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Energy level decay processes in Ho$^{3+}$-doped tellurite glass relevant to the 3 $\mu$m transition

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The primary excited state decay processes relating to the $^5I_6 \rightarrow ^5I_7$ $\sim 2.9$ $\mu$m laser transition in singly Ho$^{3+}$-doped tellurite (TZBG) glass have been investigated in detail using time-resolved fluorescence spectroscopy. Selective laser excitation of the $^5I_6$ energy level at 1151 nm and $^5I_7$ energy level at 1958 nm has established that the rate of energy transfer up-conversion between holmium ions excited to the $^5I_7$ level is negligible for Ho$^{3+}$ concentrations up to 4 mol. %. Excited state absorption was not observed from either the $^5I_7$ or $^4I_{15/2}$ levels and the luminescence from the $^7I_2$ and $^7I_6$ energy levels was measured to peak at $\sim$2050 nm and $\sim$2930 nm, respectively. The $^5I_6$ level has a low luminescence efficiency of $\sim$8.9% due to strong nonradiative multiphonon relaxation. In contrast, decay from the $^5I_7$ level is essentially fully radiative. A linear decrease in the decay time of the $^5I_6$ level with Ho$^{3+}$ concentration augmentation results from energy transfer to OH$^-$ ions in the glass (with $N_{OH}/C_0 \sim 8.2 \times 10^{17}$ ions cm$^{-3}$) and reduces the luminescence efficiency of the $^5I_6$ level to 8% for [Ho$^{3+}$] = 4 mol. %. Numerical simulation of a fiber laser incorporating 4 mol. % Ho$^{3+}$ showed that a population inversion of $\sim$7.8% is reached for square pulses of 100 $\mu$s duration and a repetition frequency of 20 Hz at a moderate pump intensity of 418 kW cm$^{-2}$ if energy transfer to OH$^-$ radicals is neglected. © 2011 American Institute of Physics. [doi:10.1063/1.3587476]

I. INTRODUCTION

Rare earth doped fiber lasers developed for emission in the infrared wavelength region longer than 2 $\mu$m are of great interest for several applications, e.g., single frequency sources in this wavelength region would allow eye-safe coherent detection at long distance as a result of a number of atmospheric transmission windows that are available for $\lambda > 2\mu$m. The efficient detection of various molecules would also be possible because they have strong vibrational absorption lines in this wavelength region, e.g., in the so-called molecular “fingerprint” region.1,2 The use of the fiber geometry for the generation of shortwave and midwave infrared laser radiation introduces good thermal management and a comparatively low threshold because of the extended longitudinal dimension and small transverse cross section relevant to optical fibers.3

The $^5I_6 \rightarrow ^5I_7$ and $^4I_{11/2} \rightarrow ^4I_{13/2}$ $\sim 3\mu$m four-level transitions of Ho$^{3+}$ and Er$^{3+}$ offer broadband fluorescence and they have been successfully used to produce fiber lasers emitting in the shortwave infrared region in fluoride glasses.4,6 The Er$^{3+}$ transition has received the most interest because fiber lasers based on this transition can be pumped with commercial diode lasers and an energy transfer up-conversion (ETU) process offers the potential for slope efficiencies beyond the Stokes limit. Pump excited state absorption (ESA) is strong, however, and may limit the maximum achievable output power as a result of thermal and photo-induced absorption losses.

The Ho$^{3+}$ ion, on the other hand, offers a longer free-running wavelength and little ESA, however, ETU cannot recycle the excitation and pump excitation is achieved with narrowly available diode lasers. Despite these limitations, the potential for high-power efficient operation of a $\sim 3\mu$m fiber laser based on the Ho$^{3+}$ ion is strong and it is useful to study the characteristics of the $^5I_6 \rightarrow ^5I_7$ transition of Ho$^{3+}$ when doped in robust materials that could allow for high-power pumping without the risk of optical or thermal damage. This investigation therefore involves a detailed spectroscopic study of Ho$_2$O$_3$-doped tellurite (TZBG) glasses with varying Ho$_2$O$_3$ concentrations. Tellurite glasses have received a lot of interest because of their low maximum photon energy (of $\sim$800 cm$^{-1}$) and robust thermomechanical properties. In the this study, we reveal the important radiative and nonradiative energy level decay processes that relate to the $^5I_6 \rightarrow ^5I_7$ transition in tellurite glass after selective energy level excitation. Numerical simulations have been used to calculate the population inversion for the $^5I_6 \rightarrow ^5I_7$ laser emission near 3 $\mu$m in a Ho$^{3+}$-doped (4 mol. %) tellurite (TZBG) glass fiber laser for cw pumping at 1153 nm.

