Supporting information

Unraveling the local structure and luminescence evolution in Nd^{3+}-doped $LiYF_4$ laser crystals: a new theoretical approach

Yang Xiao,‡[a] Xiaoyu Kuang,‡*[a] Yauyuen Yeung*[b] and Meng Ju*[c]

[a] Institute of Atomic and Molecular Physics, Sichuan University, Chengdu 610065, China

[b] Department of Science and Environmental Studies, The Education University of Hong Kong, 10 Lo Ping Road, Tai Po, NT, Hong Kong, China

[c] School of Physical Science and Technology, Southwest University, Chongqing 400715, China

*Correspondence author.

‡These two authors contributed equally to this work.

E-mail: kuangxiaoyu@scu.edu.cn (Xiaoyu Kuang) yyyeung@eduhk.hk (Yauyuen Yeung) and mengju@swu.edu.cn (Meng Ju)

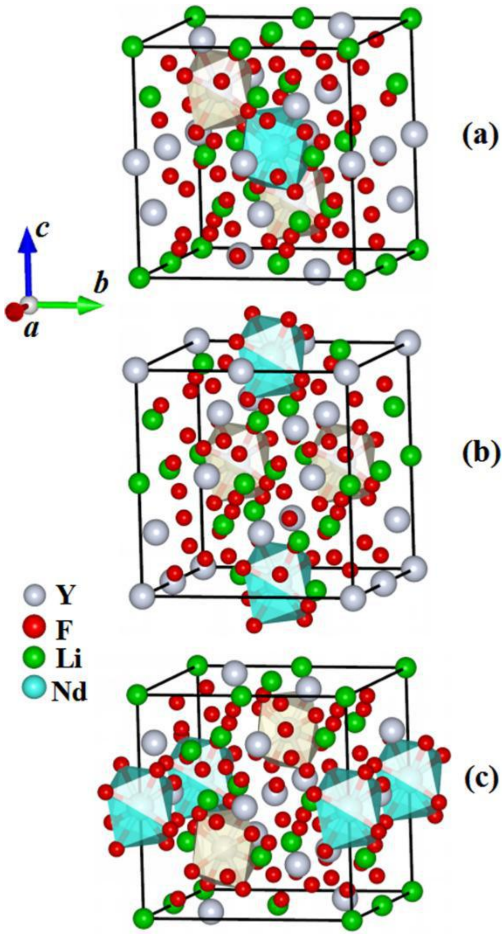

Fig. S1 Coordination structures of the predicted metastable (a), (b) and (c) for Nd^{3+}-doped YLF.

Table S1 Lattice parameters, interatomic distances and angles in the ground state structure of YLF and Nd^{3+}-doped YLF

Structure	Space group	Lattice parameters (Å)		Interatomic distances (Å)			Interatomic angles (°)		
		Theor.	Expt.[1]		Theor.	Expt.[1]		Theor.	Expt.[1]
YLF	$I4_1/a$	$a = 10.439$	$a = 10.342$	Y-F1	2.322	2.300	F1-Y-F1	76.004	75.920
		$b = 10.439$	$b = 10.342$	Y-F2	2.259	2.247	F2-Y-F2	98.594	98.700
		$c = 10.812$	$c = 10.748$				F1-Y-F2	74.685	74.453
YLF:Nd^{3+}	$P\bar{4}$	$a = 10.467$	—	Nd-F1	2.415	—	F1-Nd-F1	75.755	—
		$b = 10.467$	—	Nd-F2	2.360	—	F2-Nd-F2	99.037	—
		$c = 10.859$	—			—	F1-Nd-F2	74.566	—

