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Judd–Ofelt analysis of spectra and experimental evaluation of laser performance of Tm^{3+} doped Lu_2SiO_5 crystal*

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This paper reports that the Tm^{3+}:Lu_2SiO_5 (Tm:LSO) crystal is grown by Czochralski technique. The room-temperature absorption spectra of Tm:LSO crystal are measured on a b-cut sample with 4 at.% thulium. According to the obtained Judd–Ofelt intensity parameters Ω_2=9.3155×10^{-20} cm^2, Ω_4=8.4103×10^{-20} cm^2, Ω_6=1.5908×10^{-20} cm^2, the fluorescence lifetime is calculated to be 2.03 ms for $^3F_4 \rightarrow {}^3H_6$ transition, and the integrated emission cross section is 5.81×10^{-18} cm^2. Room-temperature laser action near 2 μm under diode pumping is experimentally evaluated in Tm:LSO. An optical-optical conversion efficiency of 9.1% and a slope efficiency of 16.2% are obtained with continuous-wave maximum output power of 0.67 W. The emission wavelengths of Tm:LSO laser are centred around 2.06 μm with spectral bandwidth of ∼13.6 nm.

Keywords: Tm:Lu_2SiO_5, Judd–Ofelt analysis, solid-state lasers, diode-pumped
PACC: 2320C, 4255R, 6170, 4260D

1. Introduction

The recent interest in diode-pumped solid state laser systems which are based on the Tm^{3+}-ion transition near 2 μm, $^3F_4 \rightarrow {}^3H_6$, is motivated by the possibility for efficient diode pumping with the widely spread AlGaAs laser diodes. The tunable 2-μm emission of Tm ions lies in the eye-safety wavelength regions and it includes strong absorption lines of water and weak absorption lines of atmosphere. Therefore, crystals doped with Tm ions are expected to have potential applications in wide range of fields, such as atmospheric remote sensing which includes Doppler radar wind sensing and water vapour profiling by differential absorption radar, spectroscopy and basic research. There are several medical applications requiring laser sources near 2 μm including lithotripsy and orthopaedics. Furthermore, high-power quasi-continuous wave 2-μm lasers with high peak power are effective pump sources of optical parametrical oscillators and solid-state lasers further in the mid-infrared region.

In recent years, research of new crystals doped with Tm ions has received considerable attention for the goal of lower threshold, higher slope efficiency and higher optical-optical efficiency. So far, efficient laser operations have already been demonstrated in many Tm^{3+}-doped crystals, such as KYW,[1] KGdW,[2] KLuW,[3] YAG,[4] YAP,[5] YLF,[6] BaYF,[7] YVO_4,[8] $GdVO_4$,[9] and $LuVO_4$.[10] To our knowledge, there are no reports on the spectrum parameters and possible laser action in Tm^{3+}-doped Lu_2SiO_5 (LSO).

Tm:LSO could be a very attractive host as its thermal conductivity value is not affected by the introduction of the thulium cation in the unit cell in substitution of the lutetium cations, which is k=5.93 Wm^{-1}K^{-1}. So high thermal conductivity can cool the crystals effectively and decrease the influences of thermal effects on output power. In this work, the room-temperature absorption (unpolarized) spectra are measured. From the absorption (unpolarized) spectra, such spectra parameters as the intensity parameters, oscillation strength, spontaneous emission probabilities, fluorescence lifetime, absorption cross

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sections and emission cross sections of several Tm^{3+} transitions are calculated according to the Judd–Ofelt (J-O) theory. And the lasing performance is evaluated in experiment under laser diode pumping.

2. Spectroscopy of Tm:LSO

The Tm:LSO single crystal was grown by the Czochralski method in Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Science. The experiment was performed with a 3-mm-long, 3 mm×7 mm of aperture, 4%-doped b-cut (along growth direct direction) Tm:LSO crystal.

