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Supporting Information

Photoluminescence and energy transfer mechanisms of Tm^{3+} doped Y_2O_3 laser crystals: Experimental and theoretical insights

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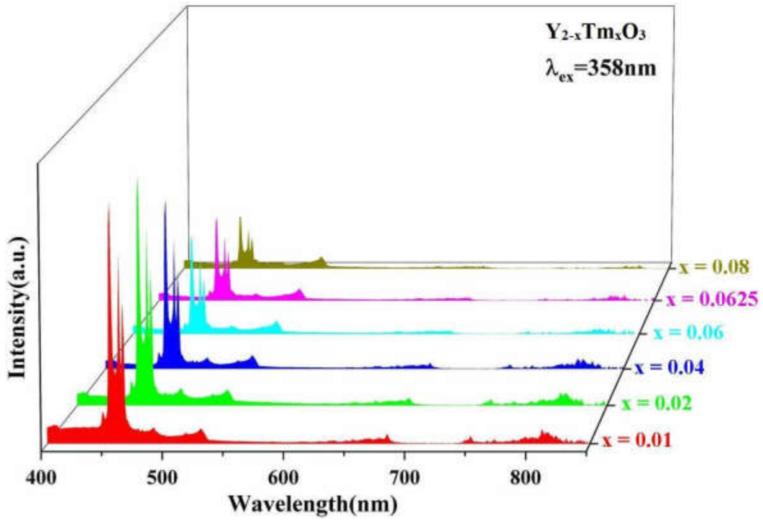


Figure S1. Dependence of photoluminescence emission intensities of $Y_{2-x}Tm_xO_3$ at λ_{ex} = 358 nm at different Tm^{3+} concentrations.

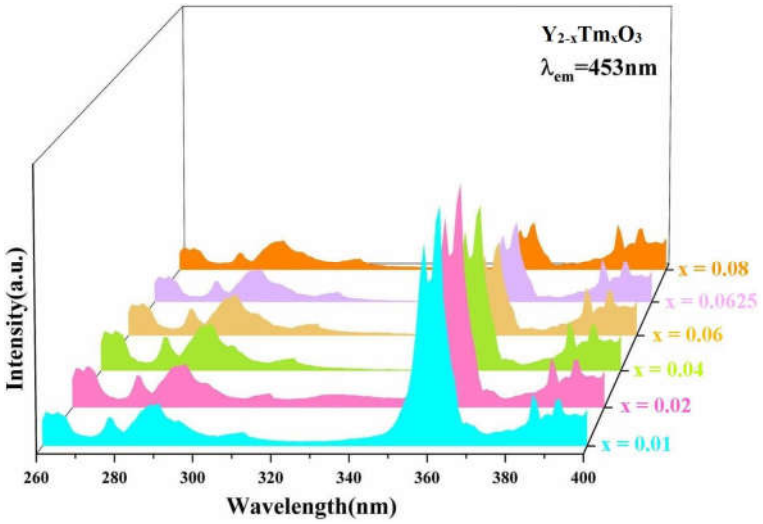


Figure S2. Dependence of photoluminescence excitation intensities of $Y_{2-x}Tm_xO_3$ at λ_{em} = 453 nm at different Tm^{3+} concentrations.

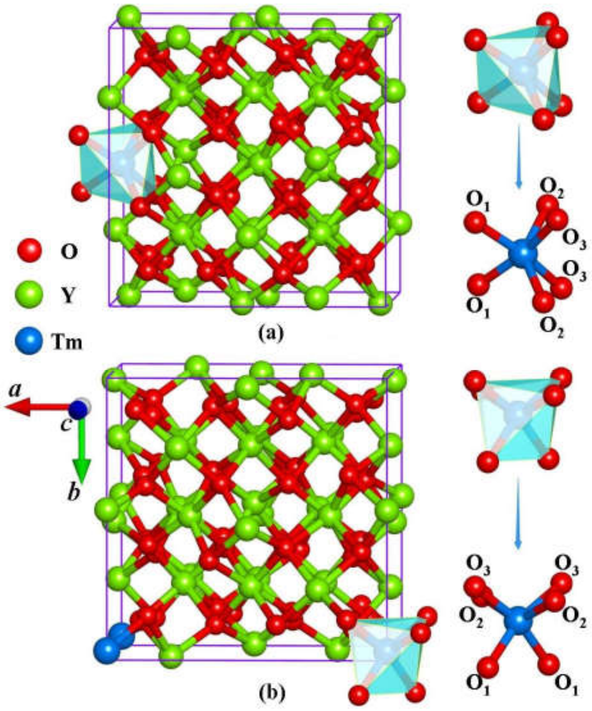


Figure S3. The metastable structures (a) and (b) for Y_2O_3:Tm^{3+}.

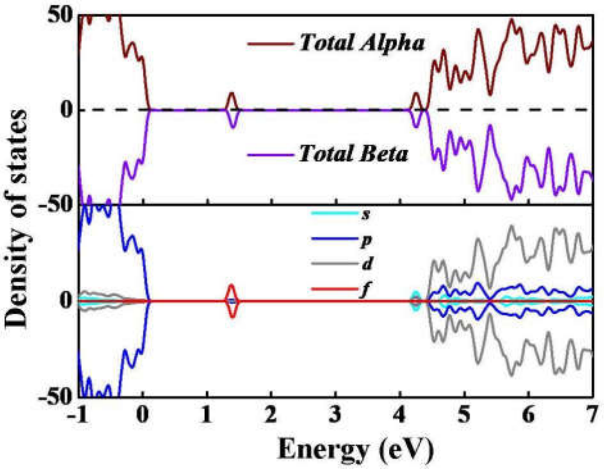


Figure S4. The calculated total and partial density of states (DOS) of Y_2O_3:Tm^{3+}.

