Ultramicroscopy 150 (2015) 65–70

Contents lists available at ScienceDirect

Ultramicroscopy

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

Prospects of linear reconstruction in atomic resolution electron holographic tomography

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ARTICLE INFO

Article history:
Received 29 September 2014
Received in revised form
3 November 2014
Accepted 29 November 2014
Available online 3 December 2014

Keywords:
Electron holography
Tomography

ABSTRACT

Tomography commonly requires a linear relation between the measured signal and the underlying specimen property; for Electron Holographic Tomography this is given by the Phase Grating Approximation (PGA). While largely valid at medium resolution, discrepancies arise at high resolution imaging conditions. We set out to investigate the artefacts that are produced if the reconstruction still assumes the PGA even with an atomic resolution tilt series. To forego experimental difficulties the holographic tilt series was simulated. The reconstructed electric potential clearly shows peaks at the positions of the atoms. These peaks have characterisitic deformations, which can be traced back to the defocus a particular atom has in the holograms of the tilt series. Exchanging an atom for one of a different atomic number results in a significant change in the reconstructed potential that is well contained within the atom's peak.

1. Introduction

Akin to any other transmission probe, Transmission Electron Microscopy (TEM) accumulates its signal while the probe (i.e. the electron wave) travels through the sample. This prohibits that the signal can be interpreted in terms of the three-dimensional specimen information (bar some special cases). Similar problems exist in many other fields as well (e.g. in X-ray microscopy, medical imaging or atmospheric sounding), so that tomography, the reconstruction of three-dimensional data from its projections, is a technique of great interest. While mathematicians started to develop the foundations [1,2] at the beginning of the 20th century only the advent of computers made its widespread application feasible [3,4]. Naturally, tomography also found its way into electron microscopy [5], where it made the three-dimensional characterisation of a multitude of specimen [6,7], from cell structures [8] to nanoparticles [9], possible.

One particular kind of tomography in a TEM is Electron Holographic Tomography (EHT) [10,11], which uses the complex wave information that can be retrieved by holography. The phase of the electron waves gives direct access to electric and magnetic potentials and EHT has been successfully used to reconstruct such potentials at the nanometer scale [12–14]. If it could be extended towards atomic resolution it could give further insight into the local structure of very small specimen. This currently attracts increasing interest as both nanoparticles and nanostructures with ever higher complexity are made. Due to their size, they differ strongly from bulk materials and their properties can only be explained when their crystal structure is known. Since that is dominated by small defects (e.g. point defects or grain boundaries) it requires a measuring method with three-dimensional atomic resolution.

The commonly used tomographic reconstruction methods depend on a linear model for the projection of the data to the signal [15,16]. In the case of EHT, the linear model is given by the Phase Grating Approximation (PGA) [17], which states that the phase reconstructed from a hologram is the integral of the local electric phase shift along the beam's direction [18]. The PGA has been proven not only to be reliable at medium resolution outside of low-order-zone-axis [19], but also to lose its validity when approaching atomic resolution imaging conditions [20]. This stems from non-linear phase shifting effects in electron scattering, which impacts the higher scattering angles stronger than lower ones.

Due to the substantial interest in atomic-resolution tomography several methods have been proposed already for several TEM imaging modes, predominately High-Angle Annular Dark-Field (HAADF) Scanning TEM. Usually, the atomicity (i.e. most of the volume inside the specimen is vacuum) of the specimen is included in the reconstruction via Compressive Sensing [21,22], a column-wise atom-counting scheme [9] or extensive filtering of conventionally reconstructed data [23]. Atomic-resolution

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http://dx.doi.org/10.1016/j.ultramic.2014.11.026
0304-3991/

tomography using Bright-Field TEM has also been investigated with nonconventional mathematical techniques for an inversion of the dynamical scattering [24,25]. An ideal inversion would allow the reconstruction of the complete three-dimensional structure from a minimal number of images; in case of a simple enough specimen this could even be a single image as proposed in Single Image Tomography [26–28]. It should be noted that these approaches are still at the proof-of-concept stage due to experimental difficulties as well as methodical problems.

It is, therefore, necessary to develop new tomographic methods that are able to cope with these non-linear effects. This work sets out to fathom the consequences of assuming linearity in reconstructing the phase of atomic resolution holograms under idealised conditions. With simulated holograms, generous sampling, an idealised specimen, (almost) no noise and the absence of experimental errors any artefacts in the reconstructed potential are results of non-linear effects. A characterisation of these artefacts gives clues onto how reconstruction methods, that allow reconstruction with atomic resolution, could look like.

