

High-temperature optical properties of sensitized Er³⁺ in Si-rich SiO₂ – implications for gain performance

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ABSTRACT

The high-temperature photoluminescence of Er-doped Si-rich SiO₂ with and without silicon nanocrystals is studied at sample temperatures in the range 20–200 °C. The optical properties of Er-doped Si-rich SiO₂ with and without silicon nanocrystals are shown to exhibit a similar temperature dependence. Based on the measured photoluminescence intensities and lifetimes it is predicted that an increase of the sample temperature from 20 to 200 °C results in a decrease of the maximum optical gain at 1535 nm by a factor of ~1.8 and ~1.6 for samples with and without silicon nanocrystals, respectively. Implementation of this material in silicon photonics requires stable operation at typical processor case temperatures up to 80–90 °C. It is demonstrated that increasing the temperature from room temperature to 90 °C leads to a predicted maximum optical gain reduction of ~1.26 for both materials. In addition, the predicted erbium related optical gain at significant inversion levels in samples processed at low temperature (600 °C) is a factor ~9 higher than for samples processed at high temperature (1060 °C). These findings demonstrate that relatively thermally stable gain performance of the Er-doped Si-rich SiO₂ up to typical processor operating temperatures is possible and indicate that low-temperature-processed erbium-doped silicon-rich SiO₂ is a technologically viable gain medium for use in silicon photonics.

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1. Introduction

The implementation of CMOS-compatible photonic devices into integrated circuits is one of the main avenues for increasing the performance of computers [1–4]. In recent years, a number of silicon compatible photonic devices have been demonstrated [5–10] however the realization of a cost efficient on-chip silicon compatible light source remains a challenge. Silicon compatible sources being considered so far are silicon Raman lasers [11], hybrid evanescent lasers [12], devices based on germanium-on-silicon [13], silicon nanocrystal doped SiO₂ [14–19], silicon nanocrystal doped silicon nitride [20], erbium-doped Si-rich silicon nitride [21], erbium-doped silicon-rich SiO₂ [22–29]. In Er-doped Si-rich SiO₂, Er³⁺ ions provide emission at 1.54 μm due to an intra-4f shell transition of Er³⁺ from its first excited state (⁴I_{13/2}) to the ground state (⁴I_{15/2}). While the Er³⁺ emission can be quite efficient, efficient excitation of Er³⁺ in SiO₂ is difficult to achieve due to intrinsically

narrow absorption lines with low absorption cross sections. To overcome this challenge, a significant amount of research has focused on sensitization of Er³⁺ ions by adding dopants with strong broadband absorption and efficient energy transfer to Er³⁺ ions. The observation of strong broadband sensitization of Er³⁺ in silicon nanocrystal doped SiO₂ led to the conclusion that Si nanocrystals are promising Er³⁺ sensitizers. Silicon sensitization of Er³⁺ in Si-rich SiO₂ has been shown to result in a dramatic increase (~3–6 orders of magnitude) of the effective Er³⁺ absorption cross section as compared to that in stoichiometric SiO₂ films [29–34]. Optical gain [25,35] and signal enhancement [36] at 1535 nm were demonstrated in waveguides based on this type of material. It should be noted that in these studies gain was achieved in composites containing rather low concentrations of excess silicon. While the presence of excess silicon has been repeatedly shown to produce significant Er³⁺ sensitization, the formation of extended Si nanocrystals is known to introduce confined carrier absorption [26,37,38] and scattering [39], both of which adversely affect optical gain. It should also be noted that annealing of Er-doped Si-rich SiO₂ at temperatures above 1000 °C necessary for silicon nanocrystal formation, results in a significant decrease of the density of optically active Er³⁺ ions [30], resulting in a significant reduction of the maximum achievable optical gain.

Recently we demonstrated that while Si nanocrystals do act as sensitizers, the dominant cause for Er³⁺ sensitization in Si-rich SiO₂

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is the presence of silicon-excess related luminescence centers [30,31] capable of sensitizing Er even in as-deposited Si-rich SiO₂ [40]. Our results imply that the use of Er-doped Si-rich SiO₂ annealed at low temperatures could provide a significant improvement in the maximum optical gain at 1.54 μm compared to Er³⁺ doped materials containing silicon nanocrystals, due to (i) a higher density of optically active Er³⁺ ions in this material, and (ii) the absence of Si nanocrystal related confined carrier absorption and optical scattering.

