

Energies of Ln^{3+} ions calculated using different approximations and spin-orbit coupling values denoted by color intensity

Basis sets:

GUESS is for ECP28MWB_GUESS (12s11p9d8f)/[5s5p4d3f], SEG is for ECP28MWB_SEG (14s13p10d8f6g)/[10s8p5d4f3g]; SEG is used unless otherwise stated

SO-CI is for Spin-Orbit CASSCF, SO-QDPT is for Spin-Orbit XMCQDPT2; SO-CI is used unless otherwise stated

4f is CASSCF and XMCQDPT2 with only 4f orbitals; 4f5f and 4f5d are for CASSCF and XMCQDPT2 with 4f and 5f or 5d orbitals, respectively; 4f is used unless otherwise stated

Table S1

$^M\text{L}_j$	Exp.	Calc. Ce^{3+} SEG basis set
		7 doublets
		^2F
$^2\text{F}_{7/2}$	2166	2292

Table S2

$^M\text{L}_j$	Exp.	Calc. Yb^{3+} SEG basis set
		7 doublets
		^2F
$^2\text{F}_{5/2}$	10282	10180

Table S3

$^M\text{L}_j$	Exp.	Calc. Pr^{3+}	
		GUESS basis set	SEG basis set
		21 triplets 28 singlets	21 triplets 28 singlets
		$^3\text{H } ^3\text{F } ^3\text{P } ^1\text{G } ^1\text{D } ^1\text{I } ^1\text{S}$	
$^3\text{H}_5$	2119	2000	2176
$^3\text{H}_6$	4307	4088	4446
$^3\text{F}_2$	4848	4826	4877
$^3\text{F}_3$	6248	6121	6324
$^3\text{F}_4$	6684	6426	6815
$^1\text{G}_4$	9704	9217	9952
$^1\text{D}_2$	16641	17646	17034
$^3\text{P}_0$	20385	22643	21502
$^3\text{P}_1$	21987	23200	22129
$^1\text{I}_6$	21327	24848	23120
$^3\text{P}_2$	22142	24227	23306
$^1\text{S}_0$	48710	50189	48722

Table S4

$^M\text{L}_j$	Exp.	Calc. Tm^{3+} SEG basis set	
		4f 21 triplets 28 singlets	4f5f 21 triplets 28 singlets
		$^3\text{H } ^3\text{F } ^3\text{P } ^1\text{G } ^1\text{D } ^1\text{I } ^1\text{S}$	
$^3\text{H}_4$	5765	5453	5506
$^3\text{H}_5$	8230	8540	8550
$^3\text{F}_4$	12617	12863	12894
$^3\text{F}_3$	14449	14746	14593
$^3\text{F}_2$	15051	15462	15328
$^1\text{G}_4$	21115	21415	21664
$^1\text{D}_2$	27701	29259	28934
$^1\text{I}_6$	34117	38594	38549
$^3\text{P}_0$	35496	38043	37222
$^3\text{P}_1$	35873	38930	38114
$^3\text{P}_2$	37484	40090	39574

Table S5

M_{L_j}	Exp.	Calc. Nd ³⁺				
		GUESS basis set SO-CI 30 quartets 11 doublets	SEG basis set SO-CI 30 quartets 11 doublets	SEG basis set SO-CI 30 quartets 56 doublets	SEG basis set SO-CI 35 quartets 56 doublets	SEG basis set SO-QDPT 35 quartets 56 doublets
		$^4G\ ^4F\ ^4S\ ^4I\ ^2H$		$^4G\ ^4F\ ^4S\ ^4I\ ^2H$ $^2G\ ^2K\ ^2D\ ^2P\ ^2I$	$^4G\ ^4F\ ^4S\ ^4I\ ^4D\ ^2H\ ^2G\ ^2K\ ^2D\ ^2P\ ^2I$	
$^4I_{11/2}$	1880	1765	1913	1925	1918	1912
$^4I_{13/2}$	3860	3713	4018	3969	3956	3942
$^4I_{15/2}$	5910	5796	6262	6085	6068	6043
$^4F_{3/2}$	11290	12362	11744	11298	11202	10794
$^4F_{5/2}$	12320	13008	12482	12322	12246	11834
$^2H_{9/2}$	12470	13655	14164	12436	12444	12154
$^4F_{7/2}$	13280	13971	13519	13481	13254	12839
$^4S_{3/2}$	13370	14068	13758	13044	13483	12990
$^4F_{9/2}$	14570	15335	15107	14729	14558	14190
$^2H_{11/2}$	15800	15600	16170	15817	15894	15658
$^4G_{5/2}$	16980	19153	17678	17694	17700	17188
$^2G_{7/2}$	17100	20110	18709	17231	17270	16842
$^4G_{7/2}$	18890	21399	20121	19352	19374	18893

