



ZnO:Ag film growth on Si substrate with ZnO buffer layer by rf sputtering

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ABSTRACT

ZnO buffer layers were deposited on *n*-Si (1 0 0) substrate by rf magnetron sputtering at a lower power of 40 W. Then Ag-doped ZnO (SZO) films were deposited on buffered and non-buffered Si at a higher sputtering power of 100 W. The effects of buffer layer on the structural, electrical and optical properties of SZO films were investigated. The three-dimensional island growth process of ZnO buffer layer was discussed. The energy band diagram of *p*-SZO/*n*-Si heterojunction was constructed based on Anderson's model. Results show the ZnO buffer layer leads to better properties of SZO film, including larger grain size, smoother surface, higher carrier mobility, better rectifying behavior, lower interface state density, and weaker deep-level emission. It is because the ZnO buffer layer effectively relaxes the partial stress induced by the large lattice mismatch between SZO and Si.

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1. Introduction

ZnO is a wide bandgap (3.37 eV at 300 K) semiconductor material of interest for short wavelength optoelectronic devices and thin film transistors. However, the difficulty of *p*-type doping inhibits applications to the devices [1,2]. *p*-Type SZO film attracted much attention very recently [3], because first-principles calculations indicated the formation energy for Ag_{Zn} is lower than that for Ag_{Si}, which may alleviate the self-compensation issue [4–6]. Moreover, strong UV emissions from SZO films were reported as a valuable characteristic for short wavelength light-emitting applications [7–10]. These reports show SZO is a promising *p*-type material.

From the viewpoint of potential integration of ZnO-based optoelectronic devices with the well-developed Si technologies, Si has been widely used as a substrate material for pure or doped ZnO films [11–16]. Furthermore, Si substrate has advantage of lower cost. However, the growth of high-quality ZnO films directly on Si substrates is difficult because of the large lattice mismatch between Si and ZnO. Using buffer layer is a good method to improve the quality of ZnO films on Si. Some groups used the homogeneous buffer layer [17], and some others used the heterogeneous buffer layers such as CaF₂ [18], MgO [19], SiC [20], or Ti [21]. SZO films have been grown on Si substrates by many groups [22–25]. How-

ever, the related buffer layers were less studied. Lu et al. and Huang et al. reported the quality of ZnO film deposited by sputtering was increased at a lower sputtering power, in spite of the film growth rate was slow [26,27]. It implies a potential homo-buffer layer preparation method.

In this study, SZO films were deposited on ZnO-buffered and non-buffered Si substrates using rf magnetron sputtering. The ZnO buffer layers were deposited beforehand at a lower sputtering power. The properties of SZO films without and with buffer layer were investigated.

2. Experimental details

ZnO buffer layers and SZO films were deposited in turn by rf magnetron sputtering. The Si substrates were *n*-Si (1 0 0) with resistivity of 80–120 Ωcm. The substrate-to-target distances were 8 cm. The sputtering target for ZnO buffer layers was a pure ceramic ZnO (99.99% purity) disk. For SZO films, a disk of ceramic ZnO target dispersed with silver dots of ~0.1% sputtering area was used. Ar and O₂ with the volume ratio of 1:1 were used as the working gas at a total pressure of 2.7 Pa. In the first step, ZnO buffer layers of about 200 nm were deposited on *n*-type Si (1 0 0) substrates at the sputtering power of 40 W. Then the buffered Si substrates were annealed at 600 °C in air for 1 h. Next, SZO films of about 1000 nm were deposited on buffered and non-buffered Si substrates. The sputtering power was 100 W. The concentration of Ag in SZO film was about 0.8 atom% determined by X-ray photoelectron spec-

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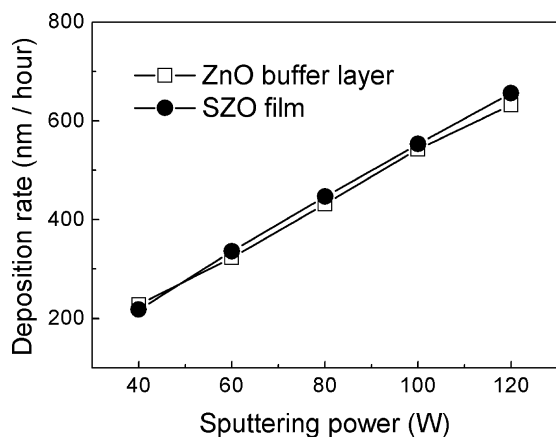


Fig. 1. Deposition rate versus sputtering power for SZO film and ZnO buffer layer.

troscopy. All the samples were annealed at 600 °C in air for 1 h after deposition.

The film thickness was measured by scanning electron microscopy (SEM). The crystal structures of samples were measured using X-ray diffraction (XRD). The surface morphologies were examined by atomic force microscopy (AFM). The elemental distributions were obtained by performing secondary ion mass spectroscopy (SIMS). The electrical properties were measured by a Hall measuring system using the Van Der Pauw technique. The optical properties were measured by room temperature (PL) using a fluorescence spectrometer with the excitation wavelength of 210 nm.

3. Results and discussion

Fig. 1 shows the deposition rate versus sputtering power for SZO film and ZnO buffer layer. The two curves are similar because the silver content in the target for SZO sputtering is very small. The deposition rate is almost directly proportional to the sputtering power in each curve. Moreover, the deposition rates of ZnO buffer layer sputtered at 40 W and SZO film sputtered at 100 W are about 230 and 550 nm/h, respectively.

