

# Utilization of AZO/Au/AZO multilayer electrodes instead of FTO for perovskite solar cells



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## ABSTRACT

Aluminum (3 at%) -doped ZnO (AZO)/Au/AZO multilayer films with low resistivity and high transmittance are in situ deposited at room temperature via facing-target sputtering for AZO and direct current (DC) sputtering for Au. The optical and electrical properties of multilayer films are compared with conventionally (*on*-axis) sputtered and facing-target sputtered (FTS) AZO films. The FTS samples show better crystallinity than that of *on*-axis sputtered samples, and the lowest value for resistivity is  $7.9 \times 10^{-5} \Omega\text{-cm}$  at an Au layer thickness of 8 nm, which is compared with that of a conventional FTO electrode. The photo-conversion efficiencies (PCE) of the perovskite solar cells based on the AZO multilayer electrodes deposited via FTS and *on*-axis sputtering are 9.5% and 8.2%, respectively. Although the PCE of AZO multilayer-based-solar cells exhibit a lower efficiency than that of the FTO based-devices (~12.3%), the perovskite solar cell performance based on the FTS AZO multilayer electrodes is very attractive for applications to flexible planar-structured solar cells.

## 1. Introduction

Lead halide perovskite solar cells have undergone an increase in power-conversion efficiencies of from 3.8% in 2009 to a certified 20.1% in 2014, which distinguishes them as having one of the most rapid rates of growth in photovoltaic cell performance [1]. Unique properties such as a high absorption coefficient, a tunable band gap, low-temperature processing, and abundant elemental constituents provide numerous advantages for their use in thin-film absorbent materials [2,3]. In addition to high-efficiency and low-cost, however, practical application is the most important prerequisite to be addressed and improved before these can be considered a sustainable energy solution. To achieve perovskite solar cells at an affordable cost, they should be prepared via roll-to-roll processing on a large area of flexible substrates. This requires device architecture that uses an electrode fabricated at low temperature. Most perovskite solar cells are fabricated using fluorine-doped tin oxide (FTO) or indium tin oxide (ITO)-based electrodes, which are expensive and difficult to etch in order to obtain a high level of efficiency [4,5].

Among various transparent conductive oxide (TCO) thin films, nanoscale Ag/Au-based Al-doped ZnO multilayer transparent-conductive oxide thin films have attracted much attention due to their rich storage, low cost, non-toxicity, abundantly available components, and simple methods of synthesis and processing [6–9]. In addition, high transmittance in the near-infrared region of AZO films could be an

advantage for the fabrication of perovskite solar cells. Besides the synthesis of epitaxial and textured AZO film using various deposition techniques [7–16], magnetron sputtering using a ceramic target has shown the most promise in practical industrial preparations, because of its high potential for the formation of uniform coatings over large areas with high reproducibility. However, various sputtering conditions such as the working pressure and the distance and location between the target and substrate strongly affect the structure, and the electrical and optical properties of AZO films. This behavior can be partially attributed to the bombardment of various energetic particles on the growing film [17,18]. Based on reports in the literature, the degradation of the crystallinity in oxide films, and the resultant electrical properties, might be due to bombardment by high-energy particles such as negative oxygen ions on the surface of the film during deposition processes [19,20]. In contrast, ZnO films deposited via a target placed in a horizontal direction to the substrate showed smooth surfaces and high-quality crystal structures [21]. Therefore, the parallel location of a substrate and a target, such as FTS for metal oxide films, is expected to be a key influence on the electrical and optical properties of TCO films [22]. Our group has reported long-term stability in damp heat-treatment for 1000 h and mechanical stability after the severe bending of Ag-based AZO multilayer films on polymer substrates, which has enabled the commercialization of TCO electrodes for flexible perovskite solar cell applications [23].

In the present study, AZO/Au/AZO multilayer films were deposited

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at room temperature onto glass substrates using *on*-axis sputtering and FTS for AZO films. We studied the effect of deposition processes on the electrical and optical properties of multilayer films and finally investigated the performance of perovskite solar cell devices prepared by AZO multilayer bottom electrodes deposited via both methods.

