cryo-ePDF to Measure the Atomic Structure of Amorphous ALD Interfaces

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Atomic layer deposition (ALD) provides uniform and conformal thin films that are of interest for a range of applications. To better understand the properties of amorphous ALD films and ALD-modified interfaces, we need improved understanding of their local atomic structure. Previous work demonstrated measurement of how the local atomic structure of ALD-grown aluminum oxide (AlO_x) evolves in operando during growth by employing synchrotron high energy X-ray diffraction (HE-XRD). In this work, we report on efforts to employ electron diffraction pair distribution function (ePDF) measurements using more broadly available transmission electron microscope (TEM) instrumentation to study the atomic structure of amorphous ALD-AlO_x and ALD-modified interfaces. We observe electron beam damage in ALD-coated samples during ePDF at ambient temperature and successfully mitigate this beam damage using ePDF at cryogenic temperatures (cryo-ePDF). We examine both wide-area electron diffraction over a ~200 nm spot size, as well as local diffraction over a <2 nm spot size using cryo-ePDF. The smaller spot size (achieved using convergent electron diffraction with a small convergence angle) allows for local electron diffraction, for example at multiple positions along the cross-section of a 10 nm thick ALD film, and enables characterization of the interfacial atomic structure of the ALD film relative to the bulk structure. We employ Reverse Monte Carlo (RMC) modeling to obtain structural models from the cryo-ePDF data. From these model structures, we derive structural metrics including stoichiometry, pair distances, and coordination environments in the ALD films and ALD-modified interfaces. The cryo-ePDF technique can be used for routine measurement of atomic structure with high spatial resolution to improve understanding of ALD materials, establish structure-property relationships, and help accelerate the timescale for the application of ALD to address technological needs.