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Different roles of similar aromatic rings in half-sandwich lanthanide single-ion magnets unravelled by *ab initio* electronic structure calculations

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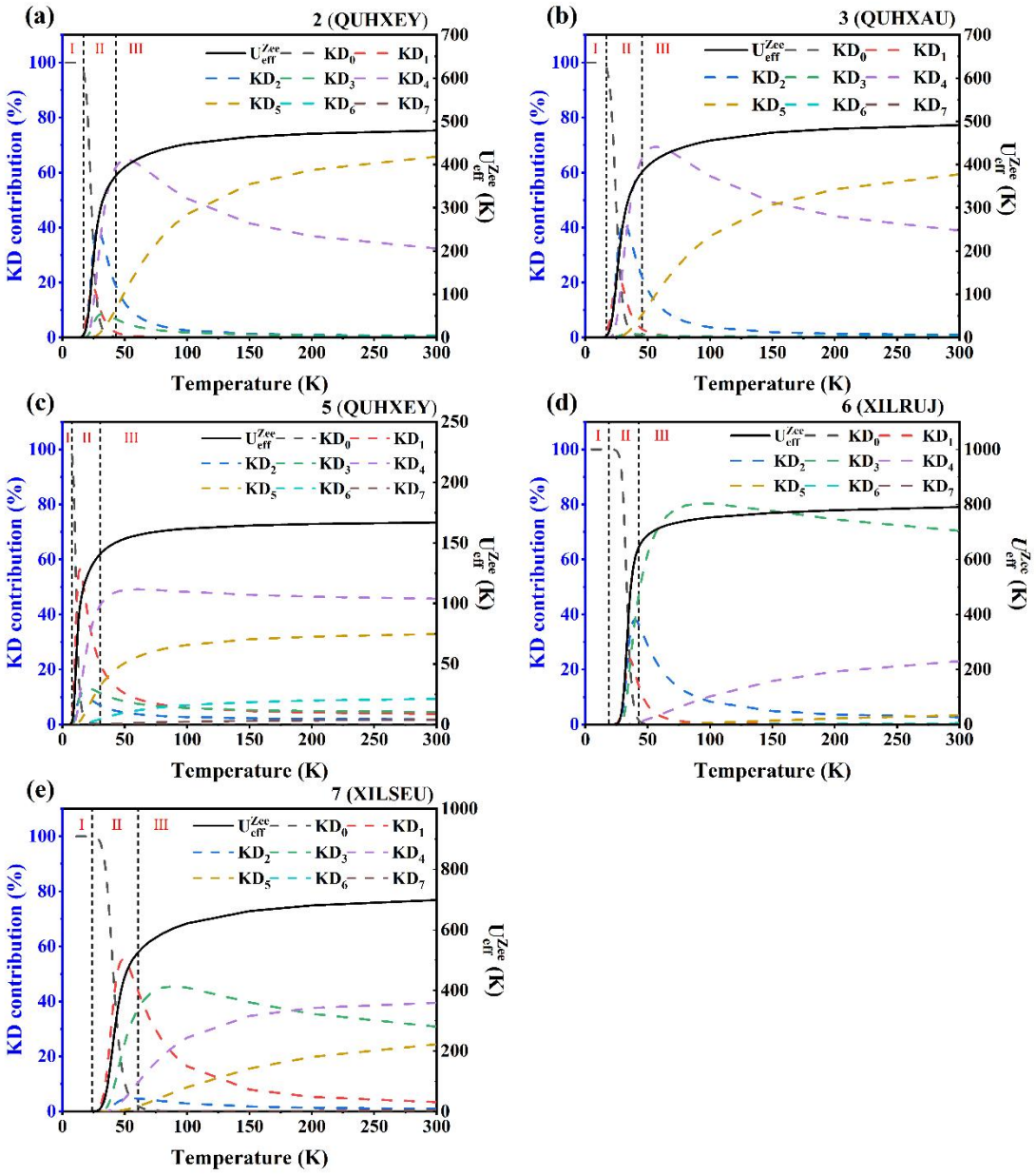

Figure S1 $U_{\mathrm{eff}}^{\mathrm{Zee}}$ and the contributions from various KDs of other Dy-SIMs

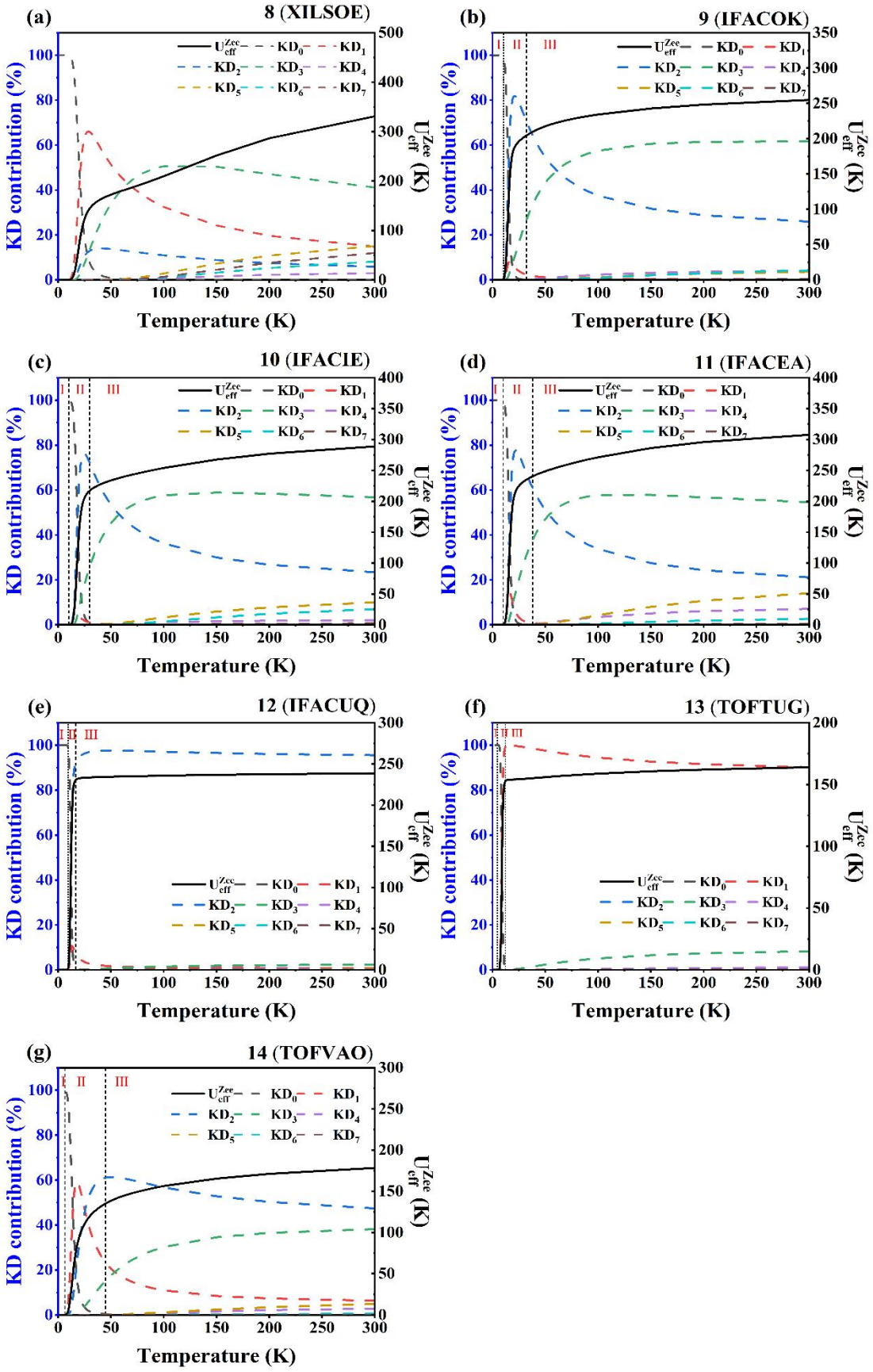

Figure S2 $U_{\mathrm{eff}}^{\mathrm{Zee}}$ and the contributions from various KDs of Er-SIMs

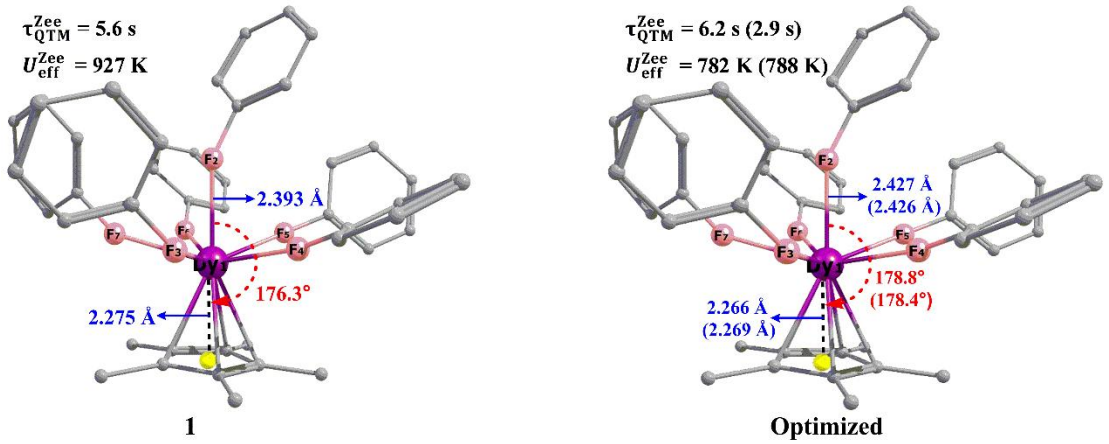

Figure S3 the comparison of geometric structural parameters and theoretical predictions values of τ_{QTM}^{Zee} and U_{eff}^{Zee} before and after optimization of **1** (results of solution-phase optimized structure are shown in parentheses)

Table S1 The atomic charge of **1** (**WUHYOR**) and **2** (**QUHXEY**) used for different model structure calculation

1 (**WUHYOR**)	Q[a]	Q1[b]	Q2[c]	**2** (**QUHXEY**)	Q[a]	Q1[b]	Q2[c]
Dy				Dy			
F	-0.4214		-0.4133	O	-0.7519		-0.7499
F	-0.4146		-0.4065	O	-0.7681		-0.7661
F	-0.4359		-0.4278	O	-0.7684		-0.7664
F	-0.3925		-0.3845	O	-0.7445		-0.7425
F	-0.4254		-0.4173	O	-0.5123		-0.5103
F	-0.4388		-0.4307	C	-0.4962	-0.5001	
C	-0.1573	-0.1782		C	-0.1109	-0.1145	
C	-0.1721	-0.1930		C	-0.1405	-0.1442	
C	-0.1532	-0.1741		C	-0.1537	-0.1573	
C	-0.1572	-0.1781		C	-0.1194	-0.1230	
C	-0.1734	-0.1943		Si	1.0457	1.0381	
C	-0.3687	-0.3902		C	-0.6728	-0.6767	
C	-0.3686	-0.3901		C	-0.6671	-0.6710	
C	-0.3716	-0.3931		C	-0.6743	-0.6782	
C	-0.3721	-0.3937		C	-0.3836	-0.3873	
C	-0.3712	-0.3927		C	-0.3836	-0.3873	
C	0.2708		0.2757	C	-0.3875	-0.3912	
C	-0.1618		-0.1566	C	-0.3841	-0.3878	
C	-0.0889		-0.0837	C	-0.1046		-0.1032
C	-0.1169		-0.1117	C	-0.1417		-0.1403
C	-0.0958		-0.0905	C	-0.1143		-0.1129
C	-0.1625		-0.1572	C	-0.1445		-0.1431
C	0.2750		0.2799	C	-0.1193		-0.1179
C	-0.1542		-0.1490	C	-0.0474		-0.0460
C	-0.1143		-0.1091	C	0.4071		0.4084
C	-0.1010		-0.0957	C	-0.4096		-0.4081
C	-0.1663		-0.1610	C	0.4075		0.4088
C	0.2780		0.2829	C	-0.0406		-0.0392
C	-0.1702		-0.1649	C	-0.1250		-0.1236
C	-0.0839		-0.0787	C	-0.1432		-0.1418
C	-0.1126		-0.1074	C	-0.1202		-0.1188
C	-0.1059		-0.1006	C	-0.1419		-0.1405
C	-0.1524		-0.1472	C	-0.1107		-0.1093
C	0.2548		0.2597	C	-0.1058		-0.1044
C	-0.1598		-0.1546	C	-0.1406		-0.1392
C	-0.1004		-0.0951	C	-0.1178		-0.1164
C	-0.1058		-0.1005	C	-0.1432		-0.1418
C	-0.1053		-0.1000	C	-0.1277		-0.1263
C	-0.0904		-0.0852	C	-0.0459		-0.0445

C	-0.1601		-0.1549	C	0.4072		0.4085
C	0.2807		0.2856	C	-0.4124		-0.4109
C	-0.1590		-0.1538	C	0.4026		0.4039
C	-0.0926		-0.0874	C	-0.0474		-0.0460
C	-0.1139		-0.1087	C	-0.1206		-0.1192
C	-0.0998		-0.0945	C	-0.1449		-0.1435
C	-0.1635		-0.1582	C	-0.1160		-0.1146
C	0.2854		0.2903	C	-0.1417		-0.1403
C	-0.1696		-0.1643	C	-0.1066		-0.1052
C	-0.0925		-0.0873	C	-0.0008		0.0005
C	-0.1214		-0.1162	C	-0.2346		-0.2331
C	-0.0914		-0.0862	C	-0.2369		-0.2354
C	-0.1557		-0.1505	C	-0.0018		-0.0005
H	0.1235	0.1205		H	0.1237	0.1232	
H	0.1201	0.1171		H	0.1195	0.1190	
H	0.1410	0.1381		H	0.1262	0.1257	
H	0.1218	0.1188		H	0.1247	0.1242	
H	0.1220	0.1190		H	0.1228	0.1223	
H	0.1402	0.1373		H	0.1308	0.1303	
H	0.1250	0.1220		H	0.1291	0.1286	
H	0.1197	0.1167		H	0.1211	0.1206	
H	0.1406	0.1377		H	0.1218	0.1213	
H	0.1202	0.1172		H	0.1256	0.1251	
H	0.1245	0.1215		H	0.1202	0.1197	
H	0.1419	0.1390		H	0.1209	0.1204	
H	0.1147	0.1117		H	0.1213	0.1208	
H	0.1263	0.1233		H	0.1212	0.1207	
H	0.1403	0.1374		H	0.1225	0.1220	
H	0.1438		0.1446	H	0.1295	0.1290	
H	0.1459		0.1467	H	0.1194	0.1189	
H	0.1453		0.1461	H	0.1215	0.1210	
H	0.1469		0.1477	H	0.1209	0.1204	
H	0.1488		0.1496	H	0.1251	0.1246	
H	0.1494		0.1502	H	0.1235	0.1230	
H	0.1461		0.1469	H	0.1463		0.1465
H	0.1453		0.1461	H	0.1303		0.1305
H	0.1427		0.1435	H	0.1301		0.1303
H	0.1407		0.1415	H	0.1285		0.1287
H	0.1498		0.1506	H	0.1288		0.1290
H	0.1505		0.1513	H	0.1213		0.1215
H	0.1475		0.1483	H	0.1298		0.1300
H	0.1449		0.1457	H	0.1290		0.1292
H	0.1499		0.1507	H	0.1296		0.1298
H	0.1440		0.1448	H	0.1302		0.1304

H	0.1483	0.1491	H	0.1452	0.1454
H	0.1463	0.1471	H	0.1465	0.1467
H	0.1445	0.1453	H	0.1303	0.1305
H	0.1403	0.1411	H	0.1300	0.1302
H	0.1471	0.1479	H	0.1290	0.1292
H	0.1465	0.1473	H	0.1300	0.1302
H	0.1455	0.1463	H	0.1206	0.1208
H	0.1442	0.1450	H	0.1288	0.1290
H	0.1457	0.1465	H	0.1289	0.1291
H	0.1501	0.1509	H	0.1300	0.1302
H	0.1450	0.1458	H	0.1300	0.1302
H	0.1472	0.1480	H	0.1470	0.1472
H	0.1476	0.1484	H	0.1368	0.1370
H	0.1544	0.1552	H	0.1113	0.1115
			H	0.1194	0.1196
			H	0.1155	0.1157
			H	0.1154	0.1156
			H	0.1199	0.1201
			H	0.1113	0.1115
			H	0.1347	0.1349

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S2 The atomic charge of **3 (QUHXAU)** and **4 (QUHWUN)** used for different model structure calculation

3 (QUHXAU)	Q[a]	Q1[b]	Q2[c]	**4 (QUHWUN)**	Q[a]	Q1[b]	Q2[c]
Dy				Dy			
O	-0.7557		-0.7556	O	-0.7635		-0.7601
O	-0.7542		-0.7541	O	-0.7585		-0.7551
O	-0.5359		-0.5357	O	-0.7527		-0.7494
O	-0.7499		-0.7498	O	-0.7559		-0.7526
O	-0.7584		-0.7583	O	-0.5172		-0.5140
C	-0.1930	-0.1690		C	-0.1499	-0.1584	
C	-0.1427	-0.1189		C	-0.1509	-0.1594	
C	-0.1795	-0.1556		C	-0.1503	-0.1588	
C	-0.1518	-0.1280		C	-0.1649	-0.1734	
C	-0.1485	-0.1247		C	-0.1615	-0.1700	
C	-0.1045		-0.1044	C	-0.3860	-0.3949	
C	-0.1376		-0.1375	C	-0.3862	-0.3951	
C	-0.1121		-0.1120	C	-0.3839	-0.3928	
C	-0.1384		-0.1383	C	-0.3874	-0.3963	

C	-0.1177		-0.1176	C	-0.3824	-0.3912	
C	-0.0427		-0.0426	C	0.4073		0.4095
C	0.4081		0.4082	C	-0.4111		-0.4086
C	-0.4089		-0.4088	C	0.4044		0.4065
C	0.4066		0.4067	C	-0.0444		-0.0421
C	-0.0413		-0.0412	C	-0.1063		-0.1040
C	-0.1107		-0.1106	C	-0.1414		-0.1391
C	-0.1349		-0.1348	C	-0.1189		-0.1166
C	-0.1152		-0.1151	C	-0.1453		-0.1430
C	-0.1346		-0.1345	C	-0.1245		-0.1221
C	-0.1190		-0.1189	C	-0.0420		-0.0397
C	0.0182		0.0183	C	-0.1227		-0.1203
C	-0.2185		-0.2184	C	-0.1455		-0.1432
C	-0.2237		-0.2236	C	-0.1189		-0.1166
C	0.0088		0.0089	C	-0.1438		-0.1415
C	-0.0961		-0.0961	C	-0.1083		-0.1060
C	-0.1391		-0.1390	C	0.4029		0.4050
C	-0.1125		-0.1124	C	-0.4145		-0.4121
C	-0.1411		-0.1410	C	0.4064		0.4086
C	-0.1178		-0.1177	C	-0.0490		-0.0467
C	-0.0485		-0.0484	C	-0.1040		-0.1017
C	0.4110		0.4111	C	-0.1452		-0.1429
C	-0.4070		-0.4069	C	-0.1170		-0.1147
C	0.4074		0.4075	C	-0.1472		-0.1449
C	-0.0457		-0.0456	C	-0.1241		-0.1217
C	-0.1190		-0.1189	C	-0.0468		-0.0445
C	-0.1395		-0.1394	C	-0.1222		-0.1198
C	-0.1136		-0.1135	C	-0.1450		-0.1427
C	-0.1349		-0.1348	C	-0.1175		-0.1152
C	-0.1079		-0.1078	C	-0.1438		-0.1415
C	0.0168	0.0400		C	-0.1085		-0.1062
C	-0.1417	-0.1179		C	0.0006		0.0029
C	-0.1433	-0.1195		C	-0.2379		-0.2356
C	-0.1479	-0.1241		C	-0.2328		-0.2305
C	-0.1355	-0.1117		C	-0.0003		0.0020
C	-0.1441	-0.1203		H	0.1192	0.1180	
C	-0.2241	-0.2000		H	0.1201	0.1189	
C	-0.1920	-0.1680		H	0.1221	0.1209	
C	-0.3596	-0.3350		H	0.1260	0.1248	
C	-0.2222	-0.1981		H	0.1235	0.1223	
C	-0.1920	-0.1680		H	0.1183	0.1171	
C	-0.3609	-0.3362		H	0.1186	0.1174	
C	-0.2158	-0.1917		H	0.1209	0.1197	
C	-0.1915	-0.1675		H	0.1208	0.1196	

