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SCANNING TRANSMISSION ELECTRON MICROSCOPY INVESTIGATION OF ZnO:Al BASED THIN FILM TRANSISTORS

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High Resolution Transmission Electron Microscopy (HRTEM), Selected Area Electron Diffraction (SAED), Scanning Transmission Electron Microscopy with High-Angle Annular Dark-Field detector (HAADF-STEM), and Energy Dispersive X-ray Spectroscopy (EDXS) studies were performed in order to investigate the nanostructure, chemical composition and elemental distribution in depth of ZnO:Al thin films used as active channel layers of the thin films transistors (TFT) as well as at the interface with the Ti/Au metallization contacts. Energy Dispersive X-ray spectra (EDXS) and elemental maps acquired in the cross section of a TFT device evidenced the composition and the localization of atomic species and revealed the local chemistry at the nanometer scale rough interfaces.

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1. Introduction

The nanometer dimensions of the today devices require the precise control of the films thickness in relationship with the microstructure and chemical composition, and of their interface properties, since these characteristics strongly affect the devices functionality [1-3]. ZnO:Al doped thin films are widely investigated for applications in transparent electronics, sensors, photodetectors, solar cells, of highest interest being optimizing the structure in relationship with opto-electrical characteristics [4-6]. Several studies have demonstrated the effect of doping elements *e.g.* Al, in improving the ZnO conduction properties, and evidenced the drop of optical properties related to structural changes induced by these impurities. The improvement of transport properties occurs for a given concentration of dopant element, while its accumulation in certain regions of ZnO thin films or at the interface with the substrates leads to deterioration of electrical characteristics of the devices.

Comprehensive analyses by high resolution microscopy techniques were performed in order to establish the crystallinity and morphology of the ZnO:-Al doped thin films as function of growth temperature [7-8]. Recently, X-ray electron spectroscopy imaging in high resolution microscopy was used for characterization of various dopants distribution and defects structures in doped ZnO systems. Several such systems ranging from powders and nanostructures to thin films were studied [9, 10]. However, the local chemical composition and elemental mapping at the level of the ZnO based devices structure are still limited.

In this study we used High Resolution Transmission Electron Microscopy (HRTEM), Selected Area Electron Diffraction (SAED), Scanning Transmission Electron Microscopy with

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High-Angle Annular Dark-Field detector (HAADF-STEM) images and STEM-Energy Dispersive X-ray Spectroscopy (EDXS) techniques for cross sectional nanoscale investigation of local chemical composition and elemental mapping of ZnO:Al thin films used as channel layers in thin films transistors (TFT). Furthermore, the interface of ZnO:Al with Ti/Au thin films (used as metallization contacts) was analysed.

2. Experimental

The TFT devices in a bottom gate configuration were fabricated by sol-gel deposition of ZnO:0.5 at.% at. Al doped films as channel layer on SiO_2 (200 nm)/Si substrates. The source, the drain, and the bottom gate contacts were prepared by thermal evaporation of Ti/Au contacts in vacuum (10^{-6} Torr), through a metallic mask. TFTs were designed with channel width (W) 1100 μm and channel length (L) 280 μm. Details of the sol-gel procedure were presented elsewhere [11].

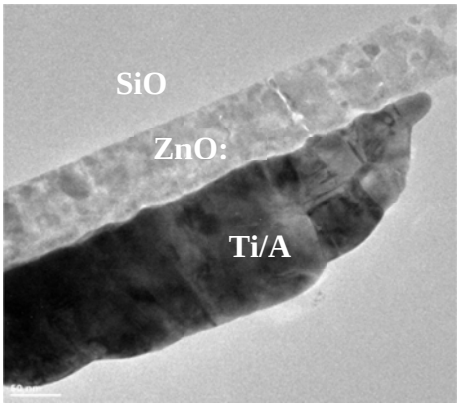
High resolution structural and chemical analysis were carried out on the cross section samples from the TFT device by using HRTEM, SAED and STEM microscopy with EDX spectroscopy. The measurements were performed using a TECNAI F30 microscope operated at 300 kV with EDAX and EELS facilities. The cross section samples for TEM analyses were prepared using an ion-milling equipment FISCHIONE with Ar ions.

