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Optimizing EELS acquisition

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Abstract

A method for spectral acquisition, called binned gain averaging, will be described and tested. Systematic or correlated noise is efficiently suppressed with this method by averaging the gain over a series of CCD pixels. As a result, improved signal-to-noise ratios are obtained that allow the detection of very weak signals. At the same time, the spectral energy resolution is not degraded—even for long acquisition periods. It will be demonstrated that with this method, it is possible to significantly enhance the acquisition speed and quality of electron energy-loss (EEL) spectra and EELS maps. Examples will be given of double ionic scattering (i.e. the detection of the second boron K-edge) and the mapping of gold surface plasmons in the near-infrared and visible energy range.

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1. Introduction

Electron energy-loss (EEL) measurements usually contain artifacts that compromise the quality of the spectra. These artifacts occur in all circumstances, whether the EEL spectrum of a specimen is measured or only a vacuum signal. Most obvious are the outlying spectral values—best known as 'X-ray spikes'. More subtle are the artifacts introduced by incomplete correction of detector gain (relative pixel sensitivity) and dark current (thermally excited electron–hole pairs), and finally, there is the unavoidable random spectral noise whose intensity solely depends on counting statistics. Correction of these artifacts is generally done using automated software routines. Gain and dark references are automatically acquired and used; outlying spectral values are removed by replacing them with the local median value and random noise is minimized by increasing the acquisition time. Here, we will show that artifacts from automated dark and gain corrections result in systematic—i.e. correlated—noise and impose significant limitations on the quality of EEL spectra. Before an alternative acquisition routine will be introduced, these artifacts will be briefly discussed.

A first source of systematic noise is the non-uniform illumination of the EELS detector during the acquisition of a gain reference image. This effect is particularly strong for dedicated scanning transmission electron microscopes (STEMs) and will lead to inaccuracies mainly at the edge of the spectra. Secondly, X-ray spikes or other outlying detector counts that occur during the acquisition of reference images will introduce inaccuracies in the spectra for which these references are used. Finally, the acquisition time for a reference image may be too short to suppress the random noise signal to appropriately low levels. The latter is undoubtedly the most important source of inaccuracies with cumulative acquisition and—more importantly—with the acquisition of EEL spectrum images [1,2].

The ubiquitous Gatan Digital Micrograph (DM) software allows the efficient acquisition of these sets of spatially distributed EEL spectra. Just before the acquisition of a spectrum image starts, a dark reference is acquired without detector illumination, under the same conditions with which the EEL spectra will be acquired. However, the

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acquisition time per spectrum for EEL spectrum images is usually less than a few tenths of seconds, and here is where the artifacts are introduced: the noisy dark reference that is acquired with this relatively short acquisition time is subsequently used for dark current subtraction in *all* spectra. The individual spectra in the EEL spectrum image do not appear to have artifacts, but when a large number of them are summed, it is clear that the channel-to-channel variation in the spectra is highly correlated.

Despite best efforts to correct for these artifacts in the spectra, some systematic noise will remain when conventional acquisition approaches are used, as illustrated in Fig. 1. For this experiment, a spectrum image was acquired and from two different regions where no specimen was present, 170 EEL spectra were summed and these summations are displayed as spectra in Fig. 1(a) and as a scatterplot in Fig. 1(b). The exposure time for each of the 170 spectra was 0.15 s; a high-quality gain reference was used together with an automatically acquired dark reference. Ideally, the summations should give spectra with intensity zero and random noise fluctuations. However, the close-up of the spectra and the scatterplot over the whole spectral range show that both summations are strongly correlated. The summation has reduced the relative intensity of the random noise, but not of the systematic noise.

