



Effects of RTA temperatures on conductivity and micro-structures of boron-doped silicon nanocrystals in Si-rich oxide thin films

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ABSTRACT

In this work, the B-doped Si rich oxide (SRO) thin films were deposited and then annealed using rapid thermal annealing (RTA) to form SiO₂-matrix silicon nanocrystals (Si NCs). The effects of the RTA temperatures on the structural properties, conduction mechanisms and electrical properties of B-doped SRO thin films (BSF) were investigated systematically using Hall measurements, Fourier transform infrared spectroscopy and Raman spectroscopy. Results showed that the crystalline fraction of annealed BSF increased from 41.3% to 62.8%, the conductivity was increased from 4.48×10^{-3} S/cm to 0.16 S/cm, the carrier concentration was increased from 8.74×10^{17} cm⁻³ to 4.9×10^{18} cm⁻³ and the carrier mobility was increased from 0.032 cm² V⁻¹ s⁻¹ to 0.2 cm² V⁻¹ s⁻¹ when the RTA temperatures increased from 1050 °C to 1150 °C. In addition, the fluctuation induced tunneling (FIT) theory was applicable to the conduction mechanisms of SiO₂-matrix boron-doped Si-NC thin films.

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1. Introduction

Si nanocrystals (Si NCs) in SiO₂ matrix are of great interest due to their inherent advantages (their strong multiple-exciton generation [1], tunable bandgap [2] and thin-film structures [3]). Impurities can drastically modify the electrical properties, micro-structures and optical properties of Si-NC thin films [4–9]. Boron (B) is one of the most popular methods for preparing p-type Si NCs [4,10]. There are many difficulties in B-doping of Si NCs [11]. For instance, the relatively low diffusion rate of boron in Si rich oxide (SRO) thin films. The diffusion rate of boron atoms is potentially as low as 7×10^{-17} cm² s⁻¹ in silicon dioxide (SiO₂) at 1100 °C [7,12]. In addition, the boron atoms can be trapped at the interface of SiO₂ and Silicon (Si), defects dangling bonds, and extended defects, which causes further impediment to diffusion [7,12,13].

Depositing B-doped SRO thin films and then high-temperature annealing is the most commonly techniques for preparing B-doped Si-NC thin films (BSF) [6–10]. The optical properties, electrical properties and crystallization properties of BSF depend on the annealing process. Hao et al. [7,8] found that the Si-NC size and the crystalline fraction of the B/P-doped silicon-rich SiO₂ thin

films was increased with the increase of tube-furnace annealing temperatures. They also found that the resistivity of BSF was reduced with the increase of furnace annealing temperatures. Several experimental studies for preparing the Si-NC thin films were reported including tube-furnace annealing [5–7], laser annealing [14,15], and rapid thermal annealing (RTA) [9,16,17]. RTA is relatively simple, cost- and time-effective approach among them [4,16,17]. RTA treatments can lead to comparable crystallographic quality and crystallization of Si-NC thin films [10,16,17]. In addition, RTA treatments was laser-radiation heating which can facilitate the diffusion of B, O and Si atoms [16,17]. Huang et al. [4] and Cheng et al. [10] reported that the fabrication of B-doped Si-NC thin films using magnetron sputtering deposition and RTA. It was found that the B-doped SRO thin film deposited under optimal substrate temperatures could significantly increase the conductivity and crystallization of Si-NC thin films. Zeng et al. [9] reported that the crystalline fraction of phosphorus (P)-doped Si_{0.85}C_{0.15} thin films was increased, and the activation energy of conductivity reduced with the increase of RTA temperatures.

It should be noted that the annealing temperatures has significant impacts on the properties of the BSF [5–8]. To our knowledge, the effects of RTA temperatures on the electrical properties and structural properties of BSF are still not well studied, and the conduction mechanisms of BSF still require further

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investigation. In this work, BSF were deposited at 100 °C and were annealed using RTA. The effects of RTA temperatures on the electrical properties, conduction mechanisms and structural properties of BSF were investigated systematically using Hall measurements, Fourier transform infrared spectroscopy (FTIR) and Raman spectroscopy.

2. Experimental details

The J-sputter8000 magnetron sputtering system was used for BSF deposition on quartz glass (1×1 cm) and double-sided polished silicon wafers (1×1 cm). Firstly, the substrates were immersed in the ultrasonic baths of acetone and distilled water for about 15 min each. The silicon substrates were dipped in a dilute (6%) HF solution for clear of the surface native oxide. The substrates were rinsed with deionized water, and then blow-dried with nitrogen gas. The substrates were immediately transferred to an ultrahigh vacuum chamber for BSF deposition. The resistivity of heavily B-doped Czochralski silicon was about $1.0 \times 10^{-3} \Omega \text{ cm}$, this resistivity corresponded to a B concentration of $\sim 1.17 \times 10^{20} \text{ cm}^{-3}$ [18]. The radio frequency (RF) power of heavily B-doped Czochralski silicon targets (4 in.) and SiO_2 targets (4 in.) were set as 120 W and 45 W, respectively. Upon achieving a base pressure of 8×10^{-6} Torr, and then the Ar was introduced to the chamber for a working gas pressure of 1.6×10^{-3} Torr. The thickness of the as-deposited B-doped SRO thin films was ~ 400 nm which measured by the profilometer (Veeco Dektak150). The O/Si ratios of the as-deposited thin films was ~ 0.7 as determined by X-ray photoelectron spectroscopy (XPS, Shimadzu, AXIS ULTRA^{DLP}). After deposition, the BSF were subjected to RTA at 1050–1150 °C for 60 s in Ar atmosphere with the heating rate of 20 °C/s and furnace cooling.