3. Results and discussion

The HRSTEM-EDXS cross sectional study of a TFT device was carried out in order to determine the in-depth elements distribution in the ZnO:Al film used as channel layer. Additionally, the interfaces with the Si/SiO_2 substrate and the Ti/Au films used for source and drain metallization contacts were investigated. The ZnO:Al thin film with 0.5 at.% Al concentration and about 50-60 nm thickness was deposited on a Si/SiO_2 substrate by a sol-gel multilayer process previously described [11]. EDX spectra recorded on different regions of the ZnO:Al film cross section showed a non-uniform distribution of Al. EDX spectra recorded at the film surface, in the central region, and at the interface with the Si/SiO_2 substrate, reveals that the Al content is about 0.28 at.%, 1.42 at.%, and 0.52 at.% respectively, and that the accumulation of Al appears in certain zones along the interface [12]. Further, we aim to investigate the interface between the ZnO:Al and Ti/Au films. Fig. 1 shows the TEM bright field image of the TFT cross section in this region. It can be observed that the polycrystalline ZnO:Al film has a rather uniform thickness with surface roughness leading to an irregular interface with the metallization contact.

TEM and HRTEM images of the TFT cross section are presented in Fig. 2 (a)-(c). Fig. 2 (a) shows the Ti/Au and the interface with the channel layer. The HRTEM image of ZnO:Al thin film presented in Fig. 2 (b), and the corresponding SAED image (Fig. 2 c), reveal the presence of 5-10 nm size nanocrystals in the polycrystalline film, and the hexagonal wurtzite structure of ZnO.

With the aim of locate the atomic species in the cross section of TFT, we acquired EDX spectra and elemental maps using HAADF-STEM technique. Figs. 3 (a) and (b) show cross sectional HAADF-STEM images of the TFT channel in the region of Ti/Au metallization contact. EDX spectrum presented in Fig. 3 (c) was acquired in the region of the ZnO:Al film, in the area pointed in the STEM image. The spectrum shows intense peaks corresponding to Zn-L and O-K, and low intensity peaks corresponding to Al-K, Si-K and Ti-K respectively.



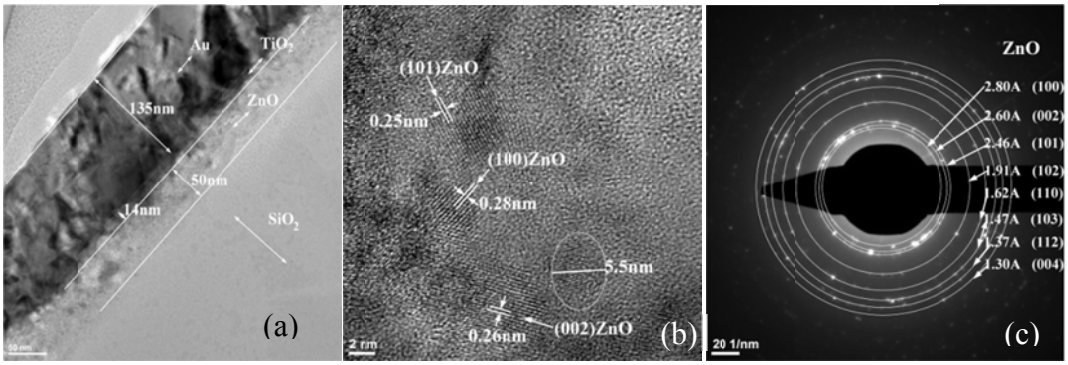
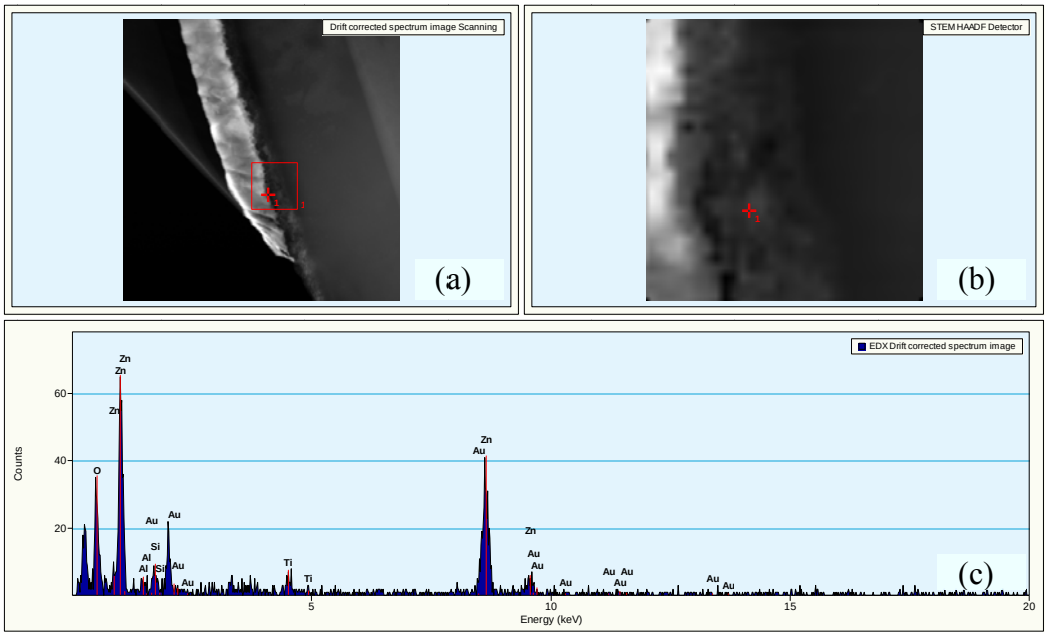


