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J.A. Hunt

Gatan Inc., 5794 W. Las Positas Blvd., Pleasanton, CA 94588 USA

In recognition of David Williams' recent induction into the ranks of septuagenarian, it is appropriate to recognize TEM and STEM techniques developed under his direction, as well as postulate future directions from these works. Development of STEM spectrum-imaging is such an example, which has become the primary method for producing EELS quantitative maps of compositional and electronic properties.

Starting in 1987 there was a close collaboration to develop the first STEM EELS spectrum imaging capability between Dave Williams and the author (at Lehigh University) and Chuck Fiori and Richard Leapman (at the National Institutes of Health). The first Gatan parallel-detection EELS system[1] was to be delivered to NIH that year, and soon another to Lehigh. Parallel-detection revolutionized the EELS technique and initially enabled collection of 80 spectra/second, a rate too fast to adequately process on-the-fly with accessible computer systems at that time. Batch collection and disk storage of "imaging spectra" acquired at each pixel of a digital image, followed by tools to iteratively process this data to produce quantitative elemental maps became a necessity to make use of this new detector. In 1988, the initial maps of 128x128 pixels required heroic patience of my colleagues as the developed tools were crude (users had to write in Fortran), and acquisition and processing were done on PCs uncomfortably limited to 640KB of memory and 80MB hard drives. Processing several EELS edges took hours and productivity was limited to a few data sets per month.

In late 1988 the Colliex group presented their vision of "spectrum-imaging"[2] and we soon adopted their more elegant nomenclature. Other groups were also working on spectrum-imaging capability, notably the Weiss et al.[3] work which was the early roots for the popular EMISPEC spectrum imaging platform.

The first detailed presentation of our system was at this conference series[4] and by this time it had become apparent that spectrum-imaging techniques provide significant advantages beyond unlinking acquisition and processing time. It allowed searching for features that were unknown a priori, and processing the same data set with a variety of techniques and parameters until optimum processing was found. Less obviously it provided a vehicle to take advantage of the statistics of thousands of data points from within a spectrum-image to measure achievable systematic and non-systematic errors, and to use this for technique development. We had become interested in developing spectrum-imaging methods to extract maximum information from these data sets, as well as designing experiments to push detection limits. Initial focus was on MLLS for trace (<100 ppm) elemental quantitation and chemical quantitation, and NLLS for plasmon energy quantification of Li in AlLi alloys and H in metal alloys. Processing times for these capabilities were published in 1991[5] and showed that processing a typical EELS edge in a 128x128 pixel SI took 11 minutes.

Active development of this system continued through 1993 and several design philosophies emerged. Spectrum-image visualization and processing was simplified by assuming that prior correction was performed for all detector artifacts, spatial, energy & current drifts and artifacts. When this pre-processing was successful, spectrum-image data sets could then be sliced and integrated over any dimension to produce consistent spectra. All metadata from acquisition and subsequent processing was archived. Maps of all possible errors and processing coefficients was available for each processing step. These design decisions and the feature sets became the template for the Gatan spectrum imaging packages.

Work at Gatan on a spectrum imaging package for DigitalMicrograph began in earnest in 1995 and by 1998 it had reached its first level of maturity with a generalized suite of acquisition (EELS, EFTEM, EDS) and on-the-fly acquisition corrections (camera artifact correction, spatial drift, energy drift, current drift, EELS chromatic figure), sub-pixel scanning (avoids under-sampling), data visualization by integrating pixels and/or energy-slices, and processing methods for EDS and EELS data sets including LLS, MLLS, and NLLS fitting.

Although this feature set looked similar to what was published in [4], the commercial version was significantly more polished and capable. Users were encouraged to build upon this framework with the ability to add or replace Gatan's acquisition code with a user's DM script via "script-callbacks" that would be called at the start and/or end of every pixel, spectrum acquisition, or artifact correction. The DM scripting language was extended to provide user script access for slicing and viewing up to 5D datasets to encourage users to improve existing processing methods. Data acquisition and processing was restricted to RAM. Macintosh and PC RAM at this time was uncomfortably limited to 2GB, which effectively limited spectrum-image data sizes to 1GB.

Over the next twenty years the Gatan spectrum imaging capability has significantly evolved in capability and performance (e.g., [6] & [7]), but essential workflows remain consistent with the system developed at Lehigh and NIH. At present, thousands of installations of Gatan's spectrum imaging tools are in active use. Spectrum images of 128x128 pixels can be collected and elemental maps processed in seconds – a several hundred-fold improvement since 1991. Spectrum-imaging has been performed using essentially every spectroscopy related to electron microscopy, some even in 3 spatial dimensions[8]. Diffraction-imaging has become commonplace[7]. CL SI has been performed in 4D per pixel (emission angles, wavelength, polarization)[9].

In future, we expect much of the routine collecting and processing of spectrum image data to be managed by machine learning and eventually AI systems. Development of these systems will require large training sets that will likely be derived from spectrum-images of individual components acquired or synthetically created to simulate variation of experimental parameters such as energy, collection angle, dose, specimen thickness and diffraction conditions.

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