3.833282375	-4.594476746	0.680606949
C	3.226750275	-5.431443227	-0.338442612
C	1.804009030	-5.561865691	-0.302412924
C	1.038402452	-5.690985057	-1.507107673
C	3.385993256	2.128989777	-2.175273689
C	4.679834084	1.532243370	-2.394153540
C	5.322777478	0.854219503	-1.288067099
C	6.060779579	-0.324125173	-1.598304127
C	6.148846221	-1.423119484	-0.671240871
C	6.212164876	-2.641103635	-1.437183197
C	5.599316004	-3.840927771	-0.960685336
C	5.048508507	-4.742921300	-1.910309785
C	3.872162505	-5.523670306	-1.602512300
C	3.106182815	-5.663410861	-2.817162940
C	1.681449446	-5.701947996	-2.784635173
C	0.967634370	-5.104843938	-3.871942363
C	-0.315093708	-4.459535046	-3.670680733
C	-0.384695845	-3.344527588	-4.601387429
C	-1.100421382	-2.142452361	-4.281262188
C	-0.530812408	-0.917417269	-4.773498470
C	-0.599067136	0.318535156	-4.028925113
C	0.573071771	1.089929288	-4.330095641
C	1.191231799	1.905186882	-3.336553631
C	2.632536186	2.025466751	-3.391595039
C	3.463386489	1.387339291	-4.402724705
C	4.769898642	1.085663108	-3.784728111
C	5.449554652	-0.186346208	-4.065500573
C	6.079190024	-0.853582434	-2.950524397
C	6.168503749	-2.299164656	-2.830929214
C	5.527564194	-3.156860243	-3.774273288
C	5.011150359	-4.405726633	-3.308801709
C	3.803777074	-4.962284248	-3.863331627
C	3.093453023	-4.288487753	-4.897988474
C	1.665706878	-4.407792922	-4.913239671
C	0.838464232	-3.324003056	-5.366933539
C	1.410861730	-2.094019008	-5.816599995
C	0.692667527	-0.897785795	-5.537160432
C	1.382275040	0.342149771	-5.267564718
C	2.817862853	0.450622333	-5.313175949
C	3.539351854	-0.753623385	-5.647087169
C	4.823635323	-1.073060219	-5.028348842
C	4.871093444	-2.519002294	-4.880072069
C	3.664270570	-3.078029094	-5.420804089
C	2.838954720	-1.998459519	-5.884382474
N	2.037193013	-1.890961839	-1.652891151
Sc	2.690505955	-2.467635783	0.103594504
Sc	0.245782238	-2.462832902	-2.202686757
Dy	3.250141654	-0.657616193	-2.974517518

Table S24. The xyz coordinates (Å) of *c3*- $Dy_2ScN@C_{80}$ compound.

C	0.824850148	2.553041413	3.140938789
C	0.442794111	1.425848983	3.961962172
C	-0.985408063	1.100895202	4.032426316
C	1.379935143	0.311936251	3.983086039
C	2.074859626	2.611349360	2.403046114
C	3.685417213	1.257496688	1.149787176
C	2.944098944	1.493953247	2.370325386

