SPECTRUM-IMAGE: THE NEXT STEP IN EELS DIGITAL ACQUISITION AND PROCESSING

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This paper defines a new concept in EELS digital acquisition and processing: the spectrum-image. This is a 3D array of nxnxn numbers, the first two axes of which correspond to the x - y position on the specimen as for any image, while the third is associated to a complete energy-loss spectrum. New images and spectra can be extracted as sections or projections of this matrix, and their respective aims and advantages will be sketched. Novel types of processing are also evoked. This approach is closely related to the parallel detection units for EELS spectroscopy now available: despite their great size, these spectrum-images can be handled with normal computing technology, and an example of data compression is introduced to illustrate how far problems in storage can be solved.

1. Introduction

There are two ways of collecting and displaying EELS data in the electron microscope [1,2] which are illustrated in fig. 1. The first method consists of selecting a specimen area, either with an aperture in the TEM mode or with a fixed probe in the STEM configuration, and recording the relevant spectrum. The local information is then distributed over a 1D sequence of s channels, which we can call “spels” for spectrum-elements: this is the electron energy-loss spectrum (fig. 1a). The alternative is to select an energy-loss band in the spectrum and record the corresponding image. The information is now distributed over a 2D matrix of numbers (i.e. the n x n “pixels” or picture-elements, fig. 1b). This is the energy-filtered image which can be directly obtained with a filtering microscope of the Castaing-Henry type [3] or with a STEM equipped with a spectrometer.

Ten years ago, several authors [4,5] pointed out that true chemical maps could only be extracted in EELS from a sequence of energy-filtered images around a core-loss edge. The pre-edge images are used to estimate the non-characteristic background, for each pixel, which then has to be subtracted from the energy-loss signal above the edge. Practical examples have been published during the past few years after the early results of Rez and Ahn [6] and Jeanguillaume et al. [7]. It then

Fig. 1. Pattern of the different conventional objects: spectrum, 2D image, 3D image and their digital data representations.

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became clear that the reliability of the data is improved if more images are recorded, and in recent work, Leapman and Ornberg [8] use about 20 or 30 rather than three. Up to now, all acquisitions have been made serially by scanning over energy loss. It has not been possible to acquire large numbers of images because of considerations of total-dose problems or lengthy recording time. The success of parallel EELS detectors [9-11] obviously opens up new possibilities. The purpose of the present contribution is to introduce a new concept in EELS digital acquisition, the spectrum-image which is particularly well adapted to the results furnished by parallel detectors in STEM. As shown in fig. 2a, the spectrum-image is a 3D array of numbers: the first two axes correspond to the \( x-y \) position on the specimen as for any image, while the third one is associated with the complete energy-loss spectrum. By analogy with the \( n \times n \times n \) "voxels" (volume-elements) handled in 3D imaging (see fig. 1c) we propose to call the elementary components of the spectrum-image "pispels", for picture-spectrum-elements, and there are \( n \times n \times s \) of them in the spectrum-image. In the following paragraphs we shall point out the interest and advantages of this new approach, then describe our projects in this field and finally discuss the questions of feasibility and practical solutions.

2. Definition of a few basic objects emanating from image-spectrum concept

One problem encountered in 3D images is to visualize all the data, and this is also a problem for spectrum-images. By analogy with 3D images, one can use either sections or projections through the whole set of numbers, as is common with tomography data. To improve the depth perception, one often displays a temporal succession of the data corresponding to different views. In the present case the three axes do not correspond to equivalent parameters, the spectrum type information being fundamentally different from the spatial one.

Conventional sections can then be classified either as \( s \) elementary images (fig. 2b) or \( n \times n \) elementary spectra (fig. 2c). More unusual sections can be described, such as the line-spectrum consisting of a 2D area of \( n \times s \) pispels (fig. 2d). They are of interest for problems with a specific variation along one direction (i.e. EELS spectra at different points through an interface [12] or at different energy loss values).
Summed spectrum

Summed image

Fig. 4. Schemes exhibiting the definition of summed spectra and summed images.

different points along an axis in a diffraction pattern [13,14].

