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Size and Interface State Dependence of the Luminescence Properties in Si Nano-crystals

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1. Introduction

Over the past few years, there has been considerable interest in semiconductor nano-crystals in SiO_2, which exhibit strong visible luminescence even at room temperature[1-4]. Although there has been a considerable amount of investigation on the basis of the quantum confinement theory, including the effects of interface states at the nano-crystal/SiO_2 interface and/or processing-induced defects, the detailed mechanism for luminescence from these Si nano-crystals is still being debated. In this work, we have shown that the photoluminescence originates from quantum confinement effects of Si nano-crystals by three kind of experiments; PL measurements with oxide etching depth, thermal oxidation of Si nano-crystals, and high temperature annealing scan. We also demonstrate that interface states at Si nano-crystal/SiO_2 interface can affect PL spectra intensity from the experiments on the hydrogen annealing.

2. Experimental

Wet thermal SiO_2 films, 200nm and 560nm thick, grown on lightly p-doped (100) Si were implanted with 130keV Si+ at does of $1\times10^{17}/cm^2$. PL depth properties were observed at the sample annealed at 1100°C for 2hr in an N_2 ambient every each 1min etch step. To observe the oxidation properties of Si nano-crystals, dry thermal oxidation was performed at 900°C for 10 ~ 60min. Also, to observe the effects of temperature, the Si implanted samples were annealed for 2hr at temperature ranging between 1000°C and 1300°C in an N_2. The hydrogen passivation effects were observed for the samples annealed for 2hr at the temperatures of 1050, 1100, and 1150°C, and followed by the treatment in a furnace for 30min at 400°C in forming gas(17% H_2 in N_2). PL spectra were taken using the 325nm line of an He-Cd laser at a pump density of $10mW/mm^2$.

3. Results and Discussions

In Fig. 1, etch rate of oxide layer monotonically decreased up to a region corresponding to the peak concentration of implanted Si. This result is consistent with the reports that nano-crystal sizes distribute into SiO_2 with the Gaussian profile[3]. PL spectra were observed every each etch step in Fig. 2. The peak of PL spectra appears the monotonically decreased intensity with etch step and blue-shift, except red-shift after first etching step. Blue components of original PL spectra are reduced due to etching of the smaller particles, located near the surface during the first etch step and as the result, PL peak seems to shift to lower energy. When etch depth increases over the region at which gets the initial average size of nano-crystals, the larger size of nano-crystals are etched out to reduce the average size of nano-crystals and to make the peak of PL spectra blue shift. From the oxidation in Fig. 3, the peak intensity of PL spectra increased and the peak energy was blue-shifted as the oxidation time increased. Blue shift of the PL peak is induced by the size reduction of nano-crystals due to the oxidation process of nano-crystals. This result indicates that PL emission depends on the size of nano-crystals. In Fig. 4, PL peaks are slightly red-shifted with increasing the annealing temperature. It is considered that the red-shift of spectra is caused by the increased average size of nano-crystals, on a basis of the previous reports that number of nano-crystals at particular size is reduced with increasing the annealing temperature and the average size of nano-crystals in SiO_2 monotonically increases with the temperature[3]. It is also observed that the intensity of PL spectra monotonically increased up to an annealing temperature of 1100°C and then decreased. As the annealing temperature increases, number of the nano-crystals to emit the visible light generally increases due to size increasing. However, size to emit the light is limited and as the result, total number of nano-crystals to emit the light finally reduces. After a hydrogen annealing treatment in Fig. 5, PL spectra did not shift and PL intensity just increased. The increase, not a red-shift, of PL peak in Fig. 5 implies that the hydrogen annealing process does not change the emission energy of the photoluminescence but increases the number of nano-crystals to emit the light. Here, the most reasonable model, which can explain our results is the introduction of the "intermediate energy state" originated from defects at Si nano-crystals/SiO_2 interface. Intermediate energy state would become a transition level of excited electrons and make Si nano-crystals non-radiative dots. The hydrogen passivated non-radiative dots can additionally contribute to enhance PL intensity. In Fig. 6, as the annealing temperature increases, the effects of hydrogen annealing on PL spectra intensity are continuously reduced. At 1150°C, almost no hydrogen annealing effects appear. It is considered that defects and/or interface states are annealed out due to the thermal healing effects at the elevated temperature. Though it is not shown in this paper, interface trap density was calculated using the C-V results. Sample annealed at 1050°C shows much higher interface state density than that of samples annealed at the elevated temperature.

5. Conclusions

We have shown that PL properties of nano-crystals in SiO_2 mainly originate from quantum confinement effects. Interface defects with the intermediate energy state in the bandgap make Si nano-crystals have non-radiative properties.

