Energy-transfer upconversion and excited-state absorption in KgdₜLuₜEr₁₋ₓ₋ₓ(WO₄)₂ waveguide amplifiers

Sergio A. Vázquez-Córdova,¹ Shanmugam Aravazhi,² Alexander M. Heuer,³ Christian Kränkel,³,⁴ Yean-Sheng Yong,¹ Sonia M. García-Blanco,¹ Jennifer L. Herek,¹ and Markus Pollnau²,⁵,*

¹Optical Sciences Group, MESA+ Institute for Nanotechnology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands
²Integrated Optical Micro Systems Group, MESA+ Institute for Nanotechnology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands
³Institut für Laser-Physik, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany
⁴Center for Laser Materials, Leibniz-Institut für Kristallzüchtung, Max-Born-Str. 2, 12489 Berlin, Germany
⁵Advanced Technology Institute, Department of Electrical and Electronic Engineering, University of Surrey, Guildford GU2 7XH, United Kingdom

Abstract: We perform a systematic spectroscopic study in channel waveguides of potassium gadolinium lutetium double tungstate doped with different Er³⁺ concentrations. Transition cross sections of ground-state absorption (GSA) and excited-state absorption (ESA), as well as stimulated emission (SE) at the pump wavelength around 980 nm are determined. ESA is directly measured by the pump-probe technique. Evaluation of GSA and ESA spectra indicates that ESA may be diminished by an appropriate choice of pump wavelength near 980 nm. Besides, GSA and SE at the signal wavelength around 1.5 μm are measured and the wavelength-dependent gain cross section as a function of excitation density is determined. Non-exponential luminescence decay curves from the ⁴I_{13/2} and ⁴I_{11/2} levels are analyzed and the probabilities of the energy-transfer-upconversion (ETU) processes (⁴I_{13/2}, ⁴I_{13/2}) → (⁴I_{15/2}, ⁴I_{9/2}) and (⁴I_{11/2}, ⁴I_{11/2}) → (⁴I_{15/2}, ⁴F_{7/2}) are quantified. Despite the large interionic distance between neighboring rare-earth sites in potassium double tungstates, the probability of ETU is comparatively large because of the large cross sections of the involved transitions. A rate-equation analysis of the influence of ETU and ESA on gain at ~1.5 μm is performed, revealing that ETU from the ⁴I_{13/2} amplifier level strongly limits the gain when the doping concentration increases above ~6 at.%. The calculated maximum achievable internal net gain per unit length amounts to ~15 dB/cm for an optimized Er³⁺ concentration of ~4 × 10²⁰ cm⁻³ and a launched pump power of 300 mW at a pump wavelength of 984.5 nm, in reasonable agreement with recent experimental results.

© 2019 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

1. Introduction

The rare-earth-doped potassium double tungstates KY(WO₄)₂, Kgd(WO₄)₂, and KLu(WO₄)₂ are widely investigated laser materials, see [1] and references therein. The thermomechanical [2] and spectroscopic properties [3] of these materials are superior to those found in many glass and crystalline hosts, mainly due to their strong anisotropy and high refractive index [3,4]. Channel waveguides in these materials have shown excellent performance in optical amplification [5] and lasing [6,7]. For Er³⁺-activated waveguide amplifiers and lasers operating in the telecom C-band at wavelengths near 1.5 μm [8], see Fig. 1, usually low Er³⁺ concentrations of ~1-2 × 10²⁰ cm⁻³ are chosen, because energy-transfer upconversion (ETU) between neighboring active ions
depopulates the upper level of the amplifier transition and imposes a maximum attainable gain value on each host material [10–15]. Nevertheless, with a long spiral waveguide an internal net gain of 20 dB was demonstrated [16]. Incorporation of Er³⁺ in potassium double tungstates is possible up to the stoichiometric compound KEr(WO₄)₂, resulting in an Er³⁺ concentration of 6.3 × 10²¹ cm⁻³ [17]. The large inter-ionic distance between neighboring rare-earth sites [18,19] in potassium double tungstates reduces the probability of ETU and potentially allows for higher doping levels [20].

Fig. 1. Simplified energy-level diagram of Er³⁺ displaying the most relevant transitions for amplification around 1530 nm: the GSA transitions ⁴Ι₁₅/₂ ↔ ⁴Ι₁₃/₂ and ⁴Ι₁₅/₂ ↔ ⁴Ι₁₁/₂ around 1480 nm, the ESA transition ⁴Ι₁₁/₂ → ⁴F₇/₂ around 980 nm, the ground-state luminescence (LUM) transitions, as well as stimulated-emission (SE) transitions ⁴Ι₁₃/₂ → ⁴Ι₁₁/₂ and ⁴Ι₁₁/₂ → ⁴Ι₁₅/₂ around 1530 nm and 980 nm, respectively, non-radiative multiphonon decay (NR), and the ETU process ETU₁ (⁴Ι₁₃/₂, ⁴Ι₁₁/₂) → (⁴Ι₁₅/₂, ⁴Ι₇/₂). τᵢ are the measured and estimated (*) [3,9] luminescence lifetimes. In high-phonon oxide materials, leading to multiphonon quenching of luminescence lifetimes, other processes usually have a smaller influence: the ETU process ETU₂ (⁴Ι₁₁/₂, ⁴Ι₁₁/₂) → (⁴Ι₁₅/₂, ⁴F₇/₂) and the cross-relaxation process CR (²H₁₁/₂, ⁴I₉/₂, ⁴I₁₅/₂) → (⁴I₉/₂, ⁴I₁₃/₂).