C	-0.3583	-0.3337		H	0.1152	0.1140	
C	-0.2199	-0.1958		H	0.1195	0.1183	
C	-0.1913	-0.1673		H	0.1258	0.1246	
C	-0.3610	-0.3363		H	0.1189	0.1177	
H	0.1399		0.1399	H	0.1181	0.1169	
H	0.1265		0.1265	H	0.1214	0.1202	
H	0.1262		0.1262	H	0.1215		0.1218
H	0.1269		0.1269	H	0.1443		0.1447
H	0.1279		0.1279	H	0.1297		0.1300
H	0.1195		0.1195	H	0.1296		0.1299
H	0.1405		0.1405	H	0.1282		0.1285
H	0.1264		0.1264	H	0.1298		0.1301
H	0.1278		0.1278	H	0.1295		0.1298
H	0.1263		0.1263	H	0.1283		0.1286
H	0.1286		0.1286	H	0.1289		0.1292
H	0.1302		0.1302	H	0.1289		0.1292
H	0.1118		0.1118	H	0.1450		0.1454
H	0.1179		0.1179	H	0.1192		0.1195
H	0.1108		0.1108	H	0.1495		0.1499
H	0.1160		0.1160	H	0.1295		0.1298
H	0.1111		0.1111	H	0.1292		0.1295
H	0.1178		0.1178	H	0.1285		0.1288
H	0.1232		0.1232	H	0.1286		0.1289
H	0.1476		0.1476	H	0.1271		0.1274
H	0.1262		0.1262	H	0.1279		0.1282
H	0.1266		0.1266	H	0.1298		0.1301
H	0.1251		0.1251	H	0.1290		0.1293
H	0.1253		0.1253	H	0.1455		0.1459
H	0.1164		0.1164	H	0.1361		0.1365
H	0.1282		0.1282	H	0.1134		0.1137
H	0.1257		0.1257	H	0.1190		0.1193
H	0.1268		0.1268	H	0.1155		0.1158
H	0.1259		0.1259	H	0.1166		0.1169
H	0.1431		0.1431	H	0.1188		0.1191
H	0.1285	0.1319		H	0.1116		0.1119
H	0.1170	0.1204		H	0.1315		0.1318
H	0.1176	0.1210					
H	0.1207	0.1241					
H	0.1232	0.1266					
H	0.1125	0.1159					
H	0.1109	0.1143					
H	0.1010	0.1044					
H	0.1004	0.1038					
H	0.1089	0.1123					

H	0.1108	0.1142	
H	0.1113	0.1147	
H	0.1112	0.1146	
H	0.1027	0.1061	
H	0.1008	0.1042	
H	0.1044	0.1078	
H	0.1120	0.1154	
H	0.1099	0.1133	
H	0.1075	0.1109	
H	0.1063	0.1097	
H	0.1029	0.1063	
H	0.1031	0.1065	
H	0.1015	0.1049	
H	0.1084	0.1118	
H	0.1110	0.1144	
H	0.1089	0.1123	
H	0.1014	0.1048	
H	0.1111	0.1111	
H	0.1093	0.1092	
H	0.1009	0.1008	
H	0.1063	0.1062	
H	0.1112	0.1112	
H	0.1110	0.1110	

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S3 The atomic charge of **5** (**QOPZIG**) and **6** (**XILRUJ**) used for different model structure calculation

5 (**QOPZIG**)	Q[a]	Q1[b]	Q2[c]	**6** (**XILRUJ**)	Q[a]	Q1[b]	Q2[c]
Dy				Dy			
Cl	-0.5066		-0.5075	O	-0.9931		-0.9807
Cl	-0.4720		-0.4729	O	-0.5656		-0.5537
Cl	-0.4888		-0.4897	C	-0.3124	-0.3497	
Cl	-0.4935		-0.4943	C	-0.3108	-0.3481	
Cl	-0.4998		-0.5007	C	-0.3113	-0.3486	
Cl	-0.4894		-0.4902	C	-0.3252	-0.3626	
C	-0.0490	-0.0481		C	-0.2897	-0.3269	
C	-0.0292	-0.0284		C	-0.2892	-0.3264	
C	-0.0439	-0.0430		C	-0.3215	-0.3589	
C	-0.0435	-0.0426		C	-0.3044	-0.3417	

C	-0.0502	-0.0494		C	0.2071	0.2151
C	-0.0559	-0.0551		C	-0.0677	-0.0593
Cl	-0.4175		-0.4184	C	-0.1719	-0.1634
Cl	-0.3902		-0.3910	C	-0.1709	-0.1624
Cl	-0.4120		-0.4129	C	-0.2266	-0.2180
Cl	-0.3949		-0.3957	C	-0.0683	-0.0599
Cl	-0.4036		-0.4045	C	0.0483	0.0566
Cl	-0.4068		-0.4077	C	-0.0086	-0.0003
Al	0.7954		0.7948	C	0.0478	0.0561
Al	0.7953		0.7947	C	-0.2299	-0.2213
Al	0.7966		0.7960	C	-0.2353	-0.2267
C	-0.3695	-0.3686		C	-0.2269	-0.2183
C	-0.3687	-0.3678		C	-0.0809	-0.0725
C	-0.3715	-0.3706		C	-0.2290	-0.2204
C	-0.3686	-0.3677		C	-0.0811	-0.0727
C	-0.3663	-0.3654		C	-0.2315	-0.2229
C	-0.3685	-0.3676		C	-0.2310	-0.2224
H	0.1357	0.1358		C	-0.3669	-0.3581
H	0.1343	0.1344		C	0.0023	0.0106
H	0.1427	0.1428		C	-0.0762	-0.0678
H	0.1341	0.1342		C	-0.2295	-0.2209
H	0.1338	0.1339		C	-0.2304	-0.2218
H	0.1449	0.1450		C	-0.2372	-0.2286
H	0.1414	0.1415		C	-0.0758	-0.0674
H	0.1357	0.1358		C	-0.0813	-0.0729
H	0.1360	0.1361		C	-0.2331	-0.2245
H	0.1354	0.1355		C	-0.0809	-0.0725
H	0.1380	0.1381		C	-0.2293	-0.2207
H	0.1369	0.1370		C	0.0019	0.0102
H	0.1352	0.1353		C	-0.2222	-0.2136
H	0.1407	0.1408		C	-0.2238	-0.2152
H	0.1358	0.1359		H	0.1149	0.1161
H	0.1363	0.1364		H	0.1155	0.1167
H	0.1407	0.1408		H	0.0949	0.0961
H	0.1350	0.1351		H	0.1041	0.1053
				H	0.0975	0.0987
				H	0.0934	0.0946
				H	0.1008	0.1020
				H	0.0985	0.0997
				H	0.0974	0.0986
				H	0.1025	0.1037
				H	0.0964	0.0976
				H	0.0969	0.0981
				H	0.1007	0.1019

	H	0.0948		0.0960
	H	0.0902		0.0914
	H	0.0996		0.1008
	H	0.0991		0.1003
	H	0.0881		0.0893
	H	0.1224		0.1236
	H	0.1169		0.1181
	H	0.1200		0.1212
	H	0.1360		0.1372
	H	0.1224		0.1236
	H	0.0938		0.0950
	H	0.0993		0.1005
	H	0.0977		0.0989
	H	0.0975		0.0987
	H	0.0978		0.0990
	H	0.0981		0.0993
	H	0.1003		0.1015
	H	0.0953		0.0965
	H	0.0852		0.0865
	H	0.0977		0.0989
	H	0.0971		0.0983
	H	0.0890		0.0902
	H	0.0970		0.0982
	H	0.0974		0.0986
	H	0.1185		0.1197
	H	0.1229		0.1241
	H	0.1054	0.1001	
	H	0.0925	0.0871	
	H	0.1000	0.0947	
	H	0.0945	0.0891	
	H	0.1067	0.1014	
	H	0.0983	0.0929	
	H	0.1034	0.0981	
	H	0.1049	0.0996	
	H	0.1161		0.1173
	H	-0.8805		0.1207
	H	-0.8885		0.1127
	H	-0.8773		0.1239

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S4 The atomic charge of **7** (**XILSEU**) and **8** (**XILSOE**) used for different model structure calculation

7 (**XILSEU**)	Q[a]	Q1[b]	Q2[c]	**8** (**XILSOE**)	Q[a]	Q1[b]	Q2[c]
Dy				Er			
O	-0.9662		-0.9578	O	-0.5667		-0.5523
O	-0.5616		-0.5536	O	-0.5737		-0.5592
O	-0.5437		-0.5358	O	-0.5748		-0.5603
C	-0.3281	-0.3603		O	-0.5757		-0.5612
C	-0.3205	-0.3526		C	-0.3178	-0.3474	
C	-0.2950	-0.3269		C	-0.3012	-0.3307	
C	-0.3024	-0.3344		C	-0.2923	-0.3218	
C	-0.2848	-0.3168		C	-0.3217	-0.3514	
C	-0.2949	-0.3268		C	-0.3153	-0.3449	
C	-0.3168	-0.3489		C	-0.2972	-0.3267	
C	-0.3121	-0.3441		C	-0.3074	-0.3370	
C	-0.0946		-0.0890	C	-0.3141	-0.3437	
C	-0.0709		-0.0652	C	-0.2298		-0.2193
C	0.2320		0.2373	C	-0.0020		0.0082
C	0.0128		0.0184	C	0.0018		0.0119
C	0.0109		0.0165	C	-0.2259		-0.2154
C	-0.0645		-0.0588	C	-0.2231		-0.2126
C	-0.1558		-0.1501	C	0.0009		0.0110
C	-0.0998		-0.0942	C	-0.0089		0.0013
C	0.0153		0.0209	C	-0.0013		0.0089
C	-0.1571		-0.1514	C	-0.0098		0.0004
C	0.0152		0.0208	C	-0.0015		0.0087
C	0.0106		0.0162	C	-0.2255		-0.2150
C	-0.0135		-0.0079	C	-0.2230		-0.2125
C	-0.1445		-0.1388	C	-0.0051		0.0051
C	0.0049		0.0105	C	-0.2273		-0.2168
C	-0.1496		-0.1439	C	-0.2260		-0.2155
C	-0.1573		-0.1516	C	-0.2310		-0.2205
C	-0.1440		-0.1383	H	0.1138		0.1153
C	-0.1474		-0.1417	H	0.1275		0.1290
C	-0.1566		-0.1509	H	0.1162		0.1177
C	-0.2195		-0.2137	H	0.1208		0.1223
C	-0.1526		-0.1469	H	0.1221		0.1236
C	-0.3631		-0.3571	H	0.1263		0.1278
C	-0.1303		-0.1246	H	0.0919	0.0876	
C	-0.2228		-0.2170	H	0.1298		0.1313
C	0.0021		0.0077	H	0.1126		0.1141
C	-0.1361		-0.1304	H	0.1213		0.1228
C	0.0087		0.0143	H	0.1269		0.1284

C	-0.1316		-0.1259	H	0.1273		0.1288
C	-0.1605		-0.1548	H	0.1198		0.1213
C	-0.1441		-0.1384	H	0.0923	0.0880	
C	-0.1429		-0.1372	H	0.1131		0.1146
C	-0.1342		-0.1285	H	0.1243		0.1258
C	-0.1303		-0.1246	H	0.0921	0.0878	
C	-0.1424		-0.1367	H	0.1255		0.1270
C	-0.2242		-0.2184	H	0.1102		0.1117
C	-0.1487		-0.1430	H	0.1202		0.1217
C	-0.1365		-0.1308	H	0.1177		0.1192
C	-0.1403		-0.1346	H	0.0937	0.0895	
C	-0.1314		-0.1257	H	0.0914	0.0871	
C	-0.2201		-0.2143	H	0.1217		0.1232
H	0.1189		0.1198	H	0.1092		0.1107
H	0.1219		0.1228	H	0.0915	0.0872	
H	0.1366		0.1374	H	0.1132		0.1147
H	0.1187		0.1196	H	0.1295		0.1310
H	0.1234		0.1243	H	0.1210		0.1225
H	0.1225		0.1234	H	0.1264		0.1279
H	0.1293		0.1301	H	0.1161		0.1176
H	0.1102		0.1111	H	0.1203		0.1218
H	0.1232		0.1241	H	0.0923	0.0880	
H	0.1292		0.1300	H	0.0925	0.0882	
H	0.1262		0.1271	H	0.1262		0.1277
H	0.1266		0.1275	H	0.1157		0.1172
H	0.1324		0.1332	H	0.1260		0.1275
H	0.1147		0.1156	H	0.1148		0.1163
H	0.1190		0.1199	H	0.1147		0.1162
H	0.1181		0.1190	H	0.1278		0.1293
H	0.1242		0.1251				
H	0.1220		0.1229				
H	0.1199		0.1208				
H	0.1206		0.1215				
H	0.1241		0.1250				
H	0.1119		0.1128				
H	0.1226		0.1235				
H	0.1229		0.1238				
H	0.1254		0.1263				
H	0.1243		0.1252				
H	0.1214		0.1223				
H	0.1174		0.1183				
H	0.1243		0.1252				
H	0.1149		0.1158				
H	0.0970	0.0924					

H	0.1133	0.1088		
H	0.1219		0.1228	
H	0.0941	0.0895		
H	0.0891	0.0845		
H	0.1043	0.0998		
H	0.1223		0.1232	
H	0.1216		0.1225	
H	0.0834	0.0787		
H	0.1249		0.1258	
H	0.1217		0.1226	
H	0.0845	0.0787		
H	0.1224		0.1233	
H	0.1102		0.1111	
H	0.1227		0.1236	
H	0.1218		0.1227	
H	0.1241		0.1250	
H	0.0820	0.0773		
H	0.1228		0.1237	
H	0.1091		0.1100	
H	0.1198		0.1207	

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S5 The atomic charge of **9** (**IFACOK**) and **10** (**IFACIE**) used for different model structure calculation

9 (**IFACOK**)	Q[a]	Q1[b]	Q2[c]	**10** (**IFACIE**)	Q[a]	Q1[b]	Q2[c]
Er				Er	-1.0246		
I	-1.0138		-0.9160	I	-0.3918		-0.9334
O	-0.5652		-0.5497	N	-0.3905		-0.3793
O	-0.5800		-0.5644	N	-0.3455		-0.3780
C	-0.3160	-0.3426		C	-0.3540	-0.3711	
C	-0.3047	-0.3312		C	-0.3442	-0.3797	
C	-0.3101	-0.3367		C	-0.3368	-0.3698	
C	-0.3067	-0.3332		C	-0.3168	-0.3623	
C	-0.2924	-0.3189		C	-0.3279	-0.3423	
C	-0.3061	-0.3326		C	-0.3398	-0.3535	
C	-0.3138	-0.3404		C	-0.3276	-0.3654	
C	-0.3180	-0.3446		C	-0.0720	-0.3532	
C	0.0200		0.0308	C	0.0135		-0.0618
C	0.0213		0.0321	C	-0.1845		0.0236

C	-0.1981		-0.1869	C	0.0217		-0.1741
C	-0.1989		-0.1877	C	-0.1769		0.0317
C	-0.1949		-0.1837	C	-0.1809		-0.1665
C	-0.2035		-0.1923	C	-0.0751		-0.1705
C	0.0221		0.0329	C	0.0234		-0.0649
C	0.0172		0.0280	C	0.0031		0.0334
H	0.1111		0.1127	C	-0.1837		0.0132
H	0.1165		0.1181	C	0.1358		-0.1733
H	0.0839	0.0800		H	0.1405		0.1373
H	0.1155		0.1171	H	0.1343		0.1420
H	0.0833		0.0850	H	0.1472		0.1358
H	0.0923		0.0940	H	0.1373		0.1486
H	0.1039		0.1055	H	0.1352		0.1388
H	0.0855	0.0817		H	0.1113		0.1367
H	0.0807	0.0768		H	0.1356	0.1077	
H	0.0952	0.0914		H	0.1499		0.1371
H	0.0843	0.0804		H	0.1357		0.1513
H	0.1019	0.0981		H	0.1336		0.1372
H	0.0952	0.0914		H	0.1243		0.1351
H	0.0843	0.0804		H	0.1092	0.1207	
H	0.0953		0.0970	H	0.1193	0.1056	
H	0.1103		0.1119	H	0.1220	0.1157	
H	0.0899		0.0916	H	0.1139	0.1184	
H	0.1014		0.1030	H	0.1155	0.1104	
H	0.1170		0.1186	H	0.1103	0.1120	
H	0.0895		0.0912				
H	0.1063		0.1079				
H	0.0984		0.1000				
H	0.0793		0.0810				
H	0.1208		0.1224				

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S6 The atomic charge of **11** (**IFACEA**) and **12** (**IFACUQ**) used for different model structure calculation

11 (**IFACEA**)	Q[a]	Q1[b]	Q2[c]	**12** (**IFACUQ**)	Q[a]	Q1[b]	Q2[c]
Er				Er	-0.4355		
I	-1.0727		-0.9685	N	-0.0999		-0.4242
N	-0.3596		-0.3454	N	-0.0933		-0.0891
N	-0.3536		-0.3394	N	-0.4547		-0.0825

C	-0.3434	-0.3641	
C	-0.3370	-0.3578	
C	-0.3350	-0.3558	
C	-0.3155	-0.3362	
C	-0.3474	-0.3682	
C	-0.3604	-0.3813	
C	-0.3320	-0.3527	
C	-0.3632	-0.3840	
C	0.1967		0.2079
C	-0.3807		-0.3684
C	0.1896		0.2008
C	-0.3745		-0.3622
H	0.1586		0.1603
H	0.1646		0.1662
H	0.1614		0.1630
H	0.1221	0.1192	
H	0.1116	0.1087	
H	0.1596		0.1613
H	0.1581		0.1598
H	0.1634		0.1650
H	0.1130	0.1101	
H	0.1108	0.1079	
H	0.1179	0.1150	
H	0.1087	0.1058	
H	0.1165	0.1136	
H	0.1226	0.1197	

N	-0.0952		-0.4433
N	-0.4514		-0.0844
N	-0.3402		-0.4400
C	-0.3188	-0.3670	
C	-0.3302	-0.3455	
C	-0.3473	-0.3570	
C	-0.3257	-0.3741	
C	-0.3445	-0.3524	
C	-0.3320	-0.3713	
C	-0.3152	-0.3588	
C	0.0764	-0.3419	
B	0.1361		0.0839
C	-0.3012		0.1451
C	-0.3088		-0.2916
C	0.1273		-0.2992
C	-0.3752		0.1362
C	0.1337		-0.3655
C	0.1270		0.1427
C	-0.3715		0.1359
C	0.1313		-0.3618
C	-0.3751		0.1403
C	-0.3011		-0.3654
C	-0.3739		-0.2915
C	-0.3715		-0.3642
C	0.1225		-0.3618
C	-0.3708		0.1314
C	0.1132		-0.3611
H	0.1224	0.1094	
H	0.1122		0.1237
H	0.1221	0.1084	
H	0.1338		0.1234
H	0.1244		0.1352
H	0.1339		0.1257
H	0.1304		0.1353
H	0.1289		0.1317
H	0.1320		0.1302
H	0.1124		0.1333
H	0.1357	0.1086	
H	0.1260		0.1371
H	0.1357		0.1273
H	0.1210		0.1371
H	0.1109		0.1223
H	0.1336	0.1071	
H	0.1235		0.1349

