



Microstructure evolution and optical properties of *c*-axis-oriented ZnO thin films incorporated with silver nanoisland layers

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Abstract

Highly *c*-axis-oriented ZnO thin films incorporated with various numbers of Ag nanoisland layers were fabricated in this study by alternating sputtering-deposited ZnO and Ag layers on Si substrates. Two such ZnO–Ag layered structures were fabricated: the first by using a 150-nm ZnO film–Ag island layer/Si, and the second by using three applications of a 50-nm ZnO film–Ag island layer/Si. The crystallographic features of the as-deposited ZnO thin film on the Si substrate exhibited a pure *c*-axis orientation. However, the degree of the *c*-axis texture of the ZnO thin films slightly decreased after inserting Ag island layers. The ZnO thin-film surface became quite rough because the surface morphology was transformed from being dense and flat to exhibit a loosely columnar grain structure. Transmittance electron microscopy (TEM) images showed that the Ag islands were clearly distributed on the ZnO/substrate and ZnO/ZnO interfaces. The photoluminescence measurement results indicated that the near-band-edge (NBE) emission peak intensity was enhanced by nearly two orders of magnitude in the 150-nm ZnO film–Ag island layer/Si compared with that of the as-deposited 150-nm ZnO thin film without an inserted Ag island layer. Inserting additional Ag island layers did not increase the NBE emission intensity; instead, the visible emission band intensity was markedly increased. This was attributed to the substantial change in the ZnO thin film microstructure caused by inserting the Ag island layers.

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1. Introduction

ZnO is a promising semiconductor for various optoelectronic and optical applications because of the following properties: a wide band gap and a high exciton binding energy (60 meV) at room temperature [1]. The excellent thin-film structure of ZnO is essential for practical device applications.

Although various physical and chemical methodologies have been proposed to prepare ZnO thin films that demonstrate desired electrical and optical properties [2–5], vacuum thin-film processes, such as sputtering, are considered the most favorable deposition methods for obtaining highly uniform films because they enable precise control of thickness, actualize large-area deposition, facilitate the doping process, and produce multilayer structures [6–8]. Sputtering is widely used in processing

semiconductor devices. This vacuum thin film process realizes the integration of ZnO thin-film processes into Si-based devices. Recently, a ZnO-based multilayer structure containing an ultrathin metal film was proposed to broaden the optical and electrical properties of ZnO for device applications [6]. These ZnO-based multilayer structures have been used as transparent conducting electrodes or optical fibers in various optoelectronic devices [9]. Several ZnO-based multilayer systems, such as ZnO/Ru/ZnO, ZnO/Cu/ZnO, ZnO/Al/ZnO, and ZnO/Ag/ZnO, have been fabricated using vacuum processes, and their optical absorption, transmittance, and electrical properties have been investigated [6,10,11].

By controlling the relative thickness of ZnO and metals, regulating the number of layers in a multilayer structure, and choosing a proper metal as an interlayer, improving the intrinsic electrical and optical properties of ZnO in ZnO-metal composite structures is possible. Recently, substantial photoluminescence (PL) enhancement of ZnO films was

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achieved by coating metal nanoparticles on ZnO surfaces through surface plasmon coupling [12]. Surface plasmons excited on thin metallic structures/dielectric interfaces have been used to enhance the luminescence of light-emitting materials and devices [12,13]. Among various plasmonic metal materials, it has been shown that Ag particles have strong interactions with visible light through the resonance of the oscillations of the electrons within them. Although a ZnO–Ag–ZnO multilayer system is promising for use in transparent contact electrodes because of its low resistivity and satisfactory optical transmittance property, reports on the correlation between the microstructure and PL properties of the layered ZnO–Ag thin-film structures are still limited. In this study, various layered structures consisting of ZnO film and Ag nanoislands were fabricated by using sputtering to investigate the microstructure-dependent PL properties. This will expand the application of ZnO–metal layered structures in optical and optoelectronic devices.

2. Experimental

ZnO–Ag single and multilayer thin films were sputter deposited on silicon substrates using ZnO and Ag targets. The substrates were supersonic clean out in acetone, rinsed in alcohol and subsequently dried in flowing air gas before deposition. The ultra-thin Ag film was deposited at 20 mTorr under an Ar atmosphere at room temperature and the Ag sputtering duration was 25 s. The Ag island film on the Si substrate can be obtained after annealing at 300 °C in vacuum for 1 h. The ZnO layer was subsequently deposited onto the Ag island film to form single ZnO–Ag layer thin film. The multilayer ZnO–Ag structure was also prepared by alternating deposition of Ag and ZnO layers. The working pressure for ZnO thin film growth is fixed at 30 mTorr with a pure Ar atmosphere. A fixed total thickness of 150 nm for ZnO was designed for all single and multilayer films.

