



Au thin films deposited on SnO₂:In and glass: Substrate effects on the optical and electrical properties

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ABSTRACT

We report on a detailed study on the optical and electrical properties of Au films made by sputter deposition onto glass substrates with and without transparent and electrically conducting layers of SnO₂:In. The Au films had thicknesses up to 10.7 nm and hence spanned the range for thin film growth from discrete islands, via large scale coalescence and formation of a meandering conducting network, to the formation of a more or less “holey” film. Scanning electron microscopy and atomic force microscopy demonstrated that the SnO₂:In films were considerably rougher than the glass itself, and this roughness influenced the Au film formation so that large scale coalescence set in at a somewhat larger thickness for films on SnO₂:In than on glass. Measurements of spectral optical transmittance and reflectance and of electrical resistance gave a fully consistent picture that could be reconciled with impeded Au film formation on the SnO₂:In layer; this led to pronounced “plateaus” in the near infrared optical spectra for Au films on SnO₂:In and a concomitant change from such two-layer films having a lower resistance than the single gold film at thicknesses below large scale coalescence to the opposite behavior for larger film thicknesses. Our work highlights the importance of the substrate roughness for transparent conductors comprising coinage metal films backed by wide band gap transparent conducting oxides.

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1. Introduction

Thin films of transparent conductors (TCs) have widespread applications in modern technology. Within the rapidly developing field of green nanotechnology [1], such films are widely used to provide low thermal emittance and solar control for architectural windows, and the films can be used to insert and extract electricity in photovoltaic and electrochromic devices [2]. Other applications regard numerous aspects of transparent electronics [3,4].

There are two main classes of TCs: those based on heavily doped wide band gap oxide semiconductors, such as SnO₂:F, In₂O₃:Sn (ITO), ZnO:Al (AZO) and ZnO:Ga (GZO), and those based on extremely thin films normally of coinage metals such as Cu, Ag and Au [2]. Each of the classes has their pros and cons. Thus Ag-based coatings can be used only in protected environments, such as in gas filled spaces between two panes in a modern window, whereas Au films are more corrosion resistant and allow uses in laminated electrochromic devices [5]. The oxide-based coatings must be thicker than the metallic ones by at least an order of magnitude to achieve comparable properties with regard to electrical resistance and thermal emittance, and the associated long thin film deposition times – as well as the frequent

use of In-based films – tend to make the oxide-based films excessively expensive for applications on commodity products.

The increasing applications of TCs have led to a recent upsurge of interest in this type of materials [2,6]. In particular, there is much contemporary work on TCs based on coinage metal films whose optical properties are boosted by transparent and antireflecting layers with high refractive indices; if these layers are electrically conducting the antireflection can come with an improvement of the electrical properties. Considerations such as these have led to numerous studies during the past few years. The breadth of these endeavors may be appreciated by looking at the number of multi-layer systems that were reported on during 2009; they include ZnO/Ag/ZnO [7], ZnO/Ag/AZO/ZnO [8], AZO/Ag/AZO [9–12], GZO/Ag/GZO [11,13], ZnS/Ag/WO₃ [14], ITO/Ag/ITO [15–20], MoO₃/Ag/MoO₃ [21], AlSiN/AgMgEu/AlSiN/AgMgEu/AlSiN [22], ZnO/Cu/ZnO [23], AZO/Cu/AZO [24], ITO/Cu/ITO [19], ZnO/Al/ZnO [25], ITO/Pt/ITO [26], ITO/Ni/ITO [27] and TiO₂/W/TiO₂ [28].

