Ultramicroscopy 138 (2014) 46–56

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Optimized imaging using non-rigid registration

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ARTICLE INFO

Article history:
Received 11 September 2013
Received in revised form
12 November 2013
Accepted 15 November 2013
Available online 25 November 2013

Keywords:
Non-rigid registration
Si-Y zeolite
Diffusion registration
IQ-factor
HAADF-STEM

ABSTRACT

The extraordinary improvements of modern imaging devices offer access to data with unprecedented information content. However, widely used image processing methodologies fall far short of exploiting the full breadth of information offered by numerous types of scanning probe, optical, and electron microscopies. In many applications, it is necessary to keep measurement intensities below a desired threshold. We propose a methodology for extracting an increased level of information by processing a series of data sets suffering, in particular, from high degree of spatial uncertainty caused by complex multiscale motion during the acquisition process. An important role is played by a non-rigid pixel-wise registration method that can cope with low signal-to-noise ratios. This is accompanied by formulating objective quality measures which replace human intervention and visual inspection in the processing chain. Scanning transmission electron microscopy of siliceous zeolite material exhibits the above-mentioned obstructions and therefore serves as orientation and a test of our procedures.

1. Introduction

"Learning from data" has become an indispensable pillar of science of ever increasing importance. Data acquisition can be broadly broken down into two categories, parallel acquisition and serial acquisition. In the case of serial acquisition when the change of signal between data points encodes the information of primary interest (e.g. images acquired one pixel at a time) extraction of the information from the raw data is difficult to achieve. Obstacles include low signal-to-noise ratios, possible changes of the observed object due to the observation process, and – perhaps commonly less acknowledged – uncertainties in the positioning of the observations. As a consequence, the various stages of data processing still often involve a high degree of subjective human intervention. This is particularly true, because standard methods cannot handle highly complex multiscale motion of the observed object. A representative example, providing the main orientation for this work, is *scanning transmission electron microscopy* (STEM). We are convinced though that the proposed methodology is *scale-invariant* and therefore relevant for a much wider scope of application.

The tremendous instrumental advances in STEM technology open new perspectives in understanding nanoscale properties of materials. However, in particular when dealing with beam sensitive materials, a full exploitation of this technology faces serious obstructions. The acquisition of the images takes time during which both material damage as well as environmental disturbances can build up. A major consequence is a significant – relative to the scale under consideration – highly complex motion intertwined with specimen distortion.

In response we propose a new data assimilation strategy that rests on the following two constituents:

(i) replacing single frame high-dose data acquisition by taking multiple low-dose, necessarily noisy, frames;
(ii) properly synthesizing the information from such a series of frames by a novel cascading registration methodology.

A few comments on the basic ingredients are needed. Taking a single high-dose frame may in principle feature a higher signal-to-noise ratio but typically increases the possibility of irreparable beam damage, prevents one from accurately tracking the combination of local and global motion during the scanning process, and possibly

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http://dx.doi.org/10.1016/j.ultramic.2013.11.007

introduces additional unwanted physical artifacts. We use STEM imaging of a water-containing siliceous zeolite Y (Si-Y) as a representative guiding example, since it poses many of the obstacles which occur in practice when determining the structure and function of important classes of materials. In particular, Si-Y mimics biological matter by rapidly deteriorating when exposed to high energy electron beams. Furthermore, the registration task for Si-Y poses difficult hurdles for registration which are often encountered in imaging materials, including a high degree of symmetry and low variety of structures in the presence of low signal-to-noise samples.

The core constituent (i) above, i.e. taking several low-dose short-exposure frames of measurements from the same physical locations of the specimen, allows us instead to better resolve the complex motion and nonstationary distortions. We demonstrate how the information from multiple frames can be used to offset such distortions as those described.

A critical ingredient of core constituent (ii) is to align the series of frames. Extracting improved information from a series of low quality images is a well-known concept, usually termed *super resolution* in the imaging community. It rests in an essential way on the ability of accurately tracking inter-frame motion, typically using feature extraction. However, the low signal-to-noise ratios prohibit accurate feature extraction. Moreover, the physical nature of the motion encountered in a series of STEM images is of a highly complex nature that can therefore not be treated satisfactorily by standard techniques. The extreme sensitivity of STEM to the measuring environment gives rise to motion which appears to exhibit at least three scales [1]: jitter on the atomic level, contortion on the unit cell level, and a macro drift. As explained later in more detail, short time exposures provide the opportunity for a sufficiently refined temporal discretization. Furthermore, being able to generate highly accurate pixel to pixel mappings, in contrast to fiducial-based registration in imaging, seems to be the only hope to avoid significant blurring at the image assimilation stage.

