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Materials Research Bulletin 40 (2005) 1915–1921
Materials Research Bulletin
www.elsevier.com/locate/matresbu

Influence of oxygen pressure on Nd:$LuVO_4$ films grown by pulsed laser deposition

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Received 14 January 2005; received in revised form 8 June 2005; accepted 9 June 2005
Available online 11 July 2005

Abstract

High quality Nd-doped lutecium vanadate thin films on silica glass substrates were fabricated successfully by using a pulsed laser deposition technique. The properties of the samples were characterized by using X-ray diffraction, Rutherford backscattering, atomic force microscopy (AFM), and prism-coupling measurements. The RBS shows that the ratio of Lu/V in the film is 0.991, which is in good agreement with the target composition. X-ray diffraction results show that the degree of crystal orientation along (2 0 0) increases with increasing oxygen pressure up to 20 Pa. The refractive indices of the films determined with dark-mode prism coupling measurements are slight, smaller than that of the bulk crystal. An optimum 20 Pa oxygen pressure, at which the oxygen was leaked into the chamber as the reactive ambient, was determined.
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PACS: 81.15.Fg; 42.79.Gn; 42.82.Et

Keywords: A. Thin films; B. Laser deposition; C. Atomic force microscopy; C. X-ray diffraction

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doi:10.1016/j.materresbull.2005.06.005

1. Introduction

In recent years, laser diode pumped solid-state lasers based on Nd-doped crystals have been extensively developed for the applications in the military, industry, scientific research, and medical treatment fields due to their high level of stability, efficiency, compactness, and longevity. At present, Nd:YAG and Nd:YVO_4 are two of the most used and studied laser crystals. Indeed, Nd:$LuVO_4$ is a new crystal material with the space group D_{4h}^{19}-$I4_1/amd$. It may be a promising candidate for lasers because of its excellent luminescence properties [1].

There is strong interest for using crystalline optical materials in the form of thin films [2]. For the fabrication of thin films, pulsed laser deposition (PLD) is a very useful and flexible tool. PLD permits a stoichiometric transfer of material from the target to the film and film growth at high temperatures in reactive ambient gases, in particular, oxygen [3]. In practice, the PLD technique has been used successfully to prepare many kinds of waveguide films, such as $KNbO_3$ [4], Ti:sapphire [5], SBN($Sr_xBa_{1-x}Nb_2O_6$) space [6], and $GdCa_4O(BO_3)_3$ [7], etc. [8–11]. In this paper, we report our work to grow Nd:$LuVO_4$ films on silica glass substrates by the PLD technique.

2. Experimental procedure

The deposition of the film was performed by using KrF excimer pulsed laser irradiation of 248 nm, 20 ns in pulse width, and 5 Hz frequency. The target used was a single bulk crystal sample of 0.9 at.% Nd-doped $LuVO_4$. The target was mounted on a rotating stage that changed the direction of rotation with a period of 1 min. The distance from the target surface to substrate was 3.5–5 cm. To reduce the surface roughness and enhance the crystallinity of the film, the substrate temperature and energy density of the excimer irradiation was optimized at 700 °C and 2.0 J/cm^2, respectively. The depositing time of the sample was 50–60 min. The pressure of the deposition chamber atmosphere was pumped down below 5.0×10^{-3} Pa prior to deposition. Oxygen was introduced into the chamber and kept constant between 2 and 20 Pa while depositing.

A Tencor Alphastep surface profiler (ET350) was used to measure film thickness. A Rigaku D/MAX-γ A X-ray diffractometer was used to investigate crystal quality and crystal structure. Rutherford backscattering was used to investigate the ratio of Lu/V in the prepared film. The backscattering analysis was conducted using 2.0 MeV helium beams. The lutecium to vanadium ratio was established from the energy spectra of backscattered helium ions, which easily resolve these elements from one another. A Digital Instruments Nanoscope IIIa atomic force microscope (AFM) was employed to measure the surface morphology of the vanadate films. All AFM images were taken in air using the contact mode. The characterization of optical waveguides has been performed at 633 nm wavelength by using a dark-line prism-coupling configuration (Metricon Modle 2010).