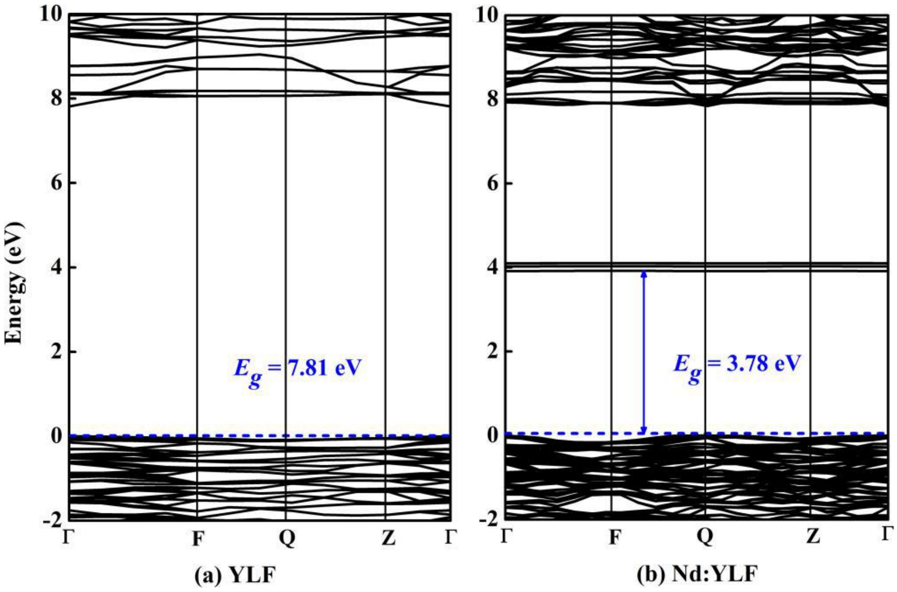

Fig. S2 Calculated band structures of (a) YLF using GGA method, (b)Nd^{3+}-doped YLF using GGA+U method. The Fermi level is indicated by the horizontal dotted lines

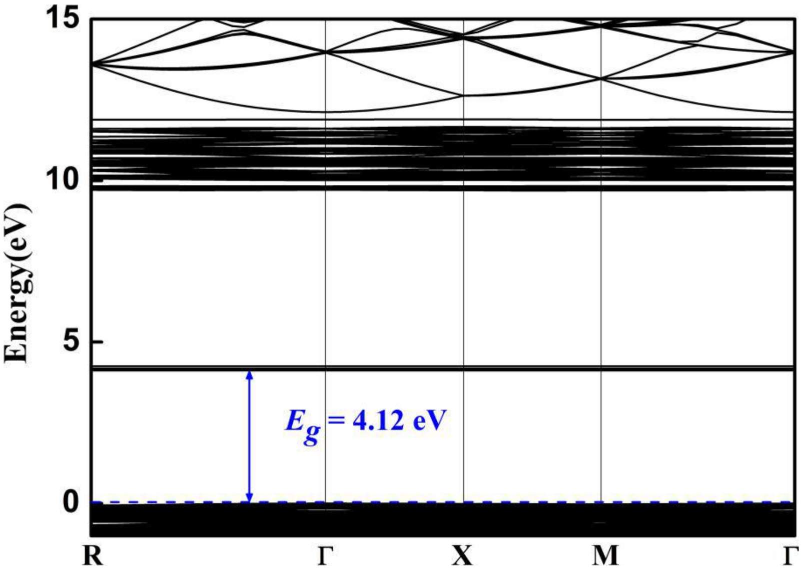

Fig. S3 Calculated band structure and DOS of Nd^{3+}-doped YLF by using the modified BJ method in Wien2k package.

Table S2 **C**oordinates of all atoms for the ground-state structure of Nd^{3+}-doped YLF.

Atom	x	y	z	Wyckoff site symmetry
F1	0.64029	-0.16793	0.33148	4h
F2	0.88839	0.08722	0.82447	4h
F3	0.60858	-0.08271	0.83023	4h
F4	0.85799	0.16656	0.33215	4h
F5	0.91784	0.14352	0.57866	4h
F6	0.66810	-0.10739	1.07993	4h
F7	0.82692	0.11350	1.08612	4h
F8	0.58355	-0.14105	0.58138	4h
F10	0.60818	0.16733	0.91951	4h
F12	0.63939	0.08364	0.42015	4h
F13	0.57969	0.10834	1.17045	4h
F15	0.66609	0.14126	0.66842	4h
F33	0.64124	0.33300	0.33047	4h
F35	0.60894	0.41677	0.83095	4h
F38	0.66674	0.39168	1.08073	4h
F40	0.58233	0.35896	0.58084	4h
Y1	0.50014	-0.24994	0.75011	4h
Y2	0.74686	0.00060	0.25218	4h
Y4	0.74958	0.74949	0.49994	4h
Li1	0.50012	0.74926	0.25036	4h
Li2	0.74890	-0.00073	0.74678	4h
Li4	0.74804	0.74858	1.00064	4h
Y3	0.50000	-0.00000	0.99997	2g
Li3	0.50000	-0.00000	0.50201	2g
Y10	0.50000	0.50000	1.00000	1c
Nd1	1.00000	-0.00000	1.00000	1a
Li7	1.00000	-0.00000	0.50000	1b
Li11	0.50000	0.50000	0.50000	1d