The crystal structures of samples were measured by XRD. A ZnO thin film was deposited at 100 W for comparing with the ZnO buffer layer deposited at 40 W. The ZnO thin film and the ZnO buffer layer have the same thickness of about 200 nm. Fig. 2 shows XRD patterns of samples. Only ZnO (002) peak is observed in each sample, indicating the highly *c*-axis orientation. It is due to the lowest sur-

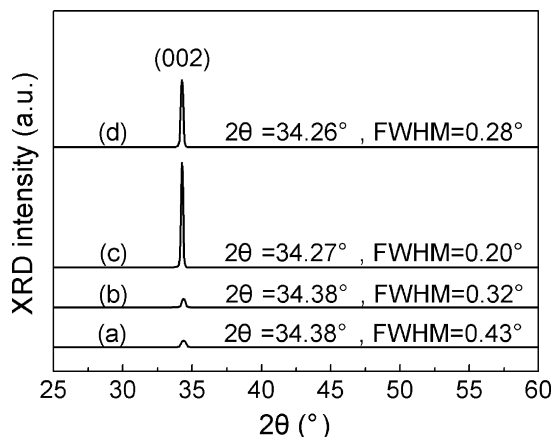


Fig. 2. XRD patterns of samples: (a) ZnO thin film deposited at 100 W, (b) ZnO buffer layer deposited at 40 W, (c) SZO film on buffer layer, and (d) SZO film on bare Si.

face energy of the (001) basal plane in ZnO [28]. Fig. 2(a) and (b) indicates the full-width at half-maximum (FWHM) values of (002) peaks of the ZnO film and the ZnO buffer layer are 0.43° and 0.32°, respectively. It confirms the lower growth rate caused the better crystalline quality of ZnO buffer layer. Fig. 2(c) and (d) indicates the FWHM values of (002) peaks of the SZO films on bare Si and ZnO buffer layer are 0.28° and 0.20°, respectively. It shows the crystalline quality of SZO film was improved by using the ZnO buffer layer. The grain size of films can be calculated by the following Scherrer's equation,

$$D = \frac{0.9\lambda}{\beta \cos \theta_0} \quad (1)$$

where D is the grain size in the thin film, λ (0.15406 nm) is the wavelength of the X-ray, β is the FWHM of XRD peak, and θ_0 is the diffraction angle of the peak. So the grain size values of SZO films on bare Si and buffer layer are calculated to be 34 and 48 nm, respectively. The SZO film on buffer layer has larger grain size. Moreover, the 2θ angle positions of (002) peak of SZO on bare Si and ZnO buffer layer are located at 34.26° and 34.27°, respectively. It is known the normal ZnO (002) peak is located at about 34.4°. The angle shift is caused by the increase of lattice constant due to the substitution of Zn^{2+} ions by Ag^+ ions [29].

Surface morphologies of samples were investigated by AFM. Fig. 3 (a) and (b) shows pictures of SZO films on Si substrate and ZnO buffer layer, respectively. The particle grains of SZO film on buffer layer are larger and more uniform than those of SZO film on Si. Compared these pictures with the results calculated by Scherrer's equation, it is noteworthy that the characteristic sizes obtained from AFM and XRD analysis are in correlation. Root mean square (RMS) roughness was also calculated from AFM. The RMS values of SZO films on Si and buffer layer are 42 and 28 nm, respectively. The surface of the SZO film on buffer layer is smoother. Fig. 3 (c) and (d) shows pictures of ZnO buffer layers. It indicates the grain size of ZnO buffer layer became larger after annealing. It is caused by the interface merging processes [30].

Compared Fig. 3(a) with (c), it is obvious that the grain structure of ZnO buffer layer is more uniform than that of SZO film on bare Si. We consider it is attributed to the change of three-dimensional island growth related to the deposition rate. At the beginning of growth process, crystals were slowly nucleated because the substrate is high quality single crystal Si. With a higher growth rate, the initial SZO crystals were enlarged quickly before the nuclei were uniformly formed. So the later nucleated crystals were much smaller. On the other hand, ZnO buffer layer deposited at a lower sputtering power had enough nucleation time. So its uniformity is better. The uniformity of ZnO buffer layer was maintained after annealing. SZO film on the ZnO buffer layer continued the arrangement [31]. Fig. 3(b) shows the SZO film on buffer layer remains the finer uniformity.

Hall effect and resistivity measurements were carried out on the SZO films with van der Pauw configuration. Silver electrodes were deposited on the surfaces of SZO films. The Ohmic contact between silver and SZO has been proved. The SZO films on buffered and non-buffered Si are both *p*-type, as shown in Table 1. We also confirmed the conductivity type of SZO by depositing another SZO film on a glass substrate under the same conditions. In this experiment, the effects of Si substrates are inevitable although the Si substrates are *n*-type and have a higher resistivity. So the values in Table 1 are not precise but could be used for a rough comparison. The SZO film on buffer layer shows a lower carrier concentration. It may be attributed to the reduction of lattice mismatch between SZO and Si due to buffer layer. The lattice constant (for the "*a*" parameter) of wurtzite ZnO is smaller than that of Si (100). So there is lattice enlarged deformation in the SZO film on Si. The deformation causes tensile strains. These tensile strains can be relaxed by sub-

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