

 ROBL-CRG	<b>Experiment title:</b> <i>In-situ x-ray diffraction during reactive magnetron sputter deposition of nanocrystalline metals (nc Au)</i>	<b>Experiment number:</b> <b>20_02_063</b> 20_02_604
<b>Beamline:</b> BM 20	<b>Date of experiment:</b> from: 02.10.2002                      to: 10.10.2002	<b>Date of report:</b> 02.12.2002
<b>Shifts:</b> 25	<b>Local contact(s):</b> Dr. Norbert Schell	<i>Received at ROBL:</i> 02.12.02
<b>Names and affiliations of applicants (* indicates experimentalists):</b> * N. Schell, ROBL-CRG, Germany * J. Bøttiger, University of Aarhus, Denmark		

## Report:

Nanocrystalline materials with their extremely small grain sizes and correspondingly large volume fraction of atoms located at grain boundaries have unique properties and thus offer a great potential for industrial applications [1]. Magnetron sputtering has turned out to be a versatile technique for their deposition in thin films. In order to tailor the films for specific applications, the dependence of the nanostructure and the mechanisms that control its formation and evolution on the deposition parameters has to be established.

Gold was chosen as a nanocrystalline model material (no oxidation), and we used it for the experimental study of the evolution of the nanostructure during growth and after subsequent annealing. In continuation of earlier work (see **Experimental Report 20\_02\_051 B**) this report describes the experimental studies at ROBL of the *texture evolution of magnetron-sputtered gold films during the non-epitaxial growth and subsequent heat treatments*.

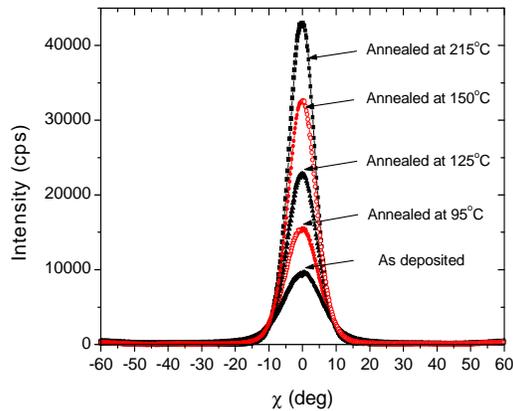
## EXPERIMENTAL

The **deposition chamber** is described in detail in **Ref. 2**. The incident *x-rays* were monochromatized to 13.120 keV /  $\lambda = 0.945 \text{ \AA}$ , and mainly one scattering geometry was used: **Bragg-Brentano** large-angle scattering (XRD) giving the out-of-plane lattice strain, grain sizes and microstrain (lattice defects). To get the *orientation distribution of the dominating (111) grains* (with (111) planes forming a small angle to the film surface),  $\chi$  scans were carried out ( $\theta$ ,  $2\theta$  fixed for Au(111) and sample tilt measured with tilt axis lying both in the film surface and in the scattering plane). As a quantitative measure of the orientation distribution (the texture), we used the widths (FWHM) of the  $\chi$  scans.

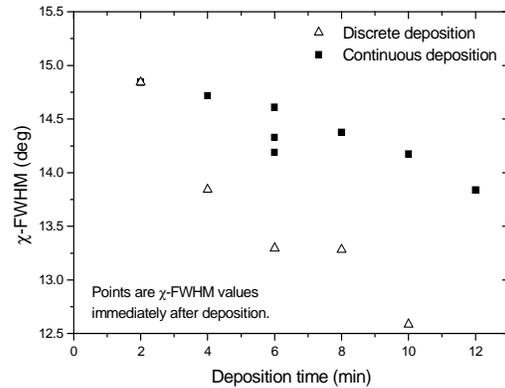
Further experimental parameters were: base pressure  $5 \times 10^{-4}$  Pa, target material Au, sputter gas Ar (99.9996%) at a pressure of 0.6 Pa. The magnetron (only one at the time) was run at a dc power of 10 W, resulting in a *deposition rate of approximately 1.3 Å/s* on Si(100) with a 180 nm amorphous oxide layer on top. The temperature varied from room temperature (deposition) up to 220°C (annealing), a negative bias voltage of -30 V was applied.

## RESULTS

Fig. 1 shows results of such  $\chi$  scans. The *orientation distributions narrowed with increasing annealing temperature*, i.e. the angles between the (111) planes and the film surface became smaller. This is probably the result of small rotations of the individual (111) grains, but recrystallization may also play a role. The development of the *orientation distribution of the (111) grains during growth at room temperature* with  $-30$  V bias voltage is displayed in Fig. 2. The FWHM of  $\chi$  scans is shown as a function of the deposition time. Also shown is the FWHM *versus* deposition time, where the growth was interrupted at regular intervals to allow for texture changes of only thermal origin. As for annealing, the orientation distribution of (111) grains also narrowed during the short times of growth.



**Fig. 1:** Au(111)  $\chi$  scans from 750 Å Au films deposited at RT with a bias voltage of  $-30$  V. The five scans correspond to measurements of a film as deposited and four films *annealed* for one hour at various temperatures.



**Fig. 2:** The development of the FWHM of  $\chi$  scans *during growth* (filled squares), and as a *function of film thickness* (i.e. deposition time), where the growth was interrupted at regular intervals to allow for annealing (open triangles).

During annealing at higher temperatures a clearly increased rate of change of the  $\chi$ -FWHM was seen. From an Arrhenius plot  $-\log(\text{initial rate of change of the FWHM})$  *versus* the reciprocal of the temperature – an *activation energy* was obtained *for the change in texture*,  $Q = 0.68 \pm 0.08$  eV.

Furthermore, a surprisingly small *activation energy of grain growth* of  $0.25 \pm 0.02$  eV was found.

During the first few minutes of growth, the *compressive film* stress was strongly reduced due to a tensile contribution arising from the coalescence of the initially formed islands. *During annealing, stress relaxation* was also observed.

The parts of the studies which include the development of the peak area of the Au(111) diffraction peak, the grain size, the microstrain and the lattice constant have been submitted for publication, while a manuscript dealing with the studies of the development of the texture is in preparation.

- [1] C. Suryanarayana, Inter. Mater. Rev. **40** (1995) 41.
- [2] W. Matz, N. Schell, W. Neumann, J. Bøttiger, and J. Chevallier, Rev. Sci. Instrum. **72** (2001) 3344.