Table S3 Structural parameters *a*, *b* and *c*, unit-cell volume, relative energies for the metastable Nd^{3+}-doped YLF.

Structure	Space group	Lattice parameters (Å)			volume	relative energies
		a	*b*	*c*	V (Å^3)	ΔE (eV)
YLF:Nd^{3+}	$P\bar{4}$	a = 10.467	b = 10.467	c = 10.859	1189.781	0
Isomer (a)	$P\bar{4}$	a = 10.467	b = 10.467	c = 10.860	1189.802	0.021
Isomer (b)	$P\bar{4}$	a = 10.468	b = 10.468	c = 10.860	1190.061	0.185
Isomer (c)	$P\bar{4}$	a = 10.468	b = 10.468	c = 10.861	1190.063	0.217

Table S4 The calculated Stark levels (all in cm^{-1}) of Nd^{3+} in $LiYF_4$. Available experimental and theoretical results are listed for comparison.

			Present work				Other[2]
			Fit 2		Fit 3		
$^{2S+1}L_J$	Level No.	E_{expt}[2]	E_{calc}	ΔE	E_{calc}	ΔE	E_{calc}
$^4I_{9/2}$	1	0	-10	10	-8	8	0
	2	130	135	-5	132	-2	144
	3	180	167	13	173	7	178
	4	247	247	0	245	2	255
	5	523	528	-5	523	0	533
$^4I_{11/2}$	6	1997	1987	10	1989	8	1985
	7	2040	2026	14	2030	10	2024
	8	2042	2028	14	2033	9	2026
	9	2077	2071	6	2071	6	2070
	10	2227	2231	-4	2228	-1	2228
	11	2262	2264	-2	2259	3	2260
$^4I_{13/2}$	12	3933	3941	-8	3945	-12	3940
	13	3949	3966	-17	3970	-21	3964
	14	3993	3989	4	3993	0	3988
	15	4026	4017	9	4020	6	4015
	16	4208	4206	2	4209	-1	4203
	17	4221	4245	-24	4238	-17	4241
	18	4241	4248	-7	4240	1	4244
$^4I_{15/2}$	19	5854	5853	1	5854	0	5864
	20	5913	5913	0	5914	-1	5922
	21	5946	5948	-2	5957	-11	5960
	22	6026	6025	1	6030	-4	6033
	23	6314	6318	-4	6309	5	6325
	24	6348	6358	-10	6350	-2	6364
	25	6392	6392	0	6393	-1	6399
	26	6434	6438	-4	6432	2	6440
$^4F_{3/2}$	27	11542	11538	4	11545	-3	11535
	28	11602	11579	23	11586	16	11574
$^4F_{5/2}$	29	12540	12527	13	12529	11	12525
$^2H(2)_{9/2}$	30	12550	12562	-12	12547	3	12560
	31	12632	12616	16	12631	1	12631
	32	12647	12669	-22	12663	-16	12669
	33	12670	12678	-8	12677	-7	12685
	34	12736	12775	-39	12743	-7	12787
	35	12809	12791	18	12823	-14	12802
	36	12843	12867	-24	12854	-11	12877

