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Optical activation of Er^{3+} in Al_2O_3 during pulsed laser deposition

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Abstract

Erbium doped Al_2O_3 thin films have been prepared by alternate pulsed laser deposition on substrates either at room temperature (RT) or heated up to 600 °C. The Er related photoluminescence intensity at 1.53 μm is enhanced by two orders of magnitude and the lifetime is increased by up to a factor of 6 when the films are deposited at temperatures $\geqslant$400 °C compared to the film grown at RT. The full width at half maximum (FWHM) of their emission spectra is as wide as 60 nm, consistently with the amorphous nature of the Al_2O_3 host. The Er photoluminescence emission in the film deposited at RT can be enhanced by post-deposition annealing at high temperature (850 °C) to achieve similar peak photoluminescence intensity and lifetime values. However, its FWHM is only 40 nm. Analysis of the photoluminescence response after post-deposition annealing treatments suggest that the microstructure of the amorphous Al_2O_3 host grown at temperatures $\geqslant$400 °C is different from that of films grown at RT, and seems to be more stable upon annealing.

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1. Introduction

The development of optical communication technologies requires Er-doped materials in thin film configuration for optical waveguide amplifiers in integrated systems [1,2]. The fabrication of such compact devices demands thin films with a high concentration of optically active Er^{3+} ions to achieve a reasonable gain, since only few centimeters are available, as compared to the hundreds of meters available in fiber optical amplifiers. A variety of methods have so far been used to obtain rare-earth doped thin films. Most of them, such as ion implantation, plasma-enhanced chemical vapor deposition, electrochemical deposition, sputtering, and sol–gel need a subsequent high temperature (>700 °C) thermal annealing [3–7] or pulsed laser annealing [8] of the films after preparation to activate the rare-earth ions and thus to optimize the photoluminescence (PL) response. This high temperature processing however restricts the flexibility and practical applicability of these methods for the development of integrated optical devices compatible with Si processing.

Pulsed laser deposition (PLD) has shown in the past decade that it is suitable for the deposition of high quality films for optical applications [9]. Moreover, in our earlier works, we have shown that Er-doped Al_2O_3 thin films with good PL performances and high Er concentrations (in the order of 10^{20} cm^{-3}) can be obtained by alternate PLD from two separate targets (for the host and the dopant material, respectively) [10]. However these films were grown at room temperature (RT), and required a thermal annealing up to 850 °C after deposition in order to achieve optimum performance. A possible route to overcome the post-deposition thermal treatment is to grow the films on heated substrates. In addition, this allows to reduce the two-step process (deposition at RT followed by film thermal

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annealing) to a one-step process (deposition at high temperature). The challenge is however to keep the temperature of the substrate low enough ($\leqslant$800 °C) to make this route compatible with other integrated functions and to prevent the crystallization of the Al_2O_3 host. The latter has been shown to produce a high increase of the propagation losses of passive Al_2O_3 waveguides produced by PLD probably related to the development of scattering centres [11]. Thus the aim of this work is to explore the feasibility of this one-step process by growing Er:Al_2O_3 films at temperatures higher than RT.

2. Experimental

An ArF excimer laser ($\lambda = 193$ nm, $\tau = 12$ ns full width at half maximum, 5 Hz repetition rate) with an energy density of 2 J cm^{-2} was used to ablate the Al_2O_3 and Er targets. They were mounted in a computer-controlled holder which allows their alternate ablation. The films were deposited in vacuum (10^{-6} Torr) on chemically cleaned Si(1 0 0) wafers held at RT (hereafter referred as the reference sample), 400, 500, and 600 °C. Some films were deposited in a 10^{-4} Torr oxygen atmosphere at 400 and 500 °C since it has recently been shown that losses could be reduced in this way [11]. Each substrate was previously heated at 500 °C for 1 h to relax stresses and to desorb the remaining impurities before starting deposition at the desired temperature.

The dopant was distributed in layers and the films were designed to have approximately 6 nm thick Al_2O_3 layers separating the Er doping layers, as this structure has been shown to lead to a good PL response [10]. To achieve this configuration, each deposition cycle consisted of 570 laser pulses on the Al_2O_3 target and 1 pulse on the Er one. This cycle was repeated 50 times with a final deposition of Al_2O_3 in order to cover the last Er doping layer. The Er content and the total film thickness were determined by Rutherford backscattering spectrometry (RBS) using a 1.0 MeV He^+ beam and a scattering angle of 162°. The RBS spectra were analyzed by means of the SIMNRA code.

