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ScienceDirect
Journal of Luminescence 121 (2006) 193–198
JOURNAL OF LUMINESCENCE
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An analysis of erbium excited state absorption in silicon-rich silica

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Available online 26 September 2006

Abstract

Erbium-doped silicon-rich silica is a material that has generated great interest, as it holds considerable promise for practical silicon-based optical sources. There are numerous reports of sensitization of erbium 1.5 μm luminescence by silicon nanoclusters (Si nc), and it is now accepted that this process increases the effective absorption cross-section of Er ions by three orders of magnitude. There have been reports of optical gain in a planar waveguide pumped at wavelengths away from Er absorption bands. However, no lasing action has yet been achieved. In this paper, we discuss a key interaction—excited state absorption (ESA)—which has been neglected in this material. Typically, Si nc responsible for sensitization have a broad photoluminescence peak centred around 800 nm. Moreover, spectral hole-burning studies suggest that resonant energy transfer occurs from the Si to the Er in this wavelength region. However, this energy transfer route may be problematic, due to an ESA transition known from early work on erbium-doped fibre amplifiers (EDFAs). Excitation around 800 nm causes the Er metastable state to be excited to higher-level states. Such transitions serve no useful purpose, but constitute an energy drain: EDFAs pumped at 800 nm require an order of magnitude more pump power to achieve the same gain as those pumped at 980 nm. We have conducted an analysis of the erbium-doped silicon-rich silica system, incorporating ESA in the model. We compare the results of our analysis with reported experimental data and extract the Si nc–Er excited state energy transfer coefficient, yielding a value of $1 \times 10^{-15}\,\mathrm{cm}^3/\mathrm{s}$. This is comparable to the Si nc–Er ground state transfer coefficient, confirming that ESA is potentially significant.

Keywords: Silicon nanoclusters; Silicon nanocrystals; Erbium; Sensitisation; Excited state absorption; ESA; Optical gain; Optical amplifier

1. Introduction

Silicon nanocluster (Si nc) sensitized Er-doped silica has generated great interest in recent years [1–4], as it harbours considerable promise for practical silicon-based lasers and optical amplifiers. In this material, pump photons are absorbed by the Si nc, which transfer the excitation to nearby erbium ions that emit in the spectral region around 1535 nm. The nature of the excitation transfer mechanism remains a matter of some debate, but it is clear that the presence of erbium ions produces a strong quenching of the luminescence from Si nc, and experiments have demonstrated a strong dependence of the exchange mechanism on the nanocluster–erbium separation [5]. The existence of a strong nanocluster–rare earth interaction is thus well established, and a number of groups are actively pursuing the development of optical amplifiers based on this material. The appeal of the Si nc-sensitized Er system is simple: using Si nc as the sensitizer for rare-earth ions enables one to take advantage of the large optical absorption cross section of silicon, which is several orders of magnitude larger than that of rare earths in silica (SiO_2). Studies have indeed demonstrated an effective cross-section for the excitation of erbium ions via Si nc some 3–4 orders of magnitude greater than that for directly excited erbium ions [6,7]. In addition, the broadband absorption spectrum of Si nc considerably relaxes the wavelength requirements of the pump source, allowing a wider variety of lasers, and even LEDs, to be used. Using LEDs instead of laser diodes as pump sources is extremely attractive, as it would reduce pump costs 10- to 100-fold, extending the scope and accessibility of many more optical applications. There are numerous reports of strong 1550 nm luminescence in this material system, and a small number of reports of gain

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0022-2313/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jlumin.2006.07.025

using both laser and LED pumping [8]. However, no lasing action has been achieved to date, and it has proved difficult to reliably produce material that exhibits gain. Indeed, evidence is accumulating that, for much of the material that has been reported, the fraction of Er ions that can be excited by Si nc is actually very low—of the order of a few percent [9,10]. For example, for the data presented by Fujii et al. [9], the Er photoluminescence (PL) for two samples with the same Er concentration, one of which contained Si nc (silicon-rich silicon dioxide (SRO)) and the other without (SiO_2), was reported to differ by a factor of 30 under the same pumping conditions. Clearly, this issue will need to be understood, and resolved, as will the poor repeatability of gain, in order for viable lasers and amplifiers to be realized using this material system.

In this paper, we present a rate equation analysis of the indirect excitation processes in erbium-doped silicon-rich silica. We show that many of the puzzling issues surrounding the apparent low fraction of excitable erbium and the difficulty in repeatably achieving optical gain may be due to excited state absorption (ESA).

