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High speed/low dose analytical electron microscopy with dynamic sampling

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ABSTRACT

Technological advances in electron microscopy, particularly improved detectors and aberration correctors, have led to higher throughput and less invasive imaging of materials and biological structures by enhancing signal-to-noise ratios at lower electron exposures. Analytical methods, such as electron energy loss spectroscopy (EELS) and energy dispersive x-ray spectrometry (EDS), have also benefitted and offer a rich set of local elemental and bonding information with nano-or atomic resolution. However, spatially resolved spectrum imaging with EELS and EDS continue to be difficult to scale due to limited detector collection angles or high signal background, requiring hours or even days for full maps. We present the principle and application of a Multi-Objective Autonomous Dynamic Sampling (MOADS) method which can accelerate spectrum mapping in EELS or EDS by over an order of magnitude. Initial guesses about the true spectrum images are constructed as measurements are collected, which allows the prediction of points which contribute information/contrast. In this fashion, an intelligently selected and reduced set of points which best approximate the true spectrum image are autonomously collected on-the-fly to save considerable time and/or radiative area dose. We implemented MOADS as a software add-on to arbitrary commercial Scanning Transmission Electron Microscopes (STEMs) equipped with a Gatan Digital Micrograph (DM, Gatan ©) interface. We demonstrate the efficacy of our proposed method on several prototypical analytical specimens, as well as dose sensitive materials. It is expected that MOADS and similar supervised dynamic sampling approaches may open the exploration of large area analytical maps or the imaging of beam reactive materials not previously thought feasible.

1. Introduction

Newer generations of electron microscopes have dramatically increased spatial resolution and analytical sensitivity by *focusing* ever higher electron currents into smaller and smaller volumes (Erni et al., 2009). Studies of a wide variety of materials which utilize new high brightness and high coherence electron sources now commonly report atomic resolution imaging and atom-by-atom elemental mapping with Electron Energy Loss Spectroscopy (EELS) and Energy Dispersive X-ray Spectrometry (EDS) (Chu et al., 2010; Kimoto et al., 2007). Commercially available MEMS-based sample holders, which can encapsulate the sample in a small volume of controllable fluid or gas, have also become widely available for most microscopes with accessible EDS or EELS signals (Holtz et al., 2013; Kelly et al., 2016). Spatially resolved EDS and EELS spectra, referred to as spectrum images (SI), have continued to be applied to the exploration of soft and hybrid soft/hard materials, often answering questions about complex and nanoscale processes unaddressable through other imaging modalities (Que et al., 2017; Wu et al., 2013). In contrast to energy filtered TEM, these spectrum images provide 'fine structure' which can be used to resolve bonding configuration and/or other information about local electronic structure (Leapman and Ornberg, 1988; Muller, 2009).

Although increased radiative fluence with a fixed cross section for signal generation has had analytical advantages (Hutchison et al., 2005; Watanabe et al., 2006), the limited detector collection angles for EDS and high background signal in EELS have prevented the routine application of spectrum imaging at reasonable doses/times outside of specialized labs. In particular, the predilection of soft, hybrid, and *in-situ* systems to be damaged under incident radiation has been an unresolved problem compounded by the increasing brightness of modern electron sources (Egerton et al., 2004; Egerton, 2011). Radiation damage with higher available beam currents and the longer acquisition times for improved pixel resolution and field of view further exasperate an already complex problem of dose-induced damage/changes to ultrastructure of materials (Hitchcock et al., 2008). Improvements in detectors have somewhat mitigated this challenge, but remain an

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expensive investment with relatively slow-moving improvements (Bammes et al., 2012; Ruskin et al., 2013). Performing additional correlated measurements after spectrum imaging, such as high-resolution imaging or nano-diffraction, is often impossible due to the distributed area dose involved in conventional spectrum imaging. Larger area or higher fidelity spectrum images of dose insensitive materials also provide challenges, as the hours or days required to complete a full map may exceed the stability of the microscope or specimen. This is especially true for *in-situ* systems, where dynamics often proceed at much faster rates (Luo et al., 2014; McDowell et al., 2012). A significant need exists for improved methods of collecting spatially resolved EELS and EDS signals at area doses low enough for the preservation of sensitive structures or subsequent investigation with a correlated technique, and/or for the acceleration of conventional spectrum images for the investigation of spatially confined biological-inorganic complex structures, engineered hybrid soft/hard ordered materials, or responsive and dynamic reconfigurable nanostructures.

Recently, algorithmic approaches have been proposed to substantially reduce the collection time or radiative dose of conventional electron, x-ray, and other radiation-based probe instruments (Choi et al., 2010; Kovarik et al., 2016; Lustig et al., 2008; Sang et al., 2017; Stevens et al., 2013). The central idea of these methods is that there is considerable redundancy in most experimental data, which can be disregarded without loss of fidelity (Candes et al., 2006). Sparse imaging involves an *under-sampled* acquisition of the signal in real space (sometimes incorrectly referred to as *compressive sensing*) (Anderson et al., 2013; Béché et al., 2016; Kovarik et al., 2016). This redundancy in combination with prior information is used to impute reasonable values for the missing entries in the image, which potentially reduces the area dose and the acquisition time. Significant redundancy means statistically accurate reconstructions from sparse acquisition can be achieved for sampling percentages low enough to reduce sample damage (Hujsak et al., 2016). It is important to note that in the case of a scanned probe, the local fluence at a single pixel in an under-sampled image is equivalent to the conventional case. However, the reduction in total area dose may allow for the subsequent investigation with a correlated technique on pristine sample in the same region-of-interest or the reduction of diffusing and damaging radical species in *in-situ* systems due to beam-media interactions. Recently, it has also been shown that for small electron probes and particular energy losses, the region of primary energy deposition in which sample damage occurs is smaller than the point spread function (PSF) for inelastic scattering (Egerton, 2017). Sparse or under-sampled EELS acquisitions may therefore take advantage of this property to maximize the volume of signal unperturbed by the beam (Egerton, 2017). So far, in all the cases described above, the scan mask is 'fixed' before observing the specimen and is referred to as static, or non-adaptive.

