Total scattering at grazing incidence to study real thin film systems at variable temperature

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Complementary to x-ray diffraction patterns that represent the crystal lattice in Q space, the atomic pair distribution function (PDF) describes the structure of a material as a histogram of interatomic distances r in real space. The total scattering (TS) approach that enables PDF analysis requires that scattering data is collected over a wide Q range of the order of 20 $Å^{-1}$ and subsequent Fourier transformation of the entire scattering pattern into direct space. While TS at high-energy beamlines has become a standard routine for bulk-type samples, the unfavorable thickness ratio of a thin film (nanometer regime) to its substrate (micrometer regime) limits the detectability of the film signal in simple transmission geometry as described *e.g.* in Ref. [1]. Therefore, we applied the high-energy surface diffraction technique established for single-crystal surfaces [2] to less ordered films and thus pushed the capabilities for PDF analysis of thin films to unprecedented limits in terms of minimum thickness and time resolution. [3,4] Besides polycrystalline and textured metal and oxide layers, we studied amorphous and naocrystalline thin films. By careful data treatment, we successfully derived PDFs of comparable data quality from different HfO_2 films with thicknesses down to 15 nm independent on their degree of ordering with domain sizes between ~5 and >30 Å. All films were deposited on fused silica which provides an easily scalable background to subtract from the sample data to isolate the film signal. Real thin film devices *e.g.* for electronic applications, however, typically consist of multiple layers, and the film growth is largely affected by the nature of the underlying layer. Therefore, we further developed grazing incidence total scattering towards a depth-resolving method by scanning the incidence angle. In this way, the technique provided insight into the structure of different types of bilayer samples studied for their use *e.g.* in next-generation computer memory applications. PDFs were successfully extracted from the individual layers of different combinations and stackings of amorphous and crystalline materials exhibiting high and low (electron) density and, hence, x-ray scattering power from TiO_2 to Pt [5]. As thermal treatment is an essential part of thin film device manufacturing, we are developing a laser-interferometer based system that, beyond data collection during isothermal heat-treatment as applied in [4], enables following structural changes during variable-temperature processes up to several hundred degrees. Fig. 1 shows data from the proof-of-concept experiment on a 30 nm HfO_2 thin film deposited by chemical vapor deposition in an amorphous state, crystallized *in situ* while continuously acquiring TS data.

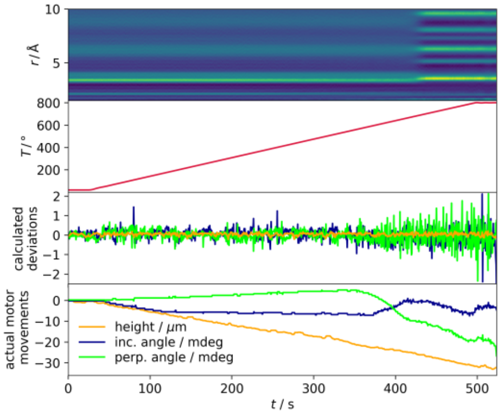

Figure 1. PDFs of an amorphous hafnium oxide film acquired during crystallization at the given heating rate of 100 K min^{-1}, incl. the fluctuations of the sample and the actual motor movements necessary to compensate for the thermal expansion.

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