2. Modelling excitation exchange

We begin by looking at the rate equations that describe the excited state population of erbium in both stoichiometric SiO_2 and SRO. It is straightforward to show that the Er excited state populations are given by

$$N_2^{SiO_2} = \frac{\sigma\tau P N_{Er}}{1+\sigma\tau P}, \quad (1)$$

$$N_2^{SRO} = \frac{k\tau n_b N_{Er}}{1+k\tau n_b}, \quad (2)$$

where σ is the Er pump absorption cross section, τ the Er lifetime, P the pump photon flux, N_{Er} the Er ion concentration. k is the Si nc to Er excitation coefficient, and n_b is the Si nc excited state population. We can then apply these equations to the data reported by Fujii et al. [9]. In their paper (data extracted and reproduced here in Fig. 1), they report PL at 1.5 μm from two samples containing the same concentration of erbium, one being a stoichiometric silica matrix whilst the other contains Si nc. At the maximum pump power reported (1.5 W/cm^2), the silicon-rich sample is in the saturation regime while the SiO_2 sample is still in the linear regime; therefore the ratio of the number of excited Er ions in the two samples can be simply estimated to be

$$\frac{N_2^{SRO}}{N_2^{SiO_2}} \cong \frac{1}{\sigma\tau P}. \quad (3)$$

Taking $\sigma_{488\,nm} \sim 10^{-20}\,cm^2$, $\tau = 10\,ms$ and $P = 4\times10^{18}\,cm^2\,s^{-1}$ (corresponding to 1.5 W/cm^2) yields an Er luminescence ratio of 2000. As the experimentally observed PL ratio is only 30, this indicates that just 1.5% of the available Er ions in the SRO sample have been excited. This conclusion seems surprising considering that, for the volume fraction of Si nc and Er concentration specified in the SRO sample, the majority of the Er ions would be expected to lie within 1.5 nm of a Si nc—the commonly accepted interaction range for Si nc to Er energy transfer [5]. Nonetheless, the small percentage inferred above is consistent with the findings of Wojdak et al. [10], as they estimate from their cw pumping data a sub-linear behaviour beyond an excited Er population of $1.1\times10^{18}\,cm^{-3}$ out of a total erbium concentration of $2.2\times10^{20}\,cm^{-3}$.

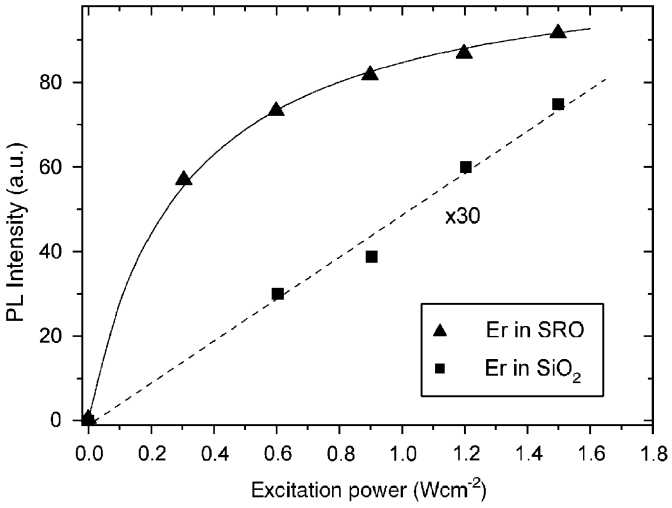

Fig. 1. Data extracted from Ref. [9] showing the dependence of Er PL on pump photon flux for a stoichiometric silica sample (SiO_2) and a silicon-rich silica sample (SRO) both containing 0.07% Er.

The small fraction of Er ions that is excited by the Si nc is related to a previous debate on whether a Si nanocrystal can excite more than one Er ion [11,12]. Initial evidence seemed to suggest that each Si nanocrystal could only excite one Er ion, leading to various hypotheses such as Auger de-excitation and pair-induced quenching to account for this limit [11]. It is now becoming clear, though, that multiple Er ions (as many as 20) can be excited if the pump power is increased sufficiently—by 10–100 times beyond the saturation power [10]. More recently, the framing of this problem has become complicated by findings that strong Er PL can be sensitized not just with Si nanocrystals, but also by an amorphous silicon network in which no distinct Si nanocrystals exist [13].

To date, no compelling explanation has been able to account satisfactorily for all these observations in a consistent manner. For example, a plausible explanation that most (over 90%) of the Er ions in the material may be optically inactive or quenched, and therefore non-luminescent, appears at odds with the ability of these ions to be excited at high power, after the initial Er PL saturation has occurred. Furthermore, the Er ions that are not excited by the Si nc can nonetheless be excited by direct resonant optical excitation, which provides further proof that these Er ions are optically active, though not susceptible to excitation exchange from Si nc. Fig. 2 shows the