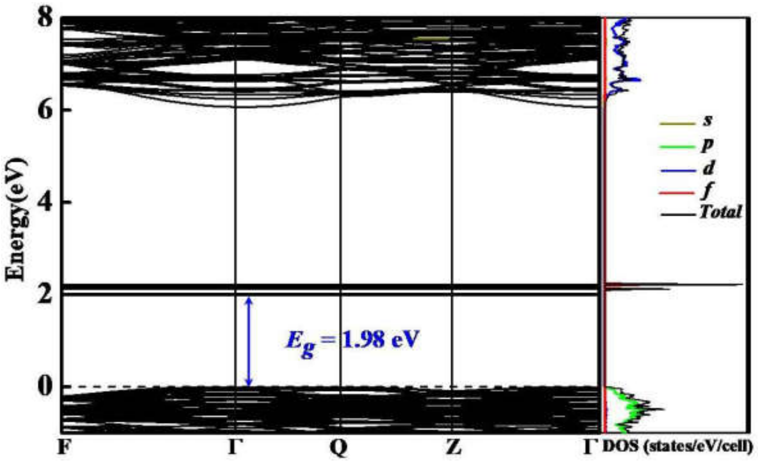
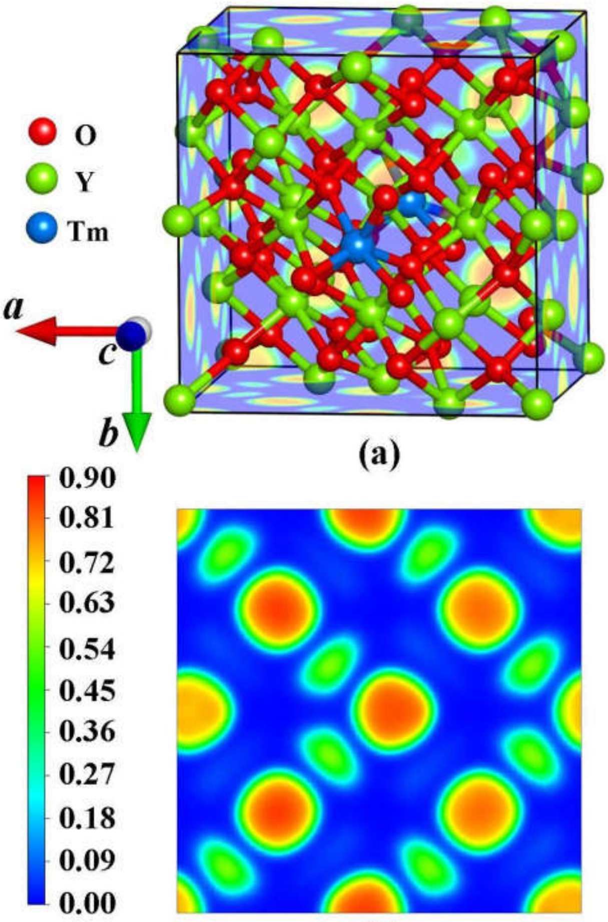


Figure S5. The calculated band structure and partial density of states (DOS) of Y_2O_3:Tm^{3+} using the modified Becke and Johnson (BJ) method as implemented in the reliable Wien2k program.



(a)

(b)

Figure S6. (a) Electron localization function (ELF) of Y_2O_3:Tm crystal. (b) The ELF of ⟨001⟩ plane for Y_2O_3:Tm^{3+}.

Table S1. Coordinates of all atoms for the ground state Y_2O_3:Tm^{3+}.

Atom	*x*	*y*	*z*	Wyckoff site symmetry
Tm	0.50000	0.53271	0.00000	1c
Y1	0.00021	0.25008	0.24996	2e
Y2	0.00011	0.74992	0.75007	2e
Y3	0.50052	0.74915	0.24916	2e
Y4	0.49929	0.25113	0.75091	2e
Y9	0.71746	-0.00002	0.24997	2e
Y11	-0.21806	0.49995	0.24946	2e
Y13	0.00000	0.96742	0.00000	1a
Y14	0.00000	0.03263	0.50000	1b
Y15	0.50000	0.46753	0.50000	1d
Y16	-0.25080	0.25101	0.96759	2e
Y17	-0.25005	0.74994	0.53285	2e
Y20	-0.21773	0.00012	0.75024	2e
Y21	0.28374	0.50046	0.24879	2e
Y24	0.50000	0.03267	0.00000	2e
Y25	0.00000	0.53242	0.50000	1b
Y26	0.00000	0.46732	0.00000	1a
Y27	0.50000	0.96727	0.50000	1d
Y28	-0.25076	0.74916	0.03229	2e
Y29	0.25014	0.25006	0.53229	2e
O1	0.14107	0.15168	0.37963	2e
O2	0.63790	0.65052	0.88192	2e
O3	-0.14159	0.84782	0.87970	2e
O4	0.35908	0.34850	0.37863	2e
O9	0.13050	0.39128	0.15146	2e
O10	0.62949	0.89073	0.65222	2e
O11	0.62948	0.11024	0.84834	2e
O12	0.13012	0.60894	0.34791	2e
O17	-0.09839	0.37978	0.39089	2e
O18	0.40155	0.87961	0.89106	2e
O21	0.40191	0.12052	0.60933	2e
O22	-0.09867	0.62018	0.10888	2e
O25	0.35889	0.84796	0.62032	2e
O26	-0.14078	0.34816	0.12034	2e
O27	0.64119	0.15153	0.12008	2e
O28	0.14134	0.65150	0.62041	2e

O33	0.37211	0.60606	0.85121	2e
O34	-0.12976	0.10908	0.34832	2e
O35	-0.12974	0.89074	0.15163	2e
O36	0.37005	0.39117	0.65103	2e
O41	0.59808	0.62023	0.60948	2e
O42	0.09882	0.12034	0.10864	2e
O45	0.09832	0.87977	0.39088	2e
O46	0.59545	0.38271	0.89394	2e

Table S2. Lattice constants *a*, *b* and *c*, unit-cell volume, relative energies for the ground state and metastable Y_2O_3:Tm^{3+} crystals.

	Space group	*a* (Å)	*b* (Å)	*c* (Å)	*V* (Å^3)	Δ*E* (meV)
Y_2O_3:Tm^{3+}	*P*2	10.6765	10.6749	10.6758	1216.73	0
Isomer (a)	*P*1	10.6748	10.6758	10.6762	1216.68	0.358
Isomer (b)	*P*2	10.6754	10.6745	10.6752	1216.49	0.401

Table S3. Comparision between the calculated and experimental energy levels of Tm^{3+} in Y_2O_3 (in units of cm^{-1}).

