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Citation: J. Appl. Phys. 84, 2352 (1998); doi: 10.1063/1.368304
View online: http://dx.doi.org/10.1063/1.368304
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Optically active Er–Yb doped glass films prepared by pulsed laser deposition

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(Received 2 March 1998; accepted for publication 5 May 1998)

Active rare-earth Er$^{3+}$–Yb$^{3+}$ co-doped phosphate glass films are produced in a single step by pulsed laser deposition. The films are multimode waveguides and exhibit the highest refractive index, optical density and 1.54 μm photoluminescence intensity and lifetime when deposited at low oxygen pressure ($P_{ox}$≤4×10$^{-5}$ Torr). The density of the films obtained under these conditions is higher than that of the target material as a consequence of the high kinetic energy of the species generated during ablation. Luminescent emission can be excited by optical pumping the Er$^{3+}$ ions either directly or through cross-relaxation of the Yb$^{3+}$. Post-deposition annealing allows us to improve the luminescence performance. © 1998 American Institute of Physics.

Optically active Er–Yb doped glass films prepared by pulsed laser deposition

Active planar waveguides are required to develop photonic integrated circuits. Pulsed laser deposition (PLD) is a recently developed technique which is compatible with deposition in gas environments and has been applied successfully to grow a wide variety of multicomponent materials in thin film configuration. It has been shown to be especially suited for the development of films for optical applications. The deposition of planar waveguides by PLD has been demonstrated, and most of the effort has been focused on achieving films of good epitaxial quality. As a result crystalline films of optical materials with large nonlinear optical coefficients (LiNbO$_3$, LiTaO$_3$, 3 Co:BaTiO$_3$, 4 optical materials with large nonlinear optical coefficients films of good epitaxial quality. As a result crystalline films of optical materials with large nonlinear optical coefficients (LiNbO$_3$, LiTaO$_3$, Co:BaTiO$_3$) and more recently, wide band gap materials doped with rare-earth ions (Eu:Y$_2$O$_3$ for electroluminescent displays) and Nd:Y$_3$AlO$_{12}$ (Nd:Gd$_3$Ga$_5$O$_{12}$) for laser-gain media) have been produced by PLD. In spite of the advantages of glasses for developing active waveguides, and although the growth of multicomponent oxides by PLD appears to be the most successful route, the number of studies devoted to the production of glassy films by PLD has been very limited.

Incorporation of the rare-earth ion erbium in waveguides allows the development of lasers and amplifiers operating at 1.5 μm, a wavelength of great interest for communication purposes. Phosphate glasses have often been used as hosts for Er$^{3+}$ because high rare-earth concentrations can be incorporated when compared to silicates, with the additional advantage of a larger emission cross section. In an earlier work, we have shown that waveguides can be produced by PLD from Er$^{3+}$–Yb$^{3+}$ co-doped phosphate glasses. In this communication we demonstrate that both the Er$^{3+}$ and Yb$^{3+}$ are active in these waveguides, and have special emphasis on the influence of the oxygen pressure ($P_{ox}$) during deposition on the refractive index (n) of the target glass is 1.5510. In order to produce the waveguides the films were grown on silicon substrates with a 3.4 μm silicon dioxide buffer layer (Si/SiO$_2$) and up to a thickness of ~1.5 μm. Their characterization has been done at 633 nm by using the dark-line prism coupling configuration. Propagation of at least two optical modes was observed for all the films, which allowed us to determine simultaneously the refractive index (n) and the thickness. Figure 1 shows the evolution of n as function of $P_{ox}$. The plot shows that n decreases as $P_{ox}$ increases, approaching the value of the target for $P_{ox}$ values close to 4.4×10$^{-2}$ Torr. In a homogeneous dielectric the n value can be related to the density (ρ) via de Lorentz–Lorenz relation. It follows that an increment of ρ leads to an increment of n:

\[ \frac{\Delta n}{n} = \frac{(n^2-1)(n^2+2)}{6n^2} \frac{\Delta \rho}{\rho}. \]  

It has been shown earlier that amorphous semiconductor films grown by PLD are denser than those grown with other deposition techniques due to the presence of energetic species during the PLD process. Moreover, studies of the plume expansion during PLD of complex oxide films have shown that $P_{ox}$ has a significant effect on the kinetic energy.
of the species ablated from the target, the higher the $P_{\text{ox}}$ or the lower the kinetic energy. The large decrease of $n$ observed at about $10^{-2}$ Torr, a pressure for which collisional processes with the foreign gas begin to be important, is most likely related to ablated species reaching the substrate with less kinetic energy. In a similar context densification of glasses under ion implantation has been known for a long time. For fused silica undergoing keV ion implantation a relative increase $\Delta n/n$ up to $10^{-2}$ has been reported. In our case $\Delta n/n$ for the films grown in vacuum compared to that for the target is $4.5 \times 10^{-2}$, which is of the same order of magnitude as that found for ion-implanted material. The results in Fig. 1 for $n$ can thus be interpreted in terms of densification, although minor variations in the deposited glass composition that may also influence $n$ cannot be completely disregarded. From an application viewpoint, the observed increase of $n$ achieved during growth at low $P_{\text{ox}}$ is beneficial for the production of waveguides with SiO$_2$ as cladding, since the larger the difference between $n$ of the cladding ($n = 1.46$) and the material of the built-in waveguide, the better the light confinement.