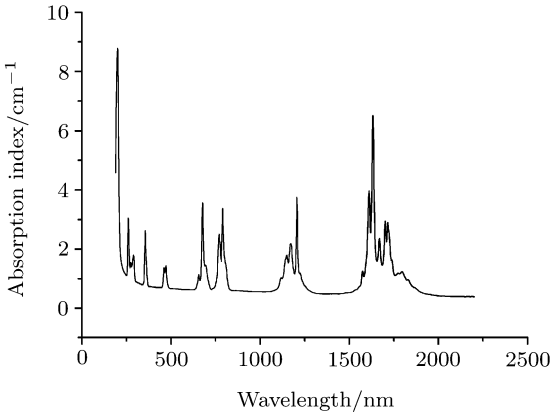


Fig.1. Absorption spectra of 4 at.% Tm^{3+} doped LSO (300 K) for unpolarized light.

The absorption spectra of Tm:LSO crystal were taken on a b-cut sample and shown in Fig.1. (JASCO V-570 type ultraviolet/visible/near-IR spectrophotometer, 1 nm resolution). As can be seen from Fig.1, some small splits in the vicinity of 472 nm, 790 nm and 1716 nm are due to the Stark energy splitting in the final state. The most intensive absorption peaks occur around at wavelengths of 678 nm, 790 nm and 1206 nm, which correspond to the transitions of Tm^{3+} ions $^3H_6 \rightarrow {}^3F_3$, $^3H_6 \rightarrow {}^3H_4$ and $^3H_6 \rightarrow {}^3H_5$. One of the strongest absorption features has a width of 13 nm, and is centred around 790 nm, which can be easily accessible with commercially available high power AlGaAs laser diodes.

3. Calculated spectra parameters and discussion

The radiative transition within the $4f^N$ configuration of a lanthanide ion can be analysed by using the J-O approach. Radiative transitions of rare earth ions in solids are predominately of electric dipole nature. For transitions between multiplets of the $4f^N$ configuration in randomly oriented systems, the experimental oscillator strengths, $f_{\rm exp}$, corresponding to absorption bands can be obtained b using the following formula:[11]

$$f_{\rm exp} = \frac{1}{N}\frac{m_{\rm e}c}{\pi e^2}\int k\,(\upsilon){\rm d}\upsilon, \tag{1}$$

where $m_{\rm e}$ and e are the electron mass and charge respectively, c is the velocity of light in vacuum , N is the unit volume concentrations of Tm^{3+}, $k\,(\upsilon)$ is the absorption index.

The absorption cross section corresponding to absorption bands can be obtained by using the following formula:

$$\sigma_{\rm abs} = 2.303 \times (L \times N)^{-1} \times {\rm OD}, \tag{2}$$

where L is the sample thickness, and OD is the optical density.

Based on the Eqs.(1), (2) and Fig.1, we calculate the oscillator strength, $f_{\rm exp}$, and the absorption cross section, $\sigma_{\rm abs}$. The results are shown in Table 1.

Table 1. Main absorption peaks corresponding to the transitions of Tm^{3+} ion in LSO crystal from $^3H_6$ ground state.

Excited state	Wavelength range/nm	Peak wavelength/nm	$\sigma_{\rm abs}/10^{-20}{\rm cm}^2$	$f_{\rm exp}/10^{-6}$
$^3P_2$	250–268	260	1.51	14.4178
$^3P_0$	280–304	290	0.88	9.7427
$^1D_2$	340–374	356	1.29	10.2149
$^3F_3$	664–690	678	1.76	3.3166
$^3H_4$	782–838	790	1.68	4.0543
$^3H_5$	1192–1218	1206	1.85	2.4210
$^3F_4$	1710–1722	1716	1.44	3.0090

According to the J-O approach, the electric-dipole line strengths, $S_{JJ'}$, from the initial state, $|(S,L)J\rangle$, to the final state, $|(S',L')J'\rangle$, can be expressed by the J-O intensity parameters,

$$S_{JJ'} = \sum_{t=2,4,6} \Omega_t \langle 4f^N(SL)J||U^{(t)}||4f^N(S'L')J'\rangle^2, \quad (3)$$

where Ω_t (t=2, 4, 6) is the J-O intensity parameters, f^N are wave functions of the states with N electrons in the $4f$ shell with total angular momentum J, J and J' are the total angular momentum of the initial and the final state, respectively, and $|<||U^{(t)}||>|^2$ are the reduced matrix elements of unit tensor operators for radiative transitions which are considered to be independent of host matrix, and their values were taken from Ref.[12].

