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ERBIUM IN SILICON: POSSIBLE LIGHT SOURCE FOR 1.5 μm AND CHALLENGE FOR DEFECT PHYSICS

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The trivalent erbium ion emits at 1.54 μm , independent of the host crystal and temperature. This fact makes Si:Er an interesting candidate for integrated optics in the optimum wavelength regime for fiber optic communication systems. Recent progress in improving the luminescence yield is reviewed and the limiting factors are discussed, namely: the low solubility of different Er centers, thermal quenching of the luminescence above 100 K, the mechanisms for energy transfer from the host crystal to the Er 4*f* shell and the process induced parasitic recombination channels.

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1. Introduction

Rare earth (RE) elements have an incompletely filled 4*f* shell, which is well screened by the outer, closed 5*s*² and 5*p*⁶ shells. When incorporated as a dopant in a semiconductor, the host crystal acts only as a weak perturbation of the free ion energy levels and the electronic states couple very weakly to lattice vibrations. Hence, internal excitations of the 4*f* shell result predominantly in radiative transitions, with extremely narrow linewidths and temperature independent wavelengths. Therefore, rare earth ions are widely used as luminescence activators in wide band gap crystals and glasses, with the desired wavelength controlled by a suitable choice of the particular RE ion.

The recent development in fiber optics communication systems has attracted interest in infrared light sources emitting close to 1.5 μm , which corresponds to minimum loss in silica based optical fibers. This wavelength coincides with the transition between the two lowest, spin-orbit split levels of the trivalent erbium ion, which has stimulated extensive investigations of Er doped semiconductors [1-17]. The wavelength makes Si:Er an interesting candidate for a Si-based, integrable light source for optical communication. A practical device application is, however, still impeded by the temperature induced quenching of the luminescence yield [7-9]

which occurs already well below room temperature when interband excitation or electron-hole pair injection is employed. Codoping with light elements like C, N, F, and particularly, O, was found to improve the luminescence intensity dramatically [10–12]. In this work, we review recent work concerning the factors governing the luminescence yield, namely:

- the abundance and properties of different types of Er centers (as distinguished by high resolution luminescence spectroscopy) and the influence of preparation conditions,
- bypassing recombination channels and their passivation,
- different possibilities for the energy transfer from the host crystal to the Er 4*f* shell and high temperature quenching.

In particular, we report the recently observed influence of hydrostatic pressure [13] which is shown to affect both the excitation channel and the photoluminescence (PL) quenching and we indicate possibilities to improve the quantum efficiency at elevated temperature.

2. Er doping and defect characterization by high resolution spectroscopy

One of the main obstacles for obtaining a high luminescence yield of Si:Er is the low equilibrium solubility of Er in silicon, estimated for 10^{17} cm^{-3} [14]. In order to overcome this limitation, non-equilibrium doping methods, such as ion implantation, molecular beam epitaxy (MBE) [15, 16] and chemical vapor deposition epitaxy, together with thermal annealing techniques are employed. Because of its versatility and accessibility, most of the work reported so far invokes ion implantation, although the method generates a variety of undesirable implantation defects, which act as non-radiative recombination centers and thus as a bypass in the de-excitation of electron-hole (e^-h^+) pairs. Such defects need to be removed by suitable annealing. The choice of the annealing procedure depends, however, critically on the doping level. In particular, the point defects created by Er implantation are unusually stable and their removal requires annealing temperatures not lower than 900°C [6, 12, 17]. For high Er doses, however, such an annealing step results in the production of a heavily dislocated and twinned material, with a high concentration of erbium precipitates decorating the dislocations. Erbium incorporation can be improved by solid state regrowth at around 600°C, in particular in silicon containing oxygen. Nevertheless, even then an additional annealing step at 900°C is necessary to remove the remaining point defects. At doses well below the amorphization limit, annealing only at 900°C gives, however, good results with respect to crystal quality and residual radiation damage, as seen in deep level transient spectroscopy (DLTS) [6] (see also Sec. 3).

