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Microarticle

Fabrication of Tm-doped Ta_2O_5 thin films using a co-sputtering method

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ABSTRACT

Thulium-doped tantalum-oxide (Ta_2O_5:Tm) thin films were prepared using a simple co-sputtering method. A remarkable photoluminescence peak having a wavelength of around 800 nm due to Tm^{3+} was observed from a film annealed at 900 °C for 20 min. The δ-Ta_2O_5 (hexagonal) phase of the Ta_2O_5:Tm sputtered film is very important for obtaining strong photoluminescence.

1. Introduction

Tantalum (V) oxide (Ta_2O_5) is a high-refractive-index material (refractive index $n > 2$) used in passive optical elements such as Ta_2O_5/SiO_2 multilayered wavelength filters for dense wavelength-division multiplexing (DWDM). Recently, it has also been used as a high-index material of autocloned (multilayered) photonic-crystal elements for the visible to near-infrared range [1–3]. However, Ta_2O_5 has recently attracted much attention as an active optical material, since broad red photoluminescence (PL) spectra at wavelengths of 600–650 nm were observed from thermal-oxidized amorphous Ta_2O_5 thin films [4]. In our previous work, we demonstrated blue PL from Ta_2O_5 thin films deposited by radio-frequency (RF) magnetron sputtering [5].

Furthermore, 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 [6]. We reported on green PL from erbium-doped Ta_2O_5 (Ta_2O_5:Er) thin films produced by a very simple co-sputtering method using RF magnetron sputtering [7,8]. Their PL spectra had main peaks at a wavelength of 550 nm due to the transition of Er^{3+} from the $^4S_{3/2}$ to the $^4I_{15/2}$ state ($^4S_{3/2} \rightarrow {}^4I_{15/2}$), and at a wavelength of 670 nm due to the transition from the $^4F_{9/2}$ to the $^4I_{15/2}$ state ($^4F_{9/2} \rightarrow {}^4I_{15/2}$). We also reported on red or orange PL from europium-doped Ta_2O_5 (Ta_2O_5:Eu) thin films deposited using the same co-sputtering method [9]. We observed four PL peaks at wavelengths of around 600, 620, 650, and 700 nm from our Ta_2O_5:Eu films after annealing at 600 to 900 °C. These four peaks seem to be the results of the $^5D_0 \rightarrow {}^7F_1$, $^5D_0 \rightarrow {}^7F_2$, $^5D_0 \rightarrow {}^7F_3$, and $^5D_0 \rightarrow {}^7F_4$ transitions of Eu^{3+}, respectively.

In this study, we fabricated thulium-doped Ta_2O_5 (Ta_2O_5:Tm) thin films using the simple co-sputtering method in order to expand the useful wavelength range of our Ta_2O_5-based light-emitting sputtered films. We obtained a remarkable PL peak around a wavelength of 800 nm from a Ta_2O_5:Tm co-sputtered film after annealing at 900 °C for 20 min.

2. Fabrication of Ta_2O_5:Tm thin films

Ta_2O_5:Tm 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 three Tm_2O_3 pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) on the Ta_2O_5 disc as illustrated in Fig. 1. The Ta_2O_5 disc and the three Tm_2O_3 pellets were co-sputtered by supplying RF power to the target. The flow rate of argon gas introduced into the vacuum chamber was 15 sccm, and the RF power supplied to the target was 200 W. Commercial fused silica plates (1 mm thick) were used as substrates, and they were not heated during sputtering. We subsequently annealed the samples in ambient air at 600, 700, 800, or 900 °C for 20 min using an electric furnace (Denken, KDF S-70). We set the annealing time to 20 min because this was the proper condition for our Ta_2O_5:Er to obtain strong PL intensities [7,8].

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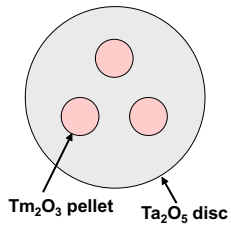

Fig. 1. Schematic diagram of a sputtering target for Tm_2O_3 and Ta_2O_5 co-sputtering.