3. Results and discussion

The XRD patterns of typical Nd-doped lutecium vanadate films deposited in oxygen ambient pressure between 2 and 20 Pa at substrate temperature of 700 °C are shown in Fig. 1. For the sample prepared in an oxygen ambient pressure of 2 Pa (Fig. 1a), the diffraction peaks are very weak, except the (2 0 0)

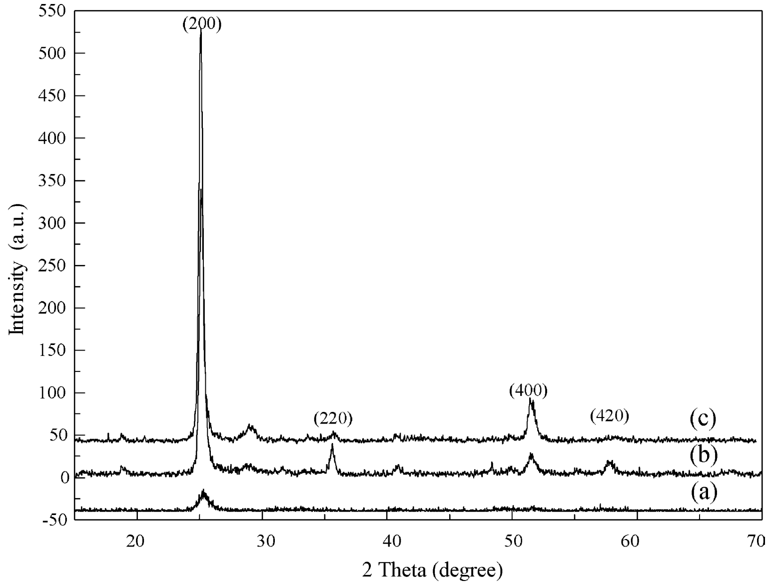

Fig. 1. X-ray diffraction pattern of Nd-doped lutecium vanadate thin films grown on silica glass substrates at substrate temperature of 700 °C and different oxygen pressures: (a) 2 Pa, (b) 10 Pa, and (c) 20 Pa.

diffraction one, some other strong peaks of Nd:$LuVO_4$ crystal like (3 1 2) and (2 1 1) cannot be observed. When the oxygen pressure increases to 10 Pa (Fig. 1 b), compared with the XRD pattern of the Fig. 1a, the peak of (2 0 0) is much stronger and sharper in addition to some weak peaks, such as (2 2 0), (4 2 0), and (4 0 0) of Nd:$LuVO_4$, indicating that grains of the film deposited in oxygen ambient pressure of 10 Pa became larger. From Fig. 1c, we can see that the (2 0 0) peak is much higher and the full width at half

