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ScienceDirect
Microelectronics Reliability 47 (2007) 781–785
MICROELECTRONICS RELIABILITY
www.elsevier.com/locate/microrel

RTA effects on the formation process of embedded luminescent Si nanocrystals in SiO_2

T.S. Iwayama [a,*], T. Hama [b], D.E. Hole [c]

[a] Department of Physics, Aichi University of Education, Igaya-cho, Kariya-shi, Aichi 448-8542, Japan
[b] Department of Information and Systems Engineering, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan
[c] Department of Engineering and Design, University of Sussex, Falmer, Brighton BN1 9QH, United Kingdom

Available online 2 March 2007

Abstract

It is well known that Si ion implantation into SiO_2 and subsequent high temperature anneals induce the formation of embedded luminescent Si nanocrystals. In this work, the potentialities of rapid thermal annealing to enhance the photoluminescence as well as those to induce low temperature formation of luminescent Si nanocrystals have been investigated. Ion implantation was used to synthesize specimens of SiO_2 containing supersaturated Si with different concentrations. The implanted samples were rapidly annealed only for a few minutes. After that, in some cases before that, the samples were annealed for a few hours using a conventional tube furnace to induce Si precipitation. Photoluminescence spectra were measured at various stages of anneal processes. The luminescence intensity is strongly enhanced with a rapid thermal annealing prior to a conventional furnace anneal. The luminescence intensity, however, decreases when rapid thermal annealing follows conventional furnace annealing. It is found that the order of heat treatment is an important factor in intensities of the luminescence. Enhancement is found to be typical for low dose samples. Moreover, the visible photoluminescence is found to be observed even after conventional furnace anneal below 1000 °C, only for rapidly thermal annealed samples. Based on our experimental results, we discuss the mechanism for the enhancement of the photoluminescence, together with the mechanism for the initial formation process of Si nanocrystals.

1. Introduction

After the discovery of the visible photoluminescence from porous silicon in the early 1990s [1,2], there has been considerable interest in Si nanocrystals embedded in SiO_2 [3–5] because of their potential applications toward Si-based optoelectronic devices due to their intense visible photoluminescence at room temperature. Recently, the attention to this material has greatly increased due to the observation of light amplification in Si nanocrystals [6,7]. Si nanocrystals have been fabricated by a variety of methods and include such techniques as ion implantation, CVD, sputtering, MBE or laser ablation.

One of the most promising approaches to producing Si nanocrystals, compatible with conventional microelectronic processing, may be by 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 doses and acceleration energies [8,9]. Ion beam synthesis of Si nanocrystals is a potential candidate for manufacturing stable and pure Si nanocrystals for applications in monolithically integrated Si-based optoelectronic devices.

It is well known that Si ion implantation into SiO_2 and subsequent high temperature annealing induce the formation of Si nanocrystals. The photoluminescence peaking in the near infrared or visible spectrum (between 1.4 and 1.8 eV) is evidently related to implanted 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].

* Corresponding author. Tel./fax: +81 566 26 2637.
E-mail address: tiwayama@auecc.aichi-edu.ac.jp (T.S. Iwayama).

0026-2714/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.microrel.2007.01.028

Although a considerable amount of research has been performed by many researchers, world wide, the detailed mechanism responsible for this photoluminescence is still unclear.

The photoluminescence arising from implanted Si nanocrystals in SiO_2 has been attributed by some investigations to quantum confinement, while others have concluded that surface states present in the interfacial layer between the Si nanocrystals and the surrounding oxide matrix play an important role in the emission process. In this work, the potentialities of rapid thermal annealing to enhance the photoluminescence as well as those to induce low temperature formation of luminescent Si nanocrystals have been investigated. Based on our experimental results, we discuss the mechanism for the enhancement of the photoluminescence, together with the mechanism for the initial formation process of Si nanocrystals, caused by rapid thermal anneals.

