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Enhanced electrical and optical properties of room temperature deposited Aluminium doped Zinc Oxide (AZO) thin films by excimer laser annealing

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ABSTRACT

High quality transparent conductive oxides (TCOs) often require a high thermal budget fabrication process. In this study, Excimer Laser Annealing (ELA) at a wavelength of 248 nm has been explored as a processing mechanism to facilitate low thermal budget fabrication of high quality aluminium doped zinc oxide (AZO) thin films. 180 nm thick AZO films were prepared by radio frequency magnetron sputtering at room temperature on fused silica substrates. The effects of the applied RF power and the sputtering pressure on the outcome of ELA at different laser energy densities and number of pulses have been investigated. AZO films deposited with no intentional heating at 180 W, and at 2 mTorr of 0.2% oxygen in argon were selected as the optimum as-deposited films in this work, with a resistivity of 1×10^{-3} Ω.cm, and an average visible transmission of 85%. ELA was found to result in noticeably reduced resistivity of 5×10^{-4} Ω.cm, and enhancing the average visible transmission to 90% when AZO is processed with 5 pulses at 125 mJ/cm^2. Therefore, the combination of RF magnetron sputtering and ELA, both low thermal budget and scalable techniques, can provide a viable fabrication route of high quality AZO films for use as transparent electrodes.

1. Introduction

Transparent conducting oxides (TCOs) are ubiquitous in a wide range of optoelectronic devices such as flat panel displays, thin film solar cells, and transparent thin film transistors [1–3]. Amongst the TCOs family, tin-doped indium oxide (ITO) has been the material of choice for transparent electrode applications. However, although ITO exhibits superior conductivity and transparency with less thickness-dependent resistivity, there is a general desire in the sector to move towards an alternative solution for advanced application of TCOs [2–4]. For instance, flexible applications that impose additional fabrication and environmental constraints such as low thermal budget fabrication and electrical conductivity stability against stretching and bending forces to which ITO is not ideally applicable [3,5]. TCO research has been directed towards exploring reduced indium content or even non-indium based transparent conducting materials such as ZnO-based transparent electrodes. ZnO has been extensively explored for various opto-electronic fields because of its attractive characteristics including, but not limited to, high exciton binding energy of 60 meV, wide and direct band gap of 3.37 eV, low growth temperature as a crystalline material, and high thermal and chemical stability. Furthermore, the ZnO characteristics can be tuned via doping in order to meet various applications requirements. For instance, doping with MgO or CdO results in larger or smaller band gap respectively [6–8]. While, doping with group III elements (Al, Ga and In) results in high quality transparent electrodes. In particular aluminium doped zinc oxide (AZO), due to its low raw material cost and comparable electrical and optical properties to ITO [2,3,8], has attracted much attention for replacing ITO in mass production of photovoltaic devices [1]. AZO was also successfully applied to organic light emitting diodes [9], and to liquid crystal displays [10].

There has been a large body of reported investigations into the fabrication of AZO via various vacuum and solution based deposition techniques such as magnetron sputtering [11–13], pulsed laser deposition [14,15], atomic layer deposition [9], sol–gel process [16], chemical vapour deposition [17], and aqueous

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solution processing [18]. Radio frequency magnetron sputtering is the most commonly used approach owing to its potential for low temperature growths, industrial applicability, as well as relatively good uniformity and reproducibility [12]. Unfortunately, to produce AZO films with high quality electro-optical characteristics for large area applications, it is typical to require high thermal budget at deposition [19], or at post-deposition processing [20]. At the same time, for further optoelectronic developments there has been an increasing interest in low thermal budget fabrication of metal oxides in order to maintain the characteristics of any underlying layers or the substrate itself, as well as to reduce the long processing times needed for conventional thermal processing. Hence, Excimer Laser Annealing (ELA) has been explored as an ultra-rapid, and spatially localised both in area and depth processing method to realise high quality metal oxide thin films at minimal substrate heating [16,21]. Only a few previously reported studies have explored the effect of ELA on AZO. R. Boukhicha et al. [22] and E. Johnson et al. [23] have reported dramatically improved AZO optical properties via ELA (XeCl 308 nm) and a chemical etching process, but only a moderate resistivity reduction was reported from 1.1×10^{-3} to 0.7×10^{-3} Ω.cm for 1 μm thick AZO samples. Q. Xu et al. [24] have reported a resistivity reduction from 3.4×10^{-3} to 2.2×10^{-3} Ω.cm via processing 200 nm thick AZO films with a cw Nd:YAG laser at 1064 nm. W.-T. Hsiao et al. [25,26] have noticeably reduced the resistivity of 200 nm thick AZO samples after annealing with a cw diode laser at 808 nm, from 4.00×10^{-2} to 2.75×10^{-2} Ω.cm. However, the achieved resistivity values by both Q. Xu et al. and W.-T. Hsiao et al. are too high for use as a practical transparent electrode. Nian et al. [15] combined room temperature pulsed laser deposition and ELA (KrF 248 nm) resulting in highly conductive ($\rho=2.23\times10^{-4}$ Ω.cm) and transparent (85% in the visible range) 250 nm thick AZO films, but a 50 nm i-ZnO was required as a buffer layer.

