

Indirect excitation of 1.5 μm emission from Er^{3+} in silicon-rich silica

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We report the observation of near-IR emission from erbium in silicon-rich silica, excited using a filtered white-light source. The characteristic ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ intra- $4f$ transition at 1535 nm is observed even when excitation wavelengths corresponding to the principal erbium optical absorption bands are removed using selective filtering. We ascribe this effect to an efficient transfer mechanism between silicon nanoclusters present in the silicon-rich silica films and the rare-earth ions. This is in good agreement with our previous work in this area and suggests the possibility of obtaining flashlamp-pumped erbium optoelectronic devices. © 2000 American Institute of Physics. [S0003-6951(00)00906-2]

Erbium-doped materials are at the heart of current optoelectronic telecommunications devices due to the correspondence between the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ Er^{3+} transition at 1.53 μm and one of the principal low-loss transmission windows in silica optical fibers. However, the integration of erbium emission with current silicon electronics remains problematic, as erbium-doped bulk crystalline silicon suffers from very low luminescence quantum efficiency. Recently, doped nanoclustered or porous silicon has shown considerable promise, and there have been a number of studies, including those by this group, showing that an efficient exchange mechanism exists between the silicon host and rare-earth ion which transfers either broad-band optical or electrical excitation into narrow-band emission at 1.53 μm .¹⁻⁶

Current erbium-based optoelectronic devices employ erbium-doped silica fibers or waveguides with a high-power laser diode pumping the 980 nm Er^{3+} absorption. Despite recent reductions in the cost of such pump sources, the development of a broad-band pumpable erbium system, or even a direct electrical injection device, would yield significant savings and constitute a major technological advance. The presence of an efficient coupling mechanism to a broad-band absorbing host suggests that such devices are feasible. In this letter we demonstrate the existence of host-to-rare-earth coupling in silicon-rich silica and report broad-band white-light pumping of erbium-doped material.

Samples were prepared by plasma-enhanced chemical vapor deposition (PECVD). The method is more fully detailed elsewhere,^{1,7,8} but briefly consisted of plasma dissociation of silane, nitrous oxide, and a volatile erbium organic chelate in a parallel-plate plasma chamber. Film stoichiometry was controlled by varying the relative flow rates of the silane and nitrous oxide gases, and by controlling both the temperature of the organic precursor and the flow rate of the carrier gas. Auger analysis of the deposited films showed them to be silicon rich (15% excess silicon) and to contain 1 at. % erbium. Film thicknesses were in the range 1–3 μm . Previous work has demonstrated that careful selection of

growth conditions produces films containing excess silicon in the form of nanoclusters, the size of which depends on growth parameters and postprocess annealing. The presence of such clusters has been inferred from optical absorption and visible photoluminescence studies of silicon-rich silica.^{1,7,8}

Photoluminescence experiments were conducted using a standard tungsten-halogen bulb to excite the samples, care being taken to eliminate any 1.5 μm output from the bulb using a low-pass filter. A series of high- and band-pass filters were inserted between the source and sample in order to selectively remove different wavelength regions corresponding to strong erbium ion absorption lines.

Photoluminescence excitation spectra were taken for all samples using an argon ion laser, a scanning monochromator, and an InGaAs photodiode. In all cases, the samples exhibited the characteristic ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ Er^{3+} transition at 1.53 μm . Spectra were obtained by tuning the argon-ion laser to each of the available lines in turn and monitoring the output from the samples at 1.53 μm . Care was taken to maintain the same laser power at each wavelength.

Absorption spectra of the samples were taken by removing the films from the silicon substrate and measuring absorption using the tungsten halogen bulb over the range 450–850 nm. For comparison and identification of the principal erbium absorption lines, an absorption spectrum was also measured for a reference erbium-doped silica fiber.

