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Improvement in crystal quality of ZnO film on Si substrate by using a homo-buffer layer

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ABSTRACT

ZnO thin films without and with a homo-buffer layer have been prepared on Si(1 1 1) substrates by pulsed laser deposition (PLD) under various conditions. Photoluminescence (PL) measurement indicates that the optical quality of ZnO thin film is dramatically improved by introducing oxygen into the growth chamber. The sample deposited at 60 Pa possesses the best optical properties among the oxygen pressure range studied. X-ray diffraction (XRD) results show that the films directly deposited on Si are of polycrystalline ZnO structures. A low-temperature (500 °C) deposited ZnO buffer layer was used to enhance the crystal quality of the ZnO film. Compared to the film without the buffer layer, the film with the buffer layer exhibits aligned spotty reflection high-energy electron diffraction (RHEED) pattern and stronger near-band-edge emission (NBE) with a smaller full-width at half-maximum (FWHM) of 98 meV. The structural properties of ZnO buffer layers grown at different temperatures were investigated by RHEED patterns. It is suggested that the present characteristics of the ZnO epilayer may be raised further by elevating the growth temperature of buffer layer to 600 °C.

1. Introduction

ZnO is a versatile II–VI compound semiconductor with wurtzite structure, which has been widely applied in gas sensors [1], transparent conducting electrodes [2], surface acoustic wave (SAW) devices [3], and ultraviolet (UV) optical detectors [4]. Because of its wide direct bandgap of 3.37 eV and large exciton binding energy of 60 meV at room temperature (RT), ZnO is considered as one of the most promising candidate material for light emitting diodes (LEDs) and laser diodes (LDs) in the UV region [5,6]. Recently, ZnO-based homojunction LEDs have been fabricated successfully [7,8]. However, the light emitting efficiency is very limited due to the poor quality of the *p*-type layer. So it is a challenge to obtain high-quality ZnO films in order to increase the light emitting efficiency of ZnO-based optical devices. Among many growth techniques for ZnO film, pulsed laser deposition (PLD) has several fundamental advantages. First, the kinetic energy of source particles is high enough to provide the required surface mobility at a low substrate temperature [9]. Second, introduction of oxygen enable it feasible to minimize the number of oxygen vacancies. Third, ablation of the target by pulsed laser beam ensures the chemical purity of deposited films. Finally, atomic-layer control can be realized by adjusting the laser energy density and pulse repetition rate.

In most studies, sapphire (α-Al_2O_3) has been adopted as substrate for ZnO growth [10–12]. However, sapphire is electrically insulating and difficult to cleave, which causes the complexity of subsequent device integration. All these problems can be conquered by using a Si substrate.

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Moreover, from the viewpoint of combination with the mature Si microelectronics technology, research on ZnO/Si heteroepitaxy is a valuable task. In this paper, ZnO films without and with homo-buffer layers have been synthesized on Si substrates by PLD technique. The structural and optical properties of the ZnO samples and buffer layers are investigated to achieve ZnO films with a quality suitable for optoelectronic device applications.

2. Experimental procedure

ZnO films were deposited on Si(1 1 1) substrates by ablating a high-purity (99.99%) ZnO ceramic target using a KrF excimer laser (248 nm, 5 Hz). The substrate was ultrasonically degreased with methylbenzene, acetone, ethanol, and deionized water in turn, then etched in 10% HF solution for 5 min and rinsed in deionized water again. After being loaded into the growth chamber, the substrate was thermally cleaned at 500 °C for 10 min with a base pressure of 4×10^{-5} Pa to remove residual contamination. During film growth, the laser fluence was around 2.5 J/cm^2. The substrate susceptor was 5 cm away from the target and was rotated at a rate of 80 rpm to acquire uniform films. In this experiment, three series of samples were prepared to enhance the crystal quality of ZnO films step by step. All the procedure and conditions mentioned above were identical for every sample growth. Firstly, at an optimized substrate temperature of 650 °C [13], a series of samples were deposited directly on Si under oxygen pressures varying from 10 to 60 Pa to study the effect of oxygen pressure on the film properties. Secondly, ZnO films without and with a homo-buffer layer were deposited at an optimized oxygen pressure of 60 Pa. The buffer layer was synthesized at a low temperature of 500 °C in vacuum. Finally, in order to characterize their structural properties, ZnO buffer layers were grown at different temperatures of 500, 550, and 600 °C. The buffer growth was carried out in vacuum with a pressure superior to 2.0×10^{-3} Pa.

