SPECTROSCOPIC PROPERTIES OF ER$^{3+}$/YB$^{3+}$ CO-DOPED TANTALUM-NIOBIUM PHOSPHATE GLASSES FOR OPTICAL WAVEGUIDE LASER AND AMPLIFIER

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Abstract- Er$^{3+}$/Yb$^{3+}$ co-doped tantalum-niobium phosphate glasses suitable for planar waveguide device applications at 1.53 $\mu$m operation were prepared and investigated. The host compositions of which were P$_2$O$_5$, Ta$_2$O$_5$, Nb$_2$O$_5$, K$_2$O, and Ga$_2$O$_3$. The measured absorption and emission spectra of these glasses were analyzed by Judd-Ofelt and McCumber theories, in order to obtain the effect of Er$^{3+}$/Yb$^{3+}$ ion concentrations on the spectroscopic properties. It turns out that the emission cross sections around 1.53 $\mu$m increase and then decrease with the increase of rare earth ion concentrations, the maximum value occur at the concentrations of 6wt%. The effective emission cross section bandwidth around 1.53 $\mu$m was about 60.0nm, which was wider than silica glasses and other phosphate glasses.

Keywords - tantalum-niobium phosphate glass, Er$^{3+}$/Yb$^{3+}$ co-doped, absorption and emission cross section, Judd-Ofelt parameters, bandwidth

1 INTRODUCTION

Er$^{3+}$/Yb$^{3+}$ co-doped glasses are interesting gain materials for microchip lasers and waveguide amplifiers due to their capability for emission of radiation at the "eye safe" wavelength of 1.53 $\mu$m. They offer the possibility of very significant applications in the areas of optical communications, laser radar and range finding. The energy levels of Er$^{3+}$ ions for optical amplification at 1.53 $\mu$m form a three-level system which requires a high pump rate to achieve population reversion, but the low absorption across section of Er$^{3+}$ ions limits the pump efficiency. Yb$^{3+}$ ions exhibit not only a large absorption cross section, but also a broad absorption band between 800 and 1100nm. Furthermore, the large spectral overlap between Yb$^{3+}$ emission and Er$^{3+}$ absorption results in an efficient resonant energy transfer from Yb$^{3+}$ to Er$^{3+}$ in Er$^{3+}$/Yb$^{3+}$ co-doped system[1].

The development of integrated optical components, allowing integration with other optoelectronic devices on the same chip and efficient waveguide-to-fiber coupling, has demonstrated that it is necessary to provide a high erbium concentration in order to achieve a large gain over a short distance. In this work we report a new tantalum-niobium phosphate glasses, which can be doped with higher concentration of rare earths and are more stable chemically and thermally than silica-phosphate and alumino-phosphate glasses. The measured absorption and emission spectra of these glasses were analyzed by Judd-Ofelt and McCumber theories and the rare earth concentration dependence of fluorescence properties was also discussed. Systematic studies on the spectral properties of Er$^{3+}$/Yb$^{3+}$ co-doped tantalum-niobium phosphate glasses can have theoretical and practical significance in developing the laser glasses with high Er$^{3+}$/Yb$^{3+}$ concentration.

2 EXPERIMENTAL

2.1 Sample synthesis

A series of glasses with 27P$_2$O$_5$-26Ta$_2$O$_5$-16Nb$_2$O$_5$-12K$_2$O-17Ga$_2$O$_3$-xEr$_2$O$_3$-yYb$_2$O$_3$ in wt%, where x and y are the content of Er$_2$O$_3$ and Yb$_2$O$_3$ (x:y=1:4), have been synthesized. The increase of the percentage of tantalum and niobium oxide in the phosphate matrix glass leads to an increase in glass transition temperature.
Batches with different weight ratio of rare earth were fully mixed and moved into aluminum crucible and melted in electronic furnace at the temperature of 1300°C for 1 h. The melt was cast into a preheated copper mold, and then the resulting glass sample was annealed at about 590°C for 20 h. The transformation temperature is higher than that of traditional phosphate glasses. Samples for optical and spectroscopic measurements were cut and polished into \(10 \times 10 \times 5\) mm\(^3\) size.

2.2 spectroscopic property measurement

Optical absorption spectra in the visible and near infrared regions were measured at room temperature by a double beam spectrometer. Room temperature emission spectra at 1.53 µm were performed with a computer-controlled monochromator and InGaAs detector by pumping with a diode laser of 976 nm. The density of samples was measured by Archimedean method.

