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Preparation of Light-Emitting Ytterbium-Doped Tantalum-Oxide Thin Films Using a Simple Co-Sputtering Method

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Abstract

Light-emitting ytterbium-doped tantalum-oxide thin films were prepared using a simple co-sputtering method for the first time. Sharp photoluminescence peaks having a wavelength of around 980 nm were observed from films annealed from 700˚C to 1000˚C for 10 to 40 min. The strongest intensity of the 980-nm peak was obtained from a film deposited using three ytterbium-oxide pellets and annealed at 800˚C for 20 min. Such rare-earth doped tantalum-oxide sputtered films can be used as high-refractive-index materials of autocloned photonic crystals that can be applied to novel light-emitting devices, and they will also be used as both anti-reflection and down-conversion layers for realizing high-efficiency silicon solar cells.

Keywords

Tantalum Oxide, Ytterbium, Co-Sputtering, Annealing, Photoluminescence

1. Introduction

Tantalum (V) oxide (Ta_2O_5) is a high-refractive-index material used in passive optical elements such as Ta_2O_5/SiO_2 multilayered wavelength filters for dense wavelength-division multiplexing (DWDM). It has also been used as a high-index material of Ta_2O_5/SiO_2 multilayered photonic-crystal elements for the visible to near-infrared range fabricated using the autocloning method based on radio-frequency (RF) bias sputtering [1]-[3], and it can additionally be used as an anti-reflection coating material for silicon solar cells [4]. However, Ta_2O_5 has recently attracted much attention as an active optical material, since broad red photoluminescence (PL) spectra at wavelengths from 600 to 650 nm were observed from thermal-oxidized amorphous Ta_2O_5 thin films [5]. In our previous work, we demonstrated blue PL from Ta_2O_5 thin films deposited by RF magnetron sputtering [6].

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In addition, many studies on rare-earth-doped Ta_2O_5 have been conducted because Ta_2O_5 is a potential host material for new phosphors due to its low phonon energy (100 - 450 cm^{-1}) compared with other oxide materials such as SiO_2 [7]. We have reported on various rare-earth (Er, Eu, Tm, and Y) doping into Ta_2O_5 thin films using simply co-sputtering of rare-earth oxide (Er_2O_3, Eu_2O_3, Tm_2O_3, and Y_2O_3) pellets and a Ta_2O_5 disc [8]-[12]. Moreover, in our recent study, we fabricated Er, Eu, and Ce co-doped Ta_2O_5 (Ta_2O_5: Er, Eu, Ce) thin films using the co-sputtering method, and observed yellow PL from the films.

In this study, in order to expand the useful wavelength range of our Ta_2O_5-based light-emitting sputtered films, we fabricated ytterbium-doped Ta_2O_5 (Ta_2O_5:Yb) thin films using the simple co-sputtering method for the first time.

2. Experimental

Ta_2O_5:Yb thin films were deposited using an RF magnetron sputtering system (ULVAC, SH-350-SE). A Ta_2O_5 disc (Furuuchi Chemical Corporation, 99.99% purity, diameter 100 mm) was used as the sputtering target. We placed Yb_2O_3 pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 21 mm) on the Ta_2O_5 disc. The Ta_2O_5 disc and Yb_2O_3 pellets were co-sputtered by supplying RF power to the target. **Figure 1** is a schematic diagram of the sputtering target, with three Yb_2O_3 pellets on the Ta_2O_5 disc. We prepared co-sputtered Ta_2O_5:Yb films with different Yb concentrations by placing two, three, four, or five Yb_2O_3 pellets on the Ta_2O_5 disc. The flow rate of argon gas introduced into the vacuum chamber was 10 sccm, and the pressure in the chamber during deposition was kept at ~1 Pa. The RF power supplied to the target was 200 W. Commercial fused silica plates (ATOCK Inc., 1 mm thick) were used as substrates. The substrates were not heated during sputtering. We subsequently annealed the samples in ambient air using an electric furnace (Denken, KDF S-70).