		This work		Other	
$^{2s+1}L_J$	E_{expt}[1]	E_{calc}	ΔE	ΔE[1]	ΔE
$^{3}H_6$	0	−51.0	51.0	73.8	−73.8
$^{3}F_4$	5644.3	5690.6	−46.3	5636.7	7.6
$^{3}H_5$	8091.6	8135.6	−44.0	8231.3	−139.7
$^{3}H_4$	12500.3	12520.7	−20.4	12535.4	−35.1
$^{3}F_3$	14349.1	14330.2	18.9	14143.8	205.3
$^{3}F_2$	14854.9	14837.3	17.6	14724.7	130.2
$^{1}G_4$	21324.0	21296.9	27.1	21319.1	4.9
$^{1}D_2$	27522.9	27515.8	7.1	27555.7	−32.8
$^{1}I_6$	—	34388.4	—	33899.0	2.3
$^{3}P_0$	34955.2	34958.6	−3.4	35095.8	140.6
$^{3}P_1$	35791.0	35781.5	9.6	35822.8	−31.8
$^{3}P_2$	37638.3	37655.6	−17.3	37673.3	−35.0
$^{1}S_0$	—	76562.5	—	77506.1	—

Table S4. Calculated wavelengths (λ), ED (A_{ED}) and MD (A_{MD}) radiative decay rates, branching ratios (β) and radiative lifetimes (τ) for spontaneous emission transitions between the first 9 excited states of Tm^{3+} in $\mathrm{Y_2O_3}$. Available theoretical and experimental results are also listed for comparison.

		λ (nm)		A_{ED} (s^{-1})		A_{MD} (s^{-1})		β		τ (μs)	
Transition		Present	Other	Present	Other	Present	Other	Present	Other	Present	Other
$^3F_4$	$^3H_6$	1742	1698[3], 1632[5]	353.6	305.5[3], 277[6]	0	0[3], 0[6]	1.00	1.00[3]	2828	3270[3], 3500[4], 3610[6]
$^3H_5$	$^3H_6$	1221	1199[3]	284.5	234.7[3], 237[6]	101.1	85.7[3], 84.1[6]	0.98	0.99[3]	2543	3090[3], 3310[6]
	$^3F_4$	4090	4080[3]	7.4	3.1[3]	0.2	0.2[3]	0.02	0.01[3]		
$^3H_4$	$^3H_6$	795	788[3], 766[4]	1568.4	1197.1[3], 1534[6]	0	0[3]	0.88	0.88[3]	560	734[3], 310[4], 639[6]
	$^3F_4$	1464	1470[3], 1550[4]	145.1	110.4[3]	26.0	19.8[3], 31.2[6]	0.10	0.09[3]		
	$^3H_5$	2280	2300[3]	35.0	26.3[3]	11.6	9.2[3]	0.02	0.03[3]		
$^3F_3$	$^3H_6$	695	690[3]	2029.1	1620.2[3], 2467[6]	0	0[3]	0.76	0.77[3]	375	473[3], 394[6]
	$^3F_4$	1157	1162[3]	52.3	40.0[3]	74.5	61.0[3], 67.8[6]	0.05	0.05[3]		
	$^3H_5$	1614	1625[3]	509.8	389.8[3]	0	0[3]	0.19	0.18[3]		
	$^3H_4$	5526		4.3		0.3		0			

$^3F_2$	$^3H_6$	672	658[3], 680[5]	502.6	424.1[3]	0	0[3]	0.28	0.03[3]	566	705[3]
	$^3F_4$	1093	1074[3]	1016.7	781.1[3]	0	0[3]	0.58	0.55[3]		
	$^3H_5$	1492	1458[3]	230.0	193.3[3]	0	0[3]	0.13	0.14[3]		
	$^3H_4$	4317	3988[3]	18.4	19.0[3]	0	0[3]	0.01	0.01[3]		
	$^3F_3$	19720		0.02		0.02		0			
$^1G_4$	$^3H_6$	468	465[3], 464[4], 476[5]	1646.4	1236.5[3]	0	0[3]	0.51	0.50[3]	312	408[3], 170[4], 286[6]
	$^3F_4$	641	640[3]	184.5	134.9[3]	14.0	11.7[3]	0.06	0.06[3]		
	$^3H_5$	760	760[3]	762.3	591.4[3]	160.7	131.2[3]	0.29	0.29[3]		
	$^3H_4$	1139	1134[3]	310.1	247.4[3]	41.6	33.3[3]	0.11	0.11[3]		
	$^3F_3$	1435	1426[3]	57.9	46.9[3]	4.8	3.7[3]	0.02	0.02[3]		
	$^3F_2$	1548		20.0		0		0.01			
$^1D_2$	$^3H_6$	363	360[3], 363[4]	10720.2	8436.6[3]	0	0[3]	0.26	0.25[3]	24	30[3], 8.5[4], 17[6]
	$^3F_4$	458	457[3], 450[5]	24981.6	20121.3[3]	0	0[3]	0.60	0.61[3]		
	$^3H_5$	516		94.5		0		0			
	$^3H_4$	667	663[3], 654[5]	2341.3	1648.0[3]	0	0[3]	0.05	0.05[3]		
	$^3F_3$	758	753[3]	1749.1	1381.1[3]	112.9	88.2[3]	0.04	0.04[3]		
	$^3F_2$	789	795[3]	1420.9	1083.1[3]	69.9	52.2[3]	0.04	0.03[3]		

	$^{1}G_{4}$	1608		235.9	0	0.01		
$^{1}I_{6}$	$^{3}H_{6}$	290	290[4]	1687.4	78.6	0.09	53	25[4], 34[6]
	$^{3}F_{4}$	348	360[4]	10374.6	0	0.55		
	$^{3}H_{5}$	381		70.6	25.1	0.01		
	$^{3}H_{4}$	457		2945.3	0	0.16		
	$^{3}F_{3}$	498		26.4	0	0		
	$^{3}F_{2}$	511		781.0	0	0.04		
	$^{1}G_{4}$	764		2683.1	0	0.14		
	$^{1}D_{2}$	1455		70.0	0	0.01		

Table S5. The calculated spontaneous emission rates and MD oscillator strengths for transitions $\psi(SLJ\Gamma_i) \rightarrow \psi(S'L'J'\Gamma'_i)$ between different levels of Tm^{3+} in Y_2O_3.