C	2.591596152	0.362291826	3.173599498
C	0.919561379	-1.086973858	3.990978307
C	-0.501335086	-1.425978080	3.985171991
C	-1.457650304	-0.309458089	4.046491359
C	-0.863650993	-2.543729536	3.139354296
C	1.863321295	-1.842812578	3.174802531
C	2.227244482	-3.245056065	1.181570737
C	1.480118016	-2.970524000	2.373465074
C	0.102901323	-3.334231004	2.397759610
C	4.070421145	-0.056135098	0.710109334
C	3.601654242	-1.195932067	1.473795801
C	2.900466894	-0.959978004	2.709472968
C	3.274594988	-2.365359516	0.743588679
C	-2.105542880	-2.609319305	2.383210963
C	-2.981473821	-1.493299667	2.342772946
C	-2.628445058	-0.360174981	3.170568613
C	-3.718770647	-1.264405545	1.128377316
C	-1.905119775	-3.433509811	1.221333414
C	-1.897434341	-3.418940851	-1.236495185
C	-2.581356451	-3.153495745	-0.013204101
C	-3.535022513	-2.089004809	-0.036175619
C	1.581555727	-3.771153333	0.008746331
C	0.180989418	-4.056395655	0.002529578
C	-0.537908626	-3.873609822	1.227797415
C	-0.531680820	-3.866202657	-1.226980745
C	-4.043038174	0.059873133	0.680803264
C	-3.605523114	1.194595399	1.430196756
C	-2.910888326	0.960340839	2.675929879
C	-3.278066647	2.364558617	0.698382572
C	-4.090117789	0.061919719	-0.769490067
C	-3.068165968	1.003168815	-2.834229656
C	-3.697497509	1.224412985	-1.529380214
C	-3.286478950	2.365530386	-0.750925021
C	-2.113333019	-2.600508587	-2.417656532
C	-3.095883230	-1.538328517	-2.485278640
C	-3.783774795	-1.290126696	-1.229255302
C	-2.772591510	-0.368995753	-3.330010306
C	-2.235934187	3.246142111	1.154983495
C	-1.504337448	2.981044083	2.357223146
C	-1.893964301	1.847200697	3.166779355
C	-0.129502400	3.349366435	2.393891780
C	-1.586286024	3.793911200	-0.005473099
C	0.532870100	3.891336259	-1.219938055
C	-0.185948262	4.075856890	0.003215045
C	0.524265270	3.890039151	1.232410220
C	-1.925629736	1.848862924	-3.166231589
C	-1.494557781	2.980877507	-2.367002889
C	-2.227218638	3.252535145	-1.175912100
C	-0.110239854	3.341495604	-2.385972173
C	1.897122616	3.444662325	-1.211051452
C	2.575119983	3.168460147	0.009736516
C	1.888564821	3.435412491	1.235865753
C	3.544455263	2.092358758	-0.011276695
C	2.116123821	2.611887994	-2.375336970
C	2.678767166	0.372255190	-3.145857713
C	3.073105519	1.538008517	-2.390381767
C	3.906469349	1.327732568	-1.199950573
C	-0.941240528	1.053768908	-3.851037475
C	1.408081295	0.308117229	-3.847052141
C	0.470539125	1.382638959	-3.803628301
C	0.860259275	2.552202460	-3.097190325
C	-0.838895086	-2.547723618	-3.117793032
C	0.951774234	-1.053047404	-3.848033566
C	-0.450951818	-1.379040844	-3.833328217
C	-1.427864257	-0.305445725	-3.926650023
C	2.234993869	-3.228343617	-1.151226431
C	1.916014148	-1.838633524	-3.122177295
C	1.504582067	-2.965626925	-2.347821187
C	0.121963065	-3.336208745	-2.390134948

C	4.257390344	-0.048180304	-0.743008764
C	3.003330628	-0.975406515	-2.697533976
C	3.754168174	-1.219765744	-1.480266976
C	3.296880738	-2.350195727	-0.710716339
N	0.262543645	0.012739085	-0.094451664
Sc	2.053693580	0.166174813	-0.719185307
Dy	-0.169852894	-0.005872378	1.978588785
Dy	-1.432686668	-0.086736055	-1.350362199

Table S25. The xyz coordinates (Å) of *csa*- $Dy_2ScN@C_{80}$ compound.

