Towards atomic scale multi-elemental quantification based on inelastic electron scattering physics

Zezhong Zhang1,2,3, Annick De Backer1,2, Ivan Lobato1,2, Sandra Van Aert1,2, Peter Nellist3

1 EMAT, University of Antwerp, Antwerp, Belgium; 2 NANOlab Center of Excellence, University of Antwerp, Antwerp, Belgium; 3 Department of Materials, University of Oxford, 16 Parks Road, Oxford OX1 3PH, United Kingdom; 4 Rosalind Franklin Institute, Harwell Research Campus, Oxfordshire OX11 0FA, United Kingdom; Email: zezhong.zhang@uantwerpen.be

Keywords: Quantitative STEM-EDX, Inelastic electron scattering, Atom counting, Scattering cross-section

To understand the structure-property relationship in nanostructured materials, we need to probe their crystal structures and compositions at the atomic scale. Advanced materials usually consist of multiple elements in a complicated structure. Significant difficulties remain to disentangle the contributions of composition and thickness in STEM due to dynamic scattering, which needs to be taken into account by detailed simulations. However, the combination of the computational cost of the multislice calculation and the enormous ordering possibilities for a given composition makes the quantification of mixed columns almost impossible.

To address these challenges, we here report the development of incoherent non-linear method (atomic lensing model) for the fast prediction of ADF-EDX scattering cross-sections of mixed columns [1]. As shown in Fig.1 for Au@Pt nanorod, one cannot distinguish the elements with adjacent atomic numbers from an ADF image but can fingerprint them with EDX. Our model predictions are in good agreement with multislice simulations, which is very different from the linear model results that simply scales with the number of atoms. Based on the atomic lensing model and correlation between ADF-EDX due to signal incoherence, we are able to count the number of atoms from an experimental ADF-EDX dataset of Au@Ag core-shell nanoparticle [2]. The fast and accurate prediction of ADF-EDX scattering cross-sections opens up new opportunities to explore the wide range of ordering possibilities of heterogeneous materials with multiple elements.

The rich information of EELS comes from the complex inelastic scattering process where fast electrons transfer their energy and momentum to the atoms by exciting the associated atomic electrons to higher unoccupied state. To quantify EELS, the common practice is to fit the observed spectrum with the scattering cross-sections calculated from the experimental parameters and general oscillation strength (GOS) database. The previous GOS was based on the Hartree-Fock solution of atomic orbitals [3], which doesn’t include the full relativistic effects. For better quantification of EELS, we developed an inelastic scattering code for GOS database generation based on Dirac solutions. The newly developed database has a much larger energy-momentum space with fine sampling for EELS cross-section quantification. We also considered the relativistic electrodynamic effect of fast incoming electron on the orbital transition. We hope the Dirac-based GOS database can benefit the EELS community for both academic use and industry integration.

![Figure 1](image-url)

**Figure 1.** (a) Atomic model of the Au@Pt core–shell nanoparticle. (b) Comparison of the scattering cross-sections (SCS) quantified from multislice simulations, the atomic lensing model (ALM), and a linear incoherent model.


*The authors acknowledge financial support from the Research Foundation Flanders (FWO, Belgium) through Project No. G.0502.18N and a post-doctoral grant to ADB. This project has received funding from the European Research Council ((Grant Agreement No. 770887 PICO METRICS) and received funding from the European Union’s Horizon 2020 research and innovation programme (No. 823717 ESTEEM3).*