II. EXPERIMENTAL PROCEDURE

The Ho$^{3+}$-doped germanium tellurite (TZBG) glass samples used for the time-resolved luminescence spectroscopy were prepared from high-purity raw materials: 99.99% purity.
TeO₂, ZnF₂, and Bi₂O₃; and 99.999% purity GeO₂. The glass composition was \((100 - x) \times [74.5 \text{ TeO}_2-12.2 \text{ ZnF}_2-4.9 \text{ Bi}_2\text{O}_3-6.4 \text{ GeO}_2] + x \text{ Ho}_2\text{O}_3\) with \(x = 0.5, 1, 2,\) and \(4\) mol. %.

The starting powder materials were melted in a Pt-Au crucible in a dry glovebox environment at 900°C for 30 min to reduce the OH⁻ content in the glass. The molten liquids were poured into polished brass molds and annealed at 315°C for 4 h to remove any mechanical stress. The glass density was 5.89 g cm⁻³ and the measured refractive index was 1.98 at 2750 nm. The samples were cut and polished into \(4.7 \times 3.2 \times 1.4\) mm³ rectangular prisms. The Ho³⁺ density was calculated to be \(1.07 \times 10^{20}\) ions cm⁻³, \(2.14 \times 10^{20}\) ions cm⁻³, \(4.27 \times 10^{20}\) ions cm⁻³, and \(8.46 \times 10^{20}\) ions cm⁻³ for the tellurite samples having \(x = 0.5, 1, 2,\) and \(4\) mol. %, respectively.

The absorption spectra in the range of 2000–10,000 nm were measured using a FTIR spectrophotometer. The decay characteristics of the excited states of Ho³⁺ were measured using pulsed 10 mJ (4 ns) laser excitation from a tunable optical parametric oscillator (OPO) pumped by the second harmonic of a Q-switched Nd-YAG laser (Brilliant B from Quantel). Tunable laser excitation from the OPO was used to excite the \(5\text{I}_6\) energy level at 1958 nm and \(5\text{I}_7\) energy level at 1958 nm. The infrared luminescence (for \(\lambda > 1000\) nm) was detected using an InSb infrared detector (Judson model J-10 D cooled to 77 K) in conjunction with a fast preamplifier with a response time of \(\sim 0.5 \mu s\), and analyzed using a digital 200 MHz oscilloscope (Tektronix TDS 410). The visible and near infrared (i.e., \(\lambda < 1100\) nm) was investigated using a photomultiplier tube (EMI) with a sensitive cathode of the S-1 or S-2 type (refrigerated to \(-20^\circ C\)); the photomultiplier has a response time of 20 ns. All the fluorescence decay characteristics were measured at 300 K. To isolate the infrared luminescence signals, bandpass filters each with \(\sim 80\%\) transmission at 1200 nm or 2750 nm with a half width of 25 nm and an extinction coefficient of \(10^{-5}\) outside this band were used.

The optical absorption spectrum of Ho³⁺ ions in tellurite glass has two main features in the near infrared, one feature at around 1958 nm from the \(5\text{I}_6 \rightarrow 5\text{I}_1\) transition and a second feature near 1151 nm due to \(5\text{I}_6 \rightarrow 5\text{I}_1\) transition. When the Ho³⁺-doped material is excited at 1151 nm the following processes are known to occur in fluoride glass:

(a) Ground state absorption (GSA); Ho³⁺ \(\left(5\text{I}_6\right) + h\nu\) (1151 nm) \(\rightarrow\) Ho³⁺ \(\left(5\text{I}_0\right)\)

(b) Energy transfer up-conversion (ETU1); Ho³⁺ \(\left(5\text{I}_7\right) + \text{ Ho}³⁺ \left(5\text{I}_6\right) \rightarrow \text{ Ho}³⁺ \left(5\text{I}_0\right) + \text{ Ho}³⁺ \left(5\text{I}_8\right)\)

(c) Cross-relaxation (CR); Ho³⁺ \(\left(5\text{S}_2\right) + \text{ Ho}³⁺ \left(5\text{I}_8\right) \rightarrow \text{ Ho}³⁺ \left(5\text{I}_0\right) + \text{ Ho}³⁺ \left(5\text{I}_1\right) + \text{ Ho}³⁺ \left(5\text{I}_3\right)\)