We have recently investigated thin films of TiO₂/Au/TiO₂ with a view to their applications in electrochromic foil technology [5], and in this present paper we describe recent progress towards the use of Au-based films in such and other devices. A critical issue is the growth of the Au film and how this growth is influenced by, in particular, the nanostructure and electrical properties of the underlying substrate. To that end we report here on the growth of Au films on indium doped tin oxide, SnO₂:In. The choice of this oxide – which has received

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comparatively scant attention in the past [29–31] – has two main reasons: (i) it can be either *n*-doped and *p*-doped depending on the deposition conditions, notably the substrate temperature, and (ii) it serves as an analogue to the much more studied tin doped indium oxide [32] and hence will enable us to compare the growth of Au on SnO₂:In and on In₂O₃:Sn in future work. Other oxides that can be *n*-doped as well as *p*-doped depending on the deposition or post treatment conditions include ZnO:P [33], ZnO:As [34] and MgZnO [35].

2. Film deposition

Thin films of Au and SnO₂:In were deposited by DC magnetron sputtering onto unheated glass plates in a versatile deposition system with a base pressure of $\sim 2.7 \times 10^{-5}$ Pa. Targets of 99.99%-pure Sn_{0.92}In_{0.08} and Au were positioned 13 cm above the substrate. All depositions were made at room temperature.

Films of SnO₂:In were prepared by reactive sputtering in argon and oxygen at an Ar/O₂ mixing ratio of 1.7 maintained via mass-flow controlled gas inlets. The power to the sputter plasma lay in the $78 < P_{\text{SnIn}} < 125$ W range, and its pressure was kept at $0.64 < p_{\text{SnIn}} < 0.71$ Pa. The films had thicknesses of $49 < d_{\text{SnIn}} < 180$ nm as recorded by scanning electron microscopy (SEM) using a LEO 1550 FEG instrument with in-lens detector and optical control via point-by-point fitting.

Gold films were deposited by sputtering in argon with $P_{\text{Au}} \approx 50$ W and $p_{\text{Au}} \approx 0.80$ Pa onto glass substrates with and without SnO₂:In layers. The mass thickness of the Au films is an essential parameter and d_{Au} s being 1.6, 5.4, and 8.1 nm were measured by Rutherford backscattering spectroscopy (RBS) at the Tandem Laboratory of Uppsala University. In practice we recorded the mass thickness of a thick Au film – made under constant deposition conditions – by RBS and determined smaller d_{Au} s by scaling with time.

3. Film characterization

3.1. Optical and electrical properties of SnO₂:In films on glass

Spectral normal transmittance $T(\lambda)$ and near-normal reflectance $R(\lambda)$ were recorded for $0.3 < \lambda < 2.5$ μm using a Perkin-Elmer Lambda 900 double-beam spectrophotometer with a 15-cm-diameter reflectance accessory and a Spectralon reflectance standard. Spectral absorbance was then obtained by $A(\lambda) = 1 - T(\lambda) - R(\lambda)$. Electrical sheet resistance R_{\square} was measured between sputter deposited Au contacts for all samples, and resistivity ρ_{SnIn} was calculated for the SnO₂:In films. Considering the deposition conditions, the conductivity was taken to be of *n*-type [29,30].

Fig. 1 reports $T(\lambda)$, $R(\lambda)$ and $A(\lambda)$ for two SnO₂:In films prepared with the shown values of P_{SnIn} and p_{SnIn} and having the stated d_{SnIn} s and ρ_{SnIn} s. Optical interference plays an important role and thicknesses could be chosen so as to maximize the luminous transmittance in the $0.4 < \lambda < 0.7$ μm range.

3.2. Nanostructural determinations

Fig. 2 shows SEM images, taken with the instrument mentioned above, for Au films having three different d_{Au} s deposited onto a substrate with a SnO₂:In film characterized by $d_{\text{SnIn}} \approx 123$ nm and $\rho_{\text{SnIn}} \approx 26$ m Ωcm and simultaneously deposited onto uncoated glass.