Instead of pre-choosing which measurements to make, dynamic sampling is performed 'on-the-fly' (also referred to as adaptive). As data is collected from a mobile probe interacting with the specimen, an algorithm uses the previously collected data to predict the next best measurement to make. By intelligently adjusting the position of the probe after each sequential acquisition, an accurate approximation of the true image can be obtained at radically lower area exposures/collection times (Godaliyadda et al., 2014; Scarborough et al., 2017). Supervised methods based on simple linear regression have also shown strong performance at minimal computational cost (milliseconds) (Godaliyadda et al., 2016, 2017). Simulations on scanning electron microscopy (SEM) EDS images have recently been carried out using a similar approach but have not been followed up with a practical implementation (Zhang et al., 2018). In addition, this previous work could not map multiple overlapping elements or continuously valued spectrum images (only integer class-based maps) and required the training of a dedicated neural-network for fitting the spectra for each potential sample. A more robust approach based on direct mapping of elements, rather than binned 'material' classes, is much needed for routine applications .

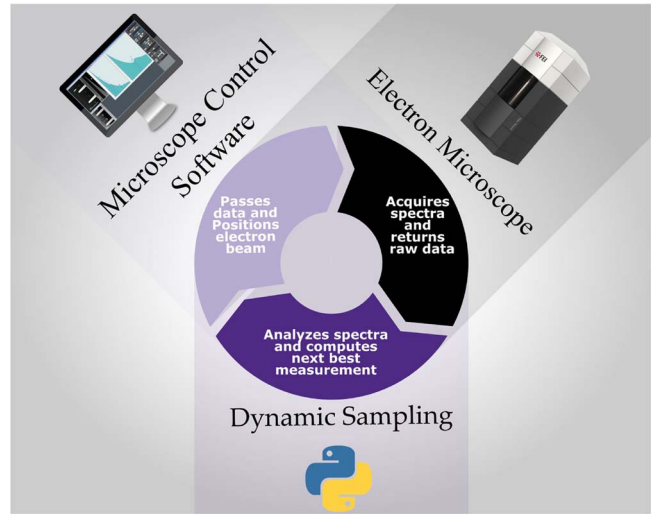

Fig. 1. A flow chart describing the interaction of MOADS, Digital Micrograph or other Microscope Control Software (TIA Envision, JEM Toolbox, etc.), and the physical microscope. All direct calls to the microscope hardware are made through the manufacturer's control and COM interface, which translates the requested measurement from the dynamic sampling code from relative coordinates to the physical DAC commands to the scan coils. After acquiring a spectrum, the raw data is sent to a support PC running the dynamic sampling code, which analyzes each spectrum and performs the necessary steps to estimate the next best measurement. In this way, the manufacturer software acts as a safety buffer, preventing bad calls to the microscope hardware.

Herein, we will show that spectrum imaging in analytical EM is amenable to dynamic sampling, where abrupt interfaces or continuous changes can be mapped with considerably fewer measurements. In particular, our multi-objective form of dynamic sampling allows for independently resolving multiple elements simultaneously, while previous methods relied on classifying groups of signals into discrete classes (Godaliyadda et al., 2017). Such an approach may potentially allow for very large spectrum images at acquisition times in the minutes, as opposed to hours or longer. In the current rendition, our approach is drop-in ready and autonomously operable on any STEM equipped with a Digital Micrograph (DM®, Gatan Inc.) control system.

2. Materials and methods

2.1. Multi-objective dynamic sampling description

Our goal in dynamic sampling will be to minimize the error between a ground truth (the true fully sampled) image X and an approximation $\hat{X}$ by sequentially sampling a reduced set of pixels that best represent the true image. In this section, we will define the relevant terminology and process in an intuitive fashion, leaving the mathematical formulation for Appendix A. At any point during an experiment we will have a set of k values sampled from the grid of pixels of X which we can interpolate to form $\hat{X}$. Our interpolation will have some error in its estimation X, which we refer to as the distortion between the interpolant and the ground truth $D(X, \hat{X}^{(k)})$. In our case we calculate D as the cumulative pixel-by-pixel error between the two images. In principle, adding any additional measurement $k + 1$ should give us more information about X and reduce the distortion, D. However, we assume that some pixels are strongly correlated with their neighbors and thus the reduction in distortion is not uniform for all possible pixels (some pixels contribute more information than others).

If we knew the ground truth image X, we could simply calculate the pixel which offers the maximum reduction in distortion by enumerating through all the possible choices and calculating D directly. During an experiment we do not have access to the ground truth elemental distribution, and thus we would like to calculate an *expected reduction in distortion*, or ERD, for all possible pixels to sample. Our second

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