$^4F_{7/2}$	37	13499	13504	-5	13502	-3	13510
$^4S_{3/2}$	38	13524	13522	2	13523	1	13527
	39	13630	13631	-1	13630	0	13639
	40	13640	13636	4	13637	3	13645
	41	13650	13651	-1	13649	1	13654
	42	13662	13652	10	13652	10	13656
$^4F_{9/2}$	43	14753	14759	-6	14752	1	14759
	44	14783	14782	1	14776	7	14781
	45	14882	14877	5	14882	0	14877
	46	14896	14895	1	14905	-9	14897
	47	14954	14945	9	14954	0	14945
$^2H(2)_{11/2}$	**48**	**15928**	**16002**	**-74**	**15940**	**-12**	**16005**
	49	**15976**	**16003**	**-27**	**15979**	**-3**	**16006**
	50	**16002**	**16022**	**-20**	**15999**	**3**	**16025**
	51	**16063**	**16036**	**27**	**16047**	**16**	**16038**
	52	**16136**	**16063**	**73**	**16116**	**20**	**16064**
	53	**16147**	**16092**	**55**	**16132**	**15**	**16094**
$^4G_{5/2}$	54	17163	17179	-16	17185	-22	17180
$^2G(1)_{7/2}$	55	17274	17275	-1	17285	-11	17274
$^4G_{7/2}$	56	17301	17326	-25	17322	-21	17328
	57	17412	17387	25	17393	19	17390
	58	17423	17396	27	17400	23	17399
	59	17479	17469	10	17457	22	17473
	60	17653	17663	-10	17647	6	17663
	61	19072	19041	31	19054	18	19037
	62	19082	19080	2	19072	10	19076
	63	19185	19195	-10	19184	1	19190
	64	19215	19213	2	19208	7	19208
$^2K_{13/2}$	65	19464	19454	10	19455	9	19456
$^4G_{9/2}$	66	19564	19547	17	19559	5	19550
	67	19618	19611	7	19612	6	19615
	68	19668	19662	6	19661	7	19664
	69	19691	19694	-3	19693	-2	19697
	70	19700	19702	-2	19706	-6	19706
	71	19719	19732	-13	19731	-12	19733
	72	19727	19733	-6	19736	-9	19737
	73	19762	19751	11	19754	8	19752
	74	19823	19799	24	19809	14	19803
	75	19950	19971	-21	19973	-23	19972
	76	19973	19987	-14	19996	-23	19987
$^2G(1)_{9/2}$	77	—	21015	—	21013	—	21013
	78	21063	21059	4	21057	6	21057

	79	21072	21085	-13	21061	11	21084
	80	21083	21092	-9	21075	8	21092
	81	21111	21178	-67	21195	-84	21175
^{2}D(1)$_{3/2}$	82	21283	21305	-22	21297	-14	21311
	83	21335	21322	13	21329	6	21329
^{4}G$_{11/2}$	84	21440	21437	3	21449	-9	21450
^{2}K$_{15/2}$	85	21462	21454	8	21463	-1	21464
	86	21555	21568	-13	21569	-14	21580
	87	21713	21679	34	21700	13	21682
	88	21730	21714	16	21715	15	21715
	89	21750	21749	1	21741	9	21749
	90	21776	21776	0	21783	-7	21780
	91	21806	21806	0	21799	7	21812
	92	21812	21813	-1	21817	-5	21820
	93	21830	21820	10	21822	8	21821
	94	21902	21905	-3	21898	4	21915
	95	21948	21947	1	21940	8	21952
	96	21985	21989	-4	21985	0	21997
	97	21994	22006	-12	22001	-7	22010
^{2}P$_{1/2}$	98	23404	23404	0	23391	13	23409
^{2}D(1)$_{5/2}$	99	23898	23912	-14	23895	3	23919
	100	23942	23947	-5	23939	3	23952
	101	24038	24016	22	24030	8	24019
^{2}P$_{3/2}$	102	26264	26285	-21	26284	-20	26282
	103	26344	26338	6	26346	-2	26333
^{4}D$_{3/2}$	104	28110	28097	13	28104	6	28093
	105	28217	28215	2	28214	3	28213
^{4}D$_{5/2}$	106	28372	28373	-1	28365	7	28366
	107	28529	28568	-39	28567	-38	28565
	108	28584	28587	-3	28587	-3	28578
^{4}D$_{1/2}$	109	28803	28812	-9	28805	-2	28810
^{2}I$_{11/2}$	110	29206	29231	-25	29222	-16	29233
	111	29296	29293	3	29293	3	29292
	112	29377	29392	-15	29390	-13	29394
	113	29467	29515	-48	29527	-60	29517
	114	29719	29689	30	29697	22	29689
	115	29734	29709	25	29724	10	29711
^{2}L$_{15/2}$	116	—	30164	—	30184	—	30161
^{4}D$_{7/2}$	117	30235	30206	29	30218	17	30202
^{2}I$_{13/2}$	118	—	30281	—	30319	—	30275
	119	30292	30329	-37	30284	8	30319