Irrespective of the substrate, the Au films go through the expected growth phases [5,36], starting with discrete Au islands at the smallest thicknesses, then entering a large scale coalescence (LSC) phase at $d_{\text{Au}} \approx 5.4$ nm where the gold film has an electrically contiguous and meandering structure, and then approaching a uniform film with voids. It is clear from Fig. 2 that the Au films grow differently on glass and on SnO₂:In. Thus the films with $d_{\text{Au}} \approx 5.4$ nm is more continuous for the former case, and the same tendency prevails at $d_{\text{Au}} \approx 8.1$ nm as

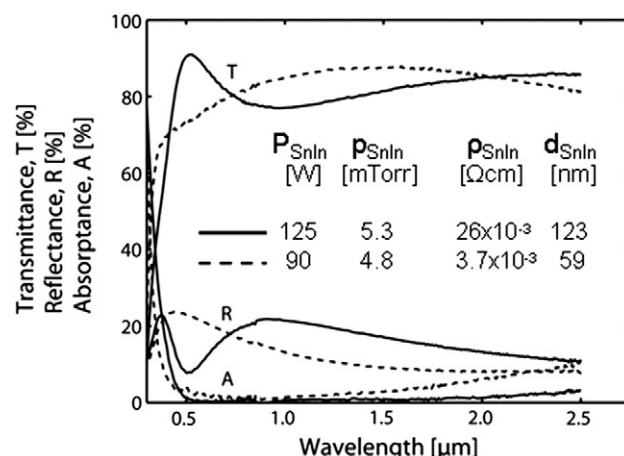


Fig. 1. Spectral optical properties for SnO₂:In films prepared with the shown values of power (P_{SnIn}) and pressure (p_{SnIn}) in the plasma and having the stated resistivity (ρ_{SnIn}) and thickness (d_{SnIn}).

evident from the smaller voids for the film on glass. Specifically, the film with $d_{\text{Au}} \approx 5.4$ nm had an uncoated area fraction of 32% for deposition onto glass and 38% for SnO₂:In on glass.

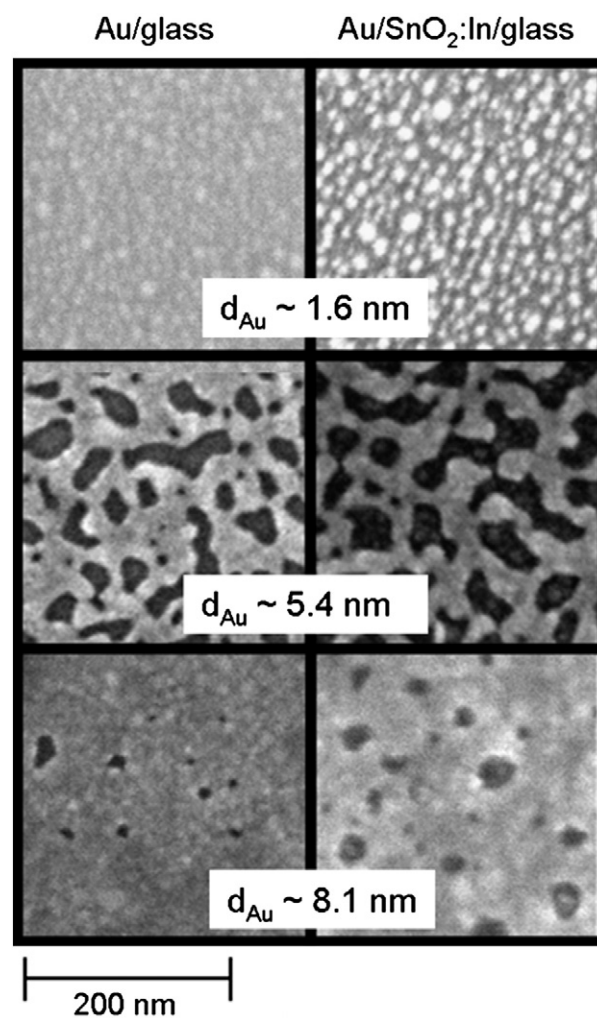


Fig. 2. Scanning electron micrographs for Au films deposited onto glass and SnO₂:In and having the shown thicknesses. The bright regions represent Au. The weak contrast for the thinnest gold film on glass is a consequence of the lack of electrical conductivity of the studied film.

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