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Temperature effect on emission spectra and fluorescence lifetime of the $^{4}I_{13/2}$ state of Er$^{3+}$-doped Gd$_2$SiO$_5$ crystal

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By measuring the emission spectra and the fluorescence lifetime of the $^{4}I_{13/2}$ state of Er$^{3+}$ ions in Gd$_2$SiO$_5$ crystal at different temperatures, the effects of temperature on the spectra and the lifetime of the $^{4}I_{13/2}$ state are investigated. When the temperature increases, the emission line width for the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ transition is broadened, and the main emission lines at 1 596, 1 609, and 1 644 nm shift toward shorter wavelengths. The measured lifetime of the $^{4}I_{13/2}$ state decreases from 13.2 to 8.4 ns with temperature increase from 13 to 300 K, which is mainly due to the temperature dependence of multiphonon relaxation between the $^{4}I_{13/2}$ and $^{4}I_{15/2}$ states and the changing population distribution among the Stark levels within the $^{4}I_{13/2}$ state. The experimental results imply that low temperature condition is better for the $\sim 1.6$-$\mu$m laser output.

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Recently, with the development of high-power InP laser diode and erbium fiber laser (EFL) in the 1.46–1.6-$\mu$m range$^{[1,2]}$, Er$^{3+}$-doped Gd$_2$SiO$_5$ crystal is considered to be an interesting material to obtain $\sim 1.6$-$\mu$m eyesafe laser based on the resonantly pumping sketch$^{[3]}$. Compared with the well-known Er$^{3+}$:YAG crystal, Er$^{3+}$: Gd$_2$SiO$_5$ is anisotropic with monoclinic structure, belonging to the space group P$_{2}$1/c$^{[4]}$, which means that it can be directly used to obtain polarized laser.

In the past several years, studies have been dedicated to the growth and spectroscopic characterization of Er$^{3+}$:Gd$_2$SiO$_5$, and some primary data are already available$^{[5–7]}$. However, such data are not sufficient to predict laser performance, especially under different temperature conditions. Generally, increasing the temperature will result in thermal broadening and shift in the laser line, which have a significant meaning for the laser action since these effects are closely related to the light amplification gain, output frequency stability, and thermal tenability of the laser. The variation of fluorescence lifetime with the temperature is also significantly important for many Q-switched and other operations of Er$^{3+}$: Gd$_2$SiO$_5$ lasers. In this letter, the emission spectra of Er$^{3+}$:Gd$_2$SiO$_5$ at 13–300 K are measured, and the temperature dependence of the fluorescence lifetime for the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ transition of Er$^{3+}$ ions in this host has been studied.

Single crystals of Er$^{3+}$:Gd$_2$SiO$_5$ were grown from iridium crucible by Czochralski technique. The highly pure oxide powders Gd$_2$O$_3$ (99.999%), SiO$_2$ (99.999%), and Er$_2$O$_3$ (99.999%) were used as the starting materials, while the concentration of Er$^{3+}$ ions in the melt was 0.3 at.%. The seed was of $<010>$ orientation and the whole growth atmosphere was high-purity nitrogen gas. The comparison of 29 diffraction peak values and their corresponding relative intensity with the standard $\chi$-ray diffraction (XRD) card of Gd$_2$SiO$_5$ indicates that the formation of the monoclinic structure of Gd$_2$SiO$_5$ was achieved in the Er$^{3+}$:Gd$_2$SiO$_5$ crystal that we have grown. The optical quality of the crystals was found to be good, as shown in Fig. 1. During the crystal growth of Er$^{3+}$:Gd$_2$SiO$_5$, the substitution of Gd$^{3+}$ by Er$^{3+}$ was accomplished easily because the dimensional, chemical, and physical properties of the two ions are quite similar. The sample used for spectroscopic measurements was optically polished, and the thickness of the sample was 1 mm.

The absorption spectrum of Er$^{3+}$:Gd$_2$SiO$_5$ was measured at 300 K by using a spectrophotometer (Lambda 900, Perkin-Elmer, USA). The emission spectra were measured with a spectrophotometer (Spect Fluorolog-3, Horiba, Japan) and were excited at 378 nm by a 450-W xenon lamp. The emission decay curves were obtained by pulse excitation with the 980-nm line of a diode laser. The emitting line of 1.528 $\mu$m for the $^{4}I_{13/2}$ manifold of Er$^{3+}$:Gd$_2$SiO$_5$ was detected through a 0.5-m monochromator with a photomultiplier. For the low temperature measurements, the Er$^{3+}$:Gd$_2$SiO$_5$ sample was cooled by a liquid-nitrogen cryogenic refrigerator, capable of controlling temperature from 13 to 300 K. For all the measurements, the spectral resolution was kept below 1 nm.

