CHIN.PHYS.LETT. Vol. 18, No. 7 (2001) 986

Dependence of 1.54 μm Photoluminescence on Excess-Si Degrees of Er-Doped Si-Rich SiO_2 Films Deposited by Magnetron Sputtering *

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(Received 13 November 2000)

Room-temperature 1.54 μm photoluminescence (PL) is observed from Er-doped Si-rich SiO_2 (SiO_2:Si:Er) films deposited by using the magnetron sputtering technique. To determine the optimum Si content in the SiO_2:Si:Er films, the percentage area of the Si target in the composite SiO_2–Si–Er target was changed from 0, to 10%, 20% and 30%. The percentage area of the Er target was fixed at 1%. It is found that the optimum annealing temperatures for Er^{3+} luminescence intensities are 900°C for the SiO_2:Er film and 900, 800 and 700°C for the SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si (percentage areas of Si target), respectively. The SiO_2:Si:Er film containing 20% excess-Si and annealed at 800°C has the most intense PL.

PACS: 78.55.−m, 73.61.Tm

Si-rich SiO_2 is a promising material for light emission not only in the visible range,[1−3] but also around 1.54 μm if Er is doped.[4−8] As is well known, it is interesting that 1.54 μm is coincident with the absorption minimum window of silica-based optical fibres. Er-doped Si-rich SiO_2 (SiO_2:Si:Er) films contain sufficient oxygen natively, which are beneficial for the optical activation of Er. Si nanoclusters in SiO_2:Si:Er films can transfer the photoexcitation energies to Er^{3+} and excite them efficiently. Thus, the sizes and concentration of Si nanoclusters have a great effect on the Er^{3+} photoluminescence (PL) yield. It is important to study the optimum excess-Si content in SiO_2:Si:Er films and the corresponding optimum annealing temperature for efficient Er^{3+} luminescence. In this study, Er-doped Si-rich SiO_2 films were deposited on p-Si substrates by the magnetron sputtering technique. The excess-Si content was controlled by the percentage area of the Si target in the composite SiO_2–Si–Er target and demonstrated by x-ray photoelectron spectroscopy (XPS). The dependence of PL intensity on excitation power was compared and studied for all the SiO_2:Si:Er films annealed at a series of temperatures.

The substrates used were the (100)-oriented 6 − 9 Ω·cm p-type Si wafers. The magnetron sputtering was performed under a base vacuum of $\sim 10^{-5}$ Pa at 200°C. A SiO_2–Si–Er composite target was used for depositing SiO_2:Si:Er films. In different magnetron sputtering processes, the percentage area of the Si target in the target was changed from 0 to 10%, 20% and 30%. The percentage area of the Er target was fixed at 1%. The XPS study demonstrated that the excess-Si content increases linearly with the increasing percentage area of the Si target in the composite target. Hereafter, the excess-Si content is described by using the percentage area of the Si target. The XPS spectra of the SiO_2 film and the SiO_2:Si films containing 10%, 20% and 30% excess-Si are shown in Fig. 1. The peaks at 103.6 ± 0.2 and 100.0 ± 0.2 eV correspond to Si in SiO_2 and Si in Si nanoclusters, respectively. All the thicknesses of the SiO_2:Er and SiO_2:Si:Er films, controlled by deposition time, were 12 nm. The films were annealed separately at 600, 700, 800, 900 and 1000°C for 30 min in a furnace in an N_2 atmosphere. The PL spectrometer consisted of a 3 W argon laser, a light chopper, a single-grating monochromator, a liquid-nitrogen-cooled Ge detector, and a lock-in amplifier. The PL was excited by the 488 nm line of an Ar laser and measured at room temperature.

Figures 2(a) and 2(b) show PL spectra of the SiO_2:Er film and the SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si, as-grown and annealed at 800°C, respectively. For clarity, compared to Fig. 2(b), the intensities of all PL spectra in Fig. 2(a) are enlarged ten times. The Er^{3+} luminescence peak due to the intra-4f transition ($^4I_{13/2} \rightarrow {}^4I_{15/2}$) around 1.54 μm appears in all the PL spectra. The PL intensity of the SiO_2:Si:Er film containing 20% excess is the most intense, and is above ten times larger than that from SiO_2:Er. For the 30% excess-Si film, the full width at the half maximum of the PL spectrum widens significantly in comparison to those of the other films with less excess-Si content.

