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Citation: Journal of Applied Physics 100, 014902 (2006); doi: 10.1063/1.2206871
View online: http://dx.doi.org/10.1063/1.2206871
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Pump-probe experiments at 1.54 μm on silicon-rich silicon oxide waveguides

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(Received 1 February 2006; accepted 28 March 2006; published online 5 July 2006)

Optical pump-probe measurements were performed on slab waveguides containing excess silicon in the form of nanoclusters or nanocrystals and erbium. The measurements were performed by prism coupling a 1.54 μm probe beam into a waveguide formed by silicon-rich oxide and monitoring its intensity and temporal response as the waveguide was optically pumped from above with a chopped 477 nm excitation source. Induced absorption (losses) of the 1.54 μm probe beam in erbium-doped and undoped silicon-rich oxide waveguides was observed in all cases. For the samples containing only well-defined nanocrystals, a fast (~60 μs) induced absorption component associated with free carriers within the silicon nanocrystals is reported, while for samples containing defective nanocrystals or nanoclusters, a much slower (>10 min) component is observed. The free carrier absorption is shown to be reduced by delaying the probe beam relative to the pump beam in cases where it dominates. © 2006 American Institute of Physics. [DOI: 10.1063/1.2206871]

I. INTRODUCTION

Silicon-based photonics has received enormous interest in recent years and has been further stimulated by recent publications reporting fast modulation in silicon-based waveguide structures1 and the fabrication of a laser based on a Raman effect.2,3 Erbium (Er)-doped photonic materials and structures also continue to attract considerable attention due to possible applications in telecommunications at a wavelength of 1.54 μm. Efficient nonresonant erbium excitation has been demonstrated in the presence of silicon nanocrystals (Si-nc), when both are present in the same silica substrate.4 The nanocrystals are believed to act as sensitizers, absorbing incident radiation over a wide spectral range and coupling it efficiently to Er3+ ions. In comparison with Er-doped bulk silicon, this medium (SiO2:Si-nc+Er) shows a strong room-temperature 1.54 μm emission due to a reduction of nonradiative recombination, thereby raising the possibility of making a room-temperature optical amplifier that operates under broadband pumping. More recent measurements have further demonstrated that this sensitization effect is present in Er-doped silicon-rich oxides (SROs) that do not contain well-defined nanocrystals.5

Despite numerous papers exploring the optical properties of Er-doped and undoped SROs and their potential for making a silicon-based amplifier, very little is reported on propagation at 1.54 μm. Moreover, the results that have been presented seem to show conflicting results. For example, Han et al.6 report optical gain at 1.54 μm in samples prepared by plasma-enhanced chemical-vapor deposition (PECVD) and rf sputtering, using an erbium concentration of 1.98 × 1019 cm−3 (or 0.03 at. %), and with a core-mode overlap of around 50%. On the other hand, Kik and Polman7 reported induced absorption using the same technique in similar rib waveguides prepared by ion implantation with a higher erbium concentration, 8.2 × 1020 cm−3, and a core-mode overlap of ~1%. With this in mind, the current study employs optical pump-probe measurements to examine the propagation of 1.5 μm light in slab waveguides constructed from Er-doped SROs as a function of annealing temperature, exploring the range of 700–1100 °C over which SROs phase separate to form Si nanoclusters and nanocrystals. The slab waveguides employed for these measurements have a high (>80%) core-mode overlap and relatively low erbium concentration (1019 cm−3) produced by multiple-energy ion implantation of silicon and erbium into silicon dioxide.

II. EXPERIMENT

Slab waveguides were fabricated by implanting multiple-energy silicon and erbium ions into a 10 μm thick thermal oxide layer grown on (100) silicon. The implants were chosen to produce a near-uniform silicon (10 at. %) and erbium (1 × 1019 cm−3) concentrations, over a depth of 1.1 μm, starting at a depth of 0.3 μm and extending to 1.4 μm. Mode-line measurements have confirmed an active layer thickness around 1 μm after analyzing multiple dips from the sample. The refractive index difference between the implanted layer and the oxide matrix allows the necessary coupling conditions (see inset of Fig. 1). Silicon nanocrystals were formed by annealing at 1100 °C for 1 h in N2 gas. From photoluminescence spectra the mean average silicon nanocrystal size distribution is estimated to be around 3 nm.8 Based on the mean size and the excess Si we have estimated a nanocrystal concentration of ~1019 cm−3. The annealed samples were subsequently implanted with erbium and annealed at 900 °C for 1 h to remove implantation damage and activate the Er3+ ions. The total Er concentration was 1019 cm−3. The mode overlap of the probe beam in the 1 μm active layer was estimated to be >80%. Pump-probe experiments were undertaken using a prism-coupling technique (see inset in Fig. 1) described elsewhere.9 Briefly, a 1.5 μm...
probe beam was prism coupled into the waveguide and its intensity monitored as it exited the edge of the guide. The intensity and temporal response of this signal were then monitored as the waveguide was optically pumped from above with a chopped frequency of 11 Hz using a standard lock-in technique. Using a prism has several advantages over edge coupling into rib waveguides. For example, beam collimation is retained along the waveguide without the loss caused by edge scattering in rib guides and no precise optics or fibers are required for the coupling. The measurement also requires little sample preparation.

