Spectroscopic characterization of Yb:Sc_2O_3 transparent ceramics*

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Yb:Sc_2O_3 transparent ceramics are fabricated by a conventional ceramic process and sintering in H_2 atmosphere. The room-temperature spectroscopic properties are investigated, and the Raman spectrum shows an obvious vibration characteristic band centred at 415 cm^{-1}. There are three broad absorption bands around 891, 937, and 971 nm, respectively. The strongest emission peak is centred at 1.04 μm with a broad bandwidth (11 nm) and an emission cross-section of 1.8×10^{-20} cm^2. The gain coefficient implies a possible laser ability in a range from 990 nm to 1425 nm. The energy-level structure shows that Yb:Sc_2O_3 ceramics have large Stark splitting at the ground state level due to their strong crystal field. All the results show that Yb:Sc_2O_3 transparent ceramics are a promising material for short pulse lasers.

Keywords: Yb:Sc_2O_3, transparent ceramics, spectroscopic properties

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1. Introduction

There has been considerable interest in trivalent ytterbium as a diode pumped laser material in recent years. This is due to its broad absorption and emission bands, very low quantum defects, reduced thermal loads and lack of excited state absorption.[1] Driven by these advantages, Yb^{3+}-doped laser materials are shown to be ideal candidates to meet the needs of high-efficiency high-power continuous wave (CW) operation and ultrafast pulse laser output.[2–4]

Among all rare earth hosts, sesquioxide Sc_2O_3 is well known for its superior thermo-mechanical properties.[5] Sc_2O_3 processes the highest thermal conductivity (16.5 W/mK) in sesquioxide, which considerably exceeds that of YAG (11 W/mK).[6,7] Its low phonon energy ensures large energy storage time by minimizing non-radiative relaxation processes. After doping with Yb^{3+} ions, Yb:Sc_2O_3 exhibits large ground-state splitting and a broad emission bandwidth, which is advantageous for laser diode (LD) pumping and femtosecond pulse generation.[8] So far, a high efficient laser output has already been realized in Yb:Sc_2O_3 single crystal.[9]

Although Sc_2O_3 has been shown to be one of the most promising laser hosts for Yb^{3+} ions, it is extremely difficult to fabricate large high-quality Sc_2O_3 single crystals because of its high melting point (2420 °C).[10] Recently, there has been growing interest in using transparent ceramics as an alternative to Sc_2O_3 single crystals.[11] By using nanocrystalline technology and the vacuum sintering (NTVS) method, it is possible to obtain large, high optical quality Sc_2O_3 transparent ceramics, and the sintering temperature of the Sc_2O_3 ceramic has been decreased to 1700 °C. A diode-pumped Yb:Sc_2O_3 ceramic laser with a slope efficiency of 9% was reported.[12]

However, to the best of our knowledge, research on the spectroscopic properties of Yb:Sc_2O_3 transparent ceramics has not been carried out in detail yet. In the present work, Yb^{3+}-doped Sc_2O_3 transparent ceramics are fabricated by a conventional ceramic process and sintering in H_2 atmosphere, and the spectroscopic properties are investigated.

2. Experiment

The starting nanopowders (10 nm–50 nm) of Sc_2O_3 and Yb_2O_3 with a purity of 99.99% were weighed according to the composition of $(Yb_{0.05}Sc_{0.95})_2O_3$. The mixed powders were ball milled in absolute ethyl alcohol for 5 h and calcined at 1200 °C for 5 h in air. Specimens were isostati-

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cally pressed into the disks, each of 15 mm in diameter and 6 mm in thickness, at 200 MPa and sintered at 1600 °C–1700 °C for 45 h in H_2 atmosphere, then the specimens were double polished for spectral analysis.

The Raman spectrum was measured using a micro-Raman spectrometer system (inVia plus, Renishaw, UK) equipped with a 514.5 nm Ar^+ laser (2000, Spectra Physics, USA). The absorption spectrum was measured with a spectrophotometer (Model V-570, JASCO, Japan) in which Xe light was used as a double polished pump source. The emission spectrum and the fluorescence lifetime of the specimen excited with a 940 nm Xe lamp were measured with a fluorescence spectra analyzer (FLSP920, Edinburgh Instruments, UK), of which the resolution is 1 nm–2 nm. All spectral analyses were made at room temperature.

