

Journal of Alloys and Compounds 217 (1995) 148-150

Journal of ALLOYS AND COMPOUNDS

Spectral characteristics of Er³⁺-activated and Ce³⁺-sensitized yttrium aluminium garnet laser crystals

Yaqin Yu, Siyuan Zhang, Shaolong Tie, Mingshu Song

Changchun Institute of Applied Chemistry, Chinese Academy of Science, Changchun 130022, China

Received 29 September 1993; in final form 6 June 1994

Abstract

We report in this paper the spectral characteristics of Er^{3+} (2 at.%)-activated and Ce^{3+} (0.3 at.%)-sensitized yttrium aluminium garnet (YAG:Er,Ce) laser crystals grown by the Czochralski technique. The absorption and emission spectra were measured at room temperature. By using absorption spectra and Judd-Ofelt theory the experimental oscillator strengths of the Er^{3+} transitions in the YAG:Er,Ce crystals were calculated. The energy transfer between the Er^{3+} and Ce^{3+} ions is also discussed.

Keywords: Spectral characteristics; Sensitized yttrium aluminium garnet; Energy transfer

1. Introduction

In recent years the laser properties of YAG:Er crystals have received considerable interest [1-4]. The Er^{3+} lases at wavelengths near 1.6 μ m or 3 μ m. The 1.6 μ m laser performance at room temperature in the YAG:Er crystals has the advantage of being eye safe for potential applications in medicine and biology. The Er laser may also be very useful for testing optical transmission properties of fibres.

In this paper we report on the spectral characteristics of Er^{3+} -activated and Ce^{3+} -sensitized yttrium aluminium garnet crystals (YAG:Er,Ce). By using Judd–Ofelt theory the experimental oscillator strengths were calculated. The energy transfer between Er^{3+} and Ce^{3+} ions was analysed and discussed.

2. Experimental details

YAG:Er,Ce and YAG:Er laser crystals were grown by the Czochralski technique. The YAG:Er,Ce crystal has been doped with Er^{3+} (2 at.%) and Ce^{3+} (0.3 at.%). The absorption spectra were recorded with a Japan UV-365 spectrophotometer and the emission spectra by a U-100 l m double grating monochromator which was made by ourselves.

3. Results and discussion

3.1. Absorption spectrum

Fig. 1 shows the absorption spectra of 0.728 nm thick YAG:Er,Ce and YAG:Er crystals for wavelengths from 200 nm to 1600 nm. The absorption spectrum consists of many lines. For the Ce^{3+} ion the 5d absorption is in the form of two broad bands, peaking at 340 nm and 455 nm, corresponding to the transitions from



Wavelength (nm)

Fig. 1. The absorption spectra of YAG:Er,Ce (spectrum a) and YAG:Er (spectrum b) at room temperature.

 ${}^{2}F_{5/2,7/2}$ to the 5d state. The 455 nm band is the strongest. These transitions allow more efficient absorption of the pump light.

By using Judd–Ofelt theory the values P_{exp} for the oscillator strengths were deduced from the absorption spectra of the YAG:Er,Ce and the YAG:Er crystals. According to Ref. [5] the oscillator strengths are given by

$$P_{\rm exp} = 4.318 \times 10^{-9} \int \epsilon_{\rm i}(\sigma) \, \mathrm{d}\sigma \tag{1}$$

where σ is the wavenumber and ϵ_i is the molar extinction coefficient of Er^{3+} absorption bands. The oscillator strengths of the YAG:Er,Ce and the YAG:Er crystals are shown in Table 1. The oscillator strengths P_{exp} of the ${}^{4}I_{15/2}$ - ${}^{4}F_{5/2,7/2}$ and ${}^{2}D_{11/2}$, ${}^{2}F_{11/2}$ transitions in the YAG:Er,Ce crystal have much larger values, 61.1×10^{6} and 15.5×10^{6} respectively. This is because the absorption bands of the Er^{3+} ion overlap those of the Ce^{3+} ions.

