Superlattices and Microstructures 75 (2014) 136–143

Contents lists available at ScienceDirect

Superlattices and Microstructures

journal homepage: www.elsevier.com/locate/superlattices

Photoluminescence decay properties of Si-rich-oxide/SiO_2 multilayer films with different Si-quantum dots densities

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ARTICLE INFO

Article history:
Received 17 July 2014
Received in revised form 21 July 2014
Accepted 22 July 2014
Available online 7 August 2014

Keywords:
Si quantum dots
Time-resolved photoluminescence
Quantum confinement effect

ABSTRACT

In this work, Si-rich-oxide/SiO_2 multilayer films have been deposited by adjusting the radio-frequency power in a PECVD system, and Si quantum dots (Si-QDs) are obtained in the Si-rich-oxide layer after annealing treatment. FTIR results show that phase separation between Si and SiO_2 occurs after annealing treatment. TEM and Raman spectra show that the size and density of Si-QDs are larger for the multilayer films deposited at 20 W. Compared with the film deposited at 40 W, a 1.7 times PL enhancement is obtained for the film deposited at 20 W, and the PL peak shifts toward low energy. Time-resolved PL spectra show the PL decay for the film deposited at 40 W can be expressed by the multi-exponential decay model, while stretched-exponential decay model can be used to describe the carrier recombination process when the density of Si-QDs is high enough. The PL decay time increases with the red shift of wavelength firstly and decreases, which suggests that quantum confinement effect is the main PL mechanism in the short wavelength region, and localized state transition in Si clusters contributes to the optical emission in the longer wavelength region. The results are further confirmed by low temperature PL spectra.

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http://dx.doi.org/10.1016/j.spmi.2014.07.035

1. Introduction

Nanoscale Si structures have been extensively studied due to their unique optoelectronic properties, and great potentials for light emitters, memory devices, and solar cells have been suggested [1–3]. The multilayer structure which comprises alternating layers of Si quantum dots (Si-QDs) and SiO_2 matrix has been demonstrated as a good candidate due to the excellent confinement effect of the oxide matrix, and the optical bandgap of Si-QDs can be large-scale adjusted due to quantum confinement effect (QCE) [4]. The Si-QDs/SiO_2 multilayer films have been prepared by plasma enhanced chemical vapor deposition technique (PECVD) followed by annealing treatment, and the structural and optoelectronic properties of Si-QDs are usually adjusted by changing the Si-QDs layer thickness and annealing parameters [5,6]. However, they are rarely changed by the plasma parameters, which is very important for the properties of Si-QDs. Hong et al. have developed Si-QDs based thin films solar cells, and the doping and size dependent photovoltaic properties have been investigated [7]. However, the photo-generated carriers transport among the Si-QDs is limited due to high resistance properties of SiO_2 matrix materials, and it should be investigated further to achieve good photovoltaic properties [8]. Photoluminescence (PL) is an effective tool to investigate the carrier transport properties in Si-QDs based system, and the carrier recombination dynamics can be studied by time-resolved PL spectra, which suggests that one can improve the carriers transport efficiency by adjusting the carrier recombination properties [9,10].

In this work, Si-rich-oxide/SiO_2 multilayer films have been deposited by PECVD technique by adjusting the radio-frequency (RF) power, and Si-QDs are obtained by the following annealing treatment. The PL intensity of the films deposited at 20 W is 1.7 times larger than that deposited at 40 W, and the PL peak shifts toward low energy. Time-resolved PL spectra show that stretched-exponential decay model can be used only when the density of Si-QDs is large enough. Further, the PL life time and low temperature PL spectra show that localized state transition in Si clusters is responsible for the carrier recombination in the long wavelength region.

2. Experiment

The Si-rich-oxide/SiO_2 multilayer films were deposited on Si substrate by a PECVD system. SiH_4, H_2 and N_2O were used as reactant gases, and the flow rates of SiH_4 and H_2 were set as 1 and 100 sccm, respectively. The flow ratio of N_2O and SiH_4 were kept at 25 for SiO_2 layer, and 0.1 for the Si rich oxide layer. Deposition pressure, substrate temperature were kept at 120 Pa, 200 °C, respectively. The radio frequency power were set as 20 W and 40 W, and they were defined as S_A and S_B, respectively. Multilayer films consisting of 30 Si-rich-oxide/SiO_2 bilayer sequences were fabricated. After deposition, the samples were annealed at 1100 °C in N_2 for 60 min.

The infrared absorptions were deduced from the transmittance measurement in a Fourier transform infrared spectrophotometer (FTIR, Perkin–Elmer 2000). The Raman spectra were measured by a Jobin Yvon T6400 Raman scattering spectrometer, and the excitation source was a 532 nm Ar^+ laser. The transmission electron microscopy (TEM) images were taken from a JEOL J2010F (S) TEM microscope operating at 200 keV. The steady and time resolved PL spectra were detected by a FLS920 fluorescence spectrometer (Edinburgh Instruments). The excitation sources are 450 W steady Xe lamp and 100 W pulse Xe lamp. The excitation wavelength was 300 nm, and the excitation and emission slits were set as 10 nm and 5 nm, respectively. Temperature dependent PL measurements were performed in a closed-cycle He refrigerator with a temperature range of 8–300 K.

3. Results and discussion

Fig. 1 shows the FTIR spectra of the Si-rich-oxide/SiO_2 multilayer films deposited at different power. The main absorption peak for the deposited film locates at around 1040 cm^{-1}, which corresponds to the asymmetric stretching vibration of oxygen atom in its twofold coordinated bridging bonding site [11]. The main absorption peak moves from 1033 to 1041 cm^{-1} with increasing the RF-power, which suggests that the oxygen content is increased in the Si-rich-oxide layer [12]. The