Structuring channel waveguides into layers provides a laterally confined propagation of signal and pump light with excellent mode overlap and high intensities within the active region [21], thereby enabling the utilization of higher doping concentrations to achieve higher gain per unit length. In this work we present a systematic study of the most relevant spectroscopic processes in Er³⁺-doped potassium double tungstate waveguides. Ground-state-absorption (GSA) and stimulated-emission cross sections of the ⁴Ι₁₅/₂ ↔ ⁴Ι₁₃/₂ and ⁴Ι₁₁/₂ ↔ ⁴Ι₁₅/₂ transitions are determined. Due to the rather high phonon energies of ∼904–935 cm⁻¹ present in potassium double tungstates [22,23], leading to fast multiphonon relaxation and accordingly short lifetime [24] of the ⁴Ι₁₁/₂ level (τ₂ ≈ 135 µs) and, particularly, the ⁴Ι₉/₂ level (τ₃ ≈ 1 µs) [3], their excitation rapidly decays to the ⁴Ι₁₁/₂ level. Decay curves and lifetimes of luminescence from the ⁴Ι₁₃/₂ and ⁴Ι₁₁/₂ levels are presented, from which microscopic parameters of migration and ETU from the ⁴Ι₁₃/₂ level are determined and the macroscopic ETU parameter is obtained. The macroscopic parameter of ETU from the ⁴Ι₁₁/₂ level is estimated. Pump excited-state absorption (ESA) on the ⁴Ι₁₁/₂ → ⁴F₇/₂ transition around 980 nm is measured. The influence of ETU and pump ESA on optical gain at ∼1.5 µm is analyzed by use of a rate-equation model.
Table 1. Optimized compositions of layers with different Er$^{3+}$ concentrations, and waveguide dimensions.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Stoichiometric formula</th>
<th>Er$^{3+}$ [10$^{20}$ cm$^{-3}$]</th>
<th>$t$ [μm]</th>
<th>$w$ [μm]</th>
<th>$d$ [μm]</th>
<th>$ℓ$ [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>KGd$0.492$LLu$_0.508$Er$_0.0075$(WO$_4$)$_2$</td>
<td>0.48</td>
<td>6.4</td>
<td>5.6</td>
<td>1.48</td>
<td>5.07</td>
</tr>
<tr>
<td>II</td>
<td>KGd$0.486$LLu$_0.515$Er$_0.015$(WO$_4$)$_2$</td>
<td>0.95</td>
<td>7.7</td>
<td>7.9</td>
<td>1.43</td>
<td>8.60</td>
</tr>
<tr>
<td>III</td>
<td>KGd$0.485$LLu$_0.515$Er$_0.015$(WO$_4$)$_2$</td>
<td>1.90</td>
<td>7.1</td>
<td>7.8</td>
<td>1.27</td>
<td>6.75</td>
</tr>
<tr>
<td>IV</td>
<td>KGd$0.475$LLu$_0.515$Er$_0.010$(WO$_4$)$_2$</td>
<td>3.81</td>
<td>8.5</td>
<td>7.5</td>
<td>1.55</td>
<td>0.75</td>
</tr>
<tr>
<td>V</td>
<td>KGd$0.462$LLu$_0.537$Er$_0.015$(WO$_4$)$_2$</td>
<td>6.36</td>
<td>3.1</td>
<td>5.6</td>
<td>1.56</td>
<td>0.58</td>
</tr>
</tbody>
</table>

2. Transition cross sections

Crystalline layers of KGd$_x$Lu$_y$Er$_{1-x-y}$(WO$_4$)$_2$ (abbreviated hereafter as KGLW:Er$^{3+}$) with five different Er$^{3+}$ concentrations, lattice matched by appropriate Gd$^{3+}$ and Lu$^{3+}$ concentrations (see Table 1), were grown by liquid-phase epitaxy (LPE) onto undoped KY(WO$_4$)$_2$ substrates [25,26]. Rib channel waveguides were microstructured by Ar$^+$ etching [21] parallel to the $N_p$ axis in the $N_m$-$N_p$ plane of the crystalline layer, hence the optical modes propagate with a polarization of either $E||N_m$ or $E||N_p$. Composition and channel thickness ($t$), width ($w$), rib height ($d$), and length ($ℓ$) of each investigated sample are shown in Table 1.

