Pole figure measurements in grazing-incidence configuration for characterizing thin film texture

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Synopsis
Grazing incidence wide-angle X-ray scattering is presented as an innovative approach to measure X-ray diffraction pole figures, enhancing sensitivity to nanoparticle and thin film texture compared to conventional Schulz geometry. Tuning the grazing incidence angle is utilized to obtain texture information at different depths in thin films.

Introduction
X-ray diffraction pole figure measurements are essential to analyse crystallographic texture in thin films and nanoparticles, with applications in a wide array of material science fields. In lab- and synchrotron-based setups, pole figures are conventionally measured using equal incident and exit angles (θ-2θ or Shulz geometry) well-beyond the critical angle of the studied material. This geometry features a large X-ray penetration depth, decreasing the pole figure sensitivity to thin films or supported nanoparticles. In addition, the necessity to tilt the sample stage out of the horizontal plane during pole figure measurements in Schulz geometry makes *in-situ* experiments rather challenging.

This study demonstrates the use of grazing-incidence wide-angle X-ray scattering (GIWAXS) to acquire pole figures that are more sensitive to the near-surface region compared to pole figures measured in classic Schulz geometry. Pt thin films and nanoparticles on a MgO single-crystal substrate are used to showcase this method.

Results
GIWAXS patterns are recorded continuously while rotating the sample azimuthally in its own plane (Fig. 1a). The near-horizontal sample orientation during GIWAXS – only tilted by the small incident angle – enables a more facile implementation of reactor chambers for in situ metrology compared to conventional Schulz geometry. Each measured 2D GIWAXS pattern contributes a line to the pole figure (Fig. 1b-c), and pole figures at different q-values (or equivalently 2θ-values) can be measured simultaneously. Figure 2 demonstrates the increased sensitivity to supported Pt nanoparticles with respect to the MgO substrate when using a grazing-incidence beam. It is seen that by decreasing the incident angle α below the MgO critical angle for total external X-ray reflection, the intensity of the Pt (111) diffraction spot is raised above the intensity of the MgO (111) diffraction spot, increasing the sensitivity to the targeted Pt signal while limiting the MgO background signal.

This increased sensitivity to the surface region is caused by the decreased X-ray penetration depth at small incident angles (Fig. 3a). This principle can also be utilized to study the crystalline texture at different depths inside a thin film. As an example, Figure 3b shows Pt 111 pole figures for a 50 nm (001)-oriented epitaxial Pt film on a MgO support recorded at an incident angle below and above the Pt critical angle. Below the critical angle, only the top few nanometers are probed, consisting exclusively of (001)-oriented grains. Above the Pt critical angle, the deeper X-ray penetration unveils extra diffraction spots, caused by (122)-oriented Pt grains presumably situated at the film-substrate interface.

Conclusion
The increased sensitivity to the near-surface region of a grazing-incidence X-ray beam, combined with the simpler measurement geometry and the capability of depth profiling, prove grazing-incidence pole figures to be powerful for future *in situ* and *ex situ* texture analysis on thin films and supported nanoparticles.

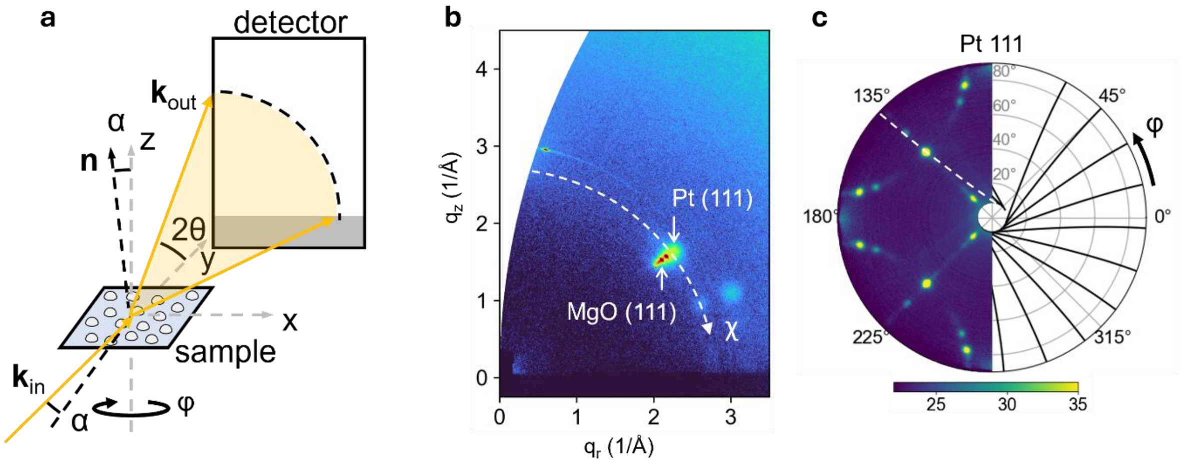

Figure 1: Grazing-incidence geometry and pole figure reconstruction. **a**: Detection of the X-ray diffraction cone. **b**: GIWAXS pattern for supported Pt nanoparticles. The white dashed line marks the contribution to the Pt 111 pole figure. **c**: Constructing a pole figure by scanning while rotating the sample. The black lines mark the contributions of individual scans.

