

# The characteristic carrier–Er interaction distance in Er-doped *a*-Si/SiO<sub>2</sub> superlattices formed by ion sputtering

Ji-Hong Jhe and Jung H. Shin<sup>a)</sup>

*Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), 373-1 Kusung-dong, Yuseong-gu, Taejeon, Korea*

Kyung Joong Kim and Dae Won Moon

*Nano Surface Group, Korea Research Institute of Standards and Science (KRISS), Taejeon 305-606, Korea*

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The characteristic interaction distance between Er<sup>3+</sup> ions and carriers that excite them in Er-doped *a*-Si/SiO<sub>2</sub> superlattices is investigated. Superlattice thin films consisting of 12 periods of *a*-Si/SiO<sub>2</sub>:Er/SiO<sub>2</sub>/SiO<sub>2</sub>:Er layers were deposited by ion sputtering and subsequent annealing at 950 °C. The dependence of the Er<sup>3+</sup> photoluminescence intensity on the thickness of the Er-doped SiO<sub>2</sub> layers is well-described by an exponentially decreasing Er-carrier interaction with a characteristic interaction distance of 0.5±0.1 nm. © 2003 American Institute of Physics.  
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Er<sup>3+</sup> luminesces at 1.54 μm due to the intra-4*f* transition from the first excited to the ground state (<sup>4</sup>I<sub>13/2</sub> → <sup>4</sup>I<sub>15/2</sub>), coincident with the absorption minimum of silica-based optical fibers. As this transition is parity-forbidden, and occurs only due to the parity-breaking effects of the crystal field, the luminescence lifetime tends to be very long (>ms). This allows for easy population inversion, and has allowed development erbium-doped fiber amplifiers that enabled the realization of all optical telecommunication networks.<sup>1</sup>

Yet, for the same reason, the optical absorption cross-section of Er<sup>3+</sup> is very small, necessitating the use of expensive lasers for excitation. However, if Er<sup>3+</sup> is doped into Si, then it can be excited via an Auger-type interaction with photogenerated excitons in Si.<sup>2–5</sup> Since only carrier generation is necessary, and since the absorption cross section of Si is several orders of magnitude greater than that of Er<sup>3+</sup>, this carrier-mediated excitation allows, in principle, the use of low-cost, broadband light sources such as light-emitting diodes for efficient excitation of Er<sup>3+</sup>. However, despite some early successes, the room-temperature Er<sup>3+</sup> luminescence efficiencies from Er-doped bulk Si remain too low to be practical.<sup>4,5</sup>

However, it has been demonstrated that by using either Si nanoclusters embedded inside an SiO<sub>2</sub> matrix<sup>6–8</sup> or Si/SiO<sub>2</sub> superlattice (SL) with nanometer-thick Si layers<sup>9,10</sup> as the host material, it is possible to obtain both the high excitation efficiency of Si and the high luminescence efficiency of SiO<sub>2</sub>. In such cases, Er<sup>3+</sup> luminescence is dominated by Er<sup>3+</sup> ions located outside, but close to, Si clusters.<sup>11–13</sup> Thus, Si clusters act as classical sensitizers, absorbing the pump photons and transferring the energy to Er<sup>3+</sup>.<sup>14,15</sup> As both optical gain<sup>16</sup> and electroluminescence<sup>17</sup> are possible in such Si nanocluster sensitized Er<sup>3+</sup>, it may open the way for a compact, active, all-silicon integrated optical devices.

However, a detailed description of the carrier–Er interaction including the characteristic interaction distance is still

lacking. Such a description is necessary, not only for designing the optimum material structure for device applications, but also for understanding the physical mechanisms underlying the Er<sup>3+</sup>–carrier interaction that is not yet fully understood. In this letter, we report on the Er<sup>3+</sup> luminescence properties of Er-doped *a*-Si/SiO<sub>2</sub> SL films whose Er-doped SiO<sub>2</sub> layer thickness was controlled with subnanometer resolution. We find that experimental data can be well-described by an Er–carrier interaction that decreases exponentially, with a characteristic carrier–Er<sup>3+</sup> interaction distance of 0.5±0.1 nm, indicating the likelihood of an exchange interaction between carriers and Er<sup>3+</sup> ions.

