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LETTER TO THE EDITOR

Optical properties of $CaGd_4Si_3O_{13}$ (CGS) crystals with Er^{3+} used as 1.5 μm laser material

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Abstract. The Stark structure of the $^4I_{13/2}$ and $^4I_{15/2}$ multiplets of the Er^{3+} ion in $CaGd_4Si_3O_{13}$ (CGS) was determined. The transition cross-sections have been obtained from the absorption and the luminescence spectra. The emission cross-section for π-polarization in the 1529.5 nm luminescence band is equal to 2.1×10^{-20} cm^2. A gain curve has been plotted for π-polarization, and positive amplification occurs at 1550 nm. A wide luminescence band makes it possible to obtain tunable generation in the range 1540–1590 nm. The spectral parameters of the crystals Er, Ce:CGS indicate ample scope for developing an effective 1.5 μm laser with semiconductor pumping.

1. Introduction

Laser emission with a wavelength of 1.5 μm is eye-safe, and appears promising as regards a wide range of applications in medicine, communications, distance measurements, and scientific research [1, 2]. To obtain such emission, solids doped with erbium ions are used. Many laser-generation schemes for different crystalline matrices have been tested in efforts to obtain efficient laser generation for Er^{3+} ions. However, in practice, glass lasers remain the main source of the 1.5 μm emission. A relatively slow excitation relaxation from the $^4I_{11/2}$ state to the upper laser level $^4I_{13/2}$ of Er^{3+} in crystals is responsible for this.

It is known that in the silicate crystals Er:CGS, with ion relaxators Ce^{3+}, which may selectively quench an erbium luminescence [3], the lifetime of the pre-laser level $^4I_{11/2}$ of Er^{3+} decreases from 14 to 1 μs and approaches that of an erbium phosphate glass [3, 4]. Therefore, crystals of CGS doped with ytterbium and erbium are good candidates for producing 1.5 μm lasers with laser diode pumping. Some properties of erbium ions in silicate crystals, such as Y_2SiO_5 and $SrY_4(SiO_4)_3O$, have been studied by Souriau *et al* [5]. As far as we know, crystals of CGS containing erbium ions have not been studied yet. At the same time, crystals of CGS have high isomorphic capacity and are of great interest for their simultaneous doping with Yb, Er, and Ce ions. For this reason, the present letter is devoted to research into spectral luminescence properties of Er:CGS.

Single crystals of $CaGd_{4-x-y-z}Yb_xEr_yCe_zSi_3O_{13}$ have been grown from the melt by the Czochralski method [3]. There are data available showing that crystals of $CaLn_4Si_3O_{13}$ exist and have the same structure for all lanthanides [6]. Crystals with such a large isomorphic

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capacity may be relatively easily co-doped with trivalent ions from different ends of the La series. Optically pure crystal samples of $8 \times 5 \times (0.3–3)$ mm^3 without cracks were used for measurements. The concentrations of erbium were defined by the formula $N_{\mathrm{Er}} = 0.6Ny$, where $N = 3.9 \times 10^{21}$ cm^{-3} [7].

The absorption and luminescence spectra have been studied using MDR-23 monochromators (reverse linear dispersion 2.6 nm mm^{-1}); the widths of the slits were 0.1–0.15 mm. Axis $\boldsymbol{c}$ was located in the plane of the crystal plate studied. We used standard polarizers to measure π-polarized ($\boldsymbol{E} \parallel \boldsymbol{c}, \boldsymbol{k} \perp \boldsymbol{c}$) and σ-polarized ($\boldsymbol{E} \perp \boldsymbol{c}, \boldsymbol{k} \perp \boldsymbol{c}$) absorption and luminescence spectra. The luminescence spectra were corrected by taking into account the spectral sensitivity of the measuring equipment. The luminescence of Er^{3+} has been excited through Yb^{3+} by 967 nm radiation of an InGaAs laser diode. The lifetimes of the excited state $^4I_{13/2}$ of the Er^{3+} ion have been determined from the kinetics of luminescence at 1530 nm wavelength after excitation by the second harmonic of the YAG:Nd laser with a 532 nm emission wavelength.

2. Experimental results and discussion

Figure 1 presents absorption spectra measured in unpolarized light and luminescence spectra of the crystals $Er_{0.05}$:CGS and $CaGd_{3.29}Yb_{0.277}Er_{0.014}Ce_{0.419}Si_3O_{13}$ in the range 1440–1650 nm.

