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Effects of hydrogen plasma annealing on the luminescence from a-Si:H/SiO_2 and nc-Si/SiO_2 multilayers

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Abstract

Effects of post-hydrogen plasma annealing (HPA) on a-Si:H/SiO_2 and nc-Si/SiO_2 multilayers have been investigated and compared. It is found that photoluminescence (PL) from hydrogen-passivated samples was improved due to the reduction of non-radiative recombination defects. Some interesting difference is that during HPA, atomic hydrogen can directly passivate defects of a-Si:H/SiO_2, which results in the reappearance of luminescence band at 760 nm, while for nc-Si/SiO_2, hydrogen passivation requires additional thermal annealing after nc-Si/SiO_2 multilayer was treated by HPA. It is indicated that higher atomic mobility is needed to passivate defects at nc-Si/SiO_2 interface compared with a-Si:H/SiO_2 interface.

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Keywords: Nanostructures; Optical properties; Plasma annealing

1. Introduction

Si/SiO_2 multilayered structure is one of the promising candidates for developing nanocrystalline Si (nc-Si)-based optoelectronic devices since the sublayer thickness can be controlled accurately and the size of the formed Si quantum dots after post-annealing can be easily adjusted by changing the Si sublayer thickness and in turn the room temperature PL peak positions can be modulated [1,2]. Despite of the extensive studies on the luminescence from amorphous-Si/SiO_2 (a-Si/SiO_2) and nc-Si/SiO_2 multilayers, the luminescence origin is still an open question [3]. Another challengeable topic is to improve the luminescence efficiency for actual device applications. Since high temperature post-annealing was usually carried out to get the structural change from a-Si/SiO_2 to nc-Si/SiO_2, many types of defects would be generated during this process which act as the non-radiative recombination sites to suppress the luminescence efficiency.

Surface passivation techniques, such as oxidation [4] and hydrogenation [5–9], have been applied to decrease defects and increase the PL efficiency. Cheylan and Elliman [7] studied the effect of hydrogen on the photoluminescence of Si nanocrystals embedded after the sample was exposed in forming gas (95% N_2 + 5% H_2, 500 °C, 1 h) because of the hydrogen passivation of non-radiative defects in nanocrystals. Many researchers studied the reaction between Si–SiO_2 interface with atomic hydrogen or molecular hydrogen [10–12]. Hydrogen plasma annealing (HPA) can be carried out at room or a low temperature, which is of benefit to the design and fabrication of Si-based devices [13,14]. In our previous work, HPA was used to treat a-SiO_2/SiO_2 multilayers under various experiment conditions, it was found that the quenched luminescence can be recovered after suitable HPA treatments [15]. In this paper, we further study the HPA effects on the luminescence of a-Si/SiO_2 and nc-Si/SiO_2 multilayers. It indicates that HPA is indeed helpful for the improvement of luminescence from the both types of samples due to the hydrogen passivation.

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2. Experimental

Fifteen periodic a-Si:H/SiO_2 (6 nm/2 nm) multilayers were prepared on the silicon substrate in r.f. plasma-enhanced chemical vapour deposition (PECVD) system by alternatively repeating the silicon deposition and *in situ* plasma oxidation process. The a-Si:H layers were deposited by using the pure silane with flow rate of 5 sccm and the chamber pressure of 10 mTorr. Subsequently, an *in situ* plasma oxidation was carried out with the pure oxygen of 20 sccm at 200 mTorr. The r.f. power and substrate temperature for both process were kept at 30 W and 250 °C, respectively.

In order to investigate the effect of hydrogen plasma annealing on the Si/SiO_2 multilayers, as-deposited samples were post-treated in N_2 ambient at the temperatures of 450 °C (30 min and 2.5 h) and 1100 °C (30 min). Following HPA at room temperature was applied with pure hydrogen gas under the reaction pressure of 46 mTorr. The hydrogen gas flow rate was 40 sccm and the r.f. power density was about 0.08 W/cm^2.

Fourier transform infrared (FTIR) transmission and Raman-scattering spectroscopy were used to study the bonding configurations and microstructures of the samples. FTIR measurements were performed by a Nexus 870 FTIR spectrometer on the sample grown on a double-polished substrate, and the signal of the substrate was subtracted from the FTIR transmission spectra. The PL signals were detected by a Jobin Yvon Horiba HR800 Spectrometer, operating with 600 g/mm, and power of 25 mW. The excitation source was a laser with wavelength of 488 nm.

