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Self-adapting denoising, alignment and reconstruction in electron tomography in materials science

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ABSTRACT

An automatic procedure for electron tomography is presented. This procedure is adapted for specimens that can be fashioned into a needle-shaped sample and has been evaluated on inorganic samples. It consists of self-adapting denoising, automatic and accurate alignment including detection and correction of tilt axis, and 3D reconstruction. We propose the exploitation of a large amount of information of an electron tomography acquisition to achieve robust and automatic mixed Poisson–Gaussian noise parameter estimation and denoising using undecimated wavelet transforms. The alignment is made by mixing three techniques, namely (i) cross-correlations between neighboring projections, (ii) common line algorithm to get a precise shift correction in the direction of the tilt axis and (iii) intermediate reconstructions to precisely determine the tilt axis and shift correction in the direction perpendicular to that axis. Mixing alignment techniques turns out to be very efficient and fast. Significant improvements are highlighted in both simulations and real data reconstructions of porous silicon in high angle annular dark field mode and agglomerated silver nanoparticles in incoherent bright field mode. 3D reconstructions obtained with minimal user-intervention present fewer artefacts and less noise, which permits easier and more reliable segmentation and quantitative analysis. After careful sample preparation and data acquisition, the denoising procedure, alignment and reconstruction can be achieved within an hour for a 3D volume of about a hundred million voxels, which is a step toward a more routine use of electron tomography.

1. Introduction

Electron Tomography (ET) is a non-invasive technique to perform 3D imaging of objects of a few hundred nanometers in size with a spatial resolution of about one nanometer [1–3]. A Scanning Transmission Electron Microscope (STEM) is used to acquire images or projections of the object at different tilt angles. Ideally a great number of projections should be acquired over a 180° angular range. After careful alignment of all the projections, a mathematical algorithm is used to reconstruct the 3D object from its 2D projections.

In ET, the direct model can be formulated as:

$$\mathbb{A}\bullet\boldsymbol{x}=\boldsymbol{b} \tag{1}$$

Here $\mathbb{A}$ is the projection matrix, $\boldsymbol{x}$ is the vector containing the voxels of the 3D object and $\boldsymbol{b}$ is the vector containing the pixels of the 2D projections. To reconstruct $\boldsymbol{x}$, an algebraic reconstruction such as Simultaneous Iterative Reconstruction Technique (SIRT) is often used in ET due to its reliability [4]. A SIRT is actually a classical steepest descent algorithm minimizing the residual error $\|\mathbb{A}\bullet\boldsymbol{x}-\boldsymbol{b}\|_2^2$. The initialization of this well-known algorithm starts with $\boldsymbol{x}^{(0)}=0$. At each iteration k, the solution $\boldsymbol{x}^{(k)}$ is updated according to the formula:

$$\boldsymbol{x}^{(k+1)}=\boldsymbol{x}^{(k)}-\lambda^{(k)}\mathbb{A}^T\bullet\left(\mathbb{A}\bullet\boldsymbol{x}^{(k)}-\boldsymbol{b}\right) \tag{2}$$

In Eq. (2), $\mathbb{A}$ is applied to a volume $\boldsymbol{x}^{(k)}$ and is equivalent to a forward projection while $\mathbb{A}^T$ is applied to a set of projections and is equivalent to a back-projection. The relaxation parameter $\lambda^{(k)}$ can be fixed or changed at every iteration.

Due to damage under electron beam and limited acquisition time, the number of projections is often limited to roughly one projection per degree. This subsequently leads to an infinite

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number of possible solutions for Eq. (1). Regularization by the application of *a priori* knowledge can be used to counterbalance this lack of information. This could be the application of a positivity constraint for SIRT, or by dedicated algorithms such as Discrete tomography (DT) [5] or Total Variation Minimization (TVM) [6] which apply further constraints, *e.g.* piecewise constant object for TVM, a finite number of known grey levels corresponding to the number of materials present in the object for DT.

In Compressed Sensing (CS) theory, a sparsity of the solution of an ill-posed inverse problem is searched in a particular dictionary such as Fourier space, wavelet dictionary, or even the gradient or finite differences – in which case it is called TVM. TVM has been used lately for ET [7–9] because it largely improves the quality of 3D reconstructions, particularly when an angular range of 180° cannot be reached, resulting in a missing wedge. Still, CS suffers severe drawbacks: Many influential parameters must be fixed by the user to get "the best" reconstruction, making the results highly user dependent. Moreover, experimentally finding the correct parameters can be time-consuming and tricky. In addition to this, solving a CS problem needs far more calculations and thus a reconstruction may be four times longer than a SIRT reconstruction [8] even with an efficient solver like TVAL3 [10].