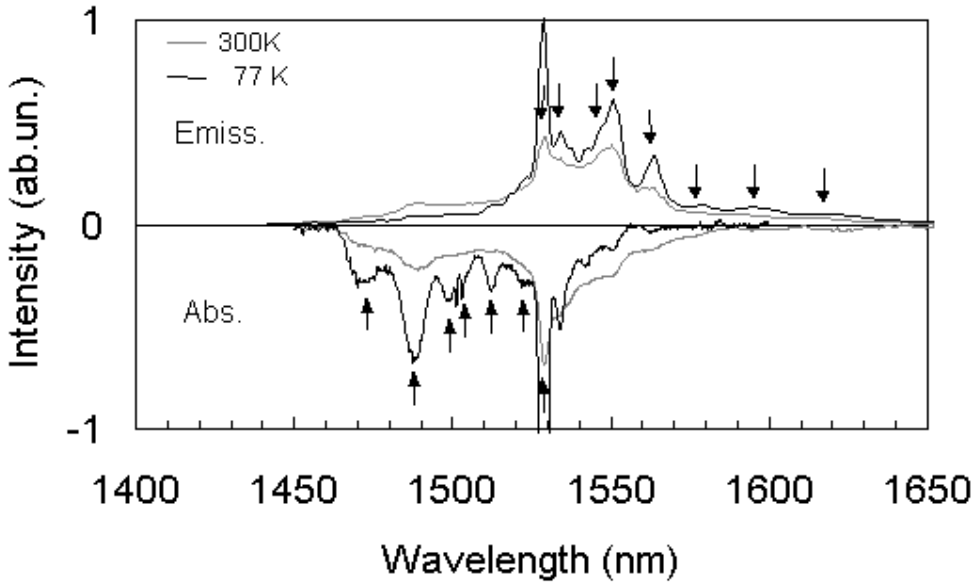

Figure 1. Unpolarized absorption and luminescence spectra for Er:CGS, measured at 300 and 77 K.

As follows from figure 1, as the temperature goes down the intensities of the erbium absorption bands at 1472, 1487.5, 1499, 1512.5, 1522, and 1529.5 nm and the erbium luminescence bands at 1529.5, 1534, 1546, 1551, 1563, 1578, 1597, and 1620 nm rise. The rise of the transition intensity is connected with the growth of the Stark-sublevel population. As the temperature goes down, the populations of the lowest levels of the fundamental and excited manifolds grow. That is why the positions of the spectral bands, presented above, give the positions of the $^4I_{13/2}$ and $^4I_{15/2}$ Stark sublevels of the erbium manifolds in CGS (table 1).

Table 1. Positions of the Stark-level components $^4I_{13/2}$ and $^4I_{15/2}$ of the Er^{3+} ion in CGS, and factors of their population.

Manifold	Energy (cm^{-1})	$b_{i\,(j)} = \exp(\Delta E_{i\,(j)}/kT)$	$\sum b_{i\,(j)}$
j, $^4I_{13/2}$	6793.48	2.95×10^{-1}	4.39×10^{0}
	6722.69	4.14×10^{-1}	
	6671.11	5.30×10^{-1}	
	6648.94	5.89×10^{-1}	
	6611.57	7.04×10^{-1}	
	6570.30	8.57×10^{-1}	
	6538.08	1.00×10^{0}	
i, $^4I_{15/2}$	365.24	1.75×10^{-1}	4.61×10^{0}
	276.34	2.67×10^{-1}	
	200.95	3.83×10^{-1}	
	140.13	5.12×10^{-1}	
	90.63	6.48×10^{-1}	
	69.78	7.16×10^{-1}	
	19.18	9.12×10^{-1}	
	0.00	1.00×10^{0}	

The transition cross-sections have been obtained from the absorption spectra and from the luminescence spectra (figures 2 and 3).

