

# Effects of Si-Layer-Thickness Ratio on UV-Light-Emission Intensity from Si/SiO<sub>2</sub> Multilayered Thin Films Prepared Using Radio-Frequency Sputtering

Kenta Miura\*, Hitomi Hoshino, Masashi Honmi, Osamu Hanaizumi

Graduate School of Science and Technology, Gunma University, Kiryu, Japan

Email: [mkenta@gunma-u.ac.jp](mailto:mkenta@gunma-u.ac.jp)

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## Abstract

We investigated the effects of Si-layer-thickness ratios on ultraviolet (UV) peak intensities of Si/SiO<sub>2</sub> multilayered films produced by alternately stacking several-nanometer-thick Si and SiO<sub>2</sub> layers using radio-frequency sputtering for the first time. The Si-layer-thickness ratio of the Si/SiO<sub>2</sub> film is a very important parameter for enhancing the peak intensity because the ratio is concerned with the size of Si nanocrystals in the film, which might affect the intensity of the UV light emission from the film. We prepared seven samples with various estimated Si-layer-thickness ratios, and measured the photoluminescence spectra of the samples after annealing at 1150°C, 1200°C, or 1250°C for 25 min. From our experiments, we estimate that the proper Si-layer-thickness ratio to obtain the strongest UV peaks from the Si/SiO<sub>2</sub> multilayered films is around 0.29. Such a UV-light-emitting thin film is expected to be used in future higher-density optical-disk systems.

## Keywords

Si, SiO<sub>2</sub>, Multilayer, Sputtering, UV-Light Emission

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## 1. Introduction

Various works on silicon (Si)-based luminescent materials utilizing the quantum-confinement effect, such as porous Si [1] [2] and Si nanocrystal (Si-nc) [3]-[5], have been reported. Ultraviolet (UV) light emission can be obtained from such materials fabricated using various methods such as laser ablation [6] and sputtering [7] [8].

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\*Corresponding author.

UV photoluminescence (PL) spectra having a peak wavelength around 370 nm were obtained from sputtered SiO<sub>2</sub> films including Si-ncs after annealing [7]. The UV light emission might originate from emission centers of interface layers (SiO<sub>x</sub>) between Si-ncs and SiO<sub>2</sub> matrices [7].

Furthermore, Si/SiO<sub>2</sub> multilayers produced by alternately stacking several-nanometer-thick Si and SiO<sub>2</sub> layers using the sputtering method are very important structures from which we can controllably and effectively produce such Si-ncs and SiO<sub>x</sub> [8]. In these multilayered structures, Si-ncs seem to be produced from nanometer-order-thick Si layers, and SiO<sub>x</sub> can clearly be formed after annealing (see **Figure 1**) [8]. In our previous study, we fabricated Si/SiO<sub>2</sub> multilayered films having nanometer-order-thick Si and SiO<sub>2</sub> layers using radio-frequency (RF) magnetron sputtering and observed UV PL having a sharp peak at a wavelength of 370 nm after annealing at 1150°C to 1250°C [8]. Such UV-light-emitting materials may be used in future higher-density optical disk systems.

However, the UV peak intensities from the Si/SiO<sub>2</sub> multilayered films should be stronger than those in our previous study in order to be used as light-emitting materials. We considered that the Si-layer-thickness ratio of the film was a very important parameter for enhancing the peak intensity because the ratio was concerned with the size of Si-nc, which might affect the intensity of the UV light emission [7]. Therefore, in this study, we investigated the effects of the Si-layer-thickness ratios on the UV peak intensity of the Si/SiO<sub>2</sub> sputtered and multilayered films.

