Modification of erbium radiative lifetime in planar silicon slot waveguides

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The authors report a systematic study of the lifetime of the 1.54 μm transition of Er^{3+}-doped SiO_2 thin film as active material in planar slot waveguides in polycrystalline silicon. The lifetime shows a strong reduction when compared with values measured in three other configurations. The experimental results, combined with a rigorous quantum-electrodynamical formalism, are consistent with a sizable increase in both the radiative and nonradiative decay rates of Er^{3+} transition in slot waveguide. The radiative efficiency is only slightly reduced with respect to Er^{3+} in the bulk oxide, this result being important for future realization of Si-compatible active optical devices. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3098072]

In the route toward the integration of photonics and silicon technology, a major and well established research field is represented by doping silicon with erbium ions to produce silicon-based optical sources.[1,2] In this respect, Er-doped SiO_2 has been shown to be a promising option to develop Si-based light emitting materials at the telecom wavelength (by exploitation of the 4-f transition at 1.54 μm of Er^{3+} ions) compatible with the complementary metal oxide semiconductor technology. A large amount of literature on modified Er^{3+} luminescence in confined structures exists;[3–8] however sizable radiative lifetime modifications are not easily demonstrated. Recently, slot configurations have been proposed to improve the waveguiding/confining properties of silicon-based optical waveguides.[9,10] In this structure, a very thin layer (few tens of nanometer thick) of SiO_2 (slot) is embedded between two silicon regions, which form the core of the optical waveguide. Planar (or horizontal) slot waveguides have been realized and experimentally studied in Refs. 11 and 12. Due to the high-index-contrast at the Si/SiO_2 interfaces, the electromagnetic (e.m.) field strongly concentrates in the narrow slot region, leading to an increase in vacuum e.m. field fluctuations. Thus an enhancement in the spontaneous emission (SE) rate is expected to occur similar to the Purcell effect of an emitter in an optical cavity,[13] as we demonstrate in the present work through theoretical analysis.

In this letter we present a detailed study of the photoluminescence (PL) decay rate of the 1.54 μm transition in Er^{3+} doped SiO_2 planar slot layers acting as active material in deposited polycrystalline silicon waveguides. We show that the confinement effects occurring in these structures considerably affect the lifetime of the erbium ions. In order to quantitatively evaluate such modification, we analyze the decay rates of Er-doped SiO_2 layers of the same thickness in three other configurations, namely, after deposition on SiO_2 and Si substrates and embedded in a SiO_2 bulk. By combining the radiative SE rates obtained by a quantum-electrodynamical formalism with time-resolved PL measurements, we evaluate nonradiative decay rates that are found to strongly increase in the slot waveguide.

A schematic of the four investigated structures is depicted in Fig. 1. Er-doped SiO_2 films, about $d_s=20$ nm thick, have been deposited by reactive cosputtering from SiO_2 and Er_2O_3 targets. The depositions have been realized in a reactive atmosphere (90% Ar and 10% O_2) by keeping the substrate heated at 300 °C. The films are located on a 1.9 μm thick thermal SiO_2 layer [structure (a)], within bulklike SiO_2 [structure (b)] by depositing a 550 nm thick SiO_2 layer on top of structure (a), on a (100) crystalline Silicon (c-Si) substrate [structure (c)], and within the silicon core of a slot waveguide [structure (d)]. The latter consists of Si(110 nm)/Er:SiO_2(20 nm)/ Si(110 nm) layers deposited on top of a 1.9 μm thick silicon dioxide layer thermally grown on a silicon substrate. The silicon layers have been realized by sputtering a Si cathode in a pure Ar atmosphere without heating the substrate. Structures (a) and (b) also lie on a Si substrate [not shown in Figs. 1(a) and 1(b) for simplicity]. After the deposition all the films have been annealed at 900 °C for 1 h in a nitrogen atmosphere. The Er content, measured by Rutherford backscattering spectrometry, is constant within the thin SiO_2 layers, with a concentration of $7.6\times10^{19}/cm^3$ for all the samples.

