Extended Abstracts of the 1999 International Conference on Solid State Devices and Materials, Tokyo, 1999, pp. 426–427

Visible Electroluminescence from Silicon Nanocrystals Embedded in CaF_2 Epilayers on Si(111) with Rapid Thermal Anneal

Takeo Maruyama, Naoto Nakamura and Masahiro Watanabe

Department of Information Processing, Tokyo Institute of Technology
2-12-1 O-okayama, Meguro-ku, Tokyo 152, Japan
Phone: +81-3-5734-3574, Fax: +81-3-5734-3574, e-mail: maruyama@pe.titech.ac.jp

1. Introduction

Light emitting materials based on silicon nanostructures are widely studied for aiming to realize optoelectronic integrated circuits (OEIC) on silicon wafers. Recently, various investigations of electroluminescence (EL) from silicon-related low dimensional structures have been reported such as porous Si [1], nanocrystalline Si (nc-Si) [2], Si/Ge superlattices [3], amorphous Si [4] and Si/CaF_2 multilayered superlattices [5].

We have studied the formation of nc-Si embedded in single-crystalline CaF_2 on a Si(111) substrate [6] and reported its photoluminescence (PL) [7] and EL properties [8]. Nc-Si embedded in a single-crystalline insulator is attractive for light emitting material due to its sharp and stable heterointerface. Moreover, large quantum confinement can be expected in nc-Si because of its large conduction band discontinuity ($\sim$2.3 eV) at the heterointerface. Very recently, we have found that a rapid thermal annealing of nc-Si/CaF_2 structures dramatically improves the intensity and uniformity of PL [9].

In this work, EL of nc-Si formed in single-crystal CaF_2 layers grown on a Si(111) substrate with rapid thermal anneal is presented.

2. Experiment

CaF_2 has a fluorite lattice structure that is well matched to the lattice structure of Si with mismatches of +0.6 % at room temperature. When Si and CaF_2 are deposited simultaneously on a CaF_2/Si(111) substrate, Si and CaF_2 were grown independently because Si and CaF_2 have different bonding mechanisms (Si: covalent, CaF_2: ionic). As a result, varying the growth temperature and the flux ratio of Si and CaF_2 can control the size and density of the nc-Si. Nc-Si of 5-10 nm in diameter embedded in the epitaxial CaF_2 layer on a Si substrate can be obtained using this technique [6].

Nc-Si embedded in CaF_2 layers were grown on a Si(111) substrate in a ultra-high vacuum molecular beam epitaxy (MBE) chamber ($\sim 10^{-9}$ Torr), an electron beam evaporation source for Si, and a resistively heated effusion crucible for CaF_2. In addition, the CaF_2 source has an ionization and acceleration unit for partially ionized epitaxy, which enhances CaF_2 epitaxy at low temperatures [10].

First, a single-crystalline CaF_2 6 monolayers (MLs) thick was grown on a p-type Si(111) substrate at a substrate temperature of 700 °C. Subsequently, a 6 MLs-thick nc-Si layer was formed by co-evaporation of Si and CaF_2 at below 80 °C with the acceleration voltage of 1 kV. Deposition rates of Si and CaF_2 were 0.4 nm/min and 1.2 nm/min, respectively. 6MLs-thick nc-Si layer and 3 MLs-thick CaF_2 multilayered superlattices were prepared.

To improve PL intensity, *ex situ* rapid thermal annealing (RTA) was carried out using infrared lamp radiation heating with N_2 ambience at atmospheric pressure. The temperature was increased from 200 °C up to an annealing temperature of 700 °C in 20 s, and then the temperature was kept relatively constant using feedback control for 20 s. RTA at this condition improves intensity and uniformity of PL by maintaining surface morphology. A longer annealing time (> 100 s) or a higher annealing temperature (> 700 °C) leads to degradation of the surface morphology and PL intensity [9].