## 2. Fabrication

A RF magnetron sputtering apparatus (ULVAC, SH-350-SE) was used in our fabrication processes. Si and SiO<sub>2</sub> plates having diameters of 100 mm were used as sputtering targets, and Si/SiO<sub>2</sub> multilayered films were deposited by alternately supplying RF power to the Si and SiO<sub>2</sub> targets. The flow rate of Ar gas introduced into the chamber was 10 sccm, and the RF powers supplied to the Si and SiO<sub>2</sub> targets were 50 and 100 W, respectively. Commercial fused-silica substrates (ATOCK Inc., 11.5 mm × 24.5 mm × 1 mm<sup>1</sup>) were used and not heated during deposition. One hundred pairs of Si and SiO<sub>2</sub> layers were alternately stacked on the substrates. The deposition rates of Si and SiO<sub>2</sub> thick films were 0.05 and 0.12 nm/s, respectively. Switching of sputtering targets and opening or closing of two shutters positioned above the targets were automatically controlled by digital timers. The total thicknesses of multilayered films were measured using a surface profiler (ULVAC, Dektak 3ST). We can estimate the Si and SiO<sub>2</sub> layer thicknesses ( $d_{\text{Si}}$  and  $d_{\text{SiO}_2}$ , see **Figure 1**) from the measured total thicknesses of the films and the ratio of their deposition rates (0.5:1.2). In this study, we prepared seven samples with estimated Si-layer-thickness ratios  $d_{\text{Si}}/(d_{\text{Si}} + d_{\text{SiO}_2})$  of 0.15, 0.23, 0.24, 0.26, 0.27, 0.29, or 0.30 (**Table 1**). Four specimens were prepared from one as-deposited sample by cutting it using a diamond-wire saw. We subsequently annealed the specimens in ambient air at 1150°C, 1200°C, or 1250°C for 25 min using a Siliconit electric furnace after sputter deposition.

## 3. Evaluation

PL spectra from the annealed samples were measured at room temperature with excitation using a He-Cd laser

**Table 1.** Seven samples prepared in this study. The Si and SiO<sub>2</sub> layer thicknesses ( $d_{\text{Si}}$  and  $d_{\text{SiO}_2}$ ) were estimated from the measured total thicknesses of the Si/SiO<sub>2</sub> multilayered films and the ratio of their deposition rates. The deposition rates of Si and SiO<sub>2</sub> thick films were 0.05 and 0.12 nm/s, respectively.

Sample No.	$d_{\text{Si}}$ [nm] (estimated)	$d_{\text{SiO}_2}$ [nm] (estimated)	Si-layer-thickness ratio: $d_{\text{Si}}/(d_{\text{Si}} + d_{\text{SiO}_2})$
1	0.6	3.4	0.15
2	0.8	2.7	0.23
3	0.9	2.9	0.24
4	1.0	2.9	0.26
5	1.0	2.6	0.27
6	0.9	2.2	0.29
7	1.1	2.3	0.32

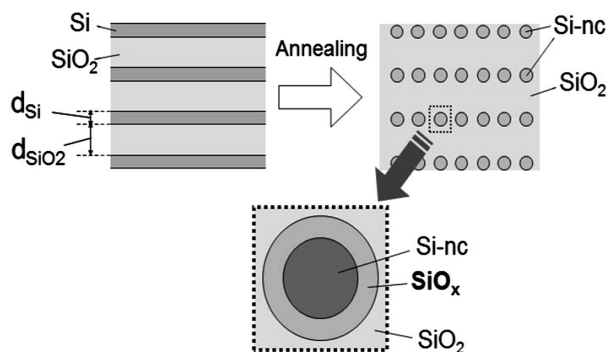
(Kimmon, IK3251R-F, wavelength  $\lambda = 325$  nm). A monochromator (Nikon, P250), a photomultiplier (Hamamatsu, R2658), and a lock-in amplifier (NF, LI-572B) were used in our measurements. The intensities of PL spectra were calibrated by using a white-light spectrum measured with an optical spectrum analyzer (Anritsu, MS9701C + MS9030A) at wavelengths from 350 to 1750 nm because sensitivities of the monochromator and the photomultiplier depend on input light wavelengths.

