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The Auger process of luminescence quenching in Si/Si:Er multinanolayers

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Abstract

Energy transfer to free carriers in an Auger process is well known to hamper emission of rare-earth dopants in semiconductors. In particular, this process limits the excitation mechanism and is partly responsible for the thermal quenching of the $\sim$1.5 μm photoluminescence from Er^{3+} ions embedded in the crystalline silicon matrix. In this contribution, we investigate the excitation cross section and the free-carrier Auger process in Er-doped silicon multinanolayer structures. This novel Si-based material has recently been shown to exhibit very interesting properties as regards photonic applications.

1. Introduction

In the past decade Si:Er has been intensively investigated with a view to emission optimization [1]. The research revealed disadvantages of this system, which could not be fully eliminated: a strong emission reduction at higher temperatures and a low percentage of optically active Er-related centres. Thermal quenching is absent for photoluminescence (PL) of Er ions embedded in a SiO_2 matrix, but this system, in turn, is characterized by a very small cross section of (direct) excitation. In view of that, a nonhomogeneous medium of Si nanocrystals (nc-Si) dispersed in a SiO_2 matrix was recently investigated as an alternative host for Er doping. In this case, the large bandgap of SiO_2 provides thermal stability for the Er emission, while Si nanocrystals facilitate efficient excitation [2–4]. Following this approach, development of room-temperature-operating LEDs was proven to be possible. However, recent results indicate that population inversion might be difficult to reach in this system [5].

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Recently a new type of Si:Er optical structure has been proposed [6]. The idea follows from a notion that since the most efficient low temperature optical excitation mechanism of Er proceeds via excitons, the generation conditions for excitons should be optimized. The requirements of a high Er^{3+} ion concentration and efficient exciton generation cannot be met simultaneously. Therefore for heavily Er-doped layers excitons are generated in the substrate rather than in the layer itself. This is evidenced by the fact that the PL intensity does not increase above a certain thickness of a Si:Er layer. It does increase, however, when a spacer of undoped Si is inserted into the Si:Er layer. Therefore, a sandwich structure of interchanged Si/Si:Er nanolayers exhibits superior optical properties. Upon illumination with a laser beam, excitons generated in spacer regions diffuse into doped layers and provide excitation of Er^{3+} ions. Such a spatially separated excitation scheme is in its concept somewhat similar to that utilized in the earlier mentioned inhomogeneous SiO_2/nc-Si:Er samples, where processes of photon absorption (nc-Si) and emission (Er^{3+} ions in the SiO_2 matrix) also take place in different spatial regions in the sample. In addition to the more efficient exciton formation, spatial separation could influence Auger quenching by free carriers generated during the excitation process. In this contribution we address the issue of Auger quenching of Er-related PL in Si/Si:Er nanolayer structures.

2. Experimental approach

The experiments were performed on an Er-doped silicon sample grown by sublimation MBE. The sample comprised 400 interchanged Si and Si:Er layers of a few nanometres thickness stacked along the ⟨100⟩ growth direction. The total Er density was 2×10^{14} cm^{-2}, with the volume concentration of [Er] $\approx 2 \times 10^{18}$ cm^{-3} in the active layer.

The PL experiments were carried out in a variable temperature continuous-flow cryostat accessing the 1.5–300 K range (Oxford Instruments Optistat CF). The samples were excited using a tunable optical parametric oscillator (OPO) laser, producing pulses of 5 ns duration at 20 Hz repetition rate. The luminescence was resolved with a 1 m F/8 monochromator (Jobin-Yvon THR-1000) equipped with a 900 grooves mm^{-1} grating blazed at 1.5 μm and detected by an infrared photomultiplier with a 30 μs response time.

3. Results and discussion

Figure 1 shows the PL spectrum of the sample in the 1.5 μm range at 4.2 K under excitation with the OPO set to a wavelength of $\lambda_{\mathrm{exc}} = 520$ nm. It has been shown before [7, 8] that this spectrum is characterized by ultranarrow emission bands. This is illustrated in the inset to the figure. For determination of the excitation cross section, the excitation power dependence of the PL intensity was measured at 4.2 K—see figure 2, for the three major components of the spectrum marked in figure 1: line L_1^1 (6502.8 cm^{-1}), L_2^1 (6443.7 cm^{-1}) and L_3^1 (6433.6 cm^{-1}), under pulsed excitation with the OPO. All the measurements were performed with the same experimental settings, so the PL intensity scale is common to all the data points. As can be seen, the behaviour of these lines is similar, consistent with the conclusion that these lines originate from the same Er-related centre [7, 8].

