Spectral properties and laser performance of Ho: Sc$_2$SiO$_5$ crystal at room temperature

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Abstract: Holmium doped scandium silicate (Ho:SSO) bulk crystal grown by Czochralski technique is reported. The absorption cross section of $4.8 \times 10^{-21}$ cm$^2$ at pumping wavelength 1940 nm and emission cross section of $5.56 \times 10^{-21}$ cm$^2$ at lasing wavelength 2112 nm were calculated, respectively. Lifetime was measured to be 1.51 ms at 300 K and 0.92 ms at 77 K. Continuous-wave laser was operated by using a diode-pumped Tm:YAP laser as pump source with central wavelength of 1940 nm. Output power of 385 mW at 2112 nm was primarily obtained.

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1. Introduction

The 2μm lasers are in the eye-safety wavelength regions, where strong absorption lines of water and weak absorption lines of atmosphere are involved. 2μm lasers has potential applications in spectroscopy, basic research as well as atmospheric remote sensing which include Doppler radar wind sensing and water vapor profiling by differential absorption radar. Furthermore, high-power quasi-continuous wave (QCW) 2μm lasers with high peak power are efficient pump sources of optical parametrical oscillators and solid-state lasers in the mid-infrared region [1–4]. Tm and Ho co-doped media with large emission cross section can meet the requirement of generating high peak power laser pulse but the gain media must be cooled to low temperature to alleviate up-conversion effects and energy transferring between Tm and Ho [5–7]. The method of alleviating the above-mentioned effects is to adulterate Ho and Tm in separate crystals. With the development of high power 1.9 μm laser as an efficient pump source [8–12], the 2 μm lasers based on Ho-doped crystals such as Y3Al5O12 (YAG) and LiYF4 (YLF) have become prominent. The output wavelength of Ho:YAG laser was centered at 2.09μm, while that of Ho:YLF laser was at 2.05μm [13–15].

In this letter, Sc2SiO5 (SSO) was chosen as a promising singly doped laser material. Up to the authors’ knowledge, it is the first time to report Ho3+ doped scandium silicate bulk crystal (Ho:SSO). SSO crystal host has been reported with the advantageous strong-field coupling, large energy splitting and wide absorption linewidth and excellent thermal property while doped with Tm [10]. SSO crystal host with negative refractive index is unique in silicate series crystals, which can limit the thermal lens effect, crystallographic sites distortion and birefrigent effects. The laser operation at 2μm in Ho:SSO crystal is performed in this letter.

2. Calculation and analyze of Ho:SSO spectroscopy

The 0.5at% Ho:SSO crystal was grown by the Czochralski technology in Shanghai Institute of Ceramics, Chinese Academy of Sciences. The real dopant of Ho3+ ions in SSO crystal (N_{Ho}) was calculated to be 8.41 × 10^{19} \text{atom} \cdot \text{cm}^{-3}. Figure 1 shows the room-temperature absorption and emission spectra of Ho:SSO. The absorption spectrum from 1750 nm to 2200 nm which was recorded by the spectrometers (PERKINELMER lambda 950) was performed with sample thickness of 1.32 mm. Absorption peaks were recorded with centered peaks at 1847 nm, 1876 nm, 1889 nm, 1906 nm, 1926 nm, 1940 nm, 1961 nm, 1983 nm, 2017 nm, 2036 nm, 2062 nm nd 2087 nm. Absorption coefficient of 0.41 cm^{-1} was determined at 1940 nm. The absorption coefficient at 2112 nm was 0.01 cm^{-1}. The re-absorption effect should be considered while designing the laser cavity.
Fig. 1. Room-temperature absorption and emission spectrum of Ho:SSO.

Emission spectrum from 1800 nm to 2200 nm was performed with sample length of 13.8 mm at room temperature. The excitation wavelength was set at 1.94 \( \mu \)m. The spectrum was recorded with a monochromator (WDM1-3) combined to a Lock-in Amplifier (SR830). The broad emission waveband was centered at 1943 nm, 2028 nm and 2087 nm with full width at half width (FWHM) of 193 nm. During the lifetime measurement, Ho:SSO crystal was excited by 1940 nm and measured at 2100 nm. Lifetime of Ho\(^{3+}\) in Sc\(_2\)SiO\(_5\) crystal host was recorded at 77K and 300K as shown in Fig. 2.

Fig. 2. Lifetime of Ho\(^{3+}\) in Sc\(_2\)SiO\(_5\) crystal host measured at 77K and 300K.