Fig. 2 TEM cross section and HRTEM images: (a) TFT with ZnO: Al channel layer and Ti/Au metallization contact, (b) ZnO: Al channel layer and nanocrystalls in the film, (c) SAED of the region (b), showing the wurtzite type structure of the film.



Figs. 4 show HAADF-STEM image (a), and EDX maps of Au-M (b), Ti-K (c), Zn-L (d) and O-K (e) elements. Variation in the colour intensity observed in the elemental maps is correlated with the number of atoms contained in a particular region, and can give an evaluation of the elemental composition at the interface between the ZnO:Al channel layer and the metallization contact.

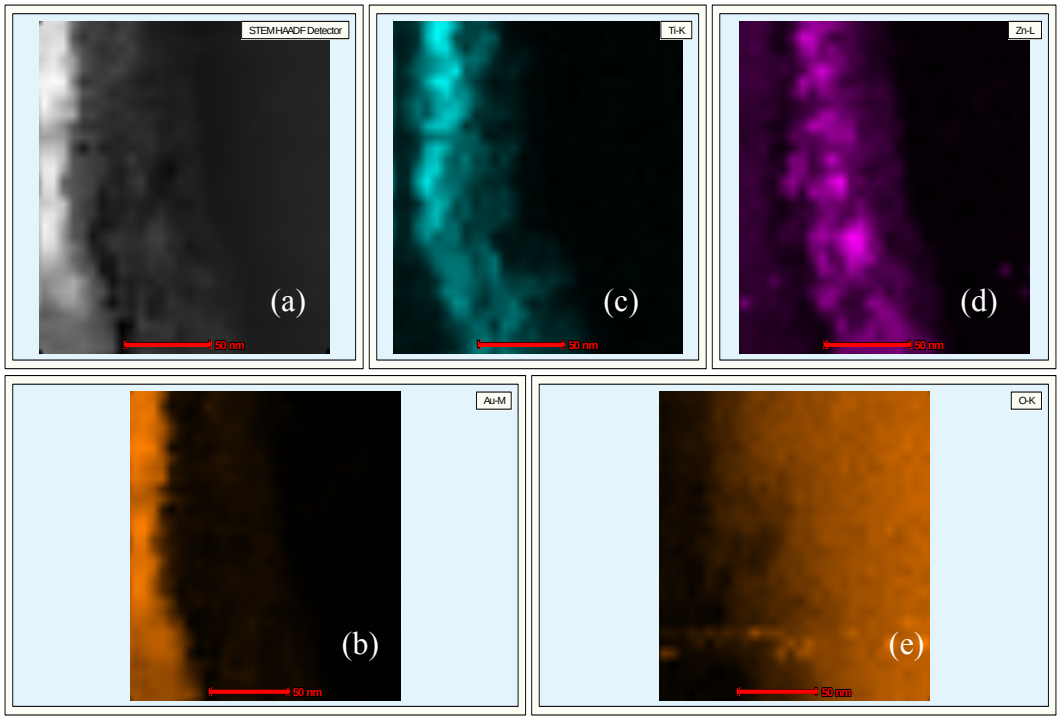


Fig. 4 Cross section of TFT device: (a) HAADF-STEM image and corresponding EDS maps of: Au (b), Ti (c), Zn (d) and O elements (e).

Figs. 4 (b) and (c) evidence a nonuniform distribution of Ti and Zn atoms in the region of Au/Ti interface with ZnO:Al film. The O map, Fig. 4 (e), reveals a lower colour intensity and a nonuniform distribution in the region of ZnO:Al film and an increased intensity in the region of the SiO_2 substrate, where the distribution is uniform.