$^{2S+1}L_J$ (Sta.)[a]	$S'L'J'$ (Sta.)	λ (nm)	A'_{MD} (s^{-1})	$P_{MD} \times 10^8$
$^3H_5$ (23)	$^3H_6$ (1)	1210	36.00	79.09
	$^3H_6$ (2)	1215	21.98	48.70
	$^3H_6$ (3)	1223	18.31	41.11
$^3H_5$ (24)	$^3H_6$ (1)	1203	16.00	34.72
	$^3H_6$ (2)	1208	36.74	80.44
	$^3H_6$ (4)	1235	12.18	27.89
	$^3H_6$ (5)	1236	14.00	32.09
$^3H_5$ (25)	$^3H_6$ (1)	1200	11.26	24.29
	$^3H_6$ (3)	1212	31.73	69.96
	$^3H_6$ (4)	1231	16.59	37.71
	$^3H_6$ (5)	1232	18.96	43.16
	$^3H_6$ (8)	1264	3.01	7.21
$^3H_5$ (26)	$^3H_6$ (3)	1193	8.28	17.66
	$^3H_6$ (4)	1211	32.12	70.62
	$^3H_6$ (6)	1233	6.75	15.38
	$^3H_6$ (7)	1236	15.85	36.32
	$^3H_6$ (8)	1243	7.14	16.54
	$^3H_6$ (9)	1252	18.61	43.71
$^3H_5$ (27)	$^3H_6$ (2)	1184	5.79	12.17
	$^3H_6$ (3)	1192	2.36	5.03
	$^3H_6$ (5)	1211	27.67	60.83
	$^3H_6$ (6)	1232	5.05	11.49
	$^3H_6$ (7)	1235	8.25	18.89
	$^3H_6$ (8)	1242	28.43	65.79
	$^3H_6$ (9)	1251	16.99	39.86
$^3H_5$ (28)	$^3H_6$ (3)	1184	7.54	15.86
	$^3H_6$ (5)	1203	5.44	11.80
	$^3H_6$ (6)	1223	29.71	66.68
	$^3H_6$ (7)	1227	33.75	76.17
	$^3H_6$ (8)	1234	5.18	11.83
	$^3H_6$ (9)	1242	11.30	26.15
	$^3H_6$ (11)	1274	3.66	8.90
$^3H_5$ (29)	$^3H_6$ (4)	1200	5.80	12.53
	$^3H_6$ (6)	1221	29.60	66.16
	$^3H_6$ (7)	1225	20.69	46.52
	$^3H_6$ (8)	1231	16.02	36.40
	$^3H_6$ (9)	1240	18.24	42.04

$^3H_5$ (30)	$^3H_6$ (4)	1172	5.67	11.68
	$^3H_6$ (6)	1192	2.19	4.66
	$^3H_6$ (7)	1195	3.79	8.13
	$^3H_6$ (8)	1202	19.02	41.17
	$^3H_6$ (10)	1236	20.41	46.75
	$^3H_6$ (11)	1240	39.82	91.82
	$^3H_6$ (13)	1258	10.52	24.95
$^3H_5$ (31)	$^3H_6$ (5)	1172	6.25	12.88
	$^3H_6$ (9)	1210	19.58	42.97
	$^3H_6$ (10)	1236	42.71	97.82
	$^3H_6$ (11)	1240	22.66	52.24
	$^3H_6$ (12)	1257	7.81	18.50
$^3H_5$ (32)	$^3H_6$ (8)	1181	3.87	8.10
	$^3H_6$ (10)	1214	20.78	45.94
	$^3H_6$ (12)	1234	44.65	102.00
	$^3H_6$ (13)	1235	35.96	82.24
$^3H_5$ (33)	$^3H_6$ (9)	1189	4.07	8.62
	$^3H_6$ (11)	1218	22.18	49.31
	$^3H_6$ (12)	1234	36.26	82.76
	$^3H_6$ (13)	1235	44.43	101.54
$^3H_4$ (34)	$^3F_4$ (14)	1446	8.63	27.07
	$^3F_4$ (15)	1461	10.23	32.72
	$^3F_4$ (18)	1535	3.45	12.18
	$^3H_5$ (23)	2340	3.20	26.30
$^3H_4$ (35)	$^3F_4$ (16)	1456	13.13	41.75
	$^3F_4$ (20)	1529	6.76	23.68
	$^3H_5$ (24)	2309	3.41	27.23
$^3H_4$ (36)	$^3F_4$ (14)	1418	8.99	27.08
	$^3F_4$ (16)	1449	9.90	31.14
	$^3F_4$ (19)	1519	3.96	13.68
$^3H_4$ (37)	$^3F_4$ (14)	1392	3.04	8.83
	$^3F_4$ (18)	1474	9.68	31.56
	$^3F_4$ (22)	1517	6.02	20.76
	$^3H_5$ (28)	2342	3.86	31.71
	$^3H_5$ (29)	2351	3.64	30.18
$^3H_4$ (38)	$^3F_4$ (15)	1396	9.50	27.77
	$^3F_4$ (17)	1459	8.58	27.41
	$^3F_4$ (20)	1481	3.05	10.04
	$^3H_5$ (28)	2316	3.23	25.95