3 JUDD-OFELT ANALYSIS

Fig. 1 shows the absorption spectrum of the 6wt% Er\(^{3+}\)/Yb\(^{3+}\) co-doped tantalum-niobium phosphate glasses. The absorption bands are also indicated in the figure. The radiative transitions within the \(4f^n\) configuration of a rare earth ion can be analyzed by using the Judd and Ofelt theory\(^{[2,3]}\). The J-O intensity parameters \(\Omega_i\) (\(i=2, 4, 6\)) determined by using a least-square fitting method, were \(\Omega_2 = 3.70 \times 10^{20}\) cm\(^2\), \(\Omega_4 = 1.14 \times 10^{26}\) cm\(^2\) and \(\Omega_6 = 0.58 \times 10^{26}\) cm\(^2\). Using the J-O intensity parameters \(\Omega_i\), some important radiative properties are calculated. The electric dipole and magnetic dipole contributions to the spontaneous transition probability \(A\) are computed respectively as follows\(^{[4]}\):

\[
A_J (\Psi J, \Psi' J') = \frac{64\pi^2 e^2}{3h^2 (2J + 1)} n(n^2 + 2)^2 \sum_{i=2,4,6} \Omega_i \left< a^{S'L'} | J | a^{S'L}, J' \right>^2
\]

\[
A_M (\Psi J, \Psi' J') = \frac{4\pi^2 e^2 h}{3\hbar^2 m^2 c^2 (2J + 1)} \frac{\hbar^3}{n^3} \times \left< a^{S'L'} | (L + 2S) | a^{S'L'}, J' \right>^2
\]

where \(\Omega_i\) is the mean wavelength of the transition. \(\left< a^{S'L'} | J (L + 2S) \right>\) is the reduced matrix elements of tensor operators coming from \(^{[4]}\), \(\left< a^{S'L'} | (L + 2S) \right>\) is the magnetic dipole matrix elements coming from\(^{[5,6]}\).

The fluorescence branching ratio of transitions from the initial to lower levels is given by

\[
\beta_{J'} = \frac{A_{M'}}{\sum_j A_{M_j}}
\]

and the radiative lifetime is expressed as

\[
\tau_J = \frac{1}{\sum_j A_{J_j}}
\]

Table 2 reports these radiative properties calculated according to Judd-Ofelt theory.

4 RESULTS AND DISCUSSION

Stimulated emission cross-sections including their values and shape, are the essential parameters for the design and identification of amplifiers and lasers. The absorption cross sections of Er\(^{3+}\) ions were determined from the absorption spectra by

\[
\sigma_a (v) = \frac{2.303 \log (I_o / I)}{NL}
\]

Where \(\log (I_o / I)\) is absorbance, \(L\) is the sample thickness, and \(N\) is concentration of Er\(^{3+}\) ions (ion/cm\(^3\)).
Table 2 Spontaneous transition probabilities, fluorescent branch ratio and radiative lifetimes

<table>
<thead>
<tr>
<th>Transition</th>
<th>$A_{2f}$ (s⁻¹)</th>
<th>$A_{ad}$</th>
<th>$\beta_{sp}$</th>
<th>$\tau_{sp}$ (ms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^4I_{15/2}$ → $^4I_{15/2}$</td>
<td>66.9</td>
<td>46.1</td>
<td>1</td>
<td>8.85</td>
</tr>
<tr>
<td>$^4I_{15/2}$ → $^4I_{15/2}$</td>
<td>98.8</td>
<td>0.83</td>
<td>8.36</td>
<td></td>
</tr>
<tr>
<td>$^4I_{15/2}$ → $^4I_{15/2}$</td>
<td>10.6</td>
<td>10.2</td>
<td>0.17</td>
<td></td>
</tr>
<tr>
<td>$^4I_{15/2}$ → $^4I_{15/2}$</td>
<td>102.0</td>
<td>0.80</td>
<td>7.83</td>
<td></td>
</tr>
<tr>
<td>$^4I_{13/2}$ → $^4I_{13/2}$</td>
<td>24.2</td>
<td>0.19</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^4I_{13/2}$ → $^4I_{13/2}$</td>
<td>0</td>
<td>1.5</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>$^2F_{5/2}$ → $^4I_{15/2}$</td>
<td>1031.0</td>
<td>0.90</td>
<td>0.88</td>
<td></td>
</tr>
<tr>
<td>$^2F_{5/2}$ → $^4I_{15/2}$</td>
<td>55.9</td>
<td>0.05</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^2F_{5/2}$ → $^4I_{15/2}$</td>
<td>40.3</td>
<td>7.3</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>$^2F_{5/2}$ → $^4I_{15/2}$</td>
<td>2.8</td>
<td>3.1</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>$^2S_{3/2}$ → $^4I_{13/2}$</td>
<td>654.5</td>
<td>0.67</td>
<td>1.02</td>
<td></td>
</tr>
<tr>
<td>$^2S_{3/2}$ → $^4I_{13/2}$</td>
<td>266.5</td>
<td>0.27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^2S_{3/2}$ → $^4I_{13/2}$</td>
<td>21.6</td>
<td>0.02</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^2S_{3/2}$ → $^4I_{13/2}$</td>
<td>39.7</td>
<td>0.04</td>
<td></td>
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</tr>
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</table>

