ATOMIC SCALE SCANNING TRANSMISSION ELECTRON MICROSCOPY

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The Scanning Transmission Electron Microscope (STEM) is optimized to perform both imaging (Z-contrast or bright-field phase -contrast) and analysis (electron energy loss spectroscopy (EELS) or energy dispersive X-ray spectrometry (EDS)) using a small electron probe (typically 0.08-0.2nm in size). This mode of operation naturally lends the STEM to particular materials challenges where the simultaneous determination of the atomic structure (by imaging), the composition (by EDS or EELS) and the electronic structure (by EELS) is essential to understanding the overall properties. Examples of such materials challenges include interfaces in structural materials [1], dislocations in semiconductors [2] and nanoparticulate systems for optical or chemical applications [3].

In recent years, aberration correctors and monochromators have greatly improved the quality of Z-contrast images and electron energy loss spectra that can be obtained from STEM. With a monochromator incorporated into an FEI Tecnai F20 Schottky field emission STEM, spectral resolution comparable to a synchrotron can be obtained (~70meV). While the highest energy resolution cannot yet be coupled with the optimum spatial resolution of the microscope (due to source demagnification and signal levels), analysis can routinely be performed with ~1nm spatial resolution. The current combined limits of spatial and energy resolution are particularly useful for the analysis of the low-loss region of the spectrum (where delocalization reduces the spatial resolution to ~1nm anyway), permitting quantum confinement effects and optical responses of individual nanostructures to be measured directly. Higher spatial resolution EELS can be obtained from a Nion aberration corrected cold-field emission STEM, where the ~0.4eV spectral resolution of the source can be coupled with an optimum spatial resolution of <0.1nm. Such resolution is particularly useful for analyzing core-loss signals at defects/interfaces, where highly localized structural/compositional modulations are expected to have the largest effect on the structure-property relationships.

Research being performed within the Interface Physics Group at UC-Davis is utilizing the advantages of the STEM mode of operation to analyze a wide variety of organic and inorganic materials. This analysis is being performed on a series of microscopes available at UC-Davis and through collaboration with the National Center for Electron Microscopy (NCEM) at Lawrence Berkeley National Laboratory (LBNL) and Lawrence Livermore National Laboratory (LLNL). In all cases, the choice of whether to use a standard system, an in-situ gas handling stage, a monochromator for higher energy resolution spectroscopy, or an aberration corrector for higher spatial resolution and increased signal levels, depends on the particular materials system/application being studied. Assessing the type of microscope that is best suited for the analysis is the major part of the experiment, and in some cases, a lower beam current and spatial resolution can be an advantage.

An example of the use of these methods is shown in Figure 1, where a standard instrument is used to analyze intergranular films in Si_3N_4 ceramics. Here the grain boundary structures is observed to change considerably with the size of the rare-earth dopant. The use of an aberration corrector for higher spatial resolution is highlighted in figure 2(a), where the structure of a mixed dislocation can easily be observed, despite the high level of associated strain. In figure 2(b), the monochromator allows the low-loss region of the spectrum to be analyzed in detail and the band-gap of the material being studied to be determined. In this presentation, these results will be described in detail and the types of experiments suited for the wide range of instruments currently available will be discussed.

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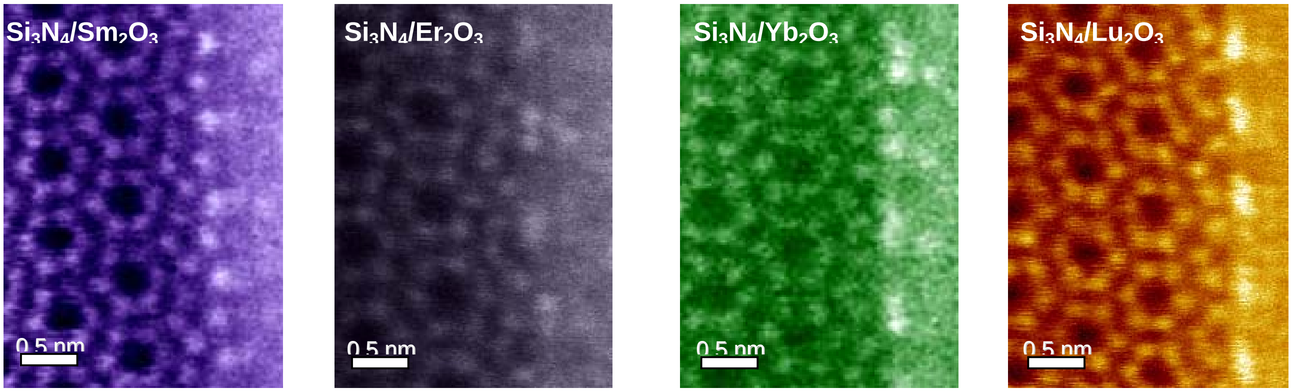

