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Er–carrier interaction and its effects on the Er³⁺ luminescence of erbium-doped Si/SiO₂ superlattices

Jung H. Shin,[a] Ji-Hong Jhe, and Se-Young Seo
Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), 373-1 Kusung-dong, Yusung-gu, Taejon, Korea

Yong Ho Ha and Dae Won Moon
Surface Analysis Group, Korea Research Institute of Standards and Science (KRISS), Taejon 305-606, Korea

The Er–carrier interaction and its effects on the Er³⁺ photoluminescence properties of erbium-doped Si/SiO₂ superlattices are investigated. The interaction between the erbium atoms and the electronic carriers was controlled by doping erbium into the SiO₂ layers only and by depositing buffer layers of pure SiO₂ between the erbium-doped SiO₂ layers and the Si layers. We demonstrate that by controlling the erbium-carrier interaction, a three orders of the magnitude enhancement of the Er³⁺ luminescence intensity and a nearly complete suppression of the temperature-induced quenching of Er³⁺ luminescence can be achieved while still allowing the Er³⁺ ions to be excited by the carriers. We identify the asymmetry between the dominant carrier-mediated excitation and the de-excitation paths of Er³⁺ ions as the possible cause for the observed effects. *© 2000 American Institute of Physics.* [S0003-6951(00)02324-X]

Since the first demonstration of 1.54 μm Er³⁺ luminescence by Ennen,[1] erbium doping of silicon has been studied intensely as a promising way to realize silicon-based integrated optoelectronics. One key point that has emerged is the importance of the interaction between the Er³⁺ $4f$ electrons and the electronic carriers in the silicon. This interaction not only allows Er³⁺ to be excited electrically, but since the electrical excitation cross sections of Er³⁺ $4f$ electrons can be up to 6 orders of magnitude larger than the resonant optical absorption,[2] it also enables, in principle, realization of very efficient optoelectronic devices. Already, light-emitting diodes using erbium-doped silcon have been demonstrated.[3,4]

However, interactions with carriers can result in a very effective quenching of the Er³⁺ luminescence as well. Excited Er³⁺ ions can Auger excite free or bound carriers if they become available, and at higher temperatures, excited Er³⁺ ions can Auger excite electrons in the valence band to form a bound exciton,[2,5,6] leading to a severe temperature quenching of the Er³⁺ luminescence. As a result, the overall Er³⁺ luminescence efficiency has so far remained too low to be practical.

One method that has been used to manipulate the Er–carrier interaction to obtain a better Er³⁺ luminescence is enlarging the band gap of Si by using nanocrystalline Si.[7–10] Unfortunately, such nanocrystalline Si films are generally very inhomogeneous such that an accurate control of the Er–carrier interaction is difficult. Recently, we have reported that a much more accurate control of the Er–carrier interaction is possible by using erbium-doped Si/SiO₂ superlattices, and that doping erbium atoms only into the SiO₂ layers is actually beneficial for the Er³⁺ luminescence.[11] Based on these observations, we have suggested that separating erbium

atoms from a direct contact with carriers may allow us to control the Er–carrier interaction in such a way that carrier-mediated de-excitation mechanisms are suppressed while the efficiency of carrier-mediated excitation mechanisms are retained. In this letter, we report on the Er³⁺ luminescence properties of erbium-doped Si/SiO₂ superlattices which have thin buffer layers of pure SiO₂ between the erbium-doped SiO₂ and Si layers. We find that by employing such buffer layers to control the Er–carrier interaction, it is possible to increase the Er³⁺ photoluminescence (PL) intensity by 3 orders of magnitude and nearly completely suppress the temperature quenching of the Er³⁺ luminescence while still retaining the efficiency of the carrier-mediated excitation. The observed results are ascribed to the asymmetry between the dominant carrier-mediated excitation and the de-excitation paths of Er³⁺ ions.