$^3H_4$ (39)	$^3F_4$ (15)	1390	4.09	11.86
	$^3F_4$ (17)	1453	7.21	22.83
	$^3F_4$ (18)	1457	4.21	13.42
	$^3F_4$ (22)	1499	4.52	15.21
	$^3H_5$ (31)	2420	4.35	38.19
$^3H_4$ (40)	$^3F_4$ (14)	1369	3.19	8.95
	$^3F_4$ (19)	1464	8.95	28.77
	$^3F_4$ (20)	1465	3.47	11.15
	$^3F_4$ (21)	1471	6.72	21.79
	$^3H_5$ (30)	2396	5.19	44.68
$^3H_4$ (41)	$^3F_4$ (17)	1416	7.73	23.22
	$^3F_4$ (18)	1420	4.67	14.11
	$^3F_4$ (22)	1460	7.65	24.44
	$^3H_5$ (32)	2399	4.75	40.97
	$^3H_5$ (33)	2401	4.92	42.54
$^3H_4$ (42)	$^3F_4$ (19)	1432	9.24	28.41
	$^3F_4$ (20)	1434	4.23	13.04
	$^3F_4$ (21)	1439	8.88	27.56
	$^3H_5$ (32)	2393	4.61	39.53
	$^3H_5$ (33)	2395	4.14	35.60
$^3F_3$ (43)	$^3F_4$ (14)	1117	14.46	27.07
	$^3F_4$ (15)	1126	8.25	15.67
	$^3F_4$ (17)	1167	15.46	31.55
	$^3F_4$ (18)	1170	7.55	15.48
	$^3F_4$ (20)	1180	6.50	13.57
	$^3F_4$ (21)	1184	15.75	33.10
	$^3F_4$ (22)	1196	5.28	11.32
$^3F_3$ (43)	$^3F_4$ (14)	1114	21.13	39.32
	$^3F_4$ (15)	1123	25.87	48.88
	$^3F_4$ (16)	1133	18.65	35.91
	$^3F_4$ (19)	1176	4.43	9.19
	$^3F_4$ (22)	1193	3.07	6.54
$^3F_3$ (45)	$^3F_4$ (14)	1109	9.66	17.82
	$^3F_4$ (16)	1128	5.20	9.93
	$^3F_4$ (17)	1158	11.44	22.99
	$^3F_4$ (18)	1161	10.84	21.90
	$^3F_4$ (19)	1170	22.46	46.13
	$^3F_4$ (21)	1175	3.85	7.96
	$^3F_4$ (22)	1187	8.13	17.17
$^3F_3$ (46)	$^3F_4$ (15)	1117	6.72	12.58
	$^3F_4$ (16)	1127	5.39	10.27
	$^3F_4$ (17)	1157	19.66	39.45

	$^3F_4$ (19)	1170	12.94	26.53
	$^3F_4$ (20)	1170	12.49	25.65
	$^3F_4$ (21)	1174	9.02	18.64
$^3F_3$ (47)	$^3F_4$ (14)	1107	3.59	6.60
	$^3F_4$ (16)	1126	13.66	25.96
	$^3F_4$ (18)	1158	13.38	26.92
	$^3F_4$ (19)	1168	11.92	24.39
	$^3F_4$ (20)	1169	7.31	14.98
	$^3F_4$ (21)	1172	7.95	16.39
	$^3F_4$ (22)	1184	13.24	27.84
$^3F_3$ (48)	$^3F_4$ (15)	1112	7.54	13.98
	$^3F_4$ (18)	1154	10.43	20.85
	$^3F_4$ (20)	1165	19.42	39.49
	$^3F_4$ (21)	1168	16.36	33.48
	$^3F_4$ (22)	1180	16.45	34.35
$^3F_3$ (49)	$^3F_4$ (16)	1119	5.69	10.67
	$^3F_4$ (17)	1148	6.91	13.64
	$^3F_4$ (18)	1151	10.42	20.68
	$^3F_4$ (19)	1160	5.25	10.59
	$^3F_4$ (20)	1161	13.84	27.96
	$^3F_4$ (21)	1164	8.25	16.78
	$^3F_4$ (22)	1176	16.11	33.43
$^1G_4$ (55)	$^3F_4$ (15)	657	7.28	4.71
	$^3F_4$ (16)	661	5.77	3.78
	$^3F_4$ (17)	671	5.95	4.02
	$^3H_5$ (23)	791	6.89	6.46
	$^3H_5$ (24)	794	34.83	32.89
	$^3H_5$ (25)	795	3.88	3.68
	$^3H_5$ (26)	804	39.27	38.07
	$^3H_5$ (27)	804	27.65	26.83
	$^3H_5$ (28)	808	14.37	14.07
	$^3H_5$ (29)	809	23.09	22.67
	$^3H_4$ (34)	1194	7.72	16.51
	$^3H_4$ (36)	1215	9.00	19.91
$^1G_4$ (56)	$^3F_4$ (14)	650	4.41	2.79
	$^3F_4$ (19)	670	3.84	2.59
	$^3H_5$ (23)	784	16.54	15.24
	$^3H_5$ (25)	789	42.25	39.39
	$^3H_5$ (26)	797	26.58	25.33
	$^3H_5$ (27)	798	38.36	36.58
	$^3H_5$ (28)	801	7.07	6.80
	$^3H_5$ (29)	802	11.17	10.77
	$^3H_4$ (34)	1179	10.89	22.69
	$^3H_4$ (38)	1225	11.94	26.86

	$^{3}H_{4}$ (39)	1230	3.39	7.69
$^{1}G_{4}$ (57)	$^{3}F_{4}$ (18)	665	6.07	4.03
	$^{3}H_{5}$ (23)	782	57.15	52.37
	$^{3}H_{5}$ (24)	785	39.66	36.62
	$^{3}H_{5}$ (25)	786	31.79	29.47
	$^{3}H_{5}$ (26)	795	3.85	3.65
	$^{3}H_{5}$ (29)	800	4.48	4.29
	$^{3}H_{4}$ (35)	1189	15.88	33.65
	$^{3}H_{4}$ (36)	1194	12.46	26.62
	$^{3}H_{4}$ (38)	1219	3.33	7.42
$^{1}G_{4}$ (58)	$^{3}F_{4}$ (20)	653	3.63	2.32
	$^{3}H_{5}$ (23)	760	23.69	20.51
	$^{3}H_{5}$ (24)	763	27.70	24.16
	$^{3}H_{5}$ (26)	772	19.25	17.22
	$^{3}H_{5}$ (27)	773	4.87	4.36
	$^{3}H_{5}$ (28)	776	41.35	37.34
	$^{3}H_{5}$ (29)	777	19.65	17.78
	$^{3}H_{5}$ (32)	798	4.07	3.89
	$^{3}H_{4}$ (34)	1126	7.19	13.66
	$^{3}H_{4}$ (37)	1161	21.20	42.82
	$^{3}H_{4}$ (40)	1177	3.67	7.63
$^{1}G_{4}$ (59)	$^{3}F_{4}$ (19)	651	3.66	2.33
	$^{3}H_{5}$ (25)	763	10.66	9.30
	$^{3}H_{5}$ (26)	771	15.13	13.48
	$^{3}H_{5}$ (28)	775	42.13	37.89
	$^{3}H_{5}$ (29)	775	40.67	36.67
	$^{3}H_{5}$ (30)	788	21.70	20.18
	$^{3}H_{5}$ (31)	788	4.10	3.81
	$^{3}H_{5}$ (33)	797	11.66	11.10
	$^{3}H_{4}$ (34)	1122	3.92	7.41
	$^{3}H_{4}$ (38)	1164	16.71	33.92
	$^{3}H_{4}$ (39)	1168	8.20	16.77
	$^{3}H_{4}$ (41)	1193	4.21	8.99
$^{1}G_{4}$ (60)	$^{3}F_{4}$ (21)	650	3.07	1.94
	$^{3}H_{5}$ (24)	758	8.56	7.37
	$^{3}H_{5}$ (25)	760	10.27	8.88
	$^{3}H_{5}$ (27)	768	48.82	43.15
	$^{3}H_{5}$ (28)	771	12.03	10.72
	$^{3}H_{5}$ (29)	772	6.12	5.47
	$^{3}H_{5}$ (30)	784	32.86	30.30
	$^{3}H_{5}$ (31)	784	29.18	26.91
	$^{3}H_{5}$ (33)	793	4.74	4.47
	$^{3}H_{4}$ (35)	1128	11.33	21.62
	$^{3}H_{4}$ (36)	1133	6.68	12.86
	$^{3}H_{4}$ (39)	1160	5.15	10.39