	H	0.1341		0.1248
	H	0.1114		0.1355
	H	0.1137	0.1076	
	H	0.1319	0.1099	
	H	0.1292		0.1332
	H	0.1334		0.1305
	H	0.1119		0.1347
	H	0.1125	0.1081	
	H	0.1325	0.1087	
	H	0.1286		0.1338
	H	0.1319		0.1299
	H	-0.0446		0.1332
	H			

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S7 The atomic charge of **13** (**TOFTUG**) and **14** (**TOFVAO**) used for different model structure calculation

13 (**TOFTUG**)	Q[a]	Q1[b]	Q2[c]	**14** (**TOFVAO**)	Q[a]	Q1[b]	Q2[c]
Er				Er			
O	-1.1039		-1.0937	O	-0.9497		-0.9408
O	-1.1016		-1.0914	O	-0.5348		-0.5262
O	-1.1016		-1.0914	O	-0.5508		-0.5423
C	-0.3228	-0.3513		C	-0.3422	-0.3758	
C	-0.3349	-0.3635		C	-0.3260	-0.3595	
C	-0.3326	-0.3612		C	-0.3144	-0.3479	
C	-0.3155	-0.3440		C	-0.3303	-0.3638	
C	-0.3345	-0.3631		C	-0.3277	-0.3612	
C	-0.3155	-0.3440		C	-0.3293	-0.3628	
C	-0.3326	-0.3612		C	-0.3252	-0.3587	
C	-0.3349	-0.3635		C	-0.3292	-0.3627	
Co	0.8817		0.9108	C	0.2638		0.2695
P	1.3527		1.3679	C	-0.0862		-0.0801
P	1.3496		1.3648	C	-0.1265		-0.1204
P	1.3527		1.3679	C	-0.1976		-0.1915
O	-0.6630		-0.6534	C	-0.1321		-0.1260
O	-0.6643		-0.6547	C	-0.0901		-0.0840
O	-0.6675		-0.6578	C	-0.0357		-0.0297
O	-0.6675		-0.6578	C	0.0128		0.0188
O	-0.6630		-0.6534	C	-0.1747		-0.1685

O	-0.6643		-0.6547	C	-0.1338		-0.1277
C	-0.2513		-0.2444	C	-0.1788		-0.1727
C	-0.1827		-0.1758	C	0.0038		0.0098
C	-0.2289		-0.2220	C	-0.0624		-0.0564
C	-0.1202		-0.1134	C	-0.3710		-0.3647
C	-0.1168		-0.1100	C	-0.3750		-0.3687
C	-0.1173		-0.1105	C	-0.0602		-0.0542
C	-0.1173		-0.1105	C	-0.3700		-0.3637
C	-0.1827		-0.1758	C	-0.3789		-0.3726
C	-0.2289		-0.2220	C	-0.0309		-0.0249
C	-0.1202		-0.1134	C	0.0118		0.0178
C	-0.1168		-0.1100	C	-0.1781		-0.1720
H	0.1445		0.1454	C	-0.1416		-0.1354
H	0.1500		0.1509	C	-0.1853		-0.1792
H	0.1470		0.1479	C	-0.0007		0.0053
H	0.1283		0.1293	C	-0.0614		-0.0554
H	0.1229		0.1239	C	-0.3767		-0.3704
H	0.1126		0.1136	C	-0.3715		-0.3652
H	0.1251		0.1261	C	-0.0646		-0.0586
H	0.1118		0.1128	C	-0.3699		-0.3636
H	0.1129		0.1139	C	-0.3800		-0.3737
H	0.1277		0.1287	C	-0.0094		-0.0034
H	0.1185		0.1195	C	-0.2352		-0.2290
H	0.1119		0.1129	C	-0.2330		-0.2268
H	0.1114	0.1074		C	-0.0043		0.0017
H	0.1103	0.1063		C	0.0025		0.0085
H	0.1102	0.1062		C	-0.2320		-0.2258
H	0.1111	0.1071		C	-0.2330		-0.2268
H	0.1094	0.1054		C	0.0002		0.0062
H	0.1277		0.1287	H	0.1148	0.1101	
H	0.1185		0.1195	H	0.1183	0.1136	
H	0.1119		0.1129	H	0.1245	0.1199	
H	0.1500		0.1509	H	0.1264	0.1218	
H	0.1470		0.1479	H	0.1131	0.1084	
H	0.1103	0.1063		H	0.1103	0.1056	
H	0.1102	0.1062		H	0.1116	0.1069	
H	0.1111	0.1071		H	0.1109	0.1062	
H	0.1283		0.1293	H	0.1285		0.1294
H	0.1229		0.1239	H	0.1221		0.1230
H	0.1126		0.1136	H	0.1255		0.1264
H	0.1251		0.1261	H	0.1221		0.1230
H	0.1118		0.1128	H	0.1249		0.1258
H	0.1129		0.1139	H	0.1196		0.1205
				H	0.1128		0.1137

	H	0.1138	0.1147
	H	0.1122	0.1131
	H	0.1239	0.1248
	H	0.1214	0.1223
	H	0.1169	0.1178
	H	0.1122	0.1131
	H	0.1087	0.1096
	H	0.1217	0.1226
	H	0.1107	0.1116
	H	0.1150	0.1159
	H	0.1141	0.1150
	H	0.1079	0.1088
	H	0.1233	0.1242
	H	0.1227	0.1236
	H	0.1241	0.1250
	H	0.1197	0.1206
	H	0.1169	0.1178
	H	0.1182	0.1191
	H	0.1240	0.1249
	H	0.1094	0.1103
	H	0.1217	0.1226
	H	0.1162	0.1171
	H	0.1123	0.1132
	H	0.1110	0.1119
	H	0.1152	0.1161
	H	0.1137	0.1146
	H	0.1225	0.1234
	H	0.1160	0.1169
	H	0.1138	0.1147
	H	0.1156	0.1165
	H	0.1466	0.1475
	H	0.1149	0.1158
	H	0.1170	0.1179
	H	0.1255	0.1264
	H	0.1149	0.1158
	H	0.1192	0.1201
	H	0.1195	0.1204
	H	0.1212	0.1221
	H	0.1232	0.1241
	H	0.1388	0.1397
	H	0.1245	0.1254
	H	0.1115	0.1124
	H	0.1176	0.1185
	H	0.1212	0.1221

	H	0.1185	0.1194
	H	0.1315	0.1324

[a] Q means the atomic charge used to replace the corresponding atom in the calculation of the Q model structure. [b] Q1 means the atomic charge used to replace the corresponding atom in the calculation of the Q1 model structure. [c] Q2 means the atomic charge used to replace the corresponding atom in the calculation of the Q2 model structure.

Table S8 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **1** (**WUHYOR**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	5.660E-05	3.784E-03	5.272E-01	3.402E-01	5.152E-01	2.113E-01	1.540E+00	1.232E+00
g_Y	9.794E-05	4.579E-03	5.370E-01	6.389E-01	8.109E-01	1.098E+00	2.400E+00	8.574E+00
g_Z	19.990	17.144	14.451	11.788	9.148	6.569	3.784	12.497
E (K)	0	404	582	671	749	835	913	962
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	3.850E-04	3.303E-02	2.164E-01	2.271E+00	1.619E+00	1.080E+00	1.105E+00	8.846E-02
g_Y	4.927E-04	4.349E-02	4.643E-01	3.527E+00	6.746E+00	3.589E+00	2.596E+00	2.033E-01
g_Z	20.003	17.252	13.840	16.022	8.525	11.825	16.796	19.719
E (K)	0	65	105	117	127	139	146	157
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.141E+00	3.757E+00	1.643E-01	3.171E-01	2.114E-01	2.390E-01	1.290E-02	8.404E-04
g_Y	6.024E+00	3.883E+00	8.586E-01	3.631E-01	2.527E-01	2.571E-01	1.444E-02	9.037E-04
g_Z	14.885	5.051	6.775	9.394	12.038	14.673	17.324	19.977
E (K)	0	27	40	102	125	141	164	181
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.000E-10	3.743E-05	3.036E-03	2.668E-05	4.758E-02	4.043E-02	9.680E-01	1.271E+00
g_Y	9.400E-08	3.830E-05	3.044E-03	5.700E-03	5.358E-02	1.366E-01	1.172E+00	9.548E+00
g_Z	20.010	17.195	14.598	11.975	9.268	6.541	3.867	11.686
E (K)	0	422	586	669	762	878	988	1057

Table S9 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **2** (**QUHXEY**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.931E-03	3.499E-02	8.528E-02	3.488E-01	2.599E+00	4.925E+00	6.183E-01	2.985E-03
g_Y	2.364E-03	4.710E-02	5.175E-01	5.690E-01	3.210E+00	5.418E+00	6.812E-01	2.626E-02
g_Z	19.925	16.901	13.645	19.089	10.937	9.413	15.172	19.151
E (K)	0	165	265	315	397	521	705	1057
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.166E-03	1.007E-02	5.076E-02	1.970E-01	5.795E-01	1.880E+00	2.538E+00	1.936E-01
g_Y	1.976E-03	1.038E-02	6.308E-02	3.022E-01	1.142E+00	3.862E+00	5.946E+00	4.838E-01
g_Z	19.601	16.797	14.523	12.127	9.577	6.472	11.407	18.395
E (K)	0	94	211	323	418	489	534	597
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

g_X	1.370E-02	1.883E-01	2.618E-01	8.206E-01	2.043E+00	1.735E+00	9.951E-01	6.537E-03
g_Y	2.411E-02	2.728E-01	6.326E-01	1.859E+00	5.710E+00	4.706E+00	2.014E+00	1.422E-02
g_Z	19.669	16.144	13.929	12.253	9.299	12.679	12.882	18.308
E (K)	0	244	416	628	796	911	1008	1259
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.559E-03	6.305E-02	2.534E-01	3.261E+00	3.366E+00	1.907E+00	2.305E-03	5.813E-03
g_Y	8.634E-03	1.652E-01	3.156E-01	3.378E+00	6.669E+00	2.156E+00	1.791E-01	1.789E-02
g_Z	19.891	19.319	19.532	14.089	9.009	13.054	16.645	19.891
E (K)	0	99	135	173	230	301	397	609

Table S10 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **3** (**QUHXAU**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	2.889E-03	5.132E-02	1.668E-01	3.653E-02	2.507E+00	4.842E+00	6.815E-01	3.158E-03
g_Y	3.123E-03	6.826E-02	5.988E-01	3.389E-01	3.761E+00	5.146E+00	8.056E-01	1.794E-02
g_Z	19.886	16.691	13.694	19.022	10.546	9.448	15.061	19.131
E (K)	0	173	273	339	412	545	735	1102
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	2.112E-03	1.812E-02	9.957E-02	4.111E-01	1.280E+00	2.642E+00	2.547E+00	3.179E-01
g_Y	3.720E-03	1.882E-02	1.171E-01	5.818E-01	2.225E+00	6.124E+00	4.256E+00	8.522E-01
g_Z	19.535	16.645	14.372	11.908	9.252	6.201	13.899	18.861
E (K)	0	100	227	350	457	541	597	657
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.511E-02	2.105E-01	3.469E-01	9.852E-01	1.069E+00	1.103E+00	2.951E+00	1.763E-01
g_Y	2.728E-02	3.131E-01	7.368E-01	2.084E+00	4.502E+00	5.164E+00	5.863E+00	3.445E-01
g_Z	19.614	15.976	13.649	11.904	9.393	6.824	10.999	17.376
E (K)	0	267	467	730	959	1140	1239	1404
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.154E-03	1.024E-01	6.691E-01	2.011E+00	3.938E+00	1.743E+00	7.962E-02	6.205E-03
g_Y	5.608E-03	2.017E-01	8.495E-01	3.625E+00	7.062E+00	1.883E+00	2.081E-01	1.631E-02
g_Z	19.849	19.409	19.104	13.619	8.715	13.270	16.903	19.881
E (K)	0	85	132	171	232	305	417	633

Table S11 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **4** (**QUHWUN**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.271E-02	1.824E-01	1.729E+00	1.234E+00	3.157E+00	4.040E+00	4.476E-01	6.059E-03
g_Y	2.187E-02	2.831E-01	3.227E+00	4.648E+00	4.849E+00	4.540E+00	5.718E-01	2.718E-02
g_Z	19.752	16.809	14.605	10.728	10.242	10.579	15.710	19.511
E (K)	0	189	316	347	469	593	760	1192
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	3.850E-04	3.303E-02	2.164E-01	2.271E+00	1.619E+00	1.080E+00	1.105E+00	8.846E-02
g_Y	4.927E-04	4.349E-02	4.643E-01	3.527E+00	6.746E+00	3.589E+00	2.596E+00	2.033E-01
g_Z	20.003	17.252	13.840	16.022	8.525	11.825	16.796	19.719
E (K)	0	122	258	385	490	568	634	745
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	6.462E-03	1.027E-01	3.201E-01	7.632E-01	1.468E+00	1.021E+00	1.693E+00	1.146E-02
g_Y	1.051E-02	1.396E-01	4.743E-01	1.564E+00	4.219E+00	5.628E+00	4.569E+00	2.790E-02
g_Z	19.726	16.484	13.997	11.958	9.382	8.278	12.322	19.082
E (K)	0	287	499	716	900	1022	1089	1391
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.764E-03	7.711E-01	1.143E+00	2.141E+00	4.560E+00	1.195E+00	7.637E-02	4.394E-03
g_Y	4.669E-03	2.173E+00	1.623E+00	3.222E+00	6.274E+00	1.408E+00	1.184E-01	1.153E-02
g_Z	19.841	18.120	16.720	13.529	9.393	13.876	17.280	19.891
E (K)	0	100	117	194	249	328	472	686

Table S12 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **5** (**QOPZIG**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.832E-02	6.054E-01	7.900E-01	5.048E-02	2.176E+00	1.285E+00	2.832E+00	1.094E+00
g_Y	3.390E-02	2.364E+00	1.492E+00	2.598E+00	7.483E+00	6.482E+00	2.994E+00	1.770E+00
g_Z	19.681	16.649	15.443	14.501	8.922	8.557	13.078	16.683
E (K)	0	96	111	124	156	184	207	243
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	7.637E-04	2.842E-02	4.040E-01	2.698E+00	4.965E+00	1.399E+00	1.853E-01	1.128E-02
g_Y	1.109E-03	3.903E-02	5.098E-01	3.075E+00	5.133E+00	1.763E+00	2.753E-01	2.043E-02
g_Z	19.911	16.989	13.970	10.307	8.221	13.233	16.698	19.712
E (K)	0	61	108	143	170	195	223	252
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

g_X	1.273E+00	1.870E+00	3.306E-01	1.003E+00	1.105E-01	2.537E-01	3.630E-03	2.101E-03
g_Y	7.812E+00	3.636E+00	2.021E+00	1.233E+00	3.650E-01	2.584E-01	7.387E-03	2.875E-03
g_Z	13.367	4.018	6.746	9.374	12.054	14.675	17.339	19.971
E (K)	0	46	129	242	367	490	597	672
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.184E-06	9.048E-04	1.077E-02	9.805E-02	9.000E-01	4.180E+00	2.077E+00	2.365E-01
g_Y	4.757E-06	1.148E-03	1.111E-02	1.150E-01	1.044E+00	5.277E+00	4.509E+00	6.638E-01
g_Z	20.003	17.291	14.491	12.255	9.521	6.278	12.989	19.036
E (K)	0	254	353	420	481	548	614	708

Table S13 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **6** (**XILRUJ**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.066E-04	6.258E-02	8.269E-01	4.677E+00	6.962E-01	1.503E+00	6.665E-01	3.996E-02
g_Y	1.645E-04	1.229E-01	1.087E+00	5.145E+00	5.567E+00	2.693E+00	1.305E+00	7.612E-02
g_Z	19.897	16.803	18.768	10.074	9.539	14.245	15.948	19.597
E (K)	0	465	620	763	907	1058	1167	1372
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.851E-05	2.621E-03	1.004E-01	1.328E+00	4.945E+00	1.993E+00	2.568E-01	1.487E-02
g_Y	2.292E-05	3.334E-03	1.271E-01	1.541E+00	6.079E+00	3.102E+00	4.230E-01	2.736E-02
g_Z	19.974	17.054	14.170	11.062	6.937	12.292	16.562	19.760
E (K)	0	427	779	1055	1265	1459	1709	1992
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	7.440E-05	2.175E-03	1.126E-01	1.945E+00	4.336E+00	1.505E+00	2.346E-01	2.377E-02
g_Y	8.882E-05	2.758E-03	1.600E-01	2.286E+00	6.943E+00	2.270E+00	3.406E-01	4.550E-02
g_Z	19.978	17.037	14.270	11.290	7.278	13.066	16.706	19.711
E (K)	0	570	996	1289	1509	1755	2064	2347
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	7.315E-05	8.010E-03	3.328E-01	2.505E+00	3.890E+00	6.031E-01	3.442E-01	1.317E-01
g_Y	8.615E-05	9.970E-03	5.720E-01	3.777E+00	4.365E+00	1.978E+00	3.614E-01	1.735E-01
g_Z	19.854	16.770	13.437	15.639	9.988	14.781	19.117	19.421
E (K)	0	396	681	783	871	1002	1090	1169

Table S14 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **7** (**XILSEU**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.500E-03	4.798E-01	1.123E-01	1.928E+00	1.288E+00	1.755E+00	1.486E-01	1.326E-01
g_Y	6.796E-03	1.041E+00	7.443E-01	3.642E+00	4.983E+00	5.466E+00	1.160E+00	1.279E+00
g_Z	19.815	15.174	17.232	11.183	8.411	13.281	15.529	16.074
E (K)	0	425	501	604	719	815	1050	1074
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.101E-06	1.739E-04	4.185E-03	7.040E-02	1.596E+00	4.029E+00	9.212E-01	4.669E-02
g_Y	1.202E-06	1.976E-04	4.609E-03	7.783E-02	1.686E+00	6.929E+00	2.019E+00	9.572E-02
g_Z	19.997	17.191	14.541	11.952	8.885	8.031	15.503	19.689
E (K)	0	357	641	858	1030	994	1288	1483
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	3.969E-04	8.386E-03	4.022E-03	5.497E-01	8.642E-01	4.249E+00	1.314E+00	6.113E-02
g_Y	5.150E-04	1.056E-02	4.466E-03	7.747E-01	2.156E+00	4.579E+00	3.488E+00	1.318E-01
g_Z	19.981	17.377	14.528	12.300	9.154	9.003	14.773	19.630
E (K)	0	389	637	840	1035	1193	1328	1583
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.484E-04	6.943E-03	5.032E-01	1.422E+00	3.881E+00	4.261E-01	8.710E-02	2.390E-03
g_Y	1.834E-04	8.892E-03	7.321E-01	3.160E+00	4.682E+00	8.787E-01	1.708E-01	4.493E-03
g_Z	19.861	16.711	12.513	15.239	11.777	15.644	19.686	19.921
E (K)	0	378	622	702	749	816	979	1169

Table S15 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **8** (**XILSOE**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.147E-01	1.882E+00	1.627E+00	4.578E+00	7.716E-01	3.483E+00	2.443E+00	2.776E+00
g_Y	1.210E-01	3.924E+00	2.334E+00	5.714E+00	2.711E+00	6.396E+00	5.432E+00	7.346E+00
g_Z	17.564	11.853	12.398	6.310	8.016	7.164	8.351	8.613
E (K)	0	141	164	227	480	508	570	582
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	2.432E-01	1.660E+00	3.531E+00	3.633E+00	1.724E+00	5.341E-01	1.637E-01	4.009E-02
g_Y	7.322E-01	2.736E+00	4.794E+00	4.647E+00	2.697E+00	8.054E-01	2.140E-01	4.936E-02
g_Z	16.589	11.856	6.566	6.913	7.902	10.555	13.032	15.487
E (K)	0	49	37	75	119	169	215	251
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

g_X	3.953E-02	1.386E+00	1.588E+00	2.587E+00	9.231E-01	5.080E-01	2.778E-01	6.236E-01
g_Y	7.646E-02	2.577E+00	5.302E+00	3.841E+00	1.895E+00	2.622E+00	1.611E+00	1.584E+00
g_Z	17.353	14.115	10.115	9.599	13.405	12.940	14.574	16.036
E (K)	0	106	153	225	306	376	411	438
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.712E-01	5.125E+00	3.378E-01	7.977E-02	8.937E-02	1.521E-01	4.130E-01	6.016E-01
g_Y	3.000E-01	7.286E+00	6.351E-01	5.555E-01	3.728E-01	6.581E-01	8.419E-01	1.769E+00
g_Z	16.565	7.520	13.955	14.627	15.074	13.627	12.606	12.629
E (K)	0	74	108	154	281	302	338	369