Each of the processes listed above has been shown to affect the operation of the \(3 \mu m\) laser in the cw pumping regime in Ho³⁺-doped ZBLAN glass.⁷

III. EXPERIMENTAL RESULTS

Figure 1 shows the visible to shortwave (a) and midwave (b) absorption spectra of Ho³⁺ (2 mol. %)-doped tellurite glass. The spectrum shows a broad strong absorption band between 2500 cm⁻¹ and 3500 cm⁻¹, which we attribute to free OH⁻ groups.⁸ The narrow absorption band due to isolated OH⁻ radicals, with maximum absorption at \(3735\) cm⁻¹, was not observed. The concentration of OH⁻ radicals is given by \(N_{\text{OH}} = N_{\text{A}} n_{\text{OH}^-},\) where \(N_{\text{A}}\) is Avogadro’s constant (\(6.02 \times 10^{23}\)), and \(n_{\text{OH}^-}\) is the absorbance coefficient (=0.0672 cm⁻¹, relevant to the OH⁻ vibration band at \(3085\) cm⁻¹), and \(\zeta\) is the absorbivity of free OH⁻ groups in the glass. Using \(\zeta = 49.1 \times 10^{3}\) cm² mol⁻¹ (Ref. 9), we estimated the OH⁻ concentration in our samples to be \(8.24 \times 10^{17}\) ions cm⁻³, a value \(\sim 29\) times smaller than the free OH⁻ density (of \(2.4 \times 10^{19}\) cm⁻³) found in Er³⁺-doped germanotellurite glass produced from drying the melt using oxygen gas and CCl₄ bubbling.¹⁰

A. Infrared fluorescence spectrum of Ho³⁺ in tellurite (TZBG) glass

The fluorescence emission spectrum of Ho³⁺ in the shortwave infrared was measured for \([\text{Ho}³⁺] = 4\) mol. %. Figure 2 shows that there are two emission bands; one from the \(5\text{I}_6\) level at \(\sim 2930\) nm and one from the \(5\text{I}_7\) level at \(\sim 2050\) nm. The emission cross section due to free OH⁻ groups is the absorptivity of free OH⁻ groups in the glass. The absorption coefficient (\(\alpha = 0.0672 \text{ cm}^{-1}\)) of the OH⁻ vibration band at \(3085\) cm⁻¹, relevant to the OH⁻ vibration band at \(3085\) cm⁻¹, and \(\zeta\) is the absorbivity of free OH⁻ groups in the glass. Using \(\zeta = 49.1 \times 10^{3}\) cm² mol⁻¹ (Ref. 9), we estimated the OH⁻ concentration in our samples to be \(8.24 \times 10^{17}\) ions cm⁻³, a value \(\sim 29\) times smaller than the free OH⁻ density (of \(2.4 \times 10^{19}\) cm⁻³) found in Er³⁺-doped germanotellurite glass produced from drying the melt using oxygen gas and CCl₄ bubbling.¹⁰
(s^{-1}) is the radiative transition probability, S(\lambda) is the line shape of the emission band, \int S(\lambda)d\lambda is the integrated line shape, n is the refractive index (=1.98), and c is the speed of light.

