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Thin gold films on SnO₂:In: Temperature-dependent effects on the optical properties

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1. Introduction

ABSTRACT

Gold films with thicknesses of 5 ± 0.5 nm were sputter deposited onto SnO₂:In-coated glass kept at different temperatures up to 140 °C, and similar films, deposited onto substrates at 25 °C, were annealing post treated at the same temperatures. Nanostructures and optical properties were recorded by scanning electron microscopy and spectrophotometry in the 0.3 to 2.5 µm wavelength range, respectively. Annealing had a minor influence on the optical transmittance despite significant changes in the scale of the nanostructure, whereas deposition onto substrates heated to 140 °C yielded granular films with strong plasmon absorption of luminous radiation. These results are of considerable interest for optical devices with gold films prepared at elevated temperature or operating at such temperature.

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Thin films combining optical transparency and electrical conductivity have many uses [1,2] and are key elements in a number of "green" nanotechnologies, including fenestration for energy efficient buildings, photovoltaic cells, and light-emitting diodes [3]. Coinage-metal-based transparent conductors are used for large-area applications and can combine excellent optical and electrical properties, sufficient inertness for applications in corrosive environments [4], and high-rate deposition. Typically a luminous transmittance exceeding 80% and a resistance per square below 20 Ω are desired for these applications.

Thin gold films have been of interest for "green" nanotechnologies during many years, and early work related to energy efficient windows date back to the 1950s [5]. Particularly detailed studies were made in the 1980s [6], and the interest in high-performance transparent conductors and infrared reflectors continues still today [3] not only for Au films but also for films based on Ag [7–12] and Cu [13,14].

An earlier paper of ours [15] explored the optical and electrical properties of thin Au films sputter deposited onto SnO₂:In; in the present sequel we present further data on this combination of materials and report on the effect of elevated substrate temperatures during and after the deposition. In particular we demonstrate the characteristic structural, optical and electrical properties occurring in Au films that are either deposited onto SnO₂:In heated to a certain temperature or deposited onto such films at room temperature and subsequently annealing post treated at nominally the same elevated temperature.

2. Sample preparation

Thin films of SnO₂:In and Au were prepared by DC magnetron sputtering as described in detail before [15]. SnO₂:In was deposited reactively from a 99.99-%-pure target of Sn_{0.92}In_{0.08} onto unheated glass; the film thickness was ~123 nm, as recorded by scanning electron microscopy (SEM) and optical interference analysis, and the surface roughness was ~1.5 nm as inferred from atomic force microscopy. The gold films were deposited onto the SnO₂:In layer from a 99.99-%-pure target with the substrate temperature τ_s kept at a constant value within the range $25 < \tau_s < 220$ °C. The Au mass thickness lay at 5 ± 0.5 nm as determined by Rutherford backscattering spectroscopy. Double-layer Au/SnO₂:In films, deposited with the substrate at room temperature, were annealing post treated in vacuum at a constant temperature τ_a in the range $45 < \tau_a < 140$ °C for 3.5 h.

The samples to be discussed below are of two types and were made by first coating $2.5 \times 5.0 \text{ cm}^2$ glass substrates with SnO₂:In and subsequently cutting them into two halves. For the *first* type of samples, one of the halves was coated with Au without substrate heating while the other was coated with the substrate kept at τ_{s} ; the deposition conditions for these Au films were as identical as possible. For the *second* type of samples, both of the halves were coated with Au without substrate heating and at the same time, and one of the halves was then heat treated at τ_{a} .

3. Results

3.1. Characterization techniques

* Corresponding author. Tel.: +46 18 4717783; fax: +46 18 4713270. *E-mail address*: pia.lansaker@angstrom.uu.se (P.C. Lansåker). Structural, optical, and electrical properties of the Au/SnO_2 :In films were recorded as in our earlier work [15]. Microstructure was

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determined by SEM using a LEO 1550 FEG instrument with in-lens detector. Spectral normal transmittance $T(\lambda)$ was determined in the 0.3 < λ < 2.5 µm wavelength range by use of a Perkin-Elmer Lambda 900 double-beam spectrophotometer equipped with a 15-cm-diameter integrating sphere. Electrical sheet resistance R_{\Box} was measured between ~110-nm-thick Au contacts sputter deposited onto the Au/SnO₂:In samples. The contacts were made under conditions so that the temperature did not exceed the lowest one at which the thin Au samples were prepared.

3.2. Nanostructure

Nanostructural features are reported in Fig. 1(a) and (b) for samples prepared with τ_s and τ_a being 45 °C, 105 °C, and 140 °C, respectively. These features looked similar irrespective of the detailed position on the sample. The six non-heated samples, presented in the left-hand panels, show reassuringly identical nanostructures with the Au films having a structure characterized by elongated and irregular voids. Deposition at $\tau_s = 45$ °C or heat treatment at $\tau_a = 45$ °C did not lead to any noticeable change in the microstructure. At $\tau_s = 105$ °C the Au film displays a contiguous meandering nanostructure, and this is the case also for $\tau_a = 105$ °C but the characteristic dimension of the structure is larger in the latter case. Setting $\tau_s = 140$ °C yields a qualitatively different nanostructure with well characterized and rather regular Au islands separated by voids, whereas at $\tau_a = 140$ °C the film still keeps its contiguity though at a still larger dimension than for $\tau_a = 105$ °C. The micrograph for

 $\tau_a = 140$ °C shows evidence for a grainy structure within the voids; this feature originates from the rough top surface of the SnO₂:In film.

3.3. Optical transmittance

Fig. 2(a) and (b) shows $T(\lambda)$ for the two-layer films whose nanostructures were displayed in Fig. 1. The data for the as-deposited Au films look practically identical, as they should since their nanostructures were indistinguishable. The transparency drops at short wavelengths due to the characteristic inter-band absorption in gold and, at the smallest wavelengths, also as a result of absorption in the SnO₂:In and glass (cf. Fig. 1 in Ref. [15]). We first make some qualitative observations: The samples having voided structures—as evident from the micrographs in Fig. 1 for $\tau_s = \tau_a = 45$ °C, $\tau_s = \tau_a = 105$ °C, and $\tau_a = 140$ °C—all display transmittance spectra with a characteristic plateau in the near-infrared, whereas the film with a clear-cut island structure, seen in Fig. 1 for $\tau_s = 140$ °C, yields a distinct minimum in the transmittance at $\lambda \approx 0.75 \,\mu$ m. The latter feature was observed also in samples prepared with $\tau_s = 220$ °C.

Fig. 2(a) refers to Au films deposited at different values of τ_s . The film produced at $\tau_s = 45$ °C has the same optical properties as for the as-deposited film, while the film made at $\tau_s = 105$ °C displays a significantly widened near-infrared plateau, which appears to extend even for $\lambda > 2.5 \,\mu\text{m}$. At $\tau_s = 140$ °C, the transmittance spectrum looks radically different, as noted above.

The results for samples treated at different magnitudes of τ_{a} , shown in Fig. 2(b), are rather similar to each other irrespectively of



Fig. 1. Transmission electron micrographs for Au/SnO₂:In two-layer films. The left-hand images in panels (a) and (b) refer to as-deposited Au films, and the right-hand images in panels (a) and (b) refer to Au films having the shown values of the substrate temperature during deposition and of the annealing post treatment temperature, respectively.

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