Materials Sciences and Applications, 2015, 6, 343-347
http://dx.doi.org/10.4236/msa.2015.65039

Fabrication of Erbium and Ytterbium Co-Doped Tantalum-Oxide Thin Films Using Radio-Frequency Co-Sputtering

Kenta Miura*, Yuki Arai, Kazusa Kano, Osamu Hanaizumi
Graduate School of Science and Technology, Gunma University, Kiryu, Japan
Email: *mkenta@gunma-u.ac.jp

Received 15 February 2015; accepted 20 April 2015; published 21 April 2015

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Abstract
An erbium and ytterbium co-doped tantalum-oxide (Ta2O5:Er, Yb) thin film was fabricated using a simple co-sputtering method for the first time, and its photoluminescence (PL) spectrum was evaluated. Energy transfers between Er³⁺ and Yb³⁺ in the Ta2O5:Er, Yb co-sputtered thin film were discussed by comparing between PL spectra of the Ta2O5:Er, Yb film and Ta2O5:Er or Ta2O5:Yb films reported in our previous works. Such a Ta2O5:Er, Yb co-sputtered film can be used as a high-refractive-index and light-emitting material of a multilayered photonic crystal that can be applied to a novel light-emitting device, and it will also be used as a multi-functional coating film having both anti-reflection and down-conversion effects for realizing a high-efficiency silicon solar cell.

Keywords
Tantalum Oxide, Erbium, Ytterbium, Co-Sputtering, Photoluminescence

1. Introduction
Many studies on rare-earth-doped tantalum (V) oxide (Ta2O5) have been conducted because Ta2O5 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 SiO2 [1]. Visible photoluminescence (PL) from erbium-doped Ta2O5 (Ta2O5:Er) produced by the sol-gel method [2] [3] and ion implantation [4] has been reported. Their PL spectra have main peaks at a wavelength of 550 nm due to the 4S3/2→4I15/2 transition of Er³⁺, and at a wavelength of 670 nm due to the 4F9/2→4I15/2 transition of Er³⁺. We previously demonstrated that Ta2O5:Er thin films deposited using a simple co-sputtering method exhibited such PL peaks at wavelengths of 550 and 670 nm after annealing at 600°C to 1100°C [5] [6].

*Corresponding author.

Recently, we also fabricated ytterbium-doped Ta$_2$O$_5$ (Ta$_2$O$_5$:Yb) thin films using the same co-sputtering method in order to expand the useful wavelength range of our rare-earth-doped and light-emitting Ta$_2$O$_5$ co-sputtered films [7]. We observed PL spectra having sharp peaks at a wavelength of 980 nm from the Ta$_2$O$_5$:Yb thin films after annealing at 700°C to 1000°C [7]. The 980-nm peaks seemed to be the result of the $^2F_{5/2} \rightarrow ^2F_{7/2}$ transition of Yb$^{3+}$ [7].

Furthermore, in our recent works, we demonstrated Tm and Ce co-doped Ta$_2$O$_5$ (Ta$_2$O$_5$:Tm, Ce) [8], Er and Ce co-doped Ta$_2$O$_5$ (Ta$_2$O$_5$:Er, Ce) [9], and Er, Eu, and Ce co-doped Ta$_2$O$_5$ (Ta$_2$O$_5$:Er, Eu, Ce) [10] thin films prepared using the co-sputtering method. In this work, we fabricated an Er and Yb co-doped Ta$_2$O$_5$ (Ta$_2$O$_5$:Er, Yb) thin film using radio-frequency (RF) magnetron co-sputtering of Ta$_2$O$_5$, Er$_2$O$_3$, and Yb$_2$O$_3$ for the first time, and evaluated its PL property.

### 2. Experimental

A Ta$_2$O$_5$:Er, Yb thin film was prepared using our co-sputtering method reported in [5][13]. A Ta$_2$O$_5$ disc (99.99% purity, diameter 100 mm), two Er$_2$O$_3$ pellets (99.9% purity, diameter 21 mm), and two Yb$_2$O$_3$ pellets (99.9% purity, diameter 21 mm) were used as co-sputtering targets. The Er$_2$O$_3$ and Yb$_2$O$_3$ pellets were placed on the Ta$_2$O$_5$ disc as shown in Figure 1. The film was deposited using a RF magnetron sputtering system (ULVAC, SH-350-SE). The flow rate of Ar gas introduced into the vacuum chamber was 10 sccm, and the RF power supplied to the targets was 200 W. A fused-silica plate (1 mm thick) was used as a substrate, and it was not heated during co-sputtering. We subsequently annealed the film in ambient air at 900°C for 20 min using an electric furnace (Denken, KDF S-70). The PL spectrum of the Ta$_2$O$_5$:Er, Yb film was measured using a dual-grating monochromator (Roper Scientific, SpectraPro 2150i) and a CCD detector (Roper Scientific, Pixis:100B, electrically cooled to $-80$ °C) under excitation with a He-Cd laser (Kimmon, IK3251R-F, wavelength $\lambda = 325$ nm).