Er-doped *a*-Si/SiO<sub>2</sub> superlattices were deposited on Si substrates using the two-target-alternation UHV-ion-beam sputtering method. One sputtering target was a 6-in.-diameter *p*-type Si wafer, and the other was a similar Si wafer with an Er metal piece (99.9% purity) centered on the wafer. The base pressure of the system is less than 1 × 10<sup>−8</sup> Torr, and a 570-eV Ar<sup>+</sup> beam from an ion gun was used to sputter the targets. The Si layers were deposited by sputtering using the Ar<sup>+</sup> beam only, and the SiO<sub>2</sub> layers were deposited by sputtering in an oxygen pressure of 4.5 × 10<sup>−4</sup> Torr. The samples were deposited at room temperature. The deposited films consisted of 12 periods of *a*-Si/SiO<sub>2</sub>:Er/SiO<sub>2</sub>/SiO<sub>2</sub>:Er layers. The thicknesses of the *a*-Si layers were fixed at 1.2 nm. The thicknesses of the SiO<sub>2</sub>:Er (Er-doped SiO<sub>2</sub>) and SiO<sub>2</sub> (pure SiO<sub>2</sub>) layers was varied such that the SiO<sub>2</sub>:Er layer thickness increased from 0 to 4.1 nm, while the SiO<sub>2</sub> layer thickness decreased accordingly to keep the thickness of the total SiO<sub>2</sub> layer fixed at 9.5 nm. Such preparation is necessary to ensure that the overall film structure is the same for all films, and that the results will not be affected by optical artifacts caused by multiple reflections. Finally, a 5-nm-thick Si cap layer was deposited on the top. Two sets of samples, one with an Er concentration of 0.27 at. % and the other with 0.1 at. %, were prepared. After deposition, all 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 in a flowing N<sub>2</sub> atmosphere to avoid cracking and spalling of the films.

<sup>a)</sup>Electronic mail: jhs@mail.kaist.ac.kr

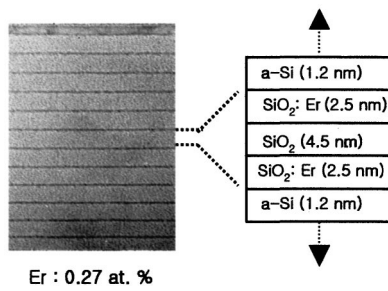


FIG. 1. Bright-field cross-section TEM image of the SL with Er-doped  $\text{SiO}_2$  layer thickness of 2.5 nm after annealing. The thin, dark bands are *a*-Si layers and gray bands are  $\text{SiO}_2$  layers. On the right is a schematic drawing showing the composition of the layers.

The  $\text{Er}^{3+}$  photoluminescence (PL) spectra were taken at room temperature using a grating monochromator, a thermoelectrically cooled InGaAs detector, and the standard lock-in technique. The 477-nm line of an Ar laser, which is not absorbed optically by  $\text{Er}^{3+}$ , was used as the excitation source to ensure that excitation of  $\text{Er}^{3+}$  occurs via carriers only. The excitation power was kept at 100 mW to remain in the weak pumping regime, as it was determined that  $\text{Er}^{3+}$  PL intensity increased linearly with the pump power up to 150 mW (not shown). Time-resolved  $\text{Er}^{3+}$  luminescence was measured using a digitizing oscilloscope. The structure and composition of the films were analyzed using transmission electron microscopy (TEM) and medium-energy ion scattering spectroscopy (MEIS). The MEIS analyses were done using a 100-keV  $\text{H}^+$  beam aligned to the [001] direction in the (011) plane.  $\text{H}^+$  ions scattered into the [111] direction were analyzed at a scattering angle of  $125^\circ$ . For MEIS analysis, a sample consisting of a single period of  $\text{SiO}_2:\text{Er}/\text{SiO}_2/\text{SiO}_2:\text{Er}$  layers with intended thicknesses of 2.2, 5, and 2.2 nm, respectively, was deposited and annealed under identical conditions.