The few existing approaches in the STEM context resort to manual *rigid* alignment (see e.g. in tomography [2] and single particle analysis [3]). Although in our experiments, from a subjective point of view, rigid alignment appears to yield good results, they are, however, as is typical, confined to a relatively small image portion that is implicitly used as a feature substitute. First, this introduces a highly subjective and therefore non-reproducible component into the data processing chain. Second, it wastes and even obscures information about the observed object carried by large parts of the image series. In particular, in STEM there is often high interest in determining possible irregularities in the atomic structure outside the small registration zone.

In order to be able to make full use of the information carried by the frames and, in particular, to be able to reliably detect material imperfections away from the subjective reference region we propose a powerful *multilevel non-rigid registration* method accurately linking the information carried by many sequentially acquired low-dose frames. This gives rise to data assimilation without significant blurring. Furthermore, it serves an important universal objective, namely to replace "human weak links" in the data processing chain by more robust, highly accurate, quantifiable, and reproducible processing modules. In this context a further important issue is to formulate and apply quantitative objective quality measures to illustrate the effectiveness of the procedures.

2. Materials and methods

2.1. Zeolites as proxy materials

Zeolites are beam sensitive, crystalline aluminosilicates consisting of AlO_4 and SiO_4 corner-sharing tetrahedra, whose arrangement allows for a large variety of structures with different pore sizes and pore connectivities. These materials are known to deteriorate rapidly, due to a combination of radiolysis and knock-on damage [4–6], when exposed to electron beams generated by accelerating voltages between 60 and 200 kV.

Knock-on damage occurs when the fast electrons impart sufficient energy to the atoms in the specimen to displace them from their equilibrium positions, thereby directly breaking the long-range order of the material. *Radiolysis* occurs when the fast electrons ionize the atoms in the material which then subsequently lose their long-range order as they seek a lower energy state. The ionization of water by the electron beam is thought to be a key step in the damage mechanism [5,6]. Several excellent review papers on the application of transmission electron microscopy to the study of the structure of zeolites and other mesoporous materials have been recently published [7–9].

Our choice of zeolites as proxies for imaging beam-sensitive materials is based on four properties: (i) high crystallographic symmetry and smaller unit cell dimensions which, compared to biological objects, simplify the quantification of results and allow us to focus on technique development, (ii) the presence of water, (iii) reduced structural variety and (iv) high beam sensitivity. Each of these properties poses a significant challenge for the registration task.

2.2. STEM imaging

STEM of zeolites has been used generally for either the high-contrast imaging of small metal nanoclusters within the pores using the high-angle annular dark-field (HAADF) technique or high spatial resolution chemical analysis using either energy dispersed X-ray (EDX) spectroscopy or electron energy loss spectroscopy (EELS) [10,11]. Aberration-corrected STEM allows for the formation of sub-Å probes with greatly increased brightness [12] which results in images with substantially better signal-to-noise ratio. The aberration corrector nearly eliminates the electrons in the probe outside the central maximum which results in much more dose-efficient imaging.

Ortalan et al. used an aberration-corrected STEM to image the location of single atoms of Iridium in a zeolite using a combination of low-dose image acquisition methodology and both Fourier filtering and real-space averaging [13]. The reported dose is similar to that in our work.

For our experiments HAADF STEM images at 200 kV were recorded with a JEOL JEM 2100F TEM/STEM equipped with a CEOS CESCOR aberration corrector. All axial aberrations of the electron wave were measured and corrected up to third order. Fifth order spherical aberration was minimized as well. The illumination semi-angle was 15.5 mrad, which at 200 kV yields a nominal probe size of 0.1 nm. The HAADF detector recorded electrons scattered between 50 and 284 mrad. The probe current was 10 pA as measured with a picoammeter attached to the small fluorescent focusing screen of the microscope. The pixel size was 0.3 $Å^2$ and dwell time per pixel was 7 μs yielding an electron dose for the images of $\approx 1400\ e^-/Å^2$. Image acquisition was controlled by a custom script in Digital Micrograph (Gatan, Pleasanton, CA) which sequentially recorded and saved a series of HAADF STEM images 1024×1024 in size.

Our material test sample is a siliceous zeolite Y (cf. [14]) which was completely de-aluminated. The Si-Y zeolite powder was dispersed onto an amorphous carbon holey support film on a copper mesh TEM grid. A small particle was located and oriented along the $\langle 110 \rangle$ direction of the zeolite Y structure. Fig. 1 shows the first and ninth frames of a series of HAADF STEM images of the Si-Y zeolite sample collected with the conditions described above. Because of the relatively low electron dose the individual images in Fig. 1 are quite noisy. The largest pores of the zeolite Y structure