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Compositional influence on the electrical performance of zinc indium tin oxide transparent thin-film transistors

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ABSTRACT

In this work, zinc indium tin oxide layers with different compositions are used as the active layer of thin film transistors. This multicomponent transparent conductive oxide is gaining great interest due to its reduced content of the scarce indium element. Experimental data indicate that the incorporation of zinc promotes the creation of oxygen vacancies, which results in a higher free carrier density. In thin-film transistors this effect leads to a higher off current and threshold voltage values. The field-effect mobility is also strongly degraded, probably due to coulomb scattering by ionized defects. A post deposition annealing in air reduces the density of oxygen vacancies and improves the field-effect mobility by orders of magnitude. Finally, the electrical characteristics of the fabricated thin-film transistors have been analyzed to estimate the density of states in the gap of the active layers. These measurements reveal a clear peak located at 0.3 eV from the conduction band edge that could be attributed to oxygen vacancies.

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1. Introduction

Transparent conductive oxides (TCOs) have attracted much attention in the optoelectronic industry during the last years due to its emerging interest in transparent electronics [1,2]. TCOs are degenerately doped semiconductors with a band gap higher than 3 eV and thus, conductive and transparent in the visible wavelength range. These properties, combined with the possibility of processing layers at low temperatures, made them suitable for several applications such as solar cells or flexible electronic devices. It is important to study and characterize the films, to further understand the interaction of each layer within the device.

Among all the TCOs, indium tin oxide (ITO) is widely used because of its low resistivity (in the order of 10^{-4} $\Omega\cdot$cm) and high transmittance (higher than 80%) [1]. However, it has the disadvantage of being expensive due to the scarcity of indium. On the contrary, zinc oxide (ZnO) seems to be a promising material to substitute ITO given its comparable electrical and optical properties and the abundance of Zn in the Earth's crust. For this reason, quaternary compounds such as zinc indium tin oxide (ZITO) have attracted much interest to lower the concentration of indium [1]. The electrical, optical and morphological properties of this alloy have been previously characterized [3,4]. Moreover, its electronic band structure has been also carefully studied [5]. The aim of this work is to study the electronic properties of different ZITO layers by manufacturing thin film transistors (TFTs) [6–10].

In the present study, a series of amorphous ZITO thin films with Zn cation concentration ranging from 17% to 67%, as well as pure ITO and ZnO thin films were employed as TFT active layers in order to compare their behavior within the device. As the Zn concentration increases, the O vacancies at the layer surface are said to increase [2,5], thus indicating a higher density of charge carriers (electrons in this case). An annealing process in air atmosphere has been applied to each layer in order to reduce the carrier concentration by decreasing the number of O vacancies. According to methodologies used to describe metal-oxide semiconductor field-effect transistors [1], we have extracted parameters such as carrier mobility, drain current on–off ratio and threshold voltage, which are three of the basic parameters to quantify TFTs performance. Besides, the density-of-states (DOS) will be estimated by analyzing the temperature dependence of the channel conductance following a procedure described elsewhere [11].

2. Experimental details

All the TFTs with ZITO active channel studied herein have been fabricated using a bottom gate–top contact structure as depicted in Fig. 1. A *p*-type Si wafer ($N_A \sim 10^{16}$ cm^{-3}) with a thermally oxidized layer (250 nm-thick SiO_2) has been employed as a substrate and gate insulator, respectively. In this application, *p*-type wafers are more suitable to replicate a metallic gate electrode because ZITO semiconductors (*n*-type) operate in accumulation under a positive voltage applied to the gate [8]. SiO_2 was etched from the wafer rear-side before further deposition steps were performed.

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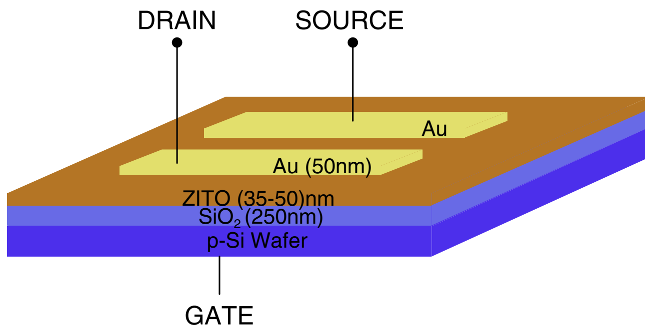

Fig. 1. Schematic cross-section of thin film transistors.

Table 1
Deposition power, cation content ratio and four point measured resistivity of the different samples.

