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

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Abstract

An erbium and ytterbium co-doped tantalum-oxide (Ta_2O_5: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 Er^{3+} and Yb^{3+} in the Ta_2O_5:Er, Yb co-sputtered thin film were discussed by comparing between PL spectra of the Ta_2O_5:Er, Yb film and Ta_2O_5:Er or Ta_2O_5:Yb films reported in our previous works. Such a Ta_2O_5: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 (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 [1]. Visible photoluminescence (PL) from erbium-doped Ta_2O_5 (Ta_2O_5: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 $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+}, and at a wavelength of 670 nm due to the $^4F_{9/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+}. We previously demonstrated that Ta_2O_5: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].

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Recently, we also fabricated ytterbium-doped Ta_2O_5 (Ta_2O_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_2O_5 co-sputtered films [7]. We observed PL spectra having sharp peaks at a wavelength of 980 nm from the Ta_2O_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_2O_5 (Ta_2O_5:Tm, Ce) [8], Er and Ce *co-doped* Ta_2O_5 (Ta_2O_5:Er, Ce) [9], and Er, Eu, and Ce *co-doped* Ta_2O_5 (Ta_2O_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_2O_5 (Ta_2O_5:Er, Yb) thin film using radio-frequency (RF) magnetron co-sputtering of Ta_2O_5, Er_2O_3, and Yb_2O_3 for the first time, and evaluated its PL property.

2. Experimental

A Ta_2O_5:Er, Yb thin film was prepared using our co-sputtering method reported in [5]-[13]. A Ta_2O_5 disc (99.99% purity, diameter 100 mm), two Er_2O_3 pellets (99.9% purity, diameter 21 mm), and two Yb_2O_3 pellets (99.9% purity, diameter 21 mm) were used as co-sputtering targets. The Er_2O_3 and Yb_2O_3 pellets were placed on the Ta_2O_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_2O_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 λ = 325 nm).

3. Results and Discussion

Figure 2 presents PL spectra of Ta_2O_5:Er, Yb (red line) and Ta_2O_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_2O_5:Er film decreased by Yb doping. In contrast, the intensity of the 980-nm peak from the Ta_2O_5:Er, Yb film was stronger than that from the Ta_2O_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**.

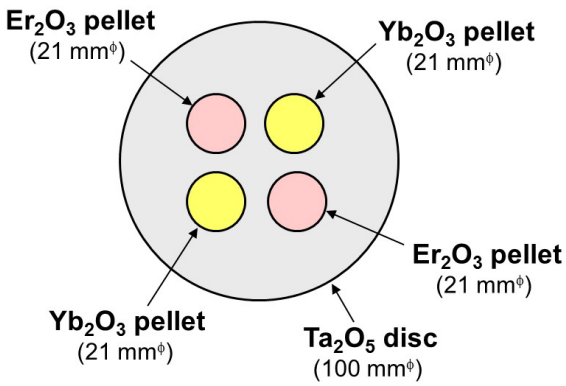

Figure 1. Schematic diagram of the sputtering target for co-sputtering of Er_2O_3, Yb_2O_3, and Ta_2O_5 used in this work.

Figure 4 presents PL spectra of the same Ta_2O_5:Er, Yb film (red line) and a Ta_2O_5:Yb film (without Er, green line) reported in [7]. The 980-nm peak from the Ta_2O_5:Yb film is much stronger than that from the Ta_2O_5:Er, Yb film. This seems to be because the opposite energy transfer from Yb^{3+} to Er^{3+} occurred in the Ta_2O_5:Er, Yb film. The energy of the $^2F_{5/2}$ state of Yb^{3+} partially transfer to the $^4I_{11/2}$ state of Er^{3+} at first, and subsequently relax to the $^4I_{13/2}$ state as presented in **Figure 3**. Finally light emission around a wavelength of 1550 nm due to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+} 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 Ta_2O_5:Er, Yb films in the near-infrared range in order to make the mechanism of the energy transfer between Er^{3+} and Yb^{3+} clearer.