To facilitate comparison with laser crystals Table 1 lists the experimental oscillator strengths. These values may be compared with that of the YAG:Er. It is noteworthy that the oscillator strengths of the YAG:Er,Ce crystal are much larger than those of the YAG:EI.

3.2. Fluorescent spectra

The fluorescent spectra of the YAG:Er,Ce crystal are shown in Fig. 2 in the range from 1500 nm to 1700

Table 1 Oscillator strengths of Er^{3+} in YAG:Er and YAG:Er,Ce laser crystals

| Transition | Spectral interval (cm ⁻¹) | Oscillator strength ($\times 10^6$) | | |
|---|--|---------------------------------------|--------|------------------|
| | | YAG:Er,Ce P _{exp} | YAG:Er | |
| | | | Pexp | P _{cal} |
| (² D, ² P) _{3/2} ⁴ L _{11/2} ⁴ D _{7/2} ⁴ D _{5/2} | 38023 40000–34482 | 7.9 | 7.29 | 7.09 |
| (² D, ⁴ F) _{3/2} | 29411 31250–27174 | 15.6 | 2.0 | 2.6 |
| ${}^{2}G_{7/2}$, ${}^{4}G_{11/2}$ | 26441 27174–25773 | 5.6 | 3.97 | 4.02 |
| (² G, ² F, ² H) _{9/2} | 24716 25126–24380 | 0.96 | 0.47 | 0.50 |
| ⁴ F _{5/2,7/2} | 21978 24380–19960 | 61.1 | 1.6 | 1.9 |
| ² H _{11/2} , ⁴ S _{3/2} | 19230 19960–17857 | 3.7 | 2.4 | 2.3 |
| ⁴ F _{9/2} | 14530 16128–14492 | 2.2 | 1.7 | 2.15 |
| ⁴ J _{9/2} | 12159 12821–12195 | 1.0 | 0.50 | 0.44 |



Fig. 2. The emission spectra of YAG:Er,Ce (spectrum a) and YAG:Er

nm. The emission intensities of the Er^{3+} in the YAG:Er,Ce are stronger than those in the YAG:Er because the UV absorption broad bands in the YAG:Er,Ce crystal are markedly enhanced owing to Ce^{3+} -doped YAG:Er.

3.3. Energy transfer from Ce^{3+} to Er^{3+} ions

The electronic configuration of Er^{3+} is $4f^{11}$ and the narrow band absorptions and emissions in the Er³⁺, which are essential for the laser activity, arise from the parity-forbidden weak transitions. This explains the poor absorption of pumped light. The parity-allowed transitions in the Ce³⁺ correspond to the absorptions of broad bands. This may be excellent for laser pumping by energy transfer processes. Energy transfer could increase the population of excited levels of Er^{3+} by about one (or two) orders of magnitude compared with direct absorption of light into the Er³⁺ ions. The lowest 5d level of the Ce^{3+} ion is in the proximity of the 4f levels of the Er³⁺ in the YAG:Er,Ce crystals and very fast non-radiative 5d to 4f relaxations would occur. The efficiency of the transfer could be found by comparing the relative intensities of the 5d bands in the absorption spectra of YAG:Er:Ce, so that a laser could be achievable.

4. Conclusion

(spectrum b).

The absorption and emission spectra of the YAG:Er,Ce crystals were measured and analysed at room temperature. By using Judd–Ofelt theory the experimental oscillator strengths were calculated according to the absorption spectra of the crystals. In

the presence of the Ce^{3+} ion two additional intense peaks appear to broad bands in the YAG:Er,Ce absorption spectra at 340 nm and 455 nm. Corresponding to the energy transfer from the Ce^{3+} to the Er^{3+} ions the mechanism would probably be of the dipole–dipole type. It can be seen that the 5d to 4f relaxation is indeed quite effective for the energy transfer. By codoping the crystals with other ions such as Ce^{3+} to absorb a larger fraction of the pump power and to achieve higher pumping densities it is possible for the YAG:Er,Ce crystals to lase under flash lamp pumping.

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