Toward 3D structural information from quantitative electron exit wave analysis

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Abstract. Simulations show that using a new