Photoluminescence (PL) measurements were performed at room temperature using a single grating monochromator (focal length 275 mm), a liquid nitrogen cooled Ge detector and standard lock-in techniques. The film PL was excited with either an Ar$^+$ (514 nm) or a tunable Ti:sapphire laser (770–880 nm) reaching the film at an incidence angle of 30°. The emitted light was collected normal to the film plane. The as-deposited PLD Er$^{3+}$–Yb$^{3+}$ co-doped phosphate glass films show broad PL spectra, typically from a glass matrix, and with a maximum at 1.54 $\mu$m corresponding to Er$^{3+}$ intra-f transition (${}^4I_{13/2} \rightarrow {}^4I_{15/2}$). Co-doping the films with Yb$^{3+}$ increases the wavelength range suitable for Er$^{3+}$ optical pumping. The Yb$^{3+}$ has a broad absorption band from approximately 800 to 1050 nm and cross-relaxation between adjacent ions Er$^{3+}$ and Yb$^{3+}$ allows the absorbed energy to be transferred to the Er$^{3+}$ system. This was observed in our films by using a Ti:sapphire tunable laser as pump source and recording both the PL excitation and emission spectra. The measurements show that the PL intensity is similar whichever the pumping wavelength in the range 800–880 nm, even at 880 nm where the Er$^{3+}$ absorption is negligible. The results thus evidence that both Er$^{3+}$ and Yb$^{3+}$ are active in the films.

Figure 1 also includes the PL intensity and lifetime at 1.54 $\mu$m when the films are excited at 514 nm. Both parameters show the same trend as the $n$ value, namely they decrease as $P_{\text{ox}}$ increases. Modifications of the PL intensity as a function of the $P_{\text{ox}}$ have been observed before in PLD Eu:Y$_2$O$_3$ films. For those films the PL intensity was observed to increase simultaneously with the surface roughness as the $P_{\text{ox}}$ was increased. The PL intensity enhancement thus was correlated to the reduced internal reflections caused by rough surfaces. In contrast no change in the surface roughness is observed for our films as a function of $P_{\text{ox}}$, while a PL intensity enhancement is found for low $P_{\text{ox}}$ ($P_{\text{ox}} \approx 4 \times 10^{-5}$ Torr). In addition it should be noted that this enhancement of the PL intensity is accompanied by an increase of the PL lifetime. For a fixed pump power and wavelength the PL intensity is proportional to the concentration of active Er$^{3+}$ ions, the pump absorption cross section, and to the ratio $\tau/\tau_r$, where $\tau$ and $\tau_r$ are the measured and the radiative lifetimes, respectively. Both lifetimes are related through the equation:

$$\frac{1}{\tau} = \frac{1}{\tau_r} + \frac{1}{\tau_{\text{nr}}},$$

where $\tau_{\text{nr}}$ is the nonradiative lifetime. Thus, the existence of nonradiative decay channels leads to a measured lifetime $\tau$ smaller than the radiative lifetime $\tau_r$. Assuming that $\tau_r$ is constant, the higher measured lifetimes ($\tau$) observed in the films deposited at low $P_{\text{ox}}$ can be interpreted in terms of a reduction of the nonradiative decay channels. Therefore the results show that there is a decrease of the concentration of defects for low $P_{\text{ox}}$ which is consistent with the discussed increase of packing density in the films.

It is worth noting, however, that the PL lifetime values measured for the films grown even at low $P_{\text{ox}}$ are unexpectedly short (hundreds of $\mu$s), one order of magnitude shorter than those observed in the target suggesting that the concentration of defects in the glass network is still not minimal. The films were thus annealed for 1 h at several temperatures at atmospheric pressure and cooled down to room temperature to perform additional PL measurements. The resulting PL intensities and lifetimes are shown in Fig. 2, where it is seen that both parameters are greatly improved upon annealing. For the films grown at low $P_{\text{ox}}$ ($P_{\text{ox}} \approx 4 \times 10^{-5}$ Torr) the PL intensity and lifetime increase for annealing up to 500–550 °C, to then decrease abruptly, at the same time that clear surface damage is observed. This decrease is not surprising since 600 °C is the glass transition temperature for most phosphate glasses. The increase of the lifetime is related to a decrease of nonradiative decay channels in the films as discussed above. Nevertheless, the relative increase in the PL lifetime (a factor of 9) is significantly lower than that of the PL intensity. This suggests that there is not only a decrease of the concentration of defects upon annealing but also an increase of the fraction of optically active Er$^{3+}$ ions. This...
decrease of the concentration of defects is not correlated to a change in density since no significant changes of $n$ were observed upon annealing. For the films grown at the highest $P_{\text{ox}}$ a much smaller and gradual increase of the PL intensity is observed up to 600 °C. For these films severe surface damage is already observed upon annealing at 400 °C. The moderate PL intensity enhancement observed in this case is most likely related to the high kinetic energy of the species reaching the substrate, and this is a remarkable result as most reports on the growth of complex oxide films by PLD are carried out at relatively high $P_{\text{ox}}$.

In conclusion, Er$^{3+}$–Yb$^{3+}$ co-doped glass films grown by PLD onto Si/SiO$_2$ substrates show guiding properties and clear Er$^{3+}$-related PL, either by optically pumping the Er$^{3+}$ directly or through cross-relaxation of the Yb$^{3+}$. Films deposited in vacuum or at low $P_{\text{ox}}$ show the highest density and the best PL performance, with lifetimes of several ms. The achieved high $n$ favors the light confinement in waveguides with SiO$_2$ cladding layers, and are thus promising for the development of Si-based optoelectronic devices. The improved performance of the films grown at low $P_{\text{ox}}$ is most likely related to the high kinetic energy of the species reaching the substrate, and this is a remarkable result as most reports on the growth of complex oxide films by PLD are carried out at relatively high $P_{\text{ox}}$.

This work has been partially supported by CICYT (Spain) under Project No. TIC96-0467. The authors thank G. C. Righini and S. Pelli (IROE, Florence, Italy) for providing the targets and useful discussions. One of the authors (J.M.B.) acknowledges financial support from the Spanish Ministry of Education.