



## Short communication

Effect of SiO<sub>2</sub> layers on electroluminescence from Si nanocrystal/SiO<sub>2</sub> superlattices prepared using argon ion beam assisted sputtering

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## ABSTRACT

This study investigated the electroluminescence (EL) properties of Si-rich oxide (SRO)/SiO<sub>2</sub> superlattices light emitting devices (LEDs). Each SiO<sub>2</sub> layer of the superlattices was prepared by using argon ion beam assisted sputtering (IBAS). Transmission electron microscopy revealed that the treatment of Ar ion beams on the SiO<sub>2</sub> layers did not affect the size or distribution of the Si nanocrystals in the SRO layers, but enhanced the thin-film quality of the SiO<sub>2</sub> and formed a clear SiO<sub>2</sub>/SRO interface. The refractive index of SiO<sub>2</sub> was increased by IBAS because of an increase in the density of SiO<sub>2</sub>. The EL efficiency was doubled for the IBAS device compared with that of a reference device. According to the retention property, the enhanced EL intensity of the IBAS device was ascribed to lower the charge loss rate through enhancing injection barrier of SiO<sub>2</sub>. The mechanism of the EL enhancement of the IBAS LED was discussed.

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Strong visible photoluminescence at room temperature from porous Si was discovered in the 1990s [1], prompting the development of solid-state light-emitting devices (LEDs) composed of the most abundant elements on earth such as Si. Moreover, Si-based luminescent devices have attracted considerable interest because of their compatibility with current Si fabrication processes that is attributable to the development of integrated circuits. Because Si-rich oxide (SRO)/SiO<sub>2</sub> superlattices structures provide higher quantum efficiency compared with bulk SRO-based devices, intense research on this structure has rapidly progressed since 1995 [2]. In SRO/SiO<sub>2</sub> systems, ultrathin SiO<sub>2</sub> barrier layers thinner than 5 nm are common [3], and the nucleation of Si nanocrystals (NCs) formed by the precipitation of the excess Si in SRO is achieved through thermal annealing.

The emission efficiency of Si-based LEDs remains too low to satisfy the needs of light sources. To date, various approaches have been developed to improve the light emission efficiency of Si-based LEDs. Previous studies have focused mostly on the properties of Si NCs produced within the SRO layer such as the size modulation [4], passivation [5], stoichiometry [6], and oxygen-related defects [7]. On the other side, investigations into barrier layers formed through Si nitride [8], Si carbide [9], and Si oxynitride [10] have extensively examined the trade-off between carrier injection and confinement.

Previously, we demonstrated that Ar ion beam treatment on the SRO layer of SRO/SiO<sub>2</sub> superlattices can improve the crystallinity of Si NCs and introduce radiative defects that yield white photoluminescence [11]. In the current study, we investigated the effect of Ar ion beam assisted sputtering (IBAS) on the SiO<sub>2</sub> layer in SRO/SiO<sub>2</sub> superlattices. Improvement of the SiO<sub>2</sub> quality was first reported as another crucial factor for enhancing the electroluminescence (EL) intensity of Si NC/SiO<sub>2</sub> LEDs. IBAS increased the density of SiO<sub>2</sub> layers and improved the interfacial quality, reducing the probability of carriers escaping the Si NCs. Consequently, the EL intensity was markedly increased. This paper discusses the electrical characteristics and light emission performance of Si NC LEDs.

The samples were developed on p-type Si substrate wafers with a resistivity of 0.01 Ω-cm. Superlattices of SRO/SiO<sub>2</sub> were alternatively deposited through dc sputtering of a pure Si target and radio-frequency sputtering of a SiO<sub>2</sub> target. The base pressure of the chamber before deposition was less than  $5 \times 10^{-6}$  Torr, and the working pressure was  $8 \times 10^{-4}$  Torr. Each SiO<sub>2</sub> layer was bombarded by an Ar ion beam with 100 eV immediately after deposition. Ar gas was fed into the chamber and an end-hole gridless ion gun at a flow rate of 20 and 3 sccm, respectively. The total thickness of the superlattices was 40 nm for both samples. In addition, a 10-nm-thick SiO<sub>2</sub> layer was prepared through sputtering or IBAS for refractive index measurement.

Post annealing was performed at 1000 °C for 3 h in a furnace with a forming gas (N<sub>2</sub> 95% and H<sub>2</sub> 5%) under the pressure of 1 Torr for Si NC nucleation. A 200-nm-thick Al-doped ZnO (AZO) electrode

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was sputtered on top of the superlattices. The front Al layer was patterned on the AZO film by using a shadow mask to form the top electrode. A 700-nm-thick Al layer was evaporated at the backside of the Si substrate and then soft-annealed in pure nitrogen (300 °C, 30 min) for ohmic contact.

The thickness and formation of Si NCs were confirmed using high-resolution transmission electron microscopy (HR-TEM, JEOL/JEM-2100). The refractive indices of the 10-nm-thin SiO<sub>2</sub> layer were determined using a variable angle spectroscopic ellipsometer (J. A. Woolam/M2000-DI). The electrical characteristics of the devices were examined according to the current–voltage (I–V) characteristics (Keithley/2400) and capacitance–voltage (C–V) measurement (Agilent/E4980A). EL measurements were performed using a Zolix omni-λ300 spectrometer coupled to a photomultiplier tube at room temperature.

