Investigation of PL properties of C-doped SiO_2/Si samples after high energy Pb ion irradiation*

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Abstract Amorphous SiO_2 thin films with about 400—500 nm in thickness were thermally grown on single crystalline silicon. These SiO_2/Si samples were firstly implanted at room temperature (RT) with 100 keV carbon ions to 2.0×10^{17}, 5.0×10^{17} or 1.2×10^{18} ions/cm^2, then irradiated at RT by 853 MeV Pb ions to 5.0×10^{11}, 1.0×10^{12}, 2.0×10^{12} or 5.0×10^{12} ions/cm^2, respectively. The variation of photoluminescence (PL) properties of these samples was analyzed at RT using a fluorescent spectroscopy. The obtained results showed that Pb-ion irradiations led to significant changes of the PL properties of the carbon ion implanted SiO_2 films. For examples, 5.0×10^{12} Pb-ions/cm^2 irradiation produced huge blue and green light-emitters in 2.0×10^{17} C-ions/cm^2 implanted samples, which resulted in the appearance of two intense PL peaks at about 2.64 and 2.19 eV. For 5.0×10^{17} carbon-ions/cm^2 implanted samples, 2.0×10^{12} Pb-ions/cm^2 irradiation could induce the formation of a strong and wide violet band at about 2.90 eV, whereas 5.0×10^{12} Pb-ions/cm^2 irradiation could create double peaks of light emissions at about 2.23 and 2.83 eV. There is no observable PL peak in the 1.2×10^{18} carbon-ions/cm^2 implanted samples whether it was irradiated with Pb ions or not. All these results implied that special light emitters could be achieved by using proper ion implantation and irradiation conditions, and it will be very useful for the synthesis of new type of SiO_2-based light-emission materials.

Key words heavy ion irradiation, carbon ion implantation, photoluminescence (PL) spectra

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1 Introduction

Since light emission from porous Si at room temperature was reported, great exertion has been spent on the discovery of new light-emitting materials to meet the demands of optoelectronic applications[1—4]. There has been a great interest in the structural and optical properties of silicon dioxide based luminous materials for their compatibility with silicon technology and huge applications on the optoelectronic materials in the future. The visible photoluminescence (PL) of silicon dioxide based materials has been reported by different authors. A number of methods including co-sputtering, CVD, chemical etching and ion implantation have been used to synthesis these luminous materials. Among them, ion implantation is an effective technique for obtaining such materials because the concentration and the depth of the implanted ions can be determined by the implantation dose and the ion energy. According to experiment

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results[5—9], annealing is quite necessary to obtain light centers from the implanted samples, and the annealing temperature and time have strong effects on the observed photoluminescence.

Swift heavy ion irradiations can change significantly the micro-structures and dynamic properties of solid materials. Recently, a novel technique[10—12], 'low energy ion implantation + swift heavy ion irradiation', was proposed to synthesize the new light-emitting materials, and had made a surprising progress on the blue-violet emission from C-doped SiO_2 films. In this paper, luminescent properties of C-doped SiO_2 films after high energy Pb-ion irradiations were extensively studied.

2 Experimental

Amorphous SiO_2 (a-SiO_2) films with about 400—500 nm in thickness were thermally grown on silicon (111) wafers by wet oxidation at 1050℃. These SiO_2/Si samples were firstly implanted at room temperature (RT) with 100 keV C-ions from LC-4 High Energy Ion Implanter (Institute of Semiconductors, CAS), then irradiated at RT by Pb ions (CIRIL-GANIL, Caen). The detailed parameters of ion implantations and irradiations were shown in Table 1, in which E, S_e, S_n, R_p and Φ are respectively the ion energy, electronic energy loss, nuclear energy loss, projected range and implantation/irradiation fluence, and S_e, S_n and R_p were estimated by SRIM2003[13]. From Table 1 we can see that the implanted carbon atoms are stopped into the SiO_2/Si samples, whereas the incident Pb ions could penetrate through the C-doped region. Because of S_e(Pb) >> S_n(Pb), the S_e plays an important role in the irradiation effects.

After C-ion implantation and high energy Pb-ion irradiations, the samples were analyzed by a fluorescent spectroscopy to investigate the light-emission properties. The used excitation light had a wavelength of 325 nm emitted from a He-Cd laser which has an output power of 15 mW.

Table 1. The parameters of C-ion implantation and Pb-ion irradiation.