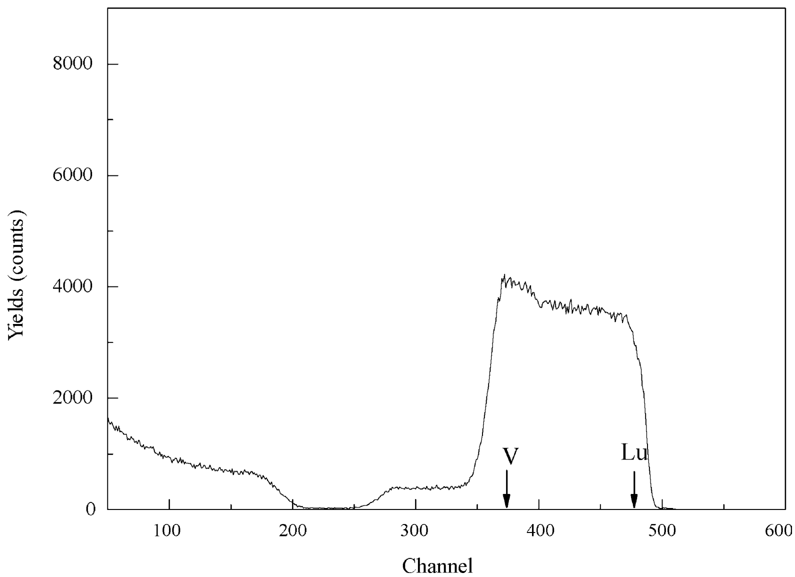

Fig. 2. Rutherford backscattering spectrum of Nd:$LuVO_4$ film grown on silica glass at the oxygen pressure of 2 Pa at 700 °C.

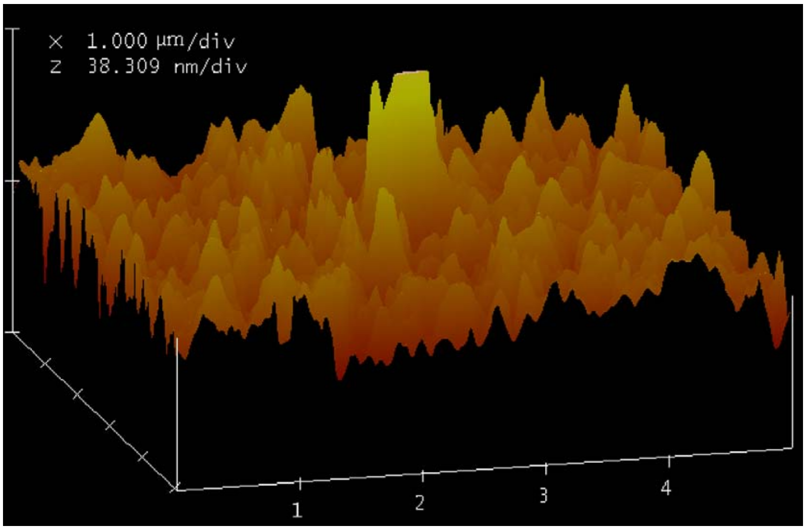

Fig. 3. AFM image for $LuVO_4$ film grown on silica glass at the oxygen pressure of 2 Pa and substrate temperature of 700 °C.

maximum (FWHM) decreases obviously in comparison with other samples, which indicates that the film fabricated at temperature of 700 °C and in oxygen ambient pressure of 20 Pa is strongly *a*-axis oriented. All the peaks revealed in Fig. 1c are compatible with $LuVO_4$ crystal. From the XRD measurement results on the film deposited in different oxygen pressures at 700 °C, we can conclude that the degree of crystal orientation increases with increasing oxygen pressure.

The stoichiometry of the Nd:$LuVO_4$ thin film was further investigated using RBS analysis. Fig. 2 shows the RBS spectrum of a film deposited at a substrate temperature of 700 °C and oxygen pressure of 2 Pa. According to Fig. 2, we can see that the layer thickness of the as-deposited Nd:$LuVO_4$ film is about 490 nm, which is in agreement with that measured by a surface profiler. The Lu/V ratio was determined to