Figure 1 shows the bright-field cross-section TEM image of a deposited and annealed SL film with 0.27 at. % Er and the schematic depiction of its intended structure. The dark and gray bands correspond to the Si and  $\text{SiO}_2$  layers, respectively. We find that the Si and the  $\text{SiO}_2$  layer thicknesses are 1.2 and 9.5 nm, respectively, in good agreement with the intended thicknesses. Note that the Si layers are amorphous despite the high-temperature annealing that crystallized the cap layer. This is consistent with reports by other researchers that nanometer-thick Si layers are very difficult to crystallize.<sup>18,19</sup>

The MEIS spectrum is shown in Fig. 2. The symbols are the experimental data, and the line is the result of the simulation. We find that thicknesses of  $\text{SiO}_2:\text{Er}/\text{SiO}_2/\text{SiO}_2:\text{Er}$  layers are 2.2, 5, and 2.2 nm, in an excellent agreement with the intended thicknesses. The Er concentration was 0.27 at. % for both  $\text{SiO}_2:\text{Er}$  layers. Taken together, Figs. 1 and 2 confirm that we can deposit *a*-Si/ $\text{SiO}_2:\text{Er}/\text{SiO}_2/\text{SiO}_2:\text{Er}$  layers with subnanometer control over the individual layers, while keeping the overall film structure the same.

Figure 3 shows the dependence of the  $\text{Er}^{3+}$  peak PL intensity from the sample with 0.27 at. % Er on the  $\text{SiO}_2:\text{Er}$  layer thickness. The inset shows that from the sample with 0.1 at. % Er. The symbols are the experimental data, and the lines are the results of the fit discussed later. We find that for

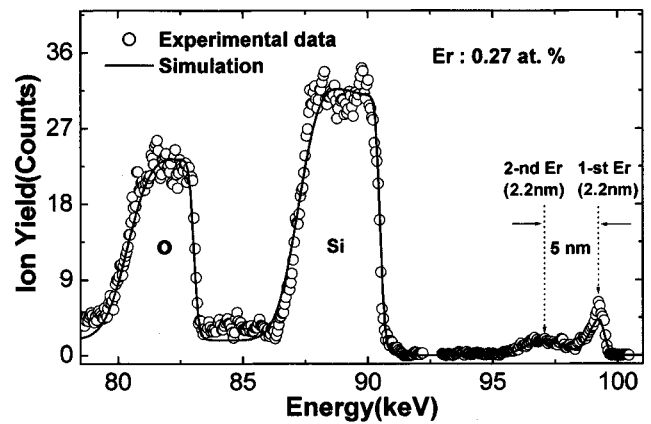


FIG. 2. MEIS spectrum for the  $\text{SiO}_2:\text{Er}/\text{SiO}_2/\text{SiO}_2:\text{Er}$  layers with thicknesses of 2.2, 5, and 2.2 nm, respectively. The symbols are experimental data and the line is the result of simulation.

both sets of samples, the  $\text{Er}^{3+}$  PL intensity increases with increasing  $\text{SiO}_2:\text{Er}$  layer thickness  $d$ , but saturates quickly after only about 1.5 nm, and remains constant thereafter.

Figure 4 shows the dependence of the  $\text{Er}^{3+}$  luminescence lifetimes on the  $\text{SiO}_2:\text{Er}$  layer thickness from the films with 0.27 at. % Er. We find that the  $\text{Er}^{3+}$  luminescence lifetime is independent of the  $\text{SiO}_2:\text{Er}$  layer thickness  $d$ , being about 0.6 ms for all films. A similar quantitative analysis on the films with 0.1 at. % Er was not possible due to the low  $\text{Er}^{3+}$  PL intensity. However, they, too, did not display any strong dependence of the  $\text{Er}^{3+}$  luminescence lifetimes on the  $\text{SiO}_2:\text{Er}$  layer thickness (data not shown).