Nd	4I	4S	4F	4G	4D	$^2H(1)$	2G	2K	2P	$^2D(1)$	2I	2L	$^2D(2)$	$^2H(2)$	2F
4I	■					■		■							
4S									■						
4F			■				■			■				■	■
4G				■		■	■							■	■
4D					■					■			■		■
$^2H(1)$	■			■		■	■					■		■	
2G			■	■		■	■							■	■
2K	■							■				■	■		
2P		■			■				■						
$^2D(1)$			■		■			■		■			■		■
2I	■					■		■			■			■	
2L							■					■			
$^2D(2)$			■		■				■				■		■
$^2H(2)$	■			■						■				■	
2F			■	■	■		■						■		■

Figure S1

Table S6

$^M L_J$	Exp.	Calc. Er ³⁺ SEG basis set			
		SO-CI 13 quartets 11 doublets	SO-CI 30 quartets 56 doublets	SO-CI 35 quartets 56 doublets	SO-QDPT 35 quartets 56 doublets
		$^4 I \ ^2 H$	$^4 I \ ^4 F \ ^4 S \ ^4 G \ ^2 H \ ^2 G \ ^2 K \ ^2 D \ ^2 P \ ^2 I$	$^4 I \ ^4 F \ ^4 S \ ^4 G \ ^4 D \ ^2 H \ ^2 G \ ^2 K \ ^2 D \ ^2 P \ ^2 I$	
$^4 I_{13/2}$	6482	6184	6674	6672	6675
$^4 I_{11/2}$	10111	9993	10516	10528	10578
$^4 I_{9/2}$	12352	13163	12709	12814	12915
$^4 F_{9/2}$	15176	-	15614	15982	16025
$^4 S_{3/2}$	18291	-	19149	18253	18323
$^2 H_{11/2}$	19000	21598	20331	20387	20577
$^4 F_{7/2}$	20373	-	21022	21580	21611
$^4 F_{5/2}$	22068	-	22946	23629	23674
$^4 F_{3/2}$	22409	-	23474	23190	23267
$^2 H_{9/2}$	24434	29827	25277	25441	25622
$^4 G_{11/2}$	26271	-	27978	28002	28090
$^2 G_{9/2}$	27238	-	29227	29243	29301
$^2 G_{7/2}$	28022	-	30381	30441	30617

Er	$^4 I$	$^4 S$	$^4 F$	$^4 G$	$^4 D$	$^2 H(1)$	$^2 G$	$^2 K$	$^2 P$	$^2 D(1)$	$^2 I$	$^2 L$	$^2 D(2)$	$^2 H(2)$	$^2 F$
$^4 I$															
$^4 S$															
$^4 F$															
$^4 G$															
$^4 D$															
$^2 H(1)$															
$^2 G$															
$^2 K$															
$^2 P$															
$^2 D(1)$															
$^2 I$															
$^2 L$															
$^2 D(2)$															
$^2 H(2)$															
$^2 F$															