In the present work we report a complete study of RF magnetron sputter deposited AZO films at different deposition parameters on UV grade fused silica substrates, followed by ELA with a Krypton Fluoride laser (KrF) and with variable number of pulses and laser energy density. Unlike all previously published studies, the deposition and processing conditions concluded in this work offer highly transparent and highly conductive AZO thin films, produced at low thermal budget and with scalable techniques.

2. Experimental procedure

2.1. AZO thin films deposition

Three series of AZO samples, all 180 nm thick, were fabricated using an in-house built RF magnetron sputtering system for deposition at room temperature, where one parameter was changed in each series: (a) with various oxygen concentrations diluted in argon (0–0.5%), (b) with varying RF power (40–240 W) applied to the target, and (c) with varying sputtering pressure (1.5–5.0 mTorr). The UV grade fused silica glass substrates (5 cm × 5 cm) were first cleaned with Decon 90 surfactant solution in water (4% by volume) to remove any dust particles from the substrate surface, followed by sequential ultrasonic cleaning in acetone, methanol, and de-ionised water (for 10 min each). Finally, the substrates were individually demoisturised with high purity dry nitrogen. A three-inch diameter ceramic target of ZnO–Al_2O_3 (98–2 wt%) from Testbourne Ltd. was used. Prior to the deposition, a base pressure of 10^{-5} Pa was achieved in the sputtering chamber. Then, the investigated gas mixture was introduced via two mass flow controllers to produce a sputtering gas of a percentage of oxygen in argon. Pre-sputtering was always performed, with the substrate covered for 10 min at the same parameters as for the subsequent deposition. The substrate to target distance was maintained at 10 cm for all depositions in a sputter-up geometry. The substrate temperature during the deposition was monitored with temperature sensitive labels attached to the back side of the substrate. The substrate was rotated at 8 rpm to provide highly uniform films. The film thickness was monitored by an in-situ interferometric monitor and was cross-checked via Veeco Dektak 6M Stylus Profilometer measurements.

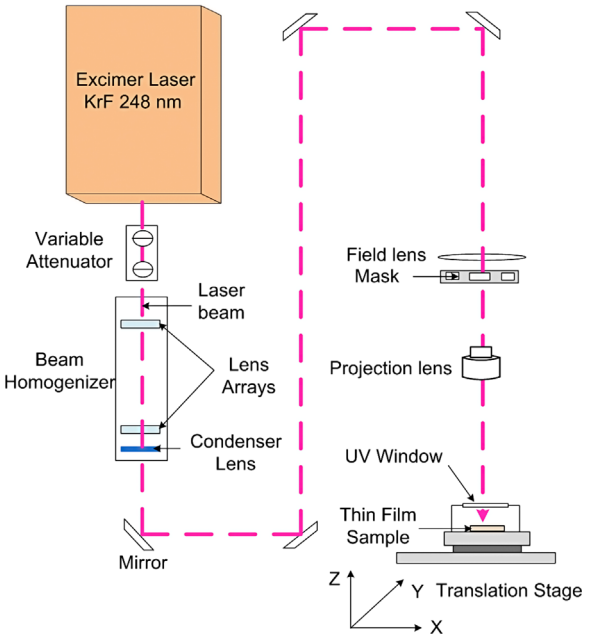

Fig. 1. Schematic diagram of the Excimer Laser Annealing system.

2.2. Excimer laser annealing

ELA was conducted in air at room temperature using the system illustrated in Fig. 1, comprising three main components: (1) the laser source, a Krypton Fluoride (KrF) excimer laser (Lambda Physik LPX 305i), which emits unpolarised light pulses at $\lambda=248$ nm and with a pulse duration of 25 ns. (2) The laser beam delivery and shaping system including an attenuator, a beam homogeniser (Exitech Ltd., type EX-HS-700D, providing laser spots with uniformity better than 2%), mirrors, a field lens, a mask stage, and a projection lens with 1:1 magnification. The laser spot defined by the optics was a square area of 1 cm × 1 cm. (3) An *XYZ* moving stage for samples mounting and manipulation during processing.

Initially, single pulse investigations were conducted in order to identify the threshold energy density required for the onset of an effect on the AZO characteristics, as well as the limit above which material ablation occurs. The investigated energy density range was from 25 to 500 mJ/cm^2 ± 2%, in steps of 25 mJ/cm^2. Clear enhancements in the AZO conductivity and transparency were observed when processed at laser energy densities ≥ 50 mJ/cm^2, while a visible ablation was observed at energy densities ≥ 150 mJ/cm^2. Following the identification of this energy density processing window further investigations were conducted with different number of pulses up to 100.

2.3. AZO thin films characterisation

The essential requirements for TCO applications are low electrical resistivity and high optical transmittance. Thus, this study is focused on these characteristics pre and post ELA application. The