The luminescence yield of Si:Er depends strongly on preparation conditions and the codoping of the host material. Thermal treatment changes the luminescence intensity and also the spectral distribution dramatically. Obviously, we are dealing here with a big variety of different luminescent centers involving erbium and other intrinsic and extrinsic defects. For any systematic improvement of the

luminescent properties of Si:Er it appears therefore necessary to be able to distinguish centers with different microscopic structure. Unfortunately, the standard method for the identification of the microscopic structure of defects in crystals, electron paramagnetic resonance, failed so far: no spectra with the characteristic Er hyperfine structure (allowing unambiguous Er identification) are reported. Therefore until now, optical spectroscopy is still the main tool which enables us to distinguish microscopically different Er complexes.

The distinction of various Er complexes is based on subtle line splitting of the $J = 13/2 \rightarrow J = 15/2$ transition. The $4f$ states of the rare earth elements are extremely localized and well screened and thus, in principle, little sensitive to the crystal field but for their immediate surrounding. On the other hand, due to the long radiative lifetime and weak coupling with lattice vibrations the linewidths are usually exceedingly narrow. Thus, even small perturbations of the local environment can be resolved by high resolution spectroscopy, provided that inhomogeneous broadening is not too bad, which for Si:Er is the case for concentrations below a few times 10^{18} cm^{-3} . Because of the narrowness of the implanted layer, however, the total number of Er ions in the sample is rather small (well below 10^{13} per cm^2 for the applied ion energies). Hence the luminescence yield is not very high and high resolution is achieved efficiently only with the use of Fourier spectroscopy [4-6].

Different surroundings of the Er ions manifest themselves by different line patterns. Without specific measures to breed one particular type of Er center, very rich spectra are obtained after ion implantation, consisting of up to 200 lines in the narrow spectral region between 1.53 and 1.54 μm . Unfortunately, the crystal field splitting is not very specific as far as the number of components is concerned. The 15-fold degenerate ground state splits into 5 in a cubic surrounding and into 8 substates for any lower site symmetry. At elevated temperatures additional, the so-called hot lines, appear due to transitions from the higher lying, crystal field split substates of the 13-fold degenerate excited state. The quasi-thermal population of the excited states causes a shift of the whole line pattern towards higher energies and thus it allows us to study the crystal field splitting also in the excited state.

The assignment of particular lines to the respective Er center requires experience since it is based on the recognition of similar behavior of lines under variation of all parameters, namely preparation conditions (implantation dose and energy, background doping, annealing procedure, etc.) and the experimental conditions, like temperature, hydrostatic pressure, time dependence, excitation power, etc. The recognition of line patterns of individual centers allows one to work out the best parameters to "breed" this particular type of Er center.

Typical PL spectra of Si:Er are shown in Fig. 1. The simplest type of spectrum is shown in Fig. 1a, top trace, with the characteristic 5-line pattern expected for isolated Er ions in the Si lattice. According to model calculations of Lea, Leask and Wolf [18] such a crystal field splitting cannot occur for substitutional incorporation. In contrast, the line splitting indicates contribution of an octahedrally coordinated crystal field, occurring, e.g., for tetrahedral interstitial location of the Er ion. On the other hand, rare earth ions are expected to form readily complexes with oxygen and it is even necessary to offer oxygen during MBE growth of Si:Er

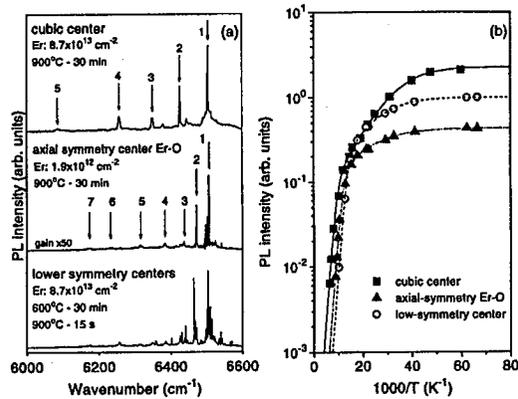


Fig. 1. (a) Three types of spectra observed in Er implanted Si as obtained for different doses, annealing temperatures and times as indicated. (b) Temperature dependence of the PL intensities of the 3 types of centers of Fig. 1a.