	120	—	30370	—	30358	—	30360
	121	30375	30379	-4	30379	-4	30367
	122	30450	30421	29	30424	26	30404
	123	30548	30539	9	30542	6	30517
	124	30574	30592	-18	30603	-29	30577
	125	30599	30609	-10	30612	-13	30587
	126	—	30614	—	30632	—	30601
	127	—	30667	—	30651	—	30649
	128	30672	30672	0	30665	7	30676
	129	30720	30717	3	30712	8	30723
	130	30758	30754	4	30741	17	30760
	131	30898	30882	16	30899	-1	30888
	132	30925	30927	-2	30923	2	30934
	133	—	31037	—	31051	—	31043
	134	31062	31048	14	31052	10	31054
$^2L_{17/2}$	135	31645	31662	-17	31686	-41	31666
	136	31743	31733	10	31739	4	31736
	137	31800	31783	17	31780	20	31783
	138	31858	31854	4	31843	15	31852
	139	31890	31884	6	31865	25	31880
	140	31934	31929	5	31907	27	31923
	141	31940	31947	-7	31938	2	31941
	142	32048	32072	-24	32089	-41	32065
	143	32096	32083	13	32102	-6	32074
$^2H(1)_{9/2}$	144	32920	32944	-24	32934	-14	32949
	145	33019	33030	-11	33030	-11	33034
	146	—	33044	—	33034	—	33047
	147	33104	33102	2	33108	-4	33104
	148	33155	33145	10	33147	8	33148
$^2D(2)_{3/2}$	149	33433	33435	-2	33433	0	33439
	150	33516	33520	-4	33513	3	33522
$^2H(1)_{11/2}$	151	34232	34238	-6	34227	5	34245
$^2D(2)_{5/2}$	152	34289	34301	-12	34302	-13	34298
	153	34338	34303	35	34312	26	34305
	154	34429	34427	2	34428	1	34431
	155	34460	34428	32	34431	29	34432
	156	—	34493	—	34498	—	34496
	157	34523	34505	18	34525	-2	34503

	158	—	34636	—	34649	—	34636
	159	—	34638	—	34653	—	34639
$^2F(2)_{5/2}$	160	38458	38531	-73	38504	-46	38533
	161	38672	38627	45	38660	12	38628
	162	38704	38695	9	38706	-2	38693
$^2F(2)_{7/2}$	163	39924	39964	-40	39945	-21	39969
	164	39970	39979	-9	39964	6	39984
	165	40070	40047	23	40051	19	40051
	166	40179	40141	38	40151	28	40145
$^2G(2)_{9/2}$	167	—	47752	—	47683	—	47789
	168	—	47836	—	47748	—	47869
	169	—	47849	—	47780	—	47882
	170	—	47929	—	47855	—	47961
	171	—	47961	—	47935	—	47993
$^2G(2)_{7/2}$	172	—	48677	—	48614	—	48689
	173	—	48739	—	48669	—	48746
	174	—	48853	—	48769	—	48858
	175	—	48928	—	48908	—	48935
$^2F(1)_{7/2}$	176	—	66339	—	66212	—	66381
	177	—	66585	—	66454	—	66622
	178	—	66772	—	66614	—	66807
	179	—	66843	—	66666	—	66879
$^2F(1)_{5/2}$	180	—	67582	—	67443	—	67631
	181	—	67916	—	67742	—	67957
	182	—	68006	—	67813	—	68051

Table S5 Calculated ED absorption lines and corresponding wavelengths (λ), ED (A_{ED}), MD (A_{MD}) radiative decay rates and line oscillator strengths from the ground to excited state level of Nd^{3+}-doped YLF.