The emission cross section of the \(^{5}\text{I}_6 \rightarrow \text{I}_7\) transition was calculated using the radiative transition probability \(A_{ij} = 138 \text{ s}^{-1}\), \(^{1,11}\) the experimental value of \(\overline{\lambda} = 2930 \text{ nm}\), and \(S(\lambda)/\int S(\lambda)d\lambda = 1.25 \times 10^{-2} \text{ nm}^{-1}\), which gave an emission cross section of \(4.30 \times 10^{-20} \text{ cm}^{2}\) at the maximum emission wavelength of 2930 nm. The emission cross section of the \(^{5}\text{I}_7 \rightarrow \text{I}_6\) transition was calculated using the radiative transition probability \(A_{ij} = \text{I}_{\text{rad}}/\text{I}_{\text{rad}}\), \(^{1,11}\) the experimental value of \(\overline{\lambda} = 2050 \text{ nm}\) and \(S(\lambda)/\int S(\lambda)d\lambda = 6.16 \times 10^{-3} \text{ nm}^{-1}\), which provides an emission cross section of \(6.37 \times 10^{-21} \text{ cm}^{2}\) at 2050 nm. The radiative lifetime of the \(^{4}\text{I}_7\) level was calculated according to the relationship \(^{12}\) \(\tau_{\text{rad}} = (\frac{2J+1}{2J_{\text{up}}+1}) \frac{\gamma^{2}}{8\pi c^{2}} \int \sigma_{\text{abs}}(\lambda)d\lambda\), where \(\tau_{\text{rad}}\) is the radiative lifetime, J and J' the total momentum for the upper and lower levels, \(\int \sigma_{\text{abs}}(\lambda)d\lambda\) is the integrated absorption cross section of the 2 \(\mu\text{m}\) band, and \(\overline{\lambda}\) the mean wavelength of the absorption band. The radiative lifetime of the \(^{4}\text{I}_7\) level was calculated using the \(\overline{\lambda} = 1964 \text{ nm}\) and the integrated absorption cross section \(\int \sigma_{\text{abs}}(\lambda)d\lambda = 7.7016 \times 10^{-26} \text{ cm}^{3}\) obtained from the spectrum shown in Fig. 1(a), giving \(\tau_{\text{rad}} = 5.8 \text{ ms}\), which is longer than the 3.52 ms for this lifetime given in Ref. 11.

**B. Emission decay from the \(^{5}\text{I}_6\) level**

Figure 3 shows the emission decay characteristic at 1200 nm for [Ho\(^{3+}\)] = 0.5 and 2 mol. % after laser excitation at 1151 nm with a mean pulse energy of 10 mJ and 4 ns pulse duration. The luminescence decay of \(^{5}\text{I}_6\) excited state was nonexponential and was fitted using the Burshtein model, \(^{13}\)

\[
I(t) = I_0 \exp(-\gamma \sqrt{t} - t/\tau_m),
\]

where \(\gamma (\text{s}^{-1/2})\) is the transfer constant due to the direct donor to acceptor transfer and \(\tau_m\) is defined by

\[
1/\tau_m = 1/\tau_R + W_{\text{nr}} + \omega,
\]

where \(\tau_R\) is the radiative lifetime and \(\omega\) is the transfer constant (s\(^{-1}\)) due to the migration assisted donor to acceptor transfer. \(W_{\text{nr}}\) is the nonradiative multiphonon decay rate (s\(^{-1}\)). Because the luminescence decay of \(^{5}\text{I}_6\) level at 1200 nm was nonexponential, the effective \(^{5}\text{I}_6\) lifetime (\(\tau\)) was obtained by integration, \(^{14}\) according to

\[
\tau = \frac{1}{I_0} \int_0^\infty I(t) dt,
\]

The best fit to the measured 1200 nm luminescence decay curve was carried out using Eq. (1); see dotted line in Fig. 3. The fitting parameters were: (i) \(\gamma = 32.4 \text{ s}^{-1/2}\), \(\tau_m = 136.8 \text{ ms}\), and \(\tau = 99.2 \text{ ms}\) for [Ho\(^{3+}\)] = 0.5 mol. % \((R^2 = 0.997)\); (ii) \(\gamma = 28 \text{ s}^{-1/2}\), \(\tau_m = 126 \text{ ms}\), and \(\tau = 96.3 \text{ ms}\) for [Ho\(^{3+}\)] = 1 mol. % \((R^2 = 0.999)\); (iii) \(\gamma = 23.4 \text{ s}^{-1/2}\), \(\tau_m = 119.3 \text{ ms}\), and \(\tau = 95.7 \text{ ms}\) for [Ho\(^{3+}\)] = 2 mol. % \((R^2 = 0.999)\); and (iv) \(\gamma = 14.8 \text{ s}^{-1/2}\), \(\tau_m = 102.1 \text{ ms}\), and \(\tau = 89.6 \text{ ms}\) for [Ho\(^{3+}\)] = 4 mol. % \((R^2 = 0.999)\).