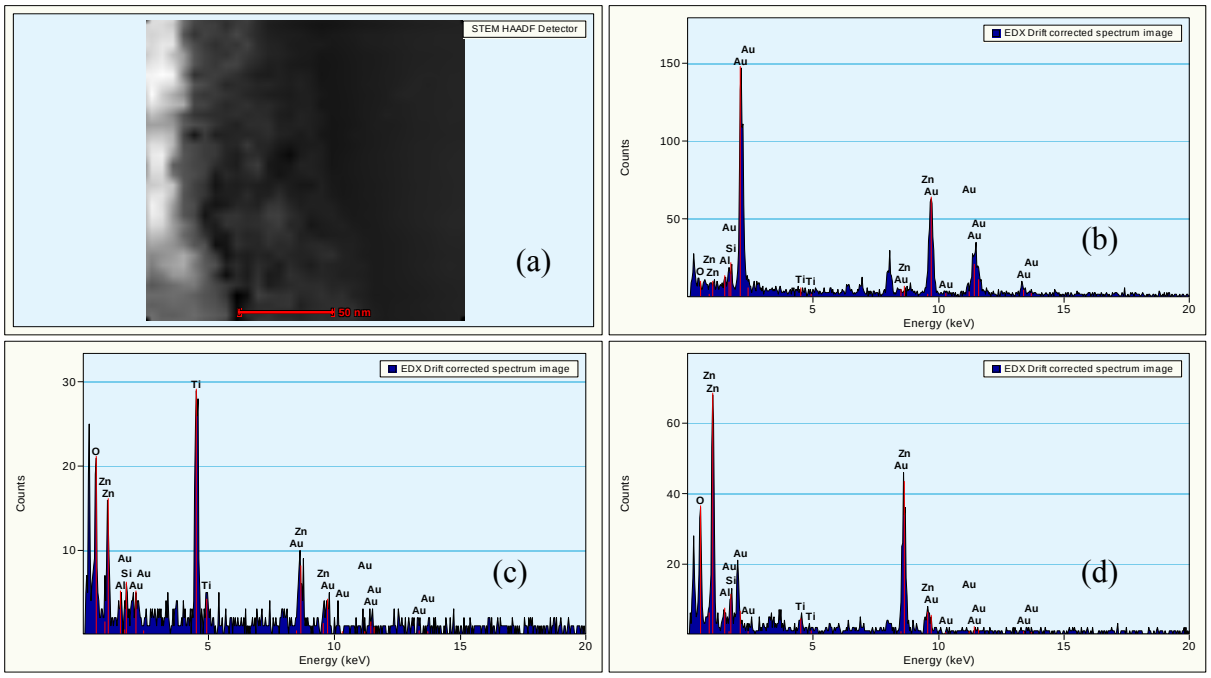


Fig. 5 HAADF-STEM image of the region presented in Fig.3 (a) and EDX spectra (b)-(d), recorded along the interface between ZnO:Al film and Ti/Au metallization films.

The HAADF-STEM image (a) and EDX spectra (b)-(d) from Figs. 5 (a)-(d) demonstrated the in depth nonuniform distribution of Ti, Zn and O atoms at the Au/Ti interface with ZnO:Al channel layer. The spectrum recorded in the region of Au thin film is presented in Fig. 5 (b). The spectrum displays that Au is the major element, and that Ti is not present in this region. Intense peaks associated with Ti and Zn appear in the spectrum recorded in the region of Ti film interface with ZnO:Al film, Fig. 5 (c), in agreement with the Ti and Zn atoms nonuniform distribution observed in the cross sectional elemental maps, Figs. 4 (c) and (d). EDX spectrum recorded in the region of ZnO:Al film is presented in Fig. 5 (d). A low intensity peak corresponding to Ti appears in the spectrum, revealing the presence of Ti atoms in the ZnO:Al film. Conversely, a low intensity peak associated with Al is also observed in the spectrum shown in Fig. 5 (d).

Previously it was reported that the nonalloyed Ti/Au contact to ZnO shows very linear current-voltage behavior with a specific contact resistivity of 2.2×10^{-5} Ωcm [13]. The Auger electron spectroscopy (AES) correlated with TEM investigation evidenced the presence of a thin interfacial Ti-O layer, whose formation was related to outdiffusion of oxygen from ZnO. The high formation energy of Ti-O phases can explain the interfacial layer development without an annealing process. Also, Auger electron spectroscopy depth profiles of the Ti/Al/Pt/Au contact on ZnO evidenced that O appears to diffuse outward and determine Ti-O phase formation after annealing at 250 ^{o}C [14]. Ti-Zn compounds were observed within the Ti layer and the out-diffusion of Zn and O occurred from ZnO to Ti in the case of annealing at 500 ^{o}C. These processes that occurred at the interface led to deterioration of contact properties [15]. In our experiment the STEM-EDX spectrum presented in Fig. 5 (c) shows an intense maximum of Ti in the region of Ti film and weak peaks associated with Zn and O. The spectrum presented in Fig. 5 (d) reveal intense peaks corresponding to Zn and O, and a very weak peak associated with Ti in the ZnO:Al film region. While it would be difficult to advance a phenomenological model of elements evolution based on the measurements presented, STEM-EDXS maps and STEM-EDX spectra revealed the atoms migration in the interfacial layer in the nonalloyed Ti/Au contact to ZnO:Al channel layer. The current-voltage characteristics of the TFT device presented a linear character, with a high specific contact resistivity [12]. A small leakage current was observed and attributed to an Al accumulation at the interface of the ZnO:Al channel layer with the substrate [12].