$^{2S+1}L_J$	Transition	A_{ED} (s^{-1})	A_{MD} (s^{-1})	λ (nm)	$S_{ED}\times10^{-20}$
$^4I_{9/2}$	$^4I_{15/2}$	2.6	0	1693	0.219
	$^4F_{3/2}$	405.6	0	882	1.192
	$^4F_{5/2}$	845.1	0	808	2.868
	$^2H_{9/2}$	110.2	1.7	799	0.604
	$^4F_{7/2}$	617.2	0	750	2.230
	$^4S_{3/2}$	643.2	0	746	1.143
	$^4F_{9/2}$	69.0	0.4	684	0.237
	$^2H_{11/2}$	18.6	0	632	0.060
	$^4G_{5/2}$	1679.8	0	584	2.148
	$^2G_{7/2}$	496.0	0.1	582	0.840
	$^4G_{7/2}$	793.5	0	527	0.995
	$^4G_{9/2}$	286.6	0.1	515	0.420
	$^2K_{13/2}$	79.2	0	513	0.161
	$^2G_{9/2}$	126.6	0	477	0.148
	$^2D_{3/2}$	177.3	0	473	0.080
	$^4G_{11/2}$	45.9	0	467	0.060
	$^2K_{15/2}$	53.0	0	466	0.092
	$^2P_{1/2}$	918.9	0	432	0.159
	$^2D_{5/2}$	19.6	0	422	0.009
	$^2P_{3/2}$	32.8	0	383	0.008

Table S6 Calculated wavelengths (λ), ED (A_{ED}), MD (A_{MD}) radiative decay rates, branching ratios (β) and radiative lifetimes (τ) for spontaneous emission transitions between J-multiplets in Nd:YLF. Available the experimental results are also listed for comparison.

Transition		λ (nm)		A_{ED} (s^{-1})		A_{MD} (s^{-1})	β	τ (μs)	
		Present	Expt.[3]	Present	Expt.[3]	Present	Present	Present	Expt.[3]
$^2H_{9/2}$	$^4I_{9/2}$	799		231.8		3.6	0.5	2409	
	$^4I_{11/2}$	940		29.3		2.2	0.1		
	$^4I_{13/2}$	1155		79.2		0.0	0.2		
	$^4I_{15/2}$	1515		69.0		0.0	0.2		
	$^4F_{3/2}$	8565		0.1		0.0	0.0		
	$^4F_{5/2}$	74203		0.0		0.0	0.0		
$^4F_{5/2}$	$^4I_{9/2}$	808		1776.8		0.0	0.7	365	
	$^4I_{11/2}$	952		326.9		0.0	0.1		
	$^4I_{13/2}$	1173		541.0		0.0	0.2		
	$^4I_{15/2}$	1546		98.5		0.0	0.0		
	$^4F_{3/2}$	9683		0.1		0.1	0.0		
$^4F_{3/2}$	$^4I_{9/2}$	882	894	852.8	617	0.0	0.4	469	525±10%
	$^4I_{11/2}$	1056	1052	1060.7	1076	0.0	0.5		
	$^4I_{13/2}$	1335	1330	210.0	210	0.0	0.1		
	$^4I_{15/2}$	1840	1830	10.6	1.08	0.0	0.0		
$^4I_{15/2}$	$^4I_{9/2}$	1693		5.5		0.0	0.2	32217	
	$^4I_{11/2}$	2477		16.7		0.0	0.5		
	$^4I_{13/2}$	4868		8.0		0.9	0.3		
$^4I_{13/2}$	$^4I_{9/2}$	2596		18.1		0.0	0.7	37918	
	$^4I_{11/2}$	5044		7.1		1.2	0.3		
$^4I_{11/2}$	$^4I_{9/2}$	5350		6.5		0.8	1.0	136509	

Table S7 Calculated MD oscillator strengths and wavelengths (λ) as well as available experimental wavelengths (λ_{expt}) for transitions from the emitting Stark levels $^4F_{3/2}$(27,28) to the Stark levels in manifolds $^4I_{9/2}$, $^4I_{11/2}$, $^4I_{13/2}$ and $^4I_{15/2}$.

