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Low temperature formation of luminescent Si nanocrystals with combined process of excimer UV-light irradiation and RTA

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ABSTRACT

Si ion implantation was widely used to synthesize specimens of SiO_2 containing supersaturated Si and subsequent high temperature annealing induces the formation of embedded luminescent Si nanocrystals. In this work, the potentialities of excimer UV-light (172 nm, 7.2 eV) irradiation and rapid thermal annealing (RTA) to achieve low temperature (below 1000 °C) formation of luminescent Si nanocrystals in SiO_2 have been investigated. The Si ions were introduced at acceleration energy of 180 keV to fluences of 7.5×10^{16} and 1.5×10^{17} ions/cm^2. The implanted samples were subsequently irradiated with an excimer-UV lamp for 2 h. After the process, the samples were rapidly thermal annealed at 1050 °C for 5 min before furnace annealing (FA) at 900 °C. Photoluminescence spectra were measured at various stages at the process. Effective visible photoluminescence is found to be observed even after FA at 900 °C, only for specimens treated with excimer-UV lamp and RTA, prior to a low temperature FA process. Based on our experimental results, we discuss the mechanism for the initial formation process of the luminescent Si nanocrystals in SiO_2, together with the effects with excimer lamp irradiation and RTA process on the luminescence.

1. Introduction

After initial reports on room temperature visible photoluminescence from porous silicon in the early 1990s [1,2], interest in the optical properties of Si nanocrystals embedded in SiO_2 has grown over the last decade [3–5] because of their potential applications toward Si-based integrated optoelectronic devices. The attention to this material is greatly increased due to the observation of light amplification in Si nanocrystals [6] and the application for nonvolatile memory [7]. One of the most promising approaches, compatible with common microelectronic device fabrication techniques, to producing Si nanocrystals may be ion implantation. This technique has the advantage that a given number of ions can be placed at a controlled depth and distribution by changing the fluences and acceleration energies [8,9].

It is well known that Si ion implantation into SiO_2 and subsequent high temperature annealing (more than 1000 °C) induce the formation of luminescent Si nanocrystals. The photoluminescence peaking in the near infrared or visible spectrum (between 1.4 and 1.8 eV) is evidently related to implant Si nanocrystals formed by decomposition of the SiO_x phase with high temperature annealing [10–14]. Moreover, the photoluminescence is enhanced with hydrogen passivation [15–19]. Although a considerable amount of research has been extensively performed by many researchers, world wide, the detailed mechanism responsible for this photoluminescence, some important issues, is still unclear.

The photoluminescence arising from implanted Si nanocrystals in SiO_2 has been attributed by some investigations to simple quantum confinement, while others have concluded that surface states present in the interfacial layer (including some types of defects) between the Si nanocrystals and the surrounding oxide matrix (localized surface states) play an important role in the emission process. In this work, the potentialities of excimer UV-light irradiation and rapid thermal annealing (RTA) processes combined with conventional furnace annealing (FA) to achieve low temperature (below 1000 °C) formation of luminescent Si nanocrystals in SiO_2 have been investigated.

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2. Experimental

The samples used were prepared by implanting Si^+ ions into oxidized commercial Si epitaxial layers (10 Ω cm, 10 μm) grown on p^+-type Si wafers (Sb-doped) with a resistance of around 0.01 Ω cm (p on p^+, oxide thickness of around 500 nm). The Si ions were introduced at an acceleration energy of 180 keV with the fluences 7.5×10^{16} and 1.5×10^{17} ions/cm^2 with a beam current of 300 μA (current density of about 15 μA/cm^2). The expected depth profiles of the implanted Si were estimated using TRIM [20] and found to be distributed in near Gaussian profiles with a peak depth around 300 nm from surface.

The implanted samples were subsequently annealed at 900 °C or 1050 °C in a flowing N_2 atmosphere for 4 h using a conventional tube furnace. Some of the samples were UV-light (172 nm, 7.2 eV, Ushio, excimer-UV lamp unit, Xe_2^* type) irradiated for 2 h with power density of 50 mW/cm^2 in vacuum, or rapidly thermal annealed at 1050 °C in N_2 atmosphere for 5 min with a rising rate of 50 °C/s (ULVAC, MILA-3000). Conventional room temperature photoluminescence spectra were measured at various stages of the processing. A He–Cd laser (325 nm, 3.82 eV) was used as the excitation source and the luminescence was detected by a cooled photomultiplier tube (Hamamatsu, R-943-02) employing the photon counting technique.

3. Results and discussion

It is well known that Si ion implantation into SiO_2 and subsequent high temperature annealing (more than 1000 °C) induce the formation of embedded luminescent Si nanocrystals by decompose of supersaturated SiO_x. The peak energy of photoluminescence is close around 1.7 eV for extreme low fluence implanted samples, but is slightly shifted to lower energies side with increasing in implanted ion fluences.