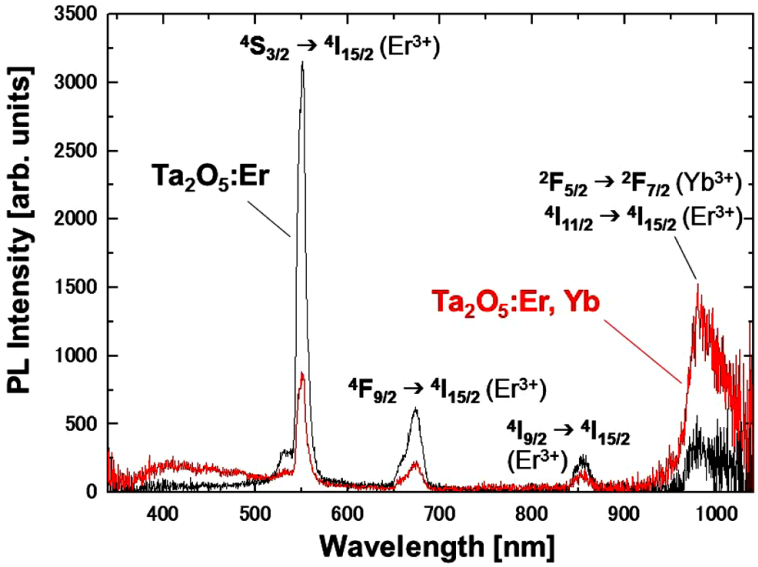

Figure 2. PL spectra of Ta_2O_5:Er, Yb and Ta_2O_5:Er [5] [6] co-sputtered thin films.

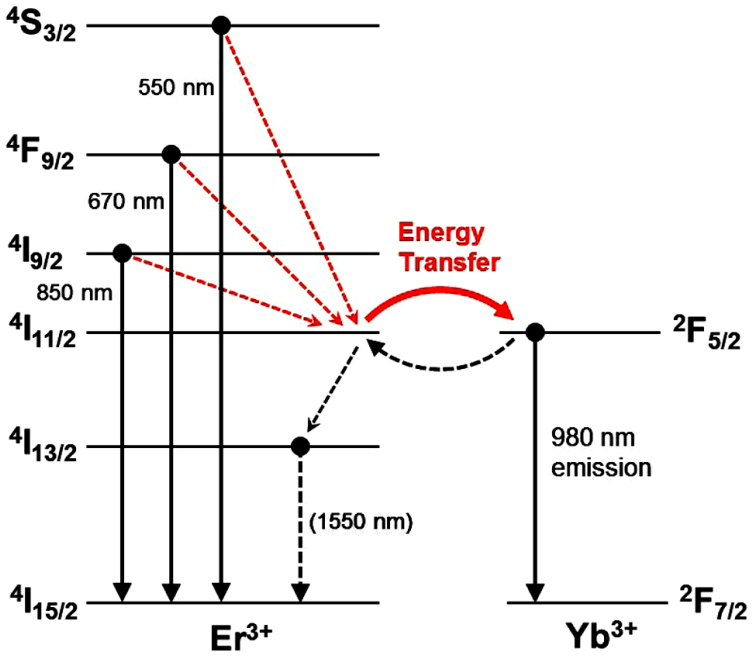

Figure 3. Energy level diagrams of Er^{3+} and Yb^{3+} [14] [15].

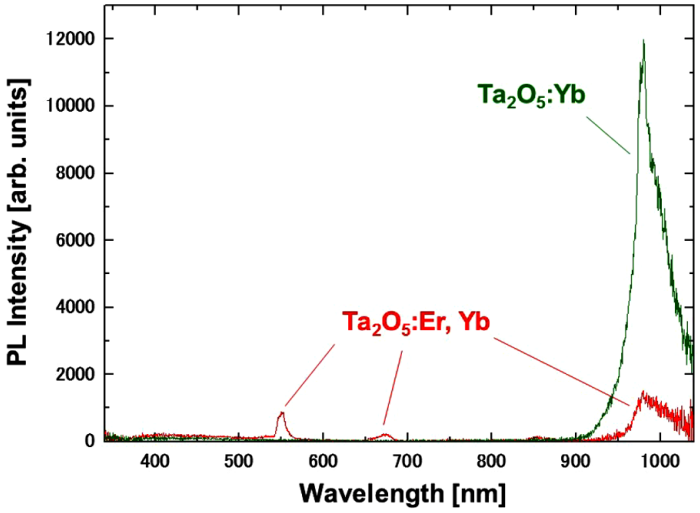

Figure 4. PL spectra of Ta_2O_5:Er, Yb and Ta_2O_5:Yb [7] co-sputtered thin films.

Such a Ta_2O_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_2O_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_2O_5:Er, Yb co-sputtered film were discussed by comparing between PL spectra of the film and our Ta_2O_5:Er or Ta_2O_5:Yb films. Such a Ta_2O_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.

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