2.1. Ground-state-absorption and -emission cross sections

Luminescence spectra, with the luminescence polarized parallel to the three principal optical axes, $E||N_p$, $E||N_m$, and $E||N_g$, on the transitions $^4$I$_{13/2}$ $→$ $^4$I$_{15/2}$ near 1.5 μm and $^4$I$_{11/2}$ $→$ $^4$I$_{15/2}$ near 1 μm were recorded during pump excitation by a continuous-wave (CW) Ti:sapphire laser operating at the wavelength of 984.5 nm or 800 nm, respectively. The sample with the lowest doping concentration (sample I) was chosen to minimize re-absorption effects. The polarized luminescence intensity $I_q(λ)$ was collected from the plane perpendicular to the excitation incidence and the spectra were dispersed by a spectrometer (Horiba-Yvon iHR550) with a resolution of 0.4 nm and detected by a cooled InAs detector. Emission cross sections were calculated from the Füchtbauer-Ladenburg equation for monoclinic biaxial crystals [27,28],

$$\sigma_{e,q}(λ) = \frac{3λ^5}{8πτ_r n_q^2 c} \int λ[I_q(λ) + I_{N_m}(λ) + I_{N_g}(λ)] dλ,$$

(1)

where $q$ stands for each of the principal axes of $N_p$, $N_m$, or $N_g$, $λ$ is the wavelength, $τ_r$ is the radiative lifetime of the emitting level, $n_q$ is the material refractive index parallel to the axis $q$, $c$ is the speed of light in vacuum, and $I_q$ is the axis-respective intensity spectrum. Refractive indexes were measured by use of the prism-coupling technique (Metricon 2010) at 633 nm, 830 nm, 1300 nm, and 1550 nm parallel to each optical axis, and the refractive index values at the pump and signal wavelength were interpolated using the single-term Sellmeier equation. For $λ_P = 980$ nm, we obtained $n_g = 2.067$, $n_m = 2.021$ and $n_p = 1.989$, and for $λ_S = 1534$ nm the values are $n_g = 2.054$, $n_m = 2.008$, and $n_p = 1.977$. The radiative lifetime of the $^4$I$_{11/2}$ level, $τ_{r,2} = 2.137$ ms [29] and the intrinsic luminescence lifetime of the $^4$I$_{13/2}$ level presented below, $τ_{r,1} ≈ τ_1 = 3.05$ ms, were considered for the calculations. The resulting polarized emission cross sections on the transitions $^4$I$_{13/2}$ $→$ $^4$I$_{15/2}$ and $^4$I$_{11/2}$ $→$ $^4$I$_{15/2}$ are displayed in Fig. 2(a) and 2(c), respectively.

Using the reciprocity theorem [30], the related absorption cross sections on the transitions $^4$I$_{15/2}$ $→$ $^4$I$_{13/2}$, see Fig. 2(b), and $^4$I$_{15/2}$ $→$ $^4$I$_{11/2}$, see Fig. 2(d), were calculated from

$$\sigma_{a,q}(λ) = \sigma_{e,q}(λ) \frac{Z_q}{Z_0} \frac{h c (λ^4 - λ^4')}{λ^2} \frac{1}{κ_0},$$

(2)
where $h$ is the Planck constant, $k_B$ is the Boltzmann constant, $\lambda_{ZL}$ = 1535 nm for $^4I_{15/2} \leftrightarrow ^4I_{13/2}$ or 981.5 nm for $^4I_{15/2} \leftrightarrow ^4I_{11/2}$ is the wavelength of the zero-phonon-line transition, $T = 300$ K is the temperature, and $Z_0 = 4.5851$, $Z_1 = 4.5395$, and $Z_2 = 4.6606$ are the partition functions of the $^4I_{15/2}$, $^4I_{13/2}$, and $^4I_{11/2}$ levels, respectively. $\lambda_{ZL}$ and the energy levels of the manifolds used to calculate $Z_i$ were extracted from the literature [31–35].

### 2.2. Pump excited-state-absorption cross sections

The Er$^{3+}$ ion is known for its large number of ESA transitions from the $^4I_{13/2}$, $^4I_{11/2}$, and $^4S_{3/2}$ levels [36]. Excitation spectra of short-wavelength luminescence obtained by GSA and ESA of a single pump source [9] provide an indication of the presence of ESA but do not deliver the actual ESA spectra, because the wavelength dependence of GSA modifies the excitation density and the ESA rate. Pump-probe ESA measurements in an Er$^{3+}$-doped KY(WO$_4$)$_2$ bulk crystal parallel to the three crystallographic axes, $E||a$, $E||b$, and $E||c$, were presented in [3], but due to the usually low excitation density in bulk materials ESA transitions were observed only from the $^4I_{13/2}$ level. With the tight mode confinement in our channel waveguides, a significantly higher excitation density is achieved in the $^4I_{11/2}$ level and ESA from this level can be detected as well.

ESA in the wavelength range 950–1030 nm was measured in a pump-probe setup [37,38], in which the signal and the pump are counter-propagating through the sample. A channel waveguide (sample II) with a length of $\ell = 8.6$ mm and an Er$^{3+}$ concentration of $0.95 \times 10^{20}$ cm$^{-3}$ was chosen. Approximately 500 mW of pump power at $\lambda_p$ = 800 nm ($E||N_m$) from a CW Ti:Sapphire laser modulated at $\sim 11$ Hz by a chopper were incident on the sample, exciting the Er$^{3+}$ ions from the ground state to the $^4I_{9/2}$ level, from which fast multi-phonon relaxation populated the $^4I_{11/2}$ level. A super-continuum white-light source (Fianium) modulated at $\sim 1$ kHz by a second chopper was used as the signal. The transmitted signal intensity $I_p(\lambda)$ or $I_u(\lambda)$ under pumped or unpumped conditions, respectively, was dispersed by a spectrometer with a resolution of 0.3 nm
and detected by a silicon detector. The difference signal \( \Delta I(\lambda) = I_p(\lambda) - I_u(\lambda) \) was amplified by a double lock-in technique. The difference signal accounts for GSA \(^4\text{I}_{15/2} \rightarrow ^4\text{I}_{11/2}\), ESA \(^4\text{I}_{11/2} \rightarrow ^4\text{F}_{7/2}\), and SE \(^4\text{I}_{11/2} \rightarrow ^4\text{I}_{15/2}\) present in the wavelength range 950–1030 nm. Following the analysis in [37], we obtain the spectra defined by

