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Absence of the enhanced intra-4f transition cross section at 1.5 μm of Er3+ in Si-rich SiO2

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We present measurements of the optical absorption cross section of the 4I15/2→4I13/2 transition at 1.5 μm of Er3+ ions embedded in SiO2 and Si-rich oxide, using cavity ringdown spectroscopy on thin films. The peak absorption cross section for Er3+ embedded in Si-rich oxide (10 at. % excess Si) was found to be (8±2)×10⁻²¹ cm² at 1536 nm, similar to typical values for Er embedded in SiO2. The data imply that the silicon nanoclusters incorporated in Si-rich oxide do not enhance the peak cross section of the Er3+ 4I15/2→4I13/2 transition by 1-2 orders of magnitude, contrary to what has been reported in earlier work. © 2005 American Institute of Physics. [DOI: 10.1063/1.1949720]

Erbium-doped Si-rich oxide (SRO:Er) is an optical gain medium in which Er3+ ions, exhibiting an intra-4f transition at 1.5 μm, are surrounded by silicon nanoclusters that serve as broadband sensitizers.1-3 A sensitizer coupled to an Er3+ ion absorbs light at a cross section several orders of magnitude larger than Er, and then transfers its energy to the Er3+ ion. Sensitizers have the potential to enable optical amplification at 1.5 μm using low-cost light-emitting diodes in top-pumping configuration, instead of a waveguide-coupled laser tuned to an Er transition.

In addition to the sensitization effect, which is of great technological interest, it has been reported that silicon nanoclusters would enhance the absorption and emission cross sections of the 4I15/2→4I13/2 transition at 1.5 μm of Er3+ embedded in SiO2 by 2 orders of magnitude.4,5 This presumed enhancement was derived from absorption spectroscopy in linear waveguides,6 and it has been invoked to explain the high signal enhancement in SRO:Er linear amplifiers.5 The enhanced emission cross section has also been used as an input parameter in a model describing the gain characteristics of SRO:Er amplifiers.6

If true, the cross section enhancement would have a broad impact on Er amplifier and laser technology as it implies an enhancement of the optical gain coefficient by the same 1-2 orders of magnitude. Thus more compact devices would be possible using SRO:Er.

Both reported cross-section enhancements are based on indirect measurements, and a direct determination is still lacking. In this letter, we present measurements of the Er3+ absorption cross section in both SRO:Er and SiO2:Er by cavity ringdown spectroscopy (CRDS) on thin films,7,8 which is an approach that does not require estimates on the overlap between an optical mode and the erbium profile as in earlier work. From the data it is concluded that the earlier reported 1-2 orders of magnitude enhancement of the cross section of the Er3+ 4I15/2→4I13/2 transition at 1.5 μm due to silicon nanoclusters is incorrect.

SRO:Er and SiO2:Er thin films were fabricated by ion implantation and thermal annealing of 2-mm-thick high-purity fused-silica substrates (Heraeus Suprasil® 300). In the first fabrication step, the substrates were implanted with 300- and 150-keV Er ions to a total fluence of 1.2×10¹⁶ at/cm². The Er peak concentration measured by Rutherford backscattering spectrometry was 2.3 at. %. Subsequently, a dual Si implant (73 and 30 keV) was applied to part of the Er-implanted samples to fabricate SRO:Er films. The Si fluences were chosen such that an excess Si concentration of ~10 at. % was achieved over the full Er depth profile. One of the SRO:Er samples was annealed at 1000 °C for 30 min in flowing Ar and at 700 °C for 30 min in forming gas (H2:N2 at 1:9) to optimize the Er3+ 1.5 μm photoluminescence (PL) intensity.9 An SiO2:Er reference sample (i.e., without Si incorporation) was exposed to the same thermal treatment. A second SRO:Er sample was annealed at 1000 °C for 30 min in flowing Ar, comparable to the thermal treatments described in Refs. 4 and 5. PL excitation spectroscopy using different lines of an Ar+ laser1 showed that Er3+ embedded in SRO was mainly excited indirectly, confirming the coupling between Er3+ and silicon nanoclusters in these samples.

