Monolayer resolved EEL spectroscopy with StripeSTEM for high-resolution interface analysis

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A simple yet powerful technique for monolayer resolved electron energy loss (EEL) spectrometry at interfaces or 1-dimensional nanostructures in the scanning transmission electron microscope (STEM) was recently proposed in ref. [1]. The technique, named StripeSTEM, is based on an isochronous collection of a series of EEL spectra while a high-angle annular dark-field (HAADF) image is taken. This contribution evaluates limitations for the spatial resolution of the technique and gives application examples in materials science.

A reference sample consisting of a monolayer basal plane of In_2O_3 in a ZnO matrix was used to measure the spatial resolution (Fig. 1). The data was taken in a probe-side corrected FEI Titan 80-300 at 300kV. Several scan lines in the HAADF image in Fig. 1a, each taken with a dwell time of around 0.1 s, contribute to a single EEL spectrum. The sequence of EEL spectra taken with a maximum of spectrometer uptime during the scan of the HAADF image is presented as a stack image in Fig. 1b. The analysis of the EEL data shows that the width of the inelastic signal of In and Zn is slightly less than 3 Å. Multislice calculations for the elastic electron beam dispersion in the sample and the convolution with and effective inelastic scattering cross sections were used to assess the residual delocalisation of the inelastic signal. Taking the remaining spatial delocalization due to the brightness limit of the Schottky field emitter used in the experiment into account, we can demonstrate that the StripeSTEM technique provides EEL signals with a spatial resolution close to the physical limit of the localization of inelastic scattering events.

Contrary to current approaches, the precise synchronisation of measurement location and EEL data is done after data collection, using the HAADF image as a simple record of the beam position during the measurement. The result of post-processing is a quantitative accuracy with a high tolerance against specimen and beam drift during the experiment. Another benefit for the practical application is the distribution of the electron dose along one spatial direction, enabling for a high signal-to-noise ratio at low dose per $Å^2$. Doses can be a kept as low as of 0.001 $nC/Å^2$ which is of considerable importance for interfaces that are sensitive to radiation damage, e.g. oxide hetero-interfaces.

Fig. 2 shows the exemplary analysis of an interface between La_2CuO_4 and $SrTiO_3$, two insulating oxides of different polarity. Previous work based on the quantification of the phase of a reconstructed wavefunction in high-resolution TEM already suggested an intermixing in the cation sub-lattice [2]. StripeSTEM now reveals a terminating TiO_2 plane and that a considerable Ti concentration is present in the first two perovskite-like lanthanum-copper-oxide layers. Thereby, intermixing in the cation lattice is an important factor for the charge balance at this interface between a polar and a non-polar layered oxide, in addition to oxygen deficiencies, valency changes and free charge carrier accumulation.

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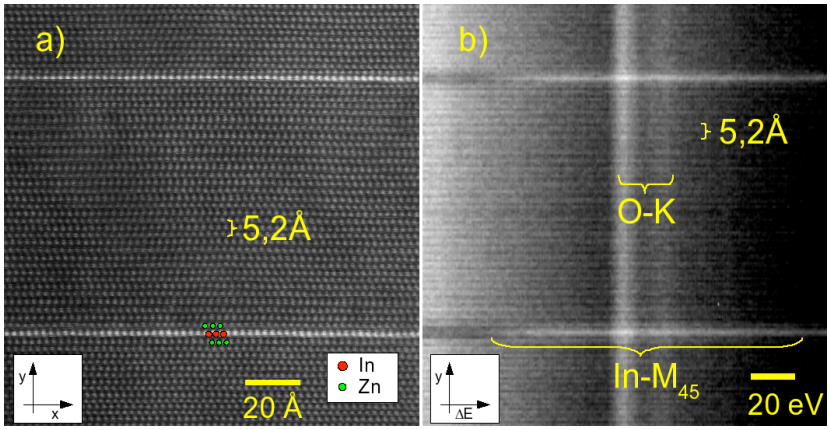

Figure 1. StripeSTEM measurement on monolayers of octahedrally coordinated In_2O_3 in a matrix of hexagonal ZnO. (a) High-resolution HAADF image. (b) The corresponding EEL spectrum-stack image shows the O-K and In-M_{45} core shell excitation. Each spectrum was taken with 1 s acquisition time, the total acquisition time was 165 s. The intensity modulation in the background just before the O-K and In-M_{45} edges is due to the excitation of the Zn M edge.

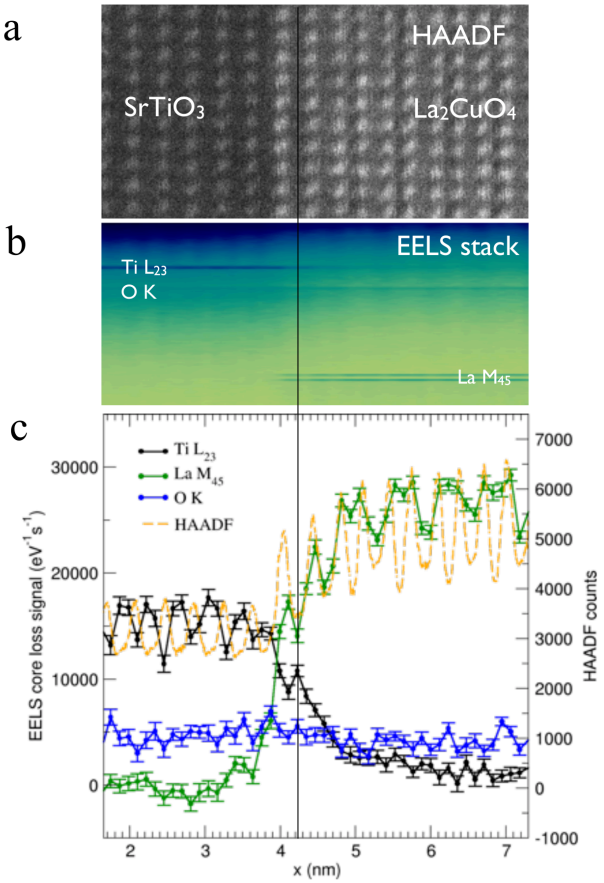

Figure 2. StripeSTEM experiment on the interface between $SrTiO_3$ and La_2CuO_4. (a) HAADF image. (b) Stack of EEL spectra. (c) Quantitative signal of core losses of Ti, O, and La after background subtraction. Error bars indicate the 1σ uncertainty due to noise and background extrapolation. The HAADF signal, integrated parallel to the interface, is displayed for comparison.