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Crystal growth and optical properties of Er:CAS ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) and Er:SLG ($\text{SrLaGa}_3\text{O}_7$)

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Abstract

The optical properties of Er^{3+} doped $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (CAS) and $\text{SrLaGa}_3\text{O}_7$ (SLG) are described in this paper. Er^{3+} solubility is determined for the two laser hosts. Cross-relaxation processes and multiphonon desexcitation are observed and the relative rates are calculated with the frame of the Judd-Ofelt analysis. Er:SLG presents some longer lifetimes and smaller multiphonon non radiative contribution than the Er:CAS. Both laser hosts present a broad and intensive emission around $1.5\ \mu\text{m}$. In this paper are also discussed the mechanisms that can reduce the laser oscillation efficiency.

Introduction

Er doped lasers operating near $1.5\ \mu\text{m}$ are useful for numerous applications (LIDAR, telecommunication with optical fiber...). The development of high power laser diodes emitting around $0.98\ \mu\text{m}$ allowed the pumping of the Er^{3+} ions through the sensitizer Yb^{3+} which strongly absorbs in that spectral range. Melilite type $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (CAS) and $\text{SrLaGa}_3\text{O}_7$ (SLG) are good matrices for RE^{3+} ions [1,2]. The structural disorder around RE^{3+} involves broad absorption and fluorescence spectra. This paper presents mainly the spectroscopic study of Er:CAS and some comparisons between absorption, lifetimes and emission cross section of Er:CAS and Er:SLG.

Results

Er^{3+} ions can be easily introduced in the melilite structure and crystals with good optical quality are obtained. Crystals with various doping rate, up to $1.3 \cdot 10^{21}\ \text{Er}^{3+}\ \text{ions cm}^{-3}$, are at first grown by the floating zone method. Their crystalline quality is sufficient to allow one to characterize the main optical properties. Some crystals grown by the Czochralski process have also been obtained. The crystal structure of gehlenite $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (CAS) is tetragonal. Half of the Al^{3+} ions occupy regular tetrahedral sites (T_1) while Si^{4+} ions and the remaining Al^{3+} ions occupy other very distorted tetrahedra (T_2). Er^{3+} cation substitutes Ca^{2+} in large distorted cubes (Thomson cubes), the charge being balanced according to the equation

$\text{Ca}^{2+} + \text{Si}^{4+} \rightarrow \text{Er}^{3+} + \text{Al}^{3+}$, leading to the formula $\text{Ca}_{2-x}\text{Er}_x\text{Al}_{2+x}\text{Si}_{1-x}\text{O}_7$. The nearest cation sites in the vicinity of the Er^{3+} doping ions are the T2 in which Al^{3+} and Si^{4+} are statistically distributed. $\text{SrLaGa}_3\text{O}_7$ is an isostructural compound where Thomson cubes are statistically occupied by Sr^{2+} and La^{3+} . Ga^{3+} is situated into the two kinds of tetrahedra. Er^{3+} cation substitutes La^{3+} , leading to the formula

$\text{SrLa}_{1-x}\text{Er}_x\text{Ga}_3\text{O}_7$. A structural disorder is observed for these matrices since several kinds of cations share statistically a same type of sites, and this disorder is responsible for the inhomogeneous broadening of the absorption bands.

The absorption spectra of the Er^{3+} in the UV and visible range are presented in figure 1 for the two hosts.

For Er:CAS, absorption spectra are obtained under polarized light leading to the determination of the energy level diagram and Judd-Ofelt phenomenological parameters Ω_t . The values calculated for that matrix are $\Omega_2 = 3.5 \times 10^{-20} \text{ cm}^2$, $\Omega_4 = 1.9 \times 10^{-20} \text{ cm}^2$ and $\Omega_6 = 0.8 \times 10^{-20} \text{ cm}^2$. In Er:CAS, the experimental lifetimes for all the excited levels, excepted the $^4\text{I}_{13/2}$ level, are considerably smaller than the radiative lifetime calculated with the frame of the Judd-Ofelt analysis. Therefore, strong non-radiative relaxation processes occur in the matrix. The value of the mean lifetime of the $^4\text{S}_{3/2}$ level for the low erbium concentration ($\approx 8 \mu\text{s}$) compared to the calculated radiative lifetime of that level ($\approx 550 \mu\text{s}$) indicates that multiphonon deexcitations occur with a probability higher than 98%. For the $^4\text{I}_{13/2}$ level the experimental lifetime is in the millisecond range, ($\approx 7.6 \text{ ms}$). For that level, the multiphonon probability is small as the energy difference between the $^4\text{I}_{13/2}$ and $^4\text{I}_{15/2}$ levels is sufficiently large: 6041 cm^{-1} and the experimental and calculated radiative lifetimes are about the same. Short lifetimes values were already observed in the gehlenite compounds doped by Ho^{3+} and Tm^{3+} ions [3]. The mean lifetime values of the $^4\text{S}_{3/2}$ level decreases from approximately $8 \mu\text{s}$ for $0.2 \times 10^{20} \text{ Er}^{3+} \text{ ions cm}^{-3}$ ($x=0.01$) to $3 \mu\text{s}$ for $1.3 \times 10^{21} \text{ Er}^{3+} \text{ ions cm}^{-3}$ ($x=0.2$ see figure 2). That behavior indicates that cross-relaxation processes also occur for concentration higher than $6.5 \times 10^{19} \text{ Er}^{3+} \text{ ions cm}^{-3}$ ($x=0.03$). The corresponding