The superlattices were deposited on oxidized Si substrates by electron cyclotron resonance plasma enhanced chemical vapor deposition of SiH₄ and O₂ with cosputtering of erbium. The base pressure, the microwave power, and the substrate temperature were 1×10^{-6} Torr, 400 W, and 450 °C, respectively. Under such conditions, the deposition rates are expected to be ~ 1 Å/s. The deposition times of both Si layers and the erbium-doped SiO₂ layers were fixed at 20 s. The deposition times of the buffer layers ranged from 0 to 15 s. The total period of layers was fixed at 30 for all samples. After deposition, the films were rapid thermal annealed in a sequence of 20 min at 600 °C, 5 min at 950 °C, and 5 min at 600 °C to avoid cracking and spalling of the films. The Er³⁺ luminescence spectra were measured using an Ar laser, a grating monochromator, a thermoelectrically cooled InGaAs detector, and the standard lock-in technique. The nominal laser power was 200 mW. Low temperature PL spectra were measured using a closed-cycle helium cryostat.

[a]Author to whom correspondence should be addressed; electronic mail: jhs@mail.kaist.ac.kr
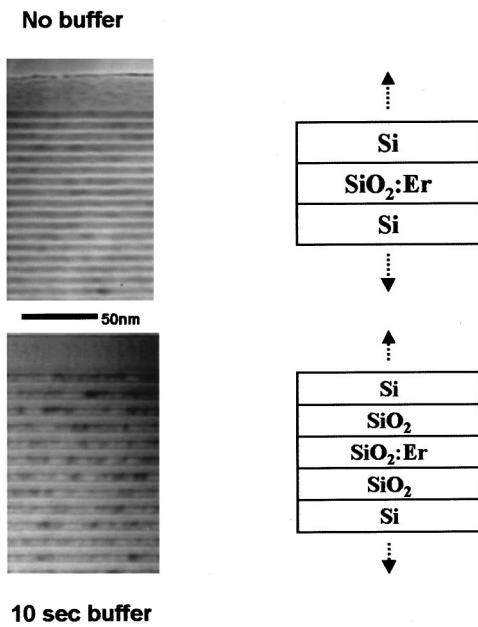
3567

No buffer

10 sec buffer

FIG. 1. Bright field cross-section transmission electron microscope images of the superlattices. On the right is a schematic drawing showing the composition of the layers.

Compositional analysis was performed by Rutherford back-scattering spectroscopy (RBS) using 2 MeV He^{++} ions and medium energy ion scattering (MEIS) spectroscopy using 100 keV H^+ ions.

Figure 1 shows the bright-field cross-section transmission electron microsopy (TEM) images of superlattice films and schematic depictions of their composition. The dark bands are the Si layers. They are polycrystalline, and some grains can be observed. The Si and SiO_2 layer thicknesses are 4 and 3 nm, respectively, for the film with no buffer layers, and 5 and 6 nm, respectively, for the film with 10 s buffer layers. Thus, we estimate a buffer layer thickness of ~1.5 nm for this particular film, in agreement with the expected deposition rates. Buffer layer deposition time was not extended beyond 15 s because doing so resulted in excessive oxidation of the deposited Si layers and decrease in the overall film quality.
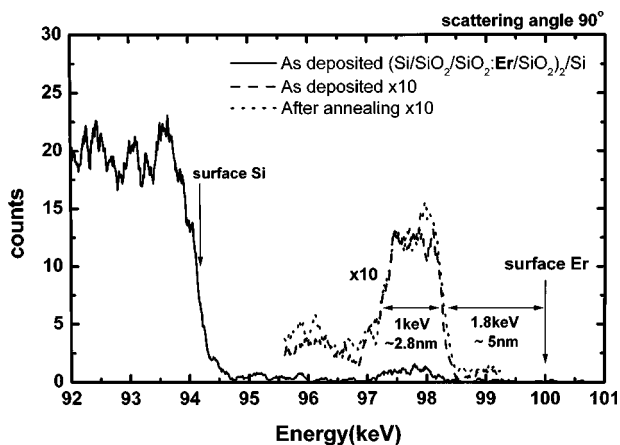
Figure 2 shows the MEIS spectra of such a buffered

