Effect of ambient oxygen pressure on structural, optical and electrical properties of SnO_2 thin films

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Abstract: Polycrystalline SnO_2 thin films were deposited on sapphire substrates at 450°C under different ambient oxygen pressures by pulsed laser deposition technique. The effect of ambient oxygen pressure on the structural, optical and electrical properties of SnO_2 thin films was studied. X-ray diffraction and Hall measurements show that increasing the ambient oxygen pressure can improve crystallization of the films and decrease resistivity of the films. A violet emission peak centered at 409 nm was observed from photoluminescence measurements for SnO_2 films at deposition ambient oxygen pressure above 5 Pa, which is related to the improvement of the crystalline of the films.

Key words: tin oxide; effect of oxygen pressure; PLD; resistivity

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1. Introduction

Tin oxide (SnO_2) is an *n*-type semiconductor with band gap of $E_g = 3.62-4.00$ eV at room temperature [1]. Over the past few decades, SnO₂ has been widely used in a broad range of important applications, including solid-state gas sensors [2-3], liquid crystal displays, thin-film heaters, photovoltaic cells and transparent conducting electrodes [4-7]. The SnO_2 research for its applications has been mainly focused on thin films. Up to date, SnO₂ thin films have been achieved by a variety of deposition techniques such as chemical vapor deposition (CVD) [8], sol-gel processing [9], reactive sputtering [10] and pulsed laser deposition (PLD) [11-13]. Among these fabrication techniques, PLD has attracted much attention. It is well known that ambient oxygen pressure is one of the key experimental parameters in the process of PLD. In this paper, SnO₂ thin films were prepared by PLD and the influence of ambient oxygen pressure (from 1 to 20 Pa) on structural, electrical and optical properties of SnO₂ thin films were reported.

2. Experimental

A KrF excimer laser ($\lambda = 248$ nm), operating at a repetition rate of 4 Hz with an energy density of about 2 J·cm⁻², was used to grow SnO₂ thin films on sapphire substrates from a polycrystalline SnO₂ target. The distance between target and substrate was kept about 5 cm, and the substrate temperature was 450°C. SnO₂ thin films were deposited under different oxygen pressures, i.e. 1, 5, 10 and 20 Pa.

The crystalline structure of SnO₂ thin films was measured by X-ray diffraction (XRD) using Cu K_{α} radiation. Photoluminescence emission measurements were performed using a 325 nm He-Cd laser excitation. All spectra were taken at room temperature using quartz lens focused into the slit of a spectrometer in air. The transport properties of the samples were determined by Hall effect measurement.

3. Results and discussion

Fig. 1 shows XRD patterns of SnO₂ films deposited at 450°C under different oxygen pressures of 1, 5, 10 and 20 Pa, respectively. It is clear that all the films are composed of SnO and SnO₂ phases. The relative content of SnO phase decreases with the oxygen pressure. For the SnO₂ phase, as the oxygen pressure is increased, the diffraction peak becomes stronger and narrower. In particular, the intensity ratio between SnO₂ (200) and SnO₂ (110) peaks increases significantly with oxygen pressure. These results indicate that the higher oxygen pressure can improve both crystallization and *a*-axis orientation of SnO₂ phase.



Fig. 1. XRD spectra of SnO₂ films on sapphire substrates at different background oxygen pressures.

Fig. 2 is photoluminescence spectra of the SnO₂ films deposited at 450°C under different oxygen pressures. A peak centered at 409 nm (3.095 eV) is observed for all the deposited films, and the intensity of this peak increases with oxygen pressure. Jin et al. [14] suggested that the peak should be attributed to electron transition from donor levels causing by oxygen vacancies to acceptor levels, while Kim et al. [15] and Jeong et al. [16] thought that this peak is related to structural defects. In our case, the peak at 3.1 eV could be caused by the improvement of the crystalline of the films. Though films fabricated under low oxygen pressure own more structural defects such as oxygen vacancies, these defects may serve as non-irradiative centers and cause the "fluorescence quenching". As the oxygen pressure rising, the enhancement of crystalline of the films will restrain the non-irradiative transitions and bring the fact of both the crystalline of SnO2 films and the intensity of the peak at 3.1 eV increasing with the oxygen pressure.



Fig. 2. Photoluminescence spectra of SnO₂ films deposited at 450°C under different oxygen ambient pressures.

Fig. 3 presents the transmittance spectra of transmittance of SnO_2 thin films deposited under different oxygen pressure. The mean transmittance (%) is large than 85% in the visible range (380-780 nm) for the films fabricated at 10 and 20 Pa, which is normalized to that of the bare substrates. It is obvious that the mean transmittance increases with the rising of the oxygen pressure. The different transmittance behavior of the film deposited at 1 Pa from others consists with the crystal difference of them.



Fig. 3. Transmittance of SnO_2 thin films as a function of oxygen pressures.

From Fig. 4, it can be found that the resistivity, carrier concentration, and carrier mobility are dependent on oxygen pressure. The resistivity of the films was decreased from 6.4 to 0.09 Ω -cm with

oxygen pressure increased from 1 to 20 Pa. The Hall effect measurement showed that the carrier concentration and the mobility of the films are $(0.224-14.770) \times 10^{18} \text{ cm}^{-3}$ and 1.0-6.0 cm²·V⁻¹·s⁻¹, respectively. When the oxygen pressure is below 1 Pa, the existence of amorphous SnO₂ phase greatly reduces the carrier density and results in the excess of the resistivity. While the oxygen pressure is above 1 Pa, the significantly increased crystallite size diminishes the grain boundary scattering effect of carriers and yet induces the increase of carrier mobility and conductivity of the film.



Fig. 4. Resistivity, carrier concentration, and carrier mobility of SnO_2 thin films as a function of oxygen pressures.

4. Conclusions

The electrical and optical properties of polycrystalline tin oxide (SnO₂) thin films by PLD have been investigated as a function of oxygen pressure. XRD results show that crystallization of thin film is improved with the increase of ambient oxygen pressure. The photoluminescence spectra indicate a violet emission peak centered at 409 nm. Furthermore, the resistivity of SnO₂ films is decreased significantly from 6 Ω ·cm to 0.09 Ω ·cm when oxygen pressure is increased from 1 to 20 Pa.

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