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Light Emitting Properties of Silicon Nanocrystallite Layer Synthesized by Pulsed Laser Ablation in Inert Background Gas

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1. Introduction

Light emitting properties of nanostructured Si have not only scientific interests but also technological advantages [1]. There have been numerous reports concerning light emission from porous Si formed by liquid phase anodization [2-3]. Besides the porous Si, various other methods forming the nanostructured Si have been actively studied. Pulsed laser ablation (PLA) is one of the promising method for synthesizing functional Si nanocrystallites [4]. However, in the case of coalesced nanocrystallites in the deposited layer by using PLA, quantum confinement effects of the Si nanocrystallites disappear as well as be weakened.

In this work, we synthesize the Si nanocrystallite layers by using PLA in inert background gases (PLA-IBG), optimizing the background gas pressure for remarkable quantum confinement effect appearance in optical properties. Finally, the Si nanocrystallite layers are adopted for active regions of visible light emitting diodes (LEDs).

2. Experimental

We adopted a tunable pulse width second harmonic Nd: YAG laser (wavelength: 532 nm) as an excitation light source for the Si ablation. The laser beam was focused onto the surface of the single crystalline Si target (*p*-type (100)). He gas was introduced into the reactor and maintained at a constant pressure. The target-substrate distance was fixed at 20mm. We surveyed the optimized condition for synthesizing the Si nanocrystallites, by varying the pulse width of the excitation laser for droplet/debris suppression and inert background gas (helium: He) pressure for the optical quantum confinement effect appearance.

For the LED fabrication, the Si nanocrystallite layers were deposited using the optimized PLA-IBG condition (pulse energy: 10 mJ, pulse width: 42 ns, He gas pressure: 3.75 Torr). The as-deposited mean diameter and standard deviation were about 4.3 nm and 1.9 nm, respectively. Substrates were (100) oriented *p*-type Si wafers with a resistivity of 0.02 Ω•cm (**Fig. 1**). Thermal oxide layers were formed with a thickness of 300 nm for the isolation dielectrics. Electrically active regions with a diameter of 1.0 mm were defined by removing the thermal oxide. The optimized Si nanocrystallite layers were deposited using the above PLA-IBG method. Thickness of the active regions were about 250 nm. Thermal annealing was carried out at 825 °C for 10 min in N_2 gas. In order to form contacts with the Si nanocrystallite layers, semitransparent platinum (Pt) films were deposited with a thickness of 12 nm.

3. Results and Discussion

In the cross-sectional HRTEM observation for the Si nanocrystallite layer (**Fig. 2**), white circles indicate lattice plane image of single crystalline Si nanoparticles. These lattice plane spacing corresponded to (111) plane of the bulk Si. The droplets and the debris were not observed at all.

Figure 3 shows the influence of the He gas pressure on optical absorption spectra of the Si nanocrystallite layer. The absorption edges of the Si nanocrystallite are blue-shifted, in comparison with that of bulk Si (1.1 eV). In particular at 3.75 and 4.0 Torr, the absorption edges shift remarkably, and the band gap energies were extracted to be about 1.5 eV. These results propose that nanometer-sizing of Si caused carrier confinement effects which increase the band gap energies.

Figure 4 shows the influence of He gas pressure on Raman scattering spectra of the Si nanocrystallite layer. At 3.75 and 4.0 Torr, the crystalline peaks are broadened and shifted to lower wave numbers. In general, phonon confinement effects cause the broadening and the lower wave number shift of the peak profiles.

Consequently, quantum confinement effects for both carriers and phonons remarkably apperared in the Si nanocrystallite layer deposited at the He pressure of 3.75-4.0 Torr. In the lower pressure region (1.0-2.5 Torr), deposited Si layers had properties of amorphous Si films. In the higher pressure region (5.0 Torr), deposited Si nanocrystallites coalesced each other. In this case, the quantum confinement effects become weak (absorption) or disappeared (Raman).

Weak rectifying behavior is observed in *I-V* characteristics of the LED (**Fig. 5**). The onset of the emission was at forward bias of 5.0 V and current density of 35 mA/cm^2. Furthermore, the integrated light emitting intensity (I_{EL}) shows abrupt nonlinear dependence on the forward current j: $I_{EL} \sim j^m$, m=1.8-2.2 (**Fig. 6**). We have observed visible spectrum of the LED at room temperature. **Figure 7** shows the spectrum measured at the dissipation power of 1.6 W. The spectrum is composed of Gaussian wave forms of a main unit peaked at 1.74 eV, and sub-units peaked at 2.07 eV and 2.25 eV.

A possible mechanism of this emission is impact ionization by tunneling hot electrons through the surface oxide layers and the subsequent radiative recombination. The nonlinearity is attributed to dependence of the impact ionization quantum efficiency on acceleration energies of the tunneling hot electrons.