to avoid surface segregation of Er [15,16]. The observation of octahedral coordination of Er with oxygen in EXAFS investigations, closely resembling that of an erbium oxide, also seems to support this view [19]. However, the PL spectra of Er_2O_3 differ considerably from that of the cubic symmetry center observed in Er implanted silicon. The latter is also not found in MBE Si grown in the presence of oxygen. The occurrence of a highly symmetric, octahedrally coordinated Er-O complex as a dominant emitting center, even in oxygen rich Czochralski (CZ) silicon, seems to be highly unlikely, the more so as it turns out to be also the dominant center in float zone (FZ) Si, which typically contains at least 2 orders of magnitude less oxygen [5]. Though the PL yield of Er in CZ-Si is usually much higher than in FZ Si, a similar relative increase in the PL intensity of the cubic center was observed upon increasing the Er dose (for a constant ion energy) in both materials, indicating that the oxygen content cannot be the limiting factor for the center formation [5]. The most likely candidate responsible for the PL spectrum shown in Fig. 1a is hence an isolated erbium ion located at the tetrahedral interstitial site. This is supported by first principle calculations, which also yield the interstitial site as the most stable configuration of a trivalent erbium impurity in silicon [20].

Other generic Er complexes seen exclusively in oxygen rich CZ-Si, particularly after coimplantation of oxygen, are attributed to axial symmetry Er-O complexes (Fig. 1a, middle trace). The same type of spectrum can be also detected in MBE Si grown in the presence of O. At high Er implant doses additional low symmetry centers occur (Fig. 1a, lower trace), whose intensities correlate neither with oxygen nor with any other additional light ion impurity introduced. We attribute them thus to Er complexes with implantation induced intrinsic defects. The temperature dependent PL intensities for these 3 types of centers are given in Fig. 1b. They will be discussed in Sec. 4.

3. Other defects and alternative recombination channels

Ion implantation of any dopants is well known to cause all kinds of "radiation defects" by displacement of host atoms. Such defects provide efficient interband recombination channels in an indirect band gap semiconductor, reducing dramatically the lifetime of excess carriers. Most of these radiation defects can be removed by suitable annealing. The annealing behavior, however, can be significantly different depending on the presence of unintentional impurities, such as oxygen. Figure 2 illustrates the influence of the same annealing step (900°C, 30 min) performed on *n*-type FZ and CZ silicon implanted with $1.9 \times 10^{12} \text{ cm}^{-2}$ erbium, upon the near band edge photoluminescence. Prior to implantation and annealing the FZ sample shows the well-known phonon-assisted transitions of free and P-bound excitons (Fig. 2a), at a much higher intensity than the more contaminated CZ-Si. After implantation of Er and annealing, however, the situation is completely reversed: the CZ sample exhibits an order of magnitude stronger excitonic emission than the FZ sample (Fig. 2b). Exactly the same intensity ratio is also found for the $1.5 \mu\text{m}$ PL of erbium in both samples. Evidently, in spite of the same implantation and annealing parameters, FZ-Si contains much more nonradiative recombination centers than CZ-Si. Obviously oxygen, as well as other light elements, can "passivate" some of the nonradiative recombination shunts.

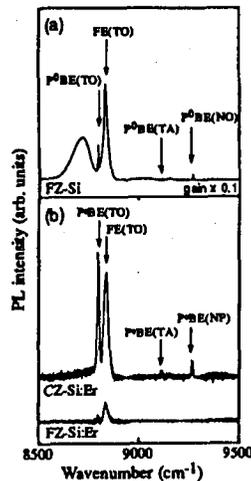


Fig. 2. Near band edge luminescence spectra of (a) FZ-Si as grown and (b) CZ-Si and FZ-Si implanted with Er and annealed for 30' at 900°C.