Figure 1. Z-contrast images (obtained with a probe size of ~0.14nm) showing the interface structure between Si_3N_4 grains and the amorphous intergranular oxide film for a range of rare-earth additives. As the atomic size of the dopants decreases (left to right across the figure), the interface structure changes from a uniform distribution of rare-earth atoms at the two distinct interface sites to a preference for the atoms to segregate to a particular site. These changes in interface structures can be correlated with a change in structural properties.

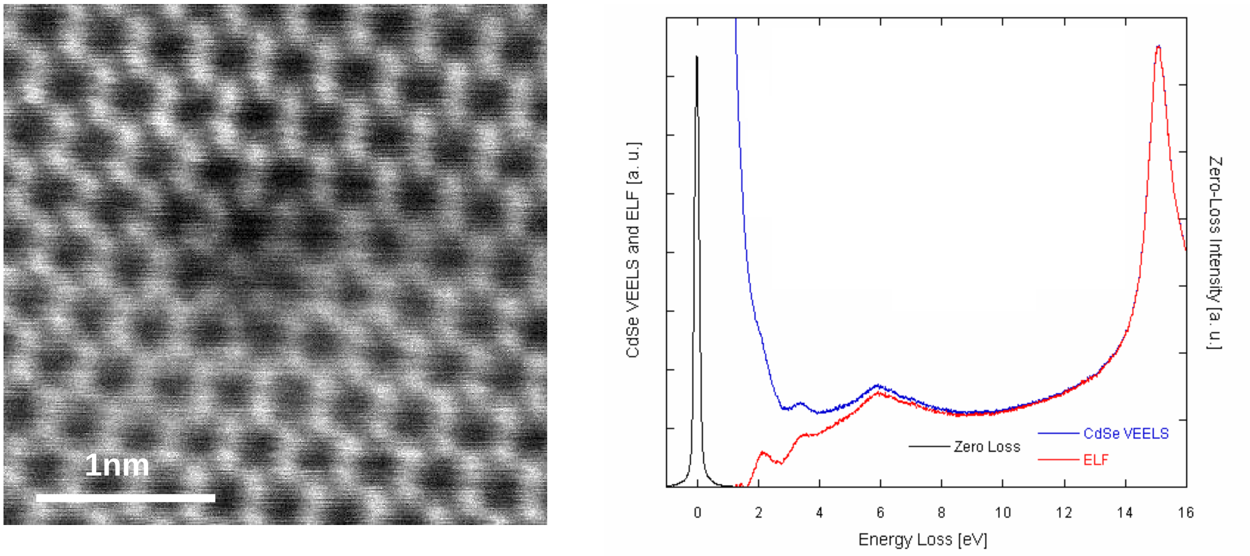

Figure 2. (a) Z-contrast image of a mixed dislocation in GaN obtained from an aberration corrected microscope with a probe size ~0.1nm. (b) Low-loss spectrum obtained from CdSe with a monochromated instrument showing the ability to measure the local band-gap.

References:

[1] A. Ziegler et al, *Science***306**, 1768-1770 (2004)
[2] I. Arslan et al, *Physical Review Letters* **94**, 025504 (2005)
[3] R. Erni and N. D. Browning, submitted *Ultramicroscopy*