\[
\ln \left( \frac{\Delta I(\lambda)}{I_u(\lambda)} + 1 \right) = \sigma_a(\lambda) N_e \Gamma \ell + (\sigma_e(\lambda) - \sigma_{ESA}(\lambda)) N_2^2 \Gamma \ell,
\]

where \( N_e \) is the total averaged excitation density, \( N_2 \) is the averaged excitation density of the \(^4\text{I}_{11/2}\) level, \( \Gamma \) is the overlap factor of the signal mode with the active layer (calculated from simulated mode profile), and \( \sigma_a(\lambda) \), \( \sigma_e(\lambda) \), and \( \sigma_{ESA}(\lambda) \) are the GSA, emission, and signal ESA cross sections, respectively. The results are presented in Fig. 3(a) and 3(b) for signal polarizations of \( E||N_m \) and \( E||N_p \), respectively. A rate-equation model [37], see later in Section 4, was used to determine the parameters \( N_e \) and \( N_2 \). Pump-GSA and emission cross sections \( (E||N_m) \) at 800 nm were taken from [9] and pump-ESA cross sections \( (E||a) \) at 800 nm from [3]. While the pump polarization was chosen as \( E||N_m \) during the experiments, due to improved pump-coupling conditions when realigning the experimental set-up between experiments different pump-coupling efficiencies were obtained during the two independent measurements for signal \( E||N_m \) and \( E||N_p \), resulting in different launched pump powers, hence also different \( N_e \) and \( N_2 \) (averaged along the entire length of the waveguides) for the two measurements (Table 2). From Eq. (3) the ESA cross sections of Fig. 3(c) and 3(d) were then calculated. The resulting ESA cross sections are nominally independent of the amount of launched pump power.

![Fig. 3. Experimental pump-probe spectra combining the contributions from ESA, GSA, and SE parallel to the (a) \( N_m \) and (b) \( N_p \) axis. Comparison of GSA and ESA cross sections for (c) \( E||N_m \) and (d) \( E||N_p \).](image)

The ESA spectra are blue-shifted with respect to the GSA spectra, see the comparison in Fig. 3(c) and 3(d), thus a longer pump wavelength will evoke less ESA. On the other hand, the
GSA cross sections at the long-wavelength side of the spectra are lower, thereby leading to lower pump absorption. Pumping at 984.5 nm instead of the GSA peak for $E[N_{a}]$ at 979 nm is promising, because the decrease in GSA is insignificant, whereas the ESA is substantially lower, see Fig. 3(c). Although a longer pump wavelength within the same multiplet-to-multiplet transition generally leads to larger stimulated emission on the pump wavelength, hence lower inversion and lower gain, this effect amounts to a difference of only a few percent between 979 nm and 984.5 nm, whereas the improvement due to diminished ESA is significantly larger.

### 3. Luminescence decay and energy-transfer processes

With a setup similar to the one presented in [10], luminescence decay on the transitions $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ at 1535 nm, $^{4}I_{11/2} \rightarrow ^{4}I_{15/2}$ at 1010 nm, and $^{4}S_{3/2} \rightarrow ^{4}I_{15/2}$ at 545 nm was measured. A fiber-coupled laser diode with $\lambda_p = 1480$ nm or 976 nm directly excited the $^{3}I_{13/2}$ or $^{1}I_{11/2}$ level, respectively. The luminescence decay at 545 nm was detected after 976 nm excitation. The diode power was square-wave modulated by a function generator at a frequency of 20 Hz and a duty cycle of 50%, which allowed the Er$^{3+}$ population densities to reach equilibrium before the pump was switched off. A glass light guide with a diameter of 0.6 mm was placed perpendicularly close to the in-coupling end of the waveguide, thereby collecting the luminescence from the region of maximum excitation density and minimizing re-absorption effects. The collected light was dispersed in a monochromator (H25 Yvon-Jobin) and detected by an InGaAs detector (ETX100 T) or a silicon detector (PIN-3CD). The current was amplified (FEMTO dhpca-100) and recorded by an oscilloscope (HP Infinium 54845A) which was triggered by the same function generator modulating the pump diode. 4096 samples were averaged by the oscilloscope.