Table S16 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **9** (**IFACOK**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.602E-03	2.376E-01	1.185E+00	2.809E+00	9.755E-01	9.465E-01	6.333E-01	2.554E-01
g_Y	7.362E-03	3.214E-01	4.039E+00	4.329E+00	1.677E+00	1.815E+00	2.290E+00	6.517E-01
g_Z	17.760	14.643	12.966	5.776	10.995	11.080	10.090	14.267
E (K)	0	143	188	254	333	405	469	506
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	3.739E-01	2.567E+00	1.364E+00	1.836E-01	2.547E-02	4.054E-03	1.668E-03	1.123E-04
g_Y	1.221E+00	6.585E+00	1.914E+00	2.273E-01	3.780E-02	6.502E-03	1.714E-03	1.327E-04
g_Z	16.487	8.448	5.771	8.370	10.786	13.110	15.507	17.899
E (K)	0	33	65	113	171	233	286	320
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.554E-01	1.246E+00	2.389E+00	2.965E+00	8.459E-01	4.380E-02	2.484E-02	1.212E-01
g_Y	5.328E-01	2.286E+00	3.674E+00	4.116E+00	1.146E+00	9.806E-02	2.468E-01	1.896E-01
g_Z	16.726	12.214	8.314	7.976	11.887	13.240	15.019	16.554
E (K)	0	57	103	156	224	342	421	435
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	4.320E-05	6.543E-04	1.133E+00	1.683E-01	2.341E+00	4.440E-01	1.783E-01	2.340E-01
g_Y	4.934E-05	8.201E-04	6.788E+00	2.063E-01	2.398E+00	8.058E-01	5.375E-01	3.575E-01
g_Z	17.951	15.532	12.153	12.412	4.675	7.140	10.568	12.762
E (K)	0	178	344	376	395	472	500	532

Table S17 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **10** (**IFACIE**) under different models

full	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	9.338E-03	8.234E-02	9.782E-01	3.276E+00	1.911E-01	1.169E+00	5.811E-01	4.160E-01
g_Y	1.564E-02	1.229E-01	4.083E+00	4.683E+00	1.489E+00	3.283E+00	3.260E+00	1.123E+00
g_Z	17.739	14.877	12.786	5.584	11.427	9.357	9.782	14.208
E (K)	0	126	203	267	359	436	502	543
Q	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	1.821E-01	2.067E+00	2.749E+00	4.921E-02	5.367E-02	5.209E-03	1.030E-03	3.890E-05
g_Y	4.912E-01	5.640E+00	3.446E+00	1.454E-01	6.393E-02	6.821E-03	1.049E-03	5.437E-05
g_Z	17.120	10.369	5.462	8.292	10.818	13.355	15.838	17.948
E (K)	0	41	71	115	169	224	277	336
Q1	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	8.117E-02	9.375E-01	3.173E+00	3.156E+00	7.273E-01	4.564E-03	2.520E-02	1.251E-01
g_Y	4.260E-01	1.158E+00	3.739E+00	4.324E+00	9.778E-01	1.310E-01	2.700E-01	2.125E-01
g_Z	16.190	12.280	9.109	8.275	12.564	13.304	15.263	16.510
E (K)	0	51	108	160	227	349	430	443
Q2	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	4.784E-05	2.003E-03	2.080E-01	9.192E-01	3.436E+00	1.569E-01	2.881E-01	1.849E-01
g_Y	5.716E-05	2.172E-03	4.350E-01	5.166E+00	4.004E+00	1.659E+00	1.598E+00	2.093E-01
g_Z	17.950	15.543	12.729	13.161	4.827	8.821	12.247	15.563
E (K)	0	180	400	430	484	539	549	619

Table S18 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **11** (**IFACEA**) under different models

full	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	1.490E-03	7.528E-02	1.057E+00	3.011E+00	1.797E+00	8.517E-01	3.691E-01	3.717E-01
g_Y	5.163E-03	1.225E-01	4.034E+00	5.192E+00	2.255E+00	3.990E+00	2.204E+00	8.197E-01
g_Z	17.692	14.902	13.390	6.884	10.634	7.766	11.189	12.670
E (K)	0	125	217	279	395	465	519	545
Q	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
g_X	1.838E-01	2.052E+00	2.894E+00	7.888E-03	5.590E-02	9.182E-03	7.243E-04	4.278E-05
g_Y	4.968E-01	5.533E+00	3.441E+00	9.907E-02	6.887E-02	1.020E-02	7.535E-04	5.470E-05
g_Z	17.120	10.443	5.488	8.326	10.872	13.400	15.796	17.953
E (K)	0	43	75	121	200	238	298	372
Q1	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7

g_X	1.439E-01	7.402E-01	4.110E+00	2.406E+00	8.492E-02	1.478E-01	8.712E-02	7.795E-04
g_Y	5.921E-01	1.315E+00	5.280E+00	2.740E+00	6.670E-01	3.625E-01	1.585E-01	2.008E-02
g_Z	16.188	12.302	7.898	9.404	12.313	13.366	16.084	17.329
E (K)	0	53	123	175	247	340	391	459
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.199E-05	4.916E-04	6.373E-01	1.080E+00	3.116E+00	1.126E-01	5.053E-01	1.508E-01
g_Y	1.673E-05	6.005E-04	1.340E+00	4.448E+00	4.241E+00	1.927E+00	9.993E-01	2.317E-01
g_Z	17.948	15.541	11.377	11.824	5.704	9.012	13.195	15.867
E (K)	0	179	401	413	474	532	553	629

Table S19 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **12** (**IFACUQ**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	2.325E-04	5.810E-01	1.773E+00	4.026E-01	2.677E-01	3.896E-01	4.415E-01	1.644E-01
g_Y	6.030E-04	6.045E-01	8.744E+00	7.665E-01	6.693E-01	6.432E-01	6.021E-01	2.525E-01
g_Z	17.716	14.407	9.075	3.655	9.194	10.000	10.323	12.540
E (K)	0	196	234	309	386	474	545	561
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.081E+00	3.155E+00	1.064E-01	2.098E-01	7.016E-03	9.588E-05	2.555E-04	1.029E-05
g_Y	6.269E+00	3.172E+00	3.251E-01	2.198E-01	7.291E-03	2.756E-04	2.625E-04	1.059E-05
g_Z	12.640	3.519	5.998	8.405	10.795	13.159	15.545	17.938
E (K)	0	21	58	113	182	258	331	386
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.029E+00	6.517E-02	4.258E-01	2.310E-01	1.530E-01	5.724E-02	2.056E-02	2.099E-02
g_Y	8.516E+00	1.774E+00	1.156E+00	5.124E-01	1.633E-01	6.489E-02	4.676E-02	1.199E-01
g_Z	10.419	3.582	5.831	9.082	13.142	12.991	15.903	15.939
E (K)	0	41	99	153	201	360	420	428
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	1.535E-05	2.299E-04	1.308E+00	8.633E-02	6.357E-01	2.929E-01	7.694E-02	1.023E-01
g_Y	2.023E-05	2.763E-04	8.790E+00	1.571E-01	7.862E-01	4.334E-01	3.283E-01	6.244E-01
g_Z	17.951	15.533	10.204	12.739	3.840	6.501	10.752	10.479
E (K)	0	174	322	362	370	442	475	495

Table S20 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **13** (**TOFTUG**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	5.107E-05	1.450E+00	1.538E-01	4.952E-01	4.637E-01	2.466E-01	9.493E-02	1.810E-01
g_Y	1.482E-03	8.529E+00	2.006E-01	1.585E+00	8.238E-01	3.611E-01	6.904E-01	4.740E-01
g_Z	17.549	10.254	13.869	3.775	7.338	13.219	10.125	11.906
E (K)	0	154	215	234	330	474	527	554
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	8.532E-01	3.275E+00	4.108E-01	4.630E-01	2.590E-03	1.562E-02	6.532E-03	3.433E-02
g_Y	3.698E+00	5.304E+00	5.946E-01	4.633E-01	1.345E-02	1.020E-01	6.744E-02	3.537E-02
g_Z	14.704	5.765	6.184	8.901	12.861	14.222	14.710	16.153
E (K)	0	24	60	110	162	244	250	281
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	8.071E-01	5.733E-01	9.121E-01	2.078E+00	2.067E-02	1.223E-02	5.272E-04	5.837E-03
g_Y	8.660E+00	1.572E+00	2.866E+00	2.081E+00	5.756E-02	1.684E-02	8.001E-03	1.721E-02
g_Z	9.992	3.883	5.375	8.508	13.034	13.042	15.820	15.385
E (K)	0	44	96	149	206	442	525	560
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	5.372E-05	6.844E-03	1.209E+00	1.704E+00	1.787E-01	3.491E-01	8.061E-02	9.626E-02
g_Y	6.609E-05	7.600E-03	7.550E+00	2.300E+00	2.373E-01	5.988E-01	7.842E-01	1.240E+00
g_Z	17.931	15.441	11.474	3.890	11.510	8.295	10.577	11.601
E (K)	0	235	367	428	450	553	607	618

Table S21 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of **14** (**TOFVAO**) under different models

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	5.123E-02	1.884E-01	2.724E+00	1.860E+00	6.369E-01	5.578E-01	6.909E-02	3.411E-02
g_Y	8.295E-02	1.761E+00	3.907E+00	3.864E+00	1.346E+00	2.235E+00	8.891E-01	1.198E-01
g_Z	17.479	14.683	10.779	8.641	9.617	11.742	13.527	17.222
E (K)	0	83	139	201	344	382	446	572
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	3.215E-02	1.129E+00	4.240E+00	3.490E-02	2.437E-01	2.397E-02	9.996E-04	6.152E-05
g_Y	6.613E-02	2.872E+00	4.737E+00	4.400E-01	2.703E-01	2.439E-02	1.344E-03	6.820E-05
g_Z	17.597	12.945	7.107	8.260	11.022	13.673	15.796	17.958
E (K)	0	82	126	180	255	343	448	641
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

g_X	2.072E-01	1.861E+00	3.138E+00	7.914E-01	1.029E-01	3.486E-01	2.753E-01	1.163E-03
g_Y	5.475E-01	3.567E+00	5.429E+00	2.049E+00	1.315E+00	1.106E+00	3.535E-01	6.219E-03
g_Z	16.935	11.798	6.276	8.692	10.772	14.424	15.377	17.886
E (K)	0	84	153	237	337	404	455	689
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	7.181E-04	4.403E-01	1.944E+00	2.686E+00	4.039E-01	5.040E-01	2.803E-01	6.267E-04
g_Y	1.044E-03	3.422E+00	2.220E+00	5.871E+00	1.411E+00	9.225E-01	4.916E-01	3.726E-03
g_Z	17.888	12.441	12.828	8.488	10.443	12.450	12.510	17.536
E (K)	0	181	191	285	337	360	425	623

Table S22 The decomposition of crystal field wave function of **1** (**WUHYOR**) in terms of $|m_J>$'s ($-15/2 \le m_J \le 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} *a*

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.95%	0.00%	0.01%	0.00%	0.00%	0.04%	0.00%	0.00%
|±13/2>	0.00%	99.52%	0.11%	0.01%	0.01%	0.01%	0.33%	0.00%
|±11/2>	0.01%	0.12%	98.21%	0.84%	0.00%	0.01%	0.03%	0.78%
|±9/2>	0.00%	0.01%	0.81%	97.78%	0.39%	0.00%	0.01%	1.01%
|±7/2>	0.00%	0.01%	0.00%	0.39%	98.42%	0.14%	0.98%	0.06%
|±5/2>	0.04%	0.01%	0.02%	0.00%	0.15%	99.65%	0.06%	0.07%
|±3/2>	0.00%	0.33%	0.02%	0.04%	1.00%	0.07%	97.65%	0.90%
|±1/2>	0.00%	0.00%	0.82%	0.94%	0.04%	0.08%	0.95%	97.17%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.89%	0.00%	0.06%	0.01%	0.01%	0.01%	0.01%	0.01%
|±13/2>	0.00%	98.88%	0.21%	0.24%	0.34%	0.13%	0.05%	0.14%
|±11/2>	0.07%	0.19%	88.07%	2.00%	6.61%	1.72%	0.25%	1.09%
|±9/2>	0.01%	0.58%	2.06%	15.33%	43.88%	28.32%	3.72%	6.10%
|±7/2>	0.00%	0.02%	5.31%	7.10%	12.90%	26.71%	29.24%	18.72%
|±5/2>	0.03%	0.01%	0.11%	22.54%	6.08%	14.36%	30.89%	25.98%
|±3/2>	0.00%	0.31%	1.49%	20.54%	20.55%	11.15%	20.72%	25.25%
|±1/2>	0.00%	0.02%	2.68%	32.25%	9.64%	17.60%	15.12%	22.70%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	95.46%	0.98%	0.19%	0.81%	1.97%	0.46%	0.11%	0.03%
|±13/2>	0.04%	18.22%	40.97%	34.57%	3.33%	1.82%	0.31%	0.73%
|±11/2>	0.20%	6.85%	6.62%	1.88%	6.31%	63.41%	5.94%	8.80%
|±9/2>	0.07%	3.51%	1.60%	1.32%	5.36%	11.32%	17.14%	59.67%
|±7/2>	0.30%	1.03%	0.38%	9.48%	7.49%	6.33%	58.53%	16.47%

|±5/2>	3.34%	1.64%	1.51%	15.38%	55.48%	8.82%	12.07%	1.77%
|±3/2>	0.09%	21.87%	17.98%	33.69%	18.13%	1.30%	4.01%	2.94%
|±1/2>	0.50%	45.91%	30.76%	2.86%	1.94%	6.54%	1.90%	9.59%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	100.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
|±13/2>	0.00%	99.87%	0.10%	0.03%	0.00%	0.00%	0.00%	0.00%
|±11/2>	0.00%	0.11%	99.29%	0.55%	0.05%	0.00%	0.00%	0.00%
|±9/2>	0.00%	0.02%	0.57%	99.19%	0.19%	0.02%	0.00%	0.00%
|±7/2>	0.00%	0.00%	0.04%	0.20%	99.65%	0.09%	0.02%	0.00%
|±5/2>	0.00%	0.00%	0.00%	0.03%	0.08%	99.72%	0.13%	0.04%
|±3/2>	0.00%	0.00%	0.00%	0.00%	0.02%	0.13%	99.40%	0.44%
|±1/2>	0.00%	0.00%	0.00%	0.00%	0.00%	0.04%	0.44%	99.51%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S23 The decomposition of crystal field wave function of **2** (**QUHXEY**) in terms of $|m_J\rangle$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.12%	0.04%	0.63%	0.08%	0.10%	0.03%	0.00%	0.00%
|±13/2>	0.00%	89.61%	0.94%	1.27%	7.89%	0.27%	0.02%	0.00%
|±11/2>	0.80%	2.58%	79.65%	8.94%	2.26%	5.17%	0.58%	0.01%
|±9/2>	0.06%	7.61%	5.34%	7.17%	76.91%	2.13%	0.68%	0.10%
|±7/2>	0.02%	0.05%	10.36%	13.68%	8.03%	59.05%	8.26%	0.55%
|±5/2>	0.00%	0.03%	1.04%	20.78%	2.18%	25.81%	42.06%	8.10%
|±3/2>	0.00%	0.04%	1.00%	23.46%	1.59%	1.69%	39.77%	32.44%
|±1/2>	0.00%	0.04%	1.03%	24.62%	1.05%	5.85%	8.62%	58.80%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	92.66%	0.08%	7.01%	0.11%	0.13%	0.00%	0.00%	0.00%
|±13/2>	0.01%	88.26%	2.67%	8.61%	0.35%	0.10%	0.01%	0.00%
|±11/2>	7.06%	0.92%	78.67%	5.82%	7.05%	0.43%	0.02%	0.03%
|±9/2>	0.05%	10.08%	1.51%	75.39%	8.61%	3.79%	0.32%	0.25%
|±7/2>	0.21%	0.37%	9.45%	2.63%	75.14%	10.18%	0.79%	1.23%
|±5/2>	0.01%	0.26%	0.47%	6.89%	4.65%	72.48%	9.47%	5.79%
|±3/2>	0.00%	0.01%	0.21%	0.38%	3.77%	9.25%	59.03%	27.35%
|±1/2>	0.00%	0.01%	0.02%	0.17%	0.30%	3.77%	30.37%	65.37%

Q1	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
|±15/2>	94.59%	0.01%	5.25%	0.05%	0.11%	0.01%	0.00%	0.00%
|±13/2>	0.00%	81.09%	0.11%	18.40%	0.19%	0.20%	0.01%	0.00%
|±11/2>	4.92%	0.07%	76.50%	1.53%	16.15%	0.65%	0.09%	0.09%
|±9/2>	0.00%	17.68%	0.43%	70.04%	3.30%	7.25%	0.74%	0.56%
|±7/2>	0.47%	0.05%	16.89%	0.63%	74.89%	5.86%	0.57%	0.65%
|±5/2>	0.00%	1.03%	0.39%	8.58%	4.40%	63.45%	18.49%	3.65%
|±3/2>	0.02%	0.04%	0.33%	0.75%	0.81%	10.66%	59.13%	28.26%
|±1/2>	0.00%	0.04%	0.10%	0.02%	0.15%	11.93%	20.98%	66.78%
Q2	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
|±15/2>	97.97%	0.04%	0.11%	0.20%	0.57%	0.67%	0.40%	0.04%
|±13/2>	0.00%	1.10%	2.55%	14.50%	39.66%	31.73%	9.78%	0.68%
|±11/2>	1.88%	4.92%	4.86%	8.89%	24.53%	30.86%	21.51%	2.55%
|±9/2>	0.05%	18.15%	19.72%	20.33%	2.33%	14.07%	20.15%	5.20%
|±7/2>	0.04%	29.64%	28.04%	6.68%	1.67%	5.73%	18.51%	9.70%
|±5/2>	0.02%	25.54%	22.71%	3.98%	12.72%	2.19%	15.69%	17.15%
|±3/2>	0.04%	14.68%	14.75%	11.66%	17.15%	3.77%	10.18%	27.76%
|±1/2>	0.00%	5.92%	7.27%	33.75%	1.39%	10.98%	3.77%	36.92%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S24 The decomposition of crystal field wave function of **3** (**QUHXAU**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
|±15/2>	98.44%	0.01%	1.22%	0.14%	0.14%	0.04%	0.00%	0.00%
|±13/2>	0.01%	89.03%	0.33%	1.55%	8.83%	0.24%	0.01%	0.00%
|±11/2>	1.41%	0.47%	83.67%	7.49%	0.91%	5.43%	0.61%	0.00%
|±9/2>	0.13%	10.29%	0.60%	9.88%	77.89%	0.48%	0.61%	0.12%
|±7/2>	0.01%	0.08%	12.10%	13.97%	5.22%	61.77%	6.45%	0.40%
|±5/2>	0.00%	0.03%	0.64%	20.66%	3.37%	25.17%	42.92%	7.22%
|±3/2>	0.01%	0.04%	0.68%	22.84%	2.02%	0.65%	41.83%	31.93%
|±1/2>	0.00%	0.05%	0.75%	23.48%	1.61%	6.22%	7.56%	60.32%
Q	KD_0	KD_1	KD_2	KD_3	KD_4	KD_5	KD_6	KD_7
|±15/2>	91.52%	0.00%	8.22%	0.00%	0.25%	0.00%	0.00%	0.00%
|±13/2>	0.00%	88.05%	0.04%	11.52%	0.02%	0.35%	0.01%	0.00%