	$^3H_4$ (41)	1185	8.04	16.93
	$^3H_4$ (42)	1187	3.31	6.99
$^1G_4$ (61)	$^3F_4$ (17)	641	3.37	2.08
	$^3H_5$ (25)	754	8.84	7.54
	$^3H_5$ (26)	762	17.47	15.21
	$^3H_5$ (29)	767	14.35	12.65
	$^3H_5$ (30)	779	35.09	31.89
	$^3H_5$ (31)	779	53.74	48.86
	$^3H_5$ (32)	788	12.71	11.82
	$^3H_4$ (36)	1121	6.15	11.59
	$^3H_4$ (37)	1138	4.49	8.71
	$^3H_4$ (40)	1154	14.08	28.10
	$^3H_4$ (42)	1174	13.08	27.02
	$^3F_3$ (45)	1438	3.41	10.55
$^1G_4$ (62)	$^3F_4$ (22)	647	5.37	3.37
	$^3H_5$ (23)	746	4.26	3.56
	$^3H_5$ (25)	750	4.74	4.00
	$^3H_5$ (30)	774	34.26	30.79
	$^3H_5$ (32)	783	44.41	40.82
	$^3H_5$ (33)	783	61.75	56.79
	$^3H_4$ (40)	1144	12.65	24.81
	$^3H_4$ (42)	1164	16.92	34.37
$^1G_4$ (63)	$^3F_4$ (21)	640	4.13	2.54
	$^3H_5$ (28)	757	3.42	2.93
	$^3H_5$ (29)	757	3.74	3.21
	$^3H_5$ (31)	769	33.99	30.15
	$^3H_5$ (32)	778	60.71	55.05
	$^3H_5$ (33)	778	46.40	42.10
	$^3H_4$ (37)	1118	4.25	7.96
	$^3H_4$ (39)	1128	10.78	20.54
	$^3H_4$ (41)	1151	20.30	40.32
$^1D_2$ (64)	$^3F_3$ (43)	763	40.34	35.19
	$^3F_3$ (44)	764	32.09	28.10
	$^3F_3$ (45)	767	3.26	2.88
	$^3F_3$ (46)	767	27.46	24.22
	$^3F_2$ (50)	793	47.53	44.79
	$^3F_2$ (51)	801	10.23	9.84
	$^3F_2$ (52)	804	4.86	4.71
	$^3F_2$ (53)	811	5.39	5.31
$^1D_2$ (65)	$^3F_3$ (43)	761	30.31	26.32
	$^3F_3$ (44)	763	19.80	17.26
	$^3F_3$ (45)	765	7.19	6.30
	$^3F_3$ (47)	766	26.92	23.67
	$^3F_3$ (49)	769	21.67	19.23

	$^3F_2$ (51)	799	25.08	24.00
	$^3F_2$ (52)	802	17.64	17.03
	$^3F_2$ (53)	809	8.43	8.26
$^1D_2$ (66)	$^3F_3$ (44)	757	13.93	11.97
	$^3F_3$ (45)	760	45.85	39.66
	$^3F_3$ (46)	760	35.18	30.45
	$^3F_3$ (47)	761	24.42	21.18
	$^3F_2$ (50)	785	39.00	36.05
	$^3F_2$ (51)	793	5.56	5.24
	$^3F_2$ (52)	797	15.18	14.45
	$^3F_2$ (54)	807	8.20	8.02
$^1D_2$ (67)	$^3F_3$ (44)	751	17.55	14.86
	$^3F_3$ (45)	754	15.34	13.06
	$^3F_3$ (47)	755	22.60	19.30
	$^3F_3$ (48)	756	35.14	30.15
	$^3F_3$ (49)	758	22.91	19.74
	$^3F_2$ (51)	787	37.43	34.74
	$^3F_2$ (52)	790	14.26	13.35
	$^3F_2$ (53)	796	19.62	18.64
$^1D_2$ (68)	$^3F_3$ (45)	752	4.56	3.87
	$^3F_3$ (46)	753	14.20	12.06
	$^3F_3$ (48)	755	36.86	31.50
	$^3F_3$ (49)	757	30.38	26.07
	$^3F_2$ (51)	785	6.50	6.00
	$^3F_2$ (52)	789	11.48	10.70
	$^3F_2$ (53)	795	9.69	9.17
	$^3F_2$ (54)	799	36.08	34.55
$^1I_6$ (69)	$^3H_6$ (11)	301	5.43	0.74
	$^3F_4$ (17)	359	3.26	0.63
	$^3H_5$ (28)	394	8.25	1.92
	$^3H_5$ (29)	395	4.95	1.16
$^1I_6$ (70)	$^3F_4$ (14)	354	3.24	0.61
	$^3H_5$ (28)	394	7.14	1.67
	$^3H_5$ (29)	395	5.97	1.39
$^1I_6$ (71)	$^3H_5$ (31)	392	8.68	1.99
	$^3H_5$ (32)	394	4.04	0.94
	$^1G_4$ (58)	777	3.22	2.91
$^1I_6$ (72)	$^3F_4$ (15)	349	3.03	0.55
	$^3H_5$ (30)	391	7.93	1.81
	$^3H_5$ (33)	393	4.27	0.99
$^1I_6$ (73)	$^3H_5$ (27)	386	5.16	1.16