Time-resolved PL at room temperature was performed by pumping with the 488 nm line of an Ar^+ laser, the laser beam being chopped through an acousto-optic modulator at a frequency of 11 Hz. The modulated luminescence signal was

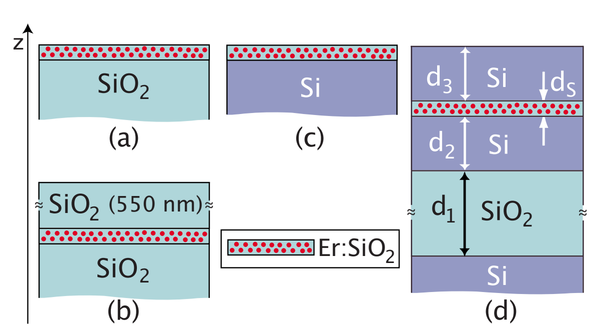

FIG. 1. (Color online) Schematic of the sample structures. Er-doped SiO_2 films $d_s=20$ nm thick on SiO_2 (a), on SiO_2 and with a 550 nm thick SiO_2 layer on its top (b), on a crystalline silicon substrate (c), and embedded in the core of a waveguide (d), with $d_1=1.9$ μm and $d_2=d_3=110$ nm.

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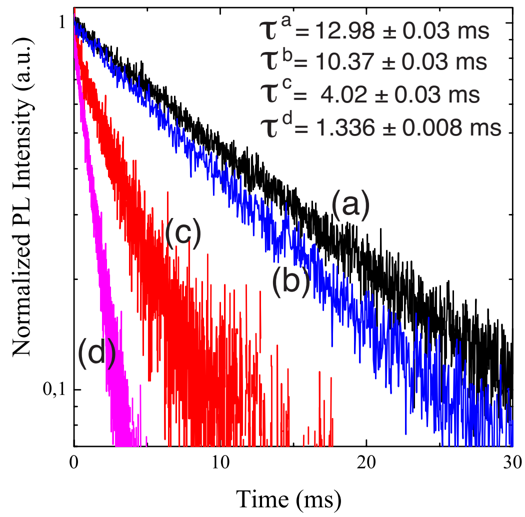

FIG. 2. (Color online) Erbium PL intensity decay curves (in log scale) for the structures depicted in Fig. 1. The intensities are normalized to their corresponding maxima.

first detected with a Hamamatsu infrared-extended photomultiplier tube and then analyzed with a photon counting multichannel scaler with an overall time resolution of 30 ns.

The PL decay curves at 1.5 μm (see Fig. 2) are all characterized by a single exponential behavior, the determined lifetimes being shown in the figure. The measured decay rate $\Gamma=1/\tau$ is enhanced with respect to Γ^a by a factor of $\Gamma^b/\Gamma^a=1.25$, $\Gamma^c/\Gamma^a=3.23$, and $\Gamma^d/\Gamma^a=9.71$. The lifetime τ^b of the Er-doped SiO_2 film embedded in SiO_2 [structure (b) in Fig. 1], i.e., the *bulk lifetime*, is similar to the value of 9.5 ms reported in Ref. 3 for films synthetized by e-beam deposition and of about 15 ms for thermally grown silicon oxide layer.[1,8]

The decrease in the observed lifetime τ^c relative to τ^a is consistent with an enhancement in the radiative SE rate. However, a full comprehension and a quantitative evaluation of the modification of the lifetime of Er^{3+} require the estimation of the radiative (Γ_{rad}) as well as the nonradiative (Γ_{nrad}) recombination rates since the measured decay rate $\Gamma=1/\tau=\Gamma_{rad}+\Gamma_{nrad}$ sums up both contributions.