#### 3.1. Luminescence decay and energy-transfer upconversion from $^{4}I_{13/2}$

Normalized luminescence-decay curves at 1535 nm for the five different Er$^{3+}$ concentrations (see Table 1) were recorded for four different pump powers, see Fig. 4(a)–4(d). The spectroscopic processes affecting luminescence decay on the $^{4}I_{13/2} \rightarrow ^{4}I_{15/2}$ transition are depicted in Fig. 1. After pump excitation into the $^{4}I_{13/2}$ level, this level is depleted via ETU$_1$ ($^{3}I_{13/2}$, $^{4}I_{11/2}$) $\rightarrow$ $^{4}I_{15/2}$, $^{4}I_{11/2}$), thereby populating the $^{4}I_{11/2}$ level, followed by a fast non-radiative multi-phonon relaxation to $^{4}I_{11/2}$. In the luminescence-decay curves of Fig. 4(a)–4(d), a fast non-exponential decay is observed during the first $\sim$1.5 ms due to migration-accelerated ETU$_1$, which is accentuated for higher Er$^{3+}$ concentrations and higher pump powers. The decay slows down and after 8 ms an exponential decay is observed. The intrinsic luminescence lifetime of the $^{4}I_{13/2}$ level was extracted from this exponential decay at delay times $> 8$ ms for all luminescence-decay curves, and an average of $\tau_1 = 3.05$ ms was obtained, see Fig. 4(e). Unlike in other materials [10,39,40], the exponential tail does not exhibit a concentration-dependent quenching for the concentration range studied. This finding could be a consequence of the high-purity raw materials and the high crystallinity of the LPE layers, as well as the large distance between neighboring rare-earth sites. Also in similar LPE-grown lattice-matched KGd$_x$Lu$_{1-x}$Y$_{1−x−y}$Yb$_{1−y}$(WO$_4$)$_2$ layers, for which concentration quenching of the Yb$^{3+}$ lifetime is usually expected in other host materials already for $\sim 10$ at.-% of Yb$^{3+}$, only a rather weak quenching was observed up to 57 at.-% [26].

### Table 2. Parameters used in the determination of ESA cross sections

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$E[N_{a}]$</th>
<th>$E[N_{p}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma$</td>
<td>0.998</td>
<td>0.9955</td>
</tr>
<tr>
<td>$N_2$ [cm$^{-3}$]</td>
<td>0.0465 $N_d$</td>
<td>0.214 $N_d$</td>
</tr>
<tr>
<td>$N_r$ [cm$^{-3}$]</td>
<td>0.352 $N_d$</td>
<td>0.726 $N_d$</td>
</tr>
</tbody>
</table>
Fig. 4. Experimental luminescence-decay curves (continuous lines) at 1535 nm and theoretical decay curves (dashed lines) simultaneously fitted for all decay curves. The incident pump power at $\lambda_p = 1480$ nm was (a) 25 mW, (b) 45 mW, (c) 108 mW, and (d) 148 mW. (e) Exponential intrinsic lifetime of the $^4I_{13/2}$ level versus Er$^{3+}$ concentration. (f) Macroscopic ETU parameter $W_{ETU,1}$ for different doping concentrations. Data points represent the coefficients for the doping concentrations of the studied samples. The dotted line is calculated from Eq. (5).

The luminescence-decay curves of Fig. 4(a)–4(d) were investigated using the analysis by Agazzi et al. [10]. The modified Zubenko equation [10,41]

$$N_1(t) = \frac{N_1(0)e^{-t/\tau_1}}{1 + N_1(0)\beta\pi^2\sqrt{\frac{C_{DA}}{\tau_0}}\frac{\tau_1}{\tau_0} \left\{ \sqrt{1 + \frac{\tau_1}{\tau_0}} \text{erf}\left(\sqrt{\frac{t}{\tau_0}}\right) - e^{-t/\tau_1}\text{erf}\left(\sqrt{\frac{t}{\tau_1}}\right) \right\}} \tag{4}$$

describes the excitation density $N_1$, to which the luminescence intensity is proportional, as a function of time $t$. In the original form of Eq. (4) [41], $\beta = 2$, which can be a reasonable approximation for materials where a large fraction of the $^4I_{13/2}$ population density decays by luminescence to the ground state. In the modified version of Eq. (4) used in the present
investigation, $\beta = 1$ is chosen, thereby assuming that the upconverted ion quickly relaxes back to the $^4I_{13/2}$ level [10], which is a reasonable approximation for high-phonon oxides. A detailed analysis considering measured luminescence lifetimes and Judd-Ofelt data for the radiative lifetimes would provide $\beta = 1 + \beta_{20} = 1.055$ for our material (see Section 4), close to our approximation of $\beta = 1$. The migration mean time $\tau_0$ and migration micro-parameter $C_{DD}$ are related by $1/\tau_0 = C_{DD}N_d^2$ [10,42], whereas the donor-acceptor micro-parameter $C_{DA}$ quantifies the ETU process. The number of donors equals the number of active ions. $N_1(0)$ is the average excitation in the geometrical cross section of the active layer where the pump intensity is $> I_0(z = 0)e^{-2}$. This value was calculated for a thin (~100 $\mu$m) longitudinal slab at the beginning of the waveguide. By an iterative estimation of $N_1(0)$ from a three-level ($^4I_{15/2}, ^4I_{13/2}, \text{and } ^4I_{11/2}$) rate-equation system [10] considering the waveguide characteristics (Table 1) and a simultaneous least-squares fit of Eq. (4) to the 20 luminescence-decay curves of Fig. 4(a)–4(d), micro-parameters of $C_{DD,1} = 5.43 \times 10^{-39}$ cm$^6$/s for energy migration ($^4I_{13/2}, ^4I_{15/2} \rightarrow ^4I_{15/2}, ^4I_{13/2}$) and $C_{DA,1} = 4.94 \times 10^{-40}$ cm$^6$/s for ETU ($^4I_{13/2}, ^4I_{13/2} \rightarrow ^4I_{15/2}, ^4I_{9/2}$) were extracted. The macroscopic ETU parameter [10] then amounts to

