Systematic Study of Background Subtraction Techniques for EELS

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Introduction

- EELS is complex in nature due to the presence of the zero-loss peak (ZLP) including phonons, plasmon scattering, near-edge structures (ELNES) and extended fine structures (EXELFS). These influence the extraction and quantification of core-loss edges through background subtraction.
- For extraction of core-losses the conventional method modelling the background only in pre-edge region is sometimes problematic and can even cross spectrum.
- Need to explore and study the statistics of inverse power-law background (AE^{-r}) models in other regions such as post-ionization range
- Two different extrapolation models provide under- and over-estimate of core-losses. An optimal background is modelled based on error bars of Poissonian statistics.
- Experimental EELS of GaAs is from region 3 with t/λ=1 [1] is used as an example. ($V = 197kV, \alpha = 16.6mrad, \beta = 15mrad$).

Background Models

Pre-edge region

- Inverse power-law fits are modelled in the pre-edge region (range >30eV).
- For As-L edge even though the R^2 values are in good agreement, the modelled background is crossing the spectrum as shown in figure 1, due to preceding Ga-L edge.
- Background crossing the spectrum predicts a negative core-loss, which is unphysical.
- As-L edge can still be quantified by integrating only the positive core-loss range.
- The Ga-L edge is straight forward as the it has very large pre-edge region.
- The background fits in pre-edge region are highly associated with large **systematic errors** if the integration ranges are large.
- Systematic errors are difficult to identify by regression and quantification.
- In figure 2, even though the background is crossing the spectrum, the Ga/As ratio is still close to unity.

Post-edge region

- Inverse power-law fits are extrapolated in the post-edge range from end of the spectrum and offset vertically to cross through the edge onset.
- With post-edge background modelling, As-L edges have very large apparent cross sections as shown in figure 1.
- This indicates an over-estimate of the core-loss edge.
- The Poissonian statistical error bars are very large.
- The Ga-L edge is not straight forward as it has very small post-edge region with varying gradient compared to pre-edge region.
- To extrapolate post-edge inverse power-law of Ga-L edge from end of the spectrum, As-L edge has to be subtracted from the spectrum.
- The background fit in post-edge regions are highly associated with **statistical errors** if the integration ranges are small.
- Statistical errors are difficult to identify in R^2 and quantification.
- In figure 2, even though the background is over-estimate the core-losses Ga/As ratio is still close to unity.

Optimal fit

- The inverse-power-law fits in pre-edge and post-edge regions provide under-estimate (B_u) and over-estimate (B_o) of the core-loss edge.
- The solution is to select backgrounds which are physically meaningful (yields positive core-loss) and have smaller error bars.
- Hence an optimal background (B_{opti}) may be given by the equation:

$$B_{opti} = \frac{\left(B_u - \sqrt{I_u(\Delta)}\right) + \left(B_o + \sqrt{I_o(\Delta)}\right)}{2}$$

where $\sqrt{I_u(\Delta)}$ and $\sqrt{I_o(\Delta)}$ are the statistical error bars associated with under- and over-estimate respectively.

- The upper (E_u) and lower error (E_l) bars associated with optimal background fit are:

$$E_u = \left(B_o + \sqrt{I_o(\Delta)}\right) - B_{opti}$$

$$E_l = B_{opti} - \left(B_u - \sqrt{I_u(\Delta)}\right)$$

- The error bars associated with optimal background are smaller when compared to Poissonian statistics.
- The quantification of the Ga/As ratio in GaAs is close to unity with less systematic and statistical errors.