Figure 4(a) shows that the decay time measured for \(^{5}\text{I}_6\) level is linearly dependent on the Ho\(^{3+}\) concentration in tellurite (TZBG) glass. This effect was attributed to energy transfer from the \(^{5}\text{I}_6\) level to OH\(^-\) radicals (i.e., free OH\(^-\) groups) that are present in the samples with the estimated concentration of \(8.24 \times 10^{17}\) ions cm\(^{-3}\). The rate of energy transfer to the radicals was obtained using

\[
W_i = 1/\tau - 1/\tau_R - W_{\text{nr}}.
\]

The intrinsic lifetime (\(\tau_0\)) of the \(^{5}\text{I}_6\) level was obtained by extrapolating the line in Fig. 4(a) to [Ho\(^{3+}\)] = 0, which gave \(\tau_0 = 100 \text{ ms}\). Assuming that for very low Ho\(^{3+}\) concentration, \(W_i\) is negligible and \(\tau_R = 1.12 \text{ ms}\) (Ref. 11) we obtain a value for the nonradiative decay rate, \(W_{\text{nr}}\), of \(9107 \text{ s}^{-1}\) for the \(^{5}\text{I}_6\) level. The following values of \(W_i\) were obtained: (i) \(76.6 \text{ s}^{-1}\) ([Ho\(^{3+}\)] = 0.5%), (ii) \(379 \text{ s}^{-1}\) ([Ho\(^{3+}\)] = 1%), and (iii) \(445 \text{ s}^{-1}\) ([Ho\(^{3+}\)] = 4%).

**FIG. 2.** Measured emission spectrum of Ho\(^{3+}\)(4 mol. %)-doped tellurite (TZBG) glass using a pulsed laser excitation at 1151 nm with an average energy of 13 mJ and pulse duration of 4 ns. Spectrum was measured using a boxcar technique.

**FIG. 3.** (Color online) Measured emission decay characteristic (solid lines) of the \(^{5}\text{I}_6\) level of Ho\(^{3+}\) (0.5 and 4 mol. %)-doped tellurite (TZBG) glass using short pulse laser excitation at 1151 nm with an average energy of 10 mJ and pulse duration of 4 ns at 10 Hz. Broken lines indicate the best fit using the Burshtein model of Eq. (1).
(\([\text{Ho}^{3+}] = 2\%\)), and (iv) 1157 s\(^{-1}\) ((\([\text{Ho}^{3+}] = 4\%)\). These values for \(W_t\) are essentially linearly dependent on \([\text{Ho}^{3+}]\) and indicate that \(5\text{I}_6\) energy level migration is important. As a consequence of energy transfer to OH\(^{-}/\text{C}_0\), the luminescence efficiency from \(5\text{I}_6\) level decreases from 8.9 to 8% for ([Ho\(^{3+}\]) = 4%. From these calculations it is clear that the main decay process for the \(5\text{I}_6\) level is multiphonon decay.

C. Emission decay from the \(5\text{I}_7\) level

Figure 5 shows the emission decay from the \(5\text{I}_7\) level measured at 2000 nm by pulsed laser excitation at 1958 nm (\(E = 10 \text{ mJ}, 4 \text{ ns}\)) for Ho\(^{3+}\)-doped TZBG glass as a function of \([\text{Ho}^{3+}]\). The solid line in (b) represents the best fit using the critical radius model given by Eq. (5), where \(\tau_{\text{rad}} = 5.8 \text{ ms}\), and the fitting parameters are \(\tau_0 = 5.3 \text{ ms}\) and \(N_C = 0.8 \text{ mol. %}\).

\[
\eta(\text{Ho}) = 1 - \exp\left(-N_{\text{Ho}}/N_C\right) \tau_d = \tau_R + \eta(\text{Ho})\tau_0 \quad (5)
\]

where \(\tau_R = 5.8 \text{ ms}\) and \(N_{\text{Ho}}\) is the Ho\(^{3+}\) concentration. The best fit using Eq. (4) gives \(\tau_0 = 5.3 \text{ ms}\) and \(N_C = 0.8 \text{ mol. %}\) [see solid line in Fig. 4(b)].

No up-conversion processes (ESA or ETU) starting from the \(5\text{I}_7\) or \(5\text{I}_6\) levels were observed in our measurements. The expected green emission from the \(5\text{S}_2\) level or the red emission from the \(5\text{F}_5\) level was not observed despite that the fact that it can be easily observed in Ho\(^{3+}\)-doped ZBLAN glass under pulsed laser excitation at 1958 nm (Ref. 16) or in Er\(^{3+}\)-doped tellurite glass with 980 nm pumping.\(^{21}\)

IV. DISCUSSION

Tellurite glasses are an interesting alternative host material for shortwave and midwave infrared fiber lasers; because they combine a relatively low phonon energy with higher resonant dipole-dipole interaction between the \(5\text{I}_7\) and \(5\text{I}_8\) energy levels. This phenomenon explains the initial increase in the decay time as the Ho\(^{3+}\) concentration is increased. Note that measuring the luminescence close to the pumped surface can minimize radiation trapping. In our experiment, we observed an increase in the decay time at distances perpendicular to the pump of \(\sim 8 \text{ mm}\), however, for distances of \(\sim 3 \text{ mm}\) and shorter (from where we took the measurements) the measured decay time was invariable.

