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Influence of oxygen pressure on elastic strain and excitonic transition energy of ZnO epilayers prepared by pulsed laser deposition

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Abstract

High quality ZnO epilayers ($\chi_{min} \sim 10\%$) were prepared on Al_2O_3 (0 0 0 1) substrates at a temperature of 750 °C by pulsed laser deposition (PLD) with oxygen pressure of 0.015, 0.15, 1.5, and 15 Pa. The best crystalline quality and strongest intensity of UV photoluminescence were observed on ZnO layer with oxygen pressure of 15 Pa. It is probable due to the higher oxygen pressure lessens oxygen deficiency in the film. The tetragonal distortion e_T, which is caused by elastic strain in the epilayer, was determined by Rutherford backscattering/channeling. It reduces as a whole (from 0.93 to 0.65%) with the increase of oxygen pressure from 0.015 to 15 Pa and the excitonic transition energy simultaneously shows a weak blue shift.

Keywords: A. Thin films; B. Laser deposition; C. X-ray diffraction; D. Crystal structure; D. Optical properties

1. Introduction

ZnO, as a promising material for ultraviolet photoelectronic devices, has attracted abundant attention for these years. The homogenous light-emitting diodes (LEDs) on $ScAlMgO_4$ and Al_2O_3 substrates grown by molecular beam epitaxy (MBE) and metalorganic chemical vapour deposition (MOCVD) techniques have been reported recently [1–3]. For practical applications, a better understanding on the factors controlling the structural and photoelectrical properties of the films is demanded. Heteroepitaxial layer always suffers from residual strain induced by the mismatch of lattice constant and thermal expansion coefficient between the substrate and epilayer. It has been confirmed that the strain can affect the energy gap, effective mass, and exciton resonance energy in GaN film grown on Al_2O_3 [4]. However, previous studies on the roles of strain in ZnO films are debative. Ghosh et al. [5] and Li et al. [6] reported a red shift of excitonic resonance energy with increasing strain. But Ong et al. [7] and Gruber et al. [8] has found an opposite change. These results show that the effects of the residual strain on electrical and optical properties of ZnO epilayer still need to be further investigated based on the precise measurements of strain in ZnO films. The tetragonal

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distortion induced by the elastic strain can be accurately determined by Rutherford backscattering spectrometry (RBS) with the angular scan around an off-normal axis. RBS is a powerful technique to characterize the composition, thickness, and interface of films directly and non-destructively [9].

Pulsed laser deposition (PLD), as one of the most widely used techniques to grow high quality thin films, is particularly well suitable for growing thin films of oxide. It allows the growth of thin films in reactive oxygen pressure with the same composition as that of the target [10]. The oxygen pressure was always considered to be one of the most important parameters of PLD growth process. It sensitively affects the epitaxial quality, surface morphology and optoelectronic properties [11]. It is a general consensus that low oxygen pressure presented a high structural quality and high oxygen pressure offered a smooth surface [12]. However, the detailed relationships between oxygen pressure during growth, the strain in the epilayer, and the band gap of ZnO film have not yet been systematically studied.

In the present study, the ZnO layers were grown under various oxygen pressure ambiences by using PLD. RBS and X-ray diffraction (XRD) were used to characterize the samples. Their optical properties and the relationships with strain were studied.

2. Experiments

ZnO films were grown on Al_2O_3 (0 0 0 1) substrates by PLD, using a KrF excimer laser with wavelength of 248 nm, energy of 150 mJ/pulse, and laser repetition rate of 10 Hz. The ZnO sintered target of high-purity 99.995% was mounted on holder and placed 4 cm away from the substrate. The vacuum chamber was evacuated to a background pressure of 1×10^{-4} Pa and the substrate temperature is maintained at 750 °C. ZnO layers were grown at different oxygen pressures of 0.015, 0.15, 1.5, and 15 Pa for 10 min, with serial number A, B, C, and D, respectively. Finally, all the samples annealed in situ in 2000 Pa oxygen ambience at 750 °C for 30 min.

A collimated 2.0 MeV He^+ beam produced by a 5SDH-2 Pelletron was used for the RBS/channeling measurements to determine the crystalline quality of ZnO layers. The backscattered particles were detected at 165° with respect to the beam direction using an Au–Si barrier detector. The energy resolution was 15 keV. X-ray diffraction measurements were carried out by using a Bruker D8 diffractometer system, using four Ge (0 2 2) as the monochromator. The X-ray wavelength was Cu Kα1 radiation at 1.5406 Å. The optical properties of films were characterized by photoluminescence (PL) spectroscopy using the 325 nm line of a He-Cd laser at room temperature.

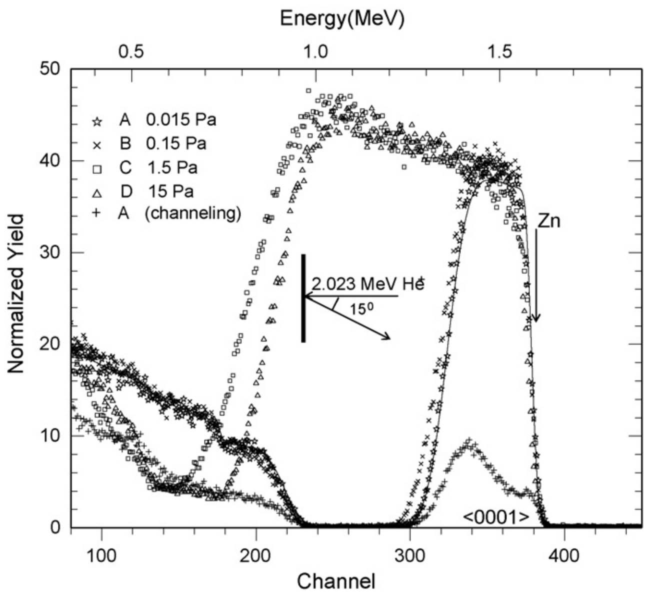

Fig. 1. Random RBS spectra of ZnO films grown at various oxygen pressures. Simulated (solid line) and aligned RBS spectra for ZnO grown under 0.015 Pa are plotted for comparison.