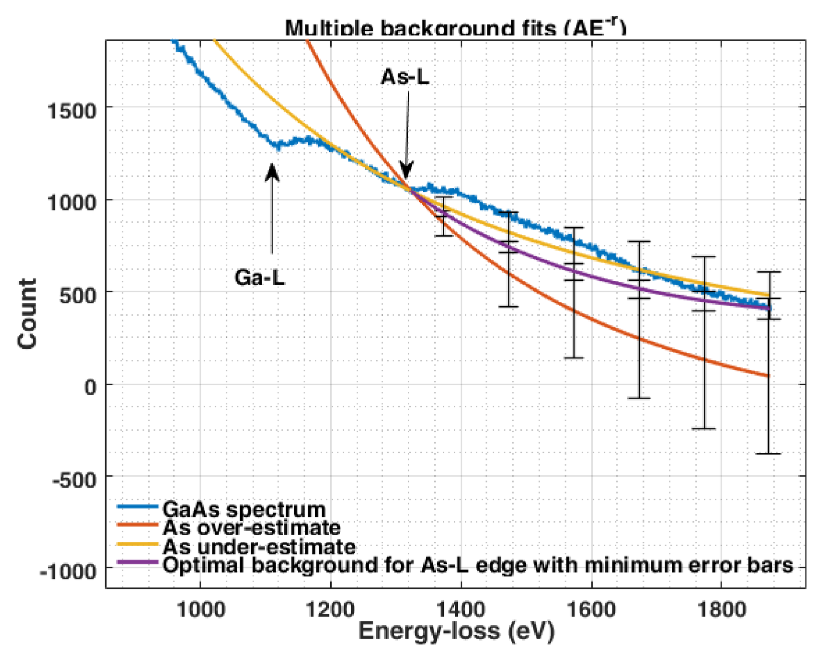

Fig 1: Experimental EELS of GaAs with t/λ=1 with different background fits with error bars for As-L (Ga-L is more straight forward).

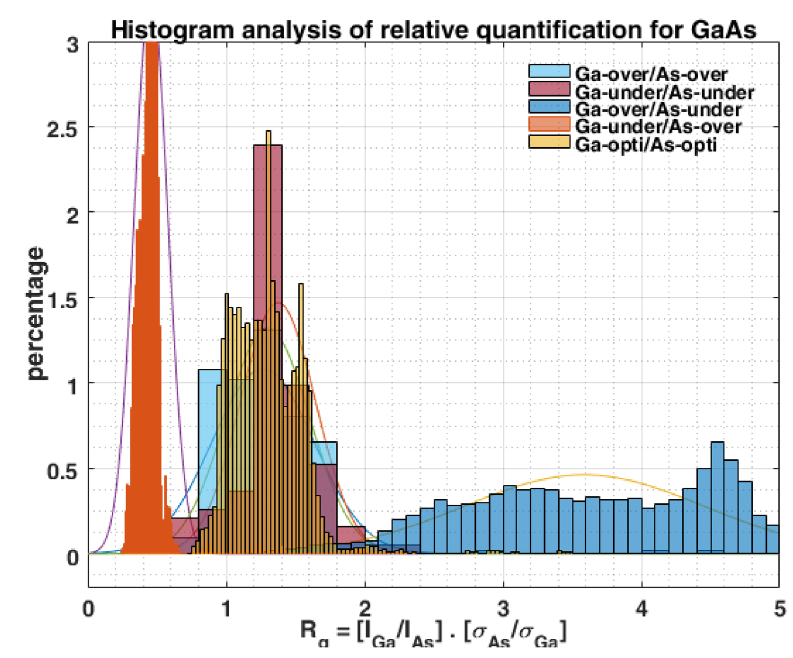

Fig 2: Normalized histograms of Ga/As quantification from all possible background subtraction routines. The combination of optimal background fitting yields $\mu = 1.2759$ $\sigma = 0.2703$

Conclusion

- Improvements in R^2 values do not guarantee more accurate quantification.
- The optimal background modelling provides quantification values with error bars below counting statistics.
- The optimal background can be used to extract core-losses from spectrum and the underlying core-losses can be quantified with better statistics using larger integration ranges (Δ) [2,3].

References

[1] V. C. Angadi, C. Abhayaratne, T. Walther. (2016), J. Microscopy, in print, doi:10.1111/jmi.12397.

[2] R. F. Egerton, Electron energy-loss spectroscopy in the electron microscope. Springer, 1996, vol. 233.

[3] Digital Micrograph. [Online]. Available: http://www.gatan.com/

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