The PL spectra of the Ta_2O_5:Yb films were measured using a dual-grating monochromator (Roper Scientific, SpectraPro 2150i) and a CCD detector (Roper Scientific, Pixis: 100B, electrically cooled to −80˚C). An He-Cd laser (Kimmon, IK3251R-F, wavelength λ = 325 nm) was used to excite the films.

3. Results and Discussion

Figure 2 presents PL spectra of the Ta_2O_5:Yb films deposited using two, three, four, or five Yb_2O_3 pellets and annealed at 900˚C for 20 min. Intense PL peaks around a wavelength of 980 nm were observed from all of the samples. The 980-nm peaks seem to be the result of the $^2F_{5/2}\rightarrow^2F_{7/2}$ transition of Yb^{3+} [13]. The peak intensity once increased and subsequently decreased with increasing the number of Yb_2O_3 pellets (the Yb concentration) as [13]. **Figure 2** indicates that the film deposited using three Yb_2O_3 pellets exhibited the strongest intensity of the 980-nm peak after annealing at 900˚C for 20 min.

Figure 3 presents PL spectra from Ta_2O_5:Yb films deposited using the standard three Yb_2O_3 pellets and annealed at 900˚C for 10, 20, 30, or 40 min. The 980-nm-peak intensities from the samples annealed for 10, 30, and 40 min were approximately the same, but the peak intensity from the sample annealed for 20 min was stronger than the others. We therefore considered that the proper annealing time is 20 min.

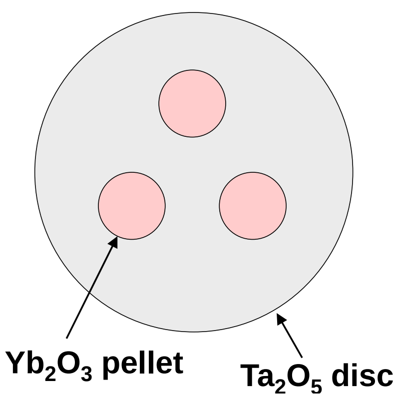

Figure 1. Schematic diagram of the sputtering target for simply co-sputtering of Yb_2O_3 and Ta_2O_5.

Figure 4 presents PL spectra from Ta_2O_5:Yb films deposited using the three Yb_2O_3 pellets and annealed at 700˚C, 800˚C, 900˚C, or 1000˚C for the standard 20 min. The 980-nm-peak intensity once increased and subsequently decreased with increasing the annealing temperature. The strongest peak intensity was observed from the sample annealed at 800˚C. Interestingly, the PL peak observed from the sample annealed at 700˚C was sharper than that of the other samples. From the X-ray diffraction (XRD) measurements, the sample annealed at 700˚C seemed to be amorphous phase, and the samples annealed at 800˚C, 900˚C, or 1000˚C seemed to be polycrystalline phase. We will continue to investigate the relationship between the width of the 980-nm peak and the crystallizability of our Ta_2O_5:Yb film.

4. Conclusion

Ta_2O_5:Yb thin films were prepared by our simple co-sputtering method for the first time, and PL spectra having sharp peaks at a wavelength of 980 nm were observed from the films after annealing from 700˚C to 1000˚C for 10 to 40 min. The reference conditions for fabricating our Ta_2O_5:Yb films (three Yb_2O_3 pellets; annealing temperature 800˚C; annealing time 20 min) determined to provide the strongest PL peak intensity. Such rare-earth-