$$W_{ETU} = \frac{\pi^2}{2} \sqrt{C_{DD}C_{DA}}N_d = C_{ETU}N_d,$$

resulting in the concentration-independent micro-parameter of the ETU process having a value of $C_{ETU,1} = 5.39 \times 10^{-39}$ cm$^6$/s and $W_{ETU,1}$ as displayed in Fig. 4(f).

In Al$_2$O$_3$:Er$^{3+}$, the concentration-independent micro-parameter of the ETU process has a value of $C_{ETU,1} = 2.5 \times 10^{-39}$ cm$^6$/s [10], which is less than half the value we find in KGLW:Er$^{3+}$. This result demonstrates that the large transition cross sections in KGLW:Er$^{3+}$ over-compensate the large distance between neighboring rare-earth ions. Using the overlap integral of the Förster-Dexter theory [43] for energy migration and ETU,

$$C_{DD} = \frac{6c}{(2\pi)^4 n^2} \int \sigma_e(\lambda)\sigma_a(\lambda) d\lambda,$$

$$C_{DA} = \frac{6c}{(2\pi)^4 n^2} \int \sigma_e(\lambda)\sigma_{ESA}(\lambda) d\lambda,$$

respectively, and apply Eq. (6) to the spectral overlap between the emission and absorption cross sections on the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition for $E|N_m$, see Fig. 2a and 2b, respectively, a value of $C_{DD,1} = 5.91 \times 10^{-39}$ cm$^6$/s is calculated for energy migration ($^4I_{13/2}, ^4I_{15/2} \rightarrow ^4I_{15/2}, ^4I_{13/2}$), which is only slightly higher than the value of $C_{DD,1} = 5.43 \times 10^{-39}$ cm$^6$/s extracted from the luminescence-decay curves. In contrast, there is no direct spectral overlap between the emission $^4I_{13/2} \rightarrow ^4I_{15/2}$ at 1470–1630 nm, see Fig. 2(a), and the ESA $^4I_{13/2} \rightarrow ^4I_{9/2}$ which appears only at wavelengths longer than 1650 nm in KY(WO$_4$)$_2$ [3]. Consequently, the value of $C_{DA,1}$ calculated from the overlap integral of Eq. (7) for ETU ($^4I_{13/2}, ^4I_{13/2} \rightarrow ^4I_{15/2}, ^4I_{9/2}$) is several orders of magnitude smaller than the value of $C_{DD,1}$ calculated from the overlap integral of Eq. (6) for energy migration ($^4I_{13/2}, ^4I_{15/2} \rightarrow ^4I_{15/2}, ^4I_{13/2}$) and the macroscopic parameter $W_{ETU,1}$ which is subsequently calculated from Eq. (5) has an extremely low value. Nevertheless, ETU 1 is a strong, albeit phonon-assisted process. However, it cannot be quantified by the overlap integral of the Förster-Dexter theory but can only be described correctly by a theory that takes into account assistance by a phonon to bridge the energy gap between the two transitions involved in the ETU process.

3.2. Luminescence decay and energy-transfer upconversion from $^4I_{11/2}$

Luminescence decay on the $^4I_{11/2} \rightarrow ^4I_{15/2}$ transition was measured at 1010 nm after direct excitation into the $^4I_{11/2}$ level at 976 nm. Luminescence decay on the $^4S_{3/2} \rightarrow ^4I_{15/2}$ transition
was measured at 545 nm after GSA ($^4I_{15/2} \rightarrow ^4I_{11/2}$) and subsequent upconversion by ESA ($^4I_{11/2} \rightarrow ^4F_{7/2}$) and ETU$_2$ ($^4I_{11/2}, ^4I_{15/2} \rightarrow ^4I_{15/2}, ^4F_{7/2}$), from where non-radiative multiphonon relaxation populates the thermally coupled $^4H_{11/2}$ and $^4S_{3/2}$ levels, see Fig. 1. Decay curves measured in the lowest-doped sample (sample I) are presented in Fig. 5(a) and 5(b). An exponential decay is observed in both cases. The exponential fits suggest $^4I_{11/2}$ and $^4S_{3/2}$ intrinsic lifetimes of $\tau_2 = 135 \pm 8$ $\mu$s and $\tau_5 = 25 \pm 2$ $\mu$s, respectively.