III. RESULTS AND DISCUSSION

Figure 1 shows the results of pump-probe measurements for two different samples: one labeled (nc+Er) containing nanocrystals and erbium and the other sample labeled (nc) containing only nanocrystals. The vertical axes show the normalized transmitted probe intensity (I), which is defined as the ratio between the probe intensities with and without the pump: I_{	ext{with pump}}/I_{	ext{without pump}}. As this is a relative measurement it does not take into account absolute changes in intensity due to internal losses in the system caused by the waveguide itself. In both cases induced absorption is observed but important differences are evident for the two samples structures. For the sample containing well-defined nanocrystals (nc) a saturation level is observed even at low pump powers, whereas for the sample containing Er (nc+Er) the induced absorption increases smoothly (the transmitted signal decreases) with increasing pump power even up to the highest pump powers used, 0.8 W cm$^{-2}$. Note that these pump powers are similar to the ones used by Han et al.$^5$ Thus, the probe beam can be absorbed almost completely in the sample containing Er (nc+Er), while a saturation level is reached for the sample containing silicon nanocrystals only (nc). The dynamics of the two systems are also quite different. Figures 2(a) and 2(b) show the intensity of the probe beam as a function of time for the samples (nc) and (nc+Er), respectively. When the pump is switched on, an immediate (fast) absorption occurs for both cases. After the pump is switched off, the probe beam recovers to its initial value. For the sample (nc+Er) the recovery time is very slow (tens of minutes), while for the sample (nc) it is much faster. In order to achieve the necessary fast time resolution, we have previously performed measurements with a pulsed (25 ns) pump beam on single-implant samples.$^10$ In pulsed experiments, the recovery time of the probe beam coincides with the nanocrystal luminescence lifetime of around 60 μs. This is consistent with the induced absorption being caused by free carriers within the nanocrystals. The time regime presented in Fig. 2(b), >20 min, cannot be explained by this phenomenon.

A similar long-lived absorption process was observed by Kik and Polman and attributed to a charge trapping mechanism responsible for nanocrystals “blinking,” which has been observed to exhibit a similar time dependence. However, if this were the mechanism one would expect the same blinking effect for samples containing nanocrystals alone (nc). From Fig. 2(a) this is clearly not the case and the difference in behavior for the nc and nc+Er samples must result from some other process. One such possibility is the effect of the implant and annealing schedule on the material structure. The Er implants used to produce the nc+Er samples damage the preformed silicon nanocrystals and these do not fully recover after annealing at 900 °C. It is therefore likely that these samples contain a fraction of nanoclusters and/or damaged nanocrystals rather than the well formed nanocrystals produced after annealing at 1100 °C. Although not depicted here, there is a direct evidence between the annealing temperature and the long recovery time effect.

In an attempt to understand the significance of material structure and free carrier absorption, experiments were un-
dertaken on samples annealed under different conditions and using a delay between the pump and probe beams, as shown in the schematics depicted in the inset of Fig. 3. The relative delay between the pump beam and probe beam is controlled by synchronizing a chopper with a signal generator so that the probe beam is switched on with a tunable delay time. The excitons produced in the nanocrystals are expected to recombine with a lifetime of about 60 μs, with some of them transferring energy to the erbium ions nonresonantly. In the steady-state regime the number of excited erbium ions is constant as they are continuously being excited and decaying. To avoid free carrier absorption the delay between pump and probe was set to around 4 times the decay lifetime of nanocrystals, at ≈ 300 μs. At this time there are no free carriers in the nanocrystals but all the erbium excited by the pump is still in its excited state as the lifetime for the excited Er is around 5.5 ms, as shown in the inset of Fig. 3. The filled and open squares are the results for samples containing only excess of silicon annealed at different temperatures (i) without and (ii) with a delay time, respectively. The filled and open stars are the results for samples containing a simultaneous implantation of Si and erbium followed by a single annealed step at different temperatures. In the vertical axes is shown the relative difference of the probe beam so we can estimate the percentage of the absorbed signal. Figure 3 presents the data for a particular low pumping regime, so the relative ratio may change at different pump powers. For the sample annealed at 1100 °C and containing perfectly formed nanocrystals, we observe a dramatic reduction in induced absorption when a delay time is applied. For the samples annealed below the formation of nanocrystals (labeled Si), we observe a reduction in the induced absorption but there is practically no difference when a delay time is applied. This is consistent with the results discussed earlier for samples containing nanocrystals and implanted with erbium (Sr+Er). In both cases, it appears that induced absorption is dominated by a long-lived (minutes) carrier trap associated with nanoclusters and/or defective nanocrystals, whereas structures containing well-defined nanocrystals (anneal temperatures >1000 °C) exhibit an effect consistent with absorption by confined carriers within the nanocrystals.

IV. CONCLUSIONS

In conclusion, induced absorption has been studied at 1.5 μm for a range of waveguide structures typically employed for the fabrication of silicon-nanocrystal-based devices, including optical amplifiers. In all cases, photoinduced absorption of the probe beam was observed but important differences were observed depending on the material structures employed. Specifically, samples containing nanoclusters and/or defective nanocrystals (samples annealed at T <1000 °C) exhibited long-lived (minutes) absorption consistent with the presence of specific charge traps, while those containing well-defined nanocrystals (samples annealed at T >1000 °C) exhibited a characteristic lifetime for absorption consistent with absorption by confined carriers within the nanocrystals. By incorporating an appropriate delay between the pump and probe beams it was possible to significantly reduce the induced absorption due to this latter process.