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Detailed investigation of defect states in Erbium doped In_2O_3 thin films

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ABSTRACT

Erbium doped Indium Oxide (In_2O_3:Er) thin films (TFs) were synthesised by spin-on technique. Secondary Ion Mass Spectrometry confirmed that Er is incorporated into the In_2O_3 lattice and formed an In-O-Er layer. The current–voltage loop produced a lower loop current window of $\sim 3.6 \times 10^{-4}$ A for In_2O_3:Er TF based devices. The Au/In_2O_3:Er/Si Schottky devices have lower ideality factor (~6) and higher barrier height (~0.63 eV) at 300 K than Au/In_2O_3/Si control samples. A blue shift in the main band-gap (~50 nm) was calculated for In_2O_3:Er TFs from 10 K photoresponse. The Au/In_2O_3:Er/Si samples show higher photosensitivity in the temperature range 10 K–300 K and maximum (~15 times) in the UV region at 10 K as compared to the Au/In_2O_3/Si devices. In addition, the Au/In_2O_3:Er/Si devices have better UV to visible cut-off ratio (~3 times). Excellent temporal responses were recorded for Au/In_2O_3:Er/Si in the UV region as compared to Au/In_2O_3/Si.

1. Introduction

Metal oxide semiconductor (MOS) thin films (TFs) are important for the fabrication of transparent optoelectronic devices [1]. Amongst MOS based materials, Indium Oxide (In_2O_3) is a better transparent oxide semiconductor (TOS) material compared to other well-known materials such as TiO_2 and ZnO because of its high carrier mobility (~43.7 cm^2/V-s) [2] and wide direct bandgap of 3.5–3.7 eV [3]. The detection of ultraviolet (UV) light is also very important for applications in flame sensors, missile plume detectors, spatial optical communication devices, UV photosensors, biological and chemical sensors, laser-based devices etc. [4]. In spite of the encouraging results reported, the carrier concentration in MOS devices is difficult to control due to the presence of large numbers of oxygen vacancies and defects [5,6] which results in non-reliable performance of optoelectronic devices. A major breakthrough in this field was the epitaxial growth of single crystal TOS (Indium Gallium Zinc Oxide) TFs with an atomically flat surface on yttria-stabilized zirconia substrates [7]. However, the reduction in oxygen vacancies in TOS as well as compatibility with existing Silicon technology by cost-effective techniques is challenging [4]. Inorganic materials activated with rare earth ions (REs) have suitable spectroscopic characteristics to design luminescent or display materials [8]. Use of REs with TOSs is receiving much interest in scientific community due to the wide range of technological applications based on their increased transparency in the visible region (Sc or Y doped ZnO) [9], intra-ionic radiative transition processes (Er doped In_2O_3) [10], increased photosensitivity or photo-emissivity (Er doped SnO_2 and Erbium Tin Oxide) [11], bandgap engineering (Ga^{3+} doped $Lu_3Al_5O_{12}$) [12], enhanced photocatalytic activity (Er^{3+} doped β-Bi_2O_3) [13], defect engineering (Praseodymium doped GaN) [14], reduction of the oxygen and N related defects by Er [15] etc. Amongst these REs, Erbium (Er) is optically active f- block element and can emit photon corresponding to the wavelength of ~1540 nm, which lies in the region of minimum losses for silica-based waveguides used in fibre optic communications [16–19] and silicon photonics [20]. Researchers also reported on the improved detection of UV light by Er doped In_2O_3 (In_2O_3:Er) TFs as compared to undoped In_2O_3 TFs and investigated the removal of oxygen-related defect states from the host In_2O_3 material using optical and electrical techniques [4]. However, it is important to find out the energy positions of the defect states within the bandgap of In_2O_3 after incorporation of Er atoms and their roles in device performance. Amongst all the defects identified in TOSs, oxygen vacancy is one of the most significant and is considered to be the dominant defect in In_2O_3. Oxygen vacancies in MOS had been investigated extensively both experimentally [21–23] and theoretically [24,25]. For example, low-temperature photo-capacitance and photocurrent measurements were performed to study defects introduced in GaAs by N incorporation [15]. However, there are no reports on the investigation of defects using temperature dependent current–voltage (I–V) characteristics, photosensitivity and low temperature temporal responses of In_2O_3:Er

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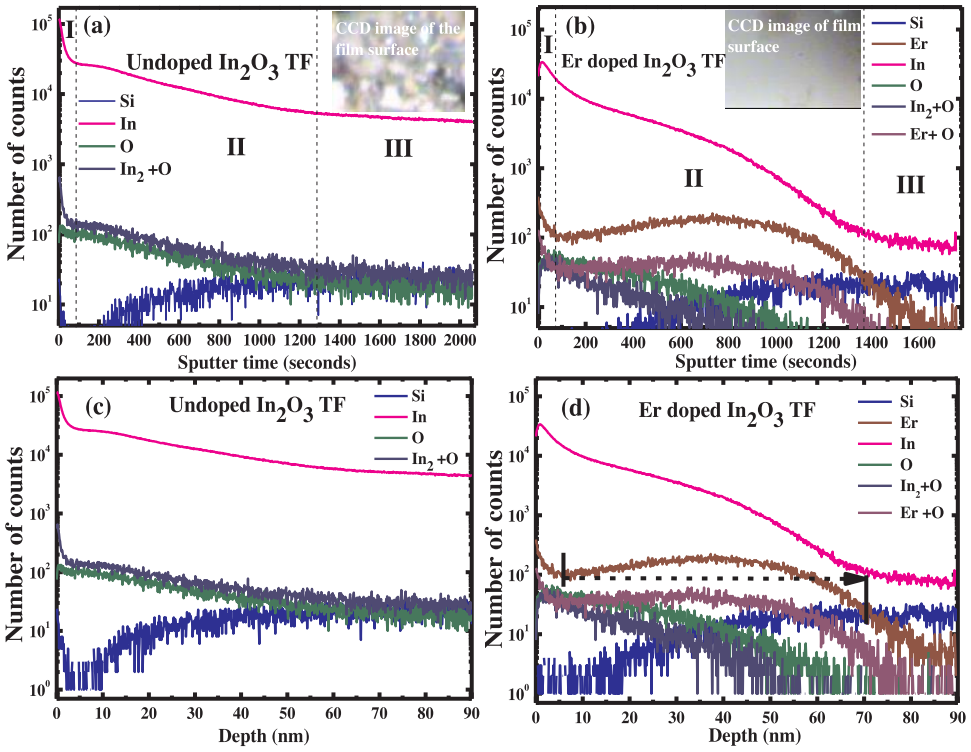

