Enhanced electroluminescence from nanocrystallite Si based MOSLED by interfacial Si nanopyramids

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The interfacial Si nano-pyramid-enhanced electroluminescence (EL) of an ITO/SiO$_x$/p-Si/Al metal-oxide-semiconductor (MOS) diode with turn-on voltage of 50 V, threshold current of 1.23 mA/cm^2, output power of 16 nW, and lifetime of 10 h is reported.

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Si nano-structures or Si nano-crystals (nc-Si) embedded in SiO$_x$ grown by the plasma enhanced chemical vapor deposition (PECVD) have attracted great interests, since they can easily deposit higher density of excess silicon and precipitate more nc-Si in oxide matrix after annealing. Such Si nano-structures or nc-Si embedded in SiO$_x$ film exhibited near-infrared (NIR) photoluminescence (PL) and electroluminescence (EL) at room temperature[1−3]. In this work, we present the growth of Si nano-pyramids at the SiO$_x$/Si interface by using low-power PECVD. The distinct modification on the carrier transport through such a new oxide system and the improved EL response at relatively low bias are compared with those obtained from a conventional device. The role of these Si nano-pyramids on the stable NIR luminescent mechanism of an indium tin oxide (ITO)/SiO$_x$/p-Si/Al metal-oxide-semiconductor (MOS) diode is also investigated.

In the experiment, the Si-rich SiO$_x$ film was deposited on p-type (100)-oriented Si substrate by PECVD at chamber pressure of 60 mtorr. The N$_2$O and SiH$_4$ fluences were controlled at 150 and 30 sccm, respectively, while the induced couple plasma (ICP) power was varied from 60 to 45 W. Typically, the sample was deposited at substrate temperature of 300 °C with an ICP power of 40 W. In our case, the substrate temperature was raising up to 400 °C and the ICP power was decreasing to near threshold, which resulted in the *in-situ* synthesis of Si nano-pyramids on the Si substrate prior to the growth of the Si-rich SiO$_x$. After deposition, the SiO$_x$ samples were annealed in a quartz furnace at 1100 °C with flowing N$_2$ gas for 1 h, which induced the formation of nc-Si embedded in the deposited SiO$_x$ film. A 200-nm ITO film layer with resistivity of 33 Ω·cm was deposited on the top of Si-rich SiO$_x$ with diameter of 0.8 mm to form the MOS diodes. A 500-nm Al contact electrode coated on the bottom of Si substrate was alloyed at 450 °C for 7.5 min.

As a result, nc-Si dependent on NIR EL from a PECVD-grown Si-rich SiO$_2$ MOS diode has been investigated. For the PECVD-grown Si-rich SiO$_x$ annealed at 1100 °C for 30 min, the peak PL at 760 nm attributed to the nc-Si is observed and a supporting evidence of the nc-Si with diameters of $4-5$ nm is given by high-resolution transmission electron microscopy (HRTEM) analysis. Longer annealing time induced re-oxidation effect of nc-Si inevitably makes the PL broaden and blue-shift. The difference between white-light and NIR ELs of the MOS diodes fabricated on 1100 °C annealed Si-rich SiO$_x$/p-Si substrate with the buried pyramid Si nano-pyramids is characterized. By changing the substrate temperature and ICP power during the PECVD of Si-rich SiO$_x$ films, the effects of growth condition on the defect-related and Si nano-pyramids related carrier transport and EL spectroscopy are also investigated. Figure 1 shows the HRTEM images of the annealed Si-rich SiO$_x$/p-Si films grown on the synthesized substrate at higher temperate of 350 °C, which show the more Si nano-pyramids precipitate near Si/SiO$_2$ interface. By decreasing the ICP power to a threshold of 30 W, the (100)-oriented triangular Si nano-pyramid significantly grows up along the Si/SiO$_x$ interface with height and bottom width of 10 and 20 nm, respectively. From the spectra shown in Fig. 2, it is concluded that the luminescent components at $700-850$ nm is mainly contributed by the recombination in Si nano-pyramids and self-trapped exciton (STE). The ITO/Si-rich SiO$_x$/p-Si/Al MOS diodes with Si-rich SiO$_x$ films under different grown conditions exhibit nearly similar PL spectra, but their completely different white-light EL spectra at wavelength from 400 to 600 nm are also observed.

