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Intense photoluminescence from erbium-doped tantalum oxide thin films deposited by sputtering

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Abstract: Erbium-doped tantalum oxide films were prepared by radio-frequency magnetron sputtering. Visible light emission was observed from the films after annealing. We obtained PL peaks at 550 and 670 nm. The effects of erbium concentration, annealing temperature, and annealing time on the light-emitting properties of the films are discussed. The strongest intensities of the 550 and 670 nm peaks were observed from the samples with 0.96 and 0.63 mol% erbium concentrations after annealing at 900°C for 20 min, respectively.

Keywords: erbium, tantalum oxide, photoluminescence, annealing, sputtering

Classification: Optoelectronics, Lasers and quantum electronics, Ultrafast optics, Silicon photonics, Planar lightwave circuits

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1 Introduction

Rare-earth doped thin films are drawing increasing attention for their use in amplifiers and lasers and their suitability for integrated optics. The optical properties of rare-earth ions in solids have been investigated widely and are well understood [1]. Er^{3+}-doped materials are attracting much attention because of the search for solid-state-laser devices operating in the green region, optical devices for 3D displays and for waveguides which can work in telecommunication window. Recently green light emitting materials are in demand because they are useful in many applications (e.g., skypointer, unmanned aerial system, and mini projection system).

Tantalum pentaoxide (Ta_2O_5) is well known because of its interesting optical and electrical properties. It's a promising material for visible-wavelength optoelectronic devices. Ta_2O_5 can also be used as a composition material for the Ta_2O_5/SiO_2 photonic crystal elements, made by the autocloning

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method [2, 3]. Moreover, Er^{3+} is more soluble in Ta_2O_5 than silica. Recently, blue and red [4, 5] light emissions have been reported from Ta_2O_5 films. Some previous works reported photoluminescence around 550 and 670 nm from Er-doped TaO_x (Er-TaO_x) [6, 7, 8] using various fabrication techniques, such as sol-gel method and ion implantation.

In this paper, we report on the fabrication of Er-TaO_x films by radio-frequency (RF) magnetron sputtering for the first time and visible light emission at 550 and 670 nm. We optimized the fabrication and annealing conditions to obtain strong light intensity from the sputtered films. Such Er-TaO_x films can be used as a high-refractive-index material for autocloning type photonic crystals to realize novel light-emitting devices.

2 Experiments

Er-TaO_x films were fabricated using an RF magnetron sputtering system, (ULVAC, SH-350-SE). A Ta_2O_5 disc (99.99% purity, diameter 100 mm) was used as a sputtering target. We placed Er_2O_3 tablets (99.9% purity, diameter 21 mm) on the Ta_2O_5 disc. Ta_2O_5 and Er_2O_3 co-sputtered films were deposited by supplying RF power to the target. The pressure in the vacuum chamber of the sputtering system was 0.54×10^{-4} to 1.06×10^{-4} Pa, the Ar flow rate in the chamber was 10 sccm, and the pressure during deposition was approximately 1.1 Pa. The RF power supplied to the target was 300 W. Fused silica substrates (ATOCK Inc., thickness 1 mm) were used. The substrates were not heated during sputtering.

The samples were annealed in ambient air from 600 to 1100°C for 10 to 40 min using an electric furnace (Denken, KDF S-70). In order to vary the Er concentration, we placed 1 to 5 tablets of Er_2O_3 on the Ta_2O_5 disc during the sputtering process. The photoluminescence (PL) of the samples was measured with a dual-grating monochromator (Roper Scientific, Spectra Pro 2150i) and a CCD detector (Roper Scientific, Pixis-100). An He-Cd laser (Kimmon, 325 nm, max power 200 mW) was used for excitation of the samples. The Er concentration in the films after annealing was measured by an electron-probe micro-analyzer (EPMA, Shimadzu EPMA-1610). The X-ray diffraction (XRD) patterns of the films annealed at various temperatures were recorded by a Rigaku diffractometer using the Cu Kα radiation at a scanning step of 0.02°.

3 Results and discussion

Fig. 1 (a) plots the PL spectra of Er-TaO_x films fabricated using two Er_2O_3 tablets and annealed from 600 to 1100°C for 20 min. The as-deposited samples did not exhibit PL. We observed two PL peaks from post annealed samples at 550 and 670 nm. The sample annealed at 900°C exhibits the strongest PL intensity. The PL peaks observed at 550 and 670 nm are due to the transitions of Er^{3+} from the $^4S_{3/2}$ to the $^4I_{15/2}$ state and from the $^4F_{9/2}$ to the $^4I_{15/2}$ state, respectively [9]. Light emission at 670 nm could not be observed in the samples annealed at 600 and 700°C.