|±11/2>	8.16%	0.01%	79.39%	0.11%	11.94%	0.12%	0.23%	0.03%
|±9/2>	0.00%	11.56%	0.02%	77.12%	0.37%	10.41%	0.32%	0.20%
|±7/2>	0.31%	0.00%	11.96%	0.05%	77.25%	1.91%	7.16%	1.35%
|±5/2>	0.00%	0.35%	0.01%	10.72%	0.33%	75.54%	6.69%	6.36%
|±3/2>	0.01%	0.00%	0.34%	0.07%	9.01%	2.25%	65.34%	22.98%
|±1/2>	0.00%	0.01%	0.02%	0.40%	0.82%	9.43%	20.24%	69.08%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	93.67%	0.00%	6.05%	0.02%	0.24%	0.00%	0.01%	0.00%
|±13/2>	0.00%	78.70%	0.18%	20.28%	0.20%	0.61%	0.02%	0.00%
|±11/2>	5.79%	0.01%	72.22%	0.83%	20.48%	0.40%	0.27%	0.01%
|±9/2>	0.01%	19.78%	0.08%	62.70%	2.30%	14.76%	0.15%	0.24%
|±7/2>	0.51%	0.07%	20.49%	0.22%	68.87%	3.31%	4.94%	1.60%
|±5/2>	0.00%	1.35%	0.21%	15.23%	1.26%	78.97%	1.37%	1.61%
|±3/2>	0.02%	0.03%	0.67%	0.57%	6.33%	1.08%	70.71%	20.58%
|±1/2>	0.00%	0.06%	0.11%	0.15%	0.32%	0.87%	22.53%	75.96%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.29%	0.32%	0.17%	0.36%	0.69%	0.74%	0.39%	0.04%
|±13/2>	0.02%	2.57%	2.48%	17.25%	40.34%	27.37%	9.26%	0.70%
|±11/2>	2.41%	7.65%	3.35%	11.31%	21.66%	28.99%	21.96%	2.68%
|±9/2>	0.14%	22.97%	13.33%	18.46%	4.11%	14.19%	21.31%	5.50%
|±7/2>	0.02%	30.37%	19.51%	10.33%	4.78%	6.65%	18.32%	10.01%
|±5/2>	0.08%	21.79%	22.70%	7.99%	9.68%	5.69%	14.73%	17.34%
|±3/2>	0.03%	11.17%	19.88%	12.44%	12.37%	6.91%	9.64%	27.55%
|±1/2>	0.01%	3.15%	18.58%	21.86%	6.36%	9.46%	4.40%	36.19%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S25 The decomposition of crystal field wave function of **4** (**QUHWUN**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	96.07%	0.02%	1.03%	2.70%	0.07%	0.10%	0.01%	0.00%
|±13/2>	0.00%	92.36%	1.62%	1.37%	4.53%	0.09%	0.03%	0.01%
|±11/2>	3.83%	0.57%	21.58%	70.12%	1.04%	2.43%	0.43%	0.00%
|±9/2>	0.01%	6.09%	5.12%	3.07%	82.93%	2.05%	0.61%	0.12%
|±7/2>	0.05%	0.10%	15.52%	4.84%	6.24%	62.00%	9.66%	1.59%

|±5/2>	0.01%	0.31%	16.10%	4.98%	1.42%	23.77%	42.65%	10.76%
|±3/2>	0.02%	0.22%	18.73%	6.69%	2.04%	1.02%	38.51%	32.77%
|±1/2>	0.01%	0.33%	20.30%	6.23%	1.73%	8.54%	8.11%	54.76%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	94.31%	0.01%	5.49%	0.02%	0.17%	0.01%	0.00%	0.00%
|±13/2>	0.00%	90.23%	0.42%	8.90%	0.10%	0.31%	0.04%	0.00%
|±11/2>	5.51%	0.14%	82.13%	1.10%	10.14%	0.56%	0.37%	0.05%
|±9/2>	0.01%	9.24%	0.25%	76.10%	2.25%	9.97%	1.79%	0.39%
|±7/2>	0.16%	0.06%	11.06%	0.38%	67.66%	8.09%	10.20%	2.37%
|±5/2>	0.00%	0.31%	0.11%	12.28%	1.01%	52.60%	23.42%	10.28%
|±3/2>	0.00%	0.00%	0.50%	0.25%	15.66%	4.66%	48.26%	30.67%
|±1/2>	0.00%	0.01%	0.03%	0.99%	3.02%	23.80%	15.92%	56.24%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	95.37%	0.01%	4.36%	0.01%	0.25%	0.01%	0.00%	0.00%
|±13/2>	0.00%	86.82%	0.06%	12.73%	0.08%	0.30%	0.01%	0.00%
|±11/2>	4.44%	0.10%	81.33%	0.20%	13.46%	0.33%	0.06%	0.07%
|±9/2>	0.00%	12.53%	0.14%	77.88%	1.05%	7.27%	0.69%	0.45%
|±7/2>	0.18%	0.00%	13.68%	0.39%	81.09%	2.73%	0.70%	1.23%
|±5/2>	0.00%	0.48%	0.08%	8.24%	2.26%	74.15%	8.22%	6.56%
|±3/2>	0.01%	0.01%	0.22%	0.44%	1.58%	3.97%	63.76%	30.00%
|±1/2>	0.00%	0.05%	0.13%	0.11%	0.22%	11.23%	26.57%	61.68%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.23%	0.33%	0.68%	0.47%	0.66%	0.36%	0.24%	0.03%
|±13/2>	0.03%	3.24%	2.56%	19.42%	43.71%	21.71%	8.54%	0.78%
|±11/2>	2.30%	4.86%	8.75%	15.19%	22.12%	21.48%	22.08%	3.22%
|±9/2>	0.28%	19.92%	19.77%	17.51%	5.16%	10.01%	20.96%	6.39%
|±7/2>	0.01%	26.04%	25.65%	5.22%	6.63%	10.21%	15.59%	10.66%
|±5/2>	0.13%	18.38%	25.44%	2.61%	9.06%	15.32%	11.79%	17.28%
|±3/2>	0.01%	17.21%	10.18%	14.77%	5.63%	15.01%	10.48%	26.71%
|±1/2>	0.01%	10.02%	6.97%	24.81%	7.03%	5.92%	10.30%	34.94%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S26 The decomposition of crystal field wave function of **5** (**QOPZIG**) in terms of $|m_J\rangle$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} *a*

full	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	97.52%	0.97%	0.58%	0.48%	0.31%	0.01%	0.07%	0.07%
|±13/2>	0.05%	4.45%	0.99%	1.37%	4.31%	10.71%	63.08%	15.03%
|±11/2>	0.03%	1.65%	3.38%	1.93%	4.69%	18.93%	3.80%	65.59%
|±9/2>	0.02%	2.60%	5.98%	15.52%	12.83%	37.69%	14.67%	10.69%
|±7/2>	0.43%	11.53%	16.60%	31.44%	19.31%	15.74%	4.16%	0.79%
|±5/2>	1.92%	26.96%	15.19%	22.25%	25.83%	7.30%	0.41%	0.14%
|±3/2>	0.03%	40.23%	13.45%	11.43%	22.59%	1.58%	9.69%	1.01%
|±1/2>	0.00%	11.63%	43.83%	15.57%	10.12%	8.03%	4.12%	6.69%
Q	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	98.23%	0.00%	1.62%	0.06%	0.06%	0.02%	0.01%	0.00%
|±13/2>	0.00%	94.17%	0.08%	4.89%	0.40%	0.33%	0.11%	0.02%
|±11/2>	1.64%	0.04%	85.62%	0.65%	8.91%	2.10%	0.83%	0.20%
|±9/2>	0.06%	5.26%	0.30%	68.31%	8.00%	12.77%	4.13%	1.17%
|±7/2>	0.06%	0.15%	10.66%	1.71%	43.75%	24.06%	14.65%	4.97%
|±5/2>	0.00%	0.35%	0.19%	18.00%	4.33%	32.35%	29.73%	15.04%
|±3/2>	0.00%	0.01%	1.33%	1.69%	24.17%	7.22%	33.72%	31.86%
|±1/2>	0.00%	0.03%	0.20%	4.70%	10.38%	21.15%	16.81%	46.74%
Q1	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	0.00%	0.00%	0.11%	0.01%	0.01%	0.02%	1.28%	98.58%
|±13/2>	0.01%	0.25%	0.01%	0.02%	0.03%	2.67%	95.81%	1.21%
|±11/2>	0.37%	0.01%	0.01%	0.09%	2.19%	94.67%	2.58%	0.06%
|±9/2>	0.44%	0.05%	0.13%	1.05%	96.11%	2.15%	0.04%	0.02%
|±7/2>	0.05%	0.61%	0.65%	97.54%	1.05%	0.08%	0.01%	0.01%
|±5/2>	0.31%	0.21%	98.55%	0.68%	0.13%	0.00%	0.00%	0.12%
|±3/2>	1.73%	97.08%	0.26%	0.60%	0.06%	0.00%	0.26%	0.00%
|±1/2>	97.08%	1.78%	0.28%	0.01%	0.42%	0.40%	0.02%	0.00%
Q2	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	99.85%	0.01%	0.13%	0.01%	0.00%	0.00%	0.00%	0.00%
|±13/2>	0.00%	93.52%	5.32%	1.05%	0.07%	0.04%	0.00%	0.00%
|±11/2>	0.14%	3.60%	78.12%	13.15%	4.80%	0.08%	0.10%	0.01%
|±9/2>	0.00%	2.56%	9.57%	77.02%	4.14%	6.14%	0.45%	0.11%
|±7/2>	0.00%	0.24%	5.99%	2.57%	82.30%	1.50%	6.50%	0.90%
|±5/2>	0.00%	0.05%	0.66%	5.81%	0.51%	74.86%	11.86%	6.24%

|±3/2>	0.00%	0.01%	0.17%	0.20%	7.64%	2.69%	64.09%	25.20%
|±1/2>	0.00%	0.00%	0.04%	0.20%	0.53%	14.69%	16.99%	67.54%

[a] The quantization axis is the magnetic easy axis of the pseudospin S = 1/2 representing the ground KD

Table S27 The decomposition of crystal field wave function of **6** (**XILRUJ**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	98.58%	0.00%	0.03%	0.47%	0.38%	0.50%	0.02%	0.00%
|±13/2>	0.02%	94.19%	0.85%	0.33%	0.36%	3.03%	1.15%	0.07%
|±11/2>	1.06%	1.26%	5.64%	60.63%	19.30%	9.85%	1.70%	0.55%
|±9/2>	0.33%	1.42%	6.28%	10.18%	20.22%	52.55%	7.57%	1.45%
|±7/2>	0.01%	2.70%	5.97%	5.51%	23.46%	18.26%	37.40%	6.70%
|±5/2>	0.00%	0.08%	18.50%	6.36%	15.78%	7.86%	33.22%	18.20%
|±3/2>	0.00%	0.23%	26.32%	12.15%	8.01%	5.33%	16.02%	31.93%
|±1/2>	0.00%	0.12%	36.41%	4.37%	12.49%	2.62%	2.91%	41.09%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.40%	0.00%	0.56%	0.02%	0.01%	0.00%	0.00%	0.00%
|±13/2>	0.00%	96.47%	1.41%	1.63%	0.39%	0.08%	0.02%	0.00%
|±11/2>	0.58%	0.60%	84.33%	9.70%	2.39%	1.94%	0.41%	0.04%
|±9/2>	0.01%	2.68%	3.51%	58.00%	23.14%	8.28%	3.87%	0.51%
|±7/2>	0.01%	0.12%	8.31%	5.27%	31.27%	35.09%	16.48%	3.46%
|±5/2>	0.00%	0.11%	0.93%	17.71%	2.25%	26.57%	38.78%	13.65%
|±3/2>	0.00%	0.01%	0.80%	3.48%	27.41%	1.07%	34.42%	32.81%
|±1/2>	0.00%	0.00%	0.15%	4.19%	13.14%	26.97%	6.02%	49.52%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.61%	0.01%	0.35%	0.02%	0.01%	0.00%	0.00%	0.00%
|±13/2>	0.00%	95.36%	3.42%	0.54%	0.58%	0.09%	0.01%	0.00%
|±11/2>	0.34%	1.54%	72.34%	21.23%	1.68%	2.44%	0.42%	0.01%
|±9/2>	0.04%	2.44%	7.91%	29.35%	40.19%	14.77%	4.92%	0.39%
|±7/2>	0.01%	0.42%	10.29%	7.10%	13.79%	42.31%	22.27%	3.80%
|±5/2>	0.00%	0.23%	2.93%	20.71%	1.22%	20.19%	39.39%	15.33%
|±3/2>	0.00%	0.01%	2.19%	9.13%	25.90%	0.76%	28.85%	33.16%
|±1/2>	0.00%	0.00%	0.57%	11.92%	16.63%	19.43%	4.14%	47.30%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

|±15/2>	97.70%	0.00%	1.54%	0.12%	0.12%	0.04%	0.48%	0.00%
|±13/2>	0.04%	93.21%	0.08%	0.80%	1.65%	0.66%	3.54%	0.01%
|±11/2>	1.91%	0.16%	82.09%	1.42%	1.20%	0.75%	12.09%	0.38%
|±9/2>	0.34%	4.75%	1.29%	17.61%	42.59%	12.01%	19.42%	1.98%
|±7/2>	0.00%	1.78%	5.27%	11.66%	14.85%	29.22%	31.94%	5.28%
|±5/2>	0.01%	0.02%	8.49%	6.39%	18.16%	24.84%	24.79%	17.30%
|±3/2>	0.01%	0.03%	0.36%	32.23%	2.64%	26.02%	6.31%	32.40%
|±1/2>	0.00%	0.05%	0.87%	29.76%	18.79%	6.46%	1.43%	42.64%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S28 The decomposition of crystal field wave function of **7** (**XILSEU**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.49%	0.07%	0.15%	0.60%	0.24%	0.05%	0.36%	1.04%
|±13/2>	0.05%	74.33%	10.67%	2.87%	2.89%	0.09%	3.16%	5.93%
|±11/2>	1.89%	1.94%	0.71%	26.20%	32.63%	9.75%	12.08%	14.80%
|±9/2>	0.31%	8.43%	0.71%	2.37%	17.17%	31.76%	5.20%	34.04%
|±7/2>	0.17%	3.15%	2.02%	14.26%	13.97%	32.54%	9.88%	24.01%
|±5/2>	0.02%	3.69%	8.49%	22.15%	18.33%	17.83%	26.08%	3.41%
|±3/2>	0.06%	2.54%	29.79%	22.50%	9.89%	5.65%	23.65%	5.92%
|±1/2>	0.02%	5.83%	47.46%	9.03%	4.88%	2.34%	19.60%	10.85%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.76%	0.00%	0.20%	0.03%	0.00%	0.00%	0.00%	0.00%
|±13/2>	0.00%	97.54%	1.72%	0.39%	0.29%	0.02%	0.03%	0.01%
|±11/2>	0.23%	1.05%	85.18%	12.05%	0.05%	0.98%	0.35%	0.09%
|±9/2>	0.00%	1.37%	6.57%	56.75%	30.06%	1.48%	2.98%	0.79%
|±7/2>	0.00%	0.03%	5.60%	12.49%	27.47%	37.19%	12.96%	4.26%
|±5/2>	0.00%	0.01%	0.59%	14.19%	8.61%	20.29%	41.24%	15.07%
|±3/2>	0.00%	0.00%	0.12%	3.08%	23.57%	4.48%	35.59%	33.15%
|±1/2>	0.00%	0.00%	0.02%	1.01%	9.94%	35.55%	6.85%	46.63%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.71%	0.02%	0.11%	0.08%	0.06%	0.00%	0.02%	0.01%
|±13/2>	0.00%	89.05%	9.62%	0.68%	0.07%	0.27%	0.20%	0.11%
|±11/2>	0.24%	6.78%	42.84%	37.75%	9.84%	0.38%	1.37%	0.78%

|±9/2>	0.03%	3.54%	21.07%	4.44%	38.17%	22.23%	6.87%	3.65%
|±7/2>	0.02%	0.47%	17.01%	11.09%	1.26%	26.41%	32.30%	11.46%
|±5/2>	0.01%	0.06%	7.09%	21.35%	1.90%	9.37%	38.12%	22.11%
|±3/2>	0.00%	0.07%	1.54%	17.90%	14.97%	16.99%	19.36%	29.17%
|±1/2>	0.00%	0.02%	0.71%	6.70%	33.74%	24.34%	1.77%	32.72%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.82%	0.01%	1.19%	0.12%	0.30%	0.02%	0.00%	0.54%
|±13/2>	0.03%	92.11%	0.21%	0.38%	2.71%	0.38%	0.04%	4.15%
|±11/2>	1.84%	0.61%	73.67%	7.76%	1.35%	0.79%	0.16%	13.82%
|±9/2>	0.28%	5.53%	4.29%	11.63%	37.19%	13.13%	2.50%	25.45%
|±7/2>	0.00%	1.30%	8.41%	16.52%	9.11%	27.77%	8.99%	27.89%
|±5/2>	0.02%	0.04%	6.30%	13.91%	10.93%	32.38%	18.17%	18.27%
|±3/2>	0.01%	0.18%	2.20%	26.19%	11.48%	20.87%	31.69%	7.37%
|±1/2>	0.00%	0.22%	3.74%	23.49%	26.93%	4.67%	38.44%	2.51%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S29 The decomposition of crystal field wave function of **8** (**XILSOE**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	96.13%	0.03%	0.00%	0.01%	0.24%	0.04%	3.11%	0.44%
|±13/2>	0.01%	69.44%	9.40%	2.10%	14.65%	3.78%	0.46%	0.16%
|±11/2>	0.00%	0.44%	0.48%	18.94%	20.58%	57.58%	0.58%	1.40%
|±9/2>	0.01%	0.05%	1.30%	1.10%	1.93%	1.47%	14.20%	79.92%
|±7/2>	3.82%	0.12%	0.01%	0.32%	4.32%	0.81%	78.44%	12.16%
|±5/2>	0.02%	20.31%	0.13%	0.09%	55.98%	18.71%	2.95%	1.81%
|±3/2>	0.01%	4.41%	9.44%	65.49%	1.36%	17.53%	0.12%	1.64%
|±1/2>	0.00%	5.20%	79.23%	11.96%	0.93%	0.08%	0.13%	2.46%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	77.25%	0.99%	14.40%	3.92%	2.34%	0.76%	0.26%	0.07%
|±13/2>	0.05%	51.78%	9.34%	22.60%	9.89%	4.37%	1.52%	0.45%
|±11/2>	18.59%	0.11%	20.27%	18.68%	23.99%	12.09%	4.85%	1.42%
|±9/2>	0.09%	32.51%	0.35%	7.28%	21.39%	23.31%	11.63%	3.44%
|±7/2>	3.14%	1.15%	32.55%	3.57%	5.04%	24.04%	22.88%	7.63%
|±5/2>	0.11%	8.89%	2.83%	27.79%	4.66%	8.17%	31.35%	16.19%

