Extended Abstracts of the 2010 International Conference on Solid State Devices and Materials, Tokyo, 2010, pp427-428

Migration-limited relaxation in $Er_xY_{2-x}SiO_5$ crystals

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Er_2SiO_5 crystal is a key light source material for Si-photonics. Due to the crystalline nature, Er_2SiO_5 crystal is considered to have few defects and segregations originated in spite of high Er concentration[1]. Therefore, we expect that Er_2SiO_5 crystalline compact waveguide amplifier less than 1mm will be delivered. In this system, the energy transfer can occurred between Er^{3+}-Er^{3+} in $^4I_{13/2}$. Under strong excitation, one exited Er^{3+} is excited to the upper state by another[2]. This mechanism is so-called "Cooperative Upconversion" (CUC). CUC suppresses the emission efficiency at wavelength around 1.5μm. To overcome CUC problem, an introduction $Er_xY_{2-x}SiO_5$ crystals is proposed, since $Er_xY_{2-x}SiO_5$ crystals is expected to be diminish CUC because of dilution of Er atoms with keeping the crystallinity and the crystalline field surrounding Er[3,4]. The crystalline nature has been expected to suppress the concentration quenching of exciton decay time. However, the quenching behavior in $Er_xY_{2-x}SiO_5$ crystallites has been reported[3,4]. In this paper, we investigate the decay in $Er_xY_{2-x}SiO_5$ crystallites and discuss the possible mechanisms of energy transfer occurring in Er silicate systems.

$Er_xY_{2-x}SiO_5$ crystal thin films were formed on Si substrates by sol-gel method with the mixture sol solutions of Er-O, Y-O and Si-O. The Er ratio x is varied from 2.0 to 0.06. After spin-coating, (at 2000rpm for 1min), the samples were dried in the air at 120°C for 30min and baked in Ar at 500°C for 30min. This process repeated 5 times to get films of about 100nm thickness. Thereafter, they were annealed at 1200°C for 30min in Ar atmosphere. Photoluminescence (PL) measurements were performed by using a semiconductor laser with wavelength of 654nm and pump power of 30mW/mm^2. PL emissions were collected by a cooled Ge detector for PL and an InGaAs photomultiplier for time decay measurements.

$Er_xY_{2-x}SiO_5$ thin films exhibit the same characteristic PL spectrum fine structure as that of Er_2SiO_5, which has the main sharp peak at 1.529μm. Figure.1 shows the PL peak intensity and decay time of each $Er_xY_{2-x}SiO_5$ crystal as a function of Er composition. PL efficiency, which is PL intensity divided by Er concentration N_{Er}, are also plotted on this figure. The PL decay time decreases with increasing Er content. PL efficiencies strongly depend on x value and shows the same tendency as the decay time. This suggests that the decay time quenching is due to increasing non-radiative transition.

Considering the crystalline nature, it is hard to form defects or segregation of Er to cause the concentration quenching. Then we discuss the possible mechanisms of energy transfer occurring in Er silicate systems. When the quenching center (acceptor) concentration is low, energy migration within the network of neighboring Er ions (donor) becomes the dominating process in the relaxation. Therefore the decay is governed principally by intrinsic relaxation ($1/\tau_0$) and by diffusion-limited relaxation to the acceptor ($1/\tau_D$). Yokota and Tanimoto[5] and then Weber[6] demonstrated the energy migration model in the case of dipole-dipole interaction. After that, Tang et al.[7] showed that the fluorescence of donor decays exponentially with a time constant τ_f, given by

$$\frac{1}{\tau_f} = \frac{1}{\tau_0} + \frac{1}{\tau_D} = \frac{1}{\tau_0} + CN_AN_D$$

where C is a constant related to the transfer constants, and N_A and N_D represent the acceptor and

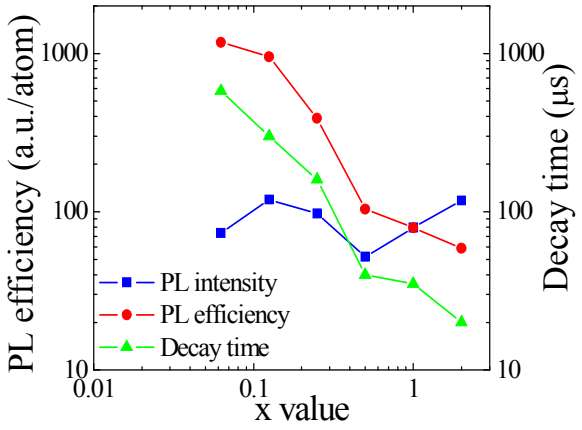

Figure.1 Photoluminescence properties of $Er_xY_{2-x}SiO_5$ thin film measured at 17K. PL efficiency is PL intensity divided by Er concentrations.

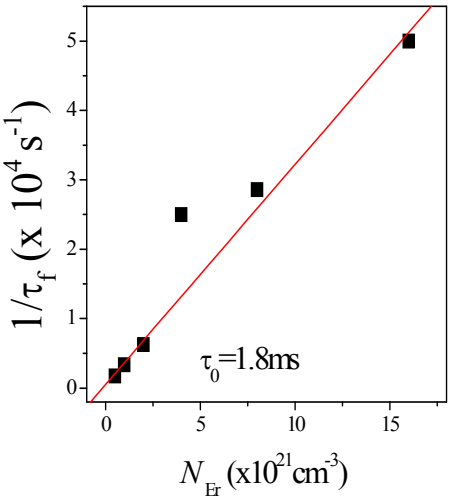

Figure.2 Exciton relaxation ratio as a function of Er concentration, which corresponds to acceptors under weak excitation.

donor concentrations, respectively. This migration indirectly increases the nonradiative deexcitation rate, enhancing the probability of decay in nonradiative defects.

Figure 2 shows the emission decay rate as a function of Er concentration. It can be approximately expressed by the linear equation with $\tau_0 = 1.8ms$. The intrinsic decay time is almost same as that of a low Er content sample independent of the energy migration. We note that the crystallinity of each $Er_xY_{2-x}SiO_5$ crystal is not so different and there is not very much change of N_A. This indicates a change in the probability of transferring the excitation to nonradiative acceptors by diffusion. Also the time-dependence decay behavior shows a distinguishing characteristic of diffusion-limited relaxation. Additionally, for crystallinity improvement of $Er_xY_{2-x}SiO_5$, we try to use layer-by-layer deposition processes of $Er_xY_{2-x}SiO_5$ films such as pulsed laser deposition (PLD) with rapid thermal annealing(RTA) crystallization. In this case, we have observed the improvement of decay[8].

Acknowledgements

This work is partly supported by Grand-in-Aid for Scientific Research of the Ministry of Education, Culture Sports, Science and Technology, Japan.

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