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Growth of Al doped ZnO thin films by a synchronized two laser system

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Abstract

We report the deposition of Al doped ZnO thin films with the aid of a synchronised two laser system. The laser system consists of an ArF* excimer laser ($\lambda = 193$ nm, $\tau \sim 12$ ns) and a Nd:YAG laser ($\lambda = 355$ nm, $\tau \sim 10$ ns), for the time-matched ablation of the host (Zn) and dopant (Al) targets in oxygen atmosphere. Our approach allows for the independent and accurate setting of the laser fluences of the two lasers, in accordance with the energy requirements of the host and dopant materials. The method proposed by us permits also an in situ change of the doping conditions throughout the thin film growth process. The controlled modification of the dopant profile inside the growing film can be obtained relatively easily by the appropriate variation of the Nd:YAG laser fluence and/or number of pulses applied to the Al dopant target during the deposition process.

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Keywords: Al doped ZnO thin films; Laser deposition; Synchronized two laser system

1. Introduction

Zinc oxide (ZnO) is a wide band-gap semiconductor, with high thermal and chemical stability and good transparency in the infrared-visible spectral region [1]. Doping of ZnO with Al proved of considerable interest in the last few years. Indeed,

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besides transparency, doped ZnO is highly conductive [2]. Thus, it finds new prospective applications as transparent electrodes in solar cells, sensor devices, or flat panel displays [3].

Doping by pulsed laser deposition (PLD) is generally obtained by irradiation of targets prepared from premixed powders of the host and dopant materials. However, this procedure meets limits in the achievement of the desired dopant concentration in the deposited films, since dopant and host materials have very often different ablation thresholds. As a consequence, a selective ablation is initiated and the homogeneous uniform doping is spoiled. The gradual change of the irradiated targets composition causes also significant non-uniformities in the dopant distribution depth profiles of the films.

Solutions to overcome these impediments resorted to build an experimental set-up consisting of two targets (host and dopant) ablated either (i) alternatively by the same laser beam [4–6] or (ii) by two laser beams, split from the initial one (also known as dual-beam configuration) [7–9]. In the present work we report the deposition of doped thin films with the aid of two synchronised laser sources. The first laser ablates the host and the second one the dopant material. This approach is more flexible and versatile than those discussed previously: it allows for the independent setting of laser fluences and repetition rates requested by the host and dopant materials, and permits the in situ change of the doping conditions throughout the film growth. The dopant profile in the growing film can be tailored by the appropriate choice of the laser fluence incident on the dopant target.

2. Experimental details

The thin film deposition was performed inside a stainless steel vacuum chamber evacuated down to a residual pressure of 7×10^{-4} Pa prior to each irradiation. For the host Zn target ablation we applied the UV laser pulses generated by a Lumonics Mo. TE-861T ArF* excimer laser ($\lambda = 193$ nm, $\tau_{FWHM} \sim 12$ ns). For the ablation of the dopant Al target we used the pulses of a Quantel Mo. YG851 Nd:YAG laser ($\lambda = 355$ nm, $\tau_{FWHM} \sim 10$ ns). The two lasers were synchronized and operated at 10 Hz pulse repetition rate. The beams incidence angle was about 30° relative to the normal of the targets surfaces. To avoid fast drilling, both targets were placed on a vacuum-compatible computer controlled *XY* table. The Si(0 0 1) substrates were positioned at 40 mm from the targets, in the place where the two ablation plasmas intersect each other under an angle of 30°. The substrates were heated during the thin films growth at temperature up to 300 °C. The depositions were performed in 10 Pa oxygen pressure. The ArF* laser fluence was 4.8 J/cm^2 for 1 5000 laser pulses. The Nd:YAG laser fluence was successively 1.8, 2.6, and 3.5 J/cm^2 for 5000 laser pulses each.

The surface morphology of the deposited films was studied by atomic force microscopy (AFM) with a PicoSPM Molecular Imaging apparatus. The crystalline status was investigated by X-ray diffraction (XRD) in θ–2θ configuration with a Philips MRD diffractometer (Cu Kα, $\lambda = 1.5418$ Å radiation). The compositional depth profile was studied by Rutherford Backscattering spectrometry (RBS) using a 1.4 MeV $^4He^{2+}$ ion beam delivered by ARAMIS accelerator at CSNSM-Orsay. The backscattered ions were recorded by a Si detector, placed at 165° with respect to the beam. The recorded RBS spectra were processed by the RUMP simulation computer program [10].

3. Results and discussion

Fig. 1 shows the top view AFM image of the film deposited in 10 Pa oxygen pressure and 200 °C substrate temperature. The surface exhibits a granular structure, and larger spherical particulates. The average grains dimension is of a few tens of nanometers, while the spherical particulates average diameter reaches a few hundreds of nm. The minimum root-mean square (r.m.s.) surface roughness is quite small, of only 15 nm. We note that the presented AFM results are characteristic for all the deposited films, since no change in the nanoparticles and particulates dimension or density as a function of substrate temperature could be detected.

The X-ray diffractograms of the thin films deposited at substrate temperatures higher than 200 °C consist of one intense line at 34.5° and two weaker lines at 36.4° and 62.9° (Fig. 2). They correspond to the (0 0 2), (1 0 1), and (1 0 3) lattice plane reflections of hexagonal phase ZnO [11], respectively. These results suggest that the films are