Figure 1 shows a photoluminescence excitation spectrum of an erbium-doped silicon-rich silica film (erbium concentration: 1 at. %). Although the peak at 488 nm indicates some degree of direct excitation of erbium, luminescence is also seen when pumped at wavelengths well away from absorption bands. A comparison of the (PLE) signals at 488 and 465 nm indicates that excitation at 488 nm is 75% indirect, 25% direct. Also shown is the absorption spectrum of the erbium-doped film over this range of wavelengths (Fig. 2). The only feature apparent is a monotonic band-edge absorption across much of the visible region. No erbium absorption lines were visible, despite the observation of characteristic erbium luminescence from these films. This is likely to be due to the strong band-edge absorption dominating and

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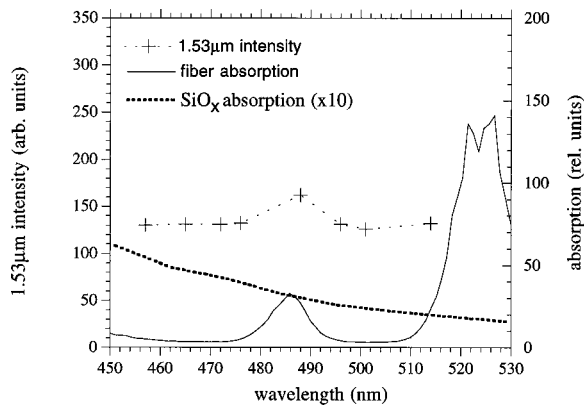


FIG. 1. Photoluminescence excitation spectrum of erbium-doped silicon-rich silica film. Also shown are absorption spectra over this wavelength range of the film and of an erbium-doped silica optical fiber.

“swamping” the erbium lines. This is in agreement with results from this group and others who have concluded that the excitation mechanism in erbium-doped bulk, nanoclustered, and porous silicon is carrier-mediated¹⁻⁶ and consists of absorption by the silicon followed by exchange to the rare-earth ion. It should be stressed at this point that stoichiometric silica films doped with similar concentrations of erbium did not exhibit a coupling effect and only showed 1.53 μm emission when pumped directly.

Figure 3(a) shows a series of emission spectra of the filtered white-light source used to excite the erbium-doped silicon-rich silica film. This illustrates the action of the low-pass filter used to remove infrared emission from the source, and the effect of the series of filters used to remove wavelengths corresponding to the erbium absorption lines around 380, 490, 520, and 650 nm. Figure 3(b) is an absorption spectrum taken of a reference erbium-doped silica fiber, included to show the positions and intensities of the erbium absorption bands. The erbium emission spectra obtained using the combinations of filters shown in Fig. 2(a) are shown in Fig. 4. Of particular interest is the spectrum obtained when the band-pass filter marked **B** was used. In this case, the range of excitation wavelengths used lies away from any of the characteristic erbium absorption bands. The overlap between pump and absorption wavelengths is therefore very small for this case. In addition, the light source was not

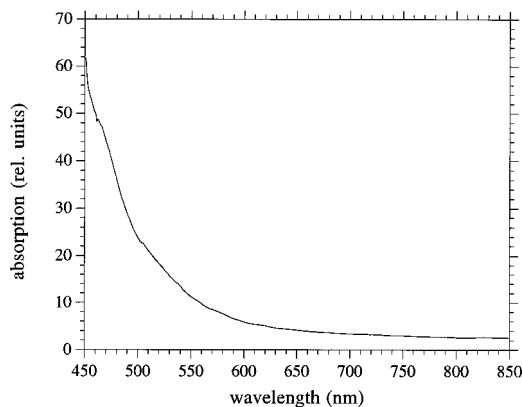


FIG. 2. An absorption spectrum of an erbium-doped silicon-rich silica thin film. The erbium absorption lines are swamped by the strong band-edge absorption of the silicon nanoclusters.

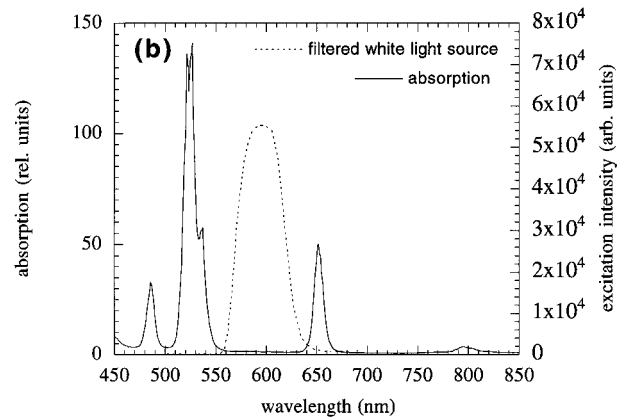
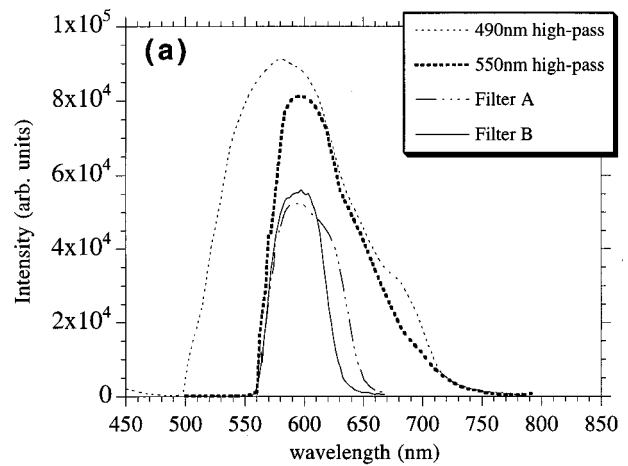