FIG. 2. Medium energy ion scattering (MEIS) spectra of erbium-doped superlattice with buffer layers before and after the anneal sequence. The erbium signal is shown enlarged tenfold for clarity.
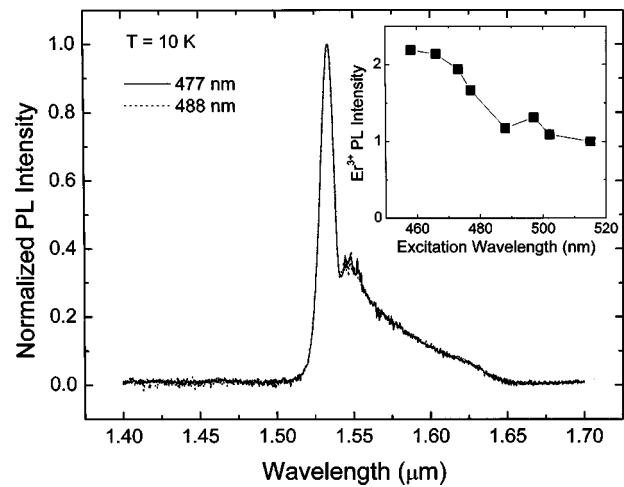

FIG. 3. The normalized Er^{3+} luminescence spectra, excited with either 477 or 488 nm line of the Ar laser and measured at 10 K of the superlattice film, which had buffer layers deposited for 15 s. The inset shows the dependence of the Er^{3+} luminescence intensity upon the excitation wavelengths at the constant pump power of 25 mW.

erbium-doped Si/SiO_2 superlattice film before and after the anneal sequence. The erbium peak is shown enlarged tenfold for clarity. The width of the erbium peak is 2.8 nm, in excellent agreement with the TEM result in Fig. 1. Moreover, the erbium peak hardly changes following the anneal sequence. This indicates that the diffusion of the doped erbium atoms during the anneal sequence is negligible, and confirms that we indeed have pure SiO_2 buffer layers separating the Si layers and the erbium-doped SiO_2 layers. Based upon this confirmation that the thickness of the erbium-containing region is the same for all samples, the erbium concentration in such regions was deduced using RBS to be ~0.1 at. % for all samples (not shown).

Figure 3 shows the normalized Er^{3+} luminescence spectra, excited with either 477 or 488 nm line of the Ar laser and measured at 10 K, of the superlattice film which had buffer layers deposited for 15 s. The two Er^{3+} luminescence spectra are identical, even though 488 nm excitation light can be absorbed resonantly by $^4I_{15/2} \rightarrow {}^2H_{11/2}$ intra-4f transition of Er^{3+} but not the 477 nm excitation light. Furthermore, as the inset shows, the Er^{3+} luminescence intensity does not show a peak near the optical resonance absorption band near the 488 and 515 nm excitation light. Taken together, Fig. 3 indicates that the Er^{3+} luminescence is dominated by Er^{3+} ions in a single class of erbium sites, and that the carrier-mediated excitation is the dominant mode of excitation even with the presence of buffer layers.

The temperature dependence of the Er^{3+} luminescence intensity is shown in Fig. 4. A 477 nm line of the Ar laser was used to ensure that the Er^{3+} ions are excited only via carriers. We find that the Er^{3+} luminescence intensity *increases* very strongly, by a factor of up to nearly 3 orders or magnitude, as the buffer layer thickness increases. Furthermore, the temperature quenching of the Er^{3+} luminescence is suppressed by the presence of buffer layers as well. Without buffer layers, the Er^{3+} luminescence intensity decreases by a factor of 3 as the temperature is increased from 10 K to room temperature, in agreement with previous results.[11] However, the temperature quenching of the Er^{3+} luminescence lessens
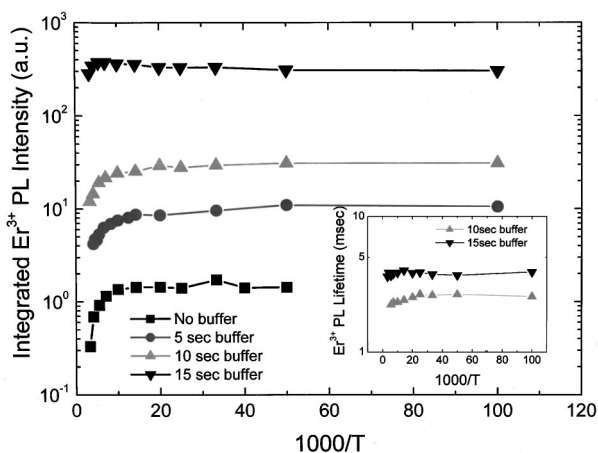
FIG. 4. The temperature dependence of integrated Er^{3+} PL intensities. The inset shows the temperature dependence of the luminescent decay lifetimes.