## 2. Experimental details

### 2.1. Preparation of AZO/Au/AZO multilayer bottom electrodes

The AZO/Au/AZO films were deposited onto Corning glass (1737) at room temperature by direct current (dc) sputtering for Au layer using an Au metal target (2 in. diameter with 99.99% purity) and radio-frequency (rf) sputtering for AZO films using two Al (3 at%)-doped ZnO ceramic targets (2 in. diameter) for AZO films. In FTS, two facing AZO targets were placed at a distance of 9 cm. The plasma existed in the space between the surfaces of the targets, while the substrates were apart from the plasma atmosphere. The substrate holder was placed in a horizontal direction far from the targets, also at 9 cm. In the case of *on*-axis sputtering, which is referred to as “the facing position”, the substrates and one AZO target were placed “face-to-face” at a distance of 9 cm, and the substrates were in direct contact with the plasma. In both methods, the Au and AZO layers were in situ-deposited without breaking the vacuum. The glass substrates were ultrasonically cleaned in an ethanol/acetone solution and then rinsed in deionized water. The AZO films deposition via FTS and *on*-axis sputtering were performed under an argon atmosphere with the same target-to-substrate distance of 9 cm. Initially, the sputtering chamber was evacuated to a base pressure of  $5 \times 10^{-3}$  Pa using a turbo molecular pump. The RF power for each target was maintained at 100 W with an Ar flow rate of 10 sccm (standard cc min<sup>-1</sup>) and a working pressure of 0.4 Pa. The Au layer was deposited at a pressure of 0.4 Pa with dc power of 6.5 W. We have confirmed the thickness of Au layer to be 10 nm- using cross-sectional TEM image with color mapping, which can be seen from Fig. S1. Further based on growth rate, the Au layers were deposited for 70, 100, and 120 s for 6, 8, and 10 nm, respectively. In addition, we have optimized the thickness of AZO layers deposited by *on*-axis and FTS method to determine the growth rate. The growth rate of AZO thin films deposited by *on*-axis and FTS was 3 nm/min and 1 nm/min, respectively. Finally, based on the growth rate controlled by deposition time, multilayer thin films were prepared. We have provided the cross-sectional images of AZO thin films deposited via both methods in Supplementary information (Fig. S1).

### 2.2. Fabrication of the perovskite solar cells on AZO/Au/AZO and FTO-coated glass substrates

For deposition of the blocking layer of TiO<sub>2</sub> (*bl*-TiO<sub>2</sub>) via nanocluster deposition (NCD), Ti(O-*i*Pr)<sub>2</sub>(*dibm*)<sub>2</sub>(Ti(Oi-C<sub>3</sub>H<sub>7</sub>)<sub>2</sub>(C<sub>9</sub>H<sub>15</sub>-O<sub>2</sub>)<sub>2</sub>) precursors for a liquid delivery process were dissolved in hexane and used as the sources of titanium at concentrations of 0.075 M. When the chemical sources entered the vaporizer, they were immediately vaporized and carried to the showerhead within the reaction chamber by argon carrier gas at a flow rate of 100 sccm. The oxygen was supplied to the showerhead as a reaction gas at a fixed-flow rate of 100 sccm. The showerhead temperature was maintained at 240–300 °C, while the substrate temperature was maintained at 400 °C for crystallization of the *bl*-TiO<sub>2</sub>. Film thicknesses of 80 nm were estimated using the deposition rate of the TiO<sub>2</sub> (~10 nm/min) and their real thickness was measured via scanning electron spectroscopy (SEM) cross-sectional image. The 300 nm-thick mesoporous TiO<sub>2</sub> (*m*-TiO<sub>2</sub>) layers were deposited onto the *bl*-TiO<sub>2</sub>/AZO/Au/AZO bottom electrode/substrates and *bl*-TiO<sub>2</sub>/FTO/substrates via spin coating at 2000 rpm (revolution per minute) and 3500 rpm, respectively, using the TiO<sub>2</sub> paste (Dyesol 18NR-T) diluted further in methanol at ratio of 1:10 by weight. The *m*-TiO<sub>2</sub> layers were sintered under an air atmo-

