Low-Energy Ion-Beam-Assisted Sputtering for Si Nanocrystals

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1. Introduction

Si-rich oxide (SRO) with embedded Si nanostructures has attracted considerable attention owing to pronounced quantum effect after annealing, regardless of the indirect band-gap nature of Si. The SRO films have been extensively studied for use in light emission devices, photodetectors, and solar cells [1-5]. Sputtering, evaporation, and plasma-enhanced chemical vapor deposition (PECVD) have been used to synthesize the SRO films. A thermal annealing is required to form the Si nanocrystals (NCs) in the oxide matrix undergoing a phase separation process [6-8]. However, the required temperature is high (>1000 °C), limiting the applicability to integrate with the Si-circuit technology. Additionally, the optical and electrical properties of the SRO films are strongly associated with the size and density of Si NCs. However, ways to control the size and density of Si NCs are still desired.

This work reports the low-energy ion-beam assisted sputtering (IBAS) for synthesis of the SRO films. The ion beams provided additional energy for the species to form crystallinic bonding and to crack the large radical in plasma. The IBAS effectively increased the density of Si NCs and lowered the required annealing temperature. Additionally, the IBAS showed capability to modulate the size and density of Si NCs in SRO films. Room-temperature photoluminescence for the un-annealed SRO films that was prepared by the IBAS was observed. The SRO films that were prepared by sputtering and IBAS were compared, and the corresponding material and optical properties were investigated.

2. Experimental details

The SRO films were deposited on P-type (100) Si wafers by sputtering and IBAS at room temperature using an identical mixed Ar/O₂ flow (Ar:O₂=20:0.3 sccm). The ion beam was generated through an end-hole gridless ion source that equipped a plasma-bridge neutralizer. The anode voltage of the ion source, which was associated with the ion energy, was 20, 40 and 66 V. The surface of Si wafer was cleaned by a standard procedure. The rf-power for sputtering was 110W, and the working pressure was 1×10⁻³ torr. The working pressure was well-chosen so that it is between the working ranges of both the sputtering and IBAS. All of the SRO films were post-annealed at 1000 °C for 3h in a furnace. The Si NCs were observed by high-resolution transmission electron microscopy (HRTEM/Hitachi HF-2000). The X-ray photoelectron spectroscopy (XPS/Ulvac-PHI) was used to determine the oxidation states and the composition of the SRO films. Room temperature Photoluminescence (PL) spectroscopy was used to observe the quantum confinement of the Si NCs, which is in the range 350-640 nm and uses a 325nm He-Cd laser as the excitation source.

3. Discussion

Figure 1 presents schematic demonstration of the IBAS. Notably, the radicals from sputtering and the Ar ions from the ion source encountered near the substrate surface. Figure 2(a)-(d) show the HR-TEM images of the SRO films that was prepared by IBAS. All samples were annealed at 1000°C for 3h.

Fig. 1 Schematic diagram of the ion-beam assisted sputter-ing system.

Fig. 2 TEM images of the SRO layer prepared by sputtering (a), and IBAS with ion beam voltages (b)20V (c) 40V (d) 66V. All samples were annealed at 1000°C for 3h.
than those prepared by sputtering as the anode increased from 0 to 40 V. The size and density decreased when the anode voltage exceeded 40 V.

Figures 4(a)-(d) show the XPS Si 2p signals of the SRO films before and after annealing at 1000°C for 3h. The annealing increased the $\text{Si}^{0+}$/$\text{Si}^{4+}$ ratio for samples that were prepared by IBAS; while the annealing decreased the $\text{Si}^{0+}$/$\text{Si}^{4+}$ ratio for those prepared by sputtering. The $\text{Si}^{0+}$/$\text{Si}^{4+}$ ratio increased monotonically as the anode voltage of the ion source was increased from 0 to 40 V. When the anode voltage was increased further to 60 V, the $\text{Si}^{0+}$/$\text{Si}^{4+}$ ratio decreased. This fact is similar with the observation of the size and density of Si NCs. We ascribed the variation of the $\text{Si}/\text{O}$ ratio to the ion energy that influenced the content of oxygen of the SRO films. The content of oxygen contributed to the volume expansion of SiO$_2$ from phase separation, changing the stress inside the SRO films. Moreover, the IBAS provided additional energy and lowered the barrier to form the Si NCs, reducing the required annealing temperature.

4. Conclusion

This work reports the low-energy ion-beam assisted sputtering (IBAS) for synthesis of the SRO films. The IBAS effectively increased the density of Si NCs and lowered the required annealing temperature. Additionally, the IBAS showed capability to modulate the size and density of Si NCs in SRO films. Room-temperature PL was observed for the un-annealed SRO films that was prepared by the IBAS (40 V). The annealing increased the PL intensity and the emission wavelength. Finally, we suggest the proposed method will be useful for future development of Si NCs.

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References