In order to evaluate the viability of Er-doped Si-rich SiO₂ as a gain medium in silicon photonics, it is important to consider its performance under realistic operating conditions. One possible approach for combining optical data transfer and processing with present day silicon technology is by incorporating photonic elements on top of the electronic device layer. This approach requires that any on-chip optical components provide reliable and stable performance under exposure to significant temperature variations ranging from room temperature up to maximum processor case temperatures of ~80–90 °C. While several studies have focused on the optical properties of Er-doped Si-rich SiO₂ at cryogenic temperatures [23,41–43], little is known about the performance of this material at elevated temperatures. In the present study, several key optical properties directly affecting optical gain are studied in the temperature range 20–200 °C. Based on the measured temperature dependence of the optical parameters the behavior of the optical gain in the samples with and without silicon nanocrystals is modeled. The results suggest that devices based on Er-doped Si-rich SiO₂ are capable of providing thermally stable gain performance under high-temperature operating conditions.

2. Experimental techniques

Er-doped Si-rich SiO₂ films containing 12 at.% of excess Si and 0.63 at.% of Er were deposited onto a P-doped Si substrate ((1 0 0), resistivity 3–7 Ω cm) by magnetron co-sputtering from Si, SiO₂ and SiO₂:Er₂O₃ targets in a multi-gun sputtering system (AJA International, Inc., ATC-2200V). Samples with different microstructure were prepared by annealing the as-deposited films for 100 s in flowing N₂ (3 SLPM) at 600 °C or 1060 °C (labeled LTA and HTA, respectively, for Low and High Temperature Anneal) using a rapid thermal processor (Modular Process Technology Corp., RTP-600S). No Si aggregates could be detected in transmission electron microscopy (TEM) measurements on LTA samples, while TEM measurements on samples annealed at $T \geq 1000$ °C (e.g. HTA sample) clearly showed the presence of NCs [30]. The processed samples were attached with silver paint to the cold finger of a closed-loop He cryostat designed for operation in the temperature range 10–800 K (ARS DE-202AE-T). Photoluminescence (PL) measurements were performed in the temperature range 20–200 °C at a pressure of $\sim 5 \times 10^{-7}$ mbar. Note that these temperatures lie well below the applied annealing temperatures. No permanent change of the optical properties was observed after extended exposure of the samples to temperatures up to 200 °C. The maximum sample temperature was limited by the ability to measure the Er³⁺ emission at 1535 nm on top of the thermal radiation background without saturating the near-infrared detectors. Photoluminescence measurements were taken under 351 nm excitation using a continuous wave (cw) Kr-ion laser (Spectra-Physics, Beam-Lok 2060) (pump irradiance 2.56 W/cm²). A single monochromator (Acton, SpectraPro 2300i) was used to disperse the collected signal. A thermo-electrically-cooled charge-coupled device camera (Andor, DU401-BR-DD) was used to record the spectra in the region 450–1050 nm. A liquid-nitrogen-cooled Ge-detector (Applied Detector Corp., 403S) in combination with standard lock-in techniques was used for spectral measurements in the region 900–

1750 nm. The obtained spectra were corrected for the system response and concatenated in the region 1030–1050 nm. The time-dependence of the photoluminescence intensity was measured by modulating the cw pump beam using an acousto-optic modulator (NEOS Technologies, 38210-6AS). Photoluminescence traces were recorded using a near-infrared photomultiplier tube (Hamamatsu R5509-73), in combination with a multi-channel scaler (Stanford Research Systems, SR430). During all measurements the samples were continuously scanned against the laser beam to prevent possible light-induced changes to their optical properties upon exposure to UV light [44]. The spectral resolution in all photoluminescence measurements was ~15 nm. The system time resolution of the lifetime measurements was 40 ns for luminescence center and nanocrystal related emission, 320 ns for the Er emission at 1535 nm in LTA sample and 1.28 μs for the Er emission at 1535 nm in HTA sample.