Figure S2

Table S7

M_{L_j}	Exp.	Calc. Pm^{3+} SEG basis set					
		4f SO-CI 30 quintets 60 triplets	4f SO-CI 30 quintets 60 triplets 44 singlets	4f SO-CI 35 quintets 99 triplets 75 singlets	4f SO-CI 35 quintets 99 triplets	4f SO-QDPT 35 quintets 99 triplets	4f/5f SO-CI 35 quintets 99 triplets
		$^5I\ ^5S\ ^5F\ ^5G\ ^3G\ ^3L\ ^3D\ ^3P\ ^3K\ ^3H$	$^5I\ ^5S\ ^5F\ ^5G\ ^3G\ ^3L\ ^3D\ ^3P\ ^3K\ ^3H\ ^1L\ ^1D\ ^1G\ ^1I$	$^5I\ ^5S\ ^5F\ ^5G\ ^5D\ ^3G\ ^3L\ ^3D\ ^3P\ ^3K\ ^3H\ ^3M\ ^3F\ ^3I\ ^1L\ ^1D\ ^1G\ ^1I\ ^1H\ ^1K\ ^1D(2)$	$^5I\ ^5S\ ^5F\ ^5G\ ^5D\ ^3G\ ^3L\ ^3D\ ^3P\ ^3K\ ^3H\ ^3M\ ^3F\ ^3I$		
5I_5	1591	1537	1528	1519	1515	1505	1515
5I_6	3212	3226	3199	3176	3180	3161	3179
5I_7	4926	4981	4941	4914	4925	4901	4922
5I_8	6690	6739	6680	6699	6728	6702	6724
5F_1	12315	12384	12382	12391	12388	12369	12187
5F_2	12715	13077	13088	13009	13039	13015	12835
5F_3	13548	14081	14083	13952	13955	13923	13753
5S_2	14139	13612	14449	13592	13613	13581	13354
5F_4	14520	15126	15107	14856	14920	14894	14721
5F_5	15885	16136	16131	16115	16133	16103	15937
3K_6	16871	18320	18435	17996	18175	18546	18091
5G_2	17459	18870	18892	18689	18694	18668	18405

Pm	$^5I\ ^5F\ ^5S\ ^5G\ ^5D$	$^3K\ ^3H\ ^3G\ ^3L\ ^3D\ ^3P\ ^3M\ ^3F\ ^3I$	$^1L\ ^1D\ ^1G\ ^1I\ ^1H\ ^1K\ ^1D(2)$
5I			
5F			
5S			
5G			
5D			
3K			
3H			
3G			
3L			
3D			
3P			
3M			
3F			
3I			
1L			
1D			
1G			
1I			
1H			
1K			
$^1D(2)$			

Figure S3

Table S8

M_{L_j}	Exp.	Calc. Ho ³⁺ SEG basis set				
		SO-CI 30 quintets 60 triplets	SO-CI 30 quintets 60 triplets 44 singlets	SO-CI 35 quintets 99 triplets 75 singlets	SO-CI 35 quintets 99 triplets	SO-QDPT 35 quintets 99 triplets
		$^5I\ ^5S\ ^5F\ ^5G\ ^3K\ ^3H\ ^3G\ ^3L\ ^3D\ ^3P$	$^5I\ ^5S\ ^5F\ ^5G\ ^3K\ ^3H\ ^3G\ ^3L\ ^3D\ ^3P\ ^1L\ ^1D\ ^1G\ ^1I$	$^5I\ ^5S\ ^5F\ ^5G\ ^5D\ ^3G\ ^3L\ ^3D\ ^3P\ ^3K\ ^3H\ ^3M\ ^3F\ ^3I\ ^1L\ ^1D\ ^1G\ ^1I\ ^1H\ ^1K\ ^1D(2)$	$^5I\ ^5S\ ^5F\ ^5G\ ^5D\ ^3K\ ^3H\ ^3G\ ^3L\ ^3D\ ^3P\ ^3M\ ^3F\ ^3I$	
5I_7	5049	5525	5549	5485	5450	5440
5I_6	8550	9484	9495	9340	9325	9318
5I_5	11110	12248	12284	12079	12110	12118
5I_4	13155	14475	14418	14401	14482	14491
5F_5	15374	16289	16594	16240	16290	16338
5S_2	18325	19769	19138	19249	19740	19800
5F_4	18444	20604	20849	20330	20358	20359
5F_3	20511	22556	22728	22007	21972	22007
5F_2	20969	23164	22871	22135	22760	22809
5F_1	-	23199	23413	23226	23202	23270
3K_8	21246	24891	23725	23233	24877	25167
5G_6	22016	24698	24760	24672	24674	24748
5G_5	23831	25756	25985	25371	25777	25918

Ho	$^5I\ ^5F\ ^5S\ ^5G\ ^5D$	$^3K\ ^3H\ ^3G\ ^3L\ ^3D\ ^3P\ ^3M\ ^3F\ ^3I$	$^1L\ ^1D\ ^1G\ ^1I\ ^1H\ ^1K\ ^1D(2)$
5I			
5F			
5S			
5G			
5D			
3K			
3H			
3G			
3L			
3D			
3P			
3M			
3F			
3I			
1L			
1D			
1G			
1I			
1H			
1K			
$^1D(2)$			