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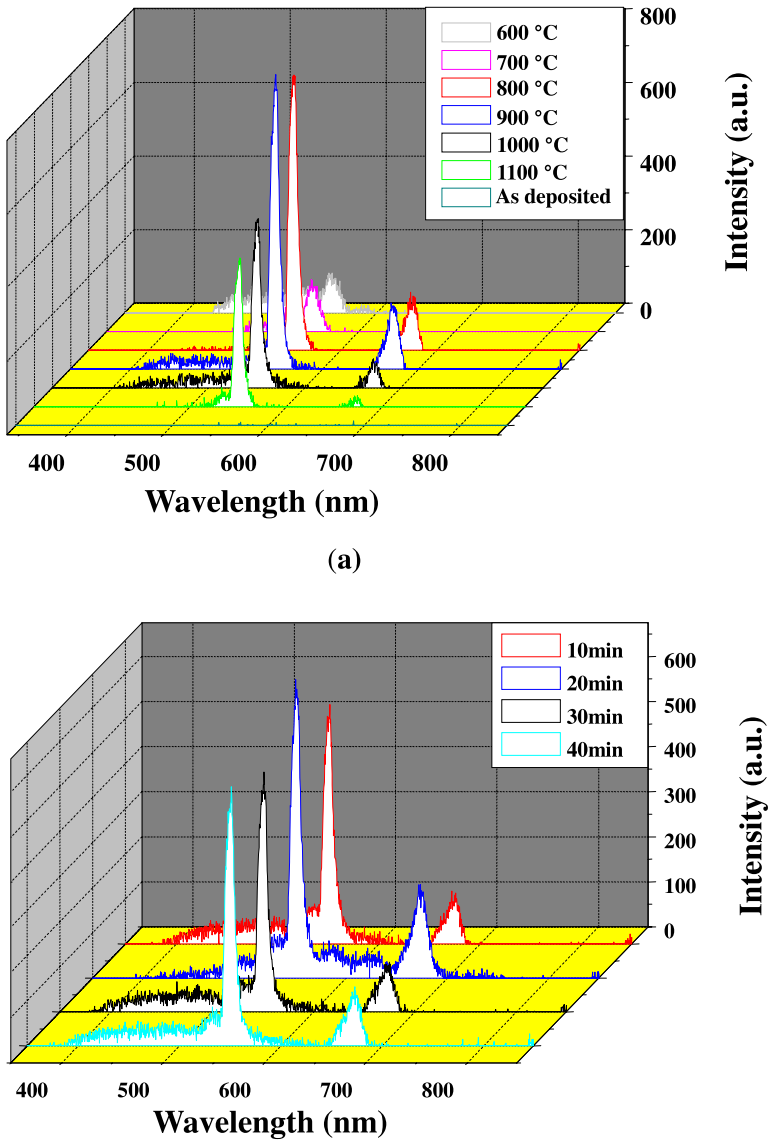

Fig. 1. PL spectra of Er-TaO_x films annealed at **(a)** 600 to 1100°C for 20 min, and **(b)** 900°C for 10 to 40 min.

To observe the effect of annealing time on PL intensity and to optimize the annealing time, we annealed samples (fabricated with two Er_2O_3 tablets) at 900°C for 10 to 40 min in ambient air. Fig. 1 (b) plots the PL spectra for different annealing times. The sample annealed for 20 min had the strongest PL intensities at both wavelengths of 550 and 670 nm.

Fig. 2 plots the PL spectra with different Er concentrations annealed at 900°C for 20 min. It has been reported that fluorescence due to Er^{3+} exhibits the strongest intensity at 1.2, 0.75, and 2 mol% Er concentration [10, 11, 12]. In our work, we observed the strongest PL intensity with 0.96 and 0.63 mol% Er concentrations at 550 and 670 nm, respectively. Table I shows the detailed results of Er concentrations in different samples measured with EPMA.

We observed an increase in Er concentration with the increasing number

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Table I. Concentration of Er in different samples measured with EPMA.

Number of Er_2O_3 tablet on Ta_2O_5 disc	1	2	3	4	5
Er concentration (mol %)	0.46	0.63	0.96	1.32	2.08

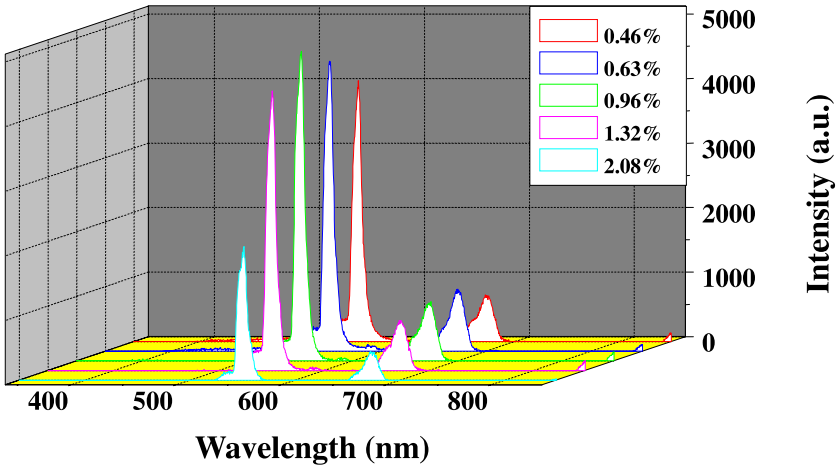

Fig. 2. PL spectra of samples with different Er concentrations (mol %) annealed at 900°C for 20 min.

