Fabrication of nc-Si/SiO_2 structure by thermal oxidation method and its luminescence characteristics

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Nano-crystalline silicon/silicon oxide (nc-Si/SiO_2) structures have been prepared from amorphous silicon films on both silicon and quartz substrates by using electron-beam evaporation approach and annealing at temperatures about 600 °C in air. As a thermal oxidation procedure, the annealing treatment is not only a crystallization process but also an oxidation process. Scanning electron microscopy is employed to characterize the surface morphology of the nc-Si/SiO_2 layers. Transmission electron microscopy study shows the sizes of nc-Si grains on the two different substrates. The nc-Si/SiO_2 structures exhibit visible luminescence at room temperature as confirmed by photoluminescence spectroscopy. Comparing the photoluminescence spectra of different samples, our results agree with the quantum confinement-luminescence center model.

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Nano-crystalline silicon/silicon oxide (nc-Si/SiO_2) structure is promising materials in the areas of optoelectronics and microelectronics due to its visible luminescence characteristics at room temperature. Possessing the unique electrical and optical properties which could not be observed in bulk silicon, these structures have the potential to be applied in the fabrication of light emitting diodes[1−3], photodetectors[4], flat-panel displays[5], light-emission memories[6], broadband laser mirrors[7], gas sensors[8], and so on. After the initial observation of photoluminescence (PL) from porous silicon layers by Canham[9], a considerable number of studies have been carried out on nanometer scale silicon structures which can exhibit luminescence[10−12]. Based on these investigations, several mechanisms have been suggested for the PL behavior of these structures. It is believed that quantum confinement-luminescence center model[13] is a mechanism that can lead to the observation of PL in such layers.

As a kind of silicon structures, which can offer visible luminescence, nc-Si/SiO_2 systems have attracted great interests[14,15]. The current methods for fabrication of nc-Si/SiO_2 systems are plasma-enhanced chemical vapor deposition (PECVD), radio frequency (RF) magnetron sputtering, and other deposition methods combined with annealing treatment which is mainly for crystallization. Most of these methods deposit composite films with Si and SiO_2 as different layers. After the annealing treatment crystallize the amorphous Si layers, the nc-Si is embedded in the SiO_2 films, and the nc-Si/SiO_2 structures are gained. As the current fabrication methods have complex procedures, we propose a method for fabrication of nc-Si/SiO_2 structures, which is simple and feasible.

In this letter, we deposit amorphous silicon on both silicon and quartz substrates suitable for the annealing treatment, using the electron-beam evaporation approach. After annealing with oxygen as part of the annealing gas content, the nc-Si/SiO_2 structures are fabricated. The luminescence of nc-Si/SiO_2 structures have been investigated by PL spectroscopy and the samples exhibit visible luminescence at room temperature. The morphology and crystalline structure of the nc-Si/SiO_2 layers are characterized by scanning electron microscopy (SEM) and transmission electron microscopy (TEM).

A 450-nm-thick layer of amorphous silicon was deposited on the silicon and quartz substrates, using electron-beam evaporation approach. The substrate temperature was kept at 300 °C and the pressure was about 2×10^{-3} Pa. Then the deposited films were annealed at 600 °C in air which contained 21% oxygen. In this situation, the annealing treatment was not only a crystallization process but also an oxidation process.

SEM study was carried out with a Quanta 400 FEG SEM system. TEM test was carried out with a JEM-2010HR TEM system. The PL spectra were measured with a laser micro-Raman spectrometer. The excitation light was from an Ar^+ laser whose output power was 2 mW and the wavelength was 514.5 nm. All the measurements were carried out at room temperature.

The microstructure of the surface of the nc-Si/SiO_2 structure prepared on the silicon substrate is shown in Fig. 1, and the one on quartz substrate is shown in Fig. 2.

As shown in Figs. 1 and 2, the thicknesses of the

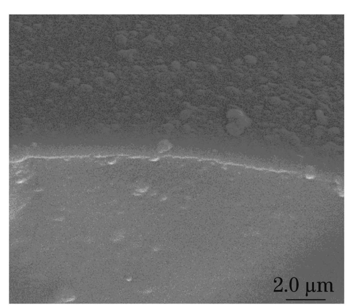

Fig. 1. SEM image of the surface of samples prepared on silicon substrate.

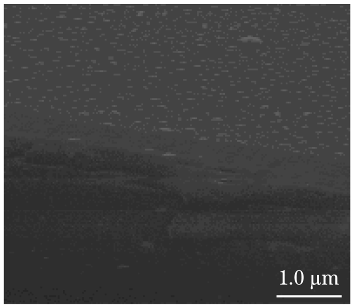

Fig. 2. SEM image of the surface of samples prepared on quartz substrate.