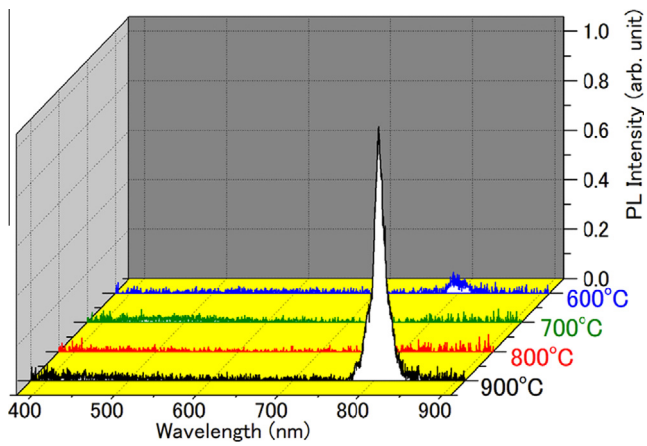

Fig. 2. PL spectra of Ta_2O_5:Tm films deposited using three Tm_2O_3 pellets and annealed at 600, 700, 800, or 900 °C for 20 min.

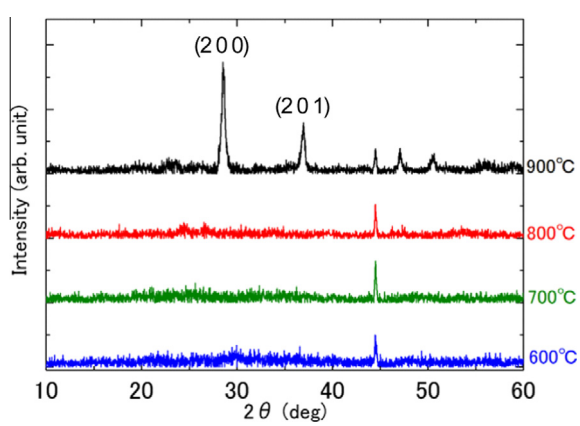

Fig. 3. XRD patterns observed from the annealed Ta_2O_5:Tm films deposited using three Tm_2O_3 pellets.

3. Evaluation of Ta_2O_5:Tm thin films

The PL spectra of Ta_2O_5:Tm 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, $\lambda = 325$ nm) was used to excite the films. The X-ray diffraction (XRD) patterns of the films were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PC system). Tm concentrations of the films after annealing were measured using an electron probe micro-analyzer (EPMA) (Shimadzu, EPMA-1610).

Fig. 2 presents PL spectra of Ta_2O_5:Tm films annealed at 600, 700, 800, or 900 °C for 20 min. A sharp PL peak around a wavelength of 800 nm was observed from the sample annealed at 900 °C. The 800-nm peak seems to be the result of the $^3H_4 \rightarrow {}^3H_6$ transition of Tm^{3+} [10]. The Tm concentration of the film annealed at 900 °C was measured to be ~2.3 mol%.

Fig. 3 presents XRD patterns of films deposited using three Tm_2O_3 pellets and annealed at 600, 700, 800, or 900 °C for 20 min. Major diffraction peaks corresponding to the (2 0 0) (δ-Ta_2O_5) and (2 0 1) phases were observed from the film annealed at 900 °C [11]. It seems that the δ-Ta_2O_5 (hexagonal) phase is very important for obtaining a strong PL peak at the wavelength of 800 nm from our Ta_2O_5:Tm co-sputtered films.

4. Conclusions

Ta_2O_5:Tm thin films were prepared using a simple co-sputtering method. A sharp PL peak having a wavelength of around 800 nm due to the $^3H_4 \rightarrow {}^3H_6$ transition of Tm^{3+} was observed from a film annealed at 900 °C for 20 min. The δ-Ta_2O_5 (hexagonal) phase of the Ta_2O_5:Tm sputtered film is very important for obtaining a strong PL. We are working to optimize the Tm concentration and annealing time of our Ta_2O_5:Tm sputtered films in order to obtain stronger PL-peak intensities. Such rare-earth-doped Ta_2O_5 sputtered films can be used as high-refractive-index materials of autocloned photonic crystals [1–3] that can be applied to novel light-emitting devices.

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