Figure S4

Table S9

M_{L_j}	Exp.	Calc. Sm^{3+} SEG basis set			
		SO-CI 21 sextets 48 quartets	SO-CI 21 sextets 80 quartets	SO-CI 18 sextets 48 quartets 42 doublets	SO-CI 18 sextets 80 quartets 98 doublets
		$^6\text{H } ^6\text{F } ^6\text{P } ^4\text{I}$ $^4\text{F } ^4\text{M } ^4\text{G}$	$^6\text{H } ^6\text{F } ^6\text{P } ^4\text{I } ^4\text{F}$ $^4\text{M } ^4\text{G } ^4\text{K } ^4\text{L}$	$^6\text{H } ^6\text{F } ^6\text{P } ^4\text{I } ^4\text{F } ^4\text{M}$ $^4\text{G } ^2\text{L } ^2\text{K } ^2\text{P } ^2\text{F}$	$^6\text{H } ^6\text{F } ^4\text{I } ^4\text{F } ^4\text{M } ^4\text{G } ^4\text{K } ^4\text{L}$ $^2\text{L } ^2\text{K } ^2\text{P } ^2\text{N } ^2\text{F } ^2\text{M } ^2\text{H } ^2\text{D}$
$^6\text{H}_{7/2}$	1045	1051	1045	1037	1055
$^6\text{H}_{9/2}$	2255	2303	2292	2275	2300
$^6\text{H}_{11/2}$	3573	3679	3663	3636	3664
$^6\text{H}_{13/2}$	4959	5094	5077	5038	5071
$^6\text{F}_{1/2}$	6254	6757	6743	6741	6769
$^6\text{H}_{15/2}$	6434	6478	6466	6399	6434
$^6\text{F}_{3/2}$	6504	6918	6911	6906	6933
$^6\text{F}_{5/2}$	7014	7310	7308	7300	7322
$^6\text{F}_{7/2}$	7874	8051	8049	8036	8064
$^6\text{F}_{9/2}$	9040	9239	9232	9209	9247
$^6\text{F}_{11/2}$	10433	10937	10918	10881	10925
$^4\text{G}_{5/2}$	17825	19628	20029	20013	18716
$^4\text{F}_{3/2}$	18821	19307	19721	19707	19414

Sm	^6H	^6F	^4I	^4F	^4M	^4G	^4K	^4L	^2L	^2K	^2P	^2N	^2F	^2M	^2H	^2D
^6H																
^6F																
^4I																
^4F																
^4M																
^4G																
^4K																
^4L																
^2L																
^2K																
^2P																
^2N																
^2F																
^2M																
^2H																
^2D																

Figure S5

Table S10

$^M L_J$	Exp.	Calc. Dy ³⁺ SEG basis set								
		4f SO-CI 18 sextets 48 quartets	4f SO-CI 21 sextets 80 quartets	4f SO-CI 18 sextets 48 quartets 42 doublets	4f SO-CI 18 sextets 80 quartets 98 doublets	4f SO-CI 18 sextets 99 quartets	4f SO-QDPT 21 sextets 96 quartets 63 doublets	4f SO-CI 21 sextets 48 quartets	4f 5f SO-CI 21 sextets 48 quartets	4f 5d SO-CI 21 sextets 48 quartets
		$^6H\ ^6F\ ^4I\ ^4F\ ^4M\ ^4G$	$^6H\ ^6F\ ^6P\ ^4I\ ^4F\ ^4M\ ^4G\ ^4K\ ^4L$	$^6H\ ^6F\ ^6P\ ^4I\ ^4F\ ^4M\ ^4G\ ^2M\ ^2P\ ^2D\ ^2L\ ^2F$	$^6H\ ^6F\ ^4I\ ^4F\ ^4M\ ^4G\ ^4K\ ^4L\ ^2L\ ^2K\ ^2P\ ^2N\ ^2F\ ^2M\ ^2H\ ^2D$	$^6H\ ^6F\ ^4I\ ^4F\ ^4M\ ^4G\ ^4K\ ^4L\ ^4D\ ^4H\ ^4P$	$^6H\ ^6F\ ^6P\ ^4I\ ^4F\ ^4M\ ^4G\ ^4K\ ^4L\ ^4D\ ^4H\ ^2L\ ^2K\ ^2P\ ^2N\ ^2F$	$^6H\ ^6F\ ^6P\ ^4I\ ^4F\ ^4M\ ^4G$		
$^6H_{13/2}$	3462	3923	3925	3955	3960	3773	3803	3919	3905	3919
$^6H_{11/2}$	5780	6772	6781	6811	6806	6500	6540	6767	6758	6767
$^6H_{9/2}$	7538	8794	8807	8845	8850	8522	8576	8791	8800	8791
$^6F_{11/2}$	7621	9066	9079	9101	9118	9086	9122	9061	8897	9030
$^6H_{7/2}$	8885	10357	10369	10407	10396	10193	10220	10353	10371	10353
$^6F_{9/2}$	8947	9930	9949	10014	10032	9937	10035	9934	9863	9911
$^6H_{5/2}$	10083	11519	11531	11568	11493	11534	11496	11515	11536	11516
$^6F_{7/2}$	10870	12045	12061	12109	12104	11835	11870	12045	11946	12019
$^6F_{5/2}$	12268	14034	14047	14079	14080	13269	13338	14031	13902	14002
$^6F_{3/2}$	13060	15584	15595	15613	15617	14151	14204	15577	15427	15547
$^6F_{1/2}$	-	16553	16562	16572	16586	14625	14621	16544	16381	16513
$^4F_{9/2}$	20987	22060	22117	22536	22543	21991	22564	22133	22739	22152
$^4I_{15/2}$	22113	28431	26968	25670	24334	26860	24454	28511	29112	28526

