Results in Physics 4 (2014) 185–186

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.journals.elsevier.com/results-in-physics

Microarticle

Fabrication and evaluation of Ta_2O_5:Y_2O_3 co-sputtered thin films

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ARTICLE INFO

Article history:
Received 14 August 2014
Accepted 23 September 2014
Available online 30 September 2014

Keywords:
Tantalum oxide
Yttrium oxide
Co-sputtering
Annealing
Photoluminescence

ABSTRACT

Co-sputtered tantalum (V) oxide and yttrium (III) oxide (Ta_2O_5:Y_2O_3) thin films were fabricated using radio-frequency magnetron sputtering for the first time, and their photoluminescence (PL) and X-ray diffraction properties were evaluated. Broad PL spectra from 380 to 800 nm were observed only from films annealed at 700 °C. The maximum PL intensities were found around a wavelength of 500 nm regardless of the Y concentrations of the films, and the films annealed at 700 °C were primarily amorphous phases. It seems that the broad PL spectra from the Ta_2O_5:Y_2O_3 films originated from oxygen vacancies of Ta_2O_5 and Y_2O_3 particles that may be produced in Ta_2O_5 by co-sputtering.

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). It has also been used as a high-index material of Ta_2O_5/SiO_2 autocloned (multilayered) photonic-crystal elements for the visible to near-infrared range [1]. 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 [2]. We demonstrated blue PL from Ta_2O_5 thin films deposited by radio-frequency (RF) magnetron sputtering [3].

Furthermore, many studies on rare-earth-doped Ta_2O_5 have been conducted because Ta_2O_5 is a potential material for new phosphors due to its low phonon energy (100–450 cm^{-1}) compared with other oxide materials such as SiO_2 [4]. We reported on green PL from erbium-doped Ta_2O_5 (Ta_2O_5:Er) produced by a simple co-sputtering method using RF magnetron sputtering [5,6]. 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 [7]. We recently demonstrated near-infrared PL from thulium-doped Ta_2O_5 (Ta_2O_5:Tm) thin films produced by co-sputtering [8].

In this study, we fabricated co-sputtered Ta_2O_5 and yttrium (III) oxide (Ta_2O_5:Y_2O_3) thin films using RF magnetron sputtering for the first time, and evaluated their PL and X-ray diffraction (XRD) properties.

2. Fabrication of Ta_2O_5:Y_2O_3 thin films

Ta_2O_5:Y_2O_3 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 two, three, or four Y_2O_3 pellets (Furuuchi Chemical Corporation, 99.9% purity, diameter 20 mm) on the Ta_2O_5 disc. The Ta_2O_5 disc and Y_2O_3 pellets were co-sputtered by supplying RF power to the target [5–8]. The flow rate of Ar gas introduced into the vacuum chamber was 15 sccm, and the RF power supplied to the target was 200 W. Commercially fused-silica plates (1 mm thick) were used as substrates, and they were not heated during sputtering. We subsequently annealed samples in ambient air at 700, 800, 900, or 1000 °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 films to obtain strong PL intensities [5,6].

3. Evaluation of Ta_2O_5:Y_2O_3 thin films

The PL spectra of the Ta_2O_5:Y_2O_3 thin 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 XRD patterns of the films were recorded using an X-ray diffractometer (RIGAKU, RINT2200VF+/PC system). The Y concentrations of the films were measured using an electron probe micro-analyzer (EPMA) (Shimadzu, EPMA-1610).

Figs. 1(a–c) present PL spectra of Ta_2O_5:Y_2O_3 films deposited using two, three, and four Y_2O_3 pellets and annealed at 700, 800,

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http://dx.doi.org/10.1016/j.rinp.2014.09.004
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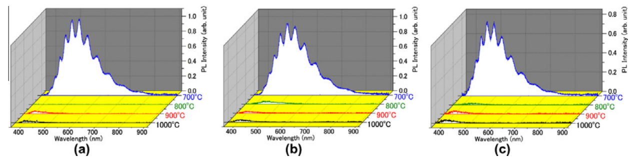

Fig. 1. PL spectra of Ta_2O_5:Y_2O_3 co-sputtered films prepared using (a) two, (b) three, and (c) four Y_2O_3 pellets and annealed at 700, 800, 900, or 1000 °C for 20 min.

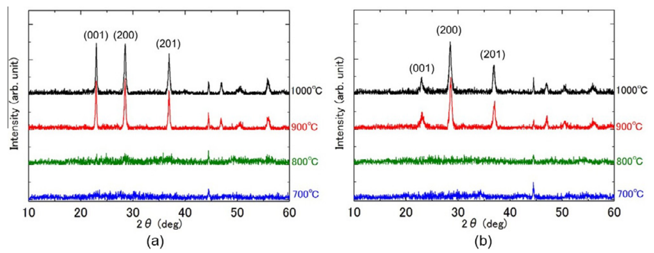

Fig. 2. XRD patterns observed from Ta_2O_5:Y_2O_3 co-sputtered films prepared using (a) two and (b) four Y_2O_3 pellets and annealed at 700, 800, 900, or 1000 °C for 20 min.