3. Results and discussion

Figure 1 shows a photograph and the transmittance of the Yb:Sc_2O_3 transparent ceramic

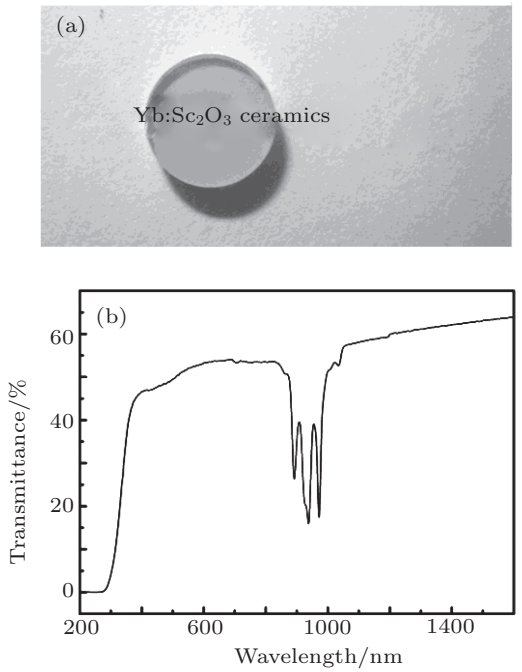


Fig. 1. Photograph (a) and transmittance (b) of a Yb:Sc_2O_3 transparent ceramic (2.0 mm in thickness).

(2.0 mm in thickness). It is obvious that the specimen has high transparency and the letters under the ceramic can be seen clearly. The highest transmittance of the Yb:Sc_2O_3 ceramic is about 64%, which is still slightly lower than that of the laser grade transparent ceramic, and laser oscillation was not realized in this specimen at the present stage. Future work should focus on improving the sintering process to increase the transmittance of the Yb:Sc_2O_3 ceramic further.

Figure 2 shows the Raman spectrum of the Yb:Sc_2O_3 ceramic. There are eight obvious characteristic Raman bands located at 190, 271, 314, 356, 388, 416, 492, and 519 cm^{-1}, respectively, and the most intense one centred at 415 cm^{-1} is the vibration characteristic band, which can correspond to the (F_g+A_g) mode of the ReO_6 (Re=Sc, Yb) octahedra.[13] Bands centred at 271, 314, 356, 388, 416, 492, and 519 cm^{-1} can be assigned to the internal vibrations of the ReO_6 octahedra. The 190 cm^{-1} band corresponds to the translational motions of the Re atoms in C_2 symmetry. The sites of all the Raman peaks show slight differences (about 2 cm^{-1}–5 cm^{-1} lower energies) from those of pure Sc_2O_3 ceramics reported in Ref. [14], which may be attributed to the doping of Yb^{3+} ions.

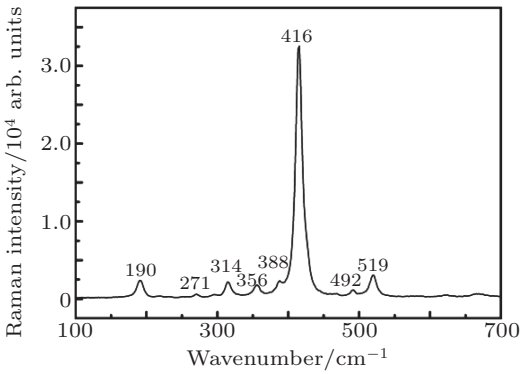


Fig. 2. Raman spectrum of the Yb:Sc_2O_3 ceramic.

The room-temperature absorption and emission spectra of the Yb:Sc_2O_3 transparent ceramics are shown in Fig. 3. There are three broad absorption bands around 891, 937, and 971 nm, respectively, corresponding to $^2F_{7/2}\rightarrow{}^2F_{5/2}$ transitions. The absorption cross-section (σ_{abs}) of the Yb^{3+} ion can be calculated from the following formula:

$$\sigma_{abs}(\lambda) = 2.303\lg(I_0/I)/LN, \quad (1)$$

where $\lg(I_0/I)$, N, L are the optical density, the number of active ions per unit volume and the thickness of the sample, respectively. The calculated results and the full width at half maximum (FWHM) values are listed in Table 1. As shown in Table 1, the Yb:Sc_2O_3 ceramic has a broad absorption band and large absorption cross section. The broadband absorption feature of the Yb:Sc_2O_3 ceramic at 937 nm and 971 nm makes it suitable to be pumped by an LD with no need for accurate temperature control.

