Comparing approaches to helium quantification by EELS

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1. INTRODUCTION

EELS is widely recognised as the only analytical technique that can detect and quantify helium present in nanometric bubbles in solids. Its unique combination of high spatial resolution and sensitivity to the UV spectral range make it particularly well adapted to this problem. Difficulties nevertheless arise when attempting to quantify the amount of helium in a given bubble. The main problem is the strong EELS signal exhibited by most solids over the range 21-25 eV in which the helium K excitation occurs. This is particularly the case in iron-based alloys (and most other cfc metals) whose plasmons (by far the strongest spectral signals in EELS) are at maximum intensity precisely in this range. The problem is therefore one of extracting the true He signal from this unwanted and intense background. Since such materials are of practical importance in reactor design, and He bubbles produced as a result of radiation damage are well known to cause fragility, the problem is of considerable practical interest. In the presentation we will compare several approaches, namely spatial difference (not shown here), curve fitting and the multivariate analysis (MVA) approach known as independent components analysis – a form of "blind source separation" (BSS).

2. RESULTS

2.1 Experimental conditions

All experiments reported here were carried out in a Nion Ultrastem 200 microscope operating at 100kV. The materials studied are ODS steels containing nanoparticles of $Y_2Ti_2O_{7-x}$ [1] irradiated with He^{2+} ions at diverse temperatures and fluxes. A Princeton EM Pro CCD camera with very fast acquisition times (>2000 spectra/s) allows collection of the unsaturated zero-loss peak without blanking, considerably simplifying the quantification process. FIB sample preparation is preferred for these iron-based materials to avoid magnetic perturbations in the microscope column. All analysis was performed using the Hypserspy suite [2].

2.2 Curve fitting

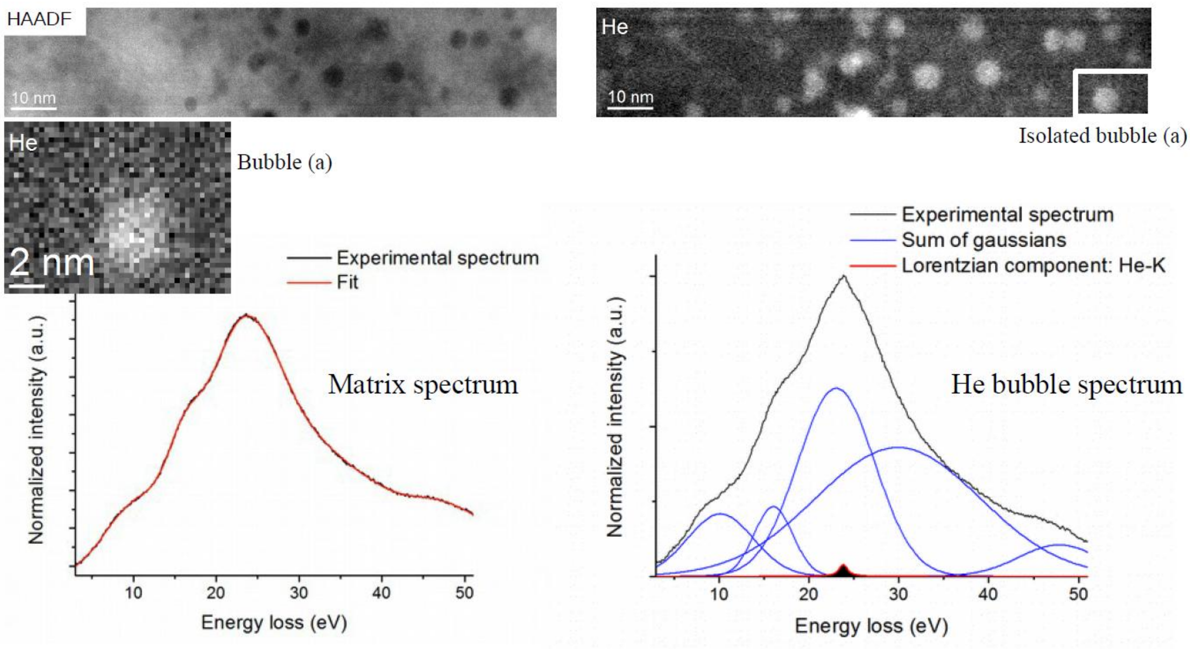

Figure 1. Illustrating the fitting process and the extraction of the He signal map (top right)

A typical example of the curve-fitting approach is summarized in figure 1 The He signal can be seen to be extracted quite satisfactorily. Statistical analysis confirms high confidence levels for the fits at nearly all pixel positions (see presentation).