### 3. Results and Discussion

Figure 2 presents PL spectra of Ta$_2$O$_5$:Er, Yb (red line) and Ta$_2$O$_5$:Er (without Yb, black line) [5] [6] co-sputtered thin films. We observed typical PL peaks around wavelengths of 550, 670, 850, and 980 nm from both the films. The 550-, 670-, and 850-nm peaks seem to be the results of the $^4S_{3/2} \rightarrow ^4I_{15/2}$, $^4F_{9/2} \rightarrow ^4I_{15/2}$, and $^4I_{9/2} \rightarrow ^4I_{15/2}$ transitions of Er$^{3+}$, respectively [5] [14] [15]. The 980-nm peaks seem to be the results of the $^4I_{11/2} \rightarrow ^4I_{15/2}$ transition of Er$^{3+}$ or the $^2F_{5/2} \rightarrow ^2F_{7/2}$ transition of Yb$^{3+}$ [7] [14]-[16]. From Figure 2, we can find that the 550-, 670-, and 850-nm peaks from the Ta$_2$O$_5$:Er film decreased by Yb doping. In contrast, the intensity of the 980-nm peak from the Ta$_2$O$_5$:Er, Yb film was stronger than that from the Ta$_2$O$_5$:Er film. This seems to be because of overlapping between the above-mentioned $^4I_{11/2} \rightarrow ^4I_{15/2}$ transition of Er$^{3+}$ and the $^2F_{5/2} \rightarrow ^2F_{7/2}$ transition of Yb$^{3+}$, and energy transfers from Er$^{3+}$ to Yb$^{3+}$ reported in [14]. Figure 3 illustrates energy level diagrams of Er$^{3+}$ and Yb$^{3+}$ [14] [15]. The energies of $^4S_{3/2}$ (the origin of the 550-nm peak), $^4F_{9/2}$ (the origin of the 670-nm peak), and $^4I_{9/2}$ (the origin of the 850-nm peak) states of Er$^{3+}$ seem to transfer through the $^4I_{11/2}$ state of Er$^{3+}$ to the $^2F_{5/2}$ state of Yb$^{3+}$ as presented in Figure 3.
Figure 4 presents PL spectra of the same Ta₂O₅:Er, Yb film (red line) and a Ta₂O₅:Yb film (without Er, green line) reported in [7]. The 980-nm peak from the Ta₂O₅:Yb film is much stronger than that from the Ta₂O₅:Er, Yb film. This seems to be because the opposite energy transfer from Yb³⁺ to Er³⁺ occurred in the Ta₂O₅:Er, Yb film. The energy of the 2F⁵/₂ state of Yb³⁺ partially transfer to the 4I₁₁/₂ state of Er³⁺ at first, and subsequently relax to the 4I₁₃/₂ state as presented in Figure 3. Finally light emission around a wavelength of 1550 nm due to the 4I₁₃/₂→2F⁷/₂ transition of Er³⁺ seems to occur [14]. The 1550-nm emission may cause the decrease of the 980-nm-peak intensity. Unfortunately, our detector did not detect the light emission in the wavelength range. We will try to evaluate the light-emission properties of our Ta₂O₅:Er, Yb films in the near-infrared range in order to make the mechanism of the energy transfer between Er³⁺ and Yb³⁺ clearer.
Such a Ta$_2$O$_5$:Er, Yb co-sputtered thin film can be used as a high-refractive-index and light-emitting material of a multilayered photonic crystal that can be applied to a novel light-emitting device [17], and it will also be used as a multi-functional coating film having both anti-reflection [18] and down-conversion [14]-[16] effects for realizing a high-efficiency silicon solar cell.

4. Summary

A Ta$_2$O$_5$:Er, Yb thin film was fabricated using our simple co-sputtering method for the first time, and its PL spectrum was evaluated. Energy transfers between Er$^{3+}$ and Yb$^{3+}$ in our Ta$_2$O$_5$:Er, Yb co-sputtered film were discussed by comparing between PL spectra of the film and our Ta$_2$O$_5$:Er or Ta$_2$O$_5$:Yb films. Such a Ta$_2$O$_5$:Er, Yb co-sputtered thin film can be used as a high-refractive-index and light-emitting material of a multilayered photonic crystal that can be applied to a novel light-emitting device, and it will also be used as a multi-functional coating film having both anti-reflection and down-conversion effects for realizing a high-efficiency silicon solar cell.

Acknowledgements

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

References


