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Effects of annealing treatments on the photoluminescence decay properties of Si-rich oxide/SiO_2 multilayer films

Xinzhan Wang, Wei Yu *, Xiang Yu, Huina Feng, Wanlei Dai, Xi Wang, Wanbing Lu, Guangsheng Fu

Hebei Key Laboratory of Optic-Electronic Information Materials, College of Physics Science and Technology, Hebei University, Baoding 071002, China

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ABSTRACT

In this work, Si-rich oxide (SRO)/SiO_2 multilayer films have been deposited and the photoluminescence (PL) decay properties of the films with different annealing temperatures are studied. The PL shifts toward low energy with increasing the annealing temperature, and intense PL at around 1.4 eV is obtained after annealing at 1100 °C. The PL decay curves can be well fitted by a multiexponential PL decay model, and the peak of the PL lifetime distribution band shifts toward longer time with increasing the annealing temperature. Two lifetime distribution bands are obtained after the formation of crystallized Si-QDs, and the proportion of slow component increases from 69.72% to 77.04% after hydrogen passivation. Analyses show that defect states recombination in the SRO layer is the main optical emission mechanism when the annealing temperature is lower than 900 °C, and interband transition in the Si-QDs due to quantum confinement effect is the main PL mechanism when the film is annealing at 1100 °C.

1. Introduction

Visible photoluminescence (PL) from porous Si was observed in 1991, and extensive research for underlying the luminescence mechanism has been done in order to realize efficient Si-based light emitters [1–3]. Considerable technologies have been used to fabricate nanoscale Si with visible PL

* Corresponding author. Tel./fax: +86 (0312)5079560.
E-mail address: yuwei_hbu@126.com (W. Yu).

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to overcome the problem of chemical and mechanical instability of porous Si [4,5]. The nanoscale Si should be incorporated to dielectric matrix due to the small size, and the multilayer structure which comprises alternating layers of Si-QDs and SiO_2 matrix has been demonstrated as a good candidate for the excellent confinement effect of the oxide matrix [6,7]. Thermal annealing in tube furnace is often employed to promote the growth of Si-QDs/SiO_2 multilayer films, and the optical emission properties of Si-QDs can be well controlled by changing the annealing environments [8].

Recent results show that quantum confinement effect (QCE) and defect states recombination are two important optical emission processes in the Si-QDs/SiO_2 multilayer films, and the QCE related PL varies with the size of Si nanocrystals, while the PL caused by defect states recombination is size insensitive [9]. Qin and Li have suggested that both QCE and interface defects contribute to the PL in nanoscale Si/Si oxide system, and the interface defect state recombination is the main PL origin when the size of Si-QDs is larger than a certain threshold, while QCE is the main optical emission mechanism when that of Si-QDs is smaller [10]. On the other hand, nonradiative defects such as P_b centers exist at the interface between the Si-QDs and SiO_2 matrix, and they are considered to quench the PL of the Si-QDs based systems [11]. Up to now, it is still a very difficult task to separate the interfacial defects from QCE in the Si-based system due to the lack of effective detection method. Time-resolved PL spectra can be used to investigate the carrier recombination processes in the Si-QDs/SiO_2 multilayer films, and stretched-exponential PL decay curve fittings are usually adopted [12]. The PL in Si-based system is sensitive to the annealing temperature, and the evolution of PL decay properties still needs further investigation. In addition, the carrier recombination in Si-QDs system containing many defects can be achieved by various paths [13]. The PL decay times cannot be expressed as one or several time constants, and PL decay time distribution bands should be adopted to describe them precisely.

In the present work, Si-rich oxide (SRO)/SiO_2 multilayer films have been prepared by plasma enhanced chemical vapor deposition (PECVD) technique, and Si-QDs/SiO_2 multilayer films are obtained by annealing in N_2. Further, the effects of annealing treatments on the PL decay properties of SRO/SiO_2 multilayer films are investigated. Analyses show that two decay time distribution bands are obtained in the crystallized Si-QDs/SiO_2 multilayer film, and they correspond to the decay times of Si-QDs and defect states related optical emission, respectively.

2. Experiment

The SRO/SiO_2 multilayer films were deposited on Si and silica substrates by the PECVD technique. SiH_4, H_2 and N_2O were used as precursor gases, and the flow rates of SiH_4 and H_2 were 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 SRO layer. Deposition pressure, substrate temperature and RF-power were kept at 120 Pa, 220 °C and 40 W, respectively. Multilayer films consisting of 30 SRO/SiO_2 bilayer sequences were fabricated. Below and on top of the multilayer structure, a 10 nm SiO_2 layer was deposited as buffer and capping layer, respectively. After deposition, the samples were annealed from 500 °C to 1100 °C in a step of 200 °C in N_2 for 1 h. In the end, hydrogen passivation is performed on the film annealed at 1100 °C, and the passivation temperature is 450 °C for 1 h.

The infrared absorptions were deduced from the transmittance measurement in a Fourier transform infrared spectrophotometer (FTIR, Perkin–Elmer2000). The transmission electron microscopy (TEM) was performed on 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), and the excitation sources are 450 W steady Xe lamp and 100 W pulse Xe lamp, respectively. The experimental data in the PL measurement were obtained from a time-correlated single photon counting system at room temperature. Distribution analysis of lifetimes was carried out by a program supplied by Edinburgh Analytical Instruments. The fitting function utilized the Fredholm integral and used up to 200 lifetime values, and no a priori assumption about the distribution shape was made.