|±3/2>	0.57%	1.66%	13.78%	3.52%	25.62%	1.80%	23.59%	29.46%
|±1/2>	0.20%	2.92%	6.48%	12.62%	7.06%	25.45%	3.92%	41.35%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	88.01%	1.14%	1.61%	2.40%	4.76%	0.46%	1.50%	0.14%
|±13/2>	0.24%	21.15%	14.41%	27.64%	16.64%	8.03%	9.96%	1.93%
|±11/2>	11.30%	9.35%	14.20%	16.33%	26.82%	4.12%	16.69%	1.19%
|±9/2>	0.25%	43.86%	13.32%	4.35%	15.32%	8.48%	14.02%	0.39%
|±7/2>	0.09%	12.43%	42.89%	4.98%	18.33%	7.69%	10.40%	3.19%
|±5/2>	0.03%	5.37%	10.38%	31.90%	11.42%	22.77%	10.25%	7.90%
|±3/2>	0.02%	3.72%	1.94%	11.36%	5.40%	35.60%	20.41%	21.55%
|±1/2>	0.06%	2.98%	1.25%	1.03%	1.33%	12.87%	16.78%	63.71%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	84.59%	0.48%	0.47%	0.63%	6.46%	4.78%	1.58%	1.01%
|±13/2>	0.40%	33.85%	9.56%	8.60%	6.35%	9.52%	17.08%	14.63%
|±11/2>	1.77%	4.01%	9.68%	11.91%	14.99%	1.32%	21.08%	35.23%
|±9/2>	1.34%	0.49%	3.73%	5.36%	37.35%	25.91%	12.91%	12.90%
|±7/2>	11.05%	3.58%	0.50%	1.22%	26.72%	40.36%	14.18%	2.39%
|±5/2>	0.15%	38.27%	6.45%	2.02%	1.27%	14.51%	22.22%	15.10%
|±3/2>	0.54%	17.90%	34.01%	18.48%	4.60%	1.00%	9.17%	14.30%
|±1/2>	0.15%	1.41%	35.60%	51.76%	2.26%	2.60%	1.77%	4.44%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S30 The decomposition of crystal field wave function of **9** (**IFACOK**) in terms of $|m_J\rangle$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.70%	0.00%	0.08%	0.01%	0.18%	0.10%	0.01%	1.92%
|±13/2>	0.03%	87.93%	2.69%	1.36%	0.32%	1.64%	4.98%	1.05%
|±11/2>	0.02%	2.16%	1.18%	10.66%	43.55%	23.37%	14.42%	4.63%
|±9/2>	1.65%	0.03%	0.65%	0.14%	5.89%	11.87%	19.78%	60.00%
|±7/2>	0.52%	3.23%	0.17%	0.50%	0.96%	13.19%	51.90%	29.52%
|±5/2>	0.04%	3.01%	0.50%	4.97%	34.74%	48.65%	6.59%	1.48%
|±3/2>	0.01%	1.46%	7.46%	74.69%	12.77%	0.74%	2.17%	0.70%
|±1/2>	0.03%	2.18%	87.27%	7.66%	1.58%	0.43%	0.14%	0.70%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

|±15/2>	80.07%	3.39%	9.55%	4.75%	1.65%	0.44%	0.11%	0.03%
|±13/2>	0.21%	37.12%	24.99%	21.53%	11.20%	3.81%	0.91%	0.23%
|±11/2>	13.24%	0.45%	12.17%	27.09%	27.15%	14.64%	4.32%	0.93%
|±9/2>	0.21%	24.41%	3.03%	3.70%	23.62%	28.50%	13.53%	2.98%
|±7/2>	3.94%	2.98%	21.54%	8.76%	2.34%	25.39%	26.83%	8.21%
|±5/2>	0.34%	15.59%	1.31%	19.15%	8.11%	5.60%	31.80%	18.09%
|±3/2>	1.38%	6.17%	18.16%	1.17%	20.69%	2.47%	19.56%	30.40%
|±1/2>	0.60%	9.90%	9.24%	13.84%	5.23%	19.14%	2.93%	39.13%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	82.42%	2.66%	7.91%	3.76%	1.00%	0.72%	0.96%	0.57%
|±13/2>	0.28%	47.85%	21.93%	12.32%	6.66%	3.06%	4.77%	3.14%
|±11/2>	11.54%	4.14%	15.03%	24.30%	17.29%	10.17%	10.03%	7.50%
|±9/2>	0.75%	17.42%	8.45%	10.09%	17.32%	21.40%	15.52%	9.04%
|±7/2>	3.26%	1.64%	12.65%	18.69%	10.90%	20.54%	22.86%	9.45%
|±5/2>	0.24%	11.23%	4.96%	11.54%	22.51%	8.36%	24.89%	16.28%
|±3/2>	0.99%	6.28%	15.80%	5.22%	20.87%	8.98%	17.64%	24.22%
|±1/2>	0.52%	8.77%	13.26%	14.09%	3.45%	26.77%	3.33%	29.80%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.96%	0.00%	0.00%	0.01%	0.00%	0.00%	0.00%	0.02%
|±13/2>	0.00%	99.79%	0.00%	0.01%	0.00%	0.03%	0.06%	0.12%
|±11/2>	0.01%	0.02%	0.06%	91.90%	4.69%	2.48%	0.72%	0.11%
|±9/2>	0.02%	0.01%	0.04%	0.07%	2.65%	2.62%	79.27%	15.33%
|±7/2>	0.01%	0.12%	0.55%	0.04%	0.07%	6.90%	13.35%	78.96%
|±5/2>	0.00%	0.06%	0.59%	1.77%	3.13%	86.16%	3.71%	4.57%
|±3/2>	0.00%	0.00%	3.21%	5.87%	86.58%	1.20%	2.85%	0.29%
|±1/2>	0.00%	0.00%	95.54%	0.33%	2.89%	0.62%	0.03%	0.60%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S31 The decomposition of crystal field wave function of **10** (**IFACIE**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	97.46%	0.01%	0.04%	0.09%	0.30%	0.12%	0.07%	1.92%
|±13/2>	0.02%	88.04%	1.73%	0.86%	0.38%	3.56%	4.88%	0.53%
|±11/2>	0.15%	4.15%	2.45%	16.93%	43.94%	11.35%	14.26%	6.77%

|±9/2>	1.56%	0.01%	0.99%	0.56%	10.56%	10.28%	18.76%	57.28%
|±7/2>	0.74%	2.64%	0.11%	0.23%	3.30%	8.13%	55.05%	29.80%
|±5/2>	0.04%	3.06%	0.67%	10.69%	14.42%	63.88%	4.72%	2.51%
|±3/2>	0.01%	0.85%	12.50%	57.29%	24.10%	2.46%	2.08%	0.70%
|±1/2>	0.01%	1.23%	81.51%	13.35%	3.00%	0.22%	0.19%	0.49%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	87.19%	1.18%	6.98%	3.38%	1.04%	0.20%	0.02%	0.00%
|±13/2>	0.05%	48.73%	20.07%	18.77%	9.29%	2.65%	0.41%	0.04%
|±11/2>	9.51%	0.62%	17.28%	29.69%	26.55%	12.85%	3.11%	0.41%
|±9/2>	0.07%	23.05%	0.87%	6.22%	27.05%	28.41%	12.03%	2.29%
|±7/2>	2.25%	2.23%	22.92%	6.63%	4.02%	28.01%	26.00%	7.94%
|±5/2>	0.10%	12.68%	2.29%	19.85%	6.59%	7.81%	32.19%	18.49%
|±3/2>	0.63%	4.23%	19.09%	1.32%	20.28%	2.28%	21.08%	31.08%
|±1/2>	0.21%	7.28%	10.50%	14.15%	5.18%	17.79%	5.16%	39.73%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	75.70%	2.77%	12.16%	5.85%	0.99%	0.69%	0.85%	0.98%
|±13/2>	0.39%	47.88%	22.40%	11.82%	6.54%	2.62%	3.79%	4.55%
|±11/2>	15.12%	2.85%	11.44%	25.19%	17.70%	10.02%	7.73%	9.96%
|±9/2>	0.86%	18.68%	5.64%	9.86%	17.56%	22.73%	13.37%	11.30%
|±7/2>	4.96%	0.83%	12.51%	16.99%	10.25%	21.19%	22.33%	10.94%
|±5/2>	0.32%	11.53%	4.45%	12.46%	22.05%	6.53%	26.42%	16.22%
|±3/2>	1.70%	6.02%	17.09%	4.29%	21.62%	7.93%	20.12%	21.22%
|±1/2>	0.94%	9.44%	14.30%	13.54%	3.28%	28.29%	5.39%	24.83%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.96%	0.00%	0.03%	0.00%	0.00%	0.00%	0.01%	0.00%
|±13/2>	0.00%	99.57%	0.27%	0.01%	0.01%	0.12%	0.00%	0.02%
|±11/2>	0.03%	0.29%	95.66%	2.37%	0.93%	0.28%	0.13%	0.33%
|±9/2>	0.01%	0.06%	0.14%	0.48%	0.50%	56.29%	35.01%	7.51%
|±7/2>	0.01%	0.05%	0.37%	0.10%	0.15%	4.72%	36.36%	58.25%
|±5/2>	0.00%	0.03%	0.70%	0.62%	15.43%	33.75%	23.12%	26.36%
|±3/2>	0.00%	0.00%	1.23%	14.89%	70.42%	2.55%	4.80%	6.12%
|±1/2>	0.00%	0.00%	1.60%	81.54%	12.56%	2.29%	0.58%	1.41%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S32 The decomposition of crystal field wave function of **11** (**IFACEA**) in terms of $|m_J\rangle$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} *a*

full	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	96.83%	0.02%	0.04%	0.24%	0.31%	0.60%	0.30%	1.65%
|±13/2>	0.02%	85.79%	0.86%	0.66%	2.46%	8.41%	0.94%	0.87%
|±11/2>	0.21%	6.24%	3.61%	21.41%	43.81%	8.44%	9.11%	7.17%
|±9/2>	1.95%	0.02%	0.49%	2.90%	11.76%	4.61%	46.12%	32.14%
|±7/2>	0.94%	3.40%	0.22%	0.77%	4.44%	17.05%	26.82%	46.36%
|±5/2>	0.01%	3.50%	4.55%	18.40%	2.79%	49.18%	10.74%	10.82%
|±3/2>	0.02%	0.47%	30.53%	27.03%	26.52%	9.02%	5.88%	0.54%
|±1/2>	0.01%	0.56%	59.69%	28.59%	7.91%	2.70%	0.09%	0.46%
Q	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	87.30%	1.16%	7.10%	3.29%	0.97%	0.16%	0.01%	0.00%
|±13/2>	0.02%	49.49%	19.60%	18.85%	9.14%	2.49%	0.36%	0.04%
|±11/2>	9.39%	0.60%	17.82%	29.78%	26.46%	12.57%	3.00%	0.38%
|±9/2>	0.04%	22.90%	0.74%	6.54%	27.53%	28.18%	11.91%	2.17%
|±7/2>	2.31%	1.97%	23.04%	6.25%	4.42%	28.31%	26.07%	7.63%
|±5/2>	0.08%	12.72%	2.01%	19.95%	6.24%	8.20%	32.64%	18.15%
|±3/2>	0.66%	3.96%	19.36%	1.03%	20.23%	2.23%	21.34%	31.19%
|±1/2>	0.21%	7.20%	10.33%	14.30%	5.02%	17.85%	4.67%	40.44%
Q1	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	77.32%	0.46%	17.14%	3.26%	0.56%	0.33%	0.71%	0.23%
|±13/2>	0.05%	59.57%	5.43%	20.67%	7.74%	2.20%	3.42%	0.93%
|±11/2>	11.73%	0.46%	23.31%	18.12%	23.58%	13.52%	8.61%	0.66%
|±9/2>	0.19%	17.66%	0.58%	10.11%	23.01%	29.32%	18.40%	0.73%
|±7/2>	8.19%	0.01%	14.09%	4.92%	7.42%	30.96%	26.18%	8.23%
|±5/2>	0.06%	15.77%	0.72%	14.77%	10.82%	10.74%	24.70%	22.42%
|±3/2>	2.16%	0.94%	26.66%	1.39%	21.03%	2.18%	13.19%	32.45%
|±1/2>	0.30%	5.13%	12.07%	26.75%	5.83%	10.76%	4.80%	34.36%
Q2	**KD$_0$**	**KD$_1$**	**KD$_2$**	**KD$_3$**	**KD$_4$**	**KD$_5$**	**KD$_6$**	**KD$_7$**
|±15/2>	99.92%	0.00%	0.06%	0.01%	0.00%	0.00%	0.00%	0.00%
|±13/2>	0.00%	99.39%	0.30%	0.09%	0.01%	0.15%	0.06%	0.01%
|±11/2>	0.07%	0.37%	81.53%	15.03%	2.09%	0.01%	0.42%	0.47%
|±9/2>	0.00%	0.15%	0.22%	0.31%	0.46%	39.50%	49.94%	9.41%
|±7/2>	0.01%	0.04%	0.77%	0.27%	1.42%	16.15%	26.11%	55.23%
|±5/2>	0.00%	0.03%	1.47%	0.85%	20.08%	30.79%	19.70%	27.08%

|±3/2>	0.00%	0.02%	2.49%	18.82%	59.61%	9.26%	3.37%	6.43%
|±1/2>	0.00%	0.00%	13.16%	64.62%	16.32%	4.15%	0.40%	1.35%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S33 The decomposition of crystal field wave function of **12** (**IFACUQ**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	96.89%	0.00%	0.00%	0.00%	0.02%	0.04%	0.54%	2.50%
|±13/2>	0.00%	88.60%	4.60%	0.08%	0.02%	0.09%	5.45%	1.16%
|±11/2>	0.04%	0.01%	0.20%	0.17%	44.91%	54.24%	0.42%	0.01%
|±9/2>	3.05%	0.06%	0.01%	0.04%	0.03%	0.27%	22.40%	74.14%
|±7/2>	0.02%	6.89%	0.01%	0.04%	0.63%	1.32%	69.06%	22.03%
|±5/2>	0.00%	0.20%	0.93%	0.83%	52.95%	43.23%	1.77%	0.10%
|±3/2>	0.00%	0.18%	0.70%	97.97%	0.43%	0.65%	0.04%	0.02%
|±1/2>	0.00%	4.06%	93.55%	0.85%	1.01%	0.15%	0.32%	0.05%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	54.44%	19.60%	15.85%	7.04%	2.39%	0.57%	0.10%	0.01%
|±13/2>	1.38%	22.64%	29.71%	26.48%	13.98%	4.73%	0.97%	0.12%
|±11/2>	18.68%	0.52%	6.48%	24.04%	28.49%	16.33%	4.80%	0.66%
|±9/2>	2.12%	22.14%	7.58%	1.50%	21.04%	28.69%	14.28%	2.64%
|±7/2>	10.06%	1.24%	18.28%	10.23%	1.38%	24.02%	26.85%	7.95%
|±5/2>	2.94%	17.08%	0.02%	17.36%	8.47%	5.08%	31.07%	17.98%
|±3/2>	6.22%	5.59%	15.91%	0.39%	19.79%	2.34%	19.07%	30.69%
|±1/2>	4.16%	11.20%	6.17%	12.97%	4.46%	18.23%	2.86%	39.94%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	0.00%	0.01%	0.02%	0.09%	33.12%	0.09%	47.59%	19.07%
|±13/2>	0.19%	0.00%	0.02%	12.37%	0.00%	0.28%	24.06%	63.07%
|±11/2>	0.14%	0.01%	2.57%	0.08%	0.08%	96.54%	0.14%	0.43%
|±9/2>	0.04%	0.07%	0.19%	0.50%	65.82%	0.17%	25.11%	8.11%
|±7/2>	2.54%	0.12%	0.45%	83.65%	0.63%	0.35%	3.02%	9.24%
|±5/2>	4.93%	0.40%	91.41%	0.39%	0.23%	2.54%	0.06%	0.04%
|±3/2>	0.36%	99.08%	0.34%	0.08%	0.11%	0.02%	0.01%	0.01%
|±1/2>	91.80%	0.31%	5.00%	2.85%	0.01%	0.00%	0.01%	0.02%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**

|±15/2>	99.96%	0.00%	0.00%	0.00%	0.00%	0.01%	0.03%	0.00%
|±13/2>	0.00%	99.79%	0.00%	0.02%	0.04%	0.00%	0.01%	0.13%
|±11/2>	0.00%	0.02%	0.98%	95.37%	1.37%	2.08%	0.11%	0.06%
|±9/2>	0.04%	0.01%	0.05%	0.04%	0.81%	1.69%	91.33%	6.03%
|±7/2>	0.00%	0.15%	0.18%	0.04%	0.07%	3.07%	5.05%	91.44%
|±5/2>	0.00%	0.00%	0.19%	2.25%	1.22%	91.73%	2.55%	2.05%
|±3/2>	0.00%	0.03%	1.38%	1.05%	95.23%	1.31%	0.89%	0.11%
|±1/2>	0.00%	0.00%	97.21%	1.23%	1.25%	0.11%	0.02%	0.17%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S34 The decomposition of crystal field wave function of **13** (**TOFTUG**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	94.63%	0.00%	0.02%	0.01%	0.01%	0.58%	0.03%	4.73%
|±13/2>	0.01%	1.82%	80.76%	1.61%	0.13%	1.11%	14.10%	0.46%
|±11/2>	0.15%	0.12%	0.01%	0.19%	18.91%	69.34%	10.00%	1.29%
|±9/2>	5.19%	0.02%	0.02%	0.09%	0.13%	3.87%	2.52%	88.17%
|±7/2>	0.02%	1.19%	14.57%	0.05%	0.05%	9.34%	70.39%	4.38%
|±5/2>	0.00%	1.05%	0.28%	0.11%	79.39%	15.58%	2.78%	0.83%
|±3/2>	0.00%	0.22%	1.64%	97.62%	0.26%	0.11%	0.01%	0.13%
|±1/2>	0.00%	95.58%	2.72%	0.32%	1.12%	0.08%	0.17%	0.00%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	66.86%	11.34%	11.50%	5.87%	2.08%	0.16%	0.97%	1.22%
|±13/2>	1.88%	26.82%	30.94%	20.48%	8.68%	2.19%	3.62%	5.38%
|±11/2>	15.05%	0.49%	8.70%	26.57%	19.92%	11.14%	6.27%	11.84%
|±9/2>	1.16%	20.93%	5.42%	3.74%	18.18%	17.61%	13.86%	19.10%
|±7/2>	7.49%	3.31%	17.17%	8.93%	3.09%	5.62%	30.60%	23.79%
|±5/2>	2.09%	16.84%	2.54%	16.01%	7.00%	3.77%	31.09%	20.67%
|±3/2>	3.04%	9.69%	14.04%	5.85%	19.10%	24.01%	12.55%	11.72%
|±1/2>	2.43%	10.57%	9.67%	12.54%	21.95%	35.51%	1.05%	6.29%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	0.00%	0.02%	0.06%	0.46%	30.30%	0.68%	61.22%	7.27%
|±13/2>	0.37%	0.02%	0.45%	10.00%	0.04%	0.01%	8.73%	80.37%
|±11/2>	0.27%	0.03%	2.60%	0.03%	0.11%	95.62%	1.04%	0.30%

|±9/2>	0.03%	0.09%	0.42%	0.53%	68.10%	0.65%	27.54%	2.65%
|±7/2>	5.28%	0.36%	3.54%	78.93%	0.96%	0.10%	1.46%	9.36%
|±5/2>	10.70%	0.20%	83.83%	2.06%	0.27%	2.93%	0.00%	0.00%
|±3/2>	0.26%	98.76%	0.49%	0.32%	0.15%	0.00%	0.00%	0.01%
|±1/2>	83.08%	0.53%	8.60%	7.67%	0.07%	0.00%	0.00%	0.04%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	99.70%	0.00%	0.00%	0.02%	0.01%	0.02%	0.17%	0.10%
|±13/2>	0.00%	98.59%	0.04%	0.01%	0.02%	0.00%	0.44%	0.91%
|±11/2>	0.01%	0.03%	0.11%	0.30%	77.51%	20.84%	1.20%	0.01%
|±9/2>	0.30%	0.00%	0.05%	4.12%	0.07%	2.62%	62.71%	30.13%
|±7/2>	0.00%	1.37%	1.10%	0.09%	0.03%	0.98%	30.55%	65.88%
|±5/2>	0.00%	0.00%	0.94%	0.06%	21.55%	74.86%	2.40%	0.18%
|±3/2>	0.00%	0.01%	1.08%	94.18%	0.39%	0.12%	2.18%	2.04%
|±1/2>	0.00%	0.01%	96.68%	1.22%	0.42%	0.55%	0.36%	0.76%

[a] The quantization axis is the magnetic easy axis of the pseudospin S = 1/2 representing the ground KD