Dy	$^6H\ ^6F\ ^4I\ ^4F\ ^4M\ ^4G\ ^4K\ ^4L$	$^2L\ ^2K\ ^2P\ ^2N\ ^2F\ ^2M\ ^2H\ ^2D$
6H		
6F		
4I		
4F		
4M		
4G		
4K		
4L		
2L		
2K		
2P		
2N		
2F		
2M		
2H		
2D		

Figure S6

Table S11

M_{L_j}	Exp.	Calc. Eu ³⁺ SEG basis set							
		4f 7 septets 5 quintets	4f 7 septets 5 quintets 3 triplets	4f 7 septets 62 quintets 3 triplets 1 singlet	4f 7 septets 62 quintets 91 triplets	4f 7 septets 62 quintets 3 triplets 86 singlets	4f 7 septets 62 quintets 3 triplets	4f5f 7 septets 62 quintets 3 triplets	4f5d 7 septets 62 quintets 3 triplets
		⁷ F ⁵ D	⁷ F ⁵ D ³ P	⁷ F ⁵ D ⁵ L ⁵ G ⁵ H ⁵ F ³ P ¹ S	⁷ F ⁵ D ⁵ L ⁵ G ⁵ H ⁵ F ⁵ I ³ P ³ O ³ M ³ K ³ H ³ F ³ I	⁷ F ⁵ D ⁵ L ⁵ G ⁵ H ⁵ F ³ P ¹ S ¹ Q ¹ N ¹ I ¹ L ¹ G	⁷ F ⁵ D ⁵ L ⁵ G ⁵ H ⁵ F ⁵ I ³ P		
⁷ F ₁	380	384	410	377	377	377	377	386	378
⁷ F ₂	1045	1103	1149	1036	1037	1036	1039	1060	1045
⁷ F ₃	1882	2078	2119	1875	1875	1875	1881	1909	1894
⁷ F ₄	2877	3224	3250	2823	2823	2822	2831	2861	2850
⁷ F ₅	3909	4478	4490	3848	3852	3847	3857	3884	3878
⁷ F ₆	4939	5805	5806	4958	4943	4956	4968	4986	4976
⁵ D ₀	17267	19841	17750	17633	17823	17633	17827	17268	17965
⁵ D ₁	19030	20698	19466	19484	19442	19484	19446	18857	19564
⁵ D ₂	21504	22461	22330	22147	22102	22147	22135	21508	22242
⁵ D ₃	24390	25209	25750	25361	25191	25359	25370	24717	25495
⁵ L ₆	25257	-	-	30771	29395	30769	30776	29379	30151
⁵ D ₄	27633	29027	29585	28954	28396	28952	28963	28168	28952