Figure 3 indicates the absorption cross section and emission cross section of Ho:SSO. The absorption cross section of Ho\(^{3+}\) ions in SSO crystal host was calculated according to Eq. (1).

\[
\sigma_{\text{abs}}(\lambda) = \frac{\alpha(\lambda)}{N_{\text{Ho}}}
\]  

(1)

where \( \sigma_{\text{abs}} \) denoting the absorption cross section, \( \alpha(\lambda) \) and \( N_{\text{Ho}} \) representing absorption coefficient and unit volume concentration of Ho\(^{3+}\) ions, respectively. The absorption cross section at 1940 nm was calculated to be \( 4.8 \times 10^{-21} \text{cm}^2 \). Broad absorption band of 59 nm at energy transition \( ^5I_8 \rightarrow ^5I_7 \) makes it easy for free laser running around 2 \( \mu \)m pumped by LD Tm-doped laser.
The emission cross section $\sigma_{em}$ was calculated according to Eq. (2).

$$\sigma_{em}(\lambda) = \frac{1}{8\pi n^2 c} \frac{1}{\tau_{rad}} \frac{\lambda^2 I(\lambda)}{\lambda I(\lambda)d\lambda}$$  \hspace{1cm} (2)$$

The laser emission spectrum has a relatively large width and a rather flat peak, which distinguishes from the emission spectra of other Ho lasers, such as the Ho:YAG laser [9] and the Ho:YAP laser [15]. The emission cross section of the Ho:SSO at 2112nm is $5.56 \times 10^{-21}$cm$^2$.

The radiative transition within the $4f^N$ configuration of a lanthanide ion can be analyzed using the Judd–Ofelt approach. Radiative transitions of rare earth ions in solids are predominant of electric dipole nature. For transitions between multiplets of the $4f^N$ configuration in randomly oriented systems, the experimental oscillator strengths $f_{exp}$ is corresponded to absorption bands that can be obtained using Eq. (3).

$$f_{exp} = \frac{m_e c^2}{\pi e^2 \lambda^2 N_q} \int \alpha(\lambda) d\lambda$$  \hspace{1cm} (3)$$

where $m_e$ and $e$ are the electron mass and charge respectively, $c$ is the light velocity in vacuum, $N$ is the unit volume concentrations of Ho$^{3+}$, and $\lambda$ is the central wavelength for different energy transitions. The line strength was calculated based on Eq. (4).

$$S_q^{\text{abs}}(J \rightarrow J') = \frac{3hc(2J+1)}{8\pi e^2 \lambda_q} \frac{9n_q}{(n_q^2 + 2)^2} \int_{J \rightarrow J'} \sigma_q^{\text{abs}}(\lambda)d\lambda$$  \hspace{1cm} (4)$$

where $q$ stands for polarized state of the absorption band, $J$ and $J'$ for the total quantum angular momentum of the initial and the final state of the transition state, $e$ for electronic charge, $c$ for light speed and $h$ for planck constant, $\lambda$ for the average wavelength between the transition $J \rightarrow J'$ and $n_q$ for the refractive index [10]. $\int \sigma_q^{\text{abs}}(\lambda)d\lambda$ stands for the integration of absorption cross section. In the following calculations, we used the unpolarized data to obtain basic J-O parameters.

According to the Judd-Ofelt approach, the calculated oscillator strength of electric-dipole absorption transition from the initial state to the final state is given by Eq. (5).

$$f_{\text{cal}}^{ed} = \frac{8\pi^2 m_e c}{3h\lambda (2J+1)} \times \frac{1}{n} \left[ \frac{(n^2 + 2)^2}{9} \right] S_{J'J}$$  \hspace{1cm} (5)$$
where \( m_e \) is the mass of the electron and \( S_{JJ'} \) is the electric-dipole line strengths. 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 Judd-Ofelt intensity parameters according to Eq. (6).

\[
S_{JJ'} = \sum_{t=2,4,6} \Omega_t \left( \frac{4 f^N [S,L]}{J(J+1)} \frac{4 f^N [S',L',J']}{J'(J'+1)} \right)^2
\]

where \( \Omega_t \) \((t=2,4,6)\) is the Judd-Ofelt intensity parameters, \( f^N \) are wavefunctions of the states with \( N \) electrons in the 4f shell with total angular momentum \( J \) and \( ||<U^{(t)}|| \) are the reduced matrix elements of unit tensor operators for radiative transitions which are considered to be independent of host matrix and referred from Ref [16]. According to Eqs. (3)–(6), the Judd-Ofelt intensity parameters \( \Omega_t \) \((t = 2, 4, 6)\) can be obtained and listed in Table 1.