Table S35 The decomposition of crystal field wave function of **14** (**TOFVAO**) in terms of $|m_J>$'s ($-15/2 \leq m_J \leq 15/2$) belonging to $^6H_{15/2}$ of Dy^{III} [a]

full	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	94.18%	0.60%	0.12%	0.44%	0.18%	3.35%	0.89%	0.25%
|±13/2>	0.00%	5.11%	59.48%	15.34%	5.86%	10.04%	2.97%	1.19%
|±11/2>	1.62%	1.86%	1.92%	8.51%	16.80%	38.88%	27.16%	3.24%
|±9/2>	2.50%	1.14%	1.03%	0.27%	3.46%	14.55%	53.68%	23.37%
|±7/2>	1.19%	1.32%	7.15%	3.26%	18.10%	25.80%	5.58%	37.61%
|±5/2>	0.10%	5.88%	14.20%	22.47%	22.30%	3.71%	8.11%	23.25%
|±3/2>	0.17%	31.71%	11.03%	21.48%	23.89%	2.88%	0.11%	8.73%
|±1/2>	0.25%	52.38%	5.07%	28.25%	9.42%	0.79%	1.49%	2.35%
Q	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	93.63%	0.10%	3.57%	1.99%	0.59%	0.10%	0.02%	0.00%
|±13/2>	0.00%	67.99%	9.68%	13.18%	7.00%	1.77%	0.32%	0.06%
|±11/2>	5.40%	0.25%	24.69%	32.18%	23.93%	10.46%	2.65%	0.45%
|±9/2>	0.00%	18.86%	0.13%	10.21%	30.80%	26.67%	11.06%	2.27%
|±7/2>	0.81%	0.62%	24.55%	4.60%	6.67%	29.56%	25.57%	7.63%
|±5/2>	0.00%	7.53%	4.50%	21.33%	5.71%	9.68%	33.24%	18.00%

|±3/2>	0.14%	1.42%	20.31%	1.73%	20.19%	2.94%	22.20%	31.08%
|±1/2>	0.03%	3.24%	12.58%	14.78%	5.11%	18.82%	4.94%	40.51%
Q1	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	85.64%	1.14%	7.58%	3.73%	0.95%	0.59%	0.32%	0.04%
|±13/2>	0.18%	58.19%	18.61%	13.61%	5.69%	2.55%	0.97%	0.21%
|±11/2>	9.31%	1.80%	18.78%	34.56%	24.77%	9.44%	0.79%	0.54%
|±9/2>	0.49%	16.85%	0.80%	9.70%	35.20%	28.72%	6.53%	1.70%
|±7/2>	2.89%	1.36%	16.80%	3.66%	9.57%	30.82%	28.07%	6.83%
|±5/2>	0.30%	10.05%	4.23%	14.61%	4.31%	14.54%	33.43%	18.53%
|±3/2>	0.86%	4.21%	17.90%	5.21%	14.98%	5.11%	19.73%	32.00%
|±1/2>	0.32%	6.39%	15.30%	14.91%	4.54%	8.23%	10.16%	40.15%
Q2	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
|±15/2>	98.78%	0.01%	0.01%	0.02%	0.78%	0.08%	0.25%	0.07%
|±13/2>	0.00%	76.82%	19.39%	0.81%	0.60%	0.42%	1.41%	0.55%
|±11/2>	0.99%	0.21%	0.28%	1.62%	58.30%	30.75%	4.48%	3.37%
|±9/2>	0.19%	1.99%	0.14%	0.04%	1.52%	29.87%	48.20%	18.05%
|±7/2>	0.03%	1.43%	0.48%	7.30%	29.44%	25.96%	1.06%	34.30%
|±5/2>	0.00%	3.27%	6.09%	49.43%	2.11%	0.44%	11.63%	27.03%
|±3/2>	0.00%	6.15%	31.08%	25.25%	1.34%	6.14%	17.79%	12.24%
|±1/2>	0.00%	10.12%	42.54%	15.52%	5.91%	6.35%	15.18%	4.40%

[a] The quantization axis is the magnetic easy axis of the pseudospin $S = 1/2$ representing the ground KD

Table S36 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of optimized structures of **1** (**WUHYOR**) [a]

gas	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	2.322E-05	7.412E-03	1.054E+00	8.140E-01	4.897E-01	1.851E-03	8.182E-01	1.261E+00
g_Y	1.047E-04	8.461E-03	1.076E+00	1.181E+00	8.554E-01	1.336E+00	2.123E+00	9.066E+00
g_Z	19.987	17.115	14.319	11.684	9.075	6.559	3.784	12.025
E (K)	0	378	519	578	635	708	782	829
solution	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	7.010E-05	9.173E-03	1.027E+00	7.758E-01	3.734E-01	7.788E-04	9.111E-01	1.261E+00
g_Y	1.402E-04	1.072E-02	1.056E+00	1.173E+00	7.699E-01	1.175E+00	2.064E+00	9.048E+00
g_Z	19.987	17.115	14.329	11.682	9.082	6.568	3.791	12.042
E (K)	0	379	523	581	639	712	785	832

[a] The optimizations were performed at BP86/def2-sv(p) plus D3BJ level. Dy atom was replace by Y to avoid possible problem in SCF convergence. **gas** means the optimization in gas phase; **solution** means the optimization in solution phase. The solution phase was accounted by implicit CPCM model with benzene as the solvent. ORCA 5.0.4 was used for the optimizations.

Table S37 *ab initio* calculated principal g values and energies (K) of the eight lowest KDs of optimized structures of **1a** [a]

gas	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	9.610E-07	2.239E-04	2.625E-02	1.336E-02	6.577E-01	2.933E-01	1.754E+00	1.240E+00
g_Y	2.135E-06	2.394E-04	2.685E-02	6.022E-02	7.211E-01	1.013E+00	2.448E+00	8.444E+00
g_Z	20.000	17.012	14.378	11.790	9.182	6.549	3.883	12.684
E (K)	0	800	1277	1542	1724	1873	1987	2052
solution	**KD_0**	**KD_1**	**KD_2**	**KD_3**	**KD_4**	**KD_5**	**KD_6**	**KD_7**
g_X	8.940E-07	1.858E-04	2.737E-02	7.996E-03	6.326E-01	2.472E-01	1.468E+00	1.250E+00
g_Y	2.067E-06	1.997E-04	2.797E-02	5.665E-02	6.898E-01	1.007E+00	2.214E+00	8.720E+00
g_Z	20.000	17.013	14.379	11.791	9.177	6.544	3.878	12.424
E (K)	0	798	1272	1536	1718	1866	1979	2043

[a] The optimizations were performed at BP86/def2-sv(p) plus D3BJ level. Dy atom was replace by Y to avoid possible problem in SCF convergence. **gas** means the optimization in gas phase; **solution** means the optimization in solution phase. The solution phase was accounted by implicit CPCM model with benzene as the solvent. ORCA 5.0.4 was used for the optimizations.

Table S38 The experimentally derived parameter values used for predicting T_B in **1a**

τ_{ref} (s)	τ_0 (s)	x_{Ra}
10.7	7.59E-11	2.11E+17

input file for MOLCAS calculation of **1**

&gateway

basis set

Dy.ANO-RCC-VTZP

Dy1	0.0000	0.0000	0.0000	Angstrom

end of basis

basis set

F.ANO-RCC-VDZP

F2	-0.4858	-1.3091	1.9438	Angstrom
F3	-0.0639	-2.2966	-0.7823	Angstrom
F4	2.0871	-1.0619	0.4700	Angstrom
F5	1.1029	1.1824	1.7232	Angstrom
F6	-1.5556	1.4528	1.1779	Angstrom
F7	-2.2643	-0.7811	-0.2711	Angstrom

end of basis

basis set

C.ANO-RCC-VDZP

C8	-0.6887	1.6131	-1.8697	Angstrom
C9	-0.3472	0.4060	-2.5035	Angstrom
C10	1.0419	0.2051	-2.3449	Angstrom
C11	1.5828	1.2770	-1.6147	Angstrom
C12	0.4848	2.1558	-1.3176	Angstrom

end of basis

basis set

C.ANO-RCC-VDZ

C13	-2.0322	2.2930	-1.9210	Angstrom
C14	-1.2675	-0.4747	-3.3563	Angstrom
C15	1.8584	-0.9112	-2.9531	Angstrom
C16	3.0205	1.4992	-1.3004	Angstrom
C17	0.5958	3.5213	-0.6718	Angstrom
C18	-0.3166	-1.5332	3.3194	Angstrom
C19	0.8206	-2.1956	3.6982	Angstrom
C20	1.0273	-2.3946	5.0190	Angstrom
C21	0.1437	-1.9407	5.9022	Angstrom
C22	-0.9757	-1.3091	5.5499	Angstrom
C23	-1.2606	-1.0986	4.1630	Angstrom
C24	-0.7994	-3.4800	-0.5645	Angstrom
C25	-1.8444	-3.7504	-1.3644	Angstrom
C26	-2.1717	-5.7397	-0.1473	Angstrom
C27	-1.1145	-5.4423	0.6374	Angstrom
C28	-0.3733	-4.2989	0.4460	Angstrom
C29	3.1413	-1.9658	0.7759	Angstrom
C30	3.2313	-3.0918	0.0368	Angstrom
C31	4.2151	-4.0034	0.3131	Angstrom

C32	5.1060	-3.6731	1.4261	Angstrom
C33	4.9717	-2.5355	2.0867	Angstrom
C34	3.9646	-1.5892	1.8065	Angstrom
C35	1.9423	1.6092	2.7141	Angstrom
C36	3.0967	2.3161	2.3962	Angstrom
C37	3.9454	2.7526	3.4067	Angstrom
C38	3.6396	2.4822	4.7355	Angstrom
C39	2.4851	1.7753	5.0538	Angstrom
C40	-2.5490	-4.8976	-1.1322	Angstrom
C41	1.6365	1.3389	4.0433	Angstrom
C42	-2.0599	2.1307	2.3133	Angstrom
C43	-1.3658	3.1505	2.8274	Angstrom
C44	-1.8686	3.7840	3.9444	Angstrom
C45	-3.0617	3.3861	4.4609	Angstrom
C46	-3.7720	2.3490	3.8803	Angstrom
C47	-3.2633	1.6962	2.7577	Angstrom
C48	-3.5307	-1.0117	0.3707	Angstrom
C49	-3.5809	-2.0005	1.2628	Angstrom
C50	-4.8219	-2.1821	1.9146	Angstrom
C51	-5.8803	-1.3941	1.5943	Angstrom
C52	-5.7639	-0.4342	0.6734	Angstrom
C53	-4.5474	-0.1812	-0.0112	Angstrom

end of basis

basis set

H.ANO-RCC-VDZ

H54	-2.2059	2.7399	-1.0675	Angstrom
H55	-2.7307	1.6249	-2.0883	Angstrom
H56	-2.0357	2.9549	-2.6441	Angstrom
H57	-2.1677	-0.4779	-2.9696	Angstrom
H58	-0.9170	-1.3904	-3.3751	Angstrom
H59	-1.3046	-0.1208	-4.2691	Angstrom
H60	1.2908	-1.6999	-3.0785	Angstrom
H61	2.6018	-1.1347	-2.3553	Angstrom
H62	2.2119	-0.6217	-3.8202	Angstrom
H63	3.3385	0.7863	-0.7075	Angstrom
H64	3.1281	2.3657	-0.8566	Angstrom
H65	3.5411	1.4897	-2.1306	Angstrom
H66	1.1340	3.4548	0.1429	Angstrom
H67	-0.3003	3.8496	-0.4471	Angstrom
H68	1.0233	4.1438	-1.2973	Angstrom
H69	1.4444	-2.5048	3.0525	Angstrom
H70	1.8011	-2.8592	5.3152	Angstrom
H71	0.3193	-2.0716	6.8262	Angstrom
H72	-1.5818	-1.0029	6.2159	Angstrom

H73 -2.0595 -0.6818 3.8638 Angstrom
H74 -2.0850 -3.1621 -2.0699 Angstrom
H75 -3.3083 -5.1031 -1.6664 Angstrom
H76 -2.6521 -6.5479 -0.0074 Angstrom
H77 -0.8761 -6.0354 1.3401 Angstrom
H78 0.3855 -4.0918 0.9800 Angstrom
H79 2.6158 -3.2510 -0.6692 Angstrom
H80 4.3130 -4.8025 -0.1914 Angstrom
H81 5.7888 -4.2839 1.6759 Angstrom
H82 5.5861 -2.3470 2.7855 Angstrom
H83 3.8677 -0.7722 2.2808 Angstrom
H84 3.3057 2.4996 1.4878 Angstrom
H85 4.7339 3.2350 3.1893 Angstrom
H86 4.2192 2.7812 5.4264 Angstrom
H87 2.2765 1.5919 5.9621 Angstrom
H88 0.8482 0.8565 4.2606 Angstrom
H89 -0.5482 3.4318 2.4345 Angstrom
H90 -1.3851 4.4951 4.3485 Angstrom
H91 -3.4125 3.8240 5.2278 Angstrom
H92 -4.6067 2.0810 4.2464 Angstrom
H93 -3.7310 0.9879 2.3311 Angstrom
H94 -2.8288 -2.5496 1.4493 Angstrom
H95 -4.9158 -2.8582 2.5754 Angstrom
H96 -6.7125 -1.5251 2.0346 Angstrom
H97 -6.5249 0.0967 0.4699 Angstrom
H98 -4.4502 0.4971 -0.6670 Angstrom
end of basis
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Threshold = 1.0d-11
Cutoff = 1.0d-11
&rasscf
lumorb
* the following is for CAS_9e_7o_21S6_RASSI_SINGLE_ANISO calculation of Dy SIM
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Nactel = 9 0 0
ras2 = 7
Inactive = 215
* Inactive is system-specific and should be input manually
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Thrs
1.0e-07,1.0e-03,1.0e-04

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>>> COPY    JOB_21S6_CAS_9e_7o    JOB001
&rassi
 Nr of JobIph
 1 21
 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21
 spin
 MEES
 PROP
 3
  'ANGMOM' 1
  'ANGMOM' 2
  'ANGMOM' 3
&SINGLE_ANISO
 mltp
 8
 2 2 2 2 2 2 2 2
 CRYS
 DY
 UBAR
 >>> RM JOB001
```

input file for MOLCAS calculation of $\mathbf{1}_Q$

```
&gateway
basis set
Dy.ANO-RCC-VTZP
Dy1        0.00000     0.00000        0.00000       Angstrom
end of basis
Xfield
97 Angstrom 0
  -0.4858    -1.3091    1.9438    -0.4214    0.    0.    0.
  -0.0639    -2.2966    -0.7823   -0.4146    0.    0.    0.
  2.0871     -1.0619    0.4700    -0.4359    0.    0.    0.
  1.1029     1.1824     1.7232    -0.3925    0.    0.    0.
  -1.5556    1.4528     1.1779    -0.4254    0.    0.    0.
  -2.2643    -0.7811    -0.2711   -0.4388    0.    0.    0.
  -0.6887    1.6131     -1.8697   -0.1573    0.    0.    0.
  -0.3472    0.4060     -2.5035   -0.1721    0.    0.    0.
  1.0419     0.2051     -2.3449   -0.1532    0.    0.    0.
  1.5828     1.2770     -1.6147   -0.1572    0.    0.    0.
  0.4848     2.1558     -1.3176   -0.1734    0.    0.    0.
```

-2.0322	2.2930	-1.9210	-0.3687	0.	0.	0.
-1.2675	-0.4747	-3.3563	-0.3686	0.	0.	0.
1.8584	-0.9112	-2.9531	-0.3716	0.	0.	0.
3.0205	1.4992	-1.3004	-0.3721	0.	0.	0.
0.5958	3.5213	-0.6718	-0.3712	0.	0.	0.
-0.3166	-1.5332	3.3194	0.2708	0.	0.	0.
0.8206	-2.1956	3.6982	-0.1618	0.	0.	0.
1.0273	-2.3946	5.0190	-0.0889	0.	0.	0.
0.1437	-1.9407	5.9022	-0.1169	0.	0.	0.
-0.9757	-1.3091	5.5499	-0.0958	0.	0.	0.
-1.2606	-1.0986	4.1630	-0.1625	0.	0.	0.
-0.7994	-3.4800	-0.5645	0.2750	0.	0.	0.
-1.8444	-3.7504	-1.3644	-0.1542	0.	0.	0.
-2.1717	-5.7397	-0.1473	-0.1143	0.	0.	0.
-1.1145	-5.4423	0.6374	-0.1010	0.	0.	0.
-0.3733	-4.2989	0.4460	-0.1663	0.	0.	0.
3.1413	-1.9658	0.7759	0.2780	0.	0.	0.
3.2313	-3.0918	0.0368	-0.1702	0.	0.	0.
4.2151	-4.0034	0.3131	-0.0839	0.	0.	0.
5.1060	-3.6731	1.4261	-0.1126	0.	0.	0.
4.9717	-2.5355	2.0867	-0.1059	0.	0.	0.
3.9646	-1.5892	1.8065	-0.1524	0.	0.	0.
1.9423	1.6092	2.7141	0.2548	0.	0.	0.
3.0967	2.3161	2.3962	-0.1598	0.	0.	0.
3.9454	2.7526	3.4067	-0.1004	0.	0.	0.
3.6396	2.4822	4.7355	-0.1058	0.	0.	0.
2.4851	1.7753	5.0538	-0.1053	0.	0.	0.
-2.5490	-4.8976	-1.1322	-0.0904	0.	0.	0.
1.6365	1.3389	4.0433	-0.1601	0.	0.	0.
-2.0599	2.1307	2.3133	0.2807	0.	0.	0.
-1.3658	3.1505	2.8274	-0.1590	0.	0.	0.
-1.8686	3.7840	3.9444	-0.0926	0.	0.	0.
-3.0617	3.3861	4.4609	-0.1139	0.	0.	0.
-3.7720	2.3490	3.8803	-0.0998	0.	0.	0.
-3.2633	1.6962	2.7577	-0.1635	0.	0.	0.
-3.5307	-1.0117	0.3707	0.2854	0.	0.	0.
-3.5809	-2.0005	1.2628	-0.1696	0.	0.	0.
-4.8219	-2.1821	1.9146	-0.0925	0.	0.	0.
-5.8803	-1.3941	1.5943	-0.1214	0.	0.	0.
-5.7639	-0.4342	0.6734	-0.0914	0.	0.	0.
-4.5474	-0.1812	-0.0112	-0.1557	0.	0.	0.
-2.2059	2.7399	-1.0675	0.1235	0.	0.	0.
-2.7307	1.6249	-2.0883	0.1201	0.	0.	0.
-2.0357	2.9549	-2.6441	0.1410	0.	0.	0.

-2.1677	-0.4779	-2.9696	0.1218	0.	0.	0.
-0.9170	-1.3904	-3.3751	0.1220	0.	0.	0.
-1.3046	-0.1208	-4.2691	0.1402	0.	0.	0.
1.2908	-1.6999	-3.0785	0.1250	0.	0.	0.
2.6018	-1.1347	-2.3553	0.1197	0.	0.	0.
2.2119	-0.6217	-3.8202	0.1406	0.	0.	0.
3.3385	0.7863	-0.7075	0.1202	0.	0.	0.
3.1281	2.3657	-0.8566	0.1245	0.	0.	0.
3.5411	1.4897	-2.1306	0.1419	0.	0.	0.
1.1340	3.4548	0.1429	0.1147	0.	0.	0.
-0.3003	3.8496	-0.4471	0.1263	0.	0.	0.
1.0233	4.1438	-1.2973	0.1403	0.	0.	0.
1.4444	-2.5048	3.0525	0.1438	0.	0.	0.
1.8011	-2.8592	5.3152	0.1459	0.	0.	0.
0.3193	-2.0716	6.8262	0.1453	0.	0.	0.
-1.5818	-1.0029	6.2159	0.1469	0.	0.	0.
-2.0595	-0.6818	3.8638	0.1488	0.	0.	0.
-2.0850	-3.1621	-2.0699	0.1494	0.	0.	0.
-3.3083	-5.1031	-1.6664	0.1461	0.	0.	0.
-2.6521	-6.5479	-0.0074	0.1453	0.	0.	0.
-0.8761	-6.0354	1.3401	0.1427	0.	0.	0.
0.3855	-4.0918	0.9800	0.1407	0.	0.	0.
2.6158	-3.2510	-0.6692	0.1498	0.	0.	0.
4.3130	-4.8025	-0.1914	0.1505	0.	0.	0.
5.7888	-4.2839	1.6759	0.1475	0.	0.	0.
5.5861	-2.3470	2.7855	0.1449	0.	0.	0.
3.8677	-0.7722	2.2808	0.1499	0.	0.	0.
3.3057	2.4996	1.4878	0.1440	0.	0.	0.
4.7339	3.2350	3.1893	0.1483	0.	0.	0.
4.2192	2.7812	5.4264	0.1463	0.	0.	0.
2.2765	1.5919	5.9621	0.1445	0.	0.	0.
0.8482	0.8565	4.2606	0.1403	0.	0.	0.
-0.5482	3.4318	2.4345	0.1471	0.	0.	0.
-1.3851	4.4951	4.3485	0.1465	0.	0.	0.
-3.4125	3.8240	5.2278	0.1455	0.	0.	0.
-4.6067	2.0810	4.2464	0.1442	0.	0.	0.
-3.7310	0.9879	2.3311	0.1457	0.	0.	0.
-2.8288	-2.5496	1.4493	0.1501	0.	0.	0.
-4.9158	-2.8582	2.5754	0.1450	0.	0.	0.
-6.7125	-1.5251	2.0346	0.1472	0.	0.	0.
-6.5249	0.0967	0.4699	0.1476	0.	0.	0.
-4.4502	0.4971	-0.6670	0.1544	0.	0.	0.

angm

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&seward
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   Threshold = 1.0d-11
   Cutoff = 1.0d-11
&rasscf
 lumorb
* the following is for CAS_9e_7o_21S6_RASSI_SINGLE_ANISO calculation of Dy SIM
 spin = 6
 Nactel = 9 0 0
 ras2 = 7
 Inactive = 27
* Inactive is system-specific and should be input manually
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 Thrs
 1.0e-07,1.0e-03,1.0e-04
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>>> COPY    JOB_21S6_CAS_9e_7o    JOB001
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 Nr of JobIph
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 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21
 spin
 MEES
 PROP
 3
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  'ANGMOM' 2
  'ANGMOM' 3
&SINGLE_ANISO
 mltp
 8
 2 2 2 2 2 2 2 2
 CRYS
 DY
 UBAR
 >>> RM JOB001
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input file for MOLCAS calculation of $\mathbf{1}_{Q1}$