![Fig. 5. Luminescence decay curves recorded at (a) 1010 nm on the transition $^4I_{11/2} \rightarrow ^4I_{15/2}$ and (b) 550 nm on the transition $^4S_{3/2} \rightarrow ^4I_{15/2}$ after excitation of sample I with a laser diode operating at 976 nm. Experimental values (data points) and exponential least-squares fit (red line).](image)

The luminescence decay curves from the $^4I_{11/2}$ level in the higher-doped samples exhibit a more complex temporal dynamics. Figure 6 displays the curves for the highest doped sample (sample V) at different pump powers. For low pump power, the first temporal component exhibits a decay time close to the intrinsic lifetime $\tau_2$ of the emitting level, whereas the second component arises, because the ETU$_1$ process re-populates the $^4I_{11/2}$ level (Fig. 1), and has a decay time similar to half the decay time of the $^4I_{13/2}$ level in which the ETU process originates [37,44]. The first component of the $^4I_{11/2}$ decay becomes faster with increasing pump power because of the ETU process ETU$_2$ ($^4I_{11/2}, ^4I_{15/2} \rightarrow ^4I_{15/2}, ^4F_{7/2}$). Assuming that after short-pulse excitation ions upconverted from $^4I_{13/2}$ by the process ETU$_1$ to $^4I_{9/2}$ relax to $^4I_{11/2}$ via multiphonon relaxation, and ions upconverted from $^4I_{11/2}$ by the process ETU$_2$ return to $^4I_{11/2}$ via multiphonon relaxation, in a simplified way the population densities $N_2(t)$ and $N_1(t)$ can be approximated by the rate equations [37]

$$\frac{dN_2}{dt} = W_{ETU,1}N_2^2 - \frac{1}{\tau_2}N_2 - W_{ETU,2}N_2^2,$$

(8)

$$\frac{dN_1}{dt} = \beta_{21}N_2 - \frac{1}{\tau_1}N_1 - 2W_{ETU,1}N_1^2.$$

(9)

From the fits to the luminescence decay curves of Fig. 6, we estimate the concentration-independent macroscopic ETU parameter as $C_{ETU,2} = 9.81 \times 10^{-39}$ cm$^3$/s. This value is a rather rough estimation, because Eqs. (8) and (9) assume infinitely fast relaxation of the energy upconverted by the ETU$_2$ process back to the $^4I_{11/2}$ level and neglect the finite migration time.

Investigations in amorphous Al$_2$O$_3$:Er$^{3+}$ [37] have shown that the microscopic parameters of migration within the $^4I_{11/2}$ level determined from the spectral-overlap integral of the resonant transitions ($^4I_{11/2} \rightarrow ^4I_{15/2}, ^4I_{15/2} \rightarrow ^4I_{11/2}$) and of ETU determined from the spectral-overlap integral of the resonant transitions ($^4I_{11/2} \rightarrow ^4I_{15/2}, ^4I_{11/2} \rightarrow ^4F_{7/2}$) provide similar results for the concentration-independent macroscopic ETU parameter $C_{ETU,2}$ as the evaluation from luminescence-decay curves from the $^4I_{11/2}$ level. Thanks to the measured ESA cross sections of
Fig. 6. Luminescence decay curves recorded at 1010 nm on the transition $^{4}I_{11/2} \rightarrow ^{4}I_{15/2}$ after excitation of sample V with a laser diode operating at 976 nm for five different incident pump powers. Experimental values (data points) and exponential least-squares fit (red line).

Fig. 3(c) and 3(d), it is possible to calculate the strength of the resonant ETU$_2$ process using the ESA, GSA, and emission cross sections from Figs. 3(c) and 3(d), 2(c), and 2(d), respectively. With Eq. (6), the microscopic parameters of migration and ETU are determined as $C_{DD,2} = 2.98 \times 10^{-39}$ cm$^6$/s and $C_{DA,2} = 2.98 \times 10^{-39}$ cm$^6$/s, respectively, which coincidentally are the same until the third digit. With these $C_{DD,2}$ and $C_{DA,2}$ values and Eq. (5), the concentration-independent macroscopic ETU parameter is then determined as $C_{ETU,2} = 9.81 \times 10^{-39}$ cm$^6$/s.

4. Influence of ETU and ESA on optical gain at 1.5 µm

By use of the obtained spectroscopic parameters, we investigate the influence of pump ESA and ETU$_1$ on the achievable optical gain. The channel-waveguide amplifier model is based on a set of rate equations describing the excitation densities of $^{4}I_{15/2}$, $^{4}I_{13/2}$, $^{4}I_{11/2}$, and $^{4}S_{3/2}$ energy levels, accounting for the relevant processes that populate/depopulate these levels:

$$\frac{dN_5}{dt} = R_{ESA} - \frac{1}{\tau_5} N_5,$$

$$\frac{dN_2}{dt} = R_P + W_{ETU,1}N_1^2 + \frac{\beta_{S2}}{\tau_5} N_5 - \frac{1}{\tau_2} N_2 - R_{ESA},$$

$$\frac{dN_1}{dt} = \frac{\beta_{S1}}{\tau_2} N_2 + \frac{\beta_{S2}}{\tau_5} N_5 - R_s - \frac{1}{\tau_1} N_1 - 2W_{ETU,1}N_1^2,$$