Sample name (thickness nm)	ZnO rf power (W)	ITO rf power (W)	Zn content ratio (%)	In content ratio (%)	Sn content ratio (%)	ρ (Ω · cm)
ZITO25 (35)	25	50	17.1	75.5	7.5	9.56E−03
ZITO50 (40)	50	50	36.6	57.6	5.8	1.95E−02
ZITO75 (45)	75	50	48.5	46.7	4.8	3.02E−02
ZITO100 (40)	100	50	53.8	41.8	4.4	2.64E−02
ZITO125 (50)	125	50	62.8	34.5	2.7	7.65E−02
ZITO150 (45)	150	50	67.3	30.2	2.6	8.10E−02

ZITO samples were deposited over the oxidized front-side of *p*-type Si wafers at the University of Barcelona by radio frequency (RF) magnetron co-sputtering of ZnO and ITO (In_sO_3 doped with 10% SnO_2) at room temperature. 3-in. target purity was of 99.995% for ZnO and 99.99% for ITO. The equipment used was an ATC ORION sputtering from AJA International, Inc. The process consists of simultaneously sputtering two targets by applying a constant RF power (50 W) to ITO target and increasing the power applied to ZnO one from 25 W to 150 W in 6 steps of 25 W, thus getting 6 kind of layers ranging from 17% to 67% of Zn content ratio to whole metal content, as determined from X-ray Photoelectron Spectroscopy (XPS) analyses. Samples were kept at 11.7 cm away from the targets, with a rotation speed of 10 rpm. The thickness of the layers ranged between 35 and 50 nm. Each deposition process was performed over 2 samples, having a total of 16 samples with 6 different Zn contents plus pure ITO and ZnO.

Increasing the Zn content increases the O vacancy density at the film surface, as it has been reported by Carreras et al. [5,12], thus resulting in a highly conductive surface layer [13]. To reduce this effect, one sample of each kind was annealed in air atmosphere. Each sample was heated during 1 h at 300 °C, with a temperature ramp-up of 30 min. The aim of the thermal treatment was to oxidize the film to decrease the carrier concentration by reducing the oxygen vacancy density [14] or by chemisorption of oxygen at the ZITOs surface and nano-grain boundaries. The morphology of the samples was previously characterized by X-Ray Diffraction Spectroscopy and High-Resolution Transmission Electron Microscopy [15]. Results showed that the structure is not completely amorphous as deduced by XRD, but it shows nanocrystals embedded in a compact amorphous structure.

Gold drain and source electrodes were thermally evaporated through a shadow mask at a pressure below 10^{-3} Pa directly over the ZITO films. The thickness of gold electrodes was 50 nm. Channel width and length were 2.6 mm and 80 μm, respectively.

Resistivity measurements were performed by a four point probe system (Jandel RM3). This measurement was performed with the same ZITO layers deposited over a glass substrate. All other electric measurements were performed inside a low-vacuum chamber (<10 Pa), in dark conditions using an Agilent 4156C source/monitor. To measure the temperature dependence of the electrical characteristics, all the devices were heated by means of a MMR Technologies temperature controller (model K-20).

3. Results

The composition of the films is summarized in Table 1 where RF magnetron power, estimated cation content ratios and resistivity are shown. All these data were obtained as described above.

All the TFTs have been electrically characterized according to standard models [16]. The current that flows along the TFT channel is a function of the gate to source (V_{GS}) and drain to source voltages (V_{DS}) distinguishing among three operational regimes (subthreshold, linear

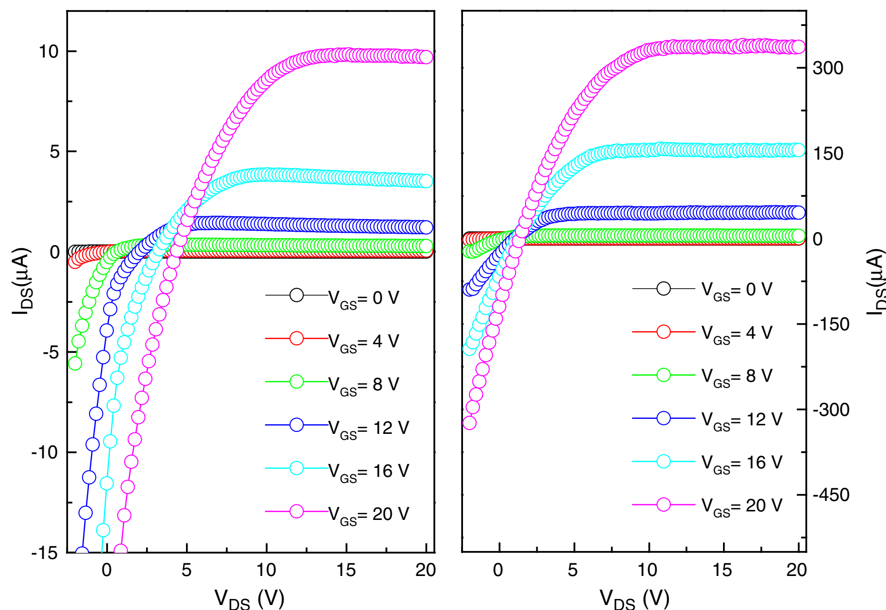

Fig. 2. $I_{DS} - V_{DS}$ characteristics for a ZITO50-channel TFT with and without annealing treatment. [V_{GS} = 0 up to 20 V in 4 V increments.]

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