The Er3+ absorption spectrum at 1.5 μm was determined by CRDS on thin films, which is a direct and highly sensitive absorption measurement method in which the sample under investigation is placed inside a high quality optical cavity (see the inset of Fig. 1).7,8 The measurements were carried out with the tunable idler output of an optical parametric oscillator (OPO), which is pumped by the third harmonic of a pulsed Nd:YAG laser operating at 30 Hz. The idler output of the OPO was tuned over the wavelength range of 1450–1630 nm with an accuracy of 2 nm, and the resulting pulse had a typical bandwidth of 2 nm, a duration of 5 ns, and a pulse energy of 10 mJ. The pulse was injected into a stable optical cavity (length 0.4 m) formed by two planocon-cave highly reflecting (R>0.9997) mirrors without using mode-matching optics, resulting in an effective spot size of 3 mm. Thus, the total injected pulse energy was less than 3 μJ. The cavity was purged with dry nitrogen to reduce the effects of H2O absorption lines. The temporal decrease in

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light intensity inside the cavity upon pulse injection was detected with a photodiode. Individual transients were sampled using a 12 bit, 100 MHz data acquisition system.

Figure 1 shows cavity ringdown transients detected at 1536 nm for both an SRO:Er sample that was annealed at 800 °C and a bare quartz substrate. After the first microsecond, the transients exhibit a single-exponential behavior of which the rate is determined by the intrinsic cavity loss and the loss generated by the sample placed in the cavity.7 The ringdown time for SRO:Er (3 μs) is significantly shorter than for quartz (8 μs) due to the stronger extinction of the SRO:Er sample. The single-exponential parts of individual transients were fitted with a standard weighted linear regression technique to extract the ringdown time. Reported values of the optical loss per pass, which includes both scattering and absorption contributions, have been corrected for the empty cavity response and were deduced from averages of the ringdown time over 100 laser shots at every wavelength.

Figure 2 shows the optical loss spectra for SRO:Er and SiO2:Er determined in this way. Every optical loss spectrum shows a characteristic Er3+ absorption spectrum superimposed on a background that is decreasing with wavelength, which is attributed to loss processes not related to Er such as scattering. Repeated measurements after sample realignment gave reproducible results for both peak shape and magnitude, as well as for the decreasing background trend, with some variations in background level. We attribute the latter to variations in both alignment and sample quality.

The estimates of the background losses indicated in Fig. 2 (dashed lines) are based on a qualitative comparison with absorption spectra derived by PL spectroscopy using McCumber theory,10,11 as is illustrated in Fig. 3. Figure 3(a) shows a PL spectrum for SRO:Er (annealed at 800 °C) taken at room temperature. The absorption spectral shape derived from this spectrum is shown in Fig. 3(b), together with the corresponding absorption spectrum derived from the optical loss data shown in Fig. 2 by subtraction of the background loss (dashed line). The spectral resemblance between the CRDS-based spectrum and the normalized PL-based spectrum validates the estimate of the background loss.

The important feature shown in Fig. 2 is that, for all three samples, the Er absorption is very similar over the entire wavelength range (within 20%). The Er3+ $^4I_{15/2} \rightarrow ^4I_{13/2}$ absorption cross section is thus found to be similar for SiO2:Er and SRO:Er annealed at either 800 or 1000 °C. Here, it is assumed that all Er ions are in the 3+ valence state, which is justified by the fact that no other valences of Er are reported for a variety of hosts.12 In addition, X-ray absorption spectroscopy of the Er $3d^{10}4f^{11} \rightarrow 3d^{9}4f^{12}$ transitions for Er-doped silicon has shown no evidence of other Er valence states.13