Fig. 1. Sputter time vs. number of counts for (a) undoped In_2O_3 TF (CCD image in inset) and (b) In_2O_3:Er TF (CCD image in inset); depth (in nm) of the TF vs. number of counts for (c) undoped In_2O_3 TF and (d) In_2O_3:Er TF.

TFs, which is important for improving their performance as UV detectors.

In this paper, the doping of Er into In_2O_3 lattice has been confirmed by Secondary Ion Mass Spectrometry (SIMS) analysis. We have performed I–V hysteresis, temperature dependent (10 K - 300 K) photosensitivity, low temperature (10 K) responsivity and low temperature (10 K) temporal response of detectors in order to probe the energy positions of the defects present in undoped In_2O_3 and In_2O_3:Er TFs. The theoretical analysis of the ideality factor (η) and barrier height (Φ_B) was performed using Schottky model by taking into consideration the presence of defects. Low-temperature photocurrent and temporal responses provided the information about the type of defects and energy level positions.

2. Experimental details

2.1. Synthesis of undoped and Er doped In_2O_3 TFs and device fabrication

In_2O_3 and In_2O_3:Er TFs were deposited on RCA cleaned p-type Si (100) substrates (resistivity ~10 Ω-cm, MTI, USA) by spin coating technique using the chemical route [4]. The sol-gel for In_2O_3 TF was prepared by dissolving 0.5 g Indium (III) Chloride ($InCl_3$) anhydrous powder (5 N purity, Sigma-Aldrich) into 30 ml of acetylacetone (purity > 99%, Merck) under ultrasonication (~50 °C, 15 min) and left for ageing for 24 h. It was then spin coated (spin NXG-P1, apex-icindia) on the Si substrate (rotation speed ~1000 rpm for 1 min). The as-deposited film was then annealed in a muffle furnace (~400 °C for 10 min) under atmospheric air condition. The entire process was repeated four times in order to get a uniform film over the substrate with final annealing at ~400 °C for 30 min. To prepare In_2O_3:Er solution, 0.04 g Erbium (III) Oxide nanopowder (purity ≥ 99.9%, Aldrich) was dissolved in 10 ml sulfuric acid (H_2SO_4) by ultrasonication (~50 °C, 15 min) and finally added to the previously prepared $InCl_3$ solution. This doped solution was ultrasonicated (~50 °C, 15 min) and left for 48 h for ageing. The In_2O_3:Er sol-gel was then spin-coated on a p-Si substrate (rotation speed ~6000 rpm for 2 min) followed by 10 min open air annealing (~400 °C) for four times with final annealing at ~400 °C for 30 min as has already been described. Gold (Au) was evaporated through circular holes of an aluminum mask, which was placed on top of the samples to form the upper electrode (diameter ~2 mm) contact of the Schottky devices by thermal evaporation method. Indium was used as an Ohmic contact on the backside of p-Si substrate. Both the undoped In_2O_3 (Au/In_2O_3/Si) and Er doped In_2O_3 devices (Au/In_2O_3:Er/Si) were prepared using the same procedure.

2.2. Characterisation techniques

Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) (PHI TRIFT V nano TOF, Physical Electronics, USA) was performed using dual beam configuration on the undoped and In_2O_3:Er TFs to analyse the depth profiles (using a stylus-based profilometer). A caesium (Cs^{2+}) primary ion beam of 0.3 μA beam current and 3.0 keV net impact energy was used as a sputtering ion source to etch the sample surface within a raster size of 600 μm x 600 μm, whereas a Gallium (Ga) gun was used to analyse the secondary ions. The SIMS aperture settings were used to restrict secondary ion analysis within the square area within the raster region of 40 μm x 40 μm using liquid metal (Ga) ion gun. This SIMS configuration is used to determine appropriate composition of the TFs [26]. The I–V characteristics of Au/In_2O_3/Si and Au/In_2O_3:Er/Si were investigated using a Keithley 2401 source meter and 300 W Ozone free xenon arc lamp (650-0047). The photocurrent spectra of both detectors were recorded at low temperature using an open beam configuration through a monochromator (120-9055, Science tech Inc., Canada) and a He cryostat (Advanced Research System).

3. Results and discussion

3.1. SIMS analysis and depth profiling interpretation

Fig. 1 shows the semi-logarithmic plot of the number of counts of different ions against sputter time and counts vs depth profiles, where the time scale is converted into depth (nanometers) by using a stylus-based profilometer. The inset of Figs. Fig. 1(a) and (b) shows the CCD images of the TF surface. The undoped In_2O_3 TF displays high surface

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