$^{2S+1}L_J$(No.)	$S'L'J'$(No.)	λ (nm)	λ_{expt} (nm)	$f_{\mathrm{MD}}\times10^{-8}$
$^4F_{3/2}$(28)	$^4I_{9/2}$(1)	863		0.029
$^4F_{3/2}$(27)	$^4I_{9/2}$(1)	866		0.587
$^4F_{3/2}$(28)	$^4I_{9/2}$(2)	873		0.084
$^4F_{3/2}$(28)	$^4I_{9/2}$(3)	876		0.094
$^4F_{3/2}$(27)	$^4I_{9/2}$(2)	876		0.062
$^4F_{3/2}$(27)	$^4I_{9/2}$(3)	879		0.247
$^4F_{3/2}$(28)	$^4I_{9/2}$(4)	882		0.043
$^4F_{3/2}$(27)	$^4I_{9/2}$(4)	885		0.049
$^4F_{3/2}$(28)	$^4I_{9/2}$(5)	904	903[4]	0.072
$^4F_{3/2}$(27)	$^4I_{9/2}$(5)	908	908[4]	0.380
$^4F_{3/2}$(28)	$^4I_{11/2}$(6)	1042		0.067
$^4F_{3/2}$(28)	$^4I_{11/2}$(7)	1046		0.293
$^4F_{3/2}$(27)	$^4I_{11/2}$(6)	1047		0.924
$^4F_{3/2}$(28)	$^4I_{11/2}$(8)	1047		0.534
$^4F_{3/2}$(27)	$^4I_{11/2}$(7)	1051		0.863
$^4F_{3/2}$(28)	$^4I_{11/2}$(9)	1051		0.095
$^4F_{3/2}$(27)	$^4I_{11/2}$(8)	1052		0.009
$^4F_{3/2}$(27)	$^4I_{11/2}$(9)	1056		0.024
$^4F_{3/2}$(28)	$^4I_{11/2}$(10)	1069		0.363
$^4F_{3/2}$(28)	$^4I_{11/2}$(11)	1072		0.001
$^4F_{3/2}$(27)	$^4I_{11/2}$(10)	1073		0.264
$^4F_{3/2}$(27)	$^4I_{11/2}$(11)	1077		0.240
$^4F_{3/2}$(28)	$^4I_{13/2}$(12)	1309		0.260
$^4F_{3/2}$(28)	$^4I_{13/2}$(13)	1313	1313[5]	0.339
$^4F_{3/2}$(27)	$^4I_{13/2}$(12)	1316		0.097
$^4F_{3/2}$(28)	$^4I_{13/2}$(14)	1317		0.051
$^4F_{3/2}$(27)	$^4I_{13/2}$(13)	1321	1321[5]	0.357
$^4F_{3/2}$(28)	$^4I_{13/2}$(15)	1322		0.022
$^4F_{3/2}$(27)	$^4I_{13/2}$(14)	1324		0.188
$^4F_{3/2}$(27)	$^4I_{13/2}$(15)	1329		0.249
$^4F_{3/2}$(28)	$^4I_{13/2}$(16)	1356		0.176
$^4F_{3/2}$(28)	$^4I_{13/2}$(17)	1361		0.183
$^4F_{3/2}$(28)	$^4I_{13/2}$(18)	1361		0.062
$^4F_{3/2}$(27)	$^4I_{13/2}$(16)	1363		0.093
$^4F_{3/2}$(27)	$^4I_{13/2}$(17)	1368		0.026

$^4F_{3/2}$(27)	$^4I_{13/2}$(18)	1369	0.077
$^4F_{3/2}$(28)	$^4I_{15/2}$(19)	1745	0.026
$^4F_{3/2}$(27)	$^4I_{15/2}$(19)	1757	0.001
$^4F_{3/2}$(28)	$^4I_{15/2}$(20)	1763	0.082
$^4F_{3/2}$(27)	$^4I_{15/2}$(20)	1776	0.038
$^4F_{3/2}$(28)	$^4I_{15/2}$(21)	1777	0.001
$^4F_{3/2}$(27)	$^4I_{15/2}$(21)	1789	0.033
$^4F_{3/2}$(28)	$^4I_{15/2}$(22)	1800	0.034
$^4F_{3/2}$(27)	$^4I_{15/2}$(22)	1813	0.054
$^4F_{3/2}$(28)	$^4I_{15/2}$(23)	1895	0.044
$^4F_{3/2}$(28)	$^4I_{15/2}$(24)	1910	0.056
$^4F_{3/2}$(27)	$^4I_{15/2}$(23)	1910	0.005
$^4F_{3/2}$(27)	$^4I_{15/2}$(24)	1925	0.013
$^4F_{3/2}$(28)	$^4I_{15/2}$(25)	1926	0.007
$^4F_{3/2}$(28)	$^4I_{15/2}$(26)	1940	0.039
$^4F_{3/2}$(27)	$^4I_{15/2}$(25)	1941	0.068
$^4F_{3/2}$(27)	$^4I_{15/2}$(26)	1955	0.014

