Applications of Ion Beam Assisted Deposition in the Synthesis of Er-doped Films

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Er-doped Al_2O_3 films and Er-doped SiO_x films have been synthesized by using ion beam assisted deposition (IBAD) methods. The microstructures of films and their annealing behaviors have been studied by using transmission electron microscopy (TEM) and x-ray diffraction (XRD). According to the results of TEM and XRD, Er-doped Al_2O_3 films are dominantly amorphous when deposited below 500°C . The films became polycrystalline γ - Al_2O_3 after annealing at 800°C and 1000°C for 6 hours and unique α - Al_2O_3 after annealing at 1200°C for 2 hours. The refractive index of Al_2O_3 films is in the range of 1.65 to 1.70 at the substrate temperature of 70 - 500°C . High substrate temperature can improve the distribution of refractive index and optical loss of the Al_2O_3 films. Er-doped SiO_x films as deposited below 500°C are amorphous and change into nano-crystalline after 800°C annealing and polycrystalline after 1100°C annealing. The photoluminescence (PL) of Er-doped SiO_x can be detected after the films annealed at 700°C and PL intensity increases with the increase of annealing temperature in the range of 700°C to 1100°C .

Key Words : Ion Beam Assisted Deposition, Er-Doped Films, Microstructure, Photoluminescence

1. Introduction

Er-doped films have attracted much attention after the success of the Er-doped fiber amplifier^(1~3) because of their potential applications in integrated optics. Er-doped planar optical waveguides offer the possibility to constructing active devices, such as optical amplifiers waveguide laser or more complicated optical integrated circuits with more compact structure than fiber-based systems. The erbium is used as optical dopant because the transition of trivalent erbium from the first excited state to the ground state in Er^{3+} occurs at the energy of 0.8 eV, corresponding to a wavelength of $1.54\text{ }\mu\text{m}$, where standard silica-based optical fibers have their maximum transparency.

In order to optimize Er^{3+} -doped films, an extensive range of film materials has been used as the host for the dopant, including silica glasses, Al_2O_3 , Y_2O_3 , SiO_2 , BaTiO_3 and LiNbO_3 . Meanwhile, various film synthesis and/or doping methods, such as high-energy ion implantation, plasma-enhanced chemical vapor deposition, metal organic chemical vapor deposition, and sol-gel method^(4~7), have been used to fabricate Er-doped films. Moreover, a net optical gain of 2.3 dB has been reached from a 4-cm long Er^{3+} doped Al_2O_3 spiral optical waveguide amplifier when pumped with light with a power of only 9 mW at a wavelength of $1.48\text{ }\mu\text{m}$ ⁽⁸⁾. In the present report, we try to apply ion beam assisted deposition (IBAD) to synthesize Er-doped Al_2O_3 films and Er-doped SiO_x films. We have studied the microstructures of the films and their annealing behaviors by using transmission electron microscopy (TEM) and x-ray diffraction (XRD). The photoluminescence of Er-doped SiO_x films and its

annealing behaviors have also been investigated.

2. Experimentals

The deposition of Er-doped Al_2O_3 films and Er-doped SiO_x films was carried out in an IBAD system, where the films can be synthesized by sputter deposition of aluminum or silicon in the oxygen atmosphere and bombarded simultaneously by low energy oxygen ions. Er was doped in the films by using metal erbium rods in the diameter of 2 mm embedded into the target. The amount of erbium rods embedded controlled the concentration of erbium in the films.

Er-doped films were deposited on Si (100) substrates and optical glasses. The silicon wafers were cleaned by the standard cleaning method. The sputtering ions were Ar ions with energy of 3 keV and beam current of 50 mA. The bombardment of oxygen ions was 500 eV and 30-50 mA. The base vacuum of the system was in the order of 10^{-3} Pa. Before deposition, Ar ion sputtering was used to clean the surfaces of samples for 15 minutes. During deposition, oxygen flow was kept at 1.8 to 6.0 sccm and the working pressure was kept at 3.0×10^{-2} Pa. The substrate temperature was controlled at 70 - 500°C by using a heater. To investigate the annealing behaviors, Er-doped films were annealed up to 1200°C in the atmosphere.

3. Results and discussions

3. 1 Optical properties of Er- Al_2O_3 films

Refractive index is one of important parameters to influence the optical properties and structure design of optical waveguide. We have investigated the influence of substrate temperature on the refractive index

of the films by using ellipsometry and photon scanning tunneling microscopy (PSTM). **Figure 1** is the change of refractive index of the films with the increase of substrate temperature. We can see that the refractive index of the films increase from 1.65 to 1.70 with the increase of substrate temperature in the range of 70°C to 500°C. This result implies that the films become more condense with the increase of substrate temperature.