Changes in the  $\text{Er}^{3+}$  PL luminescence intensity can occur due to changes in the excitation rate, the  $\text{Er}^{3+}$  luminescence efficiency, and the number of available  $\text{Er}^{3+}$  ions. In the present case, since the overall film structure is the same for all films, the generation rate of carriers that can excite  $\text{Er}^{3+}$  ions should be the same for all films. Furthermore, as Fig. 4 shows, the  $\text{Er}^{3+}$  luminescence efficiency is the same for all films as well. Thus, the increase in the  $\text{Er}^{3+}$  PL intensity with increasing  $\text{SiO}_2:\text{Er}$  layer thickness indicates that

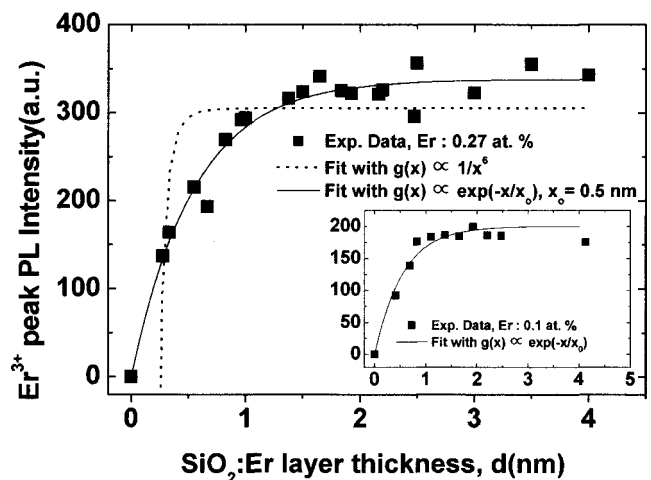


FIG. 3. The dependence of the  $\text{Er}^{3+}$  peak PL intensities on the  $\text{SiO}_2:\text{Er}$  layer thickness in Er-doped *a*-Si/ $\text{SiO}_2$  SLs. The symbols are experimental data and the lines are the results of fits using different carrier- $\text{Er}^{3+}$  interaction mechanisms. The inset shows the dependence of the  $\text{Er}^{3+}$  peak PL intensities on the  $\text{SiO}_2:\text{Er}$  layer thickness in Er-doped *a*-Si/ $\text{SiO}_2$  SLs containing 0.1 at. % Er and the line is the result of fit.

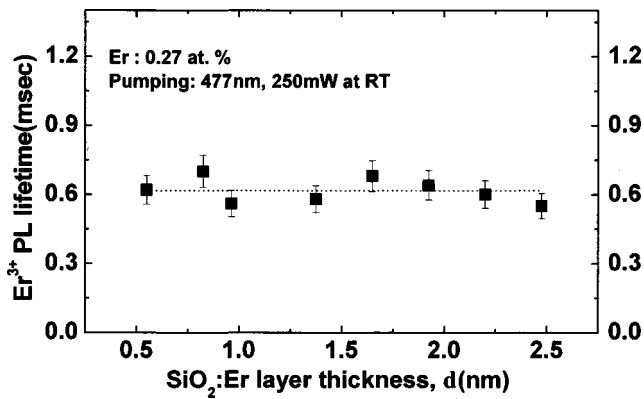


FIG. 4. The dependence of the  $\text{Er}^{3+}$  luminescence lifetimes on the  $\text{SiO}_2:\text{Er}$  layer thickness in  $\text{Er}$ -doped  $a\text{-Si}/\text{SiO}_2$  SLs containing 0.27 at %  $\text{Er}$ . The dotted line represents the average lifetime.

the number of available  $\text{Er}^{3+}$  ions is increasing, that is, excitation of  $\text{Er}^{3+}$  can occur over atomic distances and is not limited to  $a\text{-Si}/\text{SiO}_2:\text{Er}$  interfaces.