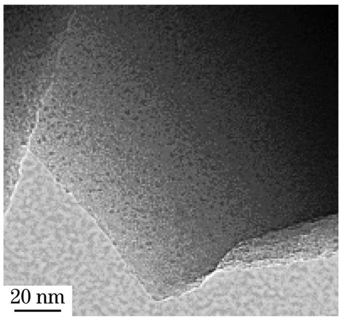

Fig. 3. TEM micrograph of films prepared on quartz substrate.

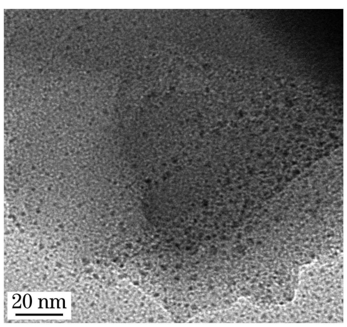

Fig. 4. TEM micrograph of films prepared on silicon substrate.

films on the two different substrates were both about 450 nm. Compared with the sample on silicon substrate, the grain size of the sample on quartz substrate was smaller: the grain size of the sample on silicon was about 3 nm and the one on quartz was about 1 nm, as shown in Figs. 3 and 4. The reason is that the silicon substrate makes benefits for the silicon atoms to organize some order structures during the depositing procedure while the quartz substrate does not. When the samples are annealed, these order structures make benefits for the growth of silicon grains.

Because of the quantum confinement effect, the band gap of nc-Si is larger than that of bulk silicon. Utilizing the channel potential well approximation model, it can be derived that the energy barrier entering the quantum wire from the quantum mechanics Schrödinger equation: $\Delta E = \Delta E_C + \Delta E_V = h^2/4m_e^* q^2 + h^2/4m_h^* q^2$[16], where ΔE_C is the energy barrier of the electron entering the quantum wire, ΔE_V is the energy barrier of the hole entering the quantum wire, h is the Planck constant, m_{e}^* is the effective mass of electron, m_{h}^* is the effective mass of hole, and q is the grain size of Si. The effective band

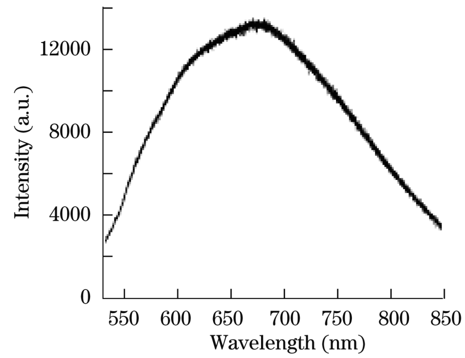

Fig. 5. PL spectra from samples prepared on silicon substrate.

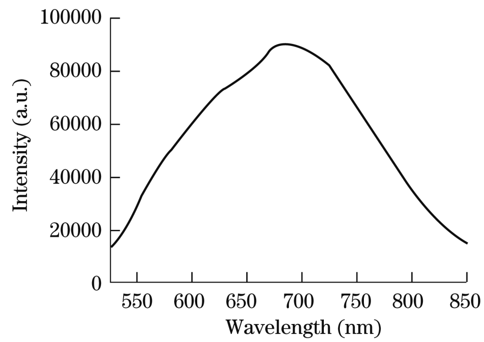

Fig. 6. PL spectra from samples prepared on quartz substrate.

gap of nc-Si is $E = E_0 + \Delta E$, where the band gap E_0 is 1.12 eV for Si.

This offers a chance for us to gain visible luminescence from silicon. In our experiments, though the sizes of Si grains on the different substrates are different, the central wavelength and wavelength range of the PL spectra from the different samples are nearly the same, as shown in Figs. 5 and 6. The carriers which were generated in the nc-Si tunneled to the luminescence center in SiO_2. And the recombination of the carriers resulted in the luminescence phenomenon. As the luminescence centers are not in the nc-Si but in SiO_2, the similarities of the PL spectra are reasonable, which agrees with the quantum confinement-luminescence center model[13].

Figures 5 and 6 also show that the PL intensity of quartz-substrate sample is much stronger than that of silicon-substrate sample. This is because, to the Si substrate samples, the photo-generated carriers in nc-Si grains tunnel through the nc-Si/SiO_2 layers, and enter the Si substrate, making nonradiative recombination. As a result, the efficiency of radiative recombination at light emission centers on the nc-Si/SiO_2 interface region is reduced. However, this does not happen in the quartz sample cases.

In summary, we report a simple and feasible method for the fabrication of nc-Si/SiO_2 structures. We deposited amorphous silicon on both silicon and quartz substrates, using the electron-beam evaporation approach. The nc-Si/SiO_2 structures were fabricated after annealing with oxygen as part of the annealing gas. The PL tests confirmed that the nc-Si/SiO_2 systems exhibited visible luminescence at room temperature. The morphology and crystalline structure of the nc-Si/SiO_2 layers were characterized by SEM and TEM. The temperature and the gas formation of the annealing ambient are important in the fabrication of nc-Si/SiO_2 structures. We believe that the structures' luminescence properties, analyzed by quantum confinement-luminescence center model, could be designed with the annealing ambient.

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