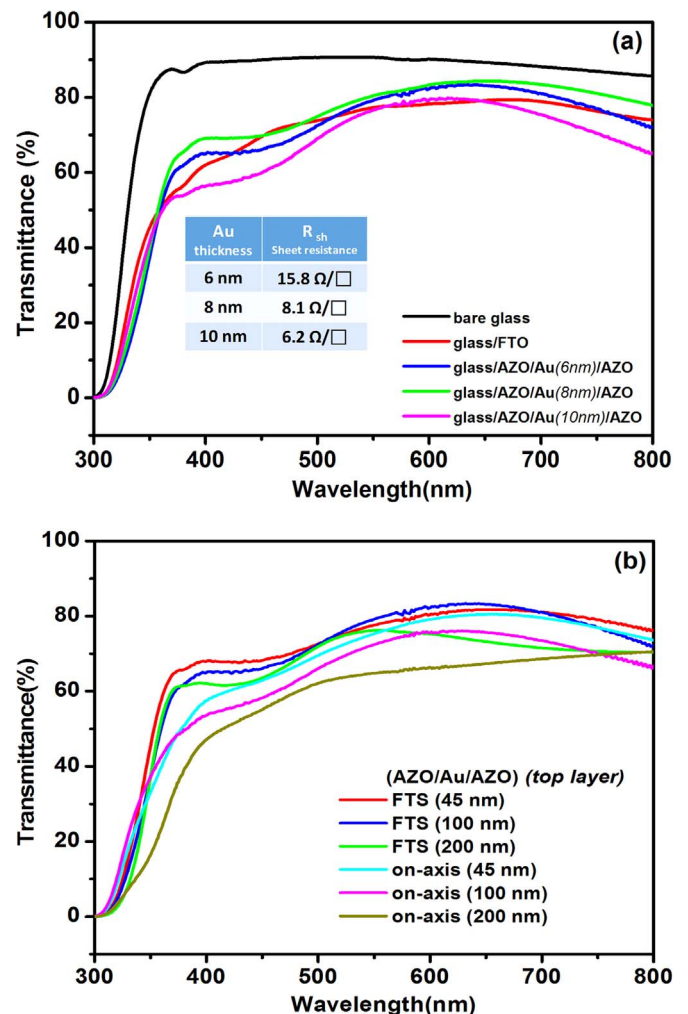


Fig. 1. Optical transmittance spectra for the AZO/Au/AZO films as a function of the (a) Au interlayer thickness, and with (b) different AZO top-layer thicknesses deposited via *on*-axis sputtering and FTS. The inset of (a) shows the sheet resistance versus Au interlayer thickness.

sphere at 450 °C for 1 h.

For preparation of the perovskite dye (CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>), a two-step process was suggested and solutions of 400 mg/mL PbI<sub>2</sub> (Sigma) in N, N-dimethylformamide (DMF) were spin-coated onto the *m*-TiO<sub>2</sub> (300 nm)/*bl*-TiO<sub>2</sub>(80 nm)/FTO and AZO/Au/AZO bottom electrode/substrates, then dried at 80 °C for 15 min. A low concentration of CH<sub>3</sub>NH<sub>3</sub>I solution (2 mg/1 mL) was spin-coated at 2000 rpm for 5 s onto the PbI<sub>2</sub>-coated *m*-TiO<sub>2</sub> layers and then a high concentration of CH<sub>3</sub>NH<sub>3</sub>I solution (40 mg/1 mL) was dropped onto the samples and was maintained for 3–4 min to promote a reaction between PbI<sub>2</sub> and CH<sub>3</sub>NH<sub>3</sub>I to form CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>. The samples were spun at 2500 rpm for 25 s. After spin coating, the samples were annealed under an Ar atmosphere at 100 °C for 1 h via RTA (rapid thermal annealing). After cooling, hole transport layers (HTL) were deposited via spin coating at 4000 rpm for 5 s on the perovskite dye/*m*-TiO<sub>2</sub>/*c*-TiO<sub>2</sub>/electrodes using a hole transport solution, employing a spiro-OMeTAD /chlorobenzene (180 mg/1 mL) solution with the addition of 50 μL Libis (trifluoromethanesulfonyl) imide (Li-TFSI, Sigma)/acetonitrile (170 mg/1 mL) and 20 μL *tert*-butylpyridine (tBP, Sigma). Finally, the 80 nm-thick Au films as a counter electrode were deposited by dc (direct current) sputtering under a working pressure of 0.4 Pa (base pressure of  $5 \times 10^{-3}$  Pa) using a 2-in. diameter gold target. The active area of the cell was approximately 0.25 cm<sup>-2</sup>. After deposition of the Au top electrode, the cells were annealed at 100 °C for 10 min under an air

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