PL measurements were performed at RT using a single grating monochromator (focal length 300 mm) with a wavelength resolution of 2.7 nm, a liquid-nitrogen cooled Ge detector, and standard lock-in techniques. The 514.5 nm line of an Ar^+ ion laser (power 200 mW) chopped at 10 Hz was used as the excitation source, the incident beam forming an angle of 25° with the sample normal. The emitted light was collected along the direction perpendicular to the film. Luminescence decay curves at the peak of the emission spectrum were averaged and recorded with a digital oscilloscope.

3. Results

The RBS spectra of all samples show that the average distribution of Er throughout the film thickness is flat. There is no evidence of segregation of Er to the surface, indicating that Er is effectively incorporated in the Al_2O_3 structure. This is similar to what has been reported for Er incorporation in lithium niobate by PLD at temperatures up to 600 °C [12]. The RBS analysis indicates that the Er areal density is similar for all the samples, the mean value being $(4.7 \pm 0.9) \times 10^{15}$ at. cm^{-2}. The average film thickness is 300 ± 10 nm.

The Er-doped Al_2O_3 films exhibit the characteristic emission band peaking at 1.53 μm, which is due to the $^4I_{13/2}$ to $^4I_{15/2}$ transition of the Er^{3+} ions [4]. The dependence of both the PL intensity and the lifetime of this emission on the substrate temperature during film growth is plotted in Fig. 1. It is clearly seen that both quantities are much higher for the films grown in vacuum at temperatures higher than RT than for the film grown at RT. Both the PL intensity and the lifetime approach saturation or undergo a maximum for a substrate temperature of 500 °C. The data of the samples deposited in oxygen atmosphere are also included in Fig. 1 and follow the same trend.

As it was reported elsewhere, annealing RT-deposited films in air in 50 °C steps from 600 to 850 °C largely improves their PL response [10]. The PL intensity and lifetime values for the reference sample after such an annealing process are also shown in Fig. 1. It can be seen in this figure

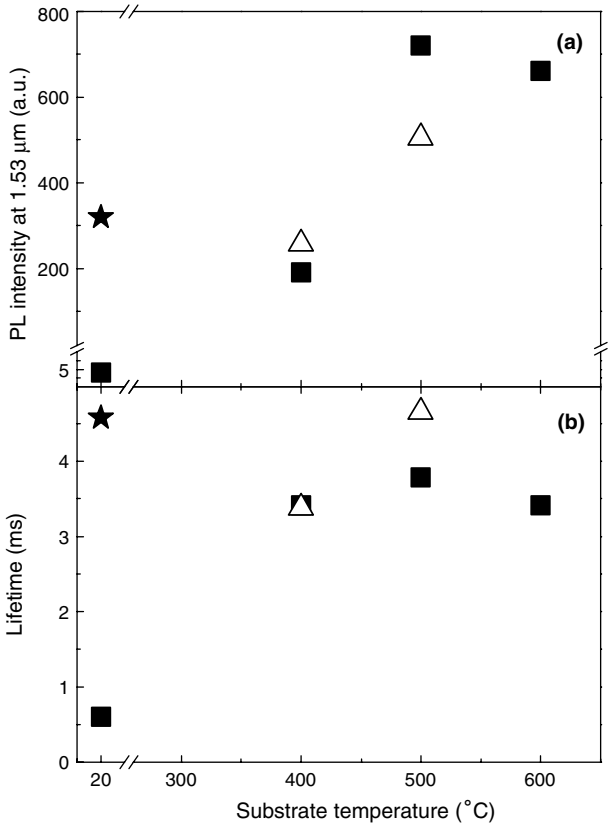

Fig. 1. Dependence on the substrate temperature of the PL intensity at 1.53 μm (a) and the lifetime (b) for Er:Al_2O_3 thin films grown in vacuum (■) and in a 10^{-4} Torr oxygen atmosphere (△). Values achieved upon annealing up to 850 °C in the sample grown at room temperature are indicated by ★.