FIG. 3. (a) Emission spectra of the white light source used to excite the erbium-doped samples, showing the wavelength ranges selected using different combinations of filters. (b) Absorption spectrum of erbium-doped silica optical fiber, showing the principal erbium absorption bands. Also shown is the output spectrum of the filtered excitation source with filter **B** in place.

tightly focused on the sample and the power density at the surface of the sample with filter **B** in place was around 25 mW cm⁻². The observation of clear erbium luminescence spectra using such low excitation levels pumping away from the rare-earth absorption lines suggests the presence of a broad-band absorbing species and an efficient exchange mechanism. The samples studied here all exhibited visible photoluminescence around 1.6 eV which we and other groups have attributed in previous work to radiative recombination of confined excitons within silicon nanoclusters,^{7,8}

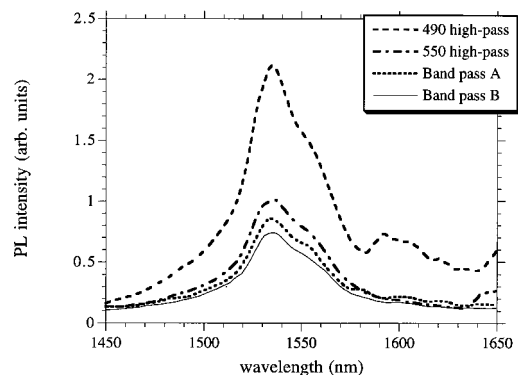


FIG. 4. Erbium emission spectra obtained using filtered white light source: labels correspond to the filters detailed in Fig. 3(a).

along with a 2 eV band which has been assigned to non-bridging oxygen hole defects in the silica matrix.^{7,9,10} Auger analysis of the films showed an excess silicon content of around 15 at. %, which we propose on the basis of the visible photoluminescence data to be in the form of silicon nanoclusters. The observation of the band-edge absorption in these films supports this contention. Assuming the primary absorbing species in these films to be nanoclustered silicon, we are led to the conclusion that an excitation exchange mechanism exists between the clustered silicon and the rare-earth ions. The nature of this exchange is unclear, but agrees qualitatively with similar observations published by this and other groups working on erbium-doped crystalline and porous silicon, silicon nanopowders, and chalcogenide glasses.^{1-6,11} For the case of doped silicon, the contention is that the exchange mechanism is carrier mediated, while for chalcogenide glasses the mechanism is presumed to proceed via the host absorption bands followed by a resonant transfer to the luminescent ion, possibly via defect states.¹¹ Recent results from other groups looking at silicon nanopowders doped with erbium during the growth phase also indicate efficient excitation exchange between the silicon host and the rare-earth ion.¹²

In conclusion, even with the 350, 490, 520, and 650 nm bands removed from our white-light pump source, erbium emission from the thin film samples is strong enough to yield a clear spectrum. We postulate an efficient excitation exchange mechanism between silicon nanoclusters and erbium ions. It is possible that the mechanism may be carrier mediated or take the form of a resonant dipole-dipole interaction. The exact nature of the transfer is unclear, and work is un-

derway within this group to investigate it more closely. In particular, the possible role of defects is being studied. However, we have demonstrated that it is possible to employ excitation exchange to pump erbium-doped silicon-based materials using broad-band sources such as flashlamps. This may reduce the need for expensive laser diode pump sources for erbium-based optical amplifiers, lossless splitters, and other optoelectronic devices.

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