XIS Working Group: Buried Interfaces

Dillon Fong, John Freeland, Hua Zhou, Christian Schlepütz, Seoyhoung Chang, John Okasinski, Phil Ryan, Paul Fenter

The strength of the XIS sector lies in the *in situ* study of materials structure and properties. A wide variety of dynamic phenomena are of interest, e.g., crystal growth, surface/interface reactions in gas/liquid environments, and phase transitions. Depending on the scientific focus, one can choose from a multitude of x-ray scattering techniques. Here we discuss x-ray measurements aimed at understanding interfacial structure and its evolution.

For monocrystalline materials, which may take the form of a heterostructure, superlattice, or an array of quantum dots, it is possible to image the atomic structure of the surface/interface with sub-Ångstrom resolution through the use of phase retrieval techniques [1-3], which may be combined with standard fitting/optimization algorithms [4,5]. The measurement, often called surface x-ray diffraction (SXRD), involves collecting a large data set of structure factors from crystal truncation rods (CTRs) and/or superstructure rods (SRs) (the latter originating from a reconstructed surface). From the viewpoint of phase retrieval analysis, the data set must be oversampled [6]. If the measurement is conducted near an x-ray absorption edge of an element in the sample, the researcher can also extract composition profiles from the data [7,8].

SXRD is typically applied to pseudo-two-dimensional epitaxial samples, and the main feature of interest is the unit cell structure along the out-of-plane direction; consequently, these particular studies do not require a particularly coherent x-ray source. (Coherent scattering from different surface domains are discussed below). Of primary importance is the ability to rapidly collect a high quality data set from samples with often very small volumes: to this end, we require high flux (> 10^{12} photons/sec), low background area detectors (e.g., Pilatus, Eiger), and high resolution diffractometers with the necessary degrees of freedom for both surface scattering and reflectivity (e.g., a six-circle diffractometer). For laterally inhomogeneous samples, studies can be carried out with a focused beam: focusing on the order of 1-10 µm would permit the study of individual nano-mesoscale structures as well as structures patterned by scanning probe techniques (e.g., piezoresponse microscopy). For many weakly scattering samples, the enhanced flux from a multilayer monochromator would be highly beneficial.

Resonant SXRD measurements must be carried out on the energy tunable beamline. For most resonant anomalous techniques (e.g., multiwavelength anomalous dispersion (MAD) phasing or resonant anomalous x-ray reflectivity (RAXR) [9]), the energy resolution from a Si (111) or diamond (111) double crystal monochromator (~10^{-4}) is necessary.

While SXRD is now a standard x-ray scattering technique, the use of phasing algorithms in their analysis is relatively new. As of today, several groups around the world utilize such algorithms, but because of their complexity, their use remains far from routine. Many users could substantially benefit from a standardized method for modeling large

SXRD data sets, and there is an ongoing, long-term effort at the APS to streamline the phase retrieval software. This effort should be aided by specialized XIS staff focused on software development. Of more immediate importance is the development of a standardized data collection scheme and data visualization software. Users should work with XIS staff in implementing the scheme and acquiring a sufficiently oversampled data set of structure factors along with their errors. It will be important to provide real-time data visualization so that users can rapidly assess the quality of the incoming data as well as establish the time-independence of a single data set assumed to represent a "snapshot" of the atomic structure. This will be of particular importance for the XIS beamline since many of the studies will be conducted *in situ*. With the development of data integration software, the collected data set, which can often range from 10-100 gigabytes in size, can be reduced to a much smaller standardized data file (often less than one megabyte in size) that the user can easily handle and analyze further. In addition to the development of SXRD collection/integration/analysis software, standard packages for low-angle reflectivity, grazing incidence small angle x-ray scattering (GISAXS), and total reflection x-ray fluorescence (TXRF) should be made available to users. We estimate that at least one full time staff member should be dedicated to software development. In addition, at least one full time staff member should be dedicated to each experimental station.

Due to large data set necessary for 3D atomic-scale imaging, one cannot conduct such a study on the ~ 1 sec time scale; as such, the time evolution of interface structure during many processes (e.g., film deposition) cannot easily be measured by these SXRD techniques. With a coherent beam, real-time studies of evolving surfaces/interfaces can be conducted by studies of surface x-ray speckle [10,11]. Furthermore, with additional x-ray optics, it is possible to directly image evolving surfaces and buried interfaces in real-time with coherent x-ray reflection imaging microscopy (XRIM) [12]; the ramifications of coherence on XRIM image interpretation have yet to be developed. Using XRIM, one could also locate particular features of interest that can subsequently be probed with higher resolution using coherent diffraction imaging (CDI) techniques. This could be carried out on the same instrument, provided that adequate sample positioning is available. In this regard, we much prefer the 10/30/30 keV configuration for the fixed-angle beamline: the 10 keV branch would be dedicated to coherent surface scattering studies.

For many of these studies, it is important to ensure that proper ancillary facilities are available. Many of the measurements will be carried *in situ* (e.g., film deposition), meaning that a sample preparation lab containing a wet lab and tube furnaces (e.g., for annealing oxide surfaces) should be near the beamline. In some instances, users may wish to modify the sample using techniques difficult to perform *in situ* – for example, using a scanning probe technique to apply a local electric field across a small region of a ferroelectric sample; since the effect of such a field may dissipate with time, it is important that such equipment be housed near the beamline (e.g., in the CNM or in the anticipated Materials Synthesis Institute).

We expect that researchers from local partners (e.g, Materials Synthesis Institute, CNM, MSD, Northwestern) will aid in staffing the XIS sector. This would strengthen the scientific tie between the XIS sector and these partners and also help provide continuity

for several anticipated projects (e.g., the oxide MBE system), where significant time investment will be necessary.

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