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Photoluminescence mechanism of erbium in silicon

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In a recent study, a physical model for the photoluminescence of erbium in silicon accounting for the energy transfer processes has been theoretically developed. The model considers the formation of free excitons, the binding of excitons to erbium ions, the excitation of 4f electrons of erbium ions and their subsequent decay by light emission. The model yields predictions for the dependence of luminescence intensity on laser excitation power and temperature. When compared with experimental observations a good quantitative agreement has been obtained. The possible mutual relation between near band-edge and erbium luminescence is presented and discussed.

1. Introduction

Over the last decade, great attention has been given to the investigation of the luminescent Er-doped-Si system. The eminent suitability of the Er light as a carrier for signal transport via glass fibers is a stimulation for such research from the application point of view. The energy transfer mechanism, which is important to understand the thermal quenching of light emission and optical efficiency of Er in Si, is a challenging topic in semiconductor physics. In this report a chain of excitation and de-excitation processes is analyzed in a physical model and compared with experimental data on excitation power and temperature dependence of photoluminescence (PL) intensity with an aim of achieving a quantitative description. At temperatures lower than 100 K the mutual transition between band-edge and erbium luminescence is observed. The origin of this effect is discussed.

2. Model and discussion

In the model the overall process of energy transfer between Er ion and Si crystal is considered. The process starts with the formation of electron-hole pairs by the incident light, at a rate G, followed by a free exciton creation with the rate $\gamma_x n^2$. Free excitons can be bound at Er-related traps, which will occur proportional to their concentration n_{Er} and the available unoccupied fraction $[(n_{Er}-n_{xb})/n_{Er}]$, where n_{xb} denotes concentration of excitons trapped at Er-related centers. Er-bound excitons can transfer their energies in an Auger process with a transfer time τ^* to the 4f electrons of an Er ion in the ground state, i.e., proportional to fraction $[(n_{Er}-n_{Er}^*)/n_{Er}]$. The Er PL follows from decay of the excited Er ions with radiative decay time τ_d: $I \propto n_{Er}^*/\tau_d$. Competing recombination paths included in the model are the electron-hole recombinations with the rate γn^2, the direct recombination of free excitons or via alternative centers with the lifetime τ_x and the thermally induced dissociation of Er-bound excitons. Two Auger processes. which remove energy from the PL path, are also accounted for in the competing paths. They are related to Er-bound excitons and excited Er ions, which dissipate their energies to free electrons in the conduction band. Based on the above discussion the following balance equations can be written for free electrons, free excitons, bound excitons and excited Er ions, respectively:

$$G + fn_x = \gamma_x n^2 + \gamma n^2, \quad (1)$$

$$\gamma_x n^2 + cf_{xb}n_{xb}N_x = cn_x n_{Er}[(n_{Er} - n_{xb})/n_{Er}] + fn_x + n_x/\tau_x, \quad (2)$$

$$cn_x n_{Er}[(n_{Er} - n_{xb})/n_{Er}] + n_{Er}^* f_1/\tau^* = n_{xb}[(n_{Er} - n_{Er}^*)/n_{Er}]/\tau^* + cf_{xb}n_{xb}N_x + c_{Ax}nn_{xb} \quad (3)$$

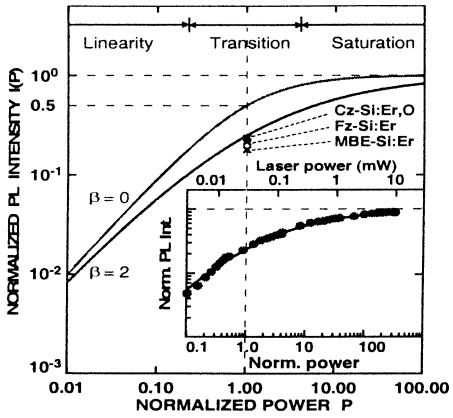

Fig. 1: Normalized PL intensity I as a function of normalized laser power P with $\beta = 0$ and $\beta = 2$. The experimental points (×) for P = 1 are given for three samples. The insert shows the Er PL versus excitation power with the solid line as the best fit to Eq. 6 for sample Cz-Si:Er,O.