Eu	⁷ F	⁵ D	⁵ L	⁵ G	⁵ H	⁵ I	⁵ F	³ P	³ O	³ M	³ K	³ H	³ F	³ I
⁷ F														
⁵ D														
⁵ L														
⁵ G														
⁵ H														
⁵ I														
⁵ F														
³ P														
³ O														
³ M														
³ K														
³ H														
³ F														
³ I														

a

Eu	⁷ F	⁵ D	⁵ L	⁵ G	⁵ H	⁵ I	⁵ F	³ P	¹ S	¹ Q	¹ N	¹ I	¹ L	¹ G
⁷ F														
⁵ D														
⁵ L														
⁵ G														
⁵ H														
⁵ I														
⁵ F														
³ P														
¹ S														
¹ Q														
¹ N														
¹ I														
¹ L														
¹ G														

b

Figure S7

Table S12

$^M L_j$	Exp.	Calc. Tb ³⁺ SEG basis set						
		$4f$ 7 septets 5 quintets	$4f$ 7 septets 5 quintets 3 triplets	$4f$ 7 septets 62 quintets 3 triplets	$4f$ 7 septets 62 quintets 3 triplets 1 singlet	$4f$ 7 septets 62 quintets 91 triplets	$4f5f$ 7 septets 62 quintets 91 triplets	$4f5d$ 7 septets 62 quintets 91 triplets
		$^7F\ ^5D$	$^7F\ ^5D\ ^3P$	$^7F\ ^5D\ ^5L\ ^5G$ $^5H\ ^5F\ ^5I\ ^3P$	$^7F\ ^5D\ ^5L\ ^5G\ ^5H\ ^5F\ ^3P$ 1S	$^7F\ ^5D\ ^5L\ ^5G\ ^5H\ ^5F\ ^5I\ ^3P\ ^3O\ ^3M\ ^3K\ ^3H\ ^3F\ ^3I$		
7F_5	2037	1942	1930	2119	2116	2145	2150	2146
7F_4	3281	3171	3164	3542	3538	3541	3538	3541
7F_3	4268	4105	4099	4703	4698	4717	4715	4717
7F_2	4931	4793	4755	5584	5578	5612	5612	5613
7F_1	5379	5249	5184	6191	6184	6225	6227	6225
7F_0	5616	5475	5396	6502	6491	6535	6539	6536
5D_4	20394	20157	20713	21683	21693	20807	20476	20772
5D_3	26187	25999	26554	27459	27450	27141	26808	27106
$^5L_{10}$	26315	-	-	32495	32495	31456	30800	31311
5D_2	28063	30393	28388	29475	29529	29056	29177	29420

Tb	7F	5D	5L	5G	5H	5I	5F	3P	3O	3M	3K	3H	3F	3I
7F														
5D														
5L														
5G														
5H														
5I														
5F														
3P														
3O														
3M														
3K														
3H														
3F														
3I														

Figure S8

Table S13

$^M L_j$	Exp.	Calc. Gd ³⁺ SEG basis set				
		4 <i>f</i> 1 octet 48 sextets 5 quartets	4 <i>f</i> 1 octet 37 sextets 5 quartets 41 doublets	4 <i>f</i> 1 octet 48 sextets 85 quartets	4 <i>f</i> 5 <i>f</i> 1 octet 48 sextets 85 quartets	4 <i>f</i> 5 <i>d</i> 1 octet 48 sextets 85 quartets
		⁸ S ⁶ P ⁶ I ⁶ D ⁶ G ⁶ F ⁶ H ⁴ D	⁸ S ⁶ P ⁶ I ⁶ D ⁶ G ⁶ F ⁴ D ² Q ² G ² F	⁸ S ⁶ P ⁶ I ⁶ D ⁶ G ⁶ F ⁶ H ⁴ D ⁴ N ⁴ H ⁴ L ⁴ K ⁴ F ⁴ G		
⁶ P _{7/2}	32100	34899	34446	34393	32834	34332
⁶ P _{5/2}	32702	35286	34930	34892	33275	34825
⁶ P _{3/2}	33282	36242	35955	35958	34349	35893
⁶ I _{7/2}	35843	45309	42476	40775	38893	40573
⁶ I _{9/2}	36185	45234	42401	41116	39226	40911
⁶ I _{7/2}	36240	44772	41939	40914	39012	40704
⁶ I _{11/2}	36461	45144	42311	41427	39528	41218
⁶ I _{5/2}	36579	44913	42079	41456	39549	41243
⁶ I _{13/2}	36614	45036	42203	41588	39682	41377
⁶ D _{9/2}	39536	45540	44036	43247	41360	43082
⁶ D _{11/2}	~40468	46160	44693	44706	42812	44537
⁶ D _{7/2}	40581	46359	44929	44760	42899	44604
⁶ D _{3/2}	~40729	46454	45048	45046	43169	44884
⁶ D _{5/2}	~40862	46614	45236	45183	43316	45025