3. Results and discussion

Fig. 1 shows the photoluminescence (PL) spectra of the LTA sample taken in the temperature range 20–200 °C. The PL spectra of the LTA sample are characterized by three emission bands: a broad rapidly decaying (lifetime < 40 ns) band peaking around 550 nm corresponding to the emission from Si-excess related luminescence centers (LC) and two relatively narrow slowly decaying emission bands at 981 and 1535 nm corresponding to the transitions from the second (⁴I_{11/2}) and first (⁴I_{13/2}) excited states, respectively, to the ground (⁴I_{15/2}) state of Er³⁺. Fig. 2 shows the corresponding PL spectra of the HTA samples plotted on the same relative intensity scale as Fig. 1. The spectrum of HTA sample is characterized by four emission bands: a fast (lifetime < 40 ns) broad LC related band peaking around ~550 nm, a rather slow (lifetime ~2 μs at room temperature) broad Si nanocrystal (NC) related band peaking at ~745 nm, and the two Er³⁺ related bands at 981 and 1535 nm also observed in the LTA sample. The insets of Figs. 1 and 2 show the spectra of the Er³⁺ transition from the first excited to the ground state, normalized to their peak value at 1535 nm for the LTA and HTA samples, respectively. Relatively small changes in spectral shape are observed in the temperature range 20–200 °C, suggesting that optical cross sections and

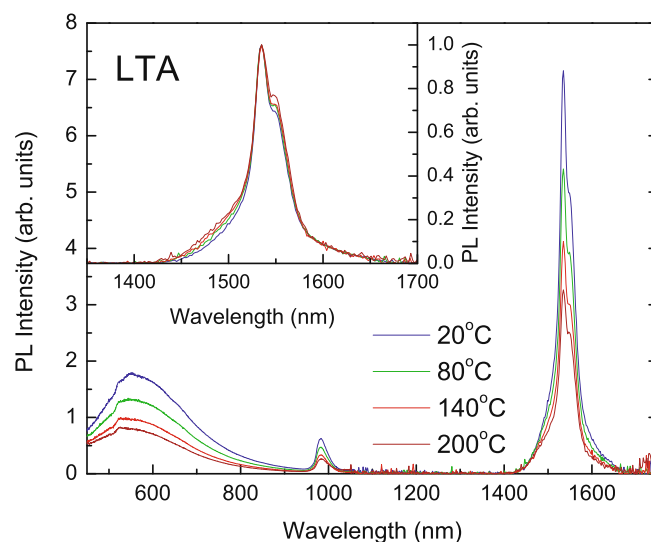


Fig. 1. (Color online) Photoluminescence spectra of Er-doped Si-rich SiO₂ annealed at 600 °C (LTA) measured in the temperature range 20–200 °C. Inset: normalized emission from the first excited state of Er³⁺ (⁴I_{13/2}).

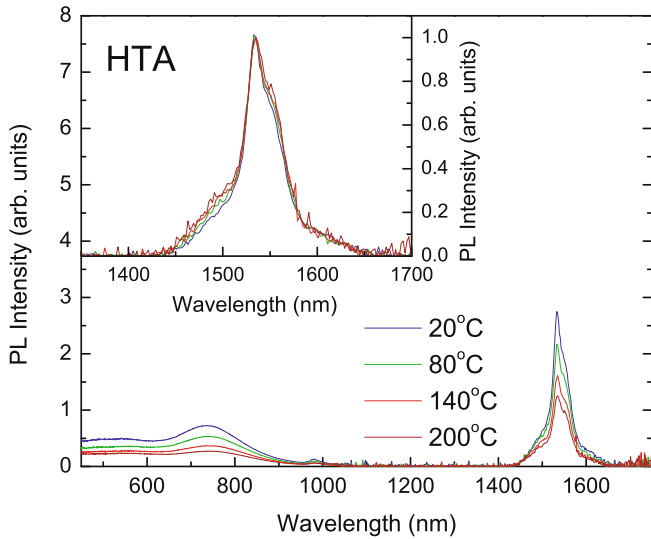


Fig. 2. (Color online) Photoluminescence spectra of Er-doped Si-rich SiO₂ annealed at 1060 °C (HTA) measured in the temperature range 20–200 °C. The values of the PL intensity may be compared to the intensities in Fig. 1. Inset: normalized emission from the first excited state of Er³⁺ (⁴I_{13/2}).