3. Results

Fig. 1 shows the FITR spectra of as-deposited sample and the samples annealed at 450 and 1100 °C, respectively. It is found that both of the absorption bands around 640 cm^{-1} (Si–H bending vibration) and 2000 cm^{-1} (Si–H stretching vibration) become weaker after thermal annealing at 450 °C, which indicates the diffusion of hydrogen from the film [16]. When the sample was annealed at 1100 °C, these two vibration bands related to Si–H bands disappeared completely. Meanwhile, it is found that the peak of Si–O band (~1050 cm^{-1}) was blue shifted and the rocking mode of Si–O band (~450 cm^{-1}) could be observed clearly. The microstructures of samples were also characterized by using Raman-scattering spectra as shown in Fig. 2, which suggested that the Si sublayers of a-Si:H/SiO_2 multilayers are still amorphous phase after 450 °C annealing because of the presence of the Si–Si TO mode at 480 cm^{-1}. However, after annealing at 1100 °C, the TO mode became sharp and shifted to 515 cm^{-1}, which indicates that the formation of nano-crystalline silicon (nc-Si) in amorphous Si sublayers. Based on the phonon confinement mode, the size of nc-Si particles embedded in Si sublayer is calculated to be 4.2 nm [17].

A broad PL band centred at 760 nm could be observed clearly from as-deposited a-Si:H/SiO_2 multilayers. However, after annealing at 450 °C for 30 min, the PL was quenched as given in Fig. 3, which could be attributed to the increase of the non-radiative recombination centres due to the effusion of hydrogen from the films. It is interesting to see that the PL was

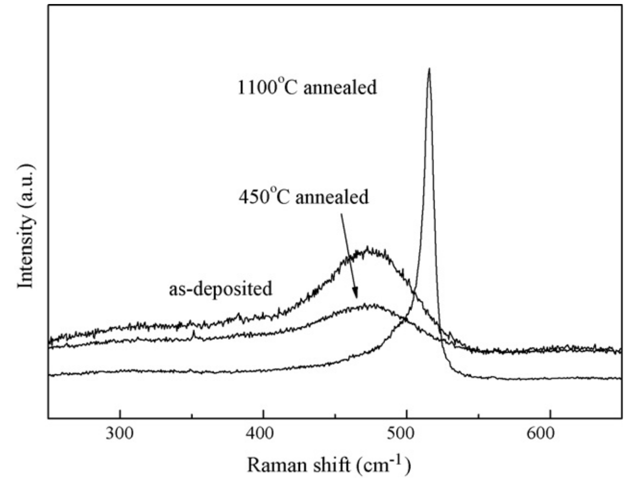

Fig. 2. Raman spectra of the as-deposited sample, the samples annealed at 450 °C for 30 min and 1100 °C for 30 min, respectively.

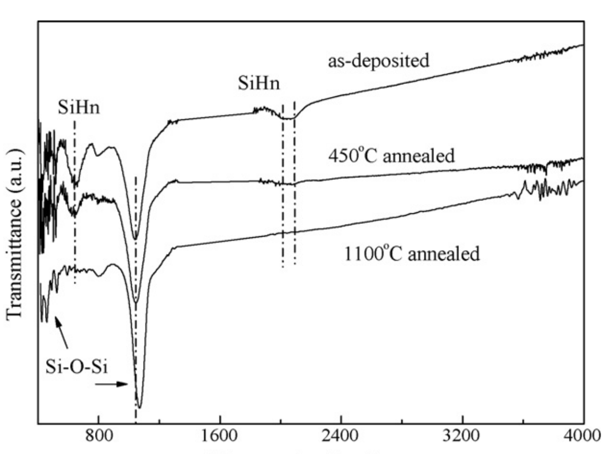

Fig. 1. FITR spectra of the as-deposited sample, the samples annealed at 450 °C for 30 min and 1100 °C for 30 min, respectively.

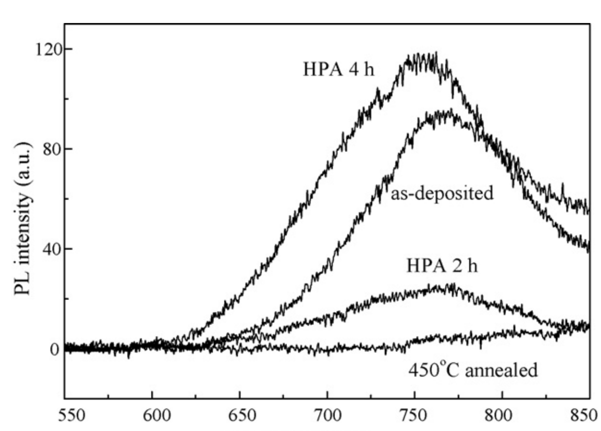

Fig. 3. Room temperature PL spectra of the as-deposited sample and the samples with and without HPA after annealing 450 °C for 30 min.