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Supporting information for article:

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Supplementary information

Local atomic structure of thin and ultrathin films *via* rapid high-energy X-ray total scattering in grazing incidence

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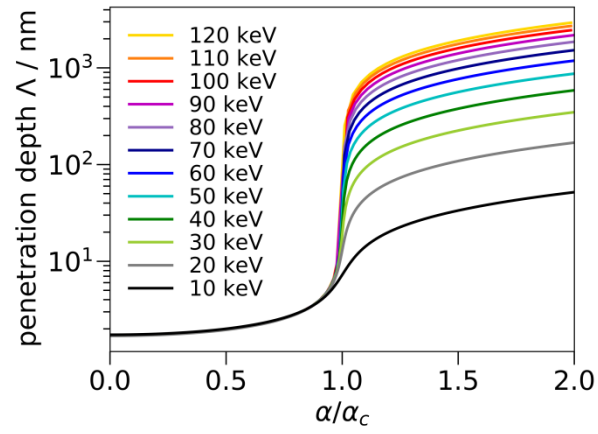


Figure S1: Penetration depth of photons of various energies calculated according to R. Feidenhans'l, (1989), Surf. Sci. Rep. 10, 105–188, exemplarily for Cu as a material without absorption edges within the illustrated energy range.

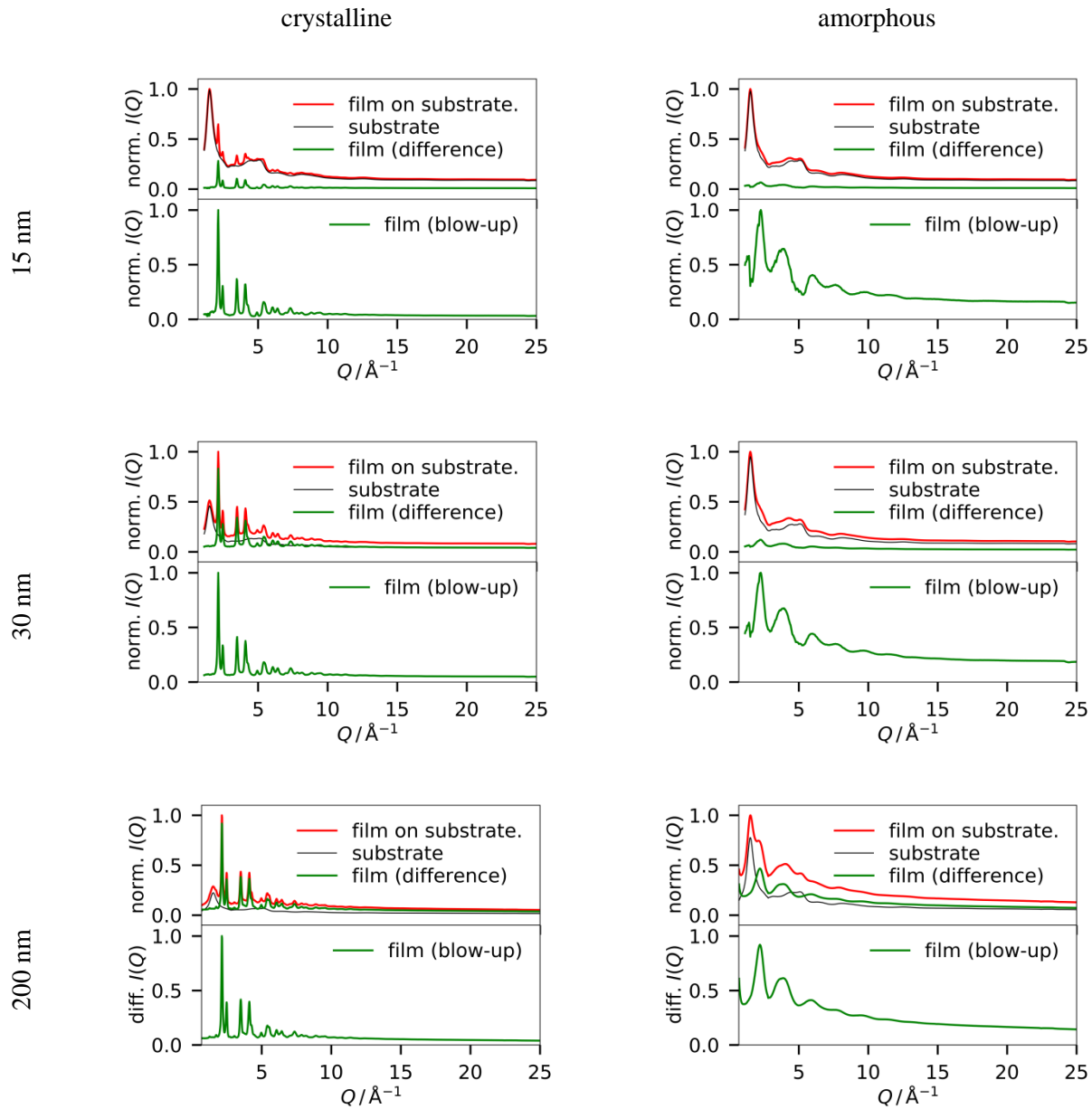


Figure S2: Integrated XRD patterns for HfO₂ layers on fused silica at different thicknesses, prepared by various repetitions of the coating process, and heat treated at 800 °C (crystalline) and 295 °C (amorphous). The plots show the integrated data of the samples and the scaled background patterns from the fused silica substrate, and the magnified subtracted signals for each case.