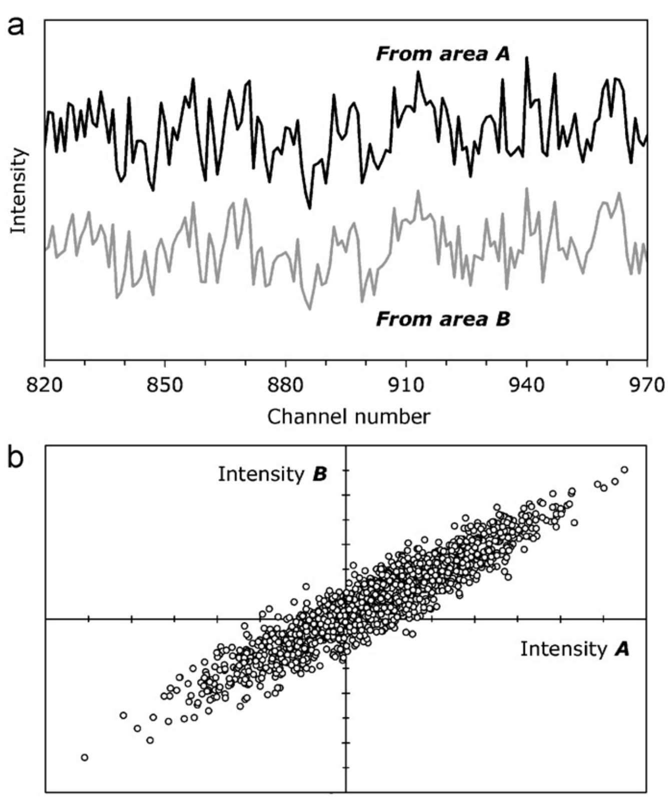

Fig. 1. Non-optimal results from conventional acquisition. Summations of 170 EEL spectra are plotted, taken from two different areas of the same spectrum image where no specimen was present. A high-quality gain reference was used and no CCD binning during readout. (a) A small part of the spectra is shown to focus on the channel-to-channel variance. The spectra are vertically shifted for clarity; the intensity fluctuates around value zero. (b) Scatterplot of the whole energy range of the same two spectra, again showing their strong correlation.

The effect shown in Fig. 1 greatly limits the application of EEL spectrum imaging. Systematic or correlated noise will prevent subtle spectral features from being resolved, even after long total acquisition times. Multivariate analysis on these data sets is also hampered; the systematic noise will always be extracted, mixed within the first few significant spectral components, and unlike random noise, it cannot be filtered from the data because it occurs in all spectra.

We will define gain variance as the channel-to-channel variation in a spectrum due to systematic and random noise. The subject of this paper is correlated gain variance that results from systematic noise due to the inaccurate dark and gain reference corrections. Its existence limits the signal-to-noise ratio (SNR) and therefore the quality of EEL spectra. Here, a modified acquisition method will be presented that can significantly improve the SNR and—in addition—will enhance the EELS acquisition speed per spectrum. This gives many advantages such as better energy and spatial resolution and less specimen drift, damage or contamination. Throughout this paper, we will give experimental examples from a VG 601UX STEM with Gatan Enfina; however, the principles we discuss will also be applicable to other parallel EELS detectors [3].

2. Optimizing the acquisition procedure

It has been suggested by Hicks et al. [4] and later demonstrated by Shuman and Kruit [5] that the method of gain averaging optimizes the SNR in EEL spectra. With this method, the location where EEL spectra are acquired is shifted to different channels of the parallel detector, after which they are aligned by correcting for the known imposed shifting value. A modified version of this method was proposed by Boothroyd et al. [6], based on an iterative correction of the gain in different channels. Schattschneider and Jonas [7] demonstrated that with the same input spectra, this 'iterative gain averaging' method would provide a further enhanced SNR compared to the method of gain averaging. In this work, a more intuitive and easily automated method will be discussed based on the non-iterative method of gain averaging. It is insightful to first briefly consider the way in which EELS data are acquired in modern CCD-based detectors [8].

With EELS, electrons that have interacted with a thin specimen and lost some of their initial speed are dispersed in energy and projected onto a thin scintillator screen. Here, they are converted to photons which are then relayed to the CCD chip, where electron–hole pairs are generated. After a user-defined time period, the electrons are 'blanked' from falling onto the scintillator and during this beam-blanking time, the charge in the CCD is read out. The readout of the CCD occurs by shifting the charge into the register, a separate row of pixels. Row-by-row, the charge is moved into the register, each row of charge then