Appendix – Method and equations for the calculations of transition intensities[6]

The ED radiative decay rates for spontaneous emission transitions can be expressed as

$$A_{ED}(SLJ \to S'L'J') = \frac{16\pi^3 e^2}{3\varepsilon_0 hc^3} \frac{\upsilon^3}{(2J+1)} \chi_{ED}^{emi} \sum_{\lambda=2,4,6} \Omega_{(\lambda)} \left| < l^N SLJ \,||\, U^{(\lambda)} \,||\, l^N S'L'J' > \right|^2 \quad (1)$$

where ν donates the transition frequency. Ω_λ (λ=2, 4, 6) are the Judd-Ofelt intensity parameters. The local field correction between χ_{ED}^{emi} in emission and χ_{ED}^{abs} in absorption with the relationship of $\chi_{ED}^{emi} = 2\chi_{ED}^{abs}$ and χ_{ED}^{abs} is the local field correction for an ED-induced absorption with the form of $\left[(n^2+2)/3\right]^2$, n is the refractive index. The summation is over the ever-rank reduced matrix-element λ=2, 4, 6 of the $U^{(\lambda)}$ tensor operator.

The MD radiative decay rates for spontaneous emission transitions be expressed as

$$A_{MD}(SLJ \to S'L'J') = \frac{\pi h e^2}{3\varepsilon_0 c^2} \frac{\upsilon^3}{(2J+1)} n^3 \left| < l^N SLJ \,||\, L+gS \,||\, l^N S'L'J' > \right|^2 \quad (2)$$

where g=2.00232 is the factor of the electron, which donates the gyromagnetic ratio of the electron.

The radiative lifetime can be expressed as

$$\tau_{SLJ} = \frac{1}{\sum_{S'L'J'} \left(A_{ED}(SLJ \to S'L'J') + A_{MD}(SLJ \to S'L'J') \right)} \quad (3)$$

where $A(SLJ \to S'L'J') = A_{ED}(SLJ \to S'L'J') + A_{MD}(SLJ \to S'L'J')$ which is the total spontaneous radiative decay rate for a transition $|l^N SLJ> \to |l^N S'L'J'>$. The total radiative rate of $|l^N SLJ>$ is simply the sum of the respective radiative rates when an excited state decays to several lower-energy final states.

The branching ratio can be expressed as

$$\beta_{SLJ\to S'L'J'}=\frac{\left(A_{ED(SLJ\to S'L'J')}+A_{MD(SLJ\to S'L'J')}\right)}{\sum\limits_{S'L'J'}\left(A_{ED(SLJ\to S'L'J')}+A_{MD(SLJ\to S'L'J')}\right)} \tag{4}$$

The oscillator strength be expressed as

$$f_{MD}=\frac{h\upsilon}{6m_e}\frac{n}{2J+1}\left|<l^N SLJ\,||\,L+gS\,||\,l^N S'L'J'>\right|^2 \tag{5}$$

References

1. A. Goryunov and A. Popov, *Zhurnal Neorganicheskoj Khimii*, 1992, **37**, 276-279.
2. H. D. Leebeeck, K. Binnemans and C. Görller-Walrand, *J. Alloys Compd.*, 1999, **291**, 300-311.
3. J. R. Ryan and R. Beach, *J. Opt. Soc. Am. B*, 1992, **9**, 1883-1887.
4. J. Zhang, J. Xia, X. Fu and P. Zhai, *Opt. Laser Technol.*, 2017, **90**,185-189.
5. X. Huang, Z. Lin, J. Lan, Y. Wang, B. Xu, H. Xu and Z. Cai, *J. Mod. Opt.*, 2016, **63**, 1-5.
6. M. P. Hehlen, M. G. Brik and K. W. Kramer, *J. Lumin.*, 2013, **136**, 221-239.