*Supported by the National Natural Science Foundation of China under Grant No. 59832100 and the State Key Laboratory on Integrated Optoelectronics.

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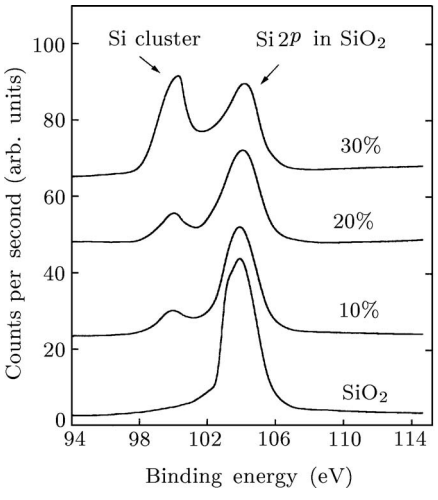


Fig. 1. XPS spectra of the SiO_2 film and SiO_2:Si films containing 10%, 20% and 30% excess-Si.

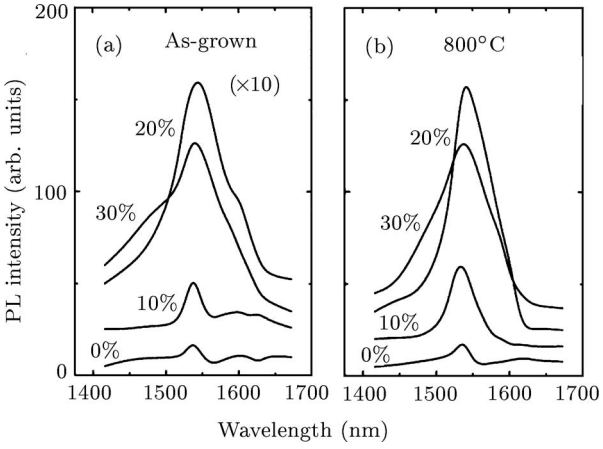


Fig. 2. PL spectra from (a) the as-grown SiO_2:Er film and SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si and (b) those annealed at 800°C. Compared to (b), the intensities of all the PL spectra in (a) had been enlarged ten times.

Figure 3 shows the PL intensities of the SiO_2:Er film and the SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si as a function of the annealing temperature. For each sample, there is an optimum annealing temperature corresponding to the maximum of PL intensity. The optimum annealing temperatures are 700, 800 and 900°C for the SiO_2:Si:Er films containing 30%, 20% and 10% excess-Si and 900°C for the SiO_2:Er film, respectively. As can be seen, the more the Si content, the lower the optimum annealing temperature.

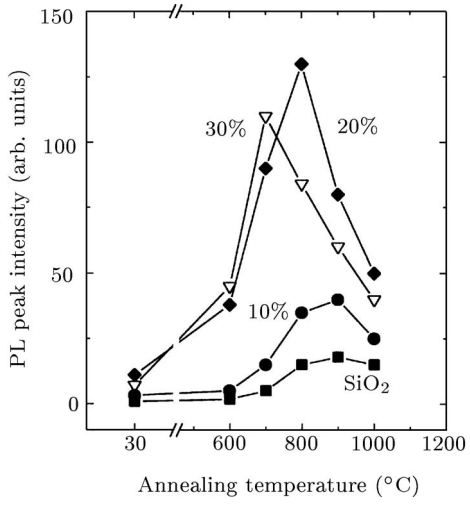


Fig. 3. PL intensities as a function of the annealing temperature for the SiO_2:Er film and SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si.