	$^3H_5$ (30)	390	6.04	1.38
	$^3H_5$ (33)	393	5.16	1.19
$^1I_6$ (74)	$^3H_5$ (26)	386	3.46	0.77
	$^3H_5$ (31)	390	4.93	1.12
	$^3H_5$ (32)	392	7.37	1.70
	$^3H_5$ (33)	392	3.07	0.71
$^1I_6$ (75)	$^3F_4$ (19)	352	4.49	0.83
	$^3H_5$ (23)	381	4.19	0.91
	$^3H_5$ (25)	382	6.58	1.44
	$^1G_4$ (61)	773	3.95	3.54
$^1I_6$ (76)	$^3H_5$ (23)	379	10.96	2.36
	$^3H_5$ (24)	379	7.50	1.62
$^1I_6$ (77)	$^3H_5$ (23)	378	7.76	1.66
	$^3H_5$ (24)	378	10.84	2.33
$^1I_6$ (78)	$^3H_5$ (25)	376	4.78	1.01
	$^3H_5$ (26)	378	4.87	1.04
	$^3H_5$ (29)	379	3.13	0.68
	$^3H_5$ (30)	382	4.66	1.02
$^1I_6$ (79)	$^3H_5$ (25)	376	6.01	1.27
	$^3H_5$ (31)	382	3.66	0.80
$^3P_0$ (80)	$^3F_4$ (17)	343	3.01	0.53
	$^3F_4$ (19)	344	6.38	1.13
	$^3F_4$ (20)	344	5.86	1.04
	$^3F_4$ (21)	344	5.45	0.97
$^1I_6$ (81)	$^3H_5$ (26)	373	3.05	0.64
	$^3H_5$ (27)	373	3.49	0.73
	$^3H_5$ (28)	374	3.26	0.68
	$^3H_5$ (29)	374	3.71	0.78
	$^3H_5$ (32)	379	4.93	1.06
$^1I_6$ (82)	$^3H_5$ (27)	373	4.00	0.83
	$^3H_5$ (28)	374	3.12	0.66
	$^3H_5$ (29)	374	3.64	0.76
	$^3H_5$ (33)	379	4.32	0.93
$^3P_1$ (83)	$^3H_5$ (28)	365	4.50	0.90
	$^3F_2$ (51)	482	5.86	2.04
	$^3F_2$ (53)	485	6.97	2.46
	$^3F_2$ (54)	487	9.53	3.39
	$^3P_2$ (66)	1227	12.92	29.16
	$^3P_2$ (67)	1242	13.96	32.30

	$^1D_2$ (68)	1246	11.57	26.95
$^3P_1$ (84)	$^3H_4$ (42)	433	3.27	0.92
	$^3F_3$ (47)	465	3.06	0.99
	$^3F_2$ (50)	474	5.02	1.69
	$^3F_2$ (51)	477	9.77	3.34
	$^3F_2$ (52)	479	10.83	3.72
	$^3F_2$ (54)	482	6.19	2.16
	$^3P_2$ (64)	1182	11.20	23.44
	$^3P_2$ (65)	1186	9.77	20.60
	$^3P_2$ (67)	1213	4.34	9.59
	$^1D_2$ (68)	1217	8.99	19.98
$^3P_1$ (85)	$^3F_3$ (49)	465	3.27	1.06
	$^3F_2$ (50)	473	12.79	4.29
	$^3F_2$ (51)	476	3.09	1.05
	$^3F_2$ (52)	477	4.54	1.55
	$^3F_2$ (53)	479	9.29	3.20
	$^3P_2$ (64)	1172	10.61	21.84
	$^3P_2$ (65)	1176	11.01	22.82
	$^3P_2$ (66)	1189	8.50	17.99
	$^3P_2$ (67)	1203	3.07	6.67
$^3P_2$ (86)	$^3F_3$ (43)	435	46.35	13.14
	$^3F_3$ (44)	435	10.01	2.84
	$^3F_3$ (45)	436	40.70	11.60
	$^3F_3$ (47)	436	38.68	11.04
	$^3F_2$ (53)	450	3.88	1.18
	$^3P_2$ (64)	1011	51.55	79.05
	$^3P_2$ (66)	1024	38.93	61.19
$^3P_2$ (87)	$^3F_3$ (43)	432	28.70	8.01
	$^3F_3$ (44)	432	16.49	4.61
	$^3F_3$ (46)	433	31.93	8.97
	$^3F_3$ (47)	433	4.92	1.39
	$^3F_3$ (48)	434	24.43	6.89
	$^3F_3$ (49)	434	24.70	6.98
	$^3F_2$ (54)	448	4.35	1.31
	$^3P_2$ (64)	994	7.08	10.49
	$^3P_2$ (65)	997	43.92	65.44
	$^3P_2$ (66)	1006	8.62	13.07
	$^3P_2$ (67)	1016	25.32	39.21
	$^1D_2$ (68)	1019	6.64	10.34
$^3P_2$ (88)	$^3F_3$ (43)	430	18.65	5.16
	$^3F_3$ (44)	430	15.57	4.32
	$^3F_3$ (45)	431	9.69	2.70
	$^3F_3$ (46)	431	34.16	9.51
	$^3F_3$ (47)	431	7.00	1.95