We will firstly show typical photoluminescence spectra obtained after FA at 1050 °C in a flowing N_2 atmosphere for several hours. The photoluminescence spectra of samples implanted to a fluence of 7.5×10^{16} ions/cm^2, and annealed with conventional furnace are shown in Fig. 1. Hereafter, all of the photoluminescence intensities are normalized with the luminescence of specimen after FA at 1050 °C for 4 h without rapid thermal annealing, i.e. only conventional FA. The photoluminescence spectrum after FA at 900 °C is also shown in the figure. It is clear from the figure that the luminescence intensity grows as the annealing time increases and the peak energy of the luminescence spectra are independent of the annealing time after annealing at 1050 °C. It is noted that only a very weak photoluminescence was obtained after FA at 900 °C. Peak height of this photoluminescence band is similar level that of as-implanted sample related to defect generated with ion implantation.

We also investigated the effects of excimer UV-light irradiation and RTA on the photoluminescence. The photoluminescence spectra of samples implanted to a fluence of 7.5×10^{16} ions/cm^2 and UV-irradiation are shown in Fig. 2. UV-light irradiation (172 nm, 7.2 eV) was carried out for 2 h in vacuum. The intensities of luminescence peaked around 2.1 eV increase with UV-light irradiation. The trend of the effects with UV-irradiation is similar for that with higher fluence implanted specimen, except appearing new defect related band around 2.7 eV.

Samples with/without UV-irradiation were further treated with conventional furnace. The results for the photoluminescence measurements of sample after FA at 900 °C in a flowing N_2 atmosphere for 4 h is shown in Fig. 3. New additional shoulders of spectra located around 1.7 eV appear without quenching of defect related peak. It is clear from figures that no difference was observed in both samples with/without UV-irradiation. It means that UV-irradiation before FA is not effective for FA at 900 °C, even generation of defects with UV-light irradiation.

We also combined the process of RTA before FA. The results for the photoluminescence measurements of samples after FA at 900 °C in a flowing N_2 atmosphere for 4 h are shown in Fig. 4(A) and (B). The photoluminescence spectrum after only FA at 1050 °C is also shown in the figure. It is clear from the figure that sufficient luminescence intensity can be obtained even after FA at 900 °C, combined with both UV-irradiation and RTA. The luminescence intensity achieves to the same level as that of after the FA process at 1050 °C or twice as high as that with RTA. However, the trend of enhancement is similar for that 1.5×10^{17} ions/cm^2 implanted samples, the enhancement is not as large as that for 7.5×10^{16} ions/cm^2 implanted samples. It is also clear that the peak energy of photoluminescence shift around 0.1 eV to lower side compared to that of 1050 °C FA sample.

Firstly, we discuss the low temperature formation of luminescent Si nanocrystals with UV-light irradiation and RTA before FA process. For the case embedded Si nanocrystals, the luminescence intensity is determined by the number of optimally-sized Si nanocrystals and their luminescence efficiency [21]. In forming the luminescent Si nanocrystals in a SiO_2 matrix, decomposition,

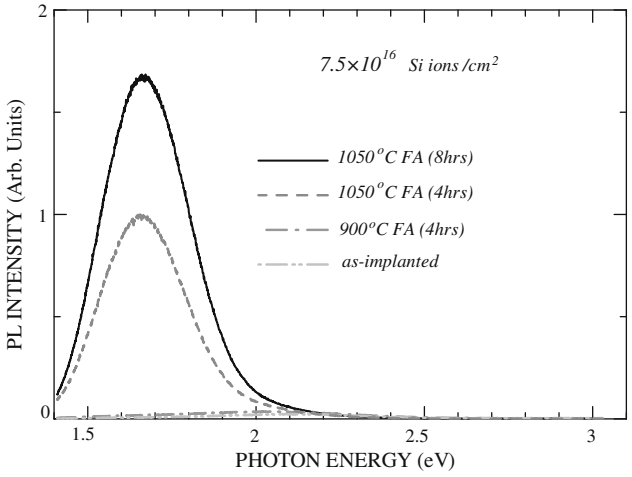

Fig. 1. Typical photoluminescence spectra of samples implanted to a fluence of 7.5×10^{16} and annealed with conventional furnace at 900 °C or 1050 °C for several hours. Each annealing conditions is indicated in the figure. Photoluminescence spectrum of sample without annealing is also showed in the figure.

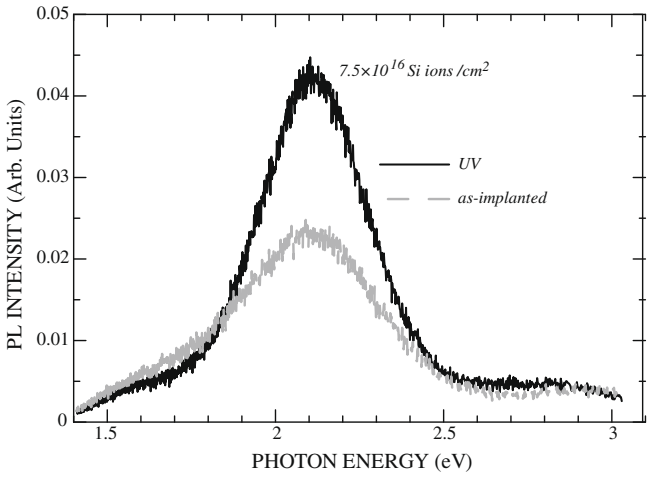

Fig. 2. Photoluminescence spectra of sample implanted to fluence of 7.5×10^{16} ions/cm^2, and before and after UV-light irradiated for 2 h in vacuum.