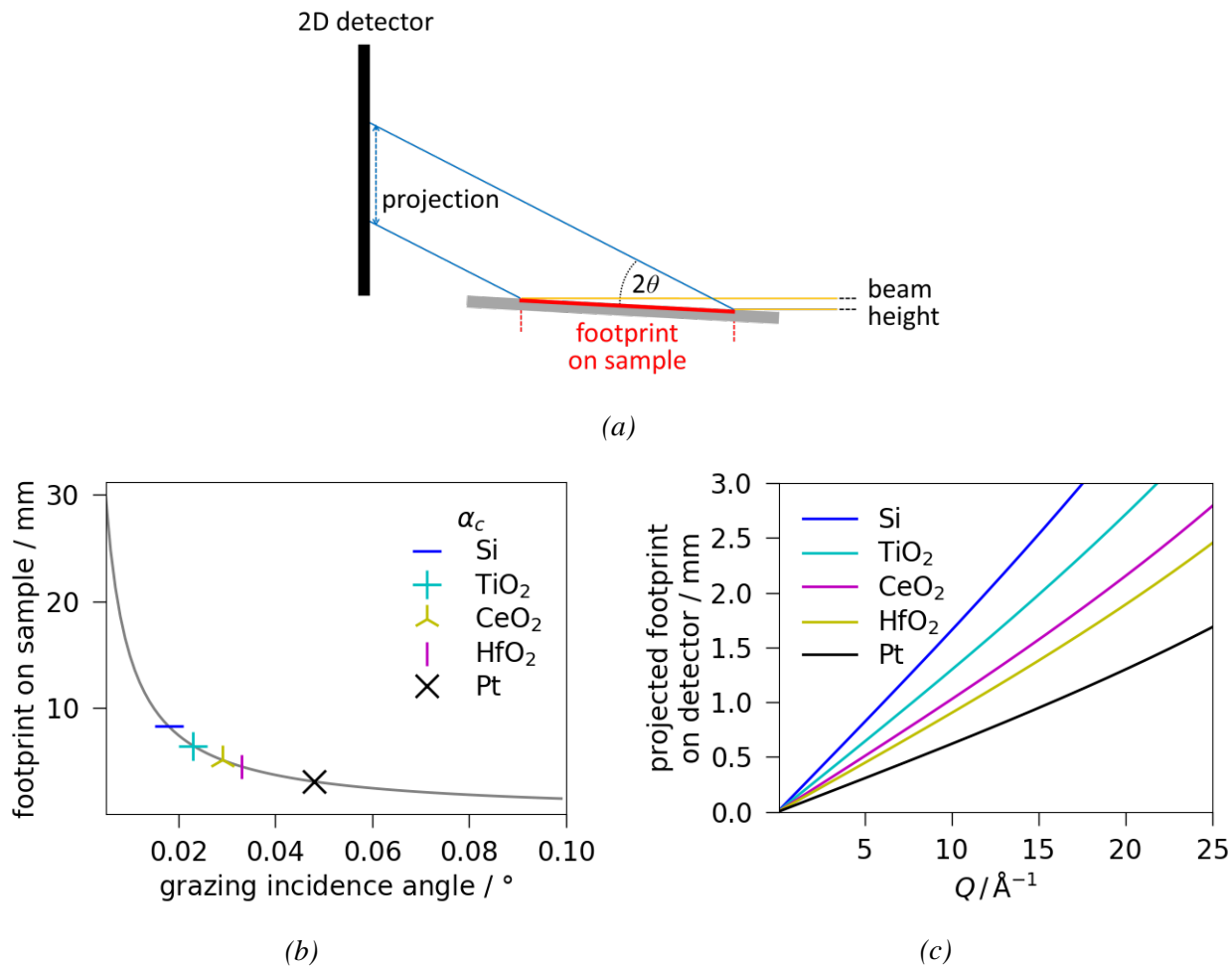


Figure S3: (a) Schematic illustration of the grazing incidence geometry (incidence angle exaggerated for clarity) and projection of the footprint on the area detector; (b) footprint in dependence on the incidence angle with marked critical angles for some selected materials; (c) projected footprint on the area detector calculated for the critical angles of the indicated materials; all values are calculated for a beam height of $3\ \mu\text{m}$ based on simple trigonometric considerations for the vertical scattering plane and a photon energy of $100\ \text{keV}$.