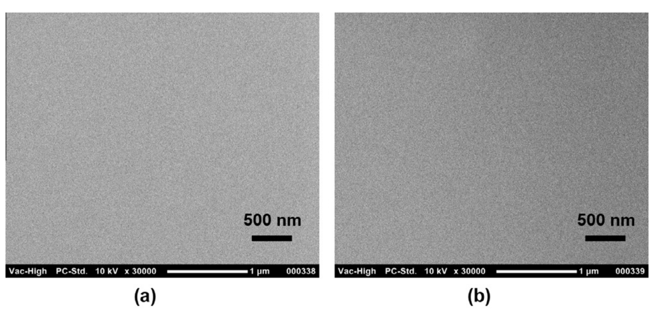

Fig. 3. SEM images of surfaces of Ta_2O_5:Y_2O_3 co-sputtered films prepared using three Y_2O_3 pellets and annealed at (a) 700 and (b) 800 °C for 20 min.

900, or 1000 °C. The Y concentrations of the films prepared using two, three, and four pellets were measured to be around 1.3, 2.4, and 3.5 mol%, respectively. Broad PL spectra from 380 to 800 nm were observed only from the films annealed at 700 °C. The periodic ripples in the spectra seem to be the result of optical interference between the two interfaces of the films. We found that the maximum PL intensities were around 500 nm regardless of the Y concentrations of the films.

Figs. 2(a) and (b) depict XRD patterns of Ta_2O_5:Y_2O_3 films deposited using two and four Y_2O_3 pellets and annealed at 700, 800, 900, or 1000 °C. Three major peaks corresponding to (001); β-Ta_2O_5 (orthorhombic), (200); δ-Ta_2O_5 (hexagonal), and (201) phases of Ta_2O_5 [6] were observed from the films annealed at 900 or 1000 °C, but the films annealed at 700 or 800 °C seem to be primarily amorphous phases because no remarkable diffraction peak was observed. In addition, Y_2O_3 in our Ta_2O_5:Y_2O_3 films seems to exist as amorphous-phase particles because no diffraction peak corresponding to crystalline Y_2O_3 [9] was also observed from the films annealed at 700, 800, 900, or 1000 °C. Therefore, it seems that there is no relationship between the broad PL and the crystallizability of the films. Figs. 3 (a) and (b) present scanning electron microscope (SEM) images of surfaces of Ta_2O_5:Y_2O_3 films deposited using three Y_2O_3 pellets and annealed at 700 °C; strong PL was observed, and 800 °C; strong PL was not observed, respectively. We found that the surfaces of the films are relatively smooth, but we could not observe clear differences between the surfaces.

Some trap levels and shallow centres of oxygen vacancies in the bandgap of Ta_2O_5 corresponding to light emission ranging from green to red wavelengths have been reported [10–12]. In addition, light emission ranging from blue to red wavelengths was also observed from Y_2O_3 [13]. Therefore, the broad PL spectra from Ta_2O_5:Y_2O_3 films annealed at 700 °C in Fig. 1 seem to originate from oxygen vacancies of Ta_2O_5 and Y_2O_3 particles that may be produced in Ta_2O_5 by co-sputtering. It is currently very difficult to distinguish and determine the origin of the broad PL from Ta_2O_5:Y_2O_3 co-sputtered thin films, but we will continue to carefully investigate its origin and attempt to clarify this.

4. Conclusions

Ta_2O_5:Y_2O_3 thin films were prepared using our simple co-sputtering method for the first time, and their PL and XRD properties were evaluated. Broad PL spectra from 380 to 800 nm were observed only from films annealed at 700 °C. The maximum PL intensities were found around a wavelength of 500 nm regardless of the Y concentrations of the films, and the films annealed at 700 °C were primarily amorphous phases. It seems that the broad PL spectra from our Ta_2O_5:Y_2O_3 films originated from oxygen vacancies of Ta_2O_5 and Y_2O_3 particles that may be produced in Ta_2O_5 by co-sputtering. Such co-sputtered films can be used as high-refractive-index materials of Ta_2O_5/SiO_2 autocloned photonic crystals that can be applied to novel light-emitting devices.

Acknowledgments

This work was supported by the "Element Innovation" Project by the Ministry of Education, Culture, Sports, Science, and Technology in Japan; JSPS KAKENHI Grant Number 26390073; and the Iketani Science and Technology Foundation. Part of this work was conducted at the Human Resources Cultivation Center (HRCC), Gunma University, Japan.

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