The chemical compositions of both as-deposited and annealed BSF were analyzed by XPS. The XPS measurements were carried out after a 120 s Ar ion sputter etch to remove surface oxidation. The Ar ion bombardment etching rate was approximately 2.0 Å/s. The XPS spectra were calibrated by using the C 1 s peak (284.5 eV). The chemical composition was also analyzed by FTIR in the spectral range between 400 and 2000 cm^{-1} with a resolution of 1 cm^{-1} . The double-sided polished silicon wafers were used as substrates. The structures of 1100 °C-annealed thin films was measured by Transmission Electron Microscopy (TEM, FEI Tecnai F20). The crystalline fraction of annealed BSF thin films were determined by confocal micro-Raman spectroscopy (Renishaw, InVia-Reflex) with the excitation of a Nd:YAG laser (532 nm). Here, the sample was measured three times, and the root mean square was used as the error bar [19]. Conductivity, carrier concentration, mobility and temperature-dependent dark conductivity (190–480 K) of thin films were determined by a Hall system (Nano-metrics, HL5500PC).

3. Results and discussion

Fig. 1(a) shows the FT-IR spectra of as-deposited BSF and were annealed using RTA at 1050–1150 °C for 60 s. The peaks at about 460, 808, 1000–1100 cm^{-1} are due to the O–Si–O rocking, Si–O–Si bending and Si–O–Si asymmetrical stretching vibrations, respectively [20]. The increased of peak position indicated that the $\text{Si-Si}_x\text{-O}_{4-x}$ ($1 < x < 3$) clusters were separated into Si-Si_4 and Si-O_4 clusters in the SRO thin films [5,6,9,20]. The peaks of O–Si–O rocking and Si–O–Si bending clearly resolved and increased when the RTA temperature increased. In addition, the asymmetrical Si–O–Si peak of thin films was positioned at 1028, 1090, 1094, 1095 cm^{-1} , respectively, as shown in Fig. 3(b). The results indicated that increasing RTA temperatures would facilitate the phase separation of BSF.

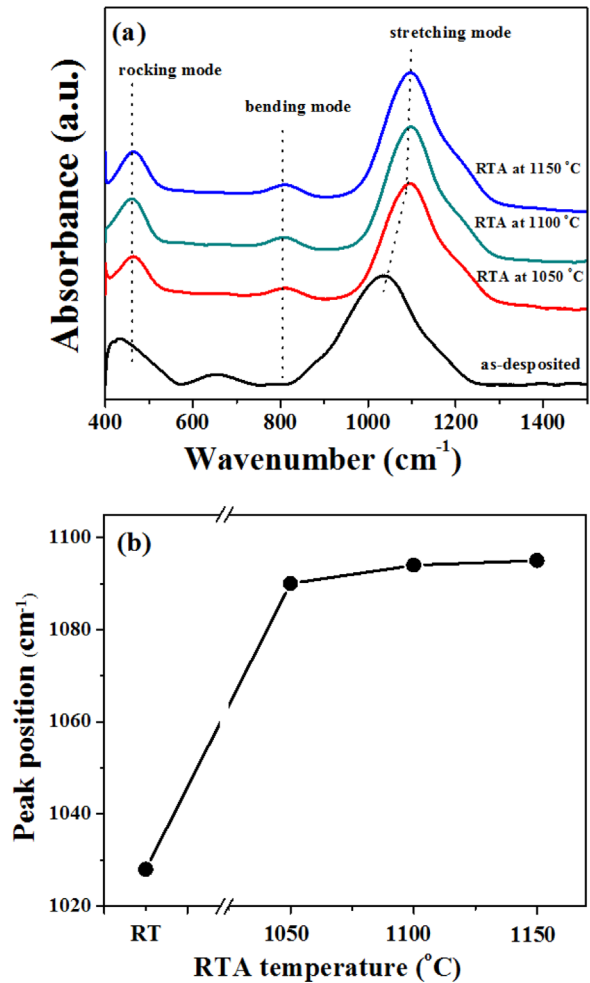


Fig. 1. (a) The FT-IR spectra of as-deposited BSF and were annealed using RTA at 1050–1150 °C for 60 s; (b) the asymmetrical Si–O–Si peak position of the annealed thin films.

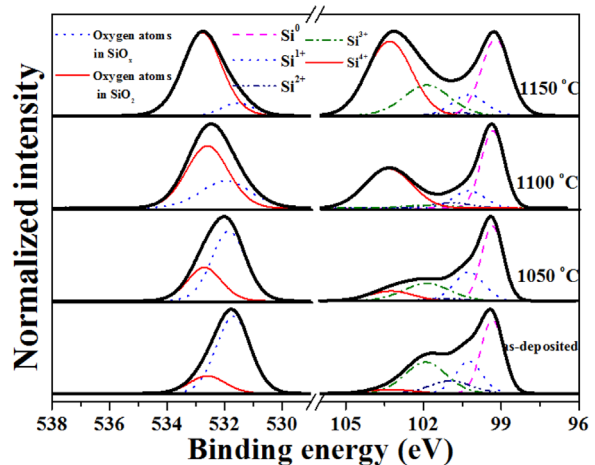


Fig. 2. The XPS spectra of O 1s and Si 2p peaks of as-deposited BSF and annealed thin films.

Fig. 2 shows the XPS spectra of Si 2p and O 1s peaks of as-deposited SRO thin films and annealed thin films. It should be mentioned that the B signal was not measured in the XPS spectra due to the low concentration and atomic sensitivity factors of B. The Si 2p spectra of samples have been deconvoluted into the five possible chemical states, corresponding to Si^0 (99.3 eV), Si^{1+}

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