In figure 2, the emission cross-section spectrum σ_e (the top part of the figure) has been obtained from the absorption cross-section spectrum $\sigma_a = k/N_{\mathrm{Er}}$ (the bottom half of the

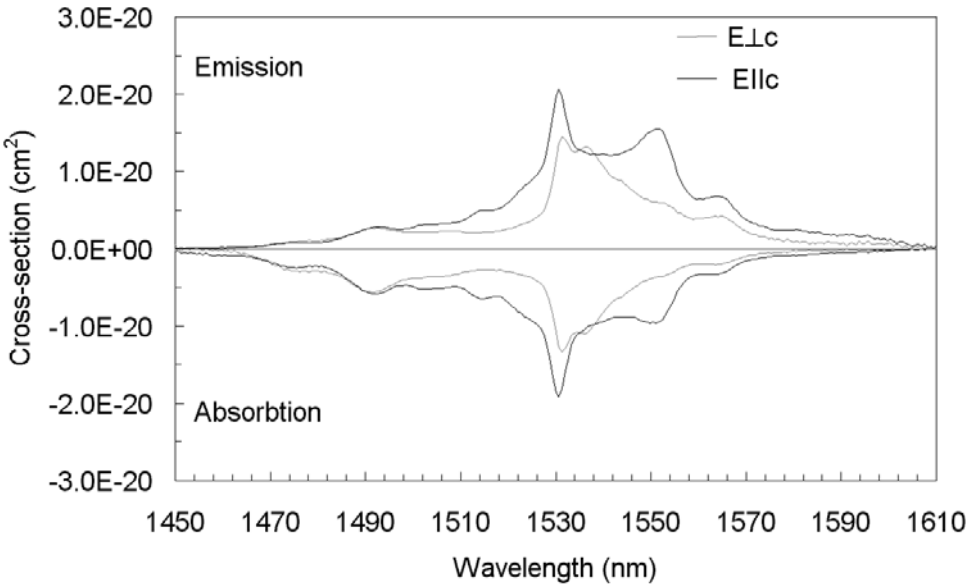

Figure 2. Absorption and emission cross-section spectra, obtained by the reciprocity method, for two polarizations in Er:CGS.

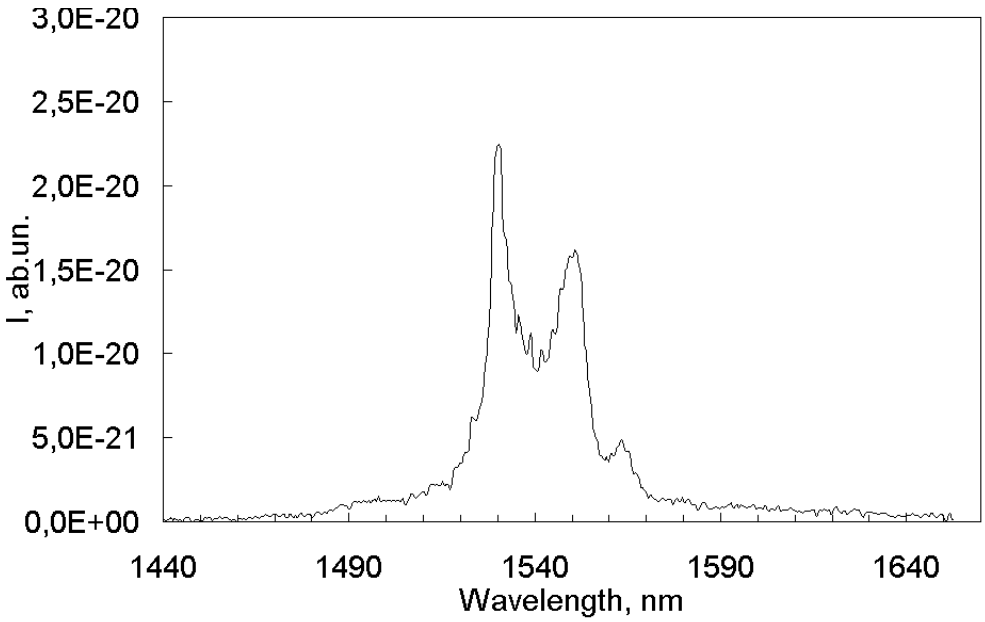

Figure 3. The emission cross-section for $\boldsymbol{E} \parallel \boldsymbol{c}$ polarization in Er:CGS, obtained from the luminescence spectra by the Fuchtbauer–Ladenburg method.

figure), using the equation

$$\sigma_e = \sigma_a \left[\sum b_i \Big/ \left(\sum b_j \right) \right] \exp((h\nu - E_0)/kT) \tag{1}$$

where

$$b_{i\,(j)} = \exp(-\Delta E_{i\,(j)}/kT)$$

are Boltzmann filling factors for Stark's sublevels in the multiplet,

$$\sum b_i \Big/ \left(\sum b_j \right) = 0.9522$$

for Er at room temperatures, and $E_0 = hc/\lambda_0$ where λ_0 is the resonance transition wavelength equal to 1529.5 nm for Er^{3+} in CGS (table 1).