```
&gateway
basis set
```

Dy.ANO-RCC-VTZP

Dy1	0.0000	0.0000	0.0000	Angstrom

end of basis

basis set

F.ANO-RCC-VDZP

F2	-0.4858	-1.3091	1.9438	Angstrom
F3	-0.0639	-2.2966	-0.7823	Angstrom
F4	2.0871	-1.0619	0.4700	Angstrom
F5	1.1029	1.1824	1.7232	Angstrom
F6	-1.5556	1.4528	1.1779	Angstrom
F7	-2.2643	-0.7811	-0.2711	Angstrom

end of basis

basis set

C.ANO-RCC-VDZ

C13	-2.0322	2.2930	-1.9210	Angstrom
C14	-1.2675	-0.4747	-3.3563	Angstrom
C15	1.8584	-0.9112	-2.9531	Angstrom
C16	3.0205	1.4992	-1.3004	Angstrom
C17	0.5958	3.5213	-0.6718	Angstrom
C18	-0.3166	-1.5332	3.3194	Angstrom
C19	0.8206	-2.1956	3.6982	Angstrom
C20	1.0273	-2.3946	5.0190	Angstrom
C21	0.1437	-1.9407	5.9022	Angstrom
C22	-0.9757	-1.3091	5.5499	Angstrom
C23	-1.2606	-1.0986	4.1630	Angstrom
C24	-0.7994	-3.4800	-0.5645	Angstrom
C25	-1.8444	-3.7504	-1.3644	Angstrom
C26	-2.1717	-5.7397	-0.1473	Angstrom
C27	-1.1145	-5.4423	0.6374	Angstrom
C28	-0.3733	-4.2989	0.4460	Angstrom
C29	3.1413	-1.9658	0.7759	Angstrom
C30	3.2313	-3.0918	0.0368	Angstrom
C31	4.2151	-4.0034	0.3131	Angstrom
C32	5.1060	-3.6731	1.4261	Angstrom
C33	4.9717	-2.5355	2.0867	Angstrom
C34	3.9646	-1.5892	1.8065	Angstrom
C35	1.9423	1.6092	2.7141	Angstrom
C36	3.0967	2.3161	2.3962	Angstrom
C37	3.9454	2.7526	3.4067	Angstrom
C38	3.6396	2.4822	4.7355	Angstrom
C39	2.4851	1.7753	5.0538	Angstrom
C40	-2.5490	-4.8976	-1.1322	Angstrom
C41	1.6365	1.3389	4.0433	Angstrom
C42	-2.0599	2.1307	2.3133	Angstrom

C43	-1.3658	3.1505	2.8274	Angstrom
C44	-1.8686	3.7840	3.9444	Angstrom
C45	-3.0617	3.3861	4.4609	Angstrom
C46	-3.7720	2.3490	3.8803	Angstrom
C47	-3.2633	1.6962	2.7577	Angstrom
C48	-3.5307	-1.0117	0.3707	Angstrom
C49	-3.5809	-2.0005	1.2628	Angstrom
C50	-4.8219	-2.1821	1.9146	Angstrom
C51	-5.8803	-1.3941	1.5943	Angstrom
C52	-5.7639	-0.4342	0.6734	Angstrom
C53	-4.5474	-0.1812	-0.0112	Angstrom

end of basis

basis set

H.ANO-RCC-VDZ

H54	-2.2059	2.7399	-1.0675	Angstrom
H55	-2.7307	1.6249	-2.0883	Angstrom
H56	-2.0357	2.9549	-2.6441	Angstrom
H57	-2.1677	-0.4779	-2.9696	Angstrom
H58	-0.9170	-1.3904	-3.3751	Angstrom
H59	-1.3046	-0.1208	-4.2691	Angstrom
H60	1.2908	-1.6999	-3.0785	Angstrom
H61	2.6018	-1.1347	-2.3553	Angstrom
H62	2.2119	-0.6217	-3.8202	Angstrom
H63	3.3385	0.7863	-0.7075	Angstrom
H64	3.1281	2.3657	-0.8566	Angstrom
H65	3.5411	1.4897	-2.1306	Angstrom
H66	1.1340	3.4548	0.1429	Angstrom
H67	-0.3003	3.8496	-0.4471	Angstrom
H68	1.0233	4.1438	-1.2973	Angstrom
H69	1.4444	-2.5048	3.0525	Angstrom
H70	1.8011	-2.8592	5.3152	Angstrom
H71	0.3193	-2.0716	6.8262	Angstrom
H72	-1.5818	-1.0029	6.2159	Angstrom
H73	-2.0595	-0.6818	3.8638	Angstrom
H74	-2.0850	-3.1621	-2.0699	Angstrom
H75	-3.3083	-5.1031	-1.6664	Angstrom
H76	-2.6521	-6.5479	-0.0074	Angstrom
H77	-0.8761	-6.0354	1.3401	Angstrom
H78	0.3855	-4.0918	0.9800	Angstrom
H79	2.6158	-3.2510	-0.6692	Angstrom
H80	4.3130	-4.8025	-0.1914	Angstrom
H81	5.7888	-4.2839	1.6759	Angstrom
H82	5.5861	-2.3470	2.7855	Angstrom
H83	3.8677	-0.7722	2.2808	Angstrom

H84	3.3057	2.4996	1.4878	Angstrom
H85	4.7339	3.2350	3.1893	Angstrom
H86	4.2192	2.7812	5.4264	Angstrom
H87	2.2765	1.5919	5.9621	Angstrom
H88	0.8482	0.8565	4.2606	Angstrom
H89	-0.5482	3.4318	2.4345	Angstrom
H90	-1.3851	4.4951	4.3485	Angstrom
H91	-3.4125	3.8240	5.2278	Angstrom
H92	-4.6067	2.0810	4.2464	Angstrom
H93	-3.7310	0.9879	2.3311	Angstrom
H94	-2.8288	-2.5496	1.4493	Angstrom
H95	-4.9158	-2.8582	2.5754	Angstrom
H96	-6.7125	-1.5251	2.0346	Angstrom
H97	-6.5249	0.0967	0.4699	Angstrom
H98	-4.4502	0.4971	-0.6670	Angstrom

end of basis

Xfield

25 Angstrom 0

-0.6887	1.6131	-1.8697	-0.1782	0.	0.	0.
-0.3472	0.4060	-2.5035	-0.1930	0.	0.	0.
1.0419	0.2051	-2.3449	-0.1741	0.	0.	0.
1.5828	1.2770	-1.6147	-0.1781	0.	0.	0.
0.4848	2.1558	-1.3176	-0.1943	0.	0.	0.
-2.0322	2.2930	-1.9210	-0.3902	0.	0.	0.
-1.2675	-0.4747	-3.3563	-0.3901	0.	0.	0.
1.8584	-0.9112	-2.9531	-0.3931	0.	0.	0.
3.0205	1.4992	-1.3004	-0.3937	0.	0.	0.
0.5958	3.5213	-0.6718	-0.3927	0.	0.	0.
-2.2059	2.7399	-1.0675	0.1205	0.	0.	0.
-2.7307	1.6249	-2.0883	0.1171	0.	0.	0.
-2.0357	2.9549	-2.6441	0.1381	0.	0.	0.
-2.1677	-0.4779	-2.9696	0.1188	0.	0.	0.
-0.9170	-1.3904	-3.3751	0.1190	0.	0.	0.
-1.3046	-0.1208	-4.2691	0.1373	0.	0.	0.
1.2908	-1.6999	-3.0785	0.1220	0.	0.	0.
2.6018	-1.1347	-2.3553	0.1167	0.	0.	0.
2.2119	-0.6217	-3.8202	0.1377	0.	0.	0.
3.3385	0.7863	-0.7075	0.1172	0.	0.	0.
3.1281	2.3657	-0.8566	0.1215	0.	0.	0.
3.5411	1.4897	-2.1306	0.1390	0.	0.	0.
1.1340	3.4548	0.1429	0.1117	0.	0.	0.
-0.3003	3.8496	-0.4471	0.1233	0.	0.	0.
1.0233	4.1438	-1.2973	0.1374	0.	0.	0.

angm

```
0.000 0.000 0.000
&seward
   medium cholesky
   Threshold = 1.0d-11
   Cutoff = 1.0d-11
&rasscf
 lumorb
* the following is for CAS_9e_7o_21S6_RASSI_SINGLE_ANISO calculation of Dy SIM
 spin = 6
 Nactel = 9 0 0
 ras2 = 7
 Inactive = 177
* Inactive is system-specific and should be input manually
 ciroot = 21,21,1
 Thrs
 1.0e-07,1.0e-03,1.0e-04
>>> COPY $Project.JobIph JOB_21S6_CAS_9e_7o
>>> COPY    JOB_21S6_CAS_9e_7o    JOB001
&rassi
 Nr of JobIph
 1 21
 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21
 spin
 MEES
 PROP
 3
  'ANGMOM' 1
  'ANGMOM' 2
  'ANGMOM' 3
&SINGLE_ANISO
 mltp
 8
 2 2 2 2 2 2 2 2
 CRYS
 DY
 UBAR
 >>> RM JOB001
```

input file for MOLCAS calculation of $\mathbf{1}_{Q2}$

```
&gateway
basis set
```

Dy.ANO-RCC-VTZP

Dy1	0.0000	0.0000	0.0000	Angstrom

end of basis

basis set

C.ANO-RCC-VDZP

C8	-0.6887	1.6131	-1.8697	Angstrom
C9	-0.3472	0.4060	-2.5035	Angstrom
C10	1.0419	0.2051	-2.3449	Angstrom
C11	1.5828	1.2770	-1.6147	Angstrom
C12	0.4848	2.1558	-1.3176	Angstrom

end of basis

basis set

C.ANO-RCC-VDZ

C13	-2.0322	2.2930	-1.9210	Angstrom
C14	-1.2675	-0.4747	-3.3563	Angstrom
C15	1.8584	-0.9112	-2.9531	Angstrom
C16	3.0205	1.4992	-1.3004	Angstrom
C17	0.5958	3.5213	-0.6718	Angstrom

end of basis

basis set

H.ANO-RCC-VDZ

H54	-2.2059	2.7399	-1.0675	Angstrom
H55	-2.7307	1.6249	-2.0883	Angstrom
H56	-2.0357	2.9549	-2.6441	Angstrom
H57	-2.1677	-0.4779	-2.9696	Angstrom
H58	-0.9170	-1.3904	-3.3751	Angstrom
H59	-1.3046	-0.1208	-4.2691	Angstrom
H60	1.2908	-1.6999	-3.0785	Angstrom
H61	2.6018	-1.1347	-2.3553	Angstrom
H62	2.2119	-0.6217	-3.8202	Angstrom
H63	3.3385	0.7863	-0.7075	Angstrom
H64	3.1281	2.3657	-0.8566	Angstrom
H65	3.5411	1.4897	-2.1306	Angstrom
H66	1.1340	3.4548	0.1429	Angstrom
H67	-0.3003	3.8496	-0.4471	Angstrom
H68	1.0233	4.1438	-1.2973	Angstrom

Xfield

72 Angstrom 0

-0.4858	-1.3091	1.9438	-0.4133	0.	0.	0.
-0.0639	-2.2966	-0.7823	-0.4065	0.	0.	0.
2.0871	-1.0619	0.4700	-0.4278	0.	0.	0.
1.1029	1.1824	1.7232	-0.3845	0.	0.	0.
-1.5556	1.4528	1.1779	-0.4173	0.	0.	0.
-2.2643	-0.7811	-0.2711	-0.4307	0.	0.	0.

-0.3166	-1.5332	3.3194	0.2757	0.	0.	0.
0.8206	-2.1956	3.6982	-0.1566	0.	0.	0.
1.0273	-2.3946	5.0190	-0.0837	0.	0.	0.
0.1437	-1.9407	5.9022	-0.1117	0.	0.	0.
-0.9757	-1.3091	5.5499	-0.0905	0.	0.	0.
-1.2606	-1.0986	4.1630	-0.1572	0.	0.	0.
-0.7994	-3.4800	-0.5645	0.2799	0.	0.	0.
-1.8444	-3.7504	-1.3644	-0.1490	0.	0.	0.
-2.1717	-5.7397	-0.1473	-0.1091	0.	0.	0.
-1.1145	-5.4423	0.6374	-0.0957	0.	0.	0.
-0.3733	-4.2989	0.4460	-0.1610	0.	0.	0.
3.1413	-1.9658	0.7759	0.2829	0.	0.	0.
3.2313	-3.0918	0.0368	-0.1649	0.	0.	0.
4.2151	-4.0034	0.3131	-0.0787	0.	0.	0.
5.1060	-3.6731	1.4261	-0.1074	0.	0.	0.
4.9717	-2.5355	2.0867	-0.1006	0.	0.	0.
3.9646	-1.5892	1.8065	-0.1472	0.	0.	0.
1.9423	1.6092	2.7141	0.2597	0.	0.	0.
3.0967	2.3161	2.3962	-0.1546	0.	0.	0.
3.9454	2.7526	3.4067	-0.0951	0.	0.	0.
3.6396	2.4822	4.7355	-0.1005	0.	0.	0.
2.4851	1.7753	5.0538	-0.1000	0.	0.	0.
-2.5490	-4.8976	-1.1322	-0.0852	0.	0.	0.
1.6365	1.3389	4.0433	-0.1549	0.	0.	0.
-2.0599	2.1307	2.3133	0.2856	0.	0.	0.
-1.3658	3.1505	2.8274	-0.1538	0.	0.	0.
-1.8686	3.7840	3.9444	-0.0874	0.	0.	0.
-3.0617	3.3861	4.4609	-0.1087	0.	0.	0.
-3.7720	2.3490	3.8803	-0.0945	0.	0.	0.
-3.2633	1.6962	2.7577	-0.1582	0.	0.	0.
-3.5307	-1.0117	0.3707	0.2903	0.	0.	0.
-3.5809	-2.0005	1.2628	-0.1643	0.	0.	0.
-4.8219	-2.1821	1.9146	-0.0873	0.	0.	0.
-5.8803	-1.3941	1.5943	-0.1162	0.	0.	0.
-5.7639	-0.4342	0.6734	-0.0862	0.	0.	0.
-4.5474	-0.1812	-0.0112	-0.1505	0.	0.	0.
1.4444	-2.5048	3.0525	0.1446	0.	0.	0.
1.8011	-2.8592	5.3152	0.1467	0.	0.	0.
0.3193	-2.0716	6.8262	0.1461	0.	0.	0.
-1.5818	-1.0029	6.2159	0.1477	0.	0.	0.
-2.0595	-0.6818	3.8638	0.1496	0.	0.	0.
-2.0850	-3.1621	-2.0699	0.1502	0.	0.	0.
-3.3083	-5.1031	-1.6664	0.1469	0.	0.	0.
-2.6521	-6.5479	-0.0074	0.1461	0.	0.	0.

```
  -0.8761   -6.0354   1.3401   0.1435   0.   0.   0.
  0.3855    -4.0918   0.9800   0.1415   0.   0.   0.
  2.6158    -3.2510   -0.6692  0.1506   0.   0.   0.
  4.3130    -4.8025   -0.1914  0.1513   0.   0.   0.
  5.7888    -4.2839   1.6759   0.1483   0.   0.   0.
  5.5861    -2.3470   2.7855   0.1457   0.   0.   0.
  3.8677    -0.7722   2.2808   0.1507   0.   0.   0.
  3.3057    2.4996    1.4878   0.1448   0.   0.   0.
  4.7339    3.2350    3.1893   0.1491   0.   0.   0.
  4.2192    2.7812    5.4264   0.1471   0.   0.   0.
  2.2765    1.5919    5.9621   0.1453   0.   0.   0.
  0.8482    0.8565    4.2606   0.1411   0.   0.   0.
  -0.5482   3.4318    2.4345   0.1479   0.   0.   0.
  -1.3851   4.4951    4.3485   0.1473   0.   0.   0.
  -3.4125   3.8240    5.2278   0.1463   0.   0.   0.
  -4.6067   2.0810    4.2464   0.1450   0.   0.   0.
  -3.7310   0.9879    2.3311   0.1465   0.   0.   0.
  -2.8288   -2.5496   1.4493   0.1509   0.   0.   0.
  -4.9158   -2.8582   2.5754   0.1458   0.   0.   0.
  -6.7125   -1.5251   2.0346   0.1480   0.   0.   0.
  -6.5249   0.0967    0.4699   0.1484   0.   0.   0.
  -4.4502   0.4971    -0.6670  0.1552   0.   0.   0.
angm
0.000 0.000 0.000
&seward
   medium cholesky
   Threshold = 1.0d-11
   Cutoff = 1.0d-11
&rasscf
  lumorb
* the following is for CAS_9e_7o_21S6_RASSI_SINGLE_ANISO calculation of Dy SIM
  spin = 6
  Nactel = 9 0 0
  ras2 = 7
  Inactive = 65
* Inactive is system-specific and should be input manually
  ciroot = 21,21,1
  Thrs
  1.0e-07,1.0e-03,1.0e-04
>>> COPY $Project.JobIph JOB_21S6_CAS_9e_7o
>>> COPY    JOB_21S6_CAS_9e_7o    JOB001
&rassi
  Nr of JobIph
  1 21
```

```
 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21
 spin
 MEES
 PROP
 3
  'ANGMOM' 1
  'ANGMOM' 2
  'ANGMOM' 3
&SINGLE_ANISO
 mltp
 8
 2 2 2 2 2 2 2 2
 CRYS
 DY
 UBAR
 >>> RM JOB001
```