In such a case, we can write the excitation dynamics of  $\text{Er}^{3+}$  as

$$\frac{dN^*(x)}{dt} = g(x)(N_o - N^*) - \frac{N^*(x)}{\tau}, \quad (1)$$

where  $x$  is the distance from the  $a\text{-Si}$  layer,  $N^*(x)$  is the number of excited  $\text{Er}^{3+}$  at  $x$ ,  $g(x)$  is the distance-dependent carrier-mediated excitation rate,  $N_o$  is the number of excitable  $\text{Er}^{3+}$  ions at  $x$  (fixed by the  $\text{Er}$  concentration), and  $\tau$  is the decay rate of excited  $\text{Er}^{3+}$  ions that include all possible nonradiative decay mechanisms. In steady state,  $dN^*/dt = 0$ , and  $N^*(x) = [g(x)\tau N_o] / [g(x)\tau + 1]$ . However, in the linear, weak pumping regime,  $g(x)\tau \ll 1$ , and we can accurately approximate:<sup>4,5,20</sup>

$$N^*(x) \cong g(x)\tau N_o. \quad (2)$$

The total  $\text{Er}^{3+}$  intensity is  $\propto \int g(x)\tau N_o$ . Clearly,  $g(x)$  has to decrease with distance, and the monotonic increase of the  $\text{Er}^{3+}$  PL intensity to a saturation value indicates that  $g(x)$  decreases monotonically with distance. The two well-known interaction mechanisms involving carriers and emission centers such as  $\text{Er}^{3+}$  with such characteristics are resonant dipole–dipole interaction and the exchange interaction.<sup>21</sup> In the former, the interactions decreases as  $x^{-6}$ , while in the latter, the interaction decreases as  $\exp(-x/x_o)$ .<sup>22</sup> Thus, Eq. (2) was integrated using both forms for  $g(x)$ , and fitted to the data shown in Fig. 3. We find that the exponential form for  $g(x)$  with a value of  $0.5 \pm 0.1$  nm for  $x_o$  describes the data much better than the polynomial form. In fact, increasing the power of  $x$  to take into account the possibility of multipole interaction did not result in any improvement of the fit (not shown). We note that, as is shown in the inset, the result for the sample with 0.1 at. %  $\text{Er}$  can also be described very well with an exponentially decreasing interaction with the same  $x_o$ , further confirming the validity of the fit.

The value of  $0.5 \pm 0.1$  nm is quite small, indicating that the  $\text{SiO}_2:\text{Er}$  layers in an  $a\text{-Si}/\text{SiO}_2:\text{Er}/a\text{-Si}$  SL must be kept very thin—less than 2 nm—for maximum efficiency. Furthermore, the exponentially decreasing interaction indicates that an exchange interaction between carriers generated in  $a\text{-Si}$  and  $\text{Er}^{3+}$  ions in the  $\text{SiO}_2$  is much more likely to be

dominant than the direct, resonant dipole–dipole interaction between them. We note that Kimura *et al.*, using pre-oxidized porous  $\text{Si}$ , have come to a similar conclusion.<sup>23</sup> However, the value of  $x_o$  was much larger in his case (2–3 versus 0.5 nm). The reason for such differences is not yet clear at this moment. We note, however, that his film contained crystalline  $\text{Si}$  rather than amorphous  $\text{Si}$ . Such differences in the  $\text{Si}$  layers may significantly impact the carrier– $\text{Er}^{3+}$  interaction. Finally, we note that the characteristic carrier– $\text{Er}^{3+}$  interaction distance may also be dependent on other parameters, such as the  $\text{Er}$  concentration and the temperature. More detailed investigations are currently on the way.

In conclusion, we have investigated the carrier– $\text{Er}^{3+}$  interaction in  $a\text{-Si}/\text{SiO}_2:\text{Er}/a\text{-Si}$  superlattice films deposited by ion-beam sputtering. We find that the experimental results are well-described by exponentially decaying carrier– $\text{Er}$  interactions with a characteristic interaction distance of  $0.5 \pm 0.1$  nm, indicating the likelihood of an exchange interaction between carriers and  $\text{Er}^{3+}$ .

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