C	-0.454948980	-0.997256542	1.212076365
C	0.747757163	-0.440985803	1.770535622
C	1.588533709	-1.525450290	2.225321953
C	0.878818520	-2.755784383	1.929735901
C	-0.385570095	-2.434179902	1.308858758
C	-1.008972791	-3.297567628	0.333746871
C	-1.832941403	-2.702705728	-0.722645043
C	-1.788376821	-1.250184480	-0.845770647
C	-1.118321988	-0.379004037	0.106797427
C	-0.590708569	0.858388038	-0.363494108
C	0.608292504	1.425152751	0.202390269
C	1.315644485	0.758712486	1.245051850
C	2.744287860	0.868064701	1.269759597
C	3.583566093	-0.208916545	1.757329661
C	3.030966297	-1.438093609	2.284723010
C	3.807976427	-2.671844577	2.082258730
C	3.053604523	-3.848346974	1.641217304
C	1.601235530	-3.925954482	1.584099083
C	0.987817023	-4.809392602	0.640456543
C	-0.289348029	-4.500867512	0.021935903
C	-0.292899361	-5.061032416	-1.323825574
C	-1.042502525	-4.454210213	-2.410891221
C	-1.881135773	-3.276577100	-2.094097497
C	-1.833181902	-2.127581961	-3.018603854
C	-1.802276936	-0.903711198	-2.243918562
C	-1.184627791	0.319487152	-2.724120924
C	-0.626110995	1.206430037	-1.763129135
C	0.549506214	1.987541408	-2.062096136
C	1.315270554	2.107978945	-0.849890124
C	2.737700393	2.135848159	-0.882461204
C	3.442878925	1.549137653	0.216984099
C	4.709911199	0.905049136	0.020528872
C	4.812476125	-0.187304867	0.970383486
C	5.539191252	-1.392545842	0.666599640
C	5.059313370	-2.643715914	1.267462830
C	5.015996445	-3.803808338	0.382336640
C	3.813535651	-4.548305799	0.640825460
C	3.199498171	-5.399544969	-0.350828138
C	1.775059385	-5.540620826	-0.311852947
C	1.007160250	-5.685277959	-1.517312221
C	3.378048264	2.120903574	-2.171010118
C	4.656517702	1.502580927	-2.378322348
C	5.304679183	0.835370510	-1.279374687
C	6.043299844	-0.337549086	-1.589156966
C	6.132581744	-1.435690007	-0.652099866
C	6.212886904	-2.649988939	-1.430480911
C	5.602470588	-3.845962664	-0.946439066
C	5.033685793	-4.730731955	-1.903105814
C	3.851285009	-5.505128752	-1.607495828
C	3.090867516	-5.650735639	-2.820642172
C	1.662578098	-5.689531179	-2.790558396
C	0.950881195	-5.095723851	-3.877506387
C	-0.336782191	-4.454210855	-3.676076776
C	-0.389770855	-3.338649467	-4.607397426
C	-1.086432079	-2.140116541	-4.271518366
C	-0.535263704	-0.914806808	-4.782705508
C	-0.594065559	0.306371561	-4.026210149

C	0.582272908	1.079480613	-4.325420221
C	1.193713016	1.897168981	-3.333477006
C	2.628303826	2.023272911	-3.391025334
C	3.452873879	1.375097023	-4.403172985
C	4.739143867	1.042688856	-3.760980902
C	5.452770026	-0.190868756	-4.071506236
C	6.074078872	-0.859376763	-2.942105192
C	6.174582659	-2.301888938	-2.823764122
C	5.527410147	-3.147016044	-3.773025374
C	4.996786178	-4.385237101	-3.304694143
C	3.792111590	-4.945095948	-3.862723650
C	3.081782273	-4.268612512	-4.890926581
C	1.656377572	-4.397694845	-4.916926273
C	0.832201480	-3.315346981	-5.375900265
C	1.409712540	-2.082530236	-5.822431926
C	0.688424339	-0.891771493	-5.548229975
C	1.395399003	0.347734741	-5.279780466
C	2.833655118	0.486828304	-5.381296735
C	3.585976503	-0.714642017	-5.760826305
C	4.879067607	-1.049313351	-5.113038860
C	4.878151415	-2.495162304	-4.890517052
C	3.656056919	-3.047532687	-5.402963193
C	2.849199181	-1.968404771	-5.902778468
N	2.358169895	-1.792521705	-1.597491157
Sc	3.315037529	-1.992847053	0.035340628
Dy	3.189482477	-0.902657152	-3.326115721
Dy	0.378063941	-2.509961850	-1.788563403

Table S26. The xyz coordinates (Å) of *c3*- $Dy_3N@C_{80}$ compound.

