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Observation of Ultraviolet-Light Emission from Si/SiO_2 Multilayer Films Prepared by Using RF Magnetron Sputtering

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1. Introduction

Various works on silicon-based (Si-based) luminescent materials utilizing the quantum confinement effect, such as porous Si [1] and Si nanocrystal (Si-nc) [2,3], have been reported. Ultraviolet-light (UV-light) emission around a wavelength of 370nm was observed from sputtered SiO_2 films including Si-nc's produced after annealing at 800°C to 1100°C [4]. The UV-light emission might originate from emission centers of interface layers between Si-nc's and SiO_2 matrices, and its intensity might be affected by sizes of Si-nc's. Sample A of oxidized porous Si in ref. [5] emitted UV light around a wavelength of 370nm after thermal oxidization at 700°C to 1150°C, and it was explained that the UV-light emission seemed to originate from Si-nc's and interface layers between Si-nc's and SiO_2 produced in the thermal oxidization process.

The UV-light emission can be useful for light sources of higher-density optical-disk systems. If we realize this kind of light source utilizing Si-based materials, we will obtain many benefits, such as matching for LSI's, lower cost, and suitability for environment application. As we explained above, the UV-light emission from Si-containing materials seems to originate from Si-nc's and interface layers between Si-nc's and surrounding SiO_2. Si/SiO_2 multilayers fabricated by alternately stacking several-nanometer-thick Si and SiO_2 layers are expected to be important structures from which we can controllably and effectively produce Si-nc's and interface layers. In these multilayer structures, Si-nc's seem to be produced from Si layers by using high-temperature annealing above 1000°C. Interface layers between Si and SiO_2 can be clearly formed, and the number of layers can be decided by selecting the numbers of Si and SiO_2 layers. Therefore, we strongly expect that UV-light emission can be obtained from such Si/SiO_2 multilayer structures. As explained in ref. [4], if sizes of Si-nc's are related to the intensity of the UV-light emission, we also expect that much stronger UV-light emission can be obtained by controlling the sizes of Si-nc's by properly selecting Si layer thickness and annealing conditions (temperatures and times) for Si/SiO_2 multilayer films.

In this paper, we fabricated Si/SiO_2 multilayer films having nanometer-order thicknesses of Si and SiO_2 layers by using radio-frequency (rf) magnetron sputtering and subsequently annealed the films at high temperatures (from 1150°C to 1250°C). We observed UV photoluminescence having a sharp peak at a wavelength of around 370nm from a sample annealed at 1200°C.

2. Fabrication

An rf-sputtering apparatus (ULVAC, SH-350-SE) was used in our sputtering processes. Si and SiO_2 plates having a diameter of 100mm are used as sputtering targets, and Si/SiO_2 multilayers were sputtered by alternately supplying rf power to Si and SiO_2 targets. The background pressure in the vacuum chamber of our apparatus was 5.3 x 10^{-5}Pa, the Ar flow rate introduced into the chamber in the sputtering process was 10sccm, and the rf power supplied to the Si (SiO_2) target was 50W (100W). Fused silica substrates (11.5 x 24.5 x $1mm^t$) were used, and they were not heated during sputtering. One hundred pairs of Si and SiO_2 layers were alternately stacked on the substrates. The sputtering rates of a Si (SiO_2) thick film was 0.05nm/sec. (0.12nm/sec.). Switching of sputtering targets and opening or closing of two shutters positioned above the targets were automatically controlled by digital timers. The total thickness of the sample was 404nm (measured using a surface profiler (ULVAC, Dektak 3ST)). The measured value differs by 3.6% from the thickness estimated from sputtering rates of Si and SiO_2 thick films. We can presume thicknesses of Si and SiO_2 layers from the measured total thickness of the sample and the ratio of 0.5:1.2 of Si and SiO_2 sputtering rates. The thickness of Si (SiO_2) layers can be estimated to be 1.1nm (2.8nm). We prepared three samples under the same sputtering conditions and annealed them in air for 25min. at 1150, 1200, 1250°C by using a siliconit furnace after sputtering.

3. Evaluation and Discussion

Photoluminescence (PL) spectra from the three samples were measured at room temperature with excitation using a He-Cd laser (Kimmon, IK3251R-F, λ=325nm). A monochromator (Nikon, P250), a photomultiplier (Hamamatsu, R2658), and a lock-in amplifier (NF, LI-572B) were used in our measurements.

Figure 1 presents the PL spectra of the three samples generated at room temperature. A sharp PL peak in the UV range was observed from the sample annealed at 1200°C. We thus demonstrated that the desired UV-light emission around a wavelength of 370nm can be obtained from a Si/SiO_2 multilayer after high-temperature annealing.