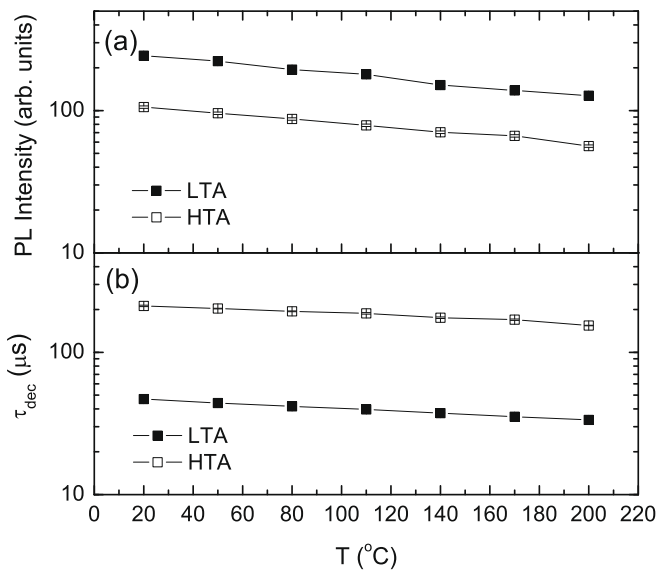


Fig. 3. Temperature dependence of (a) integrated photoluminescence intensity I_{Er} corresponding to the ⁴I_{13/2} → ⁴I_{15/2} transition of Er³⁺ and (b) decay time τ_{dec} of the first excited state of Er³⁺ (⁴I_{13/2}) in samples annealed at low (LTA) and high (HTA) temperature.

therefore the spectral dependence of the gain will be largely temperature independent across the Er³⁺ emission band.

Fig. 3a shows the PL intensity corresponding to the transition of Er³⁺ from the first excited (⁴I_{13/2}) state to the ground (⁴I_{15/2}) state in the LTA and HTA sample, integrated in the region 1350–1700 nm (I_{Er}). For both LTA and HTA samples the integrated intensity is observed to decrease by a factor of ~ 1.92 as the sample temperature is increased from 20 to 200 °C. This change was found to be reversible, i.e. upon returning the samples to room temperature, the original room-temperature PL intensity was recovered. The PL decay traces of the Er-related emission at 1535 nm (not shown) in LTA and HTA samples exhibited stretched exponential behavior. The $1/e$ decay times τ_{dec} were obtained by fitting the measured decay traces with the function $I(t) = I_0(t) \exp(-(t/\tau_{dec})^\beta)$ where β is the dispersion factor of the stretched exponent. Fig. 3b shows the

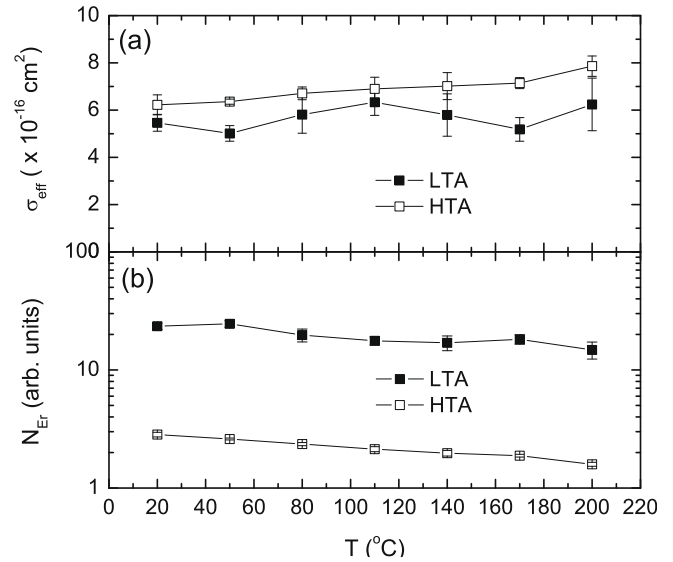


Fig. 4. Temperature dependence of the (a) effective absorption cross section of the first excited state of Er³⁺ (σ_{eff}) and (b) the density of optically active sensitized Er³⁺ ions in samples annealed at low (LTA) and high (HTA) temperature.

behavior of the Er³⁺ decay times τ_{dec} at 1535 nm in LTA and HTA samples. In the considered temperature range τ_{dec} is found to decrease by a factor of ~ 1.4 for both LTA and HTA samples. The dispersion factors for both samples were found to be approximately temperature independent and equal to $\beta_{LTA} = 0.73 \pm 0.1$ and $\beta_{HTA} = 0.76 \pm 0.1$.