3. Results and Discussions

Figure 1(a) shows a schematic cross section of the EL device fabricated in this study. On three periods of CaF_2/nc-Si multilayered superlattices, a SiO_2 layer was formed with a thickness of 100 nm for the insulation. A contact hole of 300 μm in diameter was formed by photolithography and selective wet chemical etching using BHF 7%. The indium thin oxide (ITO) films for transparent electrodes were deposited with a thickness of 100 nm using radio frequency (RF) sputtering. The ITO electrode of 500×500 μm^2 was formed by the reactive ion etching (RIE) process, and the back contact electrode was formed with Al at the bottom of the substrate.

Figure 1(b) shows the band diagram of the EL device applied forward bias. Electrons are injected from an n-type ITO electrode into nc-Si through a CaF_2 energy barrier by tunneling, and holes are injected from a p-type Si substrate resulting in a radiative recombination in the nc-Si.

Figure 2 (a) shows a optical microscope plane view of the EL device. Figure 2 (b) shows the emission of EL from ITO transparent electrode in the dark. EL was measured at a pulsed bias (duty = 1:1, frequency$\sim$ 1kHz) at room temperature. Typically, at the forward bias of 10V and the current of 2mA, EL which seemed white was clearly observed from the whole area of the contact hole through the ITO electrode. On the other hand, no light emission was observed when a reverse bias was applied because current injection cannot occur.

A current-voltage characteristic, a current-EL intensity characteristic and EL spectra are shown in Fig. 3. This I-V characteristic is reasonable as compared with the simulation results derived from the tunneling model with the band diagram in Fig. 1(b). The inset is EL spectra of the same sample .

The peak wavelength of EL spectra was not recognized by our measurement system. The EL intensity in this figure is defined by the integration of an EL spectrum from 350 nm to 700 nm. It is found that the EL intensity is proportional to the driving current, where the shape of EL spectra is not varied by bias voltage.

4. Conclusion

We have reported electroluminescence of nc-Si embedded in CaF_2 fabricated by the co-evaporation of Si and CaF_2 on Si(111) followed by RTA at 700 °C. Visible electroluminescence was clearly observed at room temperature with a forward bias voltage of approximately 10 V and an injection current of 2 mA. No luminescence was observed at for the reverse bias. The EL intensity is proportional to the driving current. We believe that the nc-Si formed in single-crystalline dielectric on a Si substrate is an attractive material for current injection light sources.

Acknowledgment

The authors would like to thank Professors K. Furuya, S. Arai, M. Asada and Associate Professor Y. Miyamoto for providing fruitful discussion. This work was supported by the Ministry of Education, Science, Sports and Culture through a Scientific Grant-in-Aid, and by The Japan Society for the Promotion of Science (JSPR-RFTF 96P00101), and by the Research Center for Ultra-High-Speed Electronics.

References

[1] N. Koshida and H. Koyama: Appl. Phys. Lett. **60** (1992) 347.
[2] G. G. Qin, A. P. Li, B. R. Zhang and B.-C. Li: J. Appl. Phys. **78** (1995) 2006.
[3] H. Presting, T. Zinke, A. Splett, H. Kibbel and M. Jaros: Appl. Phys. Lett. **69** (1996) 2376.
[4] S. Fujita and N. Sugyama: Appl. Phys. Lett. **74** (1999) 308.
[5] F. Bassani, L. Vervoort, I. Mihalcescu, J. C. Vial and F. Amaud d'Avitaya: J. Appl. Phys. **79** (1996) 4066.
[6] M. Watanabe, F. Iizuka and M. Asada: Jpn. J. Appl. Phys. **34** (1995) 4380.
[7] M. Watanabe, F. Iizuka and M. Asada: IEICE Trans. **E79-C** (1996) 1562.
[8] M. Watanebe, T. Matsunuma, T. Maruyama and Y. Maeda: Jpn. J. Appl. Phys. **37** (1998) L591.
[9] M. Watanabe, T. Maruyama and S. Ikeda: J. Lumin. **80** (1999) 253.
[10] M. Watanabe, S. Muratake, H. Fujimoto, S. Sakamori, M. Asada and S. Arai: Jpn. J. Appl. Phys. **31** (1992) L116.