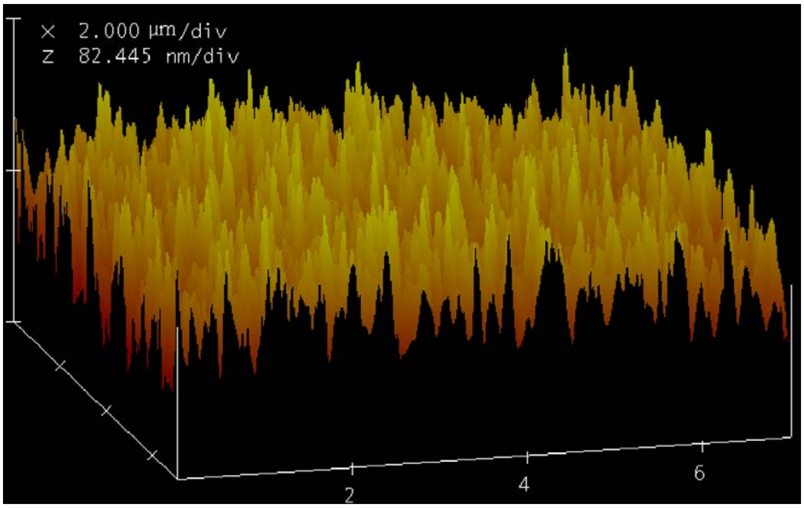

Fig. 4. AFM image for $LuVO_4$ film grown on silica glass at the oxygen pressure of 20 Pa and substrate temperature of 700 °C.

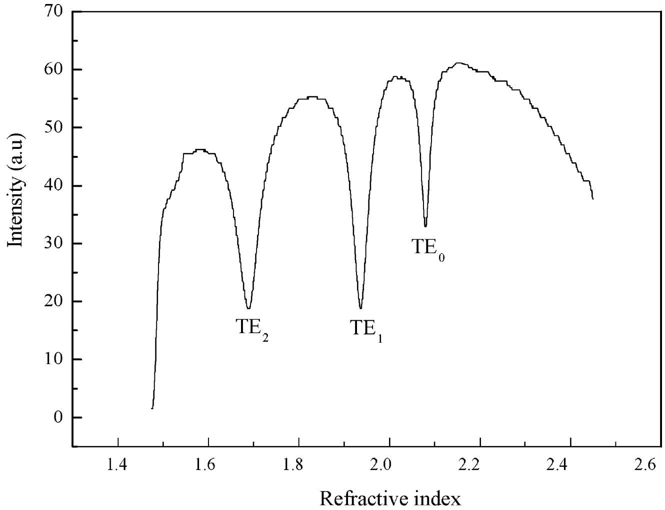

Fig. 5. Dark-line spectrum of the TE_0–TE_1 modes of Nd:$LuVO_4$ film fabricated at the oxygen pressure of 2 Pa and substrate temperature of 700 °C.

be 0.991, in accordance with the initial target composition. The oxygen concentration in the films is more difficult to determine by RBS because of the low atomic mass of oxygen.

Figs. 3 and 4 shows the surface morphology of the deposited Nd:$LuVO_4$ films observed by using an atomic force microscopy (AFM). For Fig. 3, the AFM image of the film grown on silica glass at an oxygen pressure of 2 Pa and substrate temperature of 700 °C shows a smooth surface with few droplets of particulates, which would meet the requirements for laser waveguide application. The root mean square (rms) roughness was evaluated to be 3.42 nm. This is only 0.92% of the deposited thickness (490 nm).

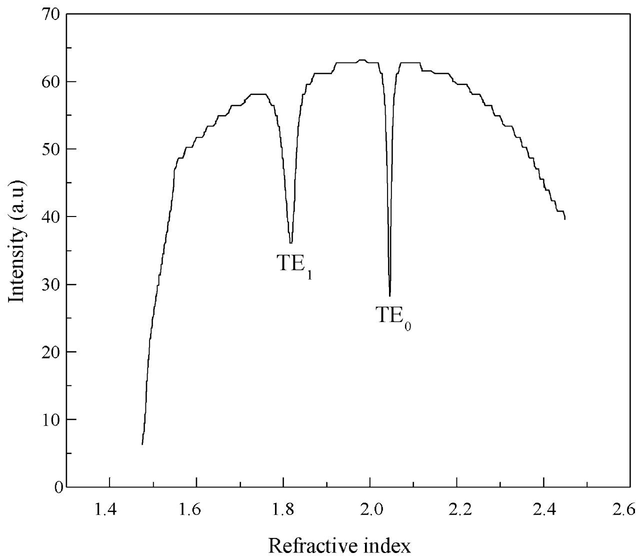

Fig. 6. Dark-line spectrum of the TE_0–TE_1 modes of Nd:$LuVO_4$ film fabricated at the oxygen pressure of 10 Pa and substrate temperature of 700 °C.