Ion	E/MeV	S_e/(keV/nm)	S_n/(keV/nm)	R_p/nm	Φ/(ions/cm^2)
C	0.10	0.25	0.044	325	(2.0, 5.0, 12) $\times10^{17}$
Pb	853	22.9	0.046	50.0 $\times10^3$	(5.0, 10, 20, 50) $\times10^{11}$

3 Results and discussion

For the SiO_2/Si samples irradiated with Pb ions (see Fig. 1), significant PL peaks centred at about 1.9, 2.6 and 3.1 eV were observed when irradiation fluences were 5.0×10^{11} and 1.0×10^{12} Pb-ions/cm^2, and the intensities of the PL peaks located at about 1.9 and 2.6 eV raised greatly with increases of the Pb-ion irradiation fluence. This PL response is similar to that studied on ion-implanted SiO_2 samples[14, 15]. In these cases, the three PL peaks are originated from the damage and defects induced by the Pb-ion irradiations, and they are corresponding to non-bridging oxygen defects (≡SiO$^\bullet$, 1.9 eV), oxygen-vacancies defects (≡Si-Si≡, 2.6 eV) and Si-O related defects (O-Si-O, 3.1 eV)[16] respectively. These imply that many point defects are induced in SiO_2 films after swift heavy ion irradiations.

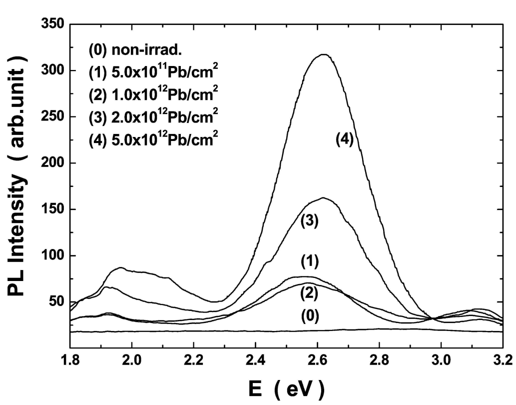

Fig. 1. PL spectra of SiO_2/Si samples after Pb-ion irradiations.

For the SiO_2/Si samples implanted with C-ions (see Fig. 2), two PL peaks centred at 2.12 and 2.71 eV was evidently observed with an implantation dose of 2.0×10^{17} C-ions/cm^2, and only one PL peak centered at about 2.12 eV was observed with an implantation dose of 5.0×10^{17} C-ions/cm^2, whereas no PL peak could be observed in the samples after 1.2×10^{18} C-ions/cm^2 implantation. Furthermore, the PL efficiency of the samples with 2.0×10^{17} C-ions/cm^2 implantation is larger than that with 5.0×10^{17} C-ions/cm^2 implantation. These PL peaks are very similar to the PL responses of oxygen-vacancies defects (≡Si-Si≡) in SiO_2 films and carbon clusters reported by Garrido[6]. These experimental results suggested that point defects and carbon clusters have been formed after C-ion implantation.

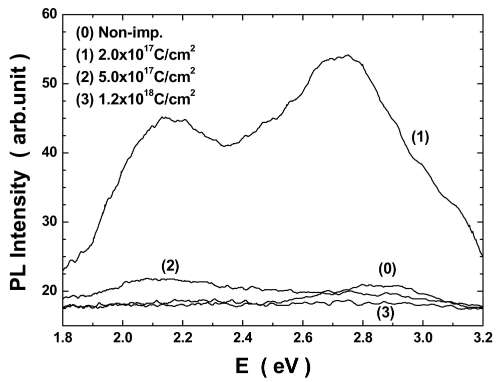

Fig. 2. PL spectra of SiO_2/Si samples after C-ion implantation.

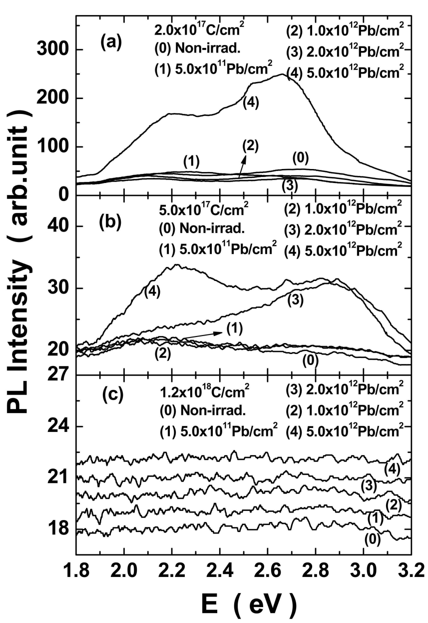

Fig. 3. PL spectra of C-doped SiO_2/Si samples after Pb-ion irradiations.

The PL spectra of C-doped samples after Pb-ion irradiations were shown in Fig. 3. For the samples implanted with 2.0×10^{17} C-ions/cm^2 (Fig. 3(a)), the PL efficiency remarkably increased when the irradiation fluence was 5.0×10^{12} Pb-ions/cm^2, and the PL bands was centered at 2.19 and 2.64 eV. The PL intensity has almost no change after 5.0×10^{11}, 1.0×10^{12} or 2.0×10^{12} Pb-ions/cm^2 irradiation. For the samples implanted with 5.0×10^{17} C-ions/cm^2(Fig. 3(b)), a significant PL band centered at 2.90 eV was observed when the irradiation fluence was 2.0×10^{12} Pb-ions/cm^2, and PL bands centered at 2.83 and 2.23 eV can also be seen when irradiation fluence was 5.0×10^{12} Pb-ions/cm^2, but no PL peaks was found for the samples un-irradiated and those after 5.0×10^{11} or 1.0×10^{12} Pb-ions/cm^2.