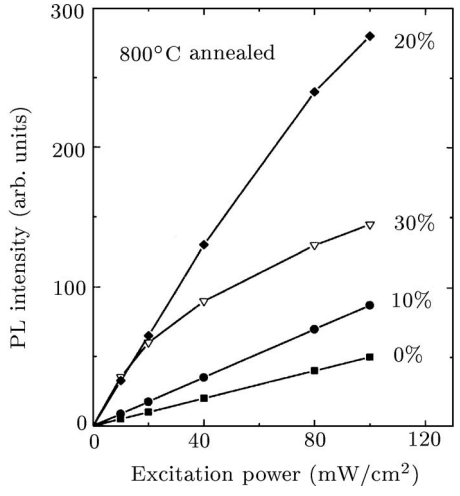


Fig. 4. PL intensities as a function of the excitation power for the SiO_2:Er film and SiO_2:Si:Er films containing 10%, 20% and 30% excess-Si annealed at 800°C.

Figure 4 shows the excitation power dependence of Er^{3+} PL intensities for the SiO_2:Er film and three types of SiO_2:Si:Er films, all annealed at 800°C. For the SiO_2:Er film and the SiO_2 film containing 10% excess-Si, the PL intensities increase linearly with excitation power in a range of 0–100 mW/cm^2. For the 20% and 30% excess-Si samples, the increase of the PL intensities slows down at higher excitation power of the range. The PL intensity of the 30% excess-Si sample is nearly equal to that of the 20% excess-Si sample at excitation power lower than 20 mW/cm^2, but evidently weaker than that of the latter at an excitation power higher than 20 mW/cm^2. The PL from the SiO_2:Si:Er samples is much more intense than that from the SiO_2:Er sample annealed at an identical annealing temperature and under an identical excitation power. These experimental results indicate that the

Si nanoclusters in the SiO_2:Si:Er films are very helpful for efficient Er^{3+} luminescence. The reason for this is that the carriers photoexcited in the Si nanoclusters can make the energy transfer excite Er^{3+} efficiently, which has been proven experimentally[5–8] and theoretically.[9] Combining Er with oxygen to form Er–O luminescence centres is a prerequisite for optical activation of Er.[10–12] In the Si oxide, an excess of Si is equivalent to a deficiency of oxygen in comparison to SiO_2, hence it is not difficult to understand why higher quantities of excess-Si in the SiO_2:Si:Er film would not always lead to a higher luminescence yield.

For the reason why the optimum annealing temperatures for effective Er^{3+} PL are lower for the SiO_2:Si:Er films containing more quantities of excess-Si, we believed that the optimum annealing temperature is determined by the annealing behaviour of the Si nanoclusters as well as Er–O luminescence centres. The Er–O luminescence centres have a lower concentration at lower annealing temperature, and begin to dissociate at temperatures higher than 900°C.[10,11] Therefore, the fact that the SiO_2:Er sample has the optimum annealing temperature around 900°C can be realized. In the case of the SiO_2:Si:Er films, the effect of the Si nanoclusters on Er^{3+} PL should be taken into account. As the annealing temperature increases, the concentration of the Si nanoclusters increases too, leading to an enhancement of PL intensity. However, as the annealing temperature increases further, the Si nanoclusters grow in size,[13] become less efficient in excitation of Er^{3+}, and suppress temperature quenching[14] because the energy gap of a larger Si nanocluster is narrower due to the quantum confinement effect. Therefore, PL intensity attains a maximum at a certain annealing temperature. The Si nanoclusters are easier to grow in the SiO_2:Si:Er films containing more excess-Si.[14] Hence, the more excess-Si in the SiO_2:Si:Er film, the lower the optimum annealing temperature.

In summary, room-temperature 1.54 μm PL is observed from SiO_2:Si:Er films. To determine the optimum Si content and the corresponding optimum annealing temperature for Er^{3+} PL, we comparatively studied Er^{3+} PL from the SiO_2:Er films and three types of SiO_2:Si:Er films containing different quantities of excess-Si and annealed at different temperatures. It is found that the more excess-Si in the SiO_2:Si:Er films, the lower the optimum annealing temperature for Er^{3+} luminescence. The 20% excess-Si sample annealed at 800°C has the most intense PL. The magnetron sputtering technique is proven to be a promising technique to grow the SiO_2:Si:Er films for efficient Er^{3+} PL.

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