Figure 2 is the PSTM images of Er-doped Al_2O_3

Fig. 1 Influence of substrate temperature on the refractive index of Er-doped Al_2O_3 films.

Fig. 2 $6 \times 6 \mu\text{m}^2$ PSTM images of Er-doped Al_2O_3 films deposited at (a) 240°C and (b) 500°C.

films synthesized at 240°C and 500°C. PSTM image is the reflection of refractive index and morphology. In the comparison with AFM images of the films, we found that the PSTM images of the films are mainly contribution of refractive index. Comparing the two PSTM images obtained in low substrate temperature (240°C) and high substrate temperature (500°C), we can see that the distribution of refractive index of the films obtained at 500°C is more uniform than that at 240°C. The uniform refractive index at high substrate temperature can attribute to the atom diffusion on the surface.

The optical losses of Er-doped Al_2O_3 films were measured by using a prism coupling the He-Ne laser beam (wavelength $\lambda = 632 \text{ nm}$) into the optical waveguide and measuring the intensity distribution of the laser beam with transportation distance in the optical waveguide. The optical loss of the film obtained at 240°C is about 0.56 dB/cm. For the films obtained at 500°C, the laser beam in the optical waveguide is much darker than that obtained at low substrate temperature and its optical loss is lower than 0.1 dB/cm.

3. 2 Microstructures and annealing behaviors of Er- Al_2O_3 films

The photoluminescence spectrum of Er-doped materials is sensitive to the thermal annealing processing. Especially for ceramic oxides and semiconductors, annealing treatment is always necessary to obtain optically active Er³⁺. Apparently, the microstructures and bonding nature of optical waveguides influence the environment and the active configuration of Er in the film. **Figure 3** presents the TEM images and their electron diffraction patterns of Er-doped Al_2O_3 films as deposited by IBAD, annealing at 800°C and 1000°C for 6 hours, and 1200°C for 2 hours, respectively. Rutherford back-scattering analysis determinates the

Fig. 3 TEM images of Er-doped Al_2O_3 films (a) as deposited and annealed at (b) 800°C for 6 hours, (c) 1000°C for 6 hours, and (d) 1200°C for 2 hours.

Fig. 4 XRD spectra of Er-doped SiO_x films as deposited and annealed at different temperatures.

Table 1 Atomic concentrations of Er-doped SiO_x films as deposited and annealed at different temperatures.

	O	Al*	Si	Er
As deposited at 500°C	18.89	12.37	68.44	0.30
Annealed at 800°C for 2 hours	40.96	12.11	46.58	0.35
Annealed at 1100°C for 2 hours	43.56	13.73	42.38	0.35

*Al is from the aluminum slices to embed Er rods

ratio of Al to O is 1.99 : 3.00, which is very close to the stoichiometric composition of Al_2O_3 . As can be seen in the figures, the films as deposited by IBAD are typical amorphous at the substrate temperature of 70°C to 500°C. After annealing at 800°C for 6 hours, the films become to polycrystalline films. By indexing the electron diffraction pattern, we find the films are composed of $\gamma\text{-Al}_2\text{O}_3$ phase in the grain size of 50-100 nm. For the case of films annealing at 1000°C for 6 hours, the microstructure of the films is still dominated by $\gamma\text{-Al}_2\text{O}_3$ phase and the grains grow up to several hundreds nm. XRD examination shows that a few mount of $\alpha\text{-Al}_2\text{O}_3$ phase have appeared in the films. When annealing at 1200°C for 2 hours, the films transit into unique $\alpha\text{-Al}_2\text{O}_3$ phase in the grain size of 5-10 μm . Based on above results, we can summary the path of phase transition of Er-doped Al_2O_3 films obtained by IBAD as amorphous $\text{Al}_2\text{O}_3 \rightarrow \gamma\text{-Al}_2\text{O}_3 \rightarrow \gamma\text{-Al}_2\text{O}_3 + \alpha\text{-Al}_2\text{O}_3 \rightarrow \alpha\text{-Al}_2\text{O}_3$ with the change of annealing temperature from room temperature to 1200°C.