**Figure 2** presents typical PL spectra of specimens prepared from the sample with a Si-layer-thickness ratio of 0.29 (sample 6) after annealing at 1150°C, 1200°C, and 1250°C. The total thickness of the multilayered film was 463 nm, and the thickness of the Si and SiO<sub>2</sub> layer can be estimated to be 0.9 and 2.2 nm, respectively. UV PL peaks around a wavelength of 370 nm were observed from all the specimens regardless of annealing temperature. The maximum arbitrary intensity of the UV PL peak was 8.45.

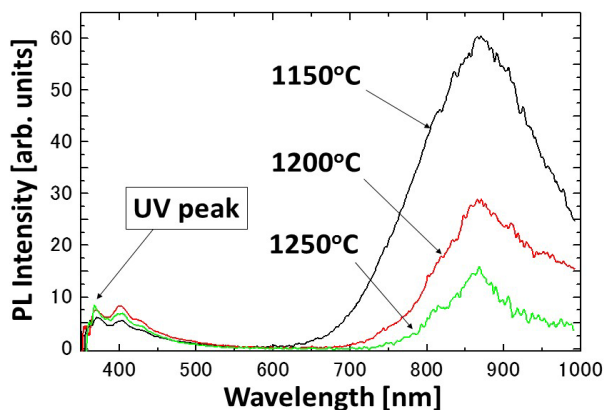
**Figure 3** plots the maximum intensities of the UV PL peaks versus the Si-layer-thickness ratios  $d_{\text{Si}}/(d_{\text{Si}} + d_{\text{SiO}_2})$  of the samples. The data in our previous work (plotted as a rectangle in **Figure 3**) [8] agreed well with the present graph. From the result, we can estimate that the proper Si-layer-thickness ratio to obtain the strongest UV peak from our Si/SiO<sub>2</sub> multilayered films is around 0.29.

#### 4. Summary

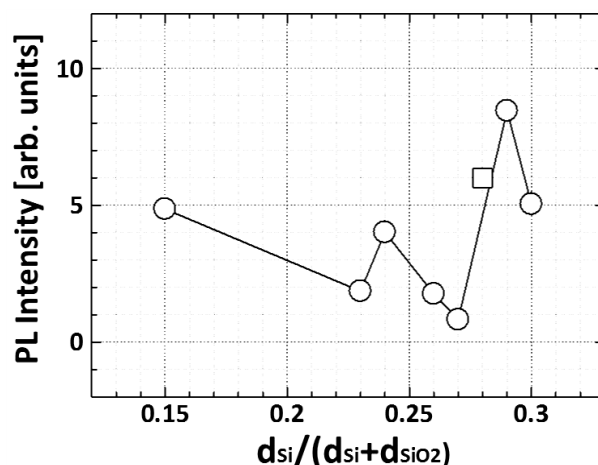
We investigated the effects of the Si-layer-thickness ratios on the UV peak intensities of Si/SiO<sub>2</sub> multilayered films produced by alternately stacking several-nanometer-thick Si and SiO<sub>2</sub> layers using RF sputtering for the first time. The proper Si-layer-thickness ratio to obtain the strongest UV peak from our Si/SiO<sub>2</sub> multilayered films was estimated to be around 0.29. The sizes of Si-ncs seem to be controllable by selecting both the amounts



**Figure 1.** Schematic figure of a Si/SiO<sub>2</sub> multilayered film and the forming process of Si-ncs and interface layers (SiO<sub>x</sub>) in the film [8].



**Figure 2.** Measured PL spectra of specimens prepared from the sample with a Si-layer-thickness ratio of 0.29 (sample 6) after annealing at 1150°C, 1200°C, and 1250°C for 25 min.



**Figure 3.** Dependence of UV PL peak intensities on Si-layer-thickness ratios of Si/SiO<sub>2</sub> multilayered films.

of Si and SiO<sub>2</sub> deposition and the annealing conditions [8]. We will try to optimize the conditions (annealing temperature and time) in order to further increase the UV light-emission intensity of the Si/SiO<sub>2</sub> multilayered films.

Such sputtered films can be used as constituent materials of autocloned photonic crystals that can be applied to novel light-emitting devices [9]-[11]. UV-light-emitting materials and devices are expected to be used in future higher-density optical-disk systems.

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