To obtain quantitative information from the pulsed laser measurements, we use the following rate equation, involving the carrier density n, the density of excitons n_{ex} and the concentration of excited Er centres N_{Er}^*:

$$\frac{\mathrm{d}N_{\mathrm{Er}}^*}{\mathrm{d}t} = \sigma\Phi\left(N_{\mathrm{Er}}^{\mathrm{ex}} - N_{\mathrm{Er}}^*\right) - \frac{N_{\mathrm{Er}}^*}{\tau}, \qquad \sigma = \alpha c_{\mathrm{A}} \tau_{\mathrm{ex}}.$$

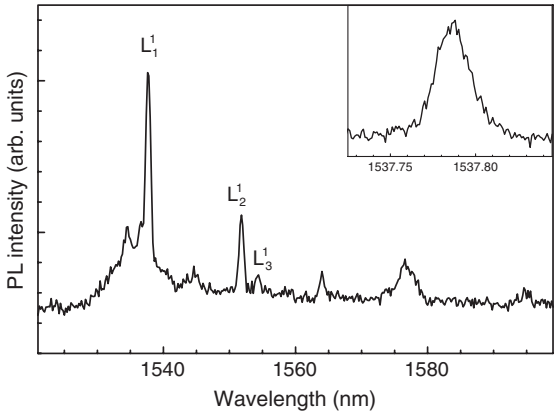

Figure 1. Photoluminescence spectrum of the multilayer sample used in the current study as obtained at $T = 4.2$ K under pulsed excitation with the OPO set to $\lambda_{exc} = 520$ nm. In the inset: the ultranarrow linewidth of the main emission band L_1^1 measured at $T = 4.2$ K under cw excitation with an Ar^+ ion laser ($\lambda_{exc} = 514$ nm).

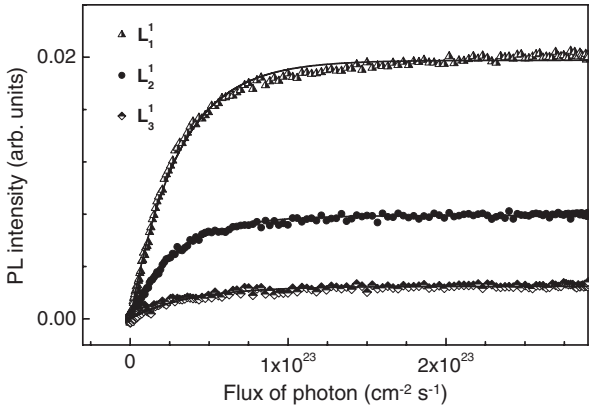

Figure 2. PL intensity dependence on the excitation photon flux for the three major lines L_1^1, L_2^1 and L_3^1 measured at $T = 4.2$ K under pulsed excitation with the OPO set to $\lambda_{exc} = 520$ nm.

N_{Er}^{ex} is the total concentration of excitable Er^{3+} ions present in the sample, α is the absorption coefficient of silicon, c_A is the coefficient of capture of free excitons by erbium-related centres with erbium excitation, τ_{ex} and τ are the effective lifetimes of the exciton and Er^{3+} in the excited state, respectively. The exciton lifetime, τ_{ex}, is controlled mainly by nonradiative Auger processes associated with the presence of Er-related donors or other impurities. We assume that the binding of free carriers into free excitons dominates at low temperatures. The decay time τ corresponds to an effective lifetime of Er^{3+} ions in the excited state due to both radiative and nonradiative recombinations:

$$\frac{1}{\tau} = \frac{1}{\tau_{rad}} + c_{A,Er}n,$$

where τ_{rad} is the time for decay of excited Er centres by the radiative transition and $c_{A,Er}$ is the coefficient of the Auger process of nonradiative recombination of excited Er^{3+} ions. In that way the proposed description concerns concentration of excited Er centres with two possible de-excitation paths: radiative emission with the spontaneous emission time τ_{rad} and

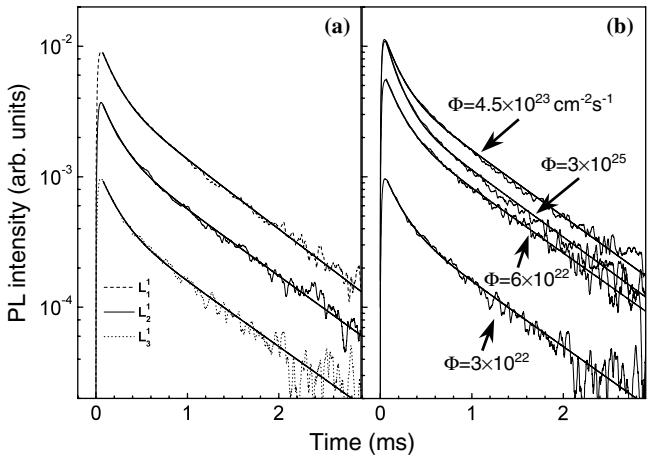

Figure 3. PL decay dynamics measured at $T = 4.2$ K under pulsed excitation with the OPO set to $\lambda_{exc} = 520$ nm for (a) the three major lines L_1^1, L_2^1 and L_3^1 with $\Phi = 3 \times 10^{22}$ cm^{-2} s^{-1} photon flux and (b) the L_1^1 line at several different photon flux values.