$$N_d = N_0 + N_1 + N_2 + N_5.$$

The pump, ESA, and signal-amplification rate are given by

$$R_P = \frac{\lambda_P}{hc} I_P(\sigma_{a,P}N_0 - \sigma_{e,P}N_2),$$

$$R_{ESA} = \frac{\lambda_P}{hc} I_P \sigma_{ESA} N_2,$$

$$R_S = \frac{\lambda_S}{hc} I_S (\sigma_{e,S} N_1 - \sigma_{a,S} N_0).$$

The evolution of pump power $P_P$ and signal power $P_S$ within the channel waveguide is considered as

$$\frac{dP_P}{dz} = P_P \left[ \int_{\lambda_{Ee}} \Phi_P(\sigma_{a,P}N_2 - \sigma_{e,P}N_0 - \sigma_{ESA}N_2)dx dy - \alpha_{\text{loss,P}} \right].$$
The results are shown in Fig. 7. Figure 7(a) displays the simulated internal net gain per unit length as a function of dopant concentration. For an optimum dopant concentration of \(4 \times 10^{20}\) cm\(^{-3}\) (= 6.3at.\%), a maximum gain of \(-15\) dB/cm is calculated, slightly depending on the assumed value of the propagation loss. The simulation overestimates the experimentally achieved gain of \(-12\) dB/cm [47] by approximately 25\%, which may be due to (i) a non-negligible influence of the ETU\(_2\) and CR processes, (ii) errors in the approximation of the relevant parameter values, and most likely (iii) heating of the waveguide when pumping in the experiment, which leads to a temperature increase and, consequently, a reduction of transitions cross sections. This reduction, which leads to line broadening, is a fundamental process in rare-earth ions. It also impacts the
performance of Yb$^{3+}$-doped waveguide amplifiers [48,49], which typically generate less heat, temperature increase, and a consequent reduction of transition cross sections than comparable Er$^{3+}$-doped devices. Nevertheless, the optimum dopant concentration is rather well predicted. Figure 7(b) indicates the influence of the processes ETU$_1$ and ESA as a function of dopant concentration. The strongest detrimental effect on gain is caused by ETU$_1$ which substantially reduces the excitation density of the $^4I_{13/2}$ level with increasing doping concentration. Although it was predicted that ETU could strongly affect the gain at $\sim$1.5 $\mu$m in Er$^{3+}$, Yb$^{3+}$-doped KY(WO$_4$)$_2$, only rough estimations on the magnitude of the macroscopic ETU parameter were reported [14]. Pump ESA from the $^4I_{11/2}$ pump level has a significantly smaller influence, partly because the pump wavelength has been chosen carefully to diminish the influence of ESA on the achievable gain. At shorter pump wavelengths the influence of pump ESA increases. ETU$_2$ from the $^4I_{11/2}$ pump level and the CR process from the $^4I_{11/2}$ level have only a small influence.

5. Summary

Spectroscopic investigations of a set of KGLW:Er$^{3+}$ channel waveguide samples with five different Er$^{3+}$ concentrations ranging from 0.45–6.36 $\times$ 10$^{20}$ cm$^{-3}$ have been performed. Investigation of the temporal dynamics of luminescence decay from the $^4I_{13/2}$ and $^4I_{11/2}$ levels has provided probabilities of the energy-transfer processes ETU$_1$ ($^4I_{13/2}$, $^4I_{11/2}$) $\rightarrow$ ($^4I_{15/2}$, $^4I_{9/2}$) and ETU$_2$ ($^4I_{11/2}$, $^4I_{11/2}$) $\rightarrow$ ($^4I_{15/2}$, $^4F_{7/2}$). The micro-parameters $C_{DD,1}$ = 5.43 $\times$ 10$^{-39}$ cm$^6$/s for migration and $C_{DA,1}$ = 4.94 $\times$ 10$^{-40}$ cm$^7$/s for upconversion from $^4I_{13/2}$ were extracted by use of Zuben’ko’s model. These values are rather high, because the reduction of ETU probability by the long interionic distances between neighboring active ions is over-compensated by the large transition cross sections in these double tungstates. The concentration-independent macro-parameters are $C_{ETU,1}$ = 5.39 $\times$ 10$^{-39}$ cm$^6$/s and $C_{ETU,2}$ = 9.81 $\times$ 10$^{-39}$ cm$^6$/s. The concentration-dependent quenching of the $^4I_{13/2}$ intrinsic lifetime observed in other Er$^{3+}$-doped materials was found to be absent in KGLW:Er$^{3+}$ for doping concentrations up to 10 at.%. A pump-probe study of ESA for the tentative pump wavelengths between 960 and 990 nm has been performed. The pump wavelength of 984.5 nm $\langle E||N_m \rangle$ is predicted to be most suitable for the amplification of $\sim$1.5 $\mu$m signals. In a rate-equation simulation, ETU$_1$ from the $^4I_{13/2}$ level appears to have the strongest detrimental effect on optical gain at 1535 nm in these Er$^{3+}$-doped channel waveguides. Measured values of internal net gain are over-estimated by $\sim$25%, partly because pump heating and the temperature dependence of transition cross sections are neglected in the simulation. The optimum dopant concentration for achieving the highest gain is confirmed in the simulation.

Funding

Stichting voor de Technische Wetenschappen (11689); H2020 European Research Council (648978).

Acknowledgment

The other authors of this paper dedicate this work to our colleague and co-author Yean-Sheng Yong who, sadly, deceased shortly after submission of this paper.

References