The radiative decay rate Γ_{rad} is generally due to the emission into leaky and guided modes, $\Gamma_{rad}=\Gamma_{rad}^{leaky}+\Gamma_{rad}^{gui}$, the latter ones being supported by the slot waveguide only. For a quantum description, a proper treatment of modes with evanescent components is needed.[14–16] Γ_{rad}^{leaky} and Γ_{rad}^{gui} can be calculated by applying a quantum-electrodynamical formalism, which is particularly suitable to the analysis of the emission processes in multilayer dielectric structures, as detailed in Ref. 14. In this model the analytical expressions $\Gamma=\Gamma_\alpha^p(z)$ are derived as a function of the emitter position z and transverse electric (TE)/transverse magnetic (TM) field polarizations (p=TE, TM) and for both in-plane (or horizontally) oriented dipoles ($\alpha=\parallel$) lying along the dielectric planes and perpendicular (or vertical) ones ($\alpha=\perp$) oriented along the vertical z-direction. The SE rates into leaky modes are[14]

$$\Gamma_\alpha^p(z) = g_\alpha^p \sum_{j=U,L} \varepsilon_j^{3/2} \int_0^{\pi/2} |E_\alpha^p(k_\parallel = k_j \sin\theta, z)|^2 \sin\theta\, d\theta,$$

$$g_\parallel^{TE} = \frac{|\mathbf{d}|^2\omega_0^3}{2\hbar c^3}, \quad g_\parallel^{TM} = g_\parallel^{TE}\frac{c^2}{\omega_0^2[\varepsilon(z)]^2}, \quad g_\perp^{TM} = 2g_\parallel^{TM}, \qquad (1)$$

with $\mathbf{d}$ being the dipole matrix element for a dipole located at z, ω_0 its frequency, $\varepsilon(z)$ the dielectric constant in the considered dielectric layer, and the field amplitudes $E_\alpha^p(k_\parallel,z)$ are found by a standard-transfer matrix method after application of proper normalization conditions.[14] To calculate the emission into the lower ($j=L$) and upper ($j=U$) cladding layers with dielectric constants ε_L and ε_U, respectively, leaky modes have to fulfill the condition $k_\parallel < k_j = \sqrt{\varepsilon_j}\omega_0/c$, $k_\parallel$ being the in-plane component of the wave vector.

The emission rates into guided modes are given by

$$\Gamma_\alpha^p(z) = \tilde{g}_\alpha^p \sum_\lambda |E_\alpha^p(k_\parallel = k_{0_\lambda}, z)|^2 \frac{k_{0_\lambda}}{v_{0_\lambda}},$$

$$\tilde{g}_\parallel^{TE} = \frac{|\mathbf{d}|^2\pi\omega_0^3}{\hbar c^2}, \quad \tilde{g}_\parallel^{TM} = \tilde{g}_\parallel^{TE}\frac{c^4}{\omega_0^4}, \quad \tilde{g}_\perp^{TM} = 2\tilde{g}_\parallel^{TM}, \qquad (2)$$

where the sum extends over all the λ guided modes supported by the waveguide and $k_{0_\lambda}=k_\parallel^\lambda(\omega=\omega_0)$ and v_{0_λ} are the in-plane wave vector and group velocity of the λth guided mode evaluated at the dipole emission frequency ω_0, respectively. It is worthy to notice that since the Er^{3+} ions are always embedded in the same SiO_2 dielectric material, local field effects[17] are not relevant and thus have not been involved in the theoretical analysis.