Gd	⁸ S	⁶ P	⁶ I	⁶ D	⁶ G	⁶ F	⁶ H	⁴ D	⁴ N	⁴ H	⁴ L	⁴ K	⁴ F	⁴ G
⁸ S														
⁶ P														
⁶ I														
⁶ D														
⁶ G														
⁶ F														
⁶ H														
⁴ D														
⁴ N														
⁴ H														
⁴ L														
⁴ K														
⁴ F														
⁴ G														

Figure S9

Table S14. Energies of some hypersensitive transitions in lanthanides (cm^{-1}) calculated in this work compared with other calculations and experimental data

Ln ($4f^n$)	Transition	Calculated (this work)	Calculated [83]	Exp. LnBr_3 [112]
Pr^{3+} ($4f^2$)	$^3\text{H}_4 \rightarrow ^3\text{F}_2$	4880	5488	4800
Nd^{3+} ($4f^3$)	$^4\text{I}_{9/2} \rightarrow ^4\text{G}_{5/2}$	17690	21748	17300
Pm^{3+} ($4f^4$)	$^5\text{I}_4 \rightarrow ^5\text{G}_2$	18870	23074	17459 ^a
Sm^{3+} ($4f^5$)	$^6\text{H}_{5/2} \rightarrow ^6\text{F}_{1/2}$	6760	7329	6200
Eu^{3+} ($4f^6$)	$^7\text{F}_0 \rightarrow ^7\text{F}_2$	1040	1132	1020 ^a
	$^7\text{F}_0 \rightarrow ^5\text{D}_2$	22140	23991	21500
Tb^{3+} ($4f^8$)	$^7\text{F}_6 \rightarrow ^7\text{F}_5$	2150	1885	2040 ^a
Dy^{3+} ($4f^9$)	$^6\text{H}_{15/2} \rightarrow ^6\text{F}_{11/2}$	8370	8739	7700
Ho^{3+} ($4f^{10}$)	$^5\text{I}_8 \rightarrow ^5\text{G}_6$	24700	26380	22200
Er^{3+} ($4f^{11}$)	$^4\text{I}_{15/2} \rightarrow ^2\text{H}_{11/2}$	20330	22591	19200
	$^4\text{I}_{15/2} \rightarrow ^4\text{G}_{11/2}$	27980	30842	26500
Tm^{3+} ($4f^{12}$)	$^3\text{H}_6 \rightarrow ^3\text{H}_4$	5450	5814	5500
	$^3\text{H}_6 \rightarrow ^3\text{F}_4$	12860	12883	12600

^a In LaCl_3 [111]**Table S15.** Energy levels of Pr^{3+} (cm^{-1}) calculated in this work compared with other calculations and experimental data

$^{\text{M}}\text{L}_j$	Our calc.	Calc. [85]	Exp. [111]
$^3\text{H}_5$	2180	2216	2152
$^3\text{H}_6$	4450	4545	4389
$^3\text{F}_2$	4880	4977	4997
$^3\text{F}_3$	6320	6463	6415
$^3\text{F}_4$	6820	7075	6855
$^1\text{G}_4$	9950	10345	9921
$^1\text{D}_2$	17030	17594	17334
$^3\text{P}_0$	21500	22589	21390
$^3\text{P}_1$	22130	22594	22007
$^1\text{I}_6$	23120	22345	22212
$^3\text{P}_2$	23310	23484	23161

Table S16. Energy $^7\text{F}_j$ levels of Tb^{3+} (cm^{-1}) calculated in this work compared with other calculations and experimental data

$^{\text{M}}\text{L}_j$	Our calc.	Calc. [85]	Exp. [111]
$^7\text{F}_5$	2145	2229	2037
$^7\text{F}_4$	3541	3321	3281
$^7\text{F}_3$	4717	4296	4268
$^7\text{F}_2$	5612	5016	4931
$^7\text{F}_1$	6225	5493	5379
$^7\text{F}_0$	6535	5732	5616