ROBL-CRG	**Experiment title:** **The evolution of texture in thin-film silver during growth and annealing – Part II**	**Experiment number**: **20_02_609**
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Report:

The nanostructural evolution during heat treatments of DC magnetron-sputtered Ag films, deposited at room temperature at different substrate bias voltages, was experimentally studied. The thermal stability of the nanostructure was sensitively dependent on the substrate bias voltage. ***Increasing the bias voltage resulted in significantly lower rates of grain growth,*** which we ascribe mainly to the formation of Ar bubbles. Furthermore, ***the grain size in the as-deposited films decreased with increasing bias voltage while the width of the one-dimensional pole figures*** (measured *ex situ*) increased.

EXPERIMENTAL

The nanocrystalline (nc) Ag thin films were synthesized by DC magnetron sputtering in the ROBL *in-situ* growth chamber [1]. The relevant parameters had been: base pressure $\approx 7\times10^{-5}$ Pa; Ar (purity 99.9996%) was used as sputter gas at a pressure of 0.5 Pa and a flow rate of 4 sccm; the substrates, mounted on a resistive heater, were Si(001) wafers (14×14 mm^2) with a 100 nm amorphous SiO_2 layer on top. The films were deposited at RT under different bias voltages (-50 V, -150 V, -250 V). The x-ray wavelength had been λ = 0.950 Å (13.051 keV).

During film deposition and subsequent annealing, Bragg-Brentano θ-2θ scans were carried out repeatedly (only planes parallel to the film surface probed). The integrated peak intensities reveal information about the texture and crystallinity, and the exact positions of the Bragg peak yield information about the out-of-plane lattice distance. From the shape and width of the diffraction peaks, out-of-plane average grain sizes and microstrain (rms strain) can be obtained. The diffraction peaks were fitted using a pseudo-Voigt peak profile, from which Gaussian β_G and Lorentzian β_L constituents to the total integral breadth were calculated according to a method described by de Keijser *et al*. [2]. The Bragg-Brentano peak profiles were found to be close to purely Lorentzian, i.e. the microstrain was negligible.

RESULTS

The table gives results obtained from *in-situ* Bragg-Brentano x-ray diffractograms shown in Fig. 1 of three Ag films. Tabulated are the integrated intensities $I_{(111)}$, the ratio $I_{(111)}/I_{(002)}$, and the (111) grain size for the as-deposited films and after 80 min. annealing at 81°C (for comparison, $I_{(111)}/I_{(002)}$ = 2.5 for a Ag powder).

Bias (V)	$I_{(111)}$ as deposited	$I_{(111)}$ annealed at 81°C	$I_{(111)}/I_{(002)}$ as deposited	$I_{(111)}/I_{(002)}$ annealed at 81°C	Grain size (nm) as deposited	Grain size (nm) Annealed at 81°C
-50	5500	9000	12.6	16.4	35	44
-150	3200	3700	9.9	9.7	18	22
-250	3300	3500	14.2	13.6	15	16

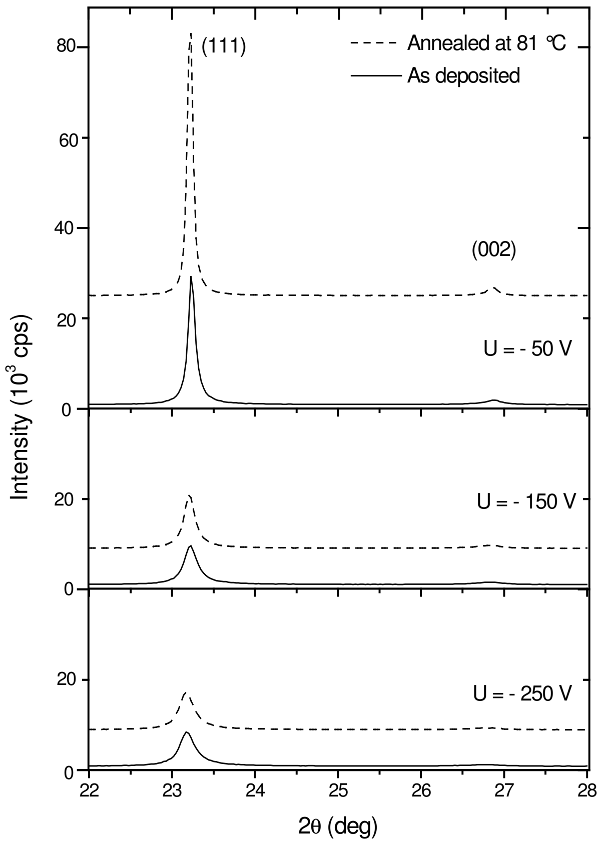

Fig. 1: *In-situ* Bragg-Brentano x-ray diffractograms obtained for three Ag films deposited with various bias voltages, recorded in the as-deposited state (RT) and after 80 min annealing at 81°C, resp. Top: -50 V; middle: -150 V; bottom: -250 V. The film thicknesses are 114, 96 and 93 nm, resp.