By applying Eqs. (1) and (2) with the same parameters (thicknesses and refractive indices) of the four considered structures and for the realistic case of a randomly oriented Er^{3+} emitter in a SiO_2 layer,[18] the ratios between the radiative emission rates $\Gamma_{rad}=1/\tau_{rad}$ are thus found to be $\Gamma_{rad}^b/\Gamma_{rad}^a=1.4$, $\Gamma_{rad}^c/\Gamma_{rad}^a=2.4$, and $\Gamma_{rad}^d/\Gamma_{rad}^a=8.9$. The increase in radiative decay rate is of course more pronounced in the slot waveguide: the high-index-contrast that develops at the $Er:SiO_2/Si$ interfaces of the slot leads to a strong field confinement for TM modes due to the discontinuity of the z-component of the electric field and thus to an enhanced radiative emission as a result of the increase in the local density of optical states.[11] Furthermore, the separate contributions to the emission rate into leaky and guided modes can be estimated, as shown in Fig. 3 for Er^{3+} ions embedded in the tiny oxide layer of the slot waveguide [structure (d) in Fig. 1]. In such a configuration, the total radiative rate $\Gamma_{rad}^{gui}+\Gamma_{rad}^{leaky}$ is mainly due to the emission into guided modes (see the black solid line), with a calculated ratio $\Gamma_{rad}^{gui}/\Gamma_{rad}^{leaky}\approx 9$. It is interesting to establish the maximum enhancement in radiative rate that can be obtained in these slot waveguides. As the slot width goes to zero, the ratio $\Gamma_{rad}^d/\Gamma_{rad}^b$ grows toward the limiting value $(\varepsilon_{Si}/\varepsilon_{SiO_2})^2\simeq 33.25$ (ε_{Si} and ε_{SiO_2} being the dielectric constants of Si and SiO_2, respectively) for TM polarization and $\simeq 12$ when averaged over polarizations; thus the ratio $\Gamma_{rad}^d/\Gamma_{rad}^a$ averaged over polarizations tends to $\simeq 16$. For the present case of a 20 nm thick slot layer, the theoretical ratio is 8.9. Taking a thinner slot layer would not necessarily yield better results for radiative lifetime reduction both because the emitter's luminescence would be even weaker and nonradiative processes due to interaction with defects at slot interfaces would be enhanced.

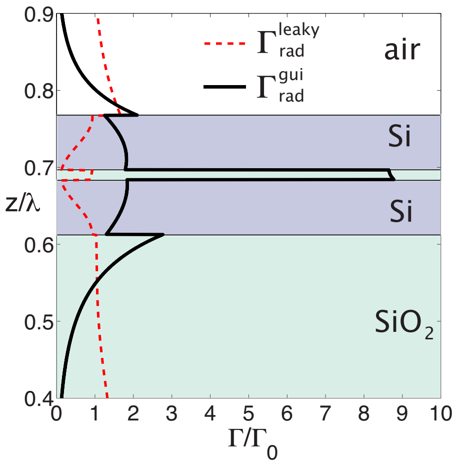

FIG. 3. (Color online) The normalized (with respect to the vacuum emission rate Γ_0) SE rates calculated as a function of the position z/λ of a randomly oriented erbium ion (Ref. 18) embedded in the slot waveguide. Solid black line: the emission rate into guided modes. Dashed red line: the emission rate into leaky modes. The refractive index of Si is taken to be $n=3.48$ and that of SiO_2 (both undoped and Er-doped) is $n=1.45$.

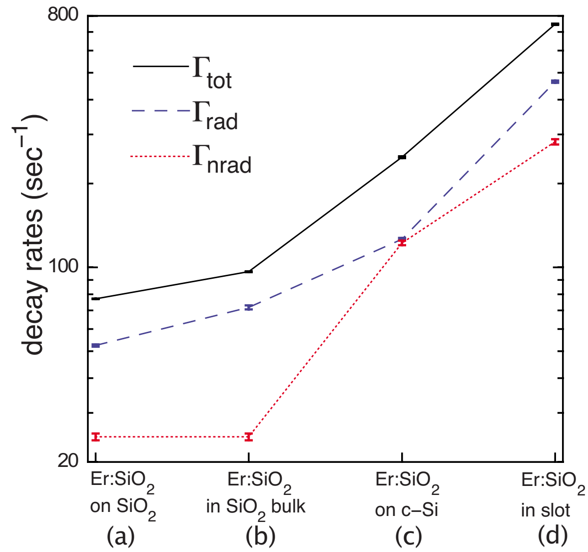

FIG. 4. (Color online) Erbium PL decay rates (in log scale). Continuous black line: the measured decay rates $\Gamma_{tot}=\Gamma_{rad}+\Gamma_{nrad}$. Dashed blue and dotted red lines: the radiative and nonradiative rates, respectively. The error bars for the experimental Γ_{tot} are those associated with the lifetimes derived from the PL decay curves (Fig. 2). The uncertainties in the radiative and nonradiative rates have been obtained by applying error propagation.

