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Investigating the Spatial Resolution of Vibrational Electron Energy-Loss Spectroscopy

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Recent work on monochromated electron energy-loss spectroscopy (EELS) has pushed the energy resolution achievable in a scanning transmission electron microscope (STEM) to around 10 meV [1]. This has enabled the detection of vibrational excitations with high spatial resolution. This high spatial and energy resolution can be leveraged to investigate local surface chemistry and analyze chemical bonding arrangements, which can in turn be used to characterize composition, structure, and chemical reactions. A fundamental understanding of this technique will have a tremendous impact on areas like catalysis. To develop this technique comprehensively, we need to perform experiments on relatively simple model systems and corroborate our experimental results with theoretical models. Here we explore the spatial variation in the SiO_2 optical phonon when the electron beam is scanned across a SiO_2/Si interface. The experimental profiles are interpreted in terms of models based on dielectric theory.

A Si wafer was subjected to thermal oxidation to give a ~900 nm layer of SiO_2 and was prepared for STEM EELS analysis by preparing a focused ion beam (FIB) lift out sample using a Nova 200 NanoLab (FEI) FIB. EELS line scans were performed across the SiO_2-Si interface using a Nion UltraSTEM 100 aberration-corrected electron microscope equipped with a monochromator, operated at an accelerating voltage of 60kV with probe convergence and collection semi-angles of 10 and 15 mrad respectively. The line scans were processed for background subtraction, signal integration, and thickness profiling using the Gatan Microscopy Suite.

Fig. 1a shows an ADF image of the Si/SiO_2 interface. The insert is the ADF intensity profile along the path followed by the electron beam during the EELS line scan and shows that the sample thickness is uniform in the SiO_2 region. Fig. 1b shows typical spectra when the beam is in SiO_2 and Si. A peak at approximately 138 meV is clearly seen in the spectra, corresponding to the SiO_2 optical phonon [1]. We also observe a background SiO_2 phonon signal when the beam is in Si due to the presence of surface oxide layers. Fig. 2a shows the integrated raw and normalized phonon signal profiles. The normalized signal was divided by the zero-loss peak to account for changes in the signal due to elastic scattering outside the entrance aperture of the EELS spectrometer. The phonon signal starts to decrease about 100 nm from the interface and drops almost to the background level right at the interface. Interestingly, there appears to be very little lateral “aloof beam” contribution from the SiO_2 layer when the probe is in the Si.

To model the behavior we are simulating spectral line scans using dielectric theory [2, 3]. Fig.2b shows the spatial variation in the SiO_2 phonon signal as a function of position for different values of q, the change in wavevector parallel to the surface. As expected, the line scan is much sharper at high q values and, at intermediate values of q, the shape of the profile matches the experiment. For all values of q, there is no significant lateral aloof beam SiO_2 intensity when the beam is in the Si. We will discuss detailed simulations of the experimental line profiles and consider the significance of relativistic effects using the models developed by Moreau et al. [4].

References:

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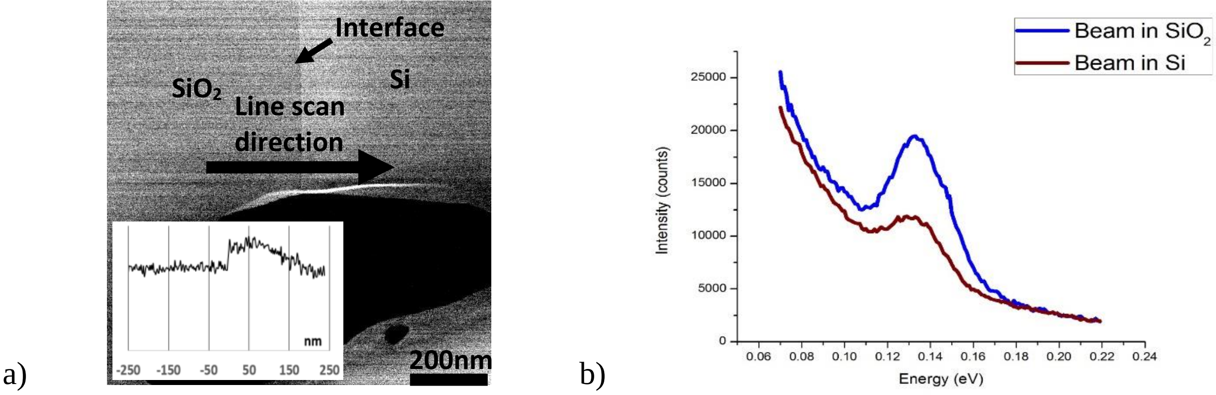

Figure 1. a) ADF image of the Si/SiO_2 interface. The insert is the ADF intensity profile followed by the electron beam during the EELS line scan and shows that the SiO_2 sample thickness is uniform. b) Typical spectra when the beam is in SiO_2 and Si.

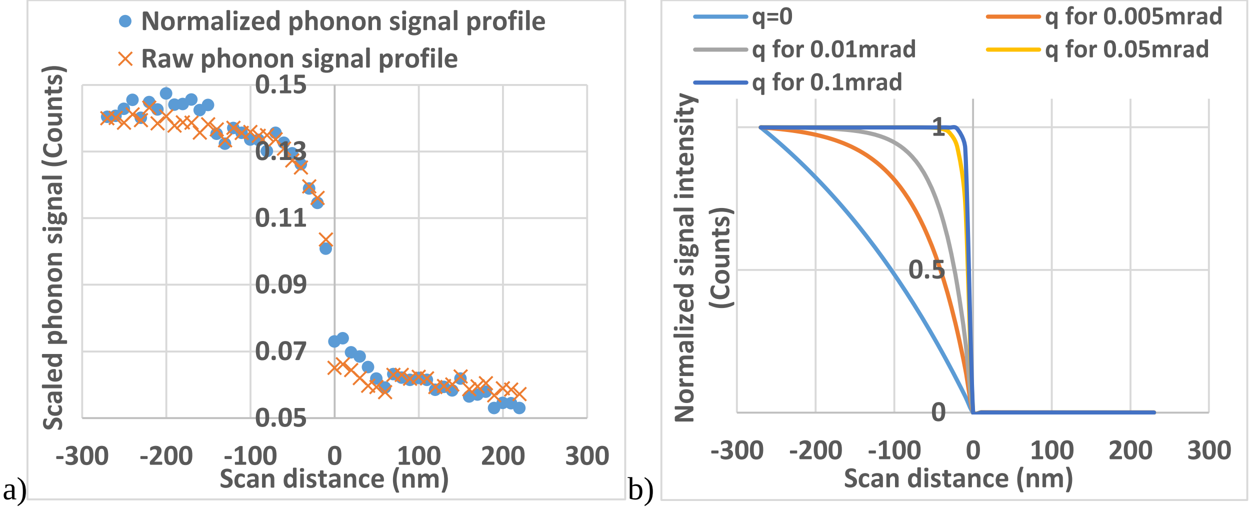

Figure 2. a) Integrated raw and normalized phonon signal as a function of position. b) Simulated spatial variation in the SiO_2 phonon signal as a function of position for different values of q (the values are scaled to match in the SiO_2 a large distance from the interface).