C	2.611859124	-1.986158076	2.496413011
C	0.414133788	3.255015391	2.496413011
C	-3.025992912	-1.268857314	2.496413011
C	3.545092571	-1.875730678	1.394885188
C	-0.148115868	4.008005564	1.394885188
C	-3.396976703	-2.132274886	1.394885188
C	4.128289289	-0.547194676	1.136545430
C	-1.590260155	3.848800737	1.136545430
C	-2.538029135	-3.301606061	1.136545430
C	3.250171306	-2.715125968	0.238380092
C	0.726282410	4.172293902	0.238380092
C	-3.976453716	-1.457167934	0.238380092
C	1.474150775	-2.893976073	2.495674038
C	1.769181410	2.723640057	2.495674038
C	-3.243332185	0.170336017	2.495674038
C	3.518739403	1.740467111	1.078945462
C	-3.266658434	2.177084156	1.078945462
C	-0.252080969	-3.917551267	1.078945462
C	1.148501823	-3.650041706	1.343735983
C	2.586777931	2.819652608	1.343735983
C	-3.735279754	0.830389098	1.343735983
C	2.052069350	-3.551743624	0.235076807
C	2.049865530	3.553015999	0.235076807
C	-4.101934881	-0.001272376	0.235076807
C	3.447608154	-2.229464866	-1.141660775
C	0.206969134	4.100448677	-1.141660775
C	-3.654577288	-1.870983811	-1.141660775
C	3.957228626	-0.882024383	-1.418043338
C	-1.214758791	3.868072710	-1.418043338
C	-2.742469835	-2.986048327	-1.418043338
C	3.291517419	-0.193449289	-2.509019733
C	-1.478226711	2.947262347	-2.509019733
C	-1.813290708	-2.753813058	-2.509019733
C	2.347754757	-2.781705236	-1.931290551
C	1.235150022	3.424067880	-1.931290551
C	-3.582904779	-0.642362643	-1.931290551
C	1.862606144	1.497310944	-3.338689455
C	-2.228012388	0.864408766	-3.338689455

C	0.365406243	-2.361719711	-3.338689455
C	1.742869405	-2.093198073	-3.035222401
C	0.941328004	2.555968217	-3.035222401
C	-2.684197409	-0.462770144	-3.035222401
C	2.258712731	-0.799091726	-3.333399356
C	-0.437322631	2.355648468	-3.333399356
C	-1.821390100	-1.556556742	-3.333399356
C	3.912939422	1.358145001	-0.251195560
C	-3.132657784	2.709632443	-0.251195560
C	-0.780281638	-4.067777444	-0.251195560
C	3.262725972	2.023538328	-1.360035015
C	-3.383798583	1.813834413	-1.360035015
C	0.121072611	-3.837372741	-1.360035015
C	1.525365425	-3.617506647	-1.095717756
C	2.370169943	3.129758532	-1.095717756
C	-3.895535367	0.487748116	-1.095717756
C	3.012896223	1.238535649	-2.513633951
C	-2.579051447	1.989976843	-2.513633951
C	-0.433844776	-3.228512492	-2.513633951
C	1.395138641	0.235192768	-3.838863862
C	-0.901252233	1.090629121	-3.838863862
C	-0.493886408	-1.325821889	-3.838863862
C	0.000000000	0.000000000	-4.055617885
C	4.337978070	-0.049573763	-0.256143967
C	-2.126056897	3.781586092	-0.256143967
C	-2.211921173	-3.732012329	-0.256143967
C	1.814066768	1.548101994	3.323854785
C	-2.247729039	0.796976908	3.323854785
C	0.433662271	-2.345078902	3.323854785
C	2.689594160	0.450472760	3.028733956
C	-1.734917934	2.104020488	3.028733956
C	-0.954676226	-2.554493248	3.028733956
C	3.611036677	0.578065844	1.919693245
C	-2.306138044	2.838216574	1.919693245
C	-1.304898632	-3.416282418	1.919693245
C	2.232976275	-0.862985422	3.332991156
C	-0.369120839	2.365306891	3.332991156
C	-1.863855436	-1.502321469	3.332991156
C	0.903919896	-1.086930843	3.836212233
C	0.489349774	1.326283014	3.836212233
C	-1.393269670	-0.239352171	3.836212233
C	0.000000000	0.000000000	4.051484965
N	0.000000000	0.000000000	0.298330093
Dy	1.976339342	-0.521533107	-0.000580621
Dy	-0.536508751	1.972326630	-0.000580621
Dy	-1.439830591	-1.450793523	-0.000580621