4. Conclusions

We have developed the visible LEDs whose active layers are Si nanocrystallite layers having remarkable quantum confinement effects, synthesized by the PLA-IBG process.

Acknowledgments

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References

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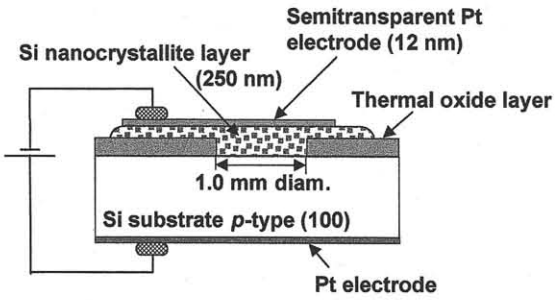

Fig. 1 Schematic cross-sectional view of the LED with the Si nanocrystallite active layer.

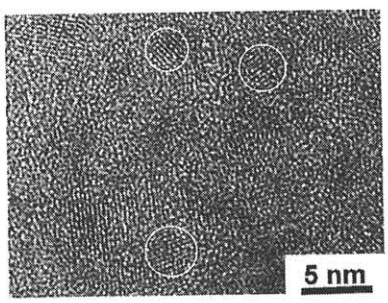

Fig. 2 Cross-sectional HRTEM micrograph of the Si nanocrystallite layer.

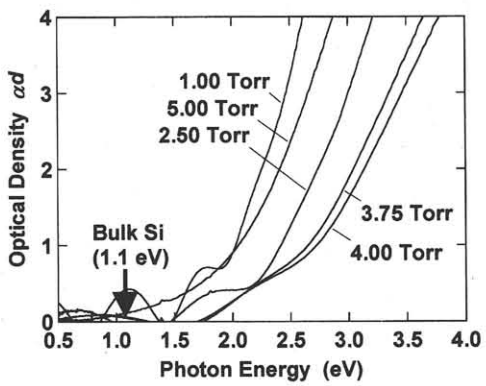

Fig. 3 Optical absorption spectra of the Si nanocrystallite layer. Influence of the He gas pressure on the optical absorption.

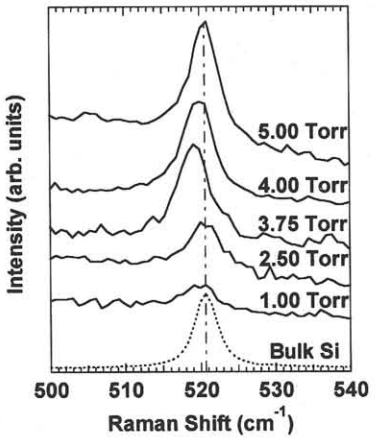

Fig. 4 Raman scattering spectra of the Si nanocrystallite layer. Influence of the He gas pressure on the Raman scattering.

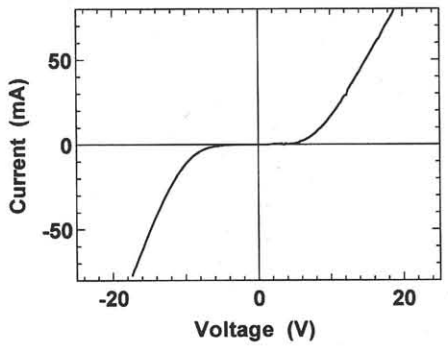

Fig. 5 Current-Voltage characteristics of the LED with the Si nanocrystallite active layer.

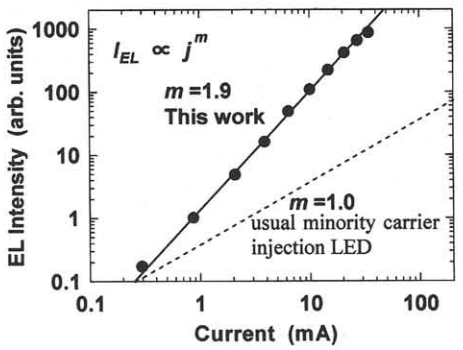

Fig. 6 Integrated light emitting intensity (I_{EL}) as a function of forward current (j).

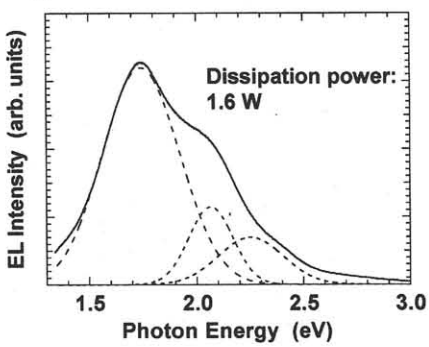

Fig. 7 Light emitting spectrum of the LED with the Si nanocrystallite active layer.