The film thickness was measured by a α-step profilometer. X-ray diffraction (XRD, λ=1.5406 Å) and *in situ* reflection high-energy electron diffraction (RHEED) were employed to evaluate the crystalline quality of the samples. The surface morphology was observed by scanning electron microscopy (SEM, JEOL JSM-6360). Photoluminescence (PL) spectra of the ZnO films were taken at RT using a He–Cd laser (325 nm, 30 mW) as excitation source.

3. Results and discussion

Fig. 1 shows XRD spectrum of the ZnO thin film deposited at 10 Pa. All the samples grown in different oxygen pressures possess similar structural properties. Apart from the (0 0 2) and (0 0 4) diffraction peaks of ZnO, a small (1 0 3) peak can also be detected, indicating a polycrystalline ZnO structure with a preferred *c*-axis orientation. This is because the lowest surface free energy of ZnO (0 0 *l*) plane makes the growth along *c*-axis easy, although the Si surface has been oxidized by ambient oxygen during the initial nucleation stage [14,15].

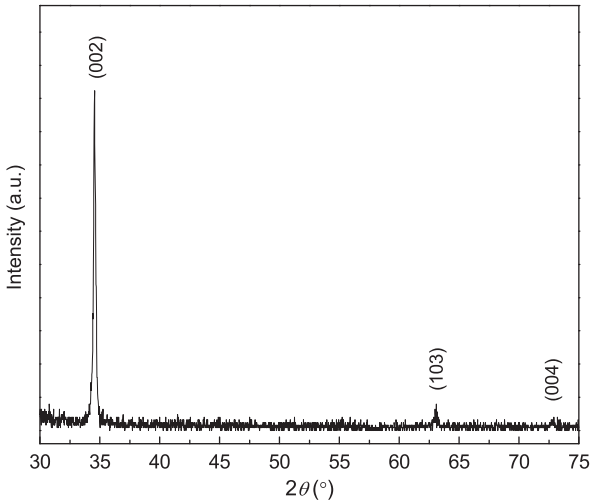

Fig. 1. XRD spectrum of the ZnO thin film deposited at 10 Pa.

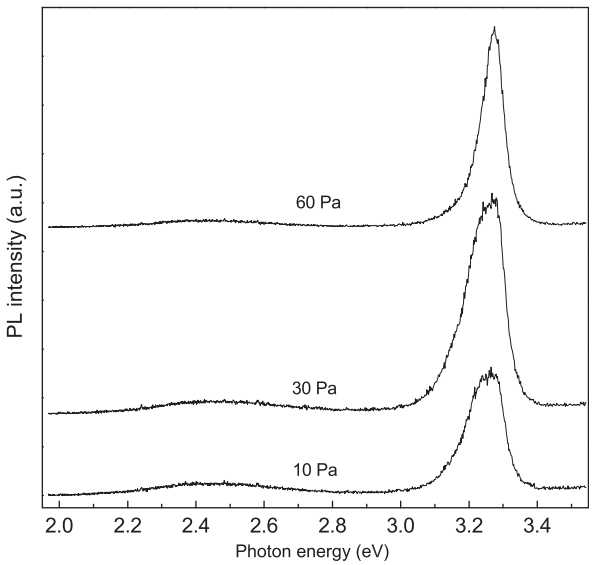

Fig. 2. RT PL spectra of ZnO thin films prepared at different oxygen pressures.

Fig. 2 presents the PL spectra of the ZnO thin films prepared at different oxygen pressures. All the spectra are composed of strong near-band-edge emission (NBE) and weak deep-level emission (DLE). With an increase in oxygen pressure from 10 to 30 Pa, the NBE intensity increases drastically and the DLE intensity decreases gradually. The PL intensity ratios of NBE to DLE for the thin films deposited at 10, 30, and 60 Pa are 10, 17, and 25, respectively. The biggest intensity ratio and the smallest FWHM of NBE peak indicate that the sample deposited at 60 Pa has better optical quality than the other ones.

The experimental results mentioned above indicate that introduction of oxygen in PLD system is a guarantee for high luminescent properties of ZnO films. However,