According to the J-O approach the calculated oscillator strength of electric-dipole absorption transition from the initial state, $|(S,L)J\rangle$, to the finial state, $|(S',L')J'\rangle$, is given by

$$f_e = \frac{8\pi^2 m_e c}{3h\bar{\lambda}(2J+1)} \frac{(n^2+2)^2}{9n} S_{JJ'}, \quad (4)$$

where $\bar{\lambda}$ is the average wavelength of the absorption band, m_e is the mass of the electron, c is the velocity of light in vacuum, h is Planck constant, J is the total angular momentum of the initial state, n is the refractive index, and $S_{JJ'}$ is the electric-dipole line strengths.

By a least-square fitting, the J-O intensity parameters for Tm^{3+} in LSO crystals were calculated to be Ω_2=9.3155×10^{-20} cm^2, Ω_4=8.4103×10^{-20} cm^2, and Ω_6=1.5908×10^{-20} cm^2. Tm^{3+}:LSO crystal has larger J-O intensity parameters Ω_t (t=2, 4, 6) in comparison with other doped crystals (see Table 2). We know that Ω_2 is very sensitive to the structure and it is associated with the asymmetry and covalence of the lanthanide sites. The larger Ω_2 value indicates the stronger covalence characteristics of this crystal.[13] The values of the J-O intensity parameters are useful to calculate the spectroscopic quality factor Ω_4/Ω_6 which is critically important in predicting the stimulated emission for the laser active medium. The spectroscopic quality factor of Tm^{3+}:LSO crystal is 5.29 which is smaller than that of YAP, but larger than that of other crystals listed in Table 2. This relatively high Ω_4/Ω_6 value indicates that Tm:LSO is a promising material for efficient laser action.

Table 2. The J-O intensity and the spectroscopic quality factor for Tm^{3+} doped in various hosts.

Crystal	$\Omega_2/10^{-20}$ cm^2	$\Omega_4/10^{-20}$ cm^2	$\Omega_6/10^{-20}$ cm^2	Ω_4/Ω_6	Refs.
LSO	9.14	8.41	1.59	5.29	this
YLF	1.67	1.93	1.10	1.75	[14]
YAG	0.7	1.2	0.5	2.4	[15]
YAP	1.46	2.07	0.32	6.47	[16]
$GdVO_4$	8.16	2.45	0.98	2.5	[17]

The electric-dipole spontaneous emission probabilities and the fluorescence lifetime of an emitting state can be expressed by

$$A_{JJ'}[(S',L')J';(S,L)J] = \frac{64\pi^4 e^2}{4h\lambda^3} \frac{n(n^2+2)^2}{9(2J'+1)} S_{JJ'}, \quad (5)$$

$$\tau = \frac{1}{A_{JJ'}}, \quad (6)$$

where $A_{JJ'}$ is the spontaneous emission probability, and τ is the fluorescence lifetime.

Emission cross section from excited state to ground state is given by

$$\sigma_{em} = \frac{\lambda^2}{8\pi c n^2} A_{JJ'}. \quad (7)$$

Based on J-O intensity parameters, the spontaneous emission probability $A_{JJ'}$, fluorescence lifetime τ and emission cross section σ_{em} were calculated from Eqs.(5)–(7) and listed in Table 3.

Table 3. Calculated spontaneous emission probability, fluorescence lifetime and emission cross section in LSO crystal.

