GISAXS Analysis of gold thin films prepared by evaporation at glancing angles

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In previous works we have study by GISAXS the microstructrure of oxide thin films prepared by evaporation at glancing angles (GLAD), as well as metal oxide composite thin films grown by deposition or infiltration of metal nanoparticles on the surface or in the bulk of the oxide films acting as hosts [1-2]. Due to their original and, some times, exotic microstructure GLAD thin films have been proposed to develop new nanostructures materials of interest for a large variation of applications [3-4]. This works addresses a basic problem in relation to the growth of metal oxide thin films, namely unravelling the transition in microstructure of the gold thin films prepared in a GLAD configuration when the deposition angle is varied from 60º to 87.5º. Besides GISAXS, the films have been studied by a large variety of techniques including SEM and UV-vis reflectometry. Their growth has been also simulated by means of Monte Carlo simulations. In general, it is found a very good correlation between the microstructure detected by SEM and accounted for by the MonteCarlo simulations and the information provided by the GISAXS analysis of the films.

Figure 1 shows a series of SEM images taken for the films prepared at 87.5º to 70º deposition angles. These images clearly show an evolution of the film microstructure from one at 70º, the lowest deposition angle, where gold forms well interconnected stripes to another at 87.5º, the highest deposition angle, where separated gold nanocolumns are clearly devised in the image. Parallel to this evolution in microstructure, quite interesting changes in the optical properties of the films were also observed by UV-vis reflectometry. According to this technique the films behave as a rather reflecting system when they are prepared at 70º to Plasmon like surfaces at 87.5º where the typical surface plasmon resonance bands of gold nanoparticles are clearly visible in the spectra.

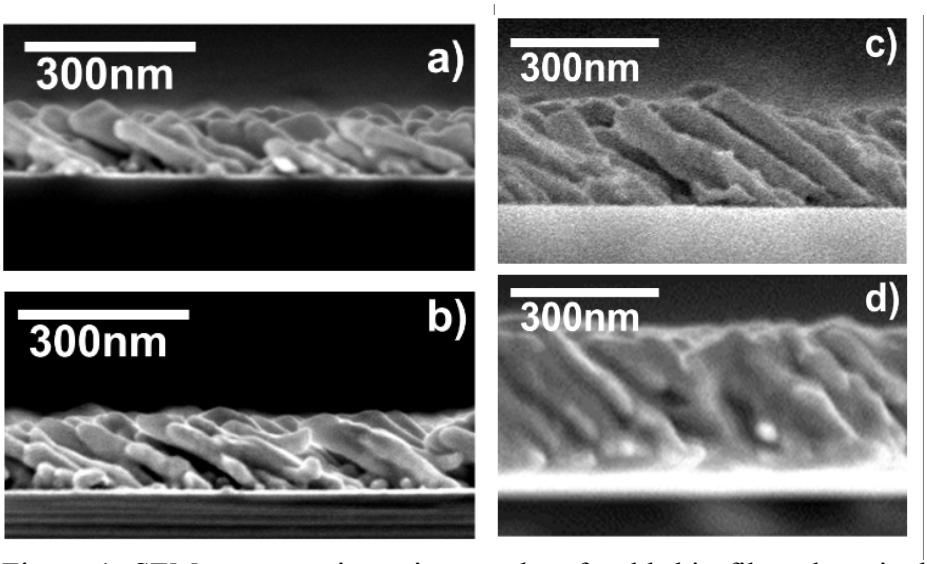

Figure 1. SEM cross section micrographs of gold thin films deposited by evaporation at different angles: a) 87.5º, b) 85º, c) 80º and d) 70º.

To clearly demonstrate this transition in the microstructure of the films, we also carried out a thorough characterization by GISAXS, a technique that provides an averaged information over the entire area examined by the X-ray beam. In addition, it may provide quantitative information about

the existence of certain critical correlation lengths that can be of much use for tailoring the fabrication of other type of materials.

GISAXS experiments were performed at the BW4 beamline (HASYLAB, Hamburg) using a wavelength of λ=0.138 nm and a sample-to-detector distance of 2.175 m. A moderate microbeam focusing was achieved using beryllium refractive lenses (beam size 42x22 μm^2). The scattering signal was recorded with a 2D detector (MAR CCD camera with 79x79 µm pixel size).

Figure 2 shows a series of selected GISAXS patterns corresponding to gold thin films prepared at evaporation angles of 70º, 80º, 85º and 87.5º. For each case, the patterns shown correspond to two different orientations of the layers, one with the nanostructured features parallel to the beam and another perpendicular to it. In this latter case it is quite clear the evolution of a significant asymmetry in the distribution of scattered intensity of the beam. In line with previous assignments of similar asymmetric GISAXS patterns obtained with oxide thin films [5], the evolution of asymmetric features in the patterns can be rationalized by assuming the progressive formation of isolated nanocolumns during the growth of the films at high incident angles.

This investigation, together with the modelling of the film growth should permit to get critical information about the processes controlling the growth of GLAD thin films and other multilayers with quite new and appealing properties.

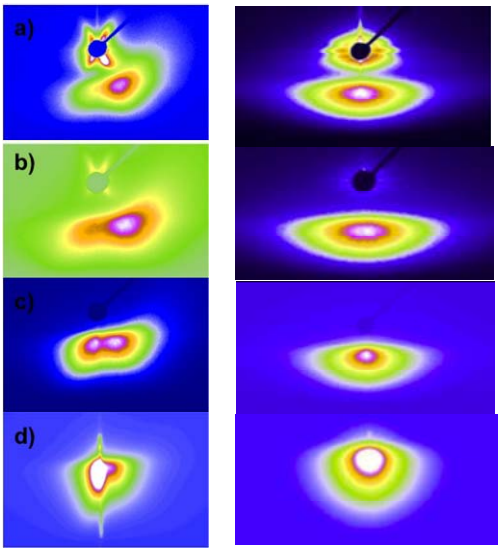

Figure 2. GISAXS patterns for 300 nm thin films of gold deposited by GLAD at increasing evaporation angles: a) 87.5º, b) 85º, c) 80º and d) 70º. Left) patterns obtained with the beam perpendicular to the plane containing the tilted nanonoculumns. Right) patterns obtained in the parallel direction to the tilting.

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