Structural analysis of novel materials via grazing-incidence high-energy total X-ray scattering

Olaf Borkiewicz[1]

[1]X-Ray Science Division, Argonne National Laboratory

borkiewicz@anl.gov

Increasing advances in the science for the discovery of new functional materials and chemistry are addressing tailored interfaces and thin-film, 2-D materials. The need for high quality ultra-thin and conformal films with precisely controlled thickness and composition at the atomic level has been growing within the last decade, with applications in a wide range of technologically and scientifically relevant fields, e.g. semiconductor industry, photovoltaics, water splitting, catalysis, energy storage and conversion. As a consequence, a detailed atom-level understanding of the structure-to-property relationship is needed to take full advantage of tailoring the emerging functional materials. An often nanostructured and disordered nature of these materials however, poses significant characterization challenges for standard crystallographic tools and analytical approaches.

Atomic pair distribution function (PDF) analyses of high-energy (> 50 keV) total X-ray scattering data have proven to be a unique technique for structural analysis capable of providing atomic-scale insights into materials structure with sub-Ångstorm spatial resolution. Whereas the application of total X-ray scattering measurements in transmission geometry and subsequent PDF analyses for functional bulk materials is well established, their application to study structure and function of 2-D, interfacial and thin-film materials are presently limited. The main impediment to routine PDF analyses of thin films using standard approach is the fact that signal generated during transmission geometry measurements is dominated by the contributions from the substrate rather than the sample (film), which consequently hinders reliable and consistent data reduction and PDF extraction. Recently, the possibility of achieving grazing-incidence high-energy total X-ray scattering data of high quality and PDF analyses (GI-PDF) has been demonstrated and suggests opportunities to develop capabilities that are poised to achieve breakthroughs for in situ and operando structure analyses of functional ultrathin materials. High resolution and surface sensitivity of GI-PDF are needed to suppress background contributions enabling structural analysis at the atomic scale and quantitative benchmarking to ab-initio structure models.

We will discuss recent upgrades to beamline 11-ID-B of the Advanced Photon Source that enabled GI-PDF and the application of this emerging methodology to gain understanding on the phase formation and evolution during early stages of the growth of indium oxide ultrathin films via sequential infiltration synthesis (SIS). Analysis of the pair distribution data has revealed that the prime nuclei, formed during the initial stages of the SIS, exhibits a structure of high ration (elongated clusters) and evolves into three dimensional network after additional SIS cycles. The annealing of the samples consolidated the as grown clusters into cubic indium oxide nanocrystals with structural details that also depend on SIS cycle number.