Table S27. The xyz coordinates (Å) of *cs*- $Dy_3N@C_{80}$ compound.

C	-1.896505855	-2.730034323	2.369354441
C	-1.896505855	-2.730034323	-2.369354441
C	-2.756521882	-2.794539435	1.217108562
C	-2.756521882	-2.794539435	-1.217108562
C	-3.646097971	-1.640110773	1.218730285
C	-3.646097971	-1.640110773	-1.218730285
C	-3.268080865	-0.849030360	2.387286445
C	-3.268080865	-0.849030360	-2.387286445
C	-2.203708679	-1.533851067	3.095798164
C	-2.203708679	-1.533851067	-3.095798164
C	-1.186570601	-0.829791794	3.826050129
C	-1.186570601	-0.829791794	-3.826050129
C	0.152430238	-1.393057273	3.925145254
C	0.152430238	-1.393057273	-3.925145254
C	0.446994496	-2.561164267	3.134525641
C	0.446994496	-2.561164267	-3.134525641
C	-0.557960268	-3.241108375	2.343951014
C	-0.557960268	-3.241108375	-2.343951014
C	-0.130460046	-3.904919373	1.162697331

C	-0.130460046	-3.904919373	-1.162697331
C	-0.978560189	-3.971007312	0.000000000
C	-2.276828384	-3.372449531	0.000000000
C	-4.183496738	-1.043937030	0.000000000
C	-4.384219322	0.432403861	0.000000000
C	-3.923857633	1.209487555	1.184139762
C	-3.923857633	1.209487555	-1.184139762
C	-3.349835723	0.579005044	2.374603566
C	-3.349835723	0.579005044	-2.374603566
C	-2.343268704	1.275251587	3.139598840
C	-2.343268704	1.275251587	-3.139598840
C	-1.297676700	0.583230727	3.861429233
C	-1.297676700	0.583230727	-3.861429233
C	-0.099654183	1.407951800	3.887303870
C	-0.099654183	1.407951800	-3.887303870
C	1.247713545	0.870127795	3.998618136
C	1.247713545	0.870127795	-3.998618136
C	1.380877745	-0.600847970	4.090502113
C	1.380877745	-0.600847970	-4.090502113
C	2.450460424	-1.315706932	3.323916634
C	2.450460424	-1.315706932	-3.323916634
C	1.830731410	-2.507502053	2.741503876
C	1.830731410	-2.507502053	-2.741503876
C	2.213858155	-3.092183912	1.466318491
C	2.213858155	-3.092183912	-1.466318491
C	1.239205697	-3.819560495	0.722609810
C	1.239205697	-3.819560495	-0.722609810
C	-3.268568128	2.422574448	0.719687868
C	-3.268568128	2.422574448	-0.719687868
C	-2.214957156	3.100435387	1.458385094
C	-2.214957156	3.100435387	-1.458385094
C	-1.796246419	2.530759684	2.701897770
C	-1.796246419	2.530759684	-2.701897770
C	-0.430594003	2.614134533	3.148823756
C	-0.430594003	2.614134533	-3.148823756
C	-1.247440840	3.837116860	0.721378454
C	-1.247440840	3.837116860	-0.721378454
C	0.120746500	3.928996922	1.168237069
C	0.120746500	3.928996922	-1.168237069
C	0.554325859	3.281971360	2.363889189
C	0.554325859	3.281971360	-2.363889189
C	1.889771762	2.750287021	2.412056908
C	1.889771762	2.750287021	-2.412056908
C	2.249520567	1.582001810	3.207668189
C	2.249520567	1.582001810	-3.207668189
C	3.302103651	0.871667137	2.461419385
C	3.302103651	0.871667137	-2.461419385
C	3.392444643	-0.576803174	2.463382358
C	3.392444643	-0.576803174	-2.463382358
C	3.761650647	-1.166857131	1.186969732
C	3.761650647	-1.166857131	-1.186969732
C	3.231850324	-2.428552158	0.720658832
C	3.231850324	-2.428552158	-0.720658832
C	0.960825743	3.968033141	0.000000000
C	2.243941421	3.357989352	0.000000000
C	2.708481757	2.782806162	1.232669101
C	2.708481757	2.782806162	-1.232669101
C	3.560313493	1.630223566	1.242339720
C	3.560313493	1.630223566	-1.242339720
C	3.949375050	1.019328429	0.000000000
C	4.074940396	-0.392534300	0.000000000
N	0.025342958	0.391697797	0.000000000
Dy	-2.032234556	0.113921725	0.000000000
Dy	1.016897737	-0.110373707	1.757215754
Dy	1.016897737	-0.110373707	-1.757215754

