



Effect of annealing temperature on PL spectrum and surface morphology of zinc oxide thin films

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ABSTRACT

Zinc oxide (ZnO) thin films were produced by thermal oxidation of Zn layers (200 nm thickness) which were coated on Si (1 0 0) substrate by DC magnetron sputtering. In order to study the effect of annealing temperature on photoluminescence (PL) properties and the surface morphology of the ZnO samples, the annealing temperature range of 500–700 °C was employed. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) for investigation of surface morphology of the ZnO samples were carried out. The surface statistical characteristics of these ZnO thin films are then evaluated against data which outcome from AFM. SEM and AFM results indicated that the annealing temperature produces larger grains and rough surfaces at higher temperatures. The results of PL spectra represent an increase in interstitial zinc with increasing annealing temperature.

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1. Introduction

Transparent conducting oxides (TCO) have been recently investigated for their interesting optical, mechanical and electrical performance [1,2]. The lasting recorded high optical transparency in the visible domain, and low electrical resistivity led to numerous applications of these materials in the new generation of optoelectronic devices [3–5]. Zinc oxide (ZnO) represents, in this context, an important basic material for the construction of nanoscale structures. Recently, zinc oxide has attracted much attention within the scientific community as a 'future material'. It has a direct band gap of 3.37 eV at room temperature and a high exciton binding energy of 60 meV [5], which is much larger than the thermal energy at room temperature and belongs to a member of hexagonal wurtzite class. Some of the optoelectronic applications of ZnO overlap with GaN ($E_g = 3.4$ eV at 300 K), which is widely used for the production of green, blue, ultraviolet and white light-emitting devices [6–9]. However, ZnO has some advantages over GaN, among which are the availability of fairly high-quality ZnO bulk single crystals and a large exciton binding energy and the ability to grown single crystal substrate. These interesting properties of ZnO thin films have been for use in many applications such as room temperature UV

laser and short-wavelength optoelectronic device [10]. Furthermore, ZnO can be use in the photo-conductors [11], gas sensors [12], and solar cell applications [13]. The highly preferential orientation a long c-axis and wide band gap energy of films are useful characteristic in optical wave-guides [14], surface acoustic wave [15] and acousto-optic device [16]. The surface, which is the first interface of the material, has an important role in the interaction between the material and the environment [17]. Surface roughness has an enormous influence on many important physical phenomena such as mechanical contact, sealing, adhesion, wave scattering and friction [18]. Most surfaces in nature are rough, and this fact is a motivation that can be studied as a random process. In fact, a surface for a special application requires specified statistical properties.

ZnO thin films have been synthesized by a variety of processes, including physical vapor deposition (PVD), chemical vapor deposition (CVD), pulsed laser deposition (PLD), molecular beam epitaxy (MBE), sputtering, sol-gel processing, etc. [19–24]. In this work we used DC magnetron sputtering for deposition of pure zinc, and method of metallic Zn thermal oxidation was employed to produce ZnO thin films. High packing density, uniform film thickness, suitable coating rate, very good adhesion of the film to substrate due to high energy of deposition particles are some of magnetron sputtering advantages. In this work the effect of the annealing temperature on the photoluminescence (PL) spectrum and the surface morphology of the ZnO layers deposited on the Si (1 0 0) substrate were investigated. To investigate surface morphology of these ZnO thin

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films, atomic force microscopy (AFM) (Park Scientific Instrument Auto Probe model CP) was employed using force constant mode. In this paper, we study the annealing process as a stochastic process. Surface roughness, roughness exponents, correlation length have been measured.

2. Experimental details

Sputtering of the layers of Zn at room temperature (RT) on the silicon substrate were performed in Ar ambient (purity 99.999%) and a 99.9% pure metal zinc target with 12.5 cm diameter and 3 mm thickness was used for sputtering. A vacuum system (Hind High Vacuum, H.H.V, 12"MSPT) with base pressure of 10^{-6} mbar was used. Si (1 0 0), *n* type wafer was cut to required dimensions (1 cm × 1 cm) and 1 mm thickness and were used as substrate for deposition of Zn thin films. Before deposition, silicon substrates were cleaned in the heated acetone ultrasonic bath for 2–3 min. A shutter was placed between the zinc target and silicon substrate for two reasons, firstly to control period of coating, secondly the shutter was removed only when the line spectra belonging to Zn, Ar (atoms, ions) was checked, and no band spectra due to contamination was detected. After many runs of discharge and sputtering, the optimum distance of 12 cm between Zn target and silicon substrate was found and this distance during this work was kept constant. The thickness of these samples was about 200 nm which were measured using a digital vibrating quartz crystal thickness monitor. Layers of zinc after thermal oxidation in air ambient at 400 °C were employed to produce ZnO thin films. In order to study the effect of annealing temperature, the samples were heated in a quartz tube furnace at various temperatures. The samples were held at each temperature for 10 min and then furnace was cooled to room temperature. The rate of the cooling was slow enough to avoid the possibility of any type of stress and strain in the thin films.