Some of the defects created by implantation and annealing, can also recombine radiatively and thus be detected in PL experiments. Figure 3 shows two typical examples of defects introduced by sample processing in CZ-Si implanted with Er and subsequently annealed at 900°C for 30' (Fig. 3a), or at 600°C for 30' followed by a rapid annealing step of 10'' at 900°C (Fig. 3b). Apart from the

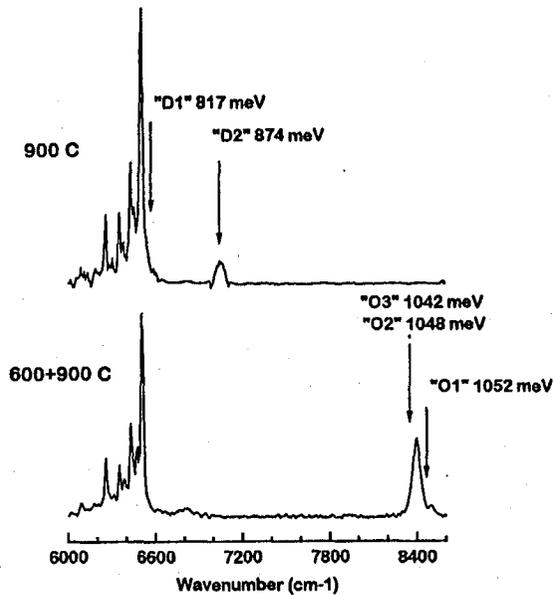


Fig. 3. Deep luminescence of CZ-Si implanted with Er after 1 stage (upper trace) and 2 stage annealing (lower trace).

different annealing procedure the material used and the Er implantation parameters were exactly the same. In the first case, the so-called *D* lines are observed, which occur also in plastically deformed material. They are attributed to radiative recombination of free electrons with holes trapped at dislocation related deep levels. One of these lines (marked "D1" in Fig. 3a) occurs at an energy very close to that of the main Er transition. In heavily Er doped samples, where considerable line broadening due to formation of Er clusters is observed, it can be even indistinguishable from the Er PL. A number of spectra ascribed in the literature to Er might be rather due to such dislocation related features. The two types of spectra can be easily distinguished, however, by their pressure dependence: the *D* lines move towards lower energies at a rate of approximately -2 meV/kbar, the value obtained also for the near band edge PL, whereas the pressure coefficient of the Er lines is by orders of magnitude smaller.

The formation of dislocations can be reduced by applying a lower annealing temperature (600°C) followed by a very rapid anneal step at 900°C , as can be seen in Fig. 3b. In this case, however, other defect related PL lines (close to 1.05 eV) are observed. Such a PL spectrum was reported for an unimplanted CZ-Si sample subjected to long time (130 hours) annealing at 500°C [21]. It is evident that the heavy damage caused by Er implantation stimulates the formation of different carrier trap centers, not necessarily related to the impurity itself. Many such defects introducing deep energy levels into the Si forbidden gap were observed, e.g., in DLTS [6, 22] and shown to be efficient carrier re-

combination centers, limiting the excitation efficiency of erbium. The concentration of most of the defects, except the one with the activation enthalpy of $E_c - 0.15$ eV which is probably oxygen related and a few others, can be significantly reduced by oxygen coimplantation [6, 22].

4. Energy transfer and temperature induced quenching of the luminescence

In the first part of this section we discuss only the case of excitation by electron hole pairs which occurs either by interband optical excitation or by a forward current in a p - n junction. In the second part we address also recent experiments with reverse biased diodes which show a quite different quenching behavior.

In Fig. 1b the temperature dependences of the three types of PL spectra shown in Fig. 1a are given. The Er PL intensity starts to decrease already at temperatures close to 25 K. In the range of 25–100 K, an activated behavior of the quenching is seen with center-dependent activation energies ranging from 5–15 meV. These energies are typical for thermal release of excitons bound at neutral, shallow impurities, suggesting that at low temperatures energy transfer from shallow bound excitons (BE) is the dominant Er excitation path. The efficiency of excitonic transport of the host excitation can be spectacularly demonstrated by directing the pump 514 nm laser line to the backside of the implanted wafer. In this case the Er PL yield is still comparable to that of the front side excitation, although the visible light penetration depth is only 1 μm and the excitation energy must travel through the whole wafer (400 μm thickness) before it reaches the Er doped layer. Only excitons can be envisioned to have such diffusion lengths. The activated behavior seen in Fig. 1b indicates, however, that the energy transfer is particularly efficient if the excitons are bound at some defect states in the neighborhood (i.e., within the diameter of the effective Bohr radius of the exciton) of the Er ion. With increasing temperature exciton localization becomes less effective and the competition of other recombination channels increases. The efficiency of such a quenching mechanism depends strongly on the sample quality.

Above 100 K, the PL intensity decays much more rapidly with a sample dependent deactivation energy of 60–150 meV, which seems, however, to be the same for all Er centers within one sample. This indicates the activation of a new recombination channel, common to all Er centers. As the Er luminescence decay is accompanied by a decrease in the Er lifetime, nonradiative recombination processes within the Er centers are also involved. So far, the nature of the quenching mechanism is not clear. One of the mechanisms under consideration which could be responsible for high temperature PL quenching is Auger-type nonradiative Er recombination due to energy transfer to free carriers. Experimental evidence supporting the occurrence of such a recombination process has been presented recently. It was shown that Er lifetime decreases upon increasing the free carrier concentration either by additional sample illumination or by increasing the current in forward biased junctions [23]. Another mechanism discussed recently is the back-transfer of the energy from the Er 4f shell to a state, which is involved also in the excitation path [12]. Such a mechanism would be thermally activated with an energy corresponding to the difference between the direct recombination energy of

that state and the intra $4f$ transition energy. This possibility was demonstrated in photo-conductivity (or photo-voltage) experiments which showed response when excited at the $1.54 \mu\text{m}$ Er transition wavelength. As a possible candidate for this state, a level found in DLTS experiments is considered, which appears only in Er and O containing samples. This level has an enthalpy of $E_c - 0.15 \text{ eV}$ (see also Sec. 3), so the energy mismatch between the direct hole recombination at that level and Er recombination would amount to about 0.15 eV , which agrees with the high temperature PL deactivation energy reported for some samples. However, there is so far no confirmation that backtransfer is indeed the dominant mechanism responsible for Er PL quenching at elevated temperatures. In contrast, the increase in the Er lifetime is governed by a much lower thermal activation energy than the decrease in the PL intensity. Moreover, the relative lifetime increase is typically by orders of magnitude lower than the drop of the normalized intensity. This suggests that the excitation path rather than nonradiative Er deexcitation is the factor limiting the PL yield. It seems, hence, that the PL deactivation energy probable relates rather to the thermal depopulation of some deep state providing otherwise an additional channel for energy transfer to the $4f$ shell.

In order to test the assumption of the participation of a deep state in the transfer process, we have performed experiments under hydrostatic pressure [13]. Hydrostatic pressure affects localized and delocalized states in different ways. A shallow state, e.g., is characterized by the effective mass of the nearby band extrema and by the dielectric constant. These two quantities depend only weakly on pressure and thus the ionization energy is practically pressure independent. Deep states, on the other hand, can be thought as composed from many band states and thus they do not follow any particular band. Consequently, their activation energy can change strongly under pressure [24].

We find actually a strong effect of pressure on the PL yield, but only in samples which were codoped with relatively high concentrations of O. The results are given in Fig. 4, where the temperature dependence of the cubic line amplitude is plotted in the same way as in Fig. 1b. It should be noted first that the Er lines themselves are practically not shifted under pressure as expected for these extremely localized states. There is, however, a systematic variation of the temperature dependence of the PL under pressure. In the intermediate temperature regime the deactivation energy does not change but the efficiency of the quenching process decreases substantially with increasing pressure, indicating an increased transfer rate to the $4f$ shell. As in this temperature range the PL quenching is governed by thermal release of shallow BE localized in the vicinity of Er ions, the lack of change in the deexcitation energy is not surprising. The BE ionization energy in silicon amounts to about 10% of the (neutral) shallow impurity ionization energy, and the latter, being a delocalized state, will depend only weakly on pressure. The strong pressure dependence of the coupling coefficient indicates, however, that some localized state is also involved in the capture of the host excitation and can transfer its recombination energy to erbium. The contribution of such a transfer channel increases evidently with increasing pressure, possibly because the energy mismatch between the deep defect state recombination (i.e.,

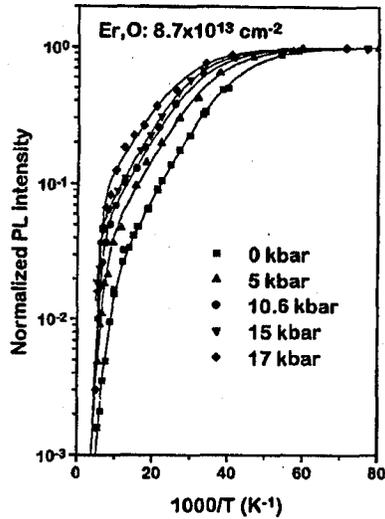


Fig. 4. Arrhenius plot of the normalized PL intensity of the cubic center for different pressures.

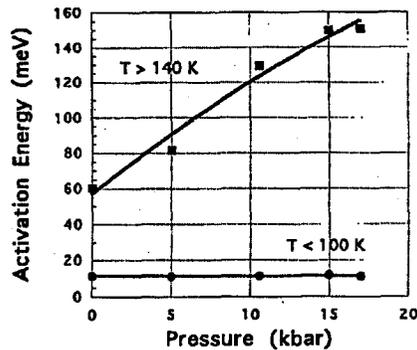


Fig. 5. Activation energy for the PL quenching as derived from Fig. 4 in the exciton detachment regime ($T < 100 \text{ K}$) and in the high temperature regime ($T > 140 \text{ K}$).

the recombination energy of an electron (hole) bound directly on a deep level with a free, or loosely bound hole (electron)) and the Er excitation becomes smaller.

Above 100 K; the PL deactivation energy increases strongly with increasing pressure, as shown in Fig. 5. The PL quenching in this regime is probably related to thermal ionization of the defect involved in the energy transfer to erbium. The increasing activation energy is in agreement with the assumption that the state becomes deeper under pressure (the pressure coefficient is about 6 meV/kbar which is of the same order of magnitude as that of the band states), therefore, its recombination energy becomes better matched to the $4f$ shell excitation.

Apparently there is a dramatic difference in the threshold temperature for quenching depending on the excitation mechanism. Diodes operated in forward direction show qualitatively the same behavior as the PL [16, 25]. In both cases, obviously the luminescence is excited by electron-hole pairs and in both cases it is quenched strongly above 100 K. Operated under reverse bias, however, the quenching can be much less effective and some samples were reported to have practically the same yield at 300 K as at low temperatures. Apparently Er excitation by hot electron impact, the mechanism envisioned for reverse bias excitation, does not suffer so much from bypassing recombination channels, which are so effective in the electron-hole pair mediated process. This seems to confirm that it is mainly the rather inefficient excitation channel which limits, so far, the room temperature Er photoluminescence yield.

5. Conclusions

The recent demonstration of practically unquenched electro-luminescence of reverse-bias operated light emitting diodes gives new hope in the future of optical emitters based on Si technology. The discovery of a significant improvement of the PL yield under hydrostatic pressure, on the other hand, shows that the energy transfer process in the electron-hole pair mediated excitation is not fully optimized as yet and that there is still an improvement possible also for this type of excitation, e.g., by use of other coactivating defects, and/or incorporation in strained superlattices. The details of the energy transfer and the quenching in the latter case are very complex and have not been fully understood as yet.

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