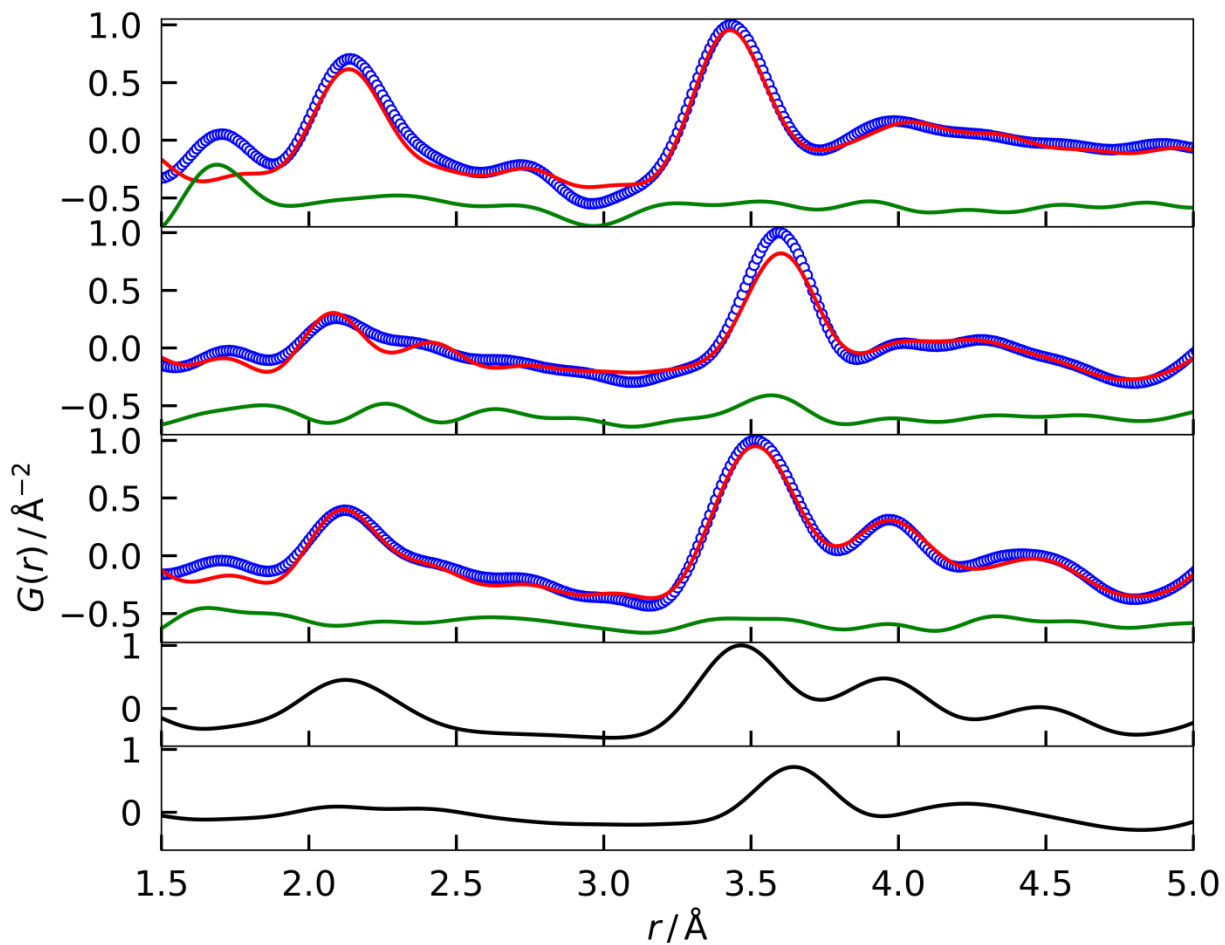


Figure S4: Zoom into Figure 4 of the main text to illustrate the local order of (from top to bottom) a spin-coated ZrO_2 film pre-annealed at $295\text{ }^\circ\text{C}$, two crystalline ZrO_2 films prepared from equally pre-annealed films and heated to $800\text{ }^\circ\text{C}$ in a rapid thermal annealing process with a heating rate of $\sim 100\text{ K min}^{-1}$, and annealed at $900\text{ }^\circ\text{C}$ in air at a slow heating rate of $\sim 10\text{ K min}^{-1}$, respectively (blue dots: data, red lines: calculated model, green line: difference curve, offset of -0.5 for clarity), along with the calculated reference PDFs from the ICSD database (references #658755 for monoclinic and #93124 for tetragonal with reduced values for U_{iso} extrapolated to room temperature).

Table S1: Q_{\max} values applied in the Fourier transformation into the GIPDFs shown in the main text (with the exception of the calibration illustrated in Figure 3 where the Q_{\max} values are included as a plot).

figure (text passage)	material	parameters	$Q_{\max} / \text{\AA}^{-1}$
Fig. 2 (Section 3.1)	Pt	3 nm thickness	17.2
		HfO ₂	15 nm, crystalline
	30 nm, crystalline		21.1
	45 nm, crystalline		21.1
	15 nm, amorphous		12.5
	30 nm, amorphous		12.5
	45 nm, amorphous	14.4	
Fig. 4 (Section 3.3)	ZrO ₂	295 °C thermal treatment	21.0
		800 °C thermal treatment	21.0
		900 °C thermal treatment	21.0
Fig. 5 (Section 3.4)	Pt	normal incidence	17.0
		grazing incidence	24.0