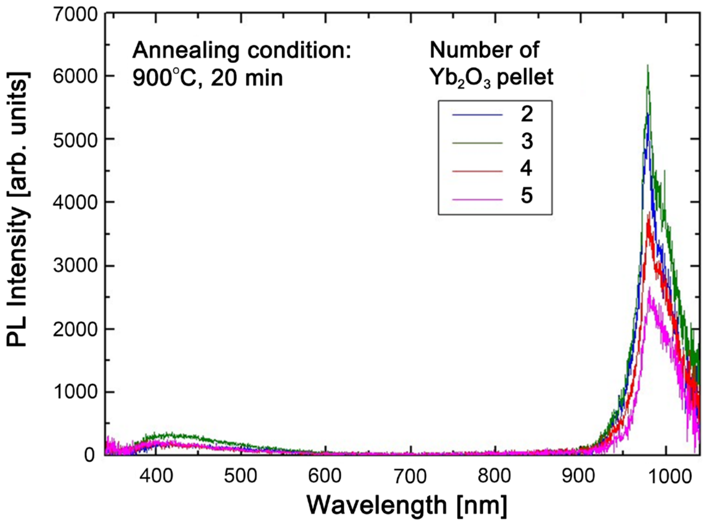

Figure 2. PL spectra of Ta_2O_5:Yb co-sputtered films deposited using two, three, four, or five Yb_2O_3 pellets and annealed at 900˚C for 20 min.

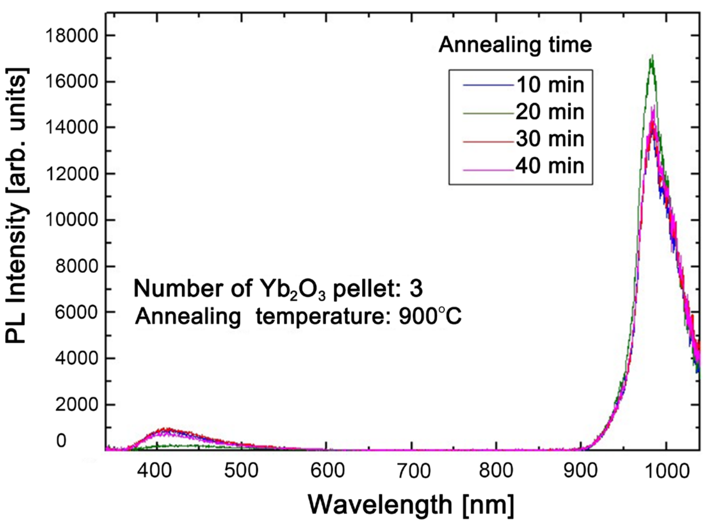

Figure 3. PL spectra of Ta_2O_5:Yb co-sputtered films deposited using three Yb_2O_3 pellets and annealed at 900˚C for 10, 20, 30, or 40 min.

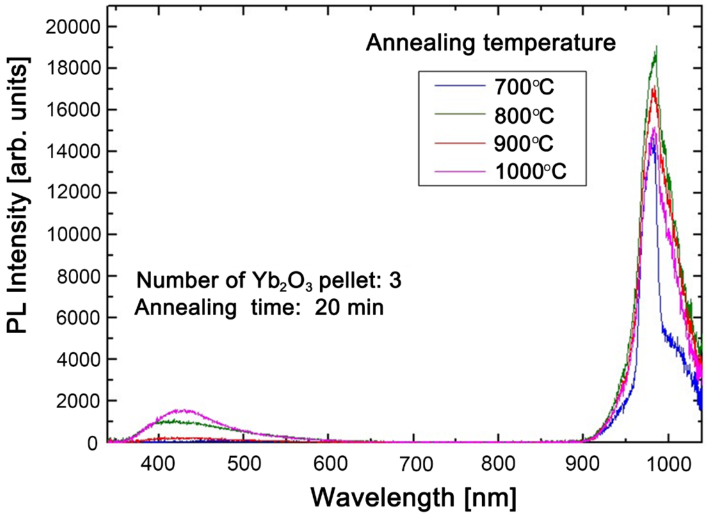

Figure 4. PL spectra of Ta_2O_5:Yb co-sputtered films deposited using three Yb_2O_3 pellets and annealed at 700˚C, 800˚C, 900˚C, or 1000˚C for 20 min.

doped Ta_2O_5 sputtered films can be used as high-refractive-index materials of Ta_2O_5/SiO_2 autocloned (multi-layered) photonic crystals that can be applied to novel light-emitting devices [1], and they will also be used as both anti-reflection [4] and down-conversion [13] [14] layers for realizing high-efficiency silicon solar cells.

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