ROBL-CRG	**Experiment title:** *In situ* **x-ray diffraction during sputter deposition of TiAlN**	**Experiment number:** **20_02_608**
Beamline: BM 20	**Date of experiment:** from: 05.03.2003 to: 11.03.2003	**Date of report:** 28.07.2003
Shifts: 18	**Local contact(s):** Dr. Norbert Schell	*Received at ROBL: 28.07.03*

Names and affiliations of applicants (* indicates experimentalists):

*M. Beckers, Forschungszentrum Rossendorf, Germany

*N. Schell, ROBL-CRG, Germany

Report:

The growth of TiAlN on oxidized silicon wafers, deposited by **reactive magnetron co-sputtering** from metallic **Al and Ti targets**, has been studied ***in situ***. The film thickness was measured with specular x-ray reflectivity. Using Bragg-Brentano geometry as well as grazing incidence in-plane wide angle scattering, the thickness dependent in-plane and off-plane texture of TiAlN was investigated. Additional examinations on texture development were carried out using a Bruker AXS SMART two-dimensional detector. Additionally, post-deposition laboratory work like transmission electron microscopy and evaluation of pole figures on laboratory x-ray instruments complemented the *in-situ* measurements. By this laboratory work the data recorded during the experiment could be accredited to strong off-plane and **in-plane texture** of the TiAlN coatings.

EXPERIMENTAL

The **deposition chamber** (together with scattering geometry and quality of data, like intensity, resolution, background, which can be obtained with the set-up) is described in detail in **Ref. 1**. Both **magnetrons** sources of 1 inch target diameter are **tilted 30 degrees** away from the **substrate normal** at a distance of 100 mm. Here, **both magnetrons were used** at the same time. The Ti magnetron was run at a conctant DC power of **80 W**, while the Al power was varied between **30 and 50 W**. The substrates were 1.5 x 1.5 cm^2 oxidized Si (001) wafer. The base pressure before deposition was appr. $5x10^{-7}$ mbar. The working pressure was varied between $3x10^{-3}$ and $5.5x10^{-3}$ mbar with a fixed **reactive sputter gas** mixture of Ar and N_2 at a ratio of 4:1. The substrate temperature was varied between 200 °C and 350 °C by a resistivity heater. A constant substrate **bias voltage of -30 V** was applied.

The chamber was mounted on the goniometer in the MRH. The incident x-rays were monochromatized to 12.8 keV (**λ = 0.9686 Å**). To study the growth of the films ***in situ***, three scattering geometries were used:

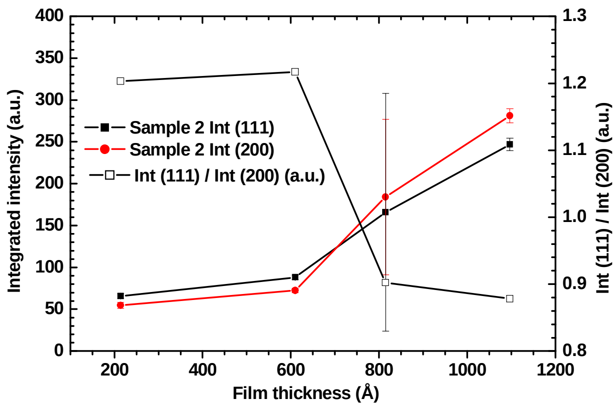
1. Vertical Bragg-Brentano large-angle scattering (**XRD**) revealing the texture. In addition, from the positions of Bragg peaks, information on out-of-plane lattice strain as well as grain sizes and microstrain (lattice defects) can be obtained.
2. Grazing incidence grazing exit in-plane large-angle scattering (**GIXS**). With an incident angle of 0.2°, the calculated x-ray penetration depth was about 200 Å, assuming a mass density of 4.3 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 can be gained.

3. Low-angle **specular reflectivity** with information on film thickness and surface roughness.
4. Grazing incidence large-angle scattering (**GID**) to improve surface sensivity compared to XRD.
5. Real-time Debye-Sherrer recordings with two-dimensional SMART detector in reflexion geometry.

For the latter one the deposition was continious while for the first 5 samples it had to be sequential.

Cross-section micrographs were obtained with a Philips CM300 **transmission electron microscope** after grinding the samples mechanically and then doing a final ion erosion step.
RBS (He^+, E_{He}=1.7 keV) acquired stochiometry was obtained by standard fitting routines (SIMNRA).



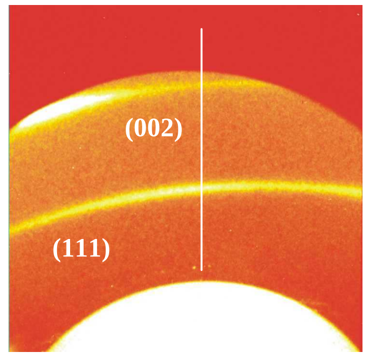
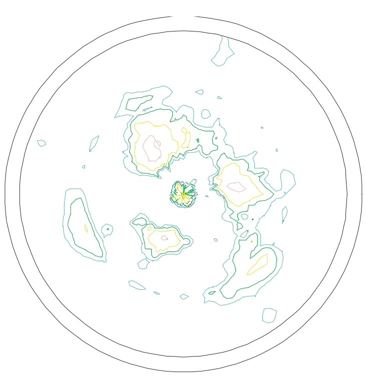


Fig. 1 (left: integrated **GIXS** intensity) and **Fig. 2** (right: SMART micrograph) are examples of the obtained data.

SUMMARY:

A total of **seven TiAlN** films were deposited according to the paramters introduced above. *In-situ* XRD scans showed very low peak intensity for low gas pressure indepent of film thickness, while there was a clear intensity rise in GIXS scans though probing the same sample thickness all the time. Also typical crossover from (002) to (111) orientation perpendicular to the sample's surface could not be observed. Increasing the total gas pressure led to higher, but still relatively low XRD intensities. Resolving SMART measurements showed (111) and (200) crystallographic planes tilted towards the sample's surface leading to low vertical off-plane intensities.
RBS showed the samples were substochiometric in nitrogen for lower gas pressure. Cross-sectional TEM at specific film thicknesses revealed texture evolution from substrate interface to surface, with strong (220) off-plane texture and interrelated in-plane (111) and (220) orientation, explaining the GIXS results. Additional pole figures approved strong off-plane (220) fiber texture but also in-plane texture directed along the incoming particle flux of the second magnetron. With increasesing pressure this texture was decreased.



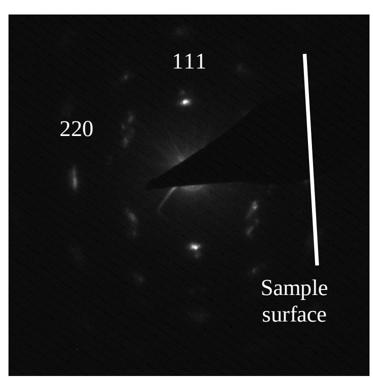


Fig. 3 (left: (111) pole figure) and **Fig. 4** (right: TEM SAED picture) are examples of the laboratory data.

[1] W. Matz, N. Schell, W. Neumann, J. Bøttiger, and J. Chevallier, Rev. Sci. Instrum. **72**, 3344 (2001).