Projections are summations along a given axis: the most common one is the projection along the energy-loss axis, which amounts to the total or unfiltered image (fig. 3a). A projection along one spatial axis corresponds to a summation of line-spectra parallel to the feature of interest in the image: it can be of great help to increase the signal (fig. 3b). Finally the double projection along the two spatial axes gives the total spectrum (fig. 3c).

A more complex way of handling the data is to combine sections and summations, as illustrated in fig. 4. A first example consists of defining an area of interest in an image and summing the spectra for the pixels within this area: this is a summed spectrum (fig. 4a). Another suggestion is to select windows or slices at different energies of interest and to add them to obtain a summed image (fig. 4b).

3. Aims and advantages

For each of the basic objects introduced there-above, we briefly discuss their novel contribution.

3.1. Elementary sections

In EELS spectrum acquisition, the recording time is divided into two parts, the recording itself and the storage of all parameters necessary for post-processing of the data. This second task is greatly reduced in the spectrum-image because it occurs only once for \( n \times n \) spectra.

The search for a given feature in an EELS spectrum is often very difficult if it is localized within a small specimen area. If all the spectra recorded from large regions are available, it is much easier to find a given feature by displaying a time sequence of 5 to 10 spectra per second. This also improves the localization after detection because it is easy to compare a posteriori the spectra with the position of the pixel in the image.

Similar arguments related to time-saving during recording, improved display and better energy definition, can be applied to the elemental images as well.

3.2. Projections

Projection is a standard manipulation, but we now have the possibility of aligning the energy-loss scale, or the position axis, before summation to compensate for energy drifts. The projection line-spectrum has not been extensively used up to now, but many new applications can be envisaged depending on the problem being considered.

3.3. Summed spectra

This is an unexplored area where connections between image and spectra can be made and morphology and chemistry can be related. As an example, one can delineate a particular “area of interest” in an image and sum all spectra originating from pixels within it. This offers great advantages if the area has a distinctive shape such as a porcupine, the branch of a tree in winter or a fractal object. A standard aperture or limited scan of the area of interest would provide spectra in which the interesting features were of low visibility because of the background contribution of the matrix. The definition of the area of interest can be achieved through an interactive procedure by...
drawing a line on the image. It can also be done automatically by following an intensity contour. This type of approach also has many applications when correlating spectra with features in the electron diffraction pattern, such as satellite or superstructure spots or diffuse scattering outside the Bragg spots.

An alternative way of benefiting from this mode is when the localization of an element is not known a priori. It is better to first calculate a chemical map and then to sum the spectra over a region of interest. This procedure enhances the signal-to-noise ratio for the edge being considered and opens up new possibilities, such as fine structure studies.

3.4. Summed images

Image summing consists of building new images which are the sum of individual images obtained through different energy-loss channels. Many applications are possible, some of them having already been used with less sophisticated techniques: total inelastic image, plasmon image, large band image for thick specimen.

3.5. Original processing schemes involving the full spectrum-image

With the addition of two extra planes of data corresponding to the incident beam measured by an electrometer and to the annular dark field signal detected through the annular detector channel, it is possible to devise a number of quantitative a posteriori processing schemes. Some of these have already been described [15,16].

3.5.1. Detection efficiency and mean free path ratio

Before interpreting the result of an intensity measurement in terms of scattering cross-section or mean free path (λ), the detection efficiency must be accurately known. Detection efficiency is governed by several effects: the geometrical factor which takes into account the optics of the column and the physical dimensions of the detectors and the detection quantum efficiency (DQE) of each detector and its amplification chain. As shown in ref. [15], the total efficiency factors can be determined from a reconstruction of the incident beam image (which shows fluctuations or decay) as the sum of the signals collected by all the detectors after suitable scaling. A knowledge of the complete EELS spectrum for each channel gives a better measurement of the inelastic image and is an important step for the calculation of a λi/λe image.

3.5.2. Chemical maps

Here again, the spectrum-image is an important advance for several reasons:

(a) Chemical maps can be obtained simultaneously from different edges and different elements in one acquisition.