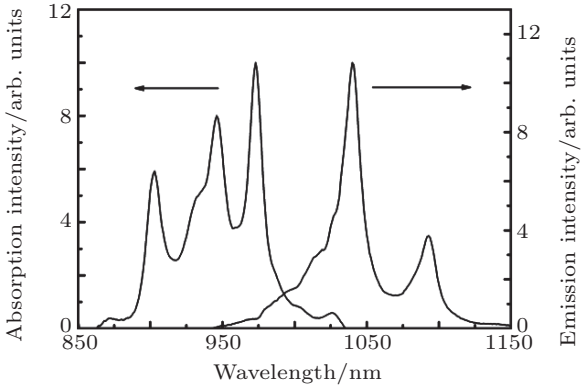


Fig. 3. Room-temperature absorption and emission spectra of the Yb:Sc_2O_3 transparent ceramic.

The emission cross section (σ_{em}) of the Yb^{3+} ion is estimated from the Füchtbauer–Ladenburg (FL) formula[15]

$$\sigma_{em}(\lambda) = \lambda_c^4(4\pi n^2 c\tau\Delta\lambda)^{-1}(\ln 2/\pi)^{1/2}, \qquad (2)$$

where λ_c is the central wavelength of the band, c is the velocity of light, $\Delta\lambda$ is the emission FWHM value, and τ is the fluorescence lifetime. According to the luminescence decay curve shown in Fig. 4, the fluorescence lifetime was calculated to be 0.56 ms. The estimated emission cross sections and the FWHMs at 1040 nm and 1093 nm are listed in Table 2. Data for the 2.5 at% Yb:Sc_2O_3 ceramic and Yb:YAG single crystal are also listed in the table for comparison.[12,16]

Table 1. The absorption cross section and FWHM of Yb:Sc_2O_3 transparent ceramics.

	Wavelength of absorption peaks/nm		
	891	937	971
Absorption cross section/10^{-20} cm^2	0.33	0.58	0.53
FWHM/nm	11	15	12

The Yb:Sc_2O_3 ceramics in this work show a shorter fluorescence lifetime than that reported in Ref. [12]. This may be attributed to the concentration quenching of Yb^{3+} ions, because the concentration of Yb^{3+} ions in this work is 5 at%, which is two times as large as that in Ref. [12]. According to formula (2), the emission cross section is inversely proportional to the fluorescence lifetime, therefore the emission cross section of the Yb:Sc_2O_3 ceramic in this work is larger than that in Ref. [12]. The emission FWHM of the Yb:Sc_2O_3 ceramic in this work is about two times larger than that of Yb:YAG. The broad emission bands of the Yb:Sc_2O_3 ceramic mean that shorter pulses were obtained probably by using mode-locking technology.

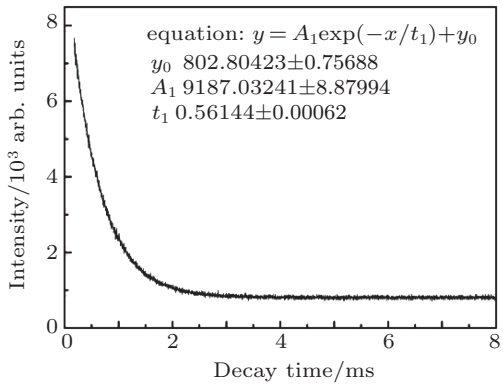


Fig. 4. Luminescence decay curve of the Yb:Sc_2O_3 ceramic at room temperature.

Table 2. The spectroscopic parameters for the Yb:Sc_2O_3 ceramics and Yb:YAG single crystal.

Parameters	5 at% Yb:Sc_2O_3 ceramics		2.5 at% Yb:Sc_2O_3 ceramics[12]		Yb:YAG[16] single crystal
Emission wavelength/nm	1040	1093	1041	1094	1030
Fluorescence lifetime/ms	0.56		0.80		0.97
Emission cross section/10^{-20} cm^2	1.8	0.85	1.3	0.25	2.1
FWHM/nm	11	16	11	17	6

Based on the absorption and emission cross sections, the gain cross section (σ_g) of the Yb:Sc_2O_3 ceramic was evaluated from the classical relation

$$\sigma_g(\lambda) = \beta\sigma_{em}(\lambda) - (1-\beta)\sigma_{abs}(\lambda), \qquad (3)$$

where β is the ratio of the number of Yb^{3+} ions in the upper level $^2F_{5/2}$ to the total number of Yb^{3+} ions. Figure 5 gives the dependences of the gain cross section with different β values. Since the 1.04 μm–1.09 μm transition of Yb^{3+} is close to that of a true three-level system, it is necessary to excite almost 50% of the Yb^{3+} ions in order to obtain the population in-

version. As shown in Fig. 5, for a 50% population inversion, the gain cross-section curve becomes positive in a wavelength range from 990 nm to 1425 nm, and the corresponding major gain cross-section peaks are centred at 1040 nm and 1093 nm. It is possible to realize laser oscillation at these two wavelengths.