The Er3+ $^4I_{15/2} \rightarrow ^4I_{13/2}$ absorption cross section for SRO:Er can directly be obtained from Fig. 3(b) by dividing the Er absorption by the Er areal density. This procedure yields a peak absorption cross section, which is related to the maximum gain coefficient for SRO:Er amplifiers, of (8.4 ± 2) × 10−21 cm2 at 1536 nm, where the error bar is determined mainly by the uncertainty in the background level of the absorption measured. This result is to typical values obtained for Er embedded in glasses:13 (4.8–7.0) × 10−21 cm2, confirming that silicon nanoclusters do not enhance the cross section of the Er3+ $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition by
been 2 orders of magnitude. Note that the cross section in SRO:Er is expected to be 10%–20% larger than in SiO$_2$:Er due to the higher refractive index of the heterogeneous SRO, but the limited signal-to-noise ratio of the SiO$_2$:Er sample does not allow one to resolve this difference from the performed measurements.

For the determination of the Er$^{3+}$ $^4I_{15/2} \rightarrow ^4I_{13/2}$ absorption cross section, bleaching of the Er$^{3+}$ ions has been neglected. This is legitimate since the effective pump power inside the cavity was adequately low, as can be derived in the following way. Upon pulse injection, the pulse energy in the cavity was lower than 3 $\mu$J. Taking into account the cavity length, ringdown time, and laser spot size, this corresponds to an effective time-integrated photon flux of $6 \times 10^{17}$ photons/cm$^2$ per pulse. This implies that for an Er$^{3+}$ absorption cross section of $8 \times 10^{-21}$ cm$^2$ less than 0.5% of the Er$^{3+}$ population was inverted per pulse. Subsequent pulses can be regarded independently, since the interval between them (33 ms) is much larger than the Er$^{3+}$ lifetime in our samples (~3 ms). The relatively short Er$^{3+}$ lifetime is ascribed to the presence of nonradiative de-excitation channels in the heavily doped matrix. If the cross section had been $2 \times 10^{-19}$ cm$^2$, still only 12% of the Er$^{3+}$ population would have been inverted, and the Er absorption peak would be 20 times larger than observed. This implies that the error in the absorption cross section induced by Er$^{3+}$ bleaching is much smaller than the error induced by the uncertainty in the background level.

Finally, we speculate on the apparent discrepancy between the cross section for SRO:Er derived here, and the much larger values claimed in earlier work. Kik’s tenfold enhanced cross-section claim was based on a calculation that involved an estimated overlap between the optical waveguide mode and the Er profile. Possibly, this overlap was underestimated. As CRDS does not require estimates on modal overlap in order to determine the absorption cross section, the method presented in the current work provides more reliable results. The reported near-hundredfold cross section enhancement derived from gain characteristics of SRO:Er linear amplifiers by Han et al. remains puzzling. In any case, such an enhancement is inconsistent with the Er luminescence lifetime of 8 ms reported in the same paper: according to the Füchtbauer-Ladenburg equation, the reported Er emission cross section of $(2 \pm 0.5) \times 10^{-19}$ cm$^2$ would correspond to a radiative lifetime of ~0.3 ms.

In conclusion, we have measured the optical absorption cross section of the Er$^{3+}$ $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition at 1.5 $\mu$m in both SRO:Er and SiO$_2$:Er by cavity ringdown spectroscopy on thin films, proving that this is a suitable technique for the determination of rare-earth absorption cross sections. The peak absorption cross section for SRO:Er was found to be $(8 \pm 2) \times 10^{-21}$ cm$^2$, which is identical (within 20%) to the measured cross section for SiO$_2$:Er, and which is a typical value for Er embedded in glasses. This result is in disagreement with earlier reported cross sections enhancements for SRO:Er. The conclusion affects modeling of the gain characteristics of SRO:Er materials that use the presumed enhanced cross section as an input parameter.

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