**Table 1. Reduced matrix elements of Ho\(^{3+}\) in different crystal hosts.**

| Crystals | \( \Omega_2 \) / \((10^{-20}\text{cm}^2)\) | \( \Omega_4 \) / \((10^{-20}\text{cm}^2)\) | \( \Omega_6 \) / \((10^{-20}\text{cm}^2)\) | \( \Omega_4 / \Omega_6 \) | References |
|----------|---------------------------------|---------------------------------|---------------------------------|----------------|-----------|
| SSO      | 4.44                            | 3.48                            | 6.82                            | 0.51           | This work |
| YAG      | 0.70                            | 1.20                            | 0.50                            | 2.40           | [9]       |
| YLF      | 1.67                            | 1.93                            | 1.10                            | 1.75           | [14]      |
| YAP      | 1.42                            | 2.92                            | 1.71                            | 1.71           | [15]      |
| GdVO\(_4\) | 8.16                           | 2.45                            | 0.98                            | 2.50           | [17]      |

As we know that \( \Omega_2 \) is very sensitive to the structure and it is associated with the asymmetry and covalence of the lanthanide sites. The larger \( \Omega_2 \) value indicates the stronger covalence characteristics of this crystal. The values of the J-O intensity parameters are useful to calculate the spectroscopic quality factor \( \Omega_4 / \Omega_6 \) which could predict the stimulated emission for the laser active medium. The spectroscopic quality factor of Ho\(^{3+}\):SSO crystal is calculated to be 0.51 based on the unpolarized spectra.

**Based on** \( S(J,J') \) **calculated from Eq. (6) and the transition probability** \( A(J;J') \) **from upper levels to lower levels that obtained from Eq. (7). The radiative lifetime of \(^5\)\( I_7 \) is calculated to be 0.99 ms according to Eq. (8) which is close to lifetime measured at 77K (0.92 ms) but lower to that at room temperature (1.51 ms).**

\[
A(J;J') = \frac{64\pi^4 e^2}{3h(2J+1)^3} \frac{n(n^2+2)^2}{9} S(J;J')
\]

\[
\tau_{rad} = \frac{1}{A(J;J')}
\]

**3. Lasing action of Ho:SSO**

![Fig. 4. Experimental setup of room-temperature Ho:SSO laser end-pumped by a Tm:YAP laser.](image-url)
The experimental setup is shown in Fig. 4. To evaluate the lasing performance of Ho:SSO crystal in room temperature, a diode-pumped Tm:YAP laser with emission wavelength of 1.94 μm was utilized for pumping, which emits 15 W of output power with beam quality $M^2$ of ~1.3. The pump beam diameter was approximately 320 μm at the input surface of the Ho:SSO crystal. The un-coated Ho:SSO crystal, doped with 0.5 at.%, had a dimension of 5mm × 5mm (in section) × 13.8mm (in length). We measure the actual absorption efficiency of the Ho:SSO crystal. It is found that 56% of the pump power is absorbed. This Ho:SSO crystal was wrapped in indium foil and held in a copper heat-sink bonded on a thermal electric cooler (TEC) for precise temperature control. In this experiment, the temperature of Ho:SSO crystal was held at 20°C. The Ho:SSO laser resonator is plano-concave with a 200 mm radius of curvature concave output coupler, whose transmissions is 2.5%. The 45° dichroic mirror provides both high reflection (HR) at 2.1 μm (R>99.5%) and high transmission (HT) at 1.94 μm (T>94%). The physical length of the resonator was about 70 mm.

The used power meter was Coherent PM2 in this experiment. The maximum continuous wave output power obtained was 385mW with incident pump power of 8.6W. The lasing threshold was 5.0W. Relative to the incident pump power, the slope efficiency was 7.1%. This slope efficiency is much lower than other Ho-doped lasers. One possible cause is intracavity loss attribute to poor quality of crystal. Another possibility is weak pump absorption by Ho:SSO crystal. By increasing the doped length of crystal and improving the quality of crystal, much better conversion efficiency would be expected. The output spectrum of Ho:SSO laser was recorded by a spectrum analyzer (Bristol 721). The output laser wavelength was centered at 2112 nm with FWHM of about 0.8 nm which was shown in Fig. 5.

4. Conclusion

In summary, the spectral characteristics of Ho:SSO crystal are investigated. The experimental evaluation of laser performance is also demonstrated. As we know, this is the first time to investigate the characteristics of Ho:SSO laser. An output power of 385mW Ho:SSO laser lasing at 2112 nm has been obtained.

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