the Auger quenching process which is proportional to the density of free carriers. In the present experiment, the duration of the OPO pulse ($\Delta t = 5$ ns) is much shorter than the characteristic lifetime τ of Er^{3+} in the excited state ($\Delta t \ll \tau$). We assume that recombination does not take place during illumination and the population N_{Er}^* reaches the level of

$$N_{Er}^*(t = \Delta t) = N_{Er}^{ex}[1 - \exp(-\sigma \Phi \Delta t)].$$

For low excitation density, when $\sigma \Phi \Delta t \ll 1$, this formula gives a linear dependence on the flux: $N_{Er}^* = \sigma \Phi N_{Er}^{ex} \Delta t$. When $\sigma \Phi \Delta t \gg 1$, the saturation regime can be obtained: $N_{Er}^* = N_{Er}^{ex}$. Since in the experiment the PL signal is integrated in time, and the PL intensity is proportional to N_{Er}^*/τ_{rad}, the result of the experiment is given by $N_{Er}^* \tau/\tau_{rad}$. By fitting the data depicted in figure 2 for the three PL lines to the photon flux dependence, we obtain the excitation cross section of Er^{3+} ions in the nanolayer structure investigated as $\sigma = (6 \pm 2) \times 10^{-15}$ cm^2. This value is in a good agreement with that reported previously from cw laser measurements on Er-implanted silicon [9]. While the physical interpretation of the experimentally determined 'effective' excitation cross section is not straightforward, this result indicates that Er^{3+} ions in Si/Si:Er nanolayers have a similar excitation mechanism to that of other Er-related optically active centres in Si, e.g. in samples prepared by ion implantation.

We have also examined decay characteristics of the PL. This is shown in figure 3(a) for a measurement performed at 4.2 K under OPO excitation with a wavelength of $\lambda_{exc} = 520$ nm with a photon flux $\Phi = 3 \times 10^{22}$ cm^{-2} s^{-1}. As can be seen, under these conditions the decay kinetics is composed of a fast and a slow component. Fitting two exponentials to the measured profiles, we obtained 1/e decay times of $\tau_F = 150$ μs and $\tau_S = 900$ μs contributing to the signal for all the four lines. The intensity ratio of the fast and slow components is found to be 1:1, the same for the three lines.

The presence of two components in the decay kinetics could indicate the formation of two different Er-related centres. In order to examine this possibility, we separated the PL spectra of the fast and the slow components by integrating the signal over time windows of $t \in [0, 100\,\mu s]$ and $t \in [100\,\mu s, 4$ ms], respectively. The two spectra were found to be identical, regardless of the different decay time constants. Taking into account the very small linewidth of the PL lines investigated, we conclude that the possibility of coexistence of two different Er-

related centres is not consistent with our findings. By looking at the decay kinetics at different excitation densities, we find that the intensity ratio of the fast and slow components increases with the laser power. While this is still under investigation, such an observation suggests that two different de-excitation processes take place for the same centre. The slow component is likely to represent the radiative decay time of Er^{3+} ion, which is generally assumed to be in the millisecond range. The fast component could be induced by an Auger process with free carriers generated by the excitation pulse.

In figure 3(b) the decay characteristic of the PL intensity at $\lambda = 1.538$ μm (line L_1^1) for $\lambda_{exc} = 520$ nm is compared for selected values of the photon flux: $\Phi = 3 \times 10^{22}$, 6×10^{22}, 4.5×10^{23} and 3×10^{25} cm^{-2} s^{-1}. The ratios of the PL intensities of the fast and slow components increase with the flux of photons and are determined, respectively, as (1:1), (1.3:1), (1.5:1), (2.2:1). At higher flux of photons the fast component is prominent. This is consistent with a clear PL quenching observed for very high flux values and, again, points to a possible Auger process with free carriers which could become significant at high excitation density, as its origin.

4. Conclusion

On the basis of this initial study, we conclude that the ultranarrow Er-related emission bands from Si/Si:Er nanolayers are characterized by the $\sigma \approx 10^{-15}$ cm^2 excitation cross section. We also find evidence for PL quenching by a free-carrier mediated Auger process. Both findings are typical for Er^{3+} ions in crystalline Si, and therefore cannot provide an explanation for the special optical properties of the Si/Si:Er nanolayers—high intensity and ultranarrow linewidth of the Er-related PL.

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