	$^3F_3$ (48)	432	29.99	8.38
	$^3F_3$ (49)	432	14.93	4.18
	$^3F_2$ (54)	446	3.29	0.98
	$^3P_2$ (64)	984	7.32	10.62
	$^3P_2$ (65)	987	21.12	30.83
	$^3P_2$ (66)	996	25.18	37.42
	$^3P_2$ (67)	1006	28.66	43.47
	$^1D_2$ (68)	1009	5.21	7.95
$^3P_2$ (89)	$^3F_3$ (44)	425	17.79	4.82
	$^3F_3$ (45)	426	34.37	9.35
	$^3F_3$ (46)	426	23.15	6.30
	$^3F_3$ (47)	426	37.51	10.22
	$^3F_3$ (49)	427	5.79	1.59
	$^3P_2$ (64)	959	12.33	17.00
	$^3P_2$ (65)	962	8.23	11.41
	$^3P_2$ (67)	980	36.83	53.00
	$^1D_2$ (68)	982	13.37	19.35
$^3P_2$ (90)	$^3F_3$ (44)	424	12.53	3.37
	$^3F_3$ (45)	424	6.57	1.77
	$^3F_3$ (47)	425	9.29	2.51
	$^3F_3$ (48)	425	42.38	11.49
	$^3F_3$ (49)	426	44.02	11.96
	$^3P_2$ (64)	950	6.55	8.86
	$^3P_2$ (66)	961	23.42	32.46
	$^1D_2$ (68)	973	40.82	57.98

[a] Only transitions between 300-2500 nm with emission rates $A'_{MD} > 3$ s^{-1} are listed.

Appendix – Method and equations for the calculations of energy levels and transition intensities

The model Hamiltonian for Tm^{3+} is defined as[7]-[8]

$$H_f = E_{AVE} + \sum_{k=2,4,6} F^k f_k + \zeta_{4f} \times \sum_i \vec{l}_i \cdot \vec{s}_i + \alpha L(L+1) + \beta G(G_2) + \gamma G(R_7)$$
$$+ \sum_{j=0,2,4} M^j m_j + \sum_{k=2,4,6} P^k p_k$$

(A1)

where E_{AVE} represents the barycenter energy of the $4f^3$ configuration. The next seven terms represent the Coulomb repulsion, spin-orbit, two-body, three-body, spin-other-orbit and electrostatically correlated spin-orbit interactions. Moreover, F^k and ζ_{4f} are the radial parts of the electrostatic and spin-orbit coupling constant. Two-body parameters are represented by α, β, and γ. $G(G2)$ and $G(R7)$ represent the eigenvalues of the Casimir's operators for the Lie groups G_2 and R_7. The remaining parameters, M^j and P^k, are used to represent the Marvin integrals and spin-orbit perturbations.

The crystal field interaction H_{CF} for Tm^{3+} in Y_2O_3, in the form of Wybourne normalization, can be expressed as[9]

$$\begin{aligned}
H_{CF} = {} & B_2^0 C_2^0 + ReB_2^2(C_2^2 + C_2^{-2}) + B_4^0 C_4^0 \\
& + ReB_4^2(C_4^2 + C_4^{-2}) + iImB_4^2(C_4^2 - C_4^{-2}) \\
& + ReB_4^4(C_4^4 + C_4^{-4}) + iImB_4^4(C_4^4 - C_4^{-4}) + B_6^0 C_6^0 \\
& + ReB_6^2(C_6^2 + C_6^{-2}) + iImB_6^2(C_6^2 - C_6^{-2}) \\
& + ReB_6^4(C_6^4 + C_6^{-4}) + iImB_6^4(C_6^4 - C_6^{-4}) \\
& + ReB_6^6(C_6^6 + C_6^{-6}) + iImB_6^6(C_6^6 - C_6^{-6})
\end{aligned} \qquad \text{(A2)}$$

where ${C_q}^k$ are the normalized spherical-tensor operators and ${B_q}^k$ are the crystal field parameters (CFPs). The values of these CFPs can be determined by the least-squares fit to the observed energy levels.[9]

The ED (A_{ED}) radiative decay rates can be written as[10]-[11]

$$A_{ED(SLJ\to S'L'J')}=\frac{16\pi^3e^2}{3\varepsilon_0hc^3}\frac{\nu^3}{(2J+1)}\chi_{ED}\sum_{\lambda=2,4,6}\Omega_{(\lambda)}\left|\left\langle l^NSLJ\left\|U^{(\lambda)}\right\|l^NS'L'J'\right\rangle\right|^2 \tag{A3}$$

where ν is the transition frequency, n is the refractive index and χ_{ED} is the local-field correction for ED induced transitions with the form of $(n^2+1)^2/(9n)$ and $n(n^2+1)^2/9$ for absorption and emission transition, respectively. The Judd-Ofelt intensity parameters $\Omega_{(\lambda)}$ should be summed over $\lambda=2,4,6$ for a product with the even-rank reduced matrix elements of the $U^{(\lambda)}$ tensor operator.

The MD (A_{MD}) radiative decay rates can be written as[10]-[11]

$$A_{MD}=\frac{\pi he^2}{3\varepsilon_0c^5m_e^2}\frac{\nu^3}{g}\chi_{MD}\left|\left\langle l^N\psi\left\|L+g_eS\right\|l^N\psi'\right\rangle\right|^2 \tag{A4}$$

where $g_e = 2.00232$ is the gyromagnetic ratio of the electron and g is the degeneracy of the initial level. χ_{MD} is the local-field correction for MD induced transitions with the form of n and n^3 for the absorption and emission transition, respectively. ψ and ψ' are the statevectors for the initial and terminating levels for the $\psi \to \psi'$ transition, respectively. For transitions between J-multiplets, the statevector takes the form of $\psi(SLJ)$ with $g = (2J+1)$ while for transitions between crystal field levels, it takes the form of $\psi(SLJ\Gamma_i)$.

The radiative lifetime can be written as[10]-[11]

$$\tau_{SLJ}=\frac{1}{\sum_{S'L'J'}(A_{ED(SLJ\to S'L'J')}+A_{MD(SLJ\to S'L'J')})} \tag{A5}$$

The branching ratio can be written as[10]-[11]

$$\beta_{(SLJ \to S'L'J')} = \tau_{SLJ} \times [A_{ED(SLJ \to S'L'J')} + A_{MD(SLJ \to S'L'J')}] \tag{A6}$$

The MD absorption oscillator strengths can be written as[10]-[11]

$$P_{MD} = \frac{h\nu}{6m_e c^2} \frac{n}{(2J+1)} \left| \left\langle l^N SLJ \middle\| L + g_e S \middle\| l^N SLJ' \right\rangle \right|^2 \tag{A7}$$

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