Compared with the EL spectra of MOS diodes made on as-grown and annealed Si-rich SiO$_x$, the Si nano-pyramids related EL spectra at $650-850$ nm are

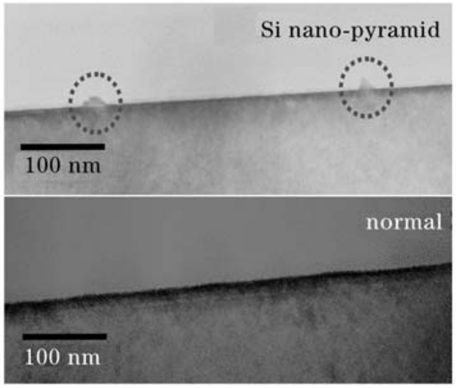

Fig. 1. HRTEM of Si-rich SiO$_x$ with and without Si nano-pyramids at the SiO$_x$/Si interface.

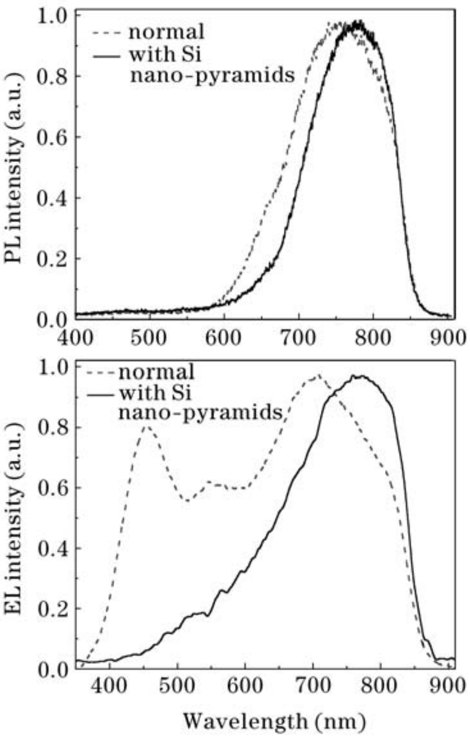

Fig. 2. EL and PL spectra of Si-rich SiO_x based MOS diode without (dashed) or with (solid) interfacial Si nano-pyramids.

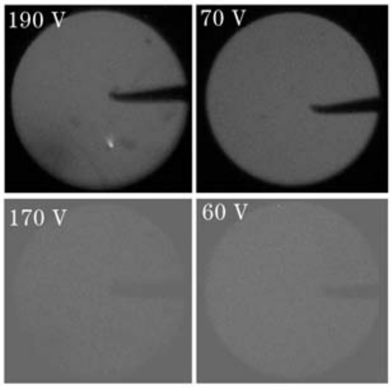

Fig. 3. EL emission patterns of Si-rich SiO_x based MOS diodes without (lower) or with (upper) interfacial Si nano-pyramids.

confirmed, whereas the EL spectra at shorter wavelengths (400 − 650 nm) are attributed to the radiative defects such as the weak-oxygen bond (415 nm), the neutral oxygen vacancy (NOV) defects (455 nm), and the E'_δ defect (520 nm). Both the radiative defects and the Si nano-pyramids can be the EL center, however, the carrier transport as well as the luminescent mechanisms among the defect centers are more pronounced than those contributed by Si nano-pyramids. These defects become an electron-preferred transporting path within the Si-rich SiO_x film, whose densities decrease with increasing substrate temperature or reducing ICP power. As shown in Fig. 3, the defect-related white-light EL occurs on the Si-rich SiO_x sample grown at lower temperature (300 °C) and higher ICP power (40 W), such a MOS diode emits the largest power at a relatively short lifetime. Lengthening the lifetime of device can be achieved by raising the substrate temperature to the system limit (350 °C), however, the EL emitting power is simultaneously attenuated to 30 nW with a reducing defect density. The PECVD process temperature increasing to 350 °C and ICP power decreasing to threshold of 30 W effectively promote the density of Si nano-pyramids in the Si-rich SiO_x film with Si concentration up to 40 at.-%.