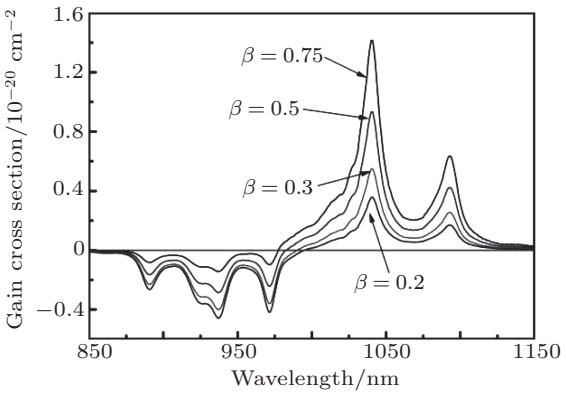


Fig. 5. Gain cross-section curves of Yb:Sc_2O_3 transparent ceramics for various population inversions.

Figure 6 shows an energy-level diagram of Yb^{3+} in the Sc_2O_3 ceramic calculated from its absorption and emission spectra. For both the $^2F_{7/2}$ and $^2F_{5/2}$ levels, the Stark splitting is much larger than in Yb:YAG, due to the strong crystal field in Yb:Sc_2O_3. The main transition lines lie on A_1–Z_3 and A_1–Z_4, and the ground state splitting is about 1150 cm^{-1}, which is larger than that of Yb:YAG (∼785 cm^{-1}).[16] This means that the Boltzmann thermal population at the ground state level in the Yb:Sc_2O_3 ceramic at room temperature is smaller than that in Yb:YAG, and population inversion can be realized in the Yb:Sc_2O_3 ceramic more easily.

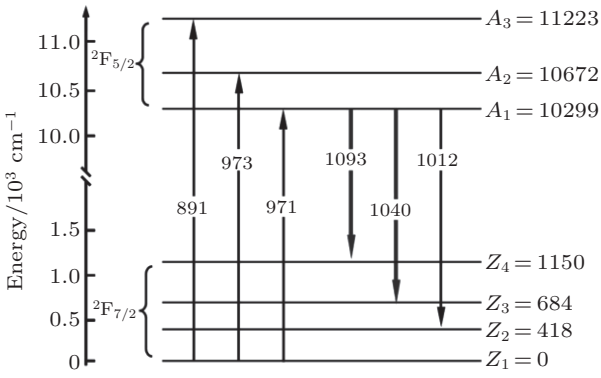


Fig. 6. Energy-level diagram of the Yb:Sc_2O_3 ceramic calculated from the absorption and emission spectra.

4. Conclusion

A 5 at% Yb^{3+}-doped Sc_2O_3 transparent ceramic with a high transparency is fabricated. The Raman spectrum of the Yb:Sc_2O_3 ceramic shows an obvious vibration characteristic band centred at 415 cm^{-1}, which corresponds to the cubic Sc_2O_3 structure. There are three broad absorption bands located at 891, 937, and 971 nm, respectively, corresponding to $^2F_{7/2}$→$^2F_{5/2}$ transitions. The two main emission peaks are centred at 1.04 μm and 1.09 μm, with broad bandwidths of 11 nm and 16 nm, respectively. The broad emission bandwidth of the Yb:Sc_2O_3 ceramic makes it a good candidate for short pulse generation using mode-locking technology. The optical gain cross sections for several populations in the conversion rates are estimated, and the gain coefficients show that possible laser oscillation can be realized at wavelengths of 1040 nm and 1093 nm. The energy-level structure of the Yb:Sc_2O_3 ceramic is also estimated. The ground state splitting of the Yb:Sc_2O_3 ceramic is about 1150 cm^{-1}, which is larger than that of Yb:YAG (∼ 785 cm^{-1}), and population inversion can be realized in the Yb:Sc_2O_3 ceramic more easily. All the results show that the Yb:Sc_2O_3 transparent ceramic is a promising material for short pulse lasers.

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