The emission cross sections of the $^4I_{13/2}$ → $^4I_{15/2}$ transition of Er³⁺ ions are calculated from McCumber theory[7]. According to the McCumber theory, the absorption and emission cross sections are related by

$$\sigma_a(v) = \sigma_e(v) \exp\left[\left(\frac{\epsilon - hv}{kT}\right)\right]$$

(6)

where $\sigma_a(v)$ and $\sigma_e(v)$ are the absorption and emission cross sections respectively, $v$ is the photon frequency, $\epsilon$ is the net free energy required to excited one Er³⁺ ion from the $^4I_{15/2}$ to $^4I_{13/2}$ state at temperature $T$, $h$ is the Planck's constant and $k$ is the Boltzmann constant. $\epsilon$ is determined from the absorption and emission spectra, using the procedure provided by Miniicale et al.[8]. Fig.2 illustrates the measured emission spectrum and the calculated absorption and emission cross sections for the $^4I_{13/2}$ → $^4I_{15/2}$ transition of Er³⁺ ions in the tantalum-niobium phosphate glasses. It shows that the shape of $\sigma_e(v)$ calculated agrees extremely well with that of the experimentally determined spectrum.

Fig.3 shows the emission cross sections around 1.53 µm for the samples a, b, c, d and e, where the concentrations of rare earth are a=4, b=5, c=6, d=7 and e=7.5wt%, respectively. From it we can see that the fluorescence intensities increase and then decrease with the increasing of the rare earth concentrations. The maximum intensity of fluorescence occurs at the concentration of 6wt%, its value is $6.8 \times 10^{-3}$ cm². It is larger than the values in alumina phosphate glasses[9]. The result shows the designed composition allowed us to add up to high concentration of rare earth to the glass batch.

As the peaks have not simple shapes, it is convenience to define a criterion to express quantitatively their bandwidths and compare them to other systems. We use the 'effective width of the peak' which is defined as[10] $\Delta \lambda = \int \sigma_e(\lambda)d\lambda / \sigma_{peak}$.
where $\sigma_\lambda(\lambda)$ is the emission cross section at wavelength $\lambda$ and $\sigma_{\text{peak}}$ is the value at the peak.

Calculated emission cross sections show that the bandwidth at 1.53 $\mu$m corresponding to Er$^{3+}_{4I_{13/2}} \rightarrow 4I_{15/2}$ transitions are about 60.0nm. The value is wider than that of Er$^{3+}$-doped Ge-P silicate glasses and Er$^{3+}$-doped aluminum phosphate glass [11].

5 CONCLUSION

The tantalum-niobium phosphate glasses with high Er$^{3+}$/Yb$^{3+}$ concentration were prepared. The effect of Er$^{3+}$/Yb$^{3+}$ ion concentrations on the spectroscopic properties was researched. The emission cross sections for $4I_{13/2} \rightarrow 4I_{15/2}$ transition and some important radiative properties of Er$^{3+}$ were calculated by McCumber theory and Judd-Oelft theory. It is found that the emission cross section around 1.53 $\mu$m varies with the variation of the rare earth concentrations, the maximum intensity of fluorescence occurs at the concentration of 6wt%.

The peak values of emission cross section is $6.8 \times 10^{-21}$ cm$^2$ and the effective emission cross section bandwidths are about 60.0nm. These values are larger than the silica glasses and other phosphate glasses, the large peak value and bandwidth of stimulated emission cross section make the tantalum-niobium phosphate glasses much more attractive candidates for microchip lasers and waveguide amplifiers.

REFERENCE