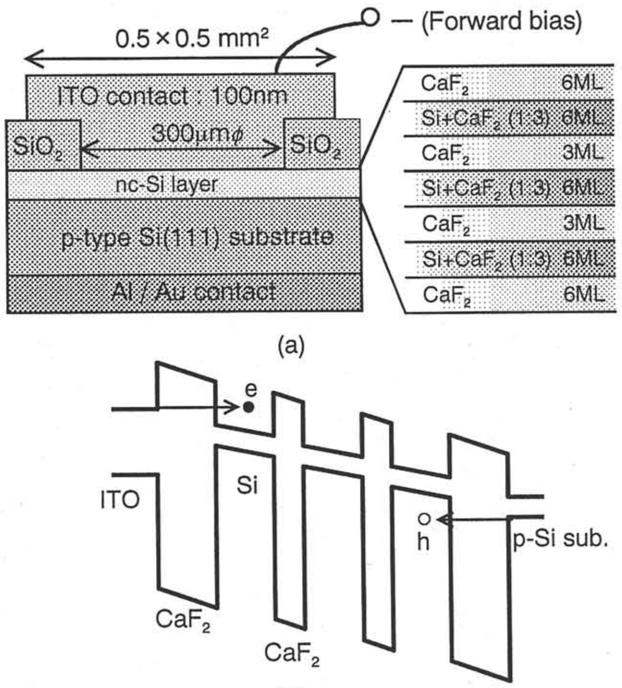

Fig. 1 Schematic cross section of the fabricated device. Current is injected by an ITO transparent electrode. Contact hole size of each electrode is 300 μm in diameter. (b) Band diagram of the EL device applied forward bias. Electrons are injected from an n-type ITO electrode into nc-Si through a CaF_2 energy barrier by tunneling, and holes are injected from a p-type Si substrate. Electrons and holes are recombined in nc-Si embedded in a a CaF_2 layer.

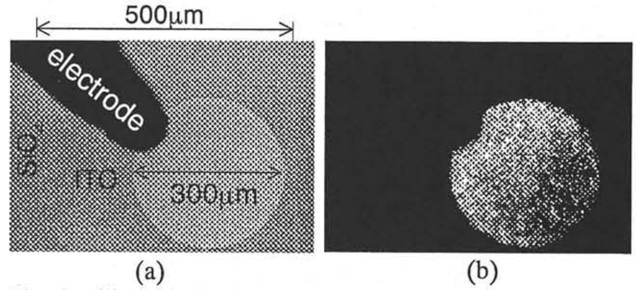

Fig. 2 (a) Optical microscope image of the EL device. (b) Photograph of EL from the device under a forward bias voltage of 10 V and a current of 2 mA at room temperature in the dark.

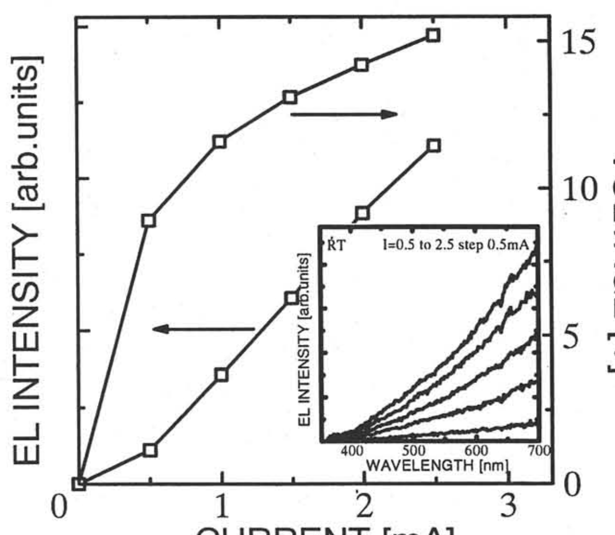

Fig. 3 The typical voltage-current and EL intensity-current characteristics of the EL device at room temperature. The inset is a EL spectrum.