A schematic of Al/P–Si/(SRO/SiO<sub>2</sub>)<sub>11</sub>/AZO/Al structures is presented in Fig. 1(a). Fig. 1(b) and (c) show the cross-sectional HR-TEM images for the superlattices prepared through sputtering and IBAS, respectively. The multilayered structure and crystalline appearance of the Si NCs in the SRO layers are clearly identifiable in the figure. The thicknesses of the SRO and SiO<sub>2</sub> layers were approximately 2.9 nm (dark region) and 1.1 nm (light region) for both samples, respectively. The size (approximately 2.6 nm) and density of the Si NCs (approximately  $8 \times 10^{17} \text{ cm}^{-3}$ ) of the two samples were similar. This result indicated that the Ar ion beam treatment of the SiO<sub>2</sub> layers did not affect the size or distribution of the Si NCs in the SRO layers. By contrast, the bright TEM image of the SiO<sub>2</sub> layer relative to the SRO layer was clear for the IBAS sample (Fig. 1(c)), whereas the SRO/SiO<sub>2</sub> interface was blurred for the sputtered sample (Fig. 1(b)).

To identify the influence of the Ar ion beam on the SiO<sub>2</sub> layer,

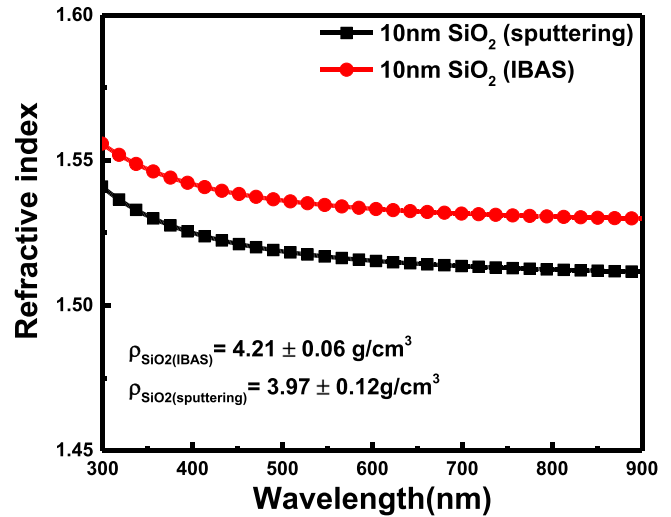


Fig. 2. Refractive index of 10-nm-thin SiO<sub>2</sub> prepared through sputtering and IBAS.

two 10-nm-thick SiO<sub>2</sub> films were separately prepared on the Si substrate through sputtering and IBAS for comparison. Fig. 2 shows the refractive index of SiO<sub>2</sub> films prepared through IBAS and sputtering. The refractive index of the SiO<sub>2</sub> in visible region was increased for the IBAS sample. For example, it increased from  $1.51 \pm 0.002$  (sputtered) to  $1.53 \pm 0.002$  (IBAS) in the visible region at 633 nm, indicating that the density of the SiO<sub>2</sub> was increased by IBAS [12]. Ar ion beams with a kinetic energy lower than 100 eV mostly affect the surface reaction during sputtering. Therefore, weak Si–O would be broken during the IBAS process, thus

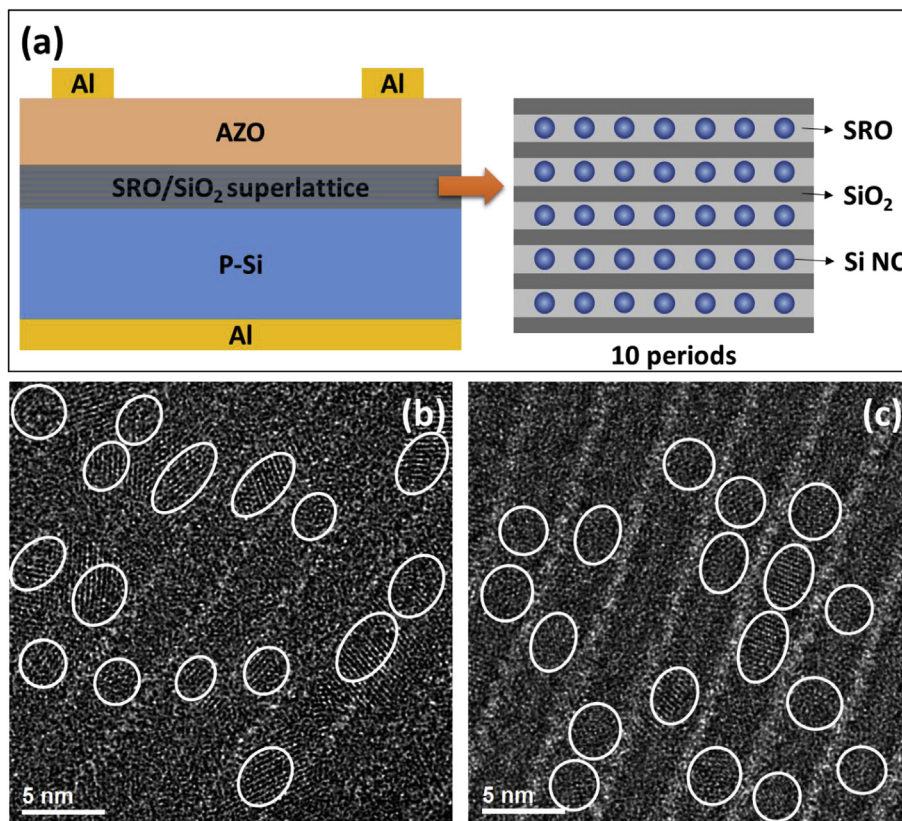


Fig. 1. (a) Schematic of the Si NC/SiO<sub>2</sub> superlattices structure. HR-TEM images of the SRO/SiO<sub>2</sub> superlattices (b) without and (c) with IBAS on SiO<sub>2</sub>.

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