In addition to UV PL peaks, lower-energy (longer wavelength from red to infrared) peaks were observed from our samples. The main PL peak from the sample annealed at 1150°C was located at around 1.44eV (λ~860nm), and the full width at half maximum (FWHM) of this peak was

0.33eV. In contrast, the main PL peak from the sample annealed at 1200°C was located in the UV range. The UV peak energy was around 3.4eV (λ~370nm), and the FWHM was 0.20eV. The UV peak from the sample annealed at 1250°C had a lower intensity but the same energy, and the peak around 1.44eV disappeared. Such lower-energy peak (longer-wavelength peak) has been observed from amorphous Si/SiO_2 superlattices and was described as originating from interface layers between Si and SiO_2 [6]. From this consideration, the two PL peaks of our samples may also originate from interface layers between Si-nc's and SiO_2 media. We have successfully demonstrated that we may be able to control the emission properties of two bands of Si/SiO_2 multilayers by selecting annealing temperatures. Therefore, only the strong UV-light emission peak seems to appear from our Si/SiO_2 multilayers due to optimization of the sputtering and annealing conditions.

Figure 2 plots the square root of the absorption coefficient of our samples. It was obtained by measuring the transmittance spectra with a spectrophotometer (Shimadzu, UV-3101PC) and fitting the spectra to the equation $T=exp(-\alpha d)$ (α, absorption coefficient; d, film thickness). We can estimate the energies of absorption edges of our samples from Fig. 2. Intersecting points of the horizontal axis and extrapolated lines correspond to absorption edges. The absorption edges of the samples are estimated to be 3.0 to 3.2eV (λ=390 to 420nm) from Fig. 2. The values of the absorption edges did not agree with the energy of the UV peak (~3.4eV) but were nearly equal to the blue PL peak energy of $Si:SiO_2$ co-sputtered films without annealing [7]. We need to investigate the relations between the UV-light emission from Si/SiO_2 multilayers and blue-light emission from $Si:SiO_2$ co-sputtered films.

4. Conclusions

We fabricated Si/SiO_2 multilayers having nanometer-order-thick Si and SiO_2 layers using rf magnetron sputtering and obtained UV-light emission from the samples after high-temperature annealing. In particular, we found a sharp UV peak around a wavelength of 370nm after annealing at 1200°C. The UV-light emission seems to originate from Si layers, which may be transformed into Si-nc's, and interface layers between Si-nc's and SiO_2 layers. The intensity of the UV peak seemed to be increase after optimization of Si-nc sizes. The sizes can be controlled by selecting the amounts of Si deposition and oxidization by annealing in air. In addition to the UV peak, we observed other PL peaks ranging from red to infrared wavelengths from our samples. However, we expect that only the UV peak will be observed under proper sputtering and annealing conditions. Therefore, we are trying to optimize the conditions in order to increase the UV-light emission intensity.

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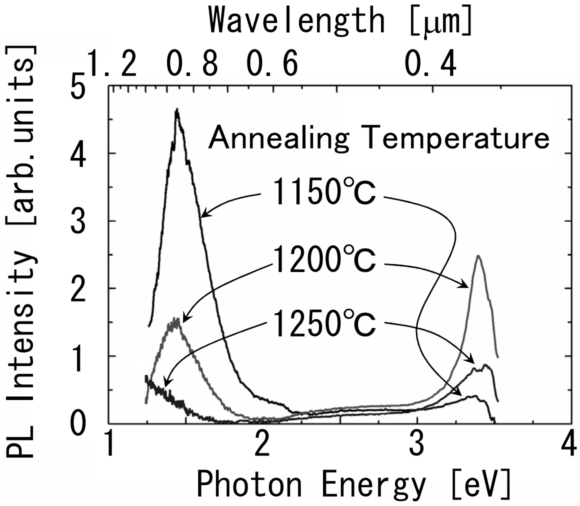

Fig. 1 Measured photoluminescence spectra.

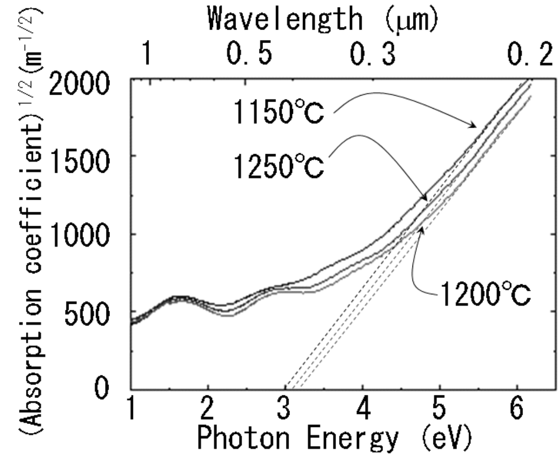

Fig. 2 Absorption coefficients deduced from transmittances.