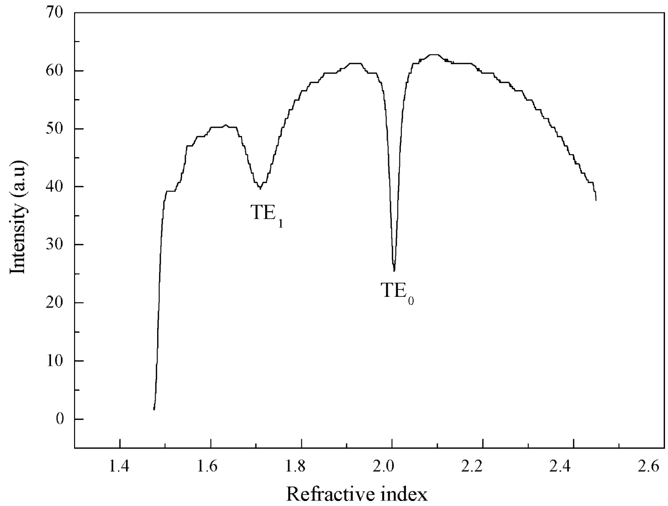

Fig. 7. Dark-line spectrum of the TE_0–TE_1 modes of Nd:$LuVO_4$ film fabricated at the oxygen pressure of 20 Pa and substrate temperature of 700 °C.

For the sample fabricated at the oxygen pressure of 20 Pa and substrate temperature of 700 °C (Fig. 4), grains became larger and denser which is consistent with the XRD results. However, the aggregation particles are not observed in these films. We can conclude that at higher oxygen pressure of 20 Pa, growth orientation becomes strong and leads to an improvement in the crystallinity of Nd:$LuVO_4$ film. This is a result of stress release due to the higher mobility of the dislocations, producing more homogeneous films.

Figs. 5–7 shows the relative intensity of the refractive light of transverse electric (TE) mode as a function of effective refractive index obtained for the Nd:$LuVO_4$ waveguide. For the films deposited in oxygen ambient pressure between 2 and 20 Pa at a substrate temperature of 700 °C, we observed three (Fig. 5), two (Fig. 6), and two (Fig. 7) TE modes for the waveguide, respectively. It should be noted that the dips become broader when the effective refractive index of the incident light decreases. Normally, a sharp dip means a good confinement of the light in the corresponding mode. The related effective mode indices are given in Table 1.The sharpness of the reflectivity dips is synonymous with a good confinement of the light into the guiding layer. The films with a preferred (2 0 0)-orientation have refractive indices lower than that of the bulk Nd:$LuVO_4$. Usually, the lower refractive index is supposed to be related to the smaller packing density of the film.

Table 1
Number of modes, effective refractive index, and refractive index for different samples

Substrate temperature (°C)	Oxygen pressure (Pa)	Mode	*M*	Effective refractive index
700	2	TE	0	2.0796
			1	1.9361
			2	1.6884
700	10	TE	0	2.0452
			1	1.8171
700	20	TE	0	2.0044
			1	1.7098

4. Conclusions

In summary, Nd-doped $LuVO_4$ waveguide films of high quality have been grown by pulsed laser deposition on the silica glass substrates. According to the results from XRD, AFM, and prism coupler measurements, we can see that the optimum oxygen pressure is 20 Pa when the substrate temperature is 700 °C. The XRD pattern shows much higher (2 0 0) peak and narrower FWHM when the oxygen pressure is increased to 20 Pa. Meanwhile, because of stress release, more homogeneous films are prepared as observed by AFM. What's more, the sharpness of the reflectivity dips is synonymous with a good confinement of the light into the guiding layer.

Acknowledgements

This work was supported by a grant of State Key program of China (No. 2004 CB 619002) and the significant research project of ministry of education of China with contract no. 03104.

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