The line of best fit to the decay time characteristic [open circles in Fig. 4(b)] with the increase in \([\text{Ho}^{3+}]\) was carried out using Eq. (5). Equation (5) predicted that the lifetime of the \(5\text{I}_7\) level increases if a Ho\(^{3+}\) ion excited to the \(5\text{I}_7\) level will transfer excitation to a ground state Ho\(^{3+}\) ion within a critical distance, \(R_C\), with a decay time augmentation efficiency, \(\eta(\tau)\), dependent on the Ho\(^{3+}\) concentration.\(^{15,16}\) This model predicts a saturation of the decay time increase to a value \(\tau_{\text{rad}} + \tau_0\), which should be reached for \(N_{\text{Ho}} > N_C\), where \(N_C\) is the critical concentration calculated from
thermals and chemical stability compared with, for example, fluoride glasses. On the other hand, tellurite glasses display lower luminescence efficiencies compared to fluoride glasses but they display higher emission cross sections and are easier to handle and splice to silica fibers. Tellurite glasses have shown efficient emission at \( \sim 2 \, \mu m \) from \( \text{Tm}^{3+} \), \( \text{Ho}^{3+} \)-co-doped tellurite glass optical fiber lasers,\(^{17-19} \) however, there has been no demonstration of laser operation beyond \( \sim 2 \, \mu m \) in tellurite glass.

The results presented here indicate that perhaps the main issue with these glasses are water incorporation and the low luminescence efficiency of the \( 5I_6 \) level. To illustrate this we have carried out a numerical simulation to predict the performance of \( \text{Ho}^{3+} \)-doped tellurite fiber lasers. The experimental observation that ETU1 is weak in \( \text{Ho}^{3+} \) TZBG glass (at least up to \( \text{Ho}^{3+} \) of 4 mol. %) implies that unlike the \( \text{Er}^{3+} \)-doped ZBLAN system,\(^{20} \) energy recycling is not possible for moderately concentrated \( \text{Ho}^{3+} \)-doped tellurite glasses when pumped into the \( 5I_6 \) level.

The rate equations were written to include the essential energy levels and pump and decay processes for laser emission at 2930 nm when the \( 5I_6 \) level is excited with pump radiation. The rate equations are

\[
\frac{dn_1}{dt} = -R_P n_1 + \frac{n_2}{\tau_2} + \frac{\beta_{31}}{\tau_{R3}} n_3, \quad (6)
\]

\[
\frac{dn_2}{dt} = -\frac{n_2}{\tau_2} + \frac{\beta_{32}}{\tau_{R3}} n_3 + W_{aR}(32)n_3 + W_l(2)n_3, \quad (7)
\]

\[
\frac{dn_3}{dt} = R_P n_1 - \frac{n_3}{\tau_{R3}} - W_{aR}(32)n_3 - W_l(2)n_3, \quad (8)
\]

where \( n_1, n_2, \) and \( n_3 \) indicate the \( 5I_8, 5I_7, \) and \( 5I_6 \) levels and \( n_1 + n_2 + n_3 = x, \) where \( x = 0.005, 0.01, 0.02, \) and 0.04 (i.e., mole fraction). The radiative lifetimes, luminescence branching ratios, and nonradiative multiphonon decay rate values that were obtained from our experiment are: \( \tau_{R3} = 1.12 \, ms, \) \( \beta_{32} = 0.16, \) \( \beta_{31} = 0.84, \) \( \tau_2 = 8.9 \, ms \) (4 mol. %), \( W_{aR}(32) = 9107 \, s^{-1}, \) and \( W_A(2) = 1157 \, s^{-1}. \) Note that \( W_{aR}(21) \sim 0. \) The pump rate \( (R_P) \) can be converted to pump irradiance \( (I_P) \) using the relation: \( R_P(x^{-1}) = \frac{\sigma_{abs}(\lambda) I_P}{W_c m^{-2}}. \)