The spectra of the effective emission cross-section in figure 3 have been plotted using luminescence spectra by the Fuchtbauer–Ladenburg method [8] in accordance with the expression

$$\sigma_e(\lambda) = \lambda^4 I(\lambda) \Big/ \left(8\pi n^2 c \tau_\lambda \int I(\lambda)\, \mathrm{d}\lambda \right) \tag{2}$$

where $\tau_\lambda = 3100\ \mu$s and $n = 1.82$. Comparison of figures 2 and 3 shows that the two methods of calculation of the emission cross-section give close results. As we can see from the figures, there is a rather high emission cross-section of erbium in CGS, namely 2.1×10^{-20} cm^2.

The spectral dependence of the gain cross-section for polarization $\boldsymbol{E} \parallel \boldsymbol{c}$ is shown in figure 4. It should be noted that the maximum on the gain curves with the relative population $n/N = 0.5$ is located at 1550 nm, and in the case of a small inversion level it is located at 1590 nm. The luminescence range of the tunable generation is predicted to be 1540–1590 nm.

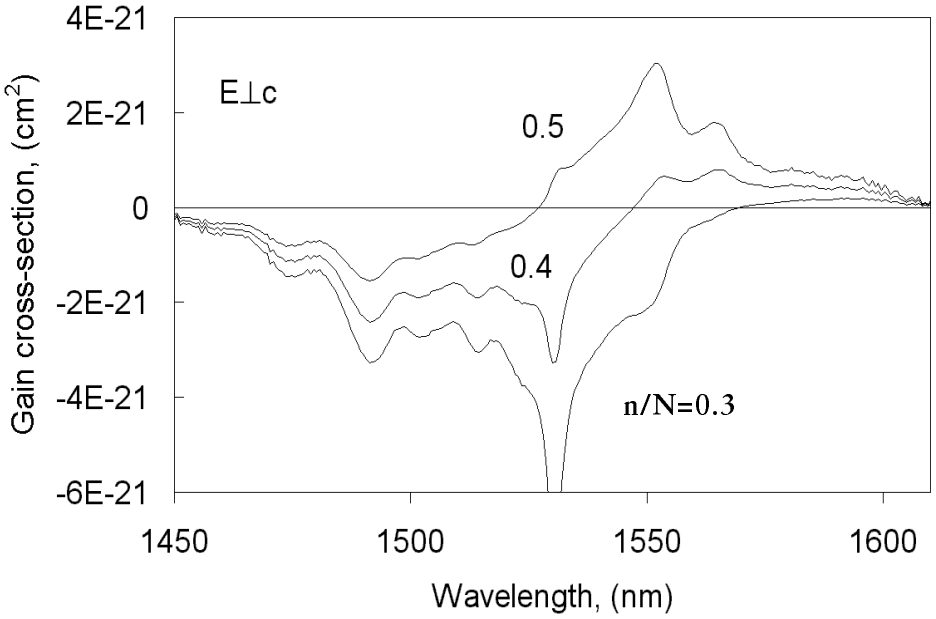

Figure 4. Spectra of the gain cross-section in Er:CGS (*n* and *N* are Er concentrations in the excited and fundamental states.

3. Conclusions

Previously it has been determined that Yb, Er, Ce:CGS crystals offer a unique possibility, by means of the introduction of Ce^{3+} ions, to reduce the lifetime of the $^4I_{11/2}$ level of the Er^{3+} ion to give the same characteristics as for erbium phosphate glasses, and thereby create conditions for the effective energy transfer Yb $\rightarrow$ $^4I_{13/2}$ Er. Spectral investigations demonstrate that crystals of Yb, Er, Ce:CGS are characterized in addition by a very high value of the Er^{3+} emission cross-section in the 1.5 μm range; this value is substantially superior to the erbium emission cross-section in the phosphate laser glasses. The wavelength of generation in crystals is supposed to be 1550 nm. A wide band of luminescence indicates possibilities for gaining tunable generation in the range 1540–1590 nm. Thus, the spectral properties are a good complement to the kinetic properties of Yb, Er, Ce:CGS crystals and show them to provide ample scope for the engineering of effective 1.5 μm lasers with diode pumping.

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