Table S28. The xyz coordinates (Å) of *opt*- $Dy_3N@C_{80}$ compound.

C	-0.150978413	-3.880919078	2.229419324
C	-0.518140517	-3.665815712	0.854095689
C	-1.731528954	-2.873406823	0.811049436
C	-2.085908079	-2.597450438	2.195747456
C	-1.120832306	-3.222003915	3.061787206
C	-0.746936274	-2.639216894	4.318237002
C	0.602649628	-2.799906754	4.743142614
C	1.576270053	-3.456890829	3.910577299
C	1.221062453	-3.964814300	2.622439257
C	2.211560920	-3.918080473	1.592437267
C	1.842952309	-3.698160309	0.219880292
C	0.470039087	-3.532991868	-0.162897564
C	0.153155590	-2.650550885	-1.268318792
C	-1.118308714	-1.938368951	-1.423786325
C	-2.063176516	-1.996991005	-0.291838425
C	-2.737267503	-0.763271654	0.090789588
C	-3.013168623	-0.471796204	1.495531749
C	-2.683595844	-1.366780080	2.563812144
C	-2.358577797	-0.810527200	3.865213316
C	-1.433465861	-1.444445400	4.767604128
C	-0.849839333	-0.435961290	5.656033047
C	0.585626404	-0.568743669	5.968701386
C	1.263807429	-1.760527000	5.506912327
C	2.662524438	-1.788425964	5.105283104
C	2.852528818	-2.832491525	4.133658313
C	3.815604038	-2.712736979	3.077344391
C	3.490007444	-3.296755848	1.819949019
C	3.941124395	-2.686146214	0.584859124
C	2.896883902	-2.937216870	-0.397723345
C	2.595858300	-1.983810212	-1.401614383
C	1.215956363	-1.863987726	-1.837858318
C	0.652524602	-0.624756582	-2.304807653
C	-0.804252617	-0.639632317	-2.096718865
C	-1.436397460	0.617133329	-1.635541544
C	-2.428145041	0.518528567	-0.563190011
C	-2.520626874	1.547869874	0.470096161
C	-2.913976679	0.942173881	1.714670126
C	-2.538751533	1.496092082	2.995181508
C	-2.331451530	0.605597145	4.121621846
C	-1.429656172	0.882055252	5.251105986
C	-0.569219183	2.083062170	5.154686423
C	0.834219452	1.939596481	5.546047915
C	1.396696555	0.640830115	5.949071682
C	2.775673331	0.603396043	5.469261562
C	3.423895419	-0.596003507	5.030891192
C	4.446741965	-0.501764400	4.004382453
C	4.687722298	-1.554990003	3.052483431
C	5.281768819	-0.989128075	1.837670032
C	4.819798646	-1.536946694	0.547547138
C	4.541550944	-0.594047880	-0.527408602
C	3.438776614	-0.828014774	-1.457165934
C	2.871208955	0.430709130	-1.845769178
C	1.484820829	0.561534663	-2.230214792
C	0.830478709	1.778776566	-1.911852698
C	-0.584702505	1.789342385	-1.561733497
C	-0.743248117	2.839871501	-0.569840077
C	-1.690505288	2.716364806	0.487206718
C	-1.372904990	3.306658003	1.756524586
C	-1.795752973	2.703973281	2.992612154
C	-0.789843529	2.956752566	4.017195047
C	0.267073120	3.710177361	3.362546169
C	1.632242926	3.537256587	3.733291067
C	1.891374303	2.647441851	4.826735155
C	3.084236921	1.842541028	4.816006156
C	4.064185582	1.928495801	3.757720946
C	4.822011393	0.745865681	3.392664890
C	5.375976568	0.488230424	2.054293208
C	5.005402830	1.430682885	0.974801138
C	4.627902015	0.859886616	-0.318881410