(b) The difficult compromise between a reduced dose and improved background modeling, which governs the detection limit in the present serial acquisition mode, is much less stringent. The fact that many points of both background and background + signal in the energy-loss domains are available reduces the extrapolation error.

(c) Quantitative chemical mapping is made possible by taking into account multiple-loss deconvolution, elastic scattering correction, incident beam modification, removing of the surface term by Kramers–Kronig analysis.

3.5.3. Novel applications

We make no claim to predict all the possible uses of spectrum image data. However, one can imagine that all parameters accessible from spectral processing will provide new images (thickness (t), elastic mean free path (λe), inelastic mean free path (λi)) and all functions accessible will produce a new spectrum-image (energy-loss function (−Im(1/ε)), dielectric constant (ε1, ε2), optical function (n, k, μ), effective number of electrons (n eff)).

The technique might also lead to improvements in the search for an element and consequently the detection limits. In the absence of a priori knowledge of the elemental composition, one can try many elementary associations of spectra 2 by 2, 3 by 3, etc., and apply all types of identification schemes either revealed on second derivative spectra, or by correlation with reference spectra to
these summed spectra. These tasks can be achieved by a computer overnight!

4. Feasibility and practical considerations

The total volume of data contained within a spectrum-image is of course dependent on the size of the spectrum and image involved. Table 1 provides some useful numbers. It shows that all cases are presently under the storage capacity of common magnetic disks. In our laboratory, the size of the computer memory permits a data volume of 1.5 Megawords (i.e. up to $24 \times 256 \times 256 \times 16$ bits) to be acquired in one image scan. This would be sufficient for all spectrum-images within the unshaded part of the table. An improvement in hardware or direct recording on magnetic disc would cover almost all the table.

The other important instrumental factor is the total recording time, which is essentially governed by the limited intensity of useful signal for each pixel. It depends on the cross-section involved, and for core losses a minimum dwell time per pixel is typically 10 ms. For a $32 \times 32$ image, the recording time then amounts to 10 s; for a $256 \times 256$ image, it corresponds to ~10 min and so on. In a sequential mode for the energy-loss scan, a sequence of 20 or more $128 \times 128$ images requires more than one hour. It appears that the performance in sensor read-out time or analog-to-digital conversion does not introduce any limitation.

Finally a few comments may be made concerning practical aspects of the construction of a well-designed digital acquisition unit. Digital control of the electron beam scan is recommended as well as readout of a parallel detection device, and all its associated electronic circuitry. In order to obtain a wide dynamic range, a separate plastic scintillator followed by a light pipe and a photomultiplier

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Table 1

<table>
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<th>128x128</th>
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Fig. 5. Example of data compression on an EELS spectrum, using the algorithm described in the text (anatase specimen, courtesy of T. Manoubi) full energy scale from 0 to 1000 eV: (a) original spectrum, (b) data compression chart, (c) expanded spectrum after data reduction (from 1028 to 120 channels).
collects the unscattered signal, the low-energy-loss spectrum is recorded on an indirect diode array through a YAG converter, while the high-energy-loss signal is directly measured. We suggest that the pispel signals be collected and stored temporarily in a fast memory for spectrum summation when required. The correction for the dark current is also made at this stage either uniformly or with the aid of a correction matrix.

Data compression is another possibility to be considered in the context of parallel EELS acquisition. In a spectrum, not all channels have the same importance. Some parts are only used for integration or extrapolation, but it is important to retain all the information. In those regions, the operator can decide that a channel in the compressed spectrum represents 4 or 8 channels in the original one by their mean value. Such a processing makes it possible to retain all the information in spectral regions of specific interest such as near-edge fine structure. Fig. 5 shows a comparison between the original spectrum (5a) and the compressed one (5c), together with the compression chart (5b). It demonstrates that with only 120 channels almost all the useful EELS information is retained.

Finally the spectrum-image is recorded in a file with the experimental acquisition parameters, the compression curve and the image statistics.

5. Conclusion

We believe that the spectrum-image is a necessary step forward in the field of analytical data acquisition and processing. Spectrum-images are easy to obtain with the new parallel recording devices, and can be handled correctly with existing computing facilities, and we think that the first results will start appearing in the near future.

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References