ROBL-CRG	**Experiment title:** **Thin film growth studies by *in situ* x-ray diffraction**	**Experiment number**: 20_02_038 EU-M02
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Report:

During growth, the **microstructural development** – especially the change in texture with film thickness – of **TiN films**, which were deposited by use of magnetrons, was studied. The films were grown in a vacuum chamber that was equipped with two magnetrons and mounted on the goniometer located in MRH of ROBL. Kapton windows enabled ***in situ* x-ray diffraction and reflection** to be carried out to follow the microstructure as a function of film thickness. With the deposition parameters that were chosen, we observed a crossover – grains with a (002) plane parallel with the film surface dominated at small thickness, while, at larger thickness, (111) grains dominated. Recrystallization was identified as a mechanism that controlled this texture development. The driving force for change of orientation of the individual grains arised from minimalisation of the sum of the surface energy and the strain energy of the individual grains.

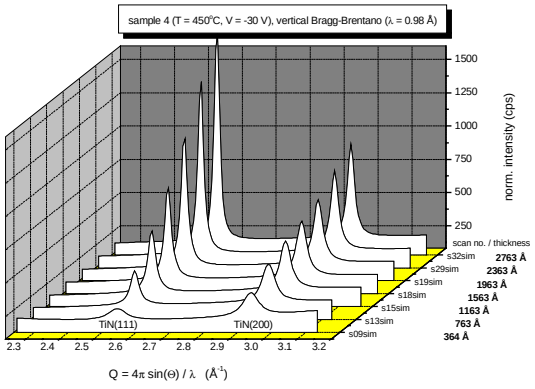
EXPERIMENTAL: The **growth chamber** (together with the detailed **scattering geometry** and the quality of the data, like intensity, resolution, background, which can be obtained with the set-up) is described in detail in Ref. 1. The **magnetrons**, commercially available from AJA International, are placed at a distance of 100 mm from the substrates and tilted 30 degrees away from the substrate normal. To avoid cross contamination of the two targets, each with a diameter of 1 inch, chimneys are mounted on the magnetrons. Air-pressure-controlled shutters are placed in front of the chimneys. The base pressure was about 2×10^{-5} Pa. The reactive sputter gas was a mixture of Ar (99.9996%) and N_2 (99.99990%) with the ratio 4:1, with a total gas pressure of 0.3 Pa. Only one magnetron was used at the time. It was run at a dc power of 80 W. The deposition rate was about 1.4 Å/s. The substrates were silicon wafers with a 1000 Å amorphous oxide layer on top. A resistive heater was mounted below the substrate so the temperature could be varied from room temperature up to 700 ^{0}C. The temperature was measured by use of a thermocouple. A negative bias voltage could be applied to the substrate.

The deposition chamber was mounted on the six-circle goniometer in MRH. The incident x-rays were monochromatized to 12.651 keV (λ = 0.980 Å). To study the growth of TiN films ***in situ***, three different scattering geometries were used:

1. Vertical Bragg-Brentano large-angle scattering (**XRD**). Such measurements reveal the texture. In addition, from the exact positions of the Bragg peaks, information on the out-of-plane lattice strain is obtained, and from the widths and shape of the peaks, out-of-plane grain sizes and microstrain (lattice defects) are obtained.
2. Grazing incidence and grazing exit in-plane large-angle scattering (**GIXS**). With an incident angle of 0.2^0, the calculated penetration depth of the x-rays was about 100 Å, assuming a mass density of TiN of 5.43 g/cm^3. Crystallographic planes perpendicular to the surface are identified and, from the positions and widths of the Bragg peaks, in-plane lattice parameters (strain), grain sizes and microstrain are calculated.
3. Low-angle **specular reflectivity**. The film thickness and information on surface roughness is obtained.

The deposited films were also checked by x-ray diffraction carried out ***ex situ*** with a Philips powder diffractometer using standard Bragg-Brentano geometry and CuK_α radiation which was filtered with a monochromator in front of the detector. The film composition was obtained by **Rutherford backscattering** spectrometry with 2.0 MeV He^+ and a scattering angle of 161°. Electron diffraction patterns and micrographs were obtained by use of a Philips CM20 **transmission electron microscope**.

We studied the growth of **five different TiN films** deposited with different deposition parameters: two were grown at a substrate temperature of **250 ^{0}C** and a bias voltage of **–30 V**, two with a bias voltage of **–30 V** and substrate temperatures of **350 ^{0}C and 450 ^{0}C**, respectively. The fifth film was grown with bias voltage **–60 V** at temperature **350 ^{0}C**. With regular intervals during growth, the deposition was interrupted, and the film was characterised by XRD, GIXS and low-angle specular reflectivity.



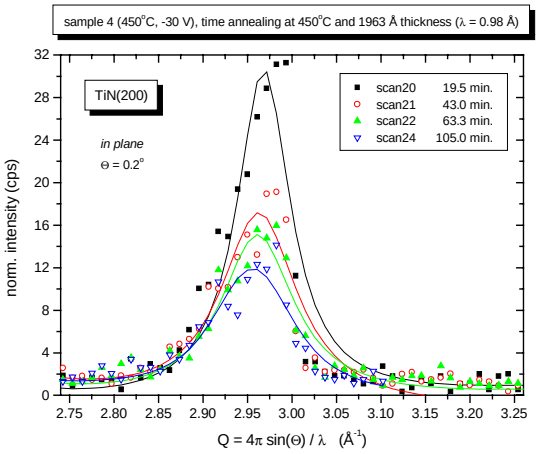


Fig. 1 (left: **crossover in vertical BB geometry**) and **Fig. 2** (right: **recrystallization of TiN(002) in GIXS** with incidence angle 0.2^0) are examples of the obtained data.

SUMMARY: Recrystallization was unambiguously identified as the **dominant mechanism that controlled the development of the texture**. The data were consistent with the driving force for the change of orientation of the grains that was suggested by **Pelleg *et al.*[2]**. ***The grain size increase with film thickness during growth was not due to normal grain growth but was probably controlled by the kinetics.***

[1] W. Matz, N. Schell, W. Neumann, J. Bøttiger, and J. Chevallier, *submitted* to Rev. Sci. Instrum.
[2] J. Pelleg, L.Z. Zevin, S. Lungo, and N. Croitoru, Thin Solid Films **197**, 117 (1991).