Fig. 4a shows the effective absorption cross section of the first excited state of Er³⁺ in LTA and HTA samples, as found from the rise time and decay time measurements (not shown) using the well known relation $\sigma_{eff} \varphi = 1/\tau_{rise} - 1/\tau_{dec}$, where σ_{eff} is the effective absorption cross section of the first excited state of Er³⁺, φ is the excitation photon flux, τ_{rise} is the rise time, and τ_{dec} is the decay time of the Er³⁺ first excited state. For both LTA and HTA samples the Er³⁺ effective absorption cross section is found to be temperature independent within the experimental error. Based on the experimentally determined values of the integrated emission intensity (I_{Er}), the decay time (τ_{dec}), and the Er³⁺ effective absorption cross section at 1535 nm (σ_{eff}) the density of optically active sensitized Er³⁺ ions N_{Er} in LTA and HTA samples can be found using the expression $N_{Er,sens} \propto I_{Er} \times (1 + 1/(\sigma_{eff} \varphi \tau_{dec}))$ [30,31]. Fig. 4b shows the thus obtained temperature dependent density of sensitized optically active Er³⁺ ions. N_{Er} is found to gradually decrease by a factor of 1.6 and 1.8 for LTA and HTA samples, respectively, as the sample temperature is increased from 20 to 200 °C.

Based on the temperature dependence of the decay time (τ_{dec}), the Er³⁺ effective absorption cross section (σ_{eff}), and the density of optically active sensitized Er³⁺ ions (N_{Er}) the effect of heating on the optical gain at 1535 nm in LTA and HTA samples can be estimated assuming a temperature independent peak emission cross section of Er³⁺, as suggested by the relatively temperature independent shape of Er³⁺ emission spectra, and excluding cooperative upconversion and excited state absorption. Under these assumptions the maximum low-signal gain coefficient can be found from the relation:

$$\gamma = \sigma_{Er,peak} \Delta N = \sigma_{Er,peak} N_{Er} \frac{\sigma_{eff} \varphi \tau_{dec} - 1}{\sigma_{eff} \varphi \tau_{dec} + 1} \quad (1)$$

where γ is the optical gain coefficient at 1535 nm, $\sigma_{Er,peak}$ is the peak emission cross section of the Er³⁺ ⁴I_{13/2} → ⁴I_{15/2} transition, assumed to be equal to the absorption cross section of the

$4I_{15/2} \rightarrow 4I_{13/2}$ transition at this same wavelength [45], and ϕ is the pump photon flux. Note that Eq. (1) does not take into account the possible effects of cooperative upconversion and excited state absorption. These effects will likely occur at high pump powers [46,47], resulting in, respectively, an increase in the pump power required to achieve Er-related gain, and a decrease in the value of the maximum gain. However, the required parameters to quantitatively predict these effects are not available for the present sample composition and processing conditions, and their determination is outside the scope of the current study.

Fig. 5a and b show the calculated photon flux dependence of γ based on Eq. (1) for LTA and HTA samples, respectively, obtained for different sample temperatures based on the experimentally determined temperature dependent values of the lifetime, Er^{3+} effective absorption cross section, and the density of sensitized Er^{3+} ions. It should be noted that these curves do not take into account ground state absorption from any unsensitized Er^{3+} ions that may be present in the film. Several key differences in gain performance of the LTA and HTA sample can be observed. First, the Er^{3+} related absorption at low pump flux and maximum achievable gain at high population inversion levels ($\sigma_{eff}\phi\tau_{dec} \gg 1$) occurring for high pump flux are a factor ~ 9 larger for the LTA sample than for the HTA sample due to the higher N_{Er} at all temperatures for the former. Second, the threshold pump flux is higher for the LTA sample mainly due to a significantly (by a factor of ~ 4.6) lower Er^{3+} lifetime compared to the HTA sample. Third, as the temperature is increased the maximum gain under significant population inversion reduces by a factor of ~ 1.6 and ~ 1.8 in the temperature region 20–200 °C for LTA and HTA samples, respectively. Note that at typical processor operating temperatures of ~ 80 – 90 °C the maximum gain is only a factor ~ 1.26 less than at room temperature for both samples. This indicates that a relatively thermally stable gain performance under significant inversion can be achieved using both low (600 °C, LTA) and high (1060 °C, HTA) temperature annealed Er-doped Si-rich SiO_2 as the gain medium under real-world operating conditions of silicon integrated circuits. Samples annealed at low temperature (LTA) not containing silicon nanocrystals are expected to provide better gain performance in real on-chip devices as compared with samples annealed at high temperature (HTA). This conclusion is based predominantly on the predicted higher optical gain, a lower ground state absorption resulting from a higher concentration of sensitizers and thus a higher fraction of sensitized Er^{3+} ions, and lower confined carrier absorption as compared