References

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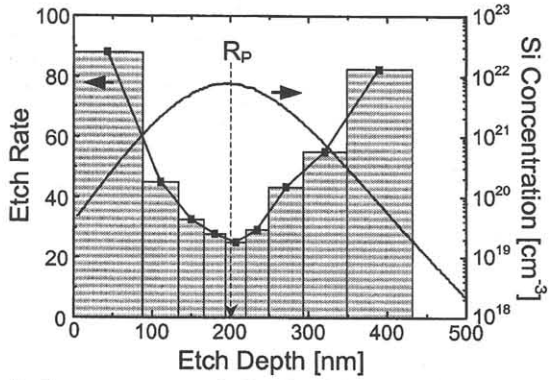

Fig. 1. Etch rate versus etch depth of the sample annealed at 1100°C for 2hr in an N_2 ambient after Si ion implantation.

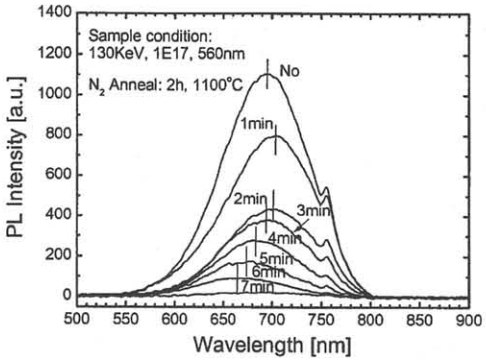

Fig. 2. The photoluminescence spectra measured every each 1min etch step for the sample of Fig. 1

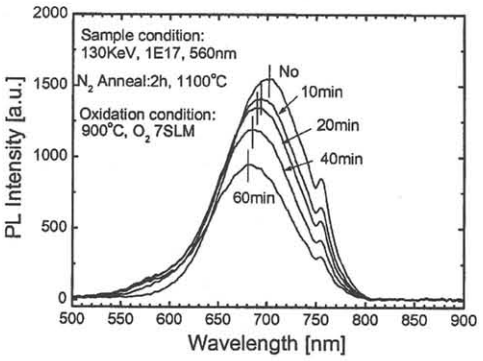

Fig. 3. PL spectra were oxidized at 900°C in an N_2 with flow rate of 7l/min for time ranging 0 to 60min

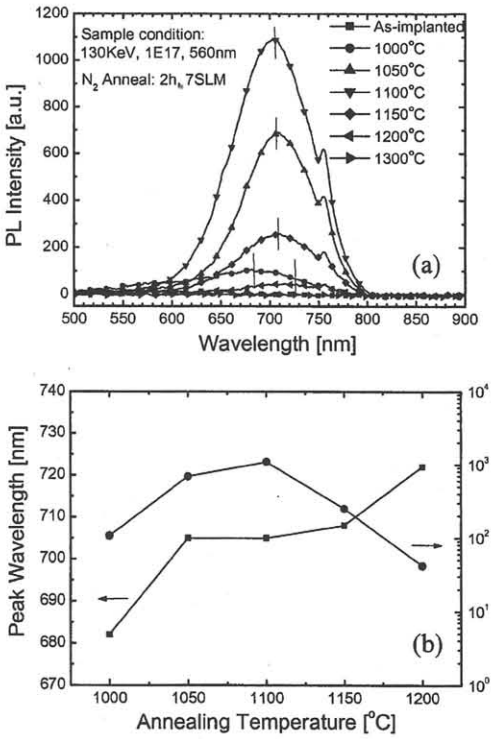

Fig. 4. PL spectra of the samples annealed for 2hr in N_2 at temperature ranging 1000 °C to 1300°C

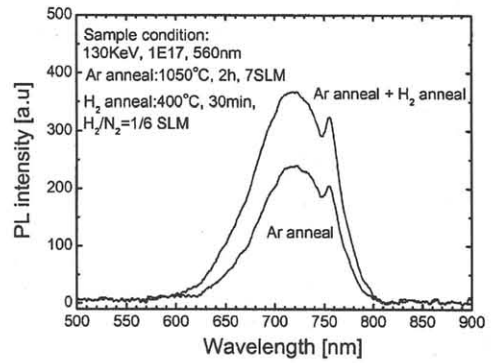

Fig. 5. PL spectra measured before and after annealing at 400°C for 30min in forming gas(17% H_2 in N_2) for the samples annealed at 1050°C for 2hr

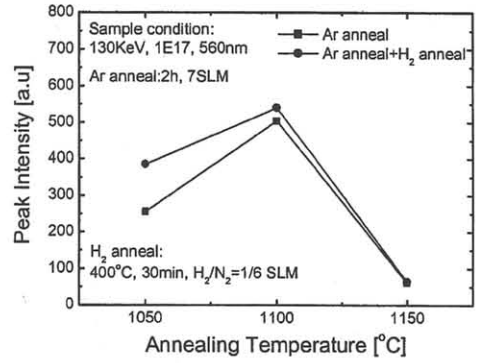

Fig. 6. The peak intensity of PL spectra, when the annealing temperature of the post-implantation was varied at 1050 °C to 1150°C in the case of Fig. 5