Transitions	λ/nm	A_{cal}/s^{-1}	τ/μs	$\sigma_{em}/10^{-18}$cm^2
$^3P_2$	260	47124	21	12.76
$^3P_0$	290	25596	39	8.63
$^1D_2$	356	17808	56	9.04
$^3F_3$	678	1594	627	2.94
$^3H_4$	790	1435	697	3.59
$^3H_5$	1206	368	2720	2.14
$^3F_4$	1716	225	2030	5.81

The measured fluorescence lifetime of $^3F_4$ excited state of Tm^{3+} is 1.82 ms, which is smaller than the calculated value (2.03 ms) according to the J-O theory. This may be understood from the fact that the

high phonon energies of LSO lead to the fluorescence lifetime of $^3F_4$ level which is probably to be quenched by nonradiative transitions.

Tm:LSO has larger emission cross sections corresponding to the $^3F_4 \rightarrow {}^3H_6$ transitions in comparison with other doped crystals (see Table 4). The longer fluorescence lifetime and larger integrated emission cross sections promise Tm:LSO lasers for the generation of 2-μm wavelength radiation.

Table 4. Calculated emission cross sections and spontaneous emission probability corresponding to the $^3F_4 \rightarrow {}^3H_6$ transitions for Tm^{3+}-doped various crystals.

Crystals	$\sigma_{em}/10^{-18}cm^2$	$A_{JJ'}/s^{-1}$	Refs.
YAG	1.49	578	[18]
YAP	2.22	972	[19]
LSO	5.81	225	this

4. Lasing actions of Tm:LSO

The geometry of fibre-coupled diode end-pumped Tm:LSO laser is shown in Fig.2. The crystal had dopant concentration of 4 at.% Tm^{3+}. The gain length of Tm:LSO crystal is 5 mm (3 mm×3 mm in aperture). Both the crystal's end faces were anti-reflection coated at 790 nm and 2 μm. The crystal was wrapped in indium foil and squeezed in a copper heat-sink for a good thermal contact, then connected with a thermal electric cooler. The crystal was end-pumped by a fibre-coupled laser diode with the emission wavelength at 790 nm (2.5 nm bandwidth). The diameter and numerical aperture of the fibre core are 200 μm and 0.22, respectively. The coupling lenses L_1 and L_2 with 25- and 45-mm focuses were used to re-focus the pump laser on the end face of the laser crystal. The resonator geometry used was plano-concave. The high reflector, M_1, acted as a resonator mirror and also transmitted the diode-pump radiation ($T > 95\%$ at 790 nm). The output coupler, M_2, with a 20-cm radius of curvature was coated for ~2.54% transmission at 2.06 μm.

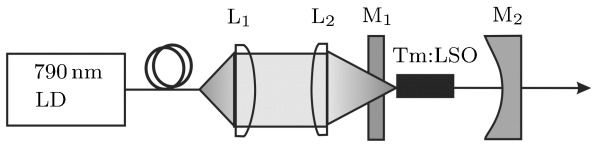


Fig.2. Schematic diagram of the diode pumped Tm:LSO laser configuration

Figure 3 shows the relationship between the input diode power and output power of single-end-pumped Tm:LSO crystal. As shown in Fig.3, the maximum output power of single-end-pumped Tm:LSO crystal is 673 mW at the crystal heat-sink temperature of 10 °C, corresponding to the optical-to-optical efficiency of 9.1%. The incident pump power threshold is about 2.68 kW/cm^2. A slope efficiency of 16.2% is obtained by linear fit of the experimental data.

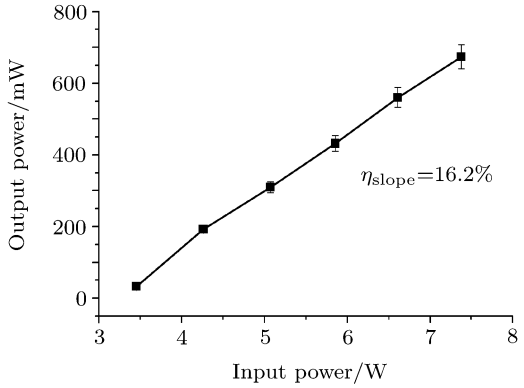


Fig.3. The output power of single-end-pumped Tm:LSO laser versus input laser diode (LD) power.