3. 3 Microstructures and annealing behaviors of Er- SiO_x films

Figure 4 shows the XRD spectra of Er-doped SiO_x films as deposited and annealed at different temperatures. In the figure, we can see that the films as deposited by IBAD below 500°C are typical amorphous. After annealing at 800°C for 2 hours, the films become to polycrystalline. By indexing the XRD spectrum, we find that the polycrystalline is silicon. TEM observation found the grains of polycrystalline silicon is in the size of 10-20 nm. For the case of samples annealing at 1100°C for 2 hours, the microstructure of the films is still dominated by polycrystal-

Fig. 5 PL spectra of Er-doped SiO_x films pumped with 980 nm laser.

line silicon and the grains grow up to 40-100 nm. The results in TEM observation and XRD analysis imply that oxygen and erbium in Er-doped SiO_x films may exist in the films in the form of dispersion. In other words, after annealed, Er-doped SiO_x films exist in the form of SiO_x with the structure of silicon.

Table 1 is the compositions of Er-doped SiO_x films as deposited and annealed at different temperatures, which were determined by using electron probe, where Al is from the aluminum slices to embed Er rods. As can be seen in the table, the concentration of oxygen increases with the increase of annealing temperature. However, no obvious evidence of silicon oxides was found in the TEM observations on the films both as deposited and annealed at 800°C and 1100°C. The possible reasons why silicon oxides were not observed in the films are considered as follows ; (1) oxygen in Er-doped SiO_x films may exist in the films in the form of dispersion, and (2) silicon oxides after annealing mainly appear in a very thin layer at the surface and were sputtered out by ions during the preparation of TEM samples.

3. 4 PL luminescence of Er- SiO_x films

Figure 5 is the PL spectra of Er-doped SiO_x films annealed at 800°C and 1100°C. The measurement of PL spectra was carried out with reflection mode and pumped by 980 nm laser. In the figure, we can see that the PL spectra are typical Er^{3+} luminescence corresponding to the transition of $^4I_{13/2} \rightarrow ^4I_{15/2}$ in the 4 f-shell of erbium ions. However, no PL spectrum was detected for Er-doped SiO_x films as deposited at 500°C. The PL was detected after annealing at 700°C. In the PL spectrum of Er-doped SiO_x films annealed at 800°C with reflection mode, two narrow lines from 980 nm laser were also detected. In the comparison between the PL spectra of Er-doped SiO_x films annealed at 800°C and 1100°C, we found the PL intensity of Er-doped SiO_x films annealed at 800°C is much lower than that annealed at 1100°C. The results mean that there are more active Er^{3+} in Er-doped SiO_x films annealed at 1100°C than that annealed at 800°C.

Figure 6 shows the change of PL intensity of Er-doped SiO_x films with laser power after the back-

Fig. 6 Change of PL intensity of Er-doped SiO_x films with laser power.

ground subtraction. As can be seen in the figure, The PL intensities of Er-doped SiO_x films annealed at 800°C and 1100°C increase with the increase of pump laser power. No obvious power saturation was observed up to 1 W for both films annealed at 800°C and 1100°C.

4. Conclusions

(1) Er-doped Al₂O₃ films and Er-doped SiO_x films synthesized by IBAD below 500°C are amorphous. The path of phase transition of Er-doped Al₂O₃ films is amorphous Al₂O₃ $\xrightarrow{800^\circ\text{C}/6\text{ h}}$ γ -Al₂O₃ $\xrightarrow{1000^\circ\text{C}/6\text{ h}}$ γ -Al₂O₃ + α -Al₂O₃ $\xrightarrow{1200^\circ\text{C}/2\text{ h}}$ α -Al₂O₃. Oxygen and Erbium in Er-doped SiO_x films may exist in the films in the form of dispersion. The annealing behavior of Er-doped SiO_x films is amorphous SiO_x $\xrightarrow{800^\circ\text{C}/2\text{ h}}$ SiO_x with structure of nanometer silicon $\xrightarrow{1100^\circ\text{C}/6\text{ h}}$ SiO_x with structure of polycrystalline silicon.

(2) The refractive index of Er-doped Al₂O₃ films

increase from 1.65 to 1.70 with the increase of substrate temperature in the range of 70°C to 500°C and high substrate temperature can improve the distribution of refractive index and optical loss of the films.

(3) No PL spectrum has been observed in Er-doped SiO_x films as deposited below 500°C. The PL intensity of Er-doped SiO_x films annealed at 1100°C is stronger than that annealed at 800°C; No obvious saturation of PL intensity was observed in Er-doped SiO_x films pumped by 980 laser with the power up to 1 W.

Acknowledgements

This work is supported by the National Natural Science Foundation of China under grant no. 69878002 and by the Education Ministry of China. The authors thank Prof. C.L. Song and his students in Dept. of physics, Dalian University of Technology, for the helps in the measurements of PL spectra.

(Received December 27, 2002 ; Accepted July 24, 2003)

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