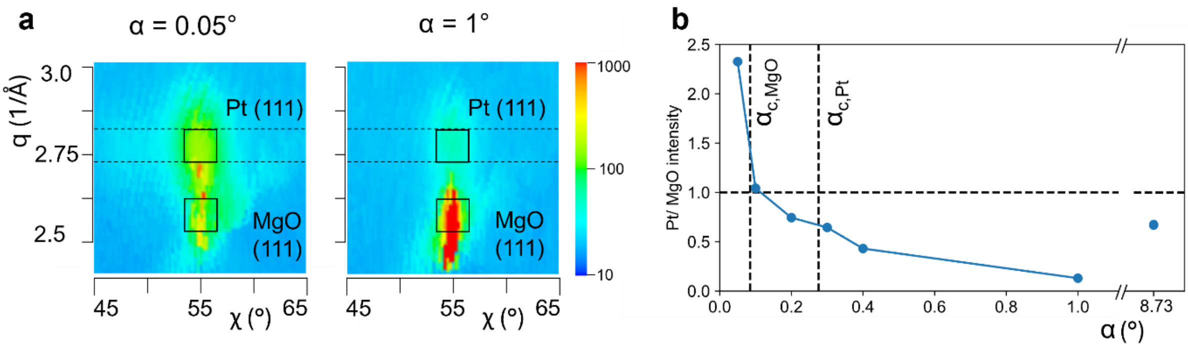

Figure 2: Pole figure sensitivity towards Pt nanoparticles compared to the MgO support. **a**: Reciprocal space cuts through the Pt and MgO (111) diffraction spots recorded at two incident angles (α=0.05° and α=1°). **b**: Ratio between the Pt and MgO (111) diffracted intensity versus incident angle, showing that the intensity of the Pt (111) diffraction spot is raised above the intensity of the MgO (111) diffraction spot when going below the critical angle of MgO ($\alpha_{c,MgO}$). The point at α=8.73° is measured in Schulz geometry.

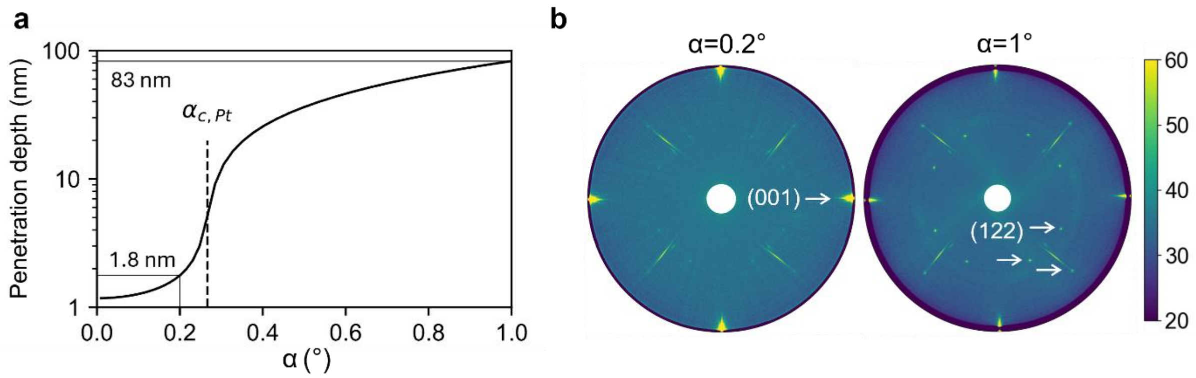

Figure 3: Selecting the information depth. **a**: X-ray penetration depth in Pt versus the incident angle. **b**: Pt 111 pole figures for a 50 nm (001)-oriented epitaxial Pt film recorded at two incident angles. At α=0.2° (i.e. below $\alpha_{c,Pt}$), the limited X-ray penetration depth in the film generates sensitivity to Pt crystals situated in the near-surface region only, leading to diffraction spots of (001)-oriented Pt. Well-beyond $\alpha_{c,Pt}$, X-rays penetrate through the entire Pt film, generating sensitivity for crystals at the near-surface, bulk and Pt-MgO interface. At α=1°, specifically, additional (122)-oriented Pt diffraction spots become visible, which can be linked to textural components at the interface region.