Er^{3+} - Er^{3+} interaction parameters corresponding to $[^4\text{S}_{3/2}, ^4\text{I}_{15/2}] \rightarrow [^4\text{I}_{13/2}, ^4\text{I}_{9/2}]$ and $[^4\text{S}_{3/2}, ^4\text{I}_{15/2}] \rightarrow [^4\text{I}_{9/2}, ^4\text{I}_{13/2}]$ cross-relaxation schemes are $C_{DA} = 2.6 \times 10^{-39} \text{ cm}^6\text{s}^{-1}$. Energy migration of the excitation has also been observed for the Er:CAS.

Er:SLG laser host presents longer lifetime values. For instance, the $^4\text{S}_{3/2}$ lifetime for the low Er^{3+} concentration is approximately $135 \mu\text{s}$. That experimental value reveals a strong decrease of the multiphonon relaxation probability when Si^{4+} ions are removed from the gehlenite matrix.

The $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ emission around $1.5 \mu\text{m}$ is reported in figure 3. For the two laser hosts, the emission is broad between 1.470 toward $1.640 \mu\text{m}$ (FWHM $\approx 40 \text{ nm}$)

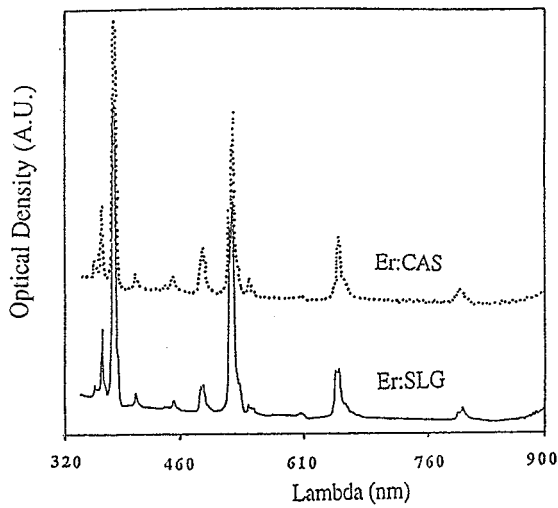


Figure 1:
Absorption spectra

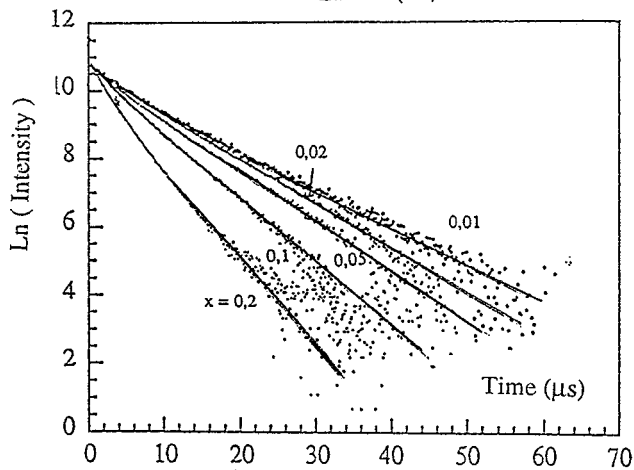


Figure 2: Room temperature fluorescence decay profiles of the $^4S_{3/2}$ level in Er:CAS $\lambda_{ex}=355$ nm and $\lambda_{em}=455$ nm