To disentangle the radiative and nonradiative contributions to the total emission rates, we use the measured PL decay rates as well as the theoretical ratios defined as $\beta_b^{a,c,d}=\Gamma_{rad}^{a,c,d}/\Gamma_{rad}^{b}$. The procedure is as follows.

First, the emission rates Γ_{rad}^{b} and Γ_{nrad}^{b} for Er-doped SiO_2 in bulk SiO_2 [structure (b) in Fig. 1] have been determined. Γ_{rad}^{b} and Γ_{nrad}^{b} are the solutions of the system given by (i) $\Gamma^a=\Gamma_{rad}^a+\Gamma_{nrad}^a=\beta_b^a\Gamma_{rad}^b+\Gamma_{nrad}^a$ and (ii) $\Gamma^b=\Gamma_{rad}^b+\Gamma_{nrad}^b\approx\Gamma_{rad}^b+\Gamma_{nrad}^a$, where $\Gamma^{a,b}=1/\tau^{a,b}$ are the SE rates obtained from the PL measurements and we have assumed $\Gamma_{nrad}^a\approx\Gamma_{nrad}^b$. This is very likely since the nonradiative decay is known to be due to short-range Förster energy transfer between the Er^{3+} transition and the resonant OH groups within the Er-doped film,[1] which is exactly the same in structures (a) and (b). Furthermore, once Γ_{rad}^{b} and Γ_{nrad}^{b} are known, the radiative efficiency for $Er:SiO_2$ in SiO_2 bulk has been estimated to be $q=\Gamma_{rad}^b/(\Gamma_{rad}^b+\Gamma_{nrad}^b)=0.74$, this value being close to that determined in Ref. 8 on similar structures. Next, the radiative emission rates $\Gamma_{rad}^{a,c,d}$ for structures (a), (c), and (d) are found as $\Gamma_{rad}^{a,c,d}=\beta_b^{a,c,d}\Gamma_{rad}^b$. The nonradiative rates $\Gamma_{nrad}^{a,c,d}$ are then given by the differences $\Gamma^{a,c,d}-\Gamma_{rad}^{a,c,d}$, $\Gamma^{a,c,d}=1/\tau^{a,c,d}$ being the decay rates experimentally found. All the decay rates are shown in Fig. 4; it is clearly seen that as the average refractive index at the Er-doped SiO_2 layer increases (see the structures from left to right), so does the radiative decay rate of Er^{3+} transition and a strong lifetime reduction does occur when the layer is embedded within the slot waveguide. Moreover, also the nonradiative decay rate increases in the Er-doped SiO_2 on c-Si and in the slot waveguide structures [(c) and (d) in Fig. 4]. This is due to interaction of Er^{3+} transition with surface defect states at the SiO_2/Si interfaces and to Auger quenching with free and bound carriers in silicon. However, the radiative quantum efficiency is only slightly smaller than in bulk SiO_2 [about 67% in structure (c) and 62% in structure (d)] so that radiative recombination is still the dominant process in the slot waveguide.

In conclusion, by using a quantum-electrodynamical formalism we have shown that the radiative lifetime of Er^{3+} is strongly reduced when the SiO_2 layer is embedded within the core of a silicon-based optical waveguide. This result is the analog of the Purcell effect in an optical cavity and is a useful step toward achieving gain and stimulated emission in these structures. Furthermore, by combining the theory and time-resolved PL measurements, we have determined the nonradiative contributions to the total decay rate, showing that radiative decay as modified by the slot effect is still the dominant decay mechanism.

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[18]For a randomly oriented dipole, the contributions Γ_{rad}^{leaky} and Γ_{rad}^{gui} to the total emission rate are given by the averaged rates $\Gamma=(2/3)\Gamma_{\parallel}+(1/3)\Gamma_{\perp}$, where $\Gamma_{\parallel}=\Gamma_{\parallel}^{TE}+\Gamma_{\parallel}^{TM}$ is the sum over the two polarizations for a planar (horizontal) dipole, while $\Gamma_{\perp}=\Gamma_{\perp}^{TM}$ for a vertical dipole (which can couple to the TM modes only).