The elemental line distribution recorded in the STEM-EDAX mode, along the cross sectional TFT device, is presented in Fig. 6. TEM image from the Fig. 6 (a) highlights the TFT cross section and the line along which were recorded the elements distribution showed in Fig. 6 (b). Distribution of the characteristic X-ray intensities Au-M, Ti-K, Zn-K, Al-K, Si-K are roughly proportional to elements concentrations along the line. It is observed that Zn concentration presents a small maximum at the interface with Ti film, then increases and is rather constant in the region of the ZnO:Al film, with a further small maximum at the interface with the SiO_2 substrate. The Al-K characteristic X-ray intensity contour pursues the Zn-K characteristic X-ray intensity contour along the line, e.g. the ratio Zn/Al is almost constant in the depth of the ZnO:Al film.

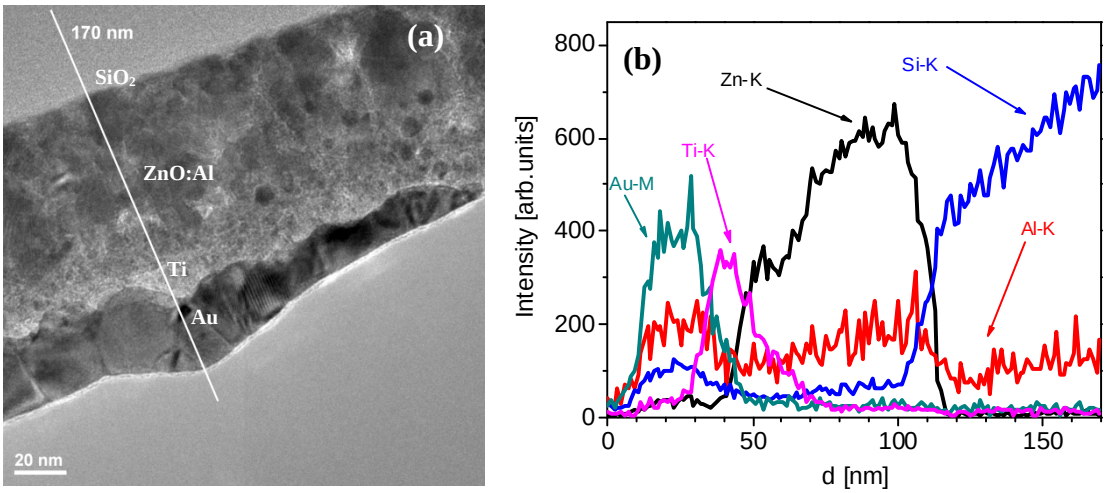


Fig. 6 Characteristic X-ray intensities Au-M, Ti-K, Zn-K, Al-K, Si-K along the line showed in (a) TEM image of TFT cross section.

It can be seen that the Au-K intensity drops rapidly at the interface with Ti film, which revealed that Au is not diffussing in this area. At the interface between the Ti film and ZnO:Al film the characteristic X-ray intensities Ti-K and Zn-K slowly decreases and increases respectively, revealing an elements interdiffusion in this region.

4. Conclusions

HRTEM, SAED, STEM-EDX, HAADF-STEM techniques were used for investigation of nanostructure, local chemical composition and elements maps in the cross section of a TFT device with ZnO:Al thin film as active layer and Ti/Au source and drain metallization contacts. The EDX spectra acquired in different points of the ZnO:Al film evidenced a variation of Al content from 0.28 at.%., at the surface, to 1.42 at.%, in the film bulk, to 0.52 at.% at the interface with Si/SiO_2 substrate. The STEM-EDAX line distribution of the characteristic X-ray intensities Au-M, Ti-K, Zn-K, Al-K in the cross section depth demonstrated an almost constant ratio Zn/Al in the ZnO:Al film. Moreover Ti and Zn interdiffusion at the interface of the channel active layer with the metallization contact was evidenced.

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