2. Experimental

The samples used were prepared by implanting Si^+ ions into oxidized Si epitaxial layers (10 Ω cm, 10 μm) grown on p^+-type Si wafers with a resistance of around 0.01 Ω cm (oxide thickness of around 600 nm). The Si ions were introduced by a Whickham ion implanter at an acceleration energy of 180 keV to doses ranging from 5.0×10^{16} to 2.0×10^{17} ions/cm^2 (corresponding to a peak excess Si concentration ranging from about 4% to 16%) with a beam current of 570 μA (current density of about 28.5 μA/cm^2). The expected depth profiles of the implanted Si atoms in the thermal oxide layers on the Si wafers were estimated using TRIM [20] and found to be distributed in near Gaussian profiles peaked around a depth of 300 nm from the surface. The implanted samples were subsequently annealed in a flowing N_2 atmosphere for several hours using a conventional tube furnace. Some of the samples were rapidly thermal annealed in N_2 atmosphere for a few minutes, with temperature rise rate of 50 °C/s. After rapid thermal anneals, samples were cooled down to below a temperature of 700 °C within 30 s. 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, employing the photon counting technique.

3. Results and discussion

The photoluminescence spectra of samples annealed at 1050 °C for 5 h are shown in Fig. 1. It is clear that the peak energies of the photoluminescence spectra are strongly affected by the dose of implanted Si ions. The peak energy is close to 1.7 eV in the samples with a dose of 5×10^{16} ions/cm^2, but is shifted to lower energies with increasing doses. The intensity of the luminescence grows and then decreases as the ion dose increases, with the maximum value being obtained at the implanted dose of 1.5×10^{17} ions/cm^2. Here, we note that the luminescence peak energy depends on the implanted ion doses, but it does not shift with prolonged annealing.

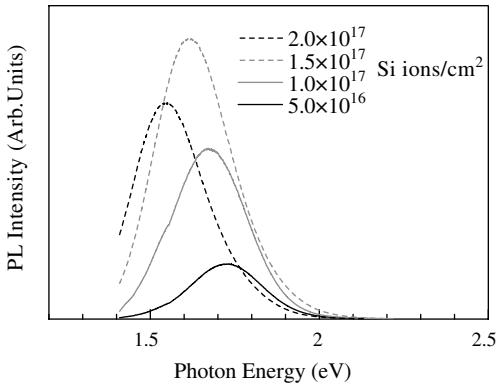

Fig. 1. Photoluminescence spectra of 180 keV Si implanted 600 nm thermal oxide films to doses of 5.0×10^{16}, 1.0×10^{17}, 1.5×10^{17} and 2.0×10^{17} ions/cm^2, after annealing at 1050 °C for 5 h.

Ion implanted samples were rapidly thermal annealed at 1100 °C in a flowing N_2 atmosphere for 5 min with a temperature rise rate of 30 °C/s, prior to a furnace anneal at 1050 °C for 1 h in N_2. The photoluminescence spectra of samples implanted to doses of 5.0×10^{16} and 1.0×10^{17} ions/cm^2, and rapidly thermally annealed prior to a furnace anneal are shown in Fig. 2a and b. It is clear from the figures that the luminescence intensity is enhanced with a rapid thermal anneal. Here, we note that the luminescence peak energy depends on the history of annealing. For samples with rapid thermal anneals, the peak energies shift to the lower side. Although not shown here, the luminescence intensity decreases with a rapid thermal annealing after a conventional furnace annealing. Moreover, we examined other samples with different ion doses up to 2.0×10^{17} ions/cm^2 and found that the rapid thermal anneals become less effective in enhancing the intensity of the luminescence with increasing ion doses, as shown Fig. 3. In Fig. 3, unity enhancement means the photoluminescence intensity without rapid thermal anneals.

Fig. 4 shows the photoluminescence spectra of samples processed with rapid thermal anneals at 1100 °C for 1 min with a rising ratio of 50 °C/s, prior to a furnace anneal at 950 °C for 3 h. In the figures, the spectra of the samples just annealed at 950 °C for 3 h are shown as gray lines and those of the samples treated with rapid thermal anneals only as dashed lines. It is clear from the figures that obvious luminescence is obtained neither only after furnace anneals nor rapid thermal anneals. However, combining these two processes, we can obtain clear photoluminescence in the visible range, even the furnace annealing below 1000 °C.

We discuss the enhancement of the photoluminescence with a rapid thermal anneal prior to a furnace anneal. For the case Si nanocrystals, the luminescence intensity