Laser wavelength measurements were performed with the use of a PbS detector, WDM1-3 monochrometer (300 mm focal length, 300 lines/mm grating blazed at 2000 nm) and digital oscilloscope. The emission wavelength of Tm:LSO laser centred at 2058.4 nm with emission band width of about 13.6 nm, which is shown in Fig.4.

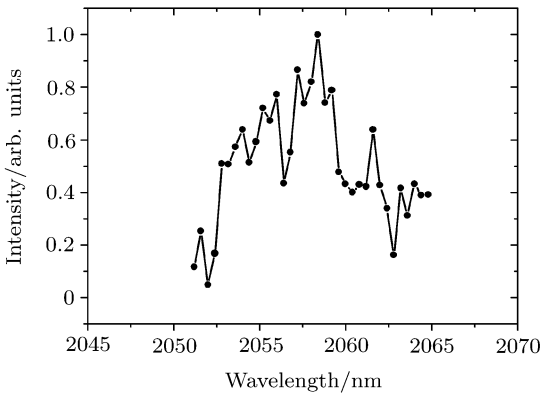


Fig.4. Spectrum of Tm:LSO laser output.

5. Conclusion

The room-temperature absorption spectra of Tm:LSO crystal were measured and investigated. The J-O intensity parameters Ω_t (t=2, 4, 6), spontaneous emission probability, fluorescence lifetime, integrated absorption cross sections and integrated emission cross

sections of several Tm^{3+} transitions were calculated from the absorption spectra based on the J-O theory. An output power of 0.67 W around 2.06 μm was attained from a diode end-pumped Tm:LSO laser with a slope efficiency of 16.2%.

References

[1] McGuckin B T, Menzies R T and Hemmati H 1991 *Appl. Phys. Lett.* **59** 2926

[2] Petrin R R, Jani M G, Powell R C and Kokta M 1992 *Opt. Mater.* **1** 111

[3] Petrov V, Liu J H, Galan M, Viera G, Pujol C, Griebner U, Aguilo M and Diza F 2006 *Proc. of SPIE* **6216** 62160K

[4] Kmetec J D, Kubo T S, Kane T J and Grund C J 1994 *Opt. Lett.* **19** 186

[5] Elder I F and Payne M J P 1998 *Optics Comm.* **145** 329

[6] Budni P A, Lemons M L, Mosto J R and Chicklis E P 2000 *IEEE J. Sel. Top. Quantum Electron.* **6** 629

[7] Galzerano G, Cornacchia F, Parisi D, Toncelli A, Tonelli M and Laporta P 2005 *Opt. Lett.* **30** 854

[8] Zayhowski J J, Harrison J, Dill C and Ochoa 1995 *J Appl. Opt.* **34** 435

[9] Mikhailov V A, Zavartsev Y D, Zagumenyi A I, Ostroumov V G, Studenikin P A, Heumann E, Huber G and Shcherbakov I 1997 *Quantum Electron.* **27** 13

[10] Tsuboi T, Ruan Y F and Kulagin N 2003 *Physics of Laser Crystals* **126** 171

[11] Kaminskii A A 1981 *Laser Crystals* (Berlin, Heidelberg, NewYork: Springer-Verlag) p19

[12] Carnail W T and Fields P R 1965 *J. Chem. Phys.* **42** 3797

[13] Chen G X, Zhang Q Y, Yang G F, Yang Z M and Jiang Z H 2007 *Acta Phys. Sin.* **56** 4200 (in Chinese)

[14] Walsh B M, Barnes N P and Bartolo B D 1998 *J. Appl. Phys.* **83** 2772

[15] Caird J A, Deshazer L G and Nella 1975 *IEEE J. Quantum Electron.* **11** 874

[16] Lu Y L, Yang Y, Wang J, Dong A P, Li S H, Sun B D, Zhou R and Dai S B 2006 *Chinese J. Lasers* **33** 968

[17] Yang Y M, Yao B Q, Chen B J, Wang C, Ren G Z and Wang X J 2007 *Opt. Mater.* **29** 1159

[18] Suni P J M and Henderson S W 1991 *Opt. Lett.* **16** 817

[19] Weber M J 1973 *Phys. Rev.* B **8** 54