SIRT with positivity constraints is relatively liberal in terms of constraints on the solution yet still yields high quality reconstructions in simulations without noise, with perfect alignment and without missing wedge and with only one projection per degree. Finally, in the case of a needle-shaped sample allowing for 180° angular range, noise and misalignment are often the most critical issues that limit the reconstruction quality. Denoising projections before the reconstruction can therefore be useful for both the alignment and the algorithm reliability. After careful alignment, a SIRT with positivity constraints can be applied to reconstruct the object with high accuracy.

In this paper, we introduce a complete methodology composed of self-adapting projection denoising, automatic and accurate alignment and reconstruction, particularly suited for needle-shaped samples in ET in materials science. In section II the denoising method using Undecimated Wavelet Transform (UWT), the alignment and the SIRT reconstruction are introduced. Three-dimensional simulations and real data acquisitions of a porous silicon sample and agglomerated silver nanoparticles – used for inkjet printing technology – are presented. Section III compares reconstructions obtained with the complete pre-processing of the projections to reconstructions obtained without denoising and with a simpler alignment made with cross-correlations.

2. Materials and methods

2.1. Porous silicon

2.1.1. Porous silicon sample

Porous silicon [11] has attracted a lot of interest since its discovery in 1956 [12] thanks to many applications ranging from electronics and optoelectronics [13], photonics [14,15] and to chemical and biomedical applications [16]. All of these applications take advantage of the different geometry and morphology of porous silicon [17] that can be obtained by varying the fabrication parameters and substrate doping level and type. ET is useful to locally determine the empty to full ratio, the distribution, size and shape of the porous structure as it is not possible to extract all of this information from a single 2D STEM projection. The porous silicon sample characterized here is prepared by electrochemical etching of (100) – p^+ oriented crystalline silicon wafer with a resistivity in the range 3–7 mΩ cm. The etching solution used is a HF:H_2O:Ethanol in a ratio of 30:30:40. Porous empty to full ratio is 55% [20].

2.1.2. Sample preparation for ET

The sample was prepared with a FEI™ Strata dual-beam Focused Ion Beam (FIB) instrument. A classical method to prepare needle-shaped sample in ET has been used [18]. First, protective layers were locally deposited on the surface of the sample to protect it from damage from incident gallium ions. Layers deposited on the surface consist first of an electron-deposited silicon-dioxide layer, then of an electron-deposited tungsten layer and finally, an ion-deposited tungsten layer. Once the region of interest is protected, a chunk is extracted using an Omniprobe®. The chunk is glued to a tip holder. It is then milled to get a needle shape using gallium ions of the FIB. Size of the annular milling is progressively decreased as well as the current of the gallium beam to get a needle-shaped sample with a diameter of around 100 nm at the top of the needle and a diameter of around 300 nm after one micron.

2.1.3. STEM HAADF tilt series acquisition

Tilt series is acquired with a FEI Titan Ultimate using an accelerating voltage of 80 kV. A convergence angle of 8 mrad is used as a compromise between resolution and depth of focus. The current of the probe was set approximately to 0.1 nA. The camera length used is 145 mm. 177 images of 2048 × 1248 pixels with a pixel size of 0.41 nm and an exposure time of 10 s are acquired between −88° and +88° using the FEI software Xplore3D™.

2.2. Agglomerated silver nanoparticles

2.2.1. Silver nanoparticle sample

Since the beginning of the twenty first century, inkjet printing technology has been extensively explored for printed flexible electronics applications. Its capability to deposit and pattern without contact and solid masks drastically reduces the process complexity for low cost electronics [19]. Advances in ink formulation have led to a large range of printable materials that were developed for several types of possible applications such as photovoltaic cells [20], light-emitting diodes, sensors [21,22], Radio-Frequency devices [23] and even smart clothing [24]. Colloidal suspensions of metallic nanoparticles are really good candidates for printing conductive patterns because of their high electrical conductivity

The sample studied here was made using SunChemical EMD5714 ink; a silver nanoparticle ink with 40 wt% silver in an ethanol and ethylene glycol solvent [22,23]. According to the supplier, the mean nanoparticle size is 70 nm. A layer of about one micron thickness is printed on a 500 nm thick thermal oxide on a silicon substrate using a Dimatix DMP2831 printer. Characterization is important to better understand the properties of this sample. Because of the size of the nanoparticles used, ET is adapted to obtain the 3D morphological parameters such as roughness, grain size and porosity.

2.2.2. Sample preparation for ET

Just like the porous silicon sample, the agglomerated silver nanoparticle sample was prepared with an annular milling process using a FEI Strata dual-beam FIB instrument. This time, the annular milling is made to get a constant diameter of around 300 nm in the region of interest of one micron height.

2.2.3. STEM IBF tilt series acquisition

Tilt series have been acquired with a FEI Titan using an accelerating voltage of 200 kV in Incoherent Bright-Field (IBF) mode [25]. Classical tomography reconstruction in STEM HAADF mode is not possible with this sample due to intensity reversal that occurs with high mass-thickness samples [9–25]. A convergence angle of 8 mrad is used as a compromise between resolution and depth of