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Formation and properties of high density Si nanodots

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ABSTRACT

Laser induced crystallization of ultrathin hydrogenated amorphous Si films or amorphous Si-based multilayered structures were used to get high density Si nanodots. The present technique can get size controllable Si nanodots embedded in various dielectric materials with uniform distribution which was revealed by cross-section transmission electron microscopy. Room temperature photoluminescence and electroluminescence were achieved with the emission wavelength in a visible light region both from a-SiN/Si nanodots/a-SiN sandwiched and Si nanodots/SiO_2 multilayered structures. The luminescence was associated with the radiative recombination of generated electron–hole pairs in Si nanodots or the luminescent surface states. The electroluminescence intensity is increased with increasing the injection current implying the bipolar carrier injection plays an important role in enhancing the luminescence efficiency. The formed Si nanodots by the present approach can be applied for many kinds of devices such as high efficient light emitting diodes and solar cells.

1. Introduction

Si nanodots have attracted much attention nowadays both because of the fundamental physical research interesting and the potential applications in future quantum devices as well as the Si-based optoelectronic devices [1,2]. So far, many approaches have been proposed to fabricate Si nanodots. For example, one can get Si nanostructures by using expensive e-beam lithography and reaction ion etching techniques [3]. Another way is to use Si ion implantation into SiO_2 films with subsequent thermally annealing or high temperature annealing (>1000 °C) of off-stoichiometric SiO_x films as well as amorphous Si/SiO_2 multilayers to get Si nanodots embedded in SiO_2 matrix [4–6]. Despite of the extensive research work, it is still an open question to achieve the high density Si nanodots with controllable size by using an approach compatible with today's mature Si technology.

In our previous work, laser induced crystallization of hydrogenated amorphous Si (a-Si:H)/amorphous SiN_x multilayers was proposed to obtain the Si nanodots with controllable sizes [7]. A strong photoluminescence and electroluminescence was achieved at room temperature [7,8]. This simple technique can avoid the high temperature process which is helpful for reducing the damages to the samples and substrates. Furthermore, the laser induced crystallization technique can be used to obtain Si nanodots on ITO coated glass plates due to its low temperature process. This point is the extremely important in fabricating Si nanodots-based photoelectronic devices such as light emitting diodes and solar cells. In order to further understand the formation process and optimize the treatment conditions, the changes of film morphology and photoluminescence behavior were also investigated for laser-annealed ultrathin a-Si:H films (<30 nm) [9,10]. Recently, the electroluminescence from a-SiN/Si nanodots/a-SiN sandwiched structures was obtained at room temperature [11].

Here, we first describe the KrF excimer laser crystallization of single layer of ultrathin a-Si:H films method to obtain Si nanodots on amorphous SiN insulator layers. The formation of Si nanodots was characterized by cross-section transmission electron microscopy (TEM) techniques. It was found experimentally that the size of formed Si nanodots can be limited by the film thickness when the initial a-Si:H film thickness is less than 15 nm [10]. It was found that high density Si nanodots can be formed by controlling the film thickness and the laser irradiation parameters. The theoretical simulation of laser crystallization was performed by using molecular dynamics method. The simulation results suggested that, when the suitable laser energy density is used to treat ultrathin a-Si films, the crystallization of amorphous Si phase can be occurred via the random nucleation and growth process which is well agreement with the experimental observations. Based on the formation of high density Si nanodots, the prototype light emitting devices containing Si nanodots were prepared and the electroluminescence properties were studied. Our results indicated that the laser crystallization of ultrathin a-Si:H films is an appropriative approach to get high density Si nanodots with controllable sizes for future device applications.

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2. Experimental

The ultrathin hydrogenated amorphous Si (a-Si:H) films were prepared on amorphous SiN (a-SiN) layers with thickness of 30 nm in conventional radio frequency (13.56 MHz) plasma enhanced chemical vapor deposition system. The thickness of a-Si:H films was changed from 4 to 30 nm. The sandwiched a-SiN/a-Si:H/a-SiN structures were deposited by successively deposition of a-SiN and a-Si:H layers. Si wafers and quartz glass plates were used as substrates. The detailed deposition parameters can be found elsewhere [9]. After deposition of samples, KrF excimer laser with wavelength of 248 nm and the pulse duration of 30 ns was used to crystallize the as-deposited samples. The laser beam was focused and passed through a rectangle quartz mask to a-Si:H films to get Si nanodots. The irradiated area on the film surface is about 3 mm × 5 mm. Only a single pulse was used to irradiate the samples and the laser fluence was change from 0 to 1.4 J/cm^2. Subsequently, the irradiated samples were thermally annealed at 700 °C for 60 min in pure N_2 ambient. The molecular dynamics (MD) simulation was performed to give the dynamical processes at atomic scale of laser induced crystallization of ultrathin a-Si:H films.

The formation of Si nanodots was revealed by using cross-sectional transmission electron microscopy (TEM) and high resolution TEM technique. Photoluminescence measurements were carried out at room temperature by using Ar^+ laser (488 nm) or He–Cd laser (325 nm) as an excitation source. The electroluminescence (EL) devices were prepared by evaporating Al electrodes both on the front surface (gate electrode, thickness around 30–50 nm) of the films and the bottom of the p-Si wafers (grounded electrode). The EL signals were recorded by the photomultiplier tubes with applying the DC voltage on the devices.