2. Electron scattering

On the atomic level, the electrons are scattered almost exclusively at the electrostatic potential of the individual atoms. This interaction is so strong that the treatment as a perturbation (as is done in the Born Series Approximation) becomes unsuitable [29]. Instead a precise model can be found by approximating the Klein–Gordon Equation, which is referred to as the high-energy paraxial approximation [17]. The wave transfer through the sample is then given thusly:

$$\Psi(z) = \hat{Z} e^{\hat{P}+\hat{V}} \Psi(0)$$

Electron small-angle scattering integral (1)

With the z-ordering operator $\hat{Z}$ (analogous to Dyson's time-ordering operator) and two terms, containing the following symbols: k the angular wave number of the electron, Δ_{xy} the Laplacian in x and y, E the electron's overall energy, e the unit charge and ϕ the electrostatic potential:

$$\hat{P} := -i\frac{1}{2k}\int dz'\, \Delta_{xy} \qquad \text{Fresnel propagator}$$

$$\hat{V} := i\frac{1}{c^2\hbar^2}\frac{E}{k}\int dz'\, e\phi(z') \qquad \text{Electronic interaction}$$

These terms represent the two fundamental phenomena influencing the electron wave: the wave propagation and the interaction with the specimen. The former is the Fresnel Propagator, a near field approximation of free wave propagation, while the latter is simply the phase shift induced by the electric potential.

3. Phase grating approximation

While the electric interaction in itself is linear in the electric potential, the propagation is not, so the information in an electron wave is non-linear in the potential. This is problematic since there is no general theory for non-linear tomographic problems but only for linear ones. Nearly all conventional methods (see [15]) depend on this linearity, so a linear approximation must be found in order to be able to utilise them. For Electron Holographic Tomography this is the Phase Grating Approximation (PGA), which states that the phase of the object exit wave is the integral of the local phase shift (due to the electric potential) along the electrons' principal direction of travel. In terms of Eq. (1) this equates to the neglection of the Fresnel propagation $\hat{P} = 0$. The effects of propagation are appreciable, since the modulation of the wave by the atomic potentials is strong, which causes a high curvature of the phase front compared to the wave number. The propagation effect is stronger for higher deflection angles and consequently increases with the spatial resolution of an image, but also appears at low resolution imaging conditions where multiple scattering leads to scattering from high angles into lower ones.

There is no single experimental parameter or criterion that determines the validity of the PGA for a particular measurement. There are some loose rules, based on experimental experience, though [19,20]. They suggest that the PGA holds at medium resolution while avoiding low-order-zone-axis orientations of the specimen. Imaging along low order zone axes of crystals causes interference effects between the partial waves from different scattering events, producing non-linear effects. While the former is unavoidable when imaging with atomic resolution, the latter can be minimised by choosing the projection directions accordingly [30].

4. Setup

4.1. Simulation

Since this is an explorative numerical study, the specimen in itself is of no particular interest, and can be chosen quite freely. We choose a Gold monocrystalline nanocrystal, since it is easy to model, quite a strong scatterer and has some relevance to practical experiments. It has the shape of a truncated octahedron with a diameter of ≈ 3.6 nm, thus it is 8 unit cells wide and contains ≈ 1300 atoms. This crystal was initially tilted about 13° out of the [1 0 0] axis in order to avoid low order crystal orientations during the tilt series.

The simulation itself used a multi-slice algorithm [31] (implemented [32] in MATLAB®). Since the algorithm includes discrete Fourier Transforms which implicate periodic repetition of the supercell, the supercell needs to be padded with vacuum. The resulting cross section of the simulated wave was 6 nm × 6 nm and sampled with 512 px × 512 px. Its interaction with the atoms was modelled for an acceleration voltage of 300 kV using the scattering factor parametrisation of Weickenmeier and Kohl [33]. Starting from the initial orientation a tilt series from −90° to 89° at 1° steps was simulated. The focus of each wave of the tilt series was set at 2 nm behind the middle of the specimen, which roughly corresponds to the object exit plane. A selection of simulated object exit waves of such a tilt series is shown in Fig. A.1 and A.2 Appendix A.

4.2. Reconstruction

The dataset, produced by the simulation, has a spatial resolution of ≈ 0.2 Å and an angular one of 1°, which is small compared to the characteristic sizes of the minimal absolute (≈ 3 Å) and angular[1] ($\approx 13°$) distance between two neighbouring atoms. This sampling is very generous, but the atom lattice on itself is not the only feature of interest. The signal-to-noise ratio (SNR) stems only from the numerical noise of the simulation, giving an SNR several orders of magnitude higher than that is experimentally possible. Since the reconstruction does not use periodic boundary conditions, the padding, used in the simulation, is not necessary any more and can be discarded, thus only the center 360 px × 360 px are used from here on.

[1] i.e. The angle equivalent of the minimal absolute distance between two atoms at the sphere enveloping the nanocrystal.