Using the absorption cross section \( \sigma_{abs}(\lambda) = 3.3 \times 10^{-21} \, cm^2 \) for the pump wavelength \( \lambda = 1153 \, nm, \) one obtains \( R_P = 1000 \, s^{-1} \) for a pump intensity \( I_P = 52.2 \, kW \, cm^{-2}. \)

The calculated evolution of the excited state populations \( n_3 \) and \( n_2 \) (in \( cm^{-3} \)) were used to determine the population difference \( (n_3 - n_2) \), as shown in Fig. 6. One can see that steady state is reached in a time \( < 2 \, ms. \) At equilibrium, the populations \( n_3, n_2, \) and \( n_1 \) were used to calculate the population inversion \( \Delta n = n_3 - n_2 \) for \([\text{Ho}^{3+}] = 0.5, 1, 2, \) and 4 mol. % for a number of pump intensities at 1153 nm. For all the simulations, we obtained a steady state but negative population inversion [even with \( W_l(2) = 0. \) A positive population inversion, however, occurs for pump times shorter than 100 \( \mu s \) with a maximum in \( \Delta n \) occurring 55 \( \mu s \) after the pump is switched on; see Fig. 7(a). This result indicates that maximum laser emission can be achieved by pump pulses of 55 \( \mu s \) duration with a minimal interval of time between pulses of 50 ms. This time interval is necessary for complete emptying of the \( 5I_7 \) level, as shown in Fig. 7(b). Using a square modulated 50 \( \mu s \) duration pump one can get \( \Delta n = 0.75 \, mol. \% \) for \([\text{Ho}^{3+}] = 4 \, mol. \%. \)

Figure 8 shows the maximum calculated population inversion using square 1153 nm pump pulses of varying

![FIG. 6. (Color online) Calculated population inversion \((n_3 - n_2)\) for cw pumping of Ho\(^{3+}\)-doped TZBG glass using a pumping intensity of \(~418 \, kW \, cm^{-2} (R_P = 8000 \, s^{-1})\) for [Ho\(^{3+}\)] = 1, 2, and 4 mol. %.](image)

![FIG. 7. Calculated (a) population inversion \((n_3 - n_2)\) for a square pump pulse of 55 \( \mu s \) duration and pulse period \(~50 \, ms\) and (b) \( 5I_7 \) population \((n_2)\). The pump intensity was \( 418 \, kW \, cm^{-2} \) at 1153 nm, [Ho\(^{3+}\)] = 4 mol. %, \( W_l = 0. \)](image)
duration for \([\text{Ho}^{3+}] = 4\text{ mol.~%}\) as a function of the pump rate \((R_p)\). It can be observed that the population inversion is greatest when \(t_{ON} = 55\mu s\) and the population inversion decreases with increasing pumping duration. Note that for these simulations, no losses were included and, hence, the simulations are only a guide to possible performance from a fiber laser arrangement.

V. SUMMARY AND CONCLUSIONS

The decay processes relating to the \(5^1I_7 \rightarrow 5^1I_6 \sim 2\mu m\) and \(5^1I_6 \rightarrow 5^1I_7 \sim 2.9\mu m\) laser transitions in singly \(\text{Ho}^{3+}\)-doped tellurite (TZBG) glass were investigated in detail using selective laser excitation of the \(5^1I_6\) energy level at 1151 nm and \(5^1I_7\) energy level at 1958 nm. The results have established that a nonradiative ETU between two excited \(\text{Ho}^{3+}\) ions in the \(5^1I_7\) level is negligibly weak for \(\text{Ho}^{3+}\) concentrations \(\leq 4\text{ mol.~%}\). Pump ESA from the \(5^1I_7\) or \(5^1I_6\) levels was not observed when these levels were excited directly. The \(5^1I_7\) and \(5^1I_6\) energy levels were measured to emit luminescence with peaks at \(\sim 2050\) nm and \(\sim 2930\) nm, respectively. The \(5^1I_6\) level has low luminescence efficiency of 8\% because of nonradiative multiphonon relaxation, however, decay from the \(5^1I_7\) level was observed to be fully radiative. The decreasing decay time of the \(5^1I_6\) level with increasing \([\text{Ho}^{3+}]\) was attributed to energy transfer to the \(\text{OH}^-\) radicals present in the glass. The numerical simulations indicate that the prospect for cw operation on the \(5^1I_6 \rightarrow 5^1I_7\) transition is low in \(\text{Ho}^{3+}\)-doped tellurite glass.

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