C	3.569394509	1.462518479	-1.125807561
C	2.865151022	2.647098102	-0.731859039
C	1.508271423	2.808794624	-1.170798423
C	0.535174943	3.456400440	-0.336654522
C	0.894399533	3.963981230	0.952668766
C	-0.095080448	3.919921378	1.986227870
C	2.269064355	3.880840717	1.342554014
C	2.635020056	3.674086748	2.715646388
C	3.835079961	2.880388061	2.731805543
C	4.243563392	2.603219794	1.360450485
C	3.246045581	3.231120699	0.510659524
N	1.045292998	0.331768564	1.715534977
Dy	3.077504390	0.005893385	1.508581004
Dy	0.284716408	0.354169222	3.640895649
Dy	-0.191836104	-0.389362455	0.222107382

VIII. The directions of the main magnetic axes of the ground KD of Dy ions

Compound		xyz coordinates (Å) of two points linking the magnetic axes
$DySc_2N@C_{80}$	*c3*	5.56414794 -0.03557966 5.89693322 -8.23775206 -0.11047966 -8.57724678
	csa	8.261456804 -0.488746247 0.00002 -11.6677032 1.193033753 -0.00002
	csb	-2.128068346 -6.227746193 3.353872482 8.628351654 4.912513807 -9.302907518
$Dy_2ScN@C_{80}$	*c3*	-1.75130289 -0.06093238 11.85259878 1.41159711 0.04918762 -7.89542122 6.69779333 0.29720394 4.4589278 -9.56316667 -0.47067606 -7.1596522
	csa	-0.95480752 -5.33482715 4.62251428 7.33377248 3.52951285 -11.27474572 9.96362394 0.27731815 -1.1986334 -9.20749606 -5.29724185 -2.3784934
$Dy_3N@C_{80}$	*c3*	-7.538801 2.329307 1.154759 11.491479 -3.372373 -1.155921 1.752161 -7.693446 1.154759 -2.825179 11.638100 -1.155921 5.786639 5.364139 1.154759 -8.666301 -8.265727 -1.155921
	cs	-11.972035 -0.980868 0.043210 7.907565 1.208712 -0.043210 5.887328 -2.338844 10.201906 -3.853532 2.118096 -6.687474 5.887328 -2.338844 -10.201906 -3.853532 2.118096 6.687474
	opt	-6.743516 1.324033 2.853961 12.898524 -1.312247 0.163201 -3.151254 0.798199 13.021566 3.720686 -0.089861 -5.739774 6.058144 2.763098 7.363517 -6.441816 -3.541822 -6.919303
	X-ray	-3.47237700 9.09694800 11.8911270 13.81408300 6.36722800 2.20986700 4.12192500 14.68957000 7.94550300 11.15754500 -3.41325000 3.17186300 1.91007700 1.43322000 7.31934400 14.39317700 16.71506000 4.05784400