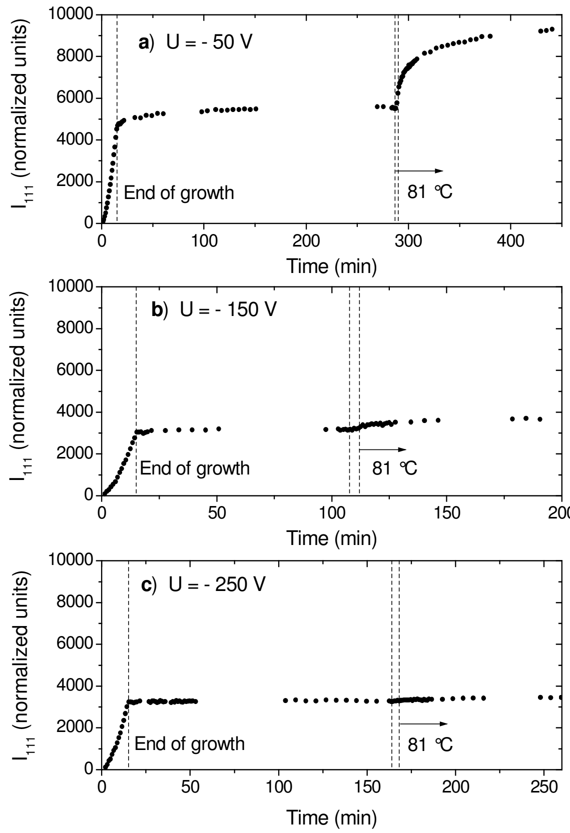

Fig. 2:. Integrated intensities of Bragg-Brentano Ag(111) diffraction peaks recorded as a function of time after start of film growth. Three films deposited at different bias voltages: (a) -50 V, (b) -150 V and (c) -250 V are shown. The film thicknesses are 114, 96 and 93 nm, resp. After a growth period of 15 min, the films were kept some hours at RT before the temperature was raised to 81°C.

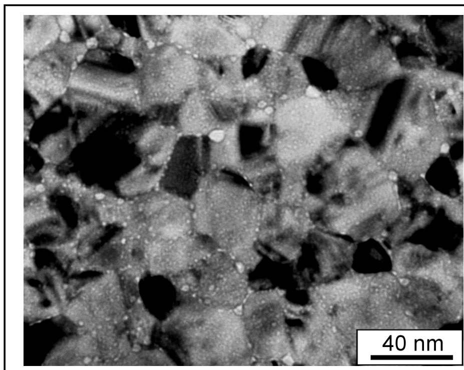

Fig. 3:. Plan-view, bright-field under-focused image of a film as deposited with -250 V bias. ***Ar bubbles*** are observed as small bright spots. Some decorate the grain boundaries while others are found in the interior of the grains. A few Ar bubbles were also observed in the films grown with -150 V bias, while in -50 V biased films no bubbles were detected. Ar bubbles were not observed in previously studied Cu films [3]. Finally, focused, plan-view bright-field micrographs (not shown) from all films revealed ***twinning***.

Grain growth and texture changes decreased with increasing bias voltage. For ***Cu*** [3] this was explained by ***decreasing defect concentrations*** (larger bias results in more energetic ion bombardments of the growing film, which, in turn, make the surface atoms more mobile, whereby the defect concentration in the form of vacancies decreases). As a result, the driving force for grain growth due to recrystallization also decreases. In the case of ***Ag***, however, the build-up of Ar atoms, which formed ***Ar bubbles***, is ***dominating*** the reduction of the grain growth (some of the Ar bubbles were spread over the interior of the grains, slowing down the recrystallization process, while others decorated the grain boundaries, reducing the grain boundary mobility due to pinning).

[1] W. Matz, N. Schell, W. Neumann, J. Bøttiger, and J. Chevallier, Rev. Sci. Instrum. **72** (2001) 3344.
[2] Th. De Keijser, J.I. Langford, E.J. Mittemeijer, and A.B.P. Vogels, J. Appl. Cryst. **15** (1982) 308.
[3] N. Schell, K. Pagh Andreasen, J. Bøttiger, and J. Chevallier, Thin Solid Films *in press* (2004).