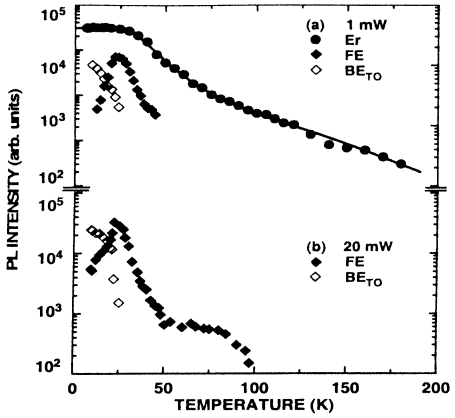

Fig. 2: Temperature dependence of the Er and the band-edge luminescence measured for sample Cz-Si:Er,O; (a) at laser excitation power of 1 mW, (b) at laser excitation power of 20 mW.

and

$$n_{xb}[(n_{Er} - n_{Er}^*)/n_{Er}]/\tau^* = n_{Er}^*/\tau_d + n_{Er}^* f_1/\tau^* + c_{AEr} n n^*_{Er}, \quad (4)$$

where $f = \gamma_x(N_c N_v/N_x)\exp(-E_x/kT)$, $f_{xb} = \exp(-E_{xb}/kT)$ and $f_1 = \exp(-E_A/kT)$; E_x, E_{xb} and E_A are the binding energies of electron and hole in a free exciton, the exciton binding energy on the Er-related trap and the energy dissipated in the creation of an excited Er ion from the bound exciton situation, respectively. N_c, N_v and N_x are $T^{3/2}$-temperature-dependent densities of states in conduction, valence and exciton bands, respectively.

2.1 Excitation power dependence

The power dependence of the PL intensity was measured at low temperature. In this case we can simplify the equations by putting $f = f_{xb} = f_1 = 0$. Solving Eqs (1-4), an approximate solution for n_{Er}^*/n_{Er} is derived as

$$n_{Er}^*/n_{Er} = c_2 G/(b_0 + b_1 G^{1/2} + b_2 G + b_3 G^{3/2}), \quad (5)$$

where b_0, b_1, b_2, b_3 and c_2 are temperature-independent factors. In the high-power limit this result yields $n_{Er}^* \sim 1/G^{1/2}$ or $n_{Er}^* \sim 1/n$ resulting in a decrease of PL intensity. This decrease has not been observed in the present experiments, nor has it been reported in the literature. A numerical estimate [1] shows that such an effect should correspond to high values of G, about 10^{26} $cm^{-3}s^{-1}$, which are not reached in actual experiments. For this reason, under the usual experimental conditions, a linear increase at low power with $(c_2/b_0)G$ is observed, followed by saturation at the level c_2/b_2. Using normalized units for power $P \equiv G/G_1$ with $G_1 = b_0/b_2$ and for PL intensity $I \equiv (n_{Er}^*/n_{Er})/(c_2/b_2)$, the solution (5) will read

$$I = P/(1+\beta\sqrt{P}+P) \quad (6)$$

with

$$\beta = \sqrt{c_{AEr}\tau_d/c_{Ax}\tau^*} + \sqrt{c_{Ax}\tau^*/c_{AEr}\tau_d} \geq 0. \quad (7)$$

The PL has its linear increase at low power with I = P and saturates at high power at I = 1. Therefore, characteristic features of the photoluminescence process are revealed at intermediate power P = 1, where $I = 1/(2+\beta)$. In Fig. 1 theoretical curves for $\beta = 0$ and 2 are presented. The power dependence of PL intensity has been measured. In the study three different kinds of samples have been used. The first sample was prepared from Czochralski