X-ray diffraction (XRD) with Cu K α radiation was used to investigate the crystalline quality of the prepared thin films. The surface morphology was investigated with a field emission scanning electron microscopy (FESEM). The detailed structure of the ZnO–Ag thin films was further characterized by high-resolution transmission electron microscopy (HRTEM). The composition analysis is performed using EDS attached to the TEM. The room temperature dependent photoluminescence (PL) spectra are obtained using the 325 nm line of a He–Cd laser.

3. Results and discussion

The insets of Fig. 1 display the ZnO–Ag thin-film configuration. The ZnO single-layer film was also grown for comparison. Fig. 1 shows the XRD patterns of the ZnO, single-ZnO–Ag-layer, and multi-ZnO–Ag-layer thin films. Fig. 1(a) shows a strong Bragg reflection by ZnO (002). The hexagonal ZnO structure has a *c*-axis lattice constant of approximately 0.52 nm calculated according to the (002) peak; no additional crystallographic planes were observed, revealing that the ZnO thin film was highly *c*-axis textured. By inserting

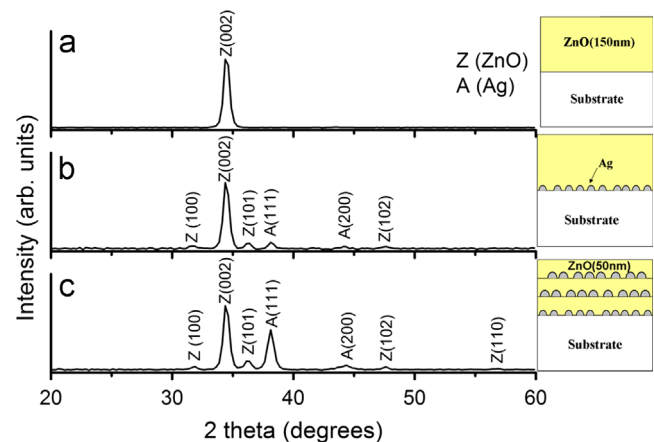


Fig. 1. XRD patterns of the ZnO thin films with and without Ag island layers: (a) without Ag island layer, (b) with one Ag island layer, and (c) with three Ag island layers. The configuration of the samples are shown in the insets.

an Ag island layer, clear Bragg reflections of (111) and (200) for the metallic Ag was observed for the single-ZnO–Ag-layer thin film. The peak intensity of Ag increased as more Ag was inserted for the triple-ZnO–Ag-layer thin film. Comparatively, a relatively strong peak intensity was observed for the (111) of the metallic Ag, revealing a preferred (111) growth of metallic Ag islands on the Si substrate and ZnO layer. No Bragg reflection from the oxidation phases of Ag was observed in this work, and pure metallic Ag–dielectric ZnO heterostructures were formed. In addition to strong Bragg reflections of ZnO (002), however, several crystallographic planes of ZnO (100), (101), (102), and (110) were observed, as shown in Fig. 1(b) and (c). This reveals that the Ag underlayer impairs the growth of *c*-axis-oriented ZnO crystals. The peak intensity ratios of (002) Bragg reflections to all crystallographic planes, $I(002)/[I(100)+I(002)+I(101)+I(102)+I(110)]$, in the polycrystalline ZnO thin films were calculated. Before the calculation, the background intensity of the Bragg reflections was initially deducted. The relative intensity ratios for the single-layer-ZnO–Ag and triple-layer-ZnO–Ag thin films were approximately 0.86 and 0.79, respectively. The triple-layer thin film exhibited a more random crystallographic orientation because the Ag islands might not have provided suitable crystallographic compatibility for the *c*-axis growth of ZnO.

Fig. 2(a)–(c) shows the surface morphology of ZnO, single-ZnO–Ag-layer, and triple-ZnO–Ag-layer thin films. The surface of the ZnO thin film without an Ag island underlayer is flat and dense, and no pores were observed on the surface. The size range of the ZnO thin-film surface grains was approximately 50–80 nm. Fig. 2 (b)–(c) illustrates how the surface of the ZnO-based thin films became rough by adding the Ag island underlayer. Regarding the single-ZnO–Ag-layer film, the surface grain size exhibited a large distribution, ranging from 50 to 150 nm. Moreover, regarding the triple-ZnO–Ag-layer thin film, the range was approximately 40–125 nm. The compactness of the film surface decreased as the number of ZnO–Ag layers increased. The triple-ZnO–Ag-layer thin film exhibited a columnar grain structure, and clear gaps among the

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