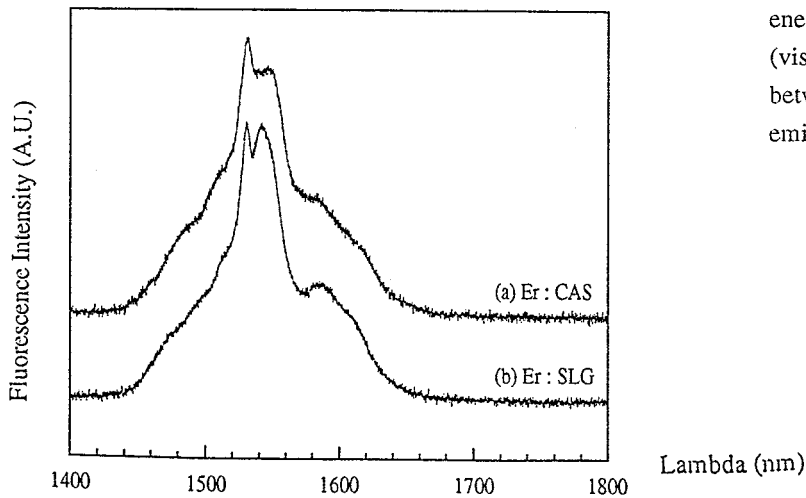


Figure 3: $^4I_{13/2} \rightarrow ^4I_{15/2}$ room temperature NIR emission

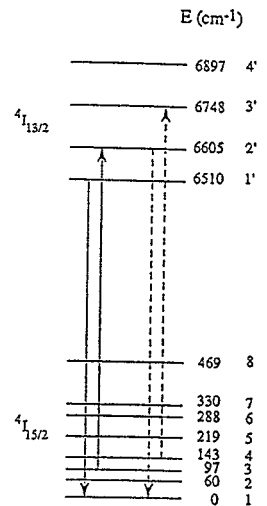


Figure 4: $^4I_{15/2}$ and $^4I_{13/2}$ energy levels of the Er:CAS (visualisation of resonances between the absorption and emission from various sublevels)

The stimulated emission cross section σ^e is related to the radiative transition rate $1/\tau_R$ and the effective linewidth $\Delta\nu_{\text{eff}}$ by :

$$\sigma^e = \frac{\lambda^2}{8\pi c \tau_R n^2 \Delta\nu_{\text{eff}}} \quad \Delta\nu_{\text{eff}} = \frac{1}{I_{\text{peak}}} \int_{\text{band}} I(\nu) d\nu$$

in which the branching ratio is one. For an inhomogeneously broadened emission line shape as the ones in Er:CAS and Er:SLG, the effective fluorescent linewidth $\Delta\nu_{\text{eff}}$ has to be determined by numerical integration of the fluorescence spectrum. The stimulated emission cross section at room temperature is estimated to be $0.4 \times 10^{-20} \text{ cm}^2$ at $1.535 \mu\text{m}$ for the Er:CAS and $0.28 \times 10^{-20} \text{ cm}^2$ at $1.540 \mu\text{m}$ for the Er:SLG. The values are smaller to the value obtained for the Er:YAG around $0.5 \times 10^{-20} \text{ cm}^2$ [4] but in the gehlenite matrices broader tunability range of the emission is expected.

In this study we have also investigated the mechanisms which can reduce the laser oscillation efficiency in the Er^{3+} :CAS. Only a small up-conversion from the $^4\text{I}_{11/2}$ level leading to a green emission from the $^4\text{S}_{3/2}$ level has been observed under CW pumping at 962 nm from a Ti:sapphire laser. This process is limited in the CAS because of the short lifetime values of the $^4\text{I}_{11/2}$ level (around 50 μs). The main process which can reduce the laser oscillation will be the reabsorption which can occur in this matrix since the emitting level of the laser transition $^4\text{I}_{13/2}$ is the first excited state and good resonances are observed (see figure 4) between the absorption and emission from various sublevels of the $^4\text{I}_{15/2}$ and $^4\text{I}_{13/2}$ levels. This effect will increase the room temperature laser threshold in the Er:CAS and reduce the tunability.

Conclusions

The two laser hosts present a broad emission around $1.55 \mu\text{m}$ with relatively high values of the emission cross-sections. In the Er:SLG the phonon relaxation processes responsible for the heating of the crystals are considerably smaller than in the Er:CAS. But, in that case, the upconversion process may be higher. The next step of this work is the study of the Yb^{3+} ions and of the Yb^{3+} - Er^{3+} interactions in these laser hosts with the knowledge of the spectroscopic properties of the two laser materials.

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