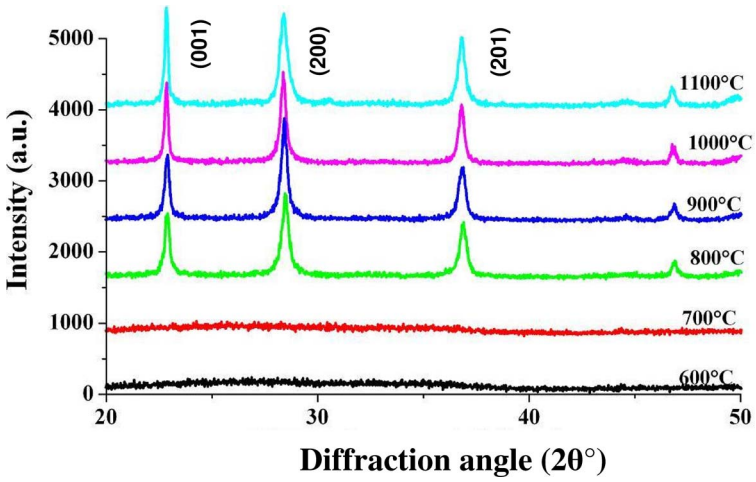

Fig. 3. XRD patterns of the films annealed at various temperatures.

of Er_2O_3 tablets on the Ta_2O_5 disc. It appears that different fabrication methods and dissolution sites of Er in the host material affect the Er concentration required to optimally enhance the PL intensity.

Fig. 3 shows the XRD patterns of the films annealed at various temperatures. The diffraction pattern fits well with references 13 and 14. The absence of 2θ peaks in samples annealed below 800°C indicates amorphous Ta_2O_5 film formation. XRD patterns of the samples annealed at 800 and 900°C probably show a mixed phase of δ-Ta_2O_5 (hexagonal, low-temperature phase) and β-Ta_2O_5 (orthorhombic, high-temperature phase) [15]. High PL intensities

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in the samples annealed at 800 and 900°C could be a result of crystallization of the films. We can observe a change in intensity of the first peak ($2\theta = 23°$) in the samples annealed at 1000 and 1100°C. In the samples annealed at 1000 and 1100°C, β-Ta_2O_5 (orthorhombic) grew more than δ-Ta_2O_5 (hexagonal), therefore a change in intensity was observed.

As discussed earlier, about some previous works reported PL from Er-TaO_x films. Rigneault et al [7] reported fluorescence spectra centered at 532 and 1530 nm from Er-TaO_x samples fabricated with ion implantation. Kojima et al [6] also observed a green fluorescence peak around 550 nm from samples fabricated with sol-gel process. Maeda et al [8] also reported fluorescence spectra around 550 nm and a weak peak around 660 nm with sol-gel process. In comparison to these previous works, we observed two very sharp PL peaks at 550 and 670 nm. Moreover, we observed visible green light emission by the naked eye which was not reported previously. This concludes that PL intensity at 550 nm is stronger in our results than previously reported works. Sputtering method has some advantages over the other deposition methods as the films deposited by sputtering have a composition close to that of the source material. Sputtered films have a better adhesion on the substrate than the evaporated films. In addition, the sputtering method is more suitable for mass production as it requires a single target and the films are relatively less contaminated.

TaO_x is superior to SiO_2 and GeO_2 in terms of both the phonon energy and Er^{3+} solubility. Amorphous Ta_2O_5 has better Er^{3+} solubility than SiO_2 because of the relatively large amount of edge oxygen having a negative charge [12]. The crystalline bulk phase of Ta_2O_5 is known to exist below 1630 K [16], but its crystal structure is not uniquely determined. Processing conditions and impurities have subtle effects on the crystal structure and light emission. It is surmised that the change in light intensity of films after annealing is the result of changes in the phase, the crystal structure, and the dissolution site of Er^{3+} in the Ta_2O_5.

4 Conclusion

We fabricated Er-TaO_x films using the RF-sputtering method and then annealed them at various temperatures and time durations. PL peaks were observed at wavelengths of 550 and 670 nm. We observed the strongest intensities of the 550 and 670 nm peaks from the samples with 0.96 and 0.63 mol% Er concentrations after annealing at 900°C for 20 min, respectively. To the best of our knowledge, this is the first report of light emission from Er-TaO_x films fabricated by the RF-sputtering method.

These results demonstrate that Er-TaO_x films fabricated by RF sputtering can serve as high-quality luminescent layers. These can easily be combined with other passive devices to realize novel active devices (e.g., a green-light-emitting photonic crystals), as only sputtering and annealing processes are needed for fabrication.

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