as the buffer layer thickness increases, and is nearly completely suppressed when the buffer layer thickness increases beyond 1.5 nm. Increasing buffer layer thickness also increases the Er^{3+} luminescence lifetime and suppresses their temperature quenching as well. This is shown in the inset, which shows the temperature dependence of the effective Er^{3+} luminescence lifetimes.

The results presented above are somewhat surprising, since the presence of buffer layers should inhibit the overall interaction between erbium and the electronic carriers in the Si layers, thus leading to the suppression of carrier-mediated excitation as well as carrier-mediated de-excitation of erbium. We note, however, that an asymmetry is known to exist between the dominant carrier-mediated excitation and de-excitation mechanisms of Er^{3+} in Si. In the following, we show that this asymmetry can be used to explain the observed effects of the buffer layers.

In general, Er^{3+} ions can interact with both free and trapped carriers. For excitation, the trapped carriers are far more efficient. The cross section for trapped-carrier mediated and free-carrier mediated excitation of Er^{3+} in silicon is 3×10^{-15} and 6×10^{-17} cm^2, respectively.[2] On the other hand, free carriers are far more efficient for de-excitation of Er^{3+}. The Auger coefficient for trapped-carrier mediated and free-carrier mediated deexcitation of excited Er^{3+} in silicon is about 1×10^{-15} and 4×10^{-13} cm^3 s^{-1}, respectively.[2]

A thin buffer layer of pure SiO_2, however, is very likely to screen effectively such interactions of Er^{3+} $4f$ electrons with free carriers in the Si layers since the $4f$ electrons remain well localized and atom-like even when doped into a host material.[12] On the other hand, if the buffer layer is thin enough, carriers photogenerated in silicon layers can tunnel through the SiO_2 buffer layers to any trap sites in the erbium-doped SiO_2 layers (e.g., Er-related defects). Indeed, using a $SiO_2/Si/SiO_2$ superlattice structure with nm-thin Si and SiO_2 layers very similar to the one used here, Qin *et al.*[14] have reported that precisely such tunneling of carriers from the Si layer to traps in the adjacent SiO_2 layer occurs. Therefore, we can expect that there exists a thickness window in which buffer layers will block the interaction of the Er^{3+} ion with the free carriers only. Given the above mentioned asymmetry between the excitation and de-excitation mechanisms, such buffer layers will then suppress carrier-mediated de-excitation of Er^{3+} while still allowing carrier-mediated excitation of Er^{3+} to proceed.

Note that this explanation does not require any band gap enlarging of the Si layers due to the quantum confinement effect. This is consistent with the fact that we observe suppression of temperature quenching of Er^{3+} luminescence even though the Si layers are too thick to show any quantum confinement effect,[13] since the temperature quenching of the Er^{3+} lumnescence, because it involves Auger excitation of electrons in the valence band, is ultimately related to interaction of Er^{3+} ions with free carriers.

In conclusion, we have demonstrated that Er^{3+} luminescence from the erbium-doped Si/SiO_2 superlattice can be greatly enhanced by doping erbium only into the SiO_2 layers and depositing thin buffer layers of pure SiO_2 between the erbium-doped SiO_2 layers and the Si layers. We ascribe the enhancement to the asymmetry between the carrier-mediated excitation and de-excitation mechanisms of Er^{3+} and to the fact that by using buffer layers, we can control the Er–carrier interaction such that only the de-excitation mechanisms are suppressed.

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