silicon by Er and O implantation (Cz-Si:Er,O) to a peak Er concentration of 10^{17} cm^{-3}, followed by annealing for 0.5 hour at temperature 900^0C. The second sample was float-zoned silicon, which was implanted with Er (Fz-Si:Er) to a peak Er concentration of 4×10^{17} cm^{-3}. The third sample was a crystalline silicon layer of 2 μm thickness, which was grown by a MBE method, followed by an annealing at 700^0C for 30 minutes (MBE-Si:Er). The sample had an Er concentration of 10^{18} cm^{-3}. The PL intensity of sample Cz-Si:Er,O as a function of Ar^+-laser power is plotted in the insert. The solid line is the best fit to the data with Eq. 6, yielding $\beta = 2.25$. Similar fits made for samples Fz-Si:Er and MBE-Si:Er gave $\beta = 2.63$ and 3.3, respectively. With Eq. 7 this result is converted to $(c_{AEr}\tau_d/c_{Ax}\tau^*)^{\pm1} \approx 4$ for the implantation samples and ≈ 9 for the MBE sample. This can be compared with the published data for $c_{AEr} = 10^{-12}$ cm^3s^{-1}, $\tau_d = 10^{-3}$ s, $c_{Ax} = 10^{-10}$ cm^3s^{-1} and $\tau^* = 4\times10^{-6}$ s [2]. One concludes that $c_{AEr}\tau_d/c_{Ax}\tau^*$ is very similar in all kinds of investigated material. Although this can be an accidental combination of parameters, one is tempted to believe that all process parameters, i.e., c_{AEr}, τ_d, c_{Ax} and τ^*, have similar values. In such a case the differences in structure of the luminescent centers in the three materials, as evidenced by their PL spectra, have little influence on the efficiency of the Er PL process.

2.2 Temperature dependence

For an analysis of the Er PL temperature dependence, the set of Eqs (1-4) is considered in its complete form. The solution of n_{Er}^*/n_{Er} is obtained also in the form of Eq. (5), but in this case the coefficients are temperature dependent as they include the state densities in conduction, valence and exciton bands and the functions f, f_{xb} and f_1. At low temperature Eq. (5) will give the solution $(n_{Er}^*/n_{Er})_{T=0}$. For the practical purpose of comparing model predictions with experimental results it is useful to express measured intensities normalized to the low temperature value, i.e., $(n_{Er}^*)_T/(n_{Er}^*)_{T=0}$. After some approximations one can obtain a final solution

$$(n_{Er}^*)_T/(n_{Er}^*)_{T=0} = 1/\{1+AN_x\tau^* \exp[-E_{xb}/kT] + BN_x\tau_d \exp[-(E_{xb}+E_A)/kT)]\} \quad (8)$$

with $A \approx B$. Note that the binding energy of an electron-hole pair in a free exciton E_x does not appear in the solution. Fig. 2(a) represents experimental data for the sample Cz-Si:Er,O, measured at an excitation power of 1 mW with the solid line as the best fit to Eq. (8). Two activation energies are necessary to fit the data. The fit is performed using a $T^{3/2}$ factor in the pre-exponents. In the literature, however, analyses with temperature-independent pre-exponential factors prevail. The first activation energy is identified as the binding energy, $E_{xb} = 15.6$ meV, of an exciton at the Er-related trap. At temperatures above 100 K the second energy of 120 meV, becomes significant. Following our analysis, this energy corresponds to $E_{xb} + E_A$, leading to the result $E_A \approx 100$ meV. By the physical model E_A is given as $E_A = E_g - E_x - E_{xb} - E_{PL} - E_d$. With estimates for silicon band gap energy $E_g = 1170$ meV, electron-hole binding energy $E_x = 15$ meV, Er-bound exciton energy $E_{xb} = 15$ meV, PL energy $E_{PL} = 800$ meV, a donor ionization energy of $E_d \approx 240$ meV is required. The ratio of magnitude between two pre-exponents obtained from the fit is $575/0.46 \approx 10^3$ in agreement with the estimated value $\tau_d/\tau^* \approx 10^3$.

2.3 Relation between erbium and silicon band-edge photoluminescence

Investigating the temperature dependence of Er PL intensity in the temperature range from 4 to 30 K evidence for the participation of excitons bound to shallow impurities of Si in the excitation process of Er ions has been experimentally found. A small growth of the Er PL intensity when increasing the temperature was observed. For the same temperatures in the wavelength region corresponding to the band-edge luminescence of silicon there was a transition from excitons bound to shallow impurities of the silicon to free excitons.

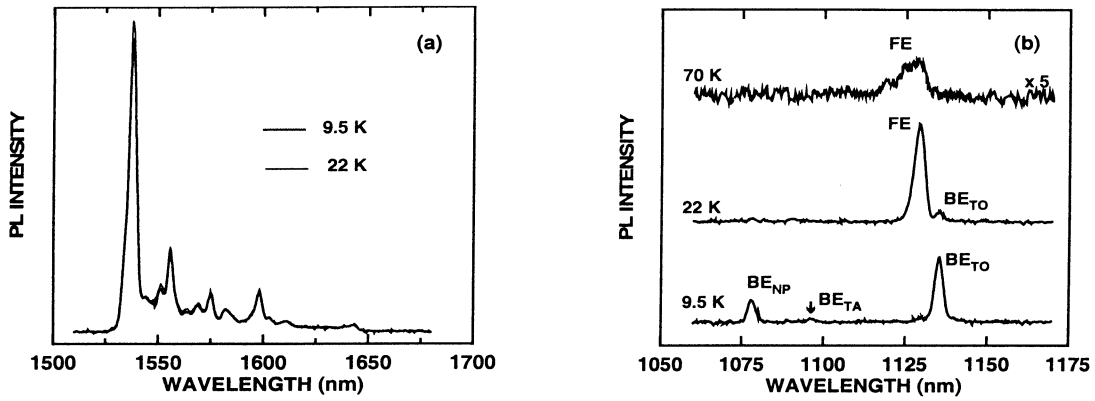

Fig. 3: PL spectra measured for the Cz-Si:Er,O sample; (a) Er PL at temperatures of 9.5 and 22 K, (b) silicon band-edge PL at temperatures of 9.5, 22 and 70 K. The 514.5 nm line of the Ar^+ laser was used for excitation.

Figs 3(a,b) show the Er and the band-edge PL spectra, respectively, which were measured for the Cz-Si:Er,O sample at several temperatures. The increase of the Er PL with increasing temperature could be explained by the mutual relation between shallow traps and deeper luminescent centers, where thermally induced energy transfer from impurity traps to Er-related centers was happening. The excitons bound to shallow-impurity dopants of silicon (BE_{TO}, λ_{BE}=1135.3 nm) were dissociated to free excitons (FE, λ_{FE}=1129.6 nm) with free-to-bound binding energy of about 3 meV [3]. The excitons freed from the shallow traps become available for capture by other, deeper, excitonic and luminescent traps; in this case these can be the Er-related luminescent centers. Consequently a rise in Er PL is observed when the temperature was raised up to about 25 K. At higher temperatures free excitons and excitons bound to Er centers are thermally dissociated, the Er PL is quenched as discussed in the previous section. The mutual relation between the Er and band-edge luminescence is presented in Fig. 2. The PL intensity of the bound excitons decreased below the detection level at a temperature of about 25 K. At temperature of about 25 K the PL intensities of Er and free excitons reached maximum values and then gradually decreased. The shape of the free-exciton line is broadened at higher temperatures and the intensity was diminished at a temperature of about 45 K for low excitation power. In Fig. 2(b) the PL intensity of the free-exciton line was relatively increased at high excitation power, 20 mW, when the bound-exciton concentration near the saturation region was achieved. At high temperatures when all excitons bound to shallow impurities had dissociated an interesting feature has been found. The free exciton luminescence is still clearly seen at temperature 70 K, as illustrated in Figs 2(b) and 3(b), then vanished at a temperature of about 100 K. This effect is explained by the dissociation of excitons bound to deep centers. The Er PL is not increased at these temperatures because of the thermal dissociation of Er-bound excitons, which is effectively active at temperatures up to 100 K. For this reason the presence of free excitons at high temperature could be the thermal release of Er-bound excitons.

In conclusion, the adequate description of the excitation power dependence of the PL requires inclusion in the model of Auger processes for decay of Er-bound excitons and erbium ions in their excited state. The thermal dependence is governed by the binding energy of excitons to Er centers in the temperature range below 100 K. At higher temperatures, the activation energy associated with the transfer energy from an excited Er ion back to an Er-bound exciton becomes more prominent. The shallow impurities of the silicon host have a contribution in the excitation process. The optical properties are found to be similar for all Er-doped crystalline samples.

References

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