3. Results

Fig. 1(a) is the cross-section TEM image of laser crystallized a-SiN/a-Si:H/a-SiN sandwiched structures. The initial a-Si:H film thickness is about 4 nm. It is clearly shown that the Si nanodots with high density were formed in the initial a-Si:H layer after laser irradiation with the fluence of 0.5 J/cm^2. The size of formed Si nanodots can be well confined by the film thickness and the area density can be as high as 10^{12}/cm^2 under the laser irradiation with a suitable fluence [11]. Fig. 1(b) gives the high resolution TEM image of formed Si nanodots. The spherical Si nanodot can be well identified. The lattice distance is 0.31 nm indicating the growth crystalline surface is (1 1 1) orientation. The TEM results also demonstrated that the amorphous SiN layers still keep their amorphous structures after laser crystallization.

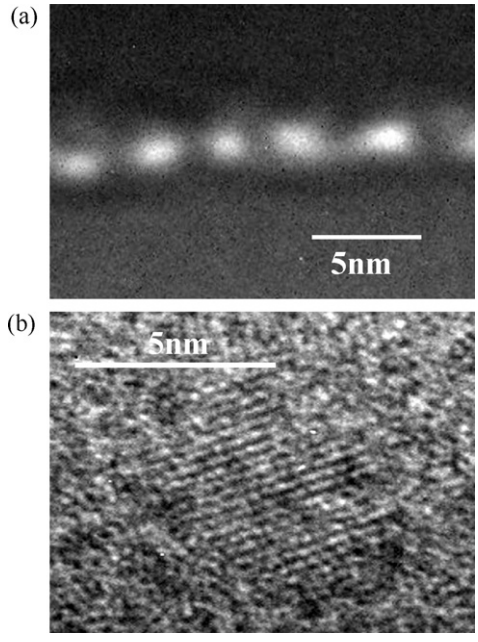

Fig. 1. Cross-section TEM image of laser crystallized a-SiN/a-Si:H/a-SiN sandwiched structures (a) and high resolution TEM image showing the formation of Si nanodot (b).

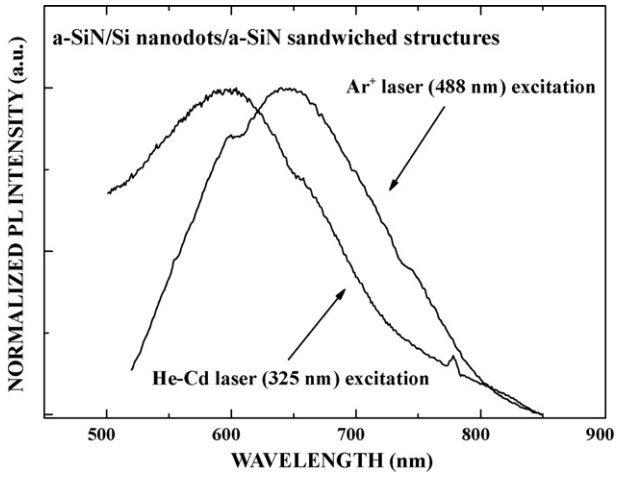

Fig. 2. Room temperature PL spectra from laser crystallized a-SiN/a-Si:H/a-SiN sandwiched structures under the excitation of Ar^+ laser (488 nm) and He–Cd laser (325 nm).

The laser crystallized a-SiN/Si nanodots/a-SiN sandwiched structures exhibit a strong photoluminescence (PL) and electroluminescence (EL) at room temperature. Fig. 2 gives the PL spectra from laser crystallized samples. The PL signals were detected under the Ar^+ laser (488 nm) or He–Cd laser (325 nm) excitation, respectively. Both of the PL band is centered at 650 nm. The PL bandwidth is about 150–200 nm and the PL band excited by He–Cd laser is slightly broaden compared with that excited by Ar^+ laser. It is found that the luminescence signals are very weak for samples only treated by the laser irradiation. After post thermal annealing, the PL intensity is obviously enhanced. It indicates that there may exist amount of defects states which act as non-radiative centers to suppress the luminescence efficiency for laser irradiated samples, post thermal annealing at the moderate temperature (700 °C in our case) can reduce the defects states and improve the luminescence intensity. Another issue is the size distribution of formed Si nanodots. It looks like that the vertical size of Si nanodots can be well confined by the film thickness. However, the broaden PL spectra and the slightly blue-shift of the PL under 325 nm laser excitation implies the existence of size distribution in lateral directions since the more dots with small sizes can be excited by 325 nm laser compared to the 488 nm laser.

4. Discussion

The laser induced crystallization process of ultrathin a-Si layer was simulated by using molecular dynamics method. According to the classical nucleation theory, the driving force of crystallization is the difference between Gibbs free energy of crystalline and amorphous phase. After the irradiation of laser, the film absorbs photons, while the temperature of film increases rapidly. The atoms obtain sufficient kinetic energy across the energy barrier of crystallization and form nuclei of nanocrystalline Si with other atoms and then these nuclei grow up with proper temperature. In the process of crystallization, Si atoms move to the site with the lowest