with the samples with silicon nanocrystals. It should be noted that due to the lower Er^{3+} decay time in LTA samples as compared to HTA samples, higher pump powers will be needed to achieve population inversion in LTA samples. Secondly, it is necessary to take into account that while the concentration of Er is the same in both LTA and HTA samples studied here, the density of optically active sensitized Er^{3+} ions is factor ~ 9 higher in the LTA sample as compared to HTA sample. This difference is expected to be even higher for the samples annealed at temperatures above 1060 °C where a steep drop of the density of optically active indirectly excitable Er^{3+} ions occurs [30]. While the predicted optical gain in the current study does not take into account any remaining unsensitized Er^{3+} in the LTA sample, the much larger density of sensitized Er^{3+} ions in the LTA sample compared to the HTA samples suggests that using low annealing temperatures can minimize ground state absorption from unsensitized Er^{3+} ions. The higher concentration of sensitized Er^{3+} in LTA samples, combined with the absence of Si nanocrystal related optical loss contributions such as confined carrier absorption [37,38] and scattering [39] make LTA the most viable candidate for the achievement of high optical gain at 1.54 μm . Based on these considerations low-temperature processed Si-sensitized Er-doped SiO_2 has the potential to become an important optical gain medium with thermally stable performance.

4. Conclusions

In summary, it was shown that in samples without (annealed at 600 °C) and with (annealed at 1060 °C) Si nanocrystals the change of the spectral shape of the Er^{3+} band at 1535 nm is insignificant in the temperature range 20–200 °C, suggesting that optical cross sections throughout the Er^{3+} emission band at 1.54 μm is relatively thermally stable. Based on measurements of the temperature dependent excitation rate, decay time and density of optically active sensitized Er^{3+} ions, it is predicted that in the temperature range 20–90 °C the peak optical gain does not change more than by a factor of ~ 1.26 in the case of LTA and HTA samples at significant inversion levels. This observation proves that relatively stable operation of on-chip photonic devices based on Er-doped Si-rich SiO_2 is possible when the material is exposed to typical processor operating temperatures. While the temperature dependence of the gain is predicted to be similar in the case of LTA and HTA samples, based on the obtained values of the density of optically active sensitized Er^{3+} ions, devices fabricated from low-temperature processed Er-doped Si-rich SiO_2 are expected to demonstrate a higher optical gain at 1535 nm as compared with high-temperature processed material.

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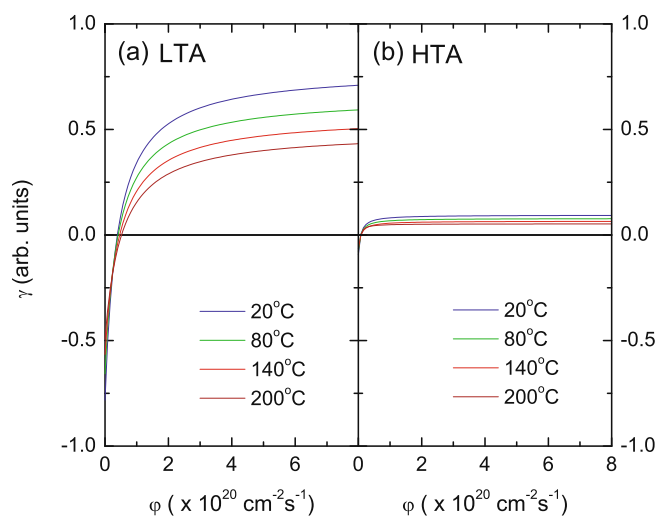


Fig. 5. (Color online) Calculated pump flux dependence of the Er^{3+} related optical gain at 1535 nm for different sample temperatures, in samples annealed at (a) low (LTA) and (b) high (HTA) temperature.

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