2.3 Independent components analysis (ICA)

ICA aims to separate signal mixtures into their independent sources. It can be viewed as an extension to the better-known principal components analysis (PCA). Indeed, PCA must first be applied to remove noise and reduce the dimensionality of the dataset to a manageable number of components (usually >10) which are then "unmixed" using BSS algorithms based on various statistical principles (including maximum entropy amongst others; see presentation). It was first applied successfully to EELS data to separate overlapping edges in the core-loss spectral region [3]. The low-loss region remains so far unexplored by this approach. A non-trivial problem is knowing where to cut off the data (how many components to retain). Figure 2 shows the results of an analysis which appears to yield only two signal-bearing components. As can be seen, the helium signal, although very prominent in the second component, has not been completely isolated from the bulk signal.

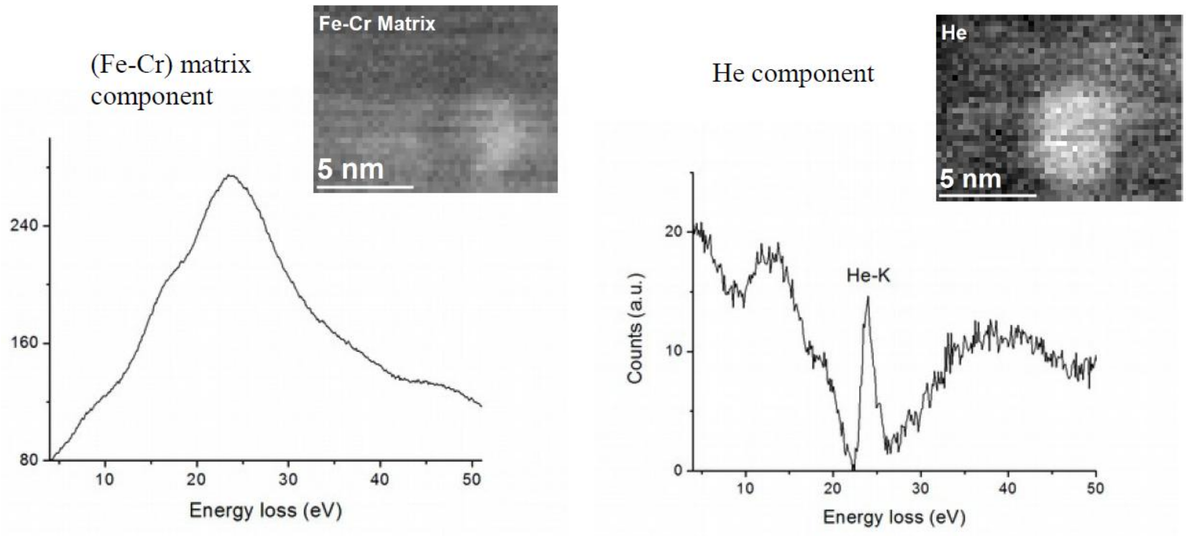

Figure 2. Two "independent components" from a spectrum-image and their intensity distributions

2.4 Comparison

The curve-fitting approach gives values for the He densities in line with those found in similar previous work. (up to 100 at.nm^{-3}). The ICA analysis appears systematically up to now to yield lower values (but which are still in the typical range). More details on this will be given in the presentation, but it seems likely that genuine signal information is being lost in the PCA and or ICA process. Even small thickness variations can lead to highly non-linear features in the data due to multiple energy losses. Deconvolution has thus far proved ineffective in completely eliminating these effects and introduces noise problems of its own. A number of other multivariate approaches exist and some are under investigation as an alternative to ICA (see presentation).

3. CONCLUSION

Curve-fitting appears thus for the moment still to represent the most reliable method for quantifying the helium signal in EELS spectra. Multivariate techniques are potentially superior but suffer too much from the effects of noise and non-linearity to be robustly reliable at present.

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