A nearly defect-free Si-rich SiO_x sample can be grown under such condition, which contributes to a most stable NIR EL with the longest lifetime although the power of pure Si nano-pyramids related EL at NIR wavelengths is slightly lower (16 nW). In Fig. 4, the forward turn-on voltage, current density, maximum output power, and optical power versus current (P-I) slope of the optimized MOS diode made on the Si-rich SiO_x sample grown at the highest substrate temperature and lowest ICP power are 50 V, 1.23 mA/cm^2, 16 nW, and 0.01 mW/A, respectively. At last, we have implemented a PECVD-grown SiO_x based distributed Bragg reflector (DBR)/SiO_x/Si (larger than 40 nm)/DBR MOS diode light emitting diode with resonance-enhanced cavity. The room-temperature PL at the wavelength range from 700 to 800 nm is enhanced by high reflectance of quarter-wavelength $\mathrm{SiO}_2/\mathrm{TiO}_2$ films. However, the 40-nm silicon electrode under the SiO_x film is too thin to work properly after 1100 °C annealing. The silicon electrode could be thinner to prevent damages from 1100 °C annealing. However, the ITO/Si-rich SiO_x/Si/Al MOS diode exhibits a smaller output power of 7 nW with a extremely high turn-on voltage of 86 V, which is due to the less pronounced effect of carriers tunneling through such a better crystalline PEVCD-grown SiO_x. In comparison, the EL at longer wavelength coincides well with the PL, which reveals that the nc-Si related PL and EL are

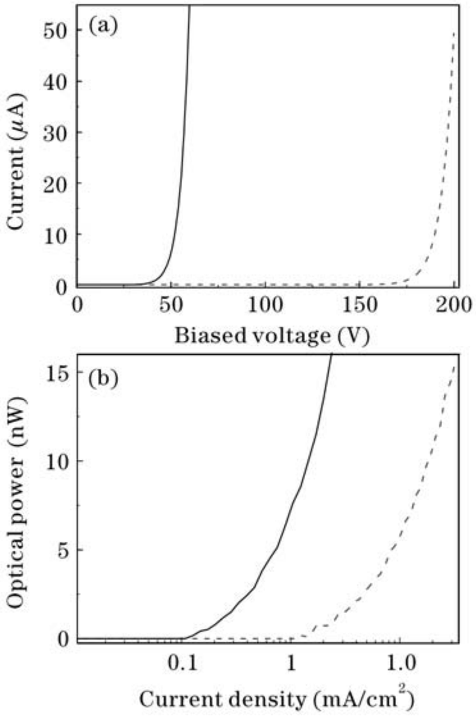

Fig. 4. (a) I-V and (b) P-I curves of Si-rich SiO_x based MOS diodes with (solid) or without (dashed) interfacial Si nano-pyramids.

attributed to the same carrier recombination mechanism in the buried nc-Si. The EL of the ITO/Si-rich SiO_x/Si/Al MOS diode is mainly due to the transfer of cold carriers by direct tunneling between adjacent nc-Si. The cold-carrier tunneling process from lower excited state to higher excited state between two adjacent nc-Si quantum dots under high electric field is primarily elucidated. The difference between PL and EL results is also explained with the proposed mechanism.

In conclusion, the enhanced EL mechanism of an ITO/SiO_x/p-Si/Al MOS diode with the premier synthesis of Si nano-pyramids at the SiO_x/Si interface using low-power PECVD is investigated. The (100)-oriented Si nano-pyramid significantly grows up at Si/SiO_x interface with 10-nm-height and 20-nm-bottom-width PECVD grown under a threshold plasma power condition. The ITO/SiO_x/p-Si/Al MOS diodes with and without Si nano-pyramids exhibit nearly similar PL spectra, but the completely different white-light EL spectra at $400-600$ nm are also observed. Increasing the substrate temperature during the PECVD process effectively promotes the density of Si nano-pyramids in the nearly defect-free Si-rich SiO_x film with Si concentration of 40 at.-%. The carrier transport as well as the luminescent efficiency in such a MOS diode with the buried Si nano-pyramids at SiO_x/Si interface become more pronounced. This contributes to a most stable NIR EL at 800 nm and facilitates the device from emitting power at relatively longer lifetime ($>$ 10 h). The forward turn-on voltage, threshold current density, maximum output power and the slope of the P-I curve of the optimized ITO/SiO_x/p-Si/Al MOS diode are 50 V, 1.23 mA/cm^2, 16 nW, and 0.01 mW/A, respectively.

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