

## Computational Aspects in Quantitative EELS

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Electron energy loss spectroscopy (EELS) offers many opportunities for microanalysis on a nanoscale and even down to atomic resolution. Combining this technique with transmission electron microscopy provides a very flexible tool for modern nanotechnology. Increased stability and detection efficiency of the instruments brings us closer than ever to the theoretical limit of the amount of information that can be obtained per incoming fast electron. Efficient interpretation of this carefully obtained information has however lagged behind and intuitive visual interpretation is commonplace.

In this talk we will describe two main areas where we attempted to improve this situation and make the interpretation of EELS results more quantitative.

In a first part we will describe the quantification of EELS spectra as a statistical parameter estimation process. We can prove that in this way we obtain the most precise result that is obtainable from a given experiment with a certain amount of noise. The parameter estimation technique requires to build a physical model with parameters which can be fitted to experimental data. When the noise model for the detection process is known, three types of output are obtained:

- an estimate of the parameters, which can be the concentrations of certain elements in an EELS experiment.
- an estimate of the precision, which depends on the amount of noise in the experiment.
- a validation of the model that was used. This indicates whether the model is statistically acceptable for describing the experiment.

This method has been implemented in a software program, EELSMODEL which is freely available to the public under a GNU licence [1,2]. We demonstrate that this method not only provides a precision up to three times better than conventional EELS quantification methods but also provides remarkably accurate results which have the potential to make EELS a standardless technique [3].

Apart from obtaining accurate and precise concentration estimates, we also can expand this method to low loss EELS and valency determination. In low loss EELS we demonstrate that we can build a parametrised model for the dielectric loss function which allows to take into account retardation effects and multiple scattering. Fitting this model to experimental low loss spectra is shown to give reasonable estimates for the complex dielectric function even for diamond which typically is problematic because of retardation effects. Currently the method is however limited to bulk-like samples with negligible surface losses. Model based quantification can also be applied to accurately estimate the formal valency in ionic materials. We will show the first results of an automated technique which is based on measuring the excitation edge onset difference between the O K-edge and the transition metal L23 edge. The advantage of this method is that it doesn't rely on the detailed shape of the L23 edge which makes it more suitable for automation.

In a second part of the talk, we will discuss the effect of elastic scattering on atomic resolution EELS [5-8]. We demonstrate a computer program which is able to calculate the combined elastic and inelastic scattering of fast electrons in a material for both energy filtered TEM and STEM EELS setups [8]. The so-called double channeling approach is used which models a single inelastic scattering event in a crystal where multiple elastic scattering occurs before and after this event. We show that delocalisation effects can seriously influence the apparent origin of an EELS signal which makes intuitive interpretation fail. This non-local imaging effect is comparable to the interference effects in conventional bright field TEM and makes image simulation necessary to interpret experiments. Only in very favorable cases a local contrast is approximately obtained. Example simulations will demonstrate these effects. Expansion of the simulations towards the simulation of chiral transitions in EELS will be discussed.

### References

- [1] J. Verbeeck, Van Aert, S., *Ultramicroscopy*, 101 2-4 (2004) 207-224.
- [2] [www.eelsmodel.ua.ac.be](http://www.eelsmodel.ua.ac.be)
- [3] J. Verbeeck, G. Bertoni, *Microchim Acta*, 161 3-4 (2008) 439-443.
- [4] L. Zhang, S. Turner, F. Brosens, and J. Verbeeck, *Phys. Rev. B*, 81 (2010) 035102
- [5] L.J. Allen et al., *Ultramicroscopy* 106 11-12 (2006) 1001-1011.
- [6] M. Bosman et al., *PRL* 99 (2007) 086102.
- [7] C. Dwyer, S.D. Findlay, L.J. Allen, *Physical Review B* 77-18 (2008) 184107-1-1.
- [8] J. Verbeeck, P. Schattschneider, A. Rosenauer, *Ultramicroscopy* 109-4 (2009) 350-360.
- [9] The FWO-Vlaanderen is acknowledged for financial support under contract nr. G.0147.06. and G.0064.10N as well as financial support from the European Union under the Framework 6 program under a contract for an Integrated Infrastructure Initiative. Reference 026019

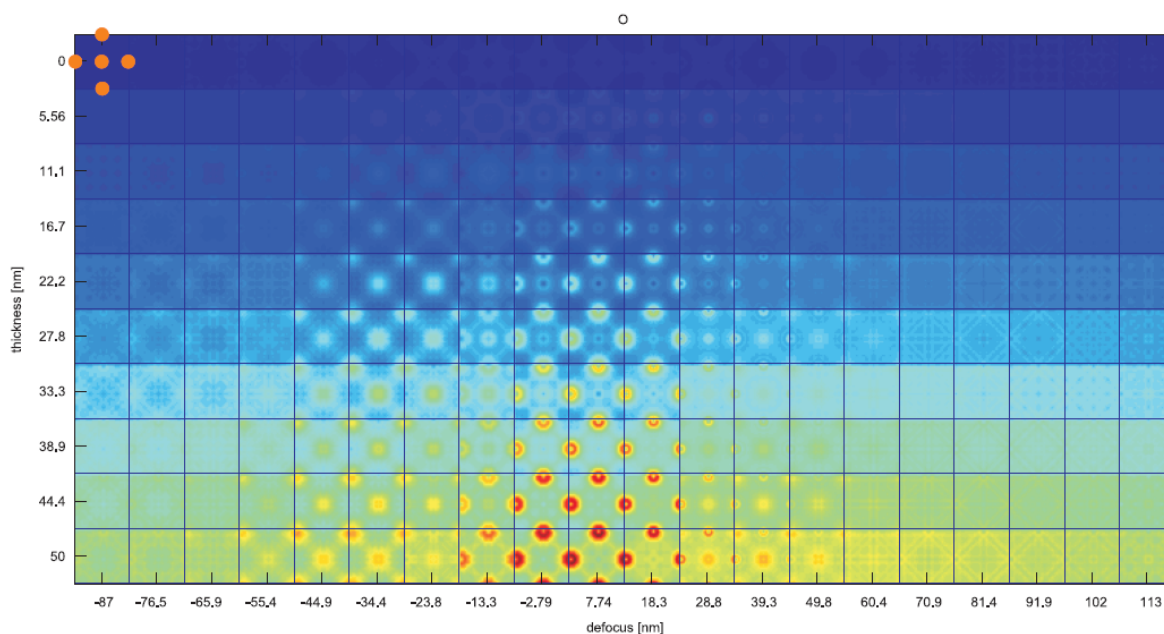


FIG. 1. Simulated O K-edge EFTEM image for SrTiO<sub>3</sub> in 100 direction showing the effect of thickness and defocus. The orange dots in the upper left corner indicate the true positions of the O atoms. Only at very low thickness of the sample and correct focus, we get local contrast. For higher thickness the central atom disappears depending on the focus which indicates strong nonlocal effects.