It is known that the light bands at 2.19 (2.23), 2.64, 2.83 and 2.90 eV are most properly related to the energy gaps of the 3C-, 10H-, 8H- and 15R-SiC[11, 17, 18]. The appearances of the PL peaks in Fig. 3(b) may suggest the formation of SiC polytypes. It was also seen that different structures have been formed at different implantation/irradiation fluences of C-ion implantations and Pb-ion irradiations. It seemed that the structures of 3C- and 10H-SiC were more easily to be produced at an implantation dose of 2.0×10^{17} C-ions/cm^2 and an irradiation fluence of 5.0×10^{12} Pb-ions/cm^2, but a 15R/8H-SiC structure was easily formed at an implantation dose of 5.0×10^{17} C-ions/cm^2 and an irradiation fluence of 2.0×10^{12} Pb-ions/cm^2, and 3C- and 8H-SiC structures were easily to be produced at an implantation dose of 5.0×10^{17} C-ions/cm^2 and an irradiation fluence of 5.0×10^{12} Pb-ions/cm^2.

It is surprising that no PL peaks could be seen in the samples at an implantation dose of 1.2×10^{18} C-ions/cm^2 no matter the samples were irradiated by Pb ions or not (as shown in Fig. 3(c)).

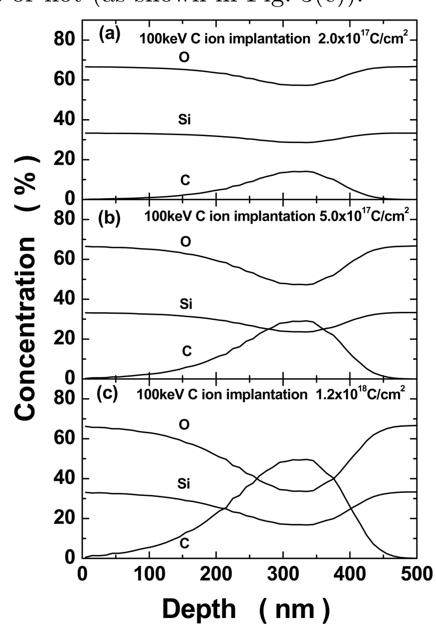

Fig. 4. Depth and concentration of C, Si and O atoms in a-SiO_2 films implanted with C ions.

For further discussion of the experimental results in detail, the depth and the concentration of C ions implanted in a-SiO_2 films were simulated with SRIM2003 code (shown in Fig. 4). One can see that the carbon atoms concentrated at a peak concentration of near 350 nm. Increasing of C ion implantation dose had increased the concentration of carbon atoms.

There are at least two factors that could affect the PL properties in the C-doped SiO_2 films. One is the damage caused by the implanted C ions, and the another one is merely the doping role of carbon which induces some new chemical bonds with silicon or/and oxygen. Both factors were equally important at an implantation dose of 2.0×10^{17} C-ions/cm^2. When the implantation dose was increased to 5.0×10^{17} C-ions/cm^2, the doping effect is more important than damage effect because the concentration of silicon is almost equal to the peak concentration of the implanted carbons. Some Si-C bonds could be induced under Pb-ion irradiations, so the luminescence bands affiliated to the new SiC micro-structures appeared. When the implantation dose of carbon was further increased to 1.2×10^{18} C-ions/cm^2, the peak concentration of the implanted carbons became more than the sum of the silicon's and oxygen's concentrations, which resulted in the combination of carbon-carbon. No matter the samples were irradiated with Pb ions or not, no light centers were produced but some carbon-related micro-structures corresponding to the broad Raman shift band within 1000—1700 cm^{-1}[19].

4 Concluding remarks

High energy Pb-ion irradiations can cause significantly modifications of photoluminescence properties in C-doped SiO_2/Si samples. A lot of blue light (2.64 eV) and green light (2.19 eV) centers were induced in the samples under 2.0×10^{17}ions/cm^{2}carbon implantation and 5.0×10^{12}Pb-ions/cm^{2}irradiation. As for 5.0×10^{17}ions/cm^{2}carbon doped samples, a notable PL peak centered at 2.90 eV was observed when the irradiation fluence was 2.0×10^{12}Pb/cm^2, and PL peaks centered at 2.83 and 2.23 eV were obviously observed when the irradiation fluence was 5.0×10^{12}Pb/cm^2. However, no PL peaks were found in SiO_2/Si samples implanted with carbon in the dose of 1.2×10^{18}C/cm^2. All the experimental results suggested that high energy heavy ion irradiation can induce new structures of photoluminescence in C-doped SiO_2/Si samples, and the specific PL structures can be selected by choosing suitable parameters of carbon implantation and heavy-ion irradiation. These can provide an important scientific basis for the development of SiO_2-based luminescent materials in a specific wavelength.

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