Fig. 1. Photo of as-grown Er$^{3+}$:Gd$_2$SiO$_5$ single crystal with size of $\Phi 35 \times 40$ (mm).
The emission spectrum at 13 K for the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ transition is shown in Fig. 2 (black curve). As most of Er$^{3+}$ ions are frozen at the lowest Stark level of the $^{4}I_{13/2}$ state, transitions from this level to all the Stark levels of the $^{4}I_{15/2}$ state give off eight main emission lines (i.e., 5,914, 6,083, 6,215, 6,266, 6,386, 6,456, 6,510, and 6,540 cm$^{-1}$), but the remaining weak emission lines (such as 6,361 cm$^{-1}$) originate from thermally populated excited levels within the $^{4}I_{13/2}$ state. The zero line is 6,540 cm$^{-1}$, and the Stark splitting of the $^{4}I_{15/2}$ state are calculated to be 0, 30, 80, 146, 270, 320, 458, and 628 cm$^{-1}$, which are in agreement with the data reported in Ref. [7]. Figure 3 shows the temperature evolution of the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ emission spectra in the temperature range of 13–300 K. The greatest influence of temperature is on the spectra shape and emission intensities. At low temperatures, the individual lines are well separated from each other; with increasing temperature, each individual line becomes broad and gradually overlaps with the neighboring lines arising from the higher Stark levels. When the temperature was increased from 13 to 300 K, the effects of thermal populations on the Stark levels within the $^{4}I_{13/2}$ state not only extended the short wavelength edge of the emission spectra from 1.520 to 1.440 $\mu$m, but also smoothened down the emission intensities of the main lines. For the relatively intense emission lines at 1.596, 1,609, and 1,644 $\mu$m, when the temperature increased from 13 to 300 K, their emission intensities decreased by 4, 4.5, and 6 times, respectively. Since their terminating levels are separated by 457, 325, and 274 cm$^{-1}$ from the lowest Stark level of the $^{4}I_{15/2}$ state (the inset of Fig. 2), thermodynamic calculation using Boltzmann distribution demonstrates that decreasing temperature will reduce the thermal population at these levels, resulting in weak reabsorption at these lines. On the contrary, the lines shorter than 1.560 $\mu$m encountered intense reabsorption, as presented in Fig. 4, by comparing the absorption and emission spectra at different temperatures. In this point of view, Er$^{3+}$:Gd$_2$SiO$_5$ may have potential to realize laser oscillation at the 1.596-, 1.609-, and 1.644-$\mu$m lines, especially on the conditions of low temperatures.

On the other hand, the line positions are also temperature dependent due to the stationary effect of the ion–phonon interaction. Electron–phonon interactions perturb various electronic states, changing their wave functions and energies. The greater the distance from neighboring atoms is, the lower will be the potential of the crystal field of an impurity ion and the smaller the stark splitting of its multiplets as the temperature of the crystal increases[8]. For Er$^{3+}$:Gd$_2$SiO$_5$ lattice, thermal expansion can reduce the crystal field of Er$^{3+}$ ions, resulting in weakened Stark splitting as temperature increases. This effect not only acts on the $^{4}I_{13/2}$ state but also on the $^{4}I_{15/2}$ state; thus, their Stark levels will shift toward lower or higher energies as the temperature increases. From Fig. 3, the emission lines at 1.529, 1.549, 1.566, 1.609, 1.644, and 1.690 $\mu$m shift to shorter wavelengths and the lines at 1.536 and 1.596 $\mu$m shift to longer wavelengths. Aside the dot line of Fig. 2 labels the line shift value ($\Delta \nu$) for each emission line; the largest red-shift occurred at 1.536 $\mu$m with 14 cm$^{-1}$ and the line at 1.609 $\mu$m encountered the largest blue-shift with 14 cm$^{-1}$ as the temperature increased from 13 to 300 K.

The emission decays of the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ transition measured at different temperatures are depicted in Fig. 5. All the decay curves have nearly single-exponential
Phonon process, the highest energy phonons are usually
fluorescence lifetime of the is temperature dependent.

The temperature dependence of the fluorescence lifetime of the
in Er$^{3+}$-doped Gd$_2$SiO$_5$ crystal (it is recorded using a spectrometer (INVIA, Renishaw, England)
equipped with a monochromator, a filter system, and charge-coupled
device detector; unpolarized 785-nm laser from a laser
diode is chosen as the excitation light).
8.4 ms at 300 K. This is mainly ascribed to the temperature dependence of multiphonon relaxation between the $^4I_{13/2}$ and $^4I_{15/2}$ states and the changing population distribution among the Stark levels within the $^4I_{13/2}$ state. All the experimental data imply that low temperature is beneficial to realize laser oscillation at the potential laser lines of 1.596, 1.609, and 1.644 µm.

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References