PL spectra were taken at room temperature under 340 nm xenon lamps (with Fabry Perot filter) as the excitation source (Stellnet model Epp-200). To investigate the surface morphology of these ZnO thin films scanning electron microscopy (SEM) (Hitachi S-4160), and AFM (Park Scientific Instrument Auto Probe model CP) were also performed using the force constant mode and digitized into 256×256 pixels. A commercial standard pyramidal Si₃N₄ tip was used. All AFM images were acquired in ambient air. For a better comparison of the effects of different interfaces, we kept all other experimental parameters unchanged. To analyses the AFM images, the topographic image data were converted into ASCII data. AFM images of samples indicated changes in surface behavior of the film.

3. Statistical analysis

A random rough surface can be described mathematically as $h(i, j)$, where h is the height of the surface relative to the reference level, that reference level defined by a mean surface height and $r(i, j)$ is the position on the surface. We assume that the distance between two adjacent discrete positions is d , and the number of surface points is N . The average surface height is the arithmetic average of surface heights. Analytically it can be expressed, for a digitized surface, as:

$$\langle h \rangle_N = \frac{1}{N} \sum_{i=1}^N h(i, j) \quad (1)$$

Root mean square (RMS) roughness is one of the most important parameters for characterizing a rough surface. Analytically, it can be estimated as:

$$\langle w \rangle_N = \left(\frac{1}{N} \sum_{i=1}^N (h(i, j) - \langle h \rangle_N)^2 \right)^{1/2} \quad (2)$$

The height–height correlation function can be expressed as [25]:

$$\langle H(md) \rangle_N = \frac{1}{N-m} \sum_{i=1}^{N-m} (h(i+m, j) - h(i, j))^2$$

$$m = 0, 1, 2, 3 \dots N-1 \quad (3)$$

For self-affine surfaces, the dynamic scaling hypothesis suggests that the height–height correlation function $H(r)$ has the scaling form of:

$$H(r) = \begin{cases} (\rho r)^{2\alpha} & \text{for } r \ll l_0 \\ 2w^2 & \text{for } r \gg l_0 \end{cases} \quad (4)$$

where $\rho = w^{1/\alpha}/l_0$ is the local slope, and α is called the roughness exponent ($0 < \alpha < 1$) which describes how wiggly the surface is. The quantity l_0 is the lateral correlation length, within which the surface heights of any two points are correlated. The roughness exponent α is related to the fractal dimension D_f of the random surface by $D_f = E + 1 - \alpha$ with $0 < \alpha < 1$, where $E + 1$ is the dimension of the embedded space ($E = 1$ for a profile; $E = 2$ for a plan). A larger value of α corresponds to a locally smooth surface structure while a smaller value of α corresponds to a more jagged local surface morphology [25,26].

The height distribution function provides a complete specification of the random variable $h(r)$ at the position r . Although different rough surfaces may have different height distributions, the most generally used height distribution is the Gaussian height distribution. The statistical analysis of AFM data was done using the height distribution histograms. The height asymmetry is described by the statistical parameters such as surface skewness and kurtosis. Unlike RMS roughness, skewness is dimensional. Skewness is a measure of the symmetry of a distribution about a mean surface level. Kurtosis is also a dimensionless quantity. It is a measure of the sharpness of the height distribution function. These two parameters represent the shape of the surface height distribution and can be estimated as [25]:

$$R_{sk} = \frac{1}{N \langle w \rangle_N^3} \sum_{i,j=1}^N (h(i, j) - \langle h \rangle_N)^3 \quad (5)$$

$$R_{ku} = \frac{1}{N \langle h \rangle_N^4} \sum_{i,j=1}^N (h(i, j) - \langle h \rangle_N)^4 \quad (6)$$

For comparison of these parameters for all samples, we used central limit theorem. This theorem mean of a sufficiently large number of independent random variables, each with finite mean and variance, will approximately have a normal distribution [27].

4. Result and discussion

4.1. Photoluminescence properties

In order to study the effects of annealing temperature on PL spectra and the surface morphology of the ZnO thin films, three samples were deposited under similar conditions (Ar pressure, substrate temperature, oxidation temperature, period of heating and deposition rate). All of them were heated at 400 °C for oxidation of zinc film then they were annealed at different temperatures (500, 600 and 700 °C), and they are named as b_1 , b_2 and b_3 respectively. It is well known that the defects in ZnO thin films which may occur include oxygen vacancy (V_O), zinc vacancy (V_{Zn}), interstitial zinc (Zn_i), interstitial oxygen (O_i) and anti-site oxygen (O_{Zn}) depend upon the method of coating and important parameters such as temperature of annealing, deposition rate, atmosphere of heating, etc.

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