ROBL-CRG	**Experiment title:** ***In-situ study of oscillations during epitaxial and non-epitaxial thin film growth studied by specular and diffuse x-ray scattering***	**Experiment number**: **20_02_661**
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Report

AIM:

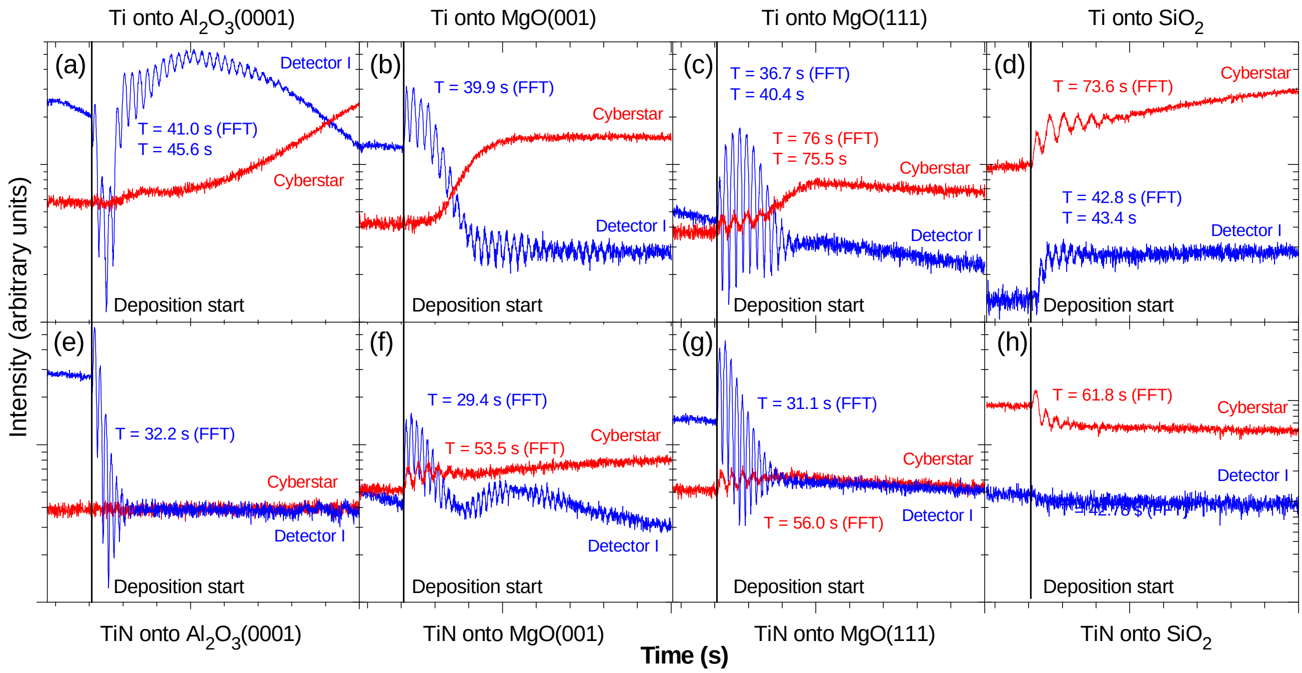
In situ x-ray growth oscillations during epitaxial depositions are mostly characterized by specular x-ray reflectivity at the so-called anti-Bragg position. They are treated as roughness oscillations between nucleation and coalescence of individual monolayers. The amplitude decay caused by roughening is fitted in the kinematical approximation using a step model for the coverages of each monolayer proposed by Cohen [1]. In another approach, measurements during non-epitaxial depositions are carried out close to the critical angle of the deposited material. Correspondingly, the growth oscillations are treated as interference effects (time-resolved Kiessig fringes). Here, the amplitude decay caused by roughening is fitted by employing dynamic scattering using the Parratt formalism together with Nevot-Croce and Debye-Waller roughness models. One way to distinguish between roughness oscillations and thickness interference effects is the simultaneous measurement of specular and diffuse scattering, since for roughness oscillations the specular and diffuse scattering intensity should be exactly out of phase. This has been carried only for a few experiments [2], but up to now is missing a systematic approach. The **aim** of this experiment is to close this gap by a **systematic study of specular and diffuse growth oscillations for a set of film-substrate combinations with epitaxial and non-epitaxial growth**.

EXPERIMENTAL:

A set of 12 samples of different film-substrate combinations was deposited by magnetron sputtering from a Ti target, employing heteroepitaxy with high adatom mobility onto smooth substrates (Ti on to Al_2O_3(0001) and TiN onto MgO(001)), heteroepitaxy with high mobility onto rough substrates (Ti onto MgO(111)), heteroepitaxy with low mobility onto smooth and rough substrates (TiN onto Al_2O_3(0001) and MgO(111)) and polycrystalline film growth (Ti and TiN onto amorphous SiO_2). All films were deposited at a substrate temperature of 300°C. Ti was deposited at a target power of 40 W with an Ar flux of 3.4 sccm and a working pressure of 0.55 Pa, leading to a deposition rate of ~0.53 Å/s. TiN was deposited at a power of 70 W with Ar/N_2 fluxes of 2.16/0.66 sccm and a working pressure of 0.35 Pa, leading to a deposition rate of ~0.76 Å/s.
Each deposited film was characterized *in situ* by specular reflectivity (XRR) for thickness and roughness determination and large angle x-ray diffraction (XRD) to determine phase formation and off-plane lattice parameters, using monochromatized x-rays of λ=1.053 Å.
During each deposition both, the time-resolved specular reflectivity at ($\theta/2\theta$=1.75°/3.5°) and diffuse scattering at the Yoneda peak position in grazing exit configuration ($\theta/2\theta$=1.75°/1.95°) was measured, to distinguish between intensity oscillations due to oscillating roughness and thickness interferences.

RESULTS

The figure shows *in situ* data obtained for specular reflectivity (labelled Detector 1) and diffuse scattering (labelled Cyberstar) during the deposition of eight Ti and TiN films onto various subsrates.



The melting point of Ti is 1668 °C, hence the homologous temperature during deposition was 0.18, yielding sufficient adatom mobility, especially on the Ti (0001) basal plane. A Ti deposition onto Al_2O_3(0001) (a) leads to an intially strained heteroepitxial layer with decrease in the specular reflectivity, but no increase in the diffuse signal, before after breakdown of the interfacial layer the specular growth oscillations decay due to slow surface roughening in accordance with slow increase of the diffuse signal. A deposition onto a relatively smooth MgO(001) substrate (b) without epitaxial matching yields a rough, non-textured polycrystalline Ti film, hence also a faster decay of the specular reflectivity right after deposition start and correspondingly to a steep increase of the diffuse signal with fast saturation before deposition stop. For epitaxial deposition of Ti onto a rough MgO(111) (c), the initial substrate roughness induces a fast decay of the specular growth oscillations with a concurrent oscillating increase of the diffuse signal, before signal saturation of both for later growth states. The ratio between the diffuse and specular oscillation periods is close to two, with a phase coherence (specular: max-max-max, diffuse: max-min-max) between them. The same doubling of the oscilation period is observed for a non-epitaxial – however still (0001)-oriented fiber-textured – deposition of Ti onto a rough amorphous SiO_2 substrate (c), although with less pronounced specular interference fringes due to interfacial reactions between the Ti and the SiO_2, as proven by *ex situ* compositional analyses.
The dissociation temperature of TiN is 2950 °C, hence the homologous temperature during deposition was reduced to 0.11. Furthermore, the surface binding energy of TiN is highly anisotropic with (111) being the high-energy surface. Correspondingly, the oscillations and overall intensity of the specular signal for epitaxial TiN(111) deposition onto smooth Al_2O_3(0001) (e) and rough MgO(111) (g) vanish quickly after deposition start. For epitaxial deposition of the low energy-surface TiN(001) onto MgO(001), the specular oscillations are disnernible for a longer period of time. Here, and for the MgO(001) case, also oscillations of the diffuse signal can be observed, with the same doubling of the period as for Ti deposition. A depositon onto rough SiO_2 yields polycristalline TiN, hence no visible oscillations in the specular signal, but oscillations in the diffuse signal which exhibit a comparable oscillation period as for depositions onto MgO(111) and MgO(001).

REFERENCES

[1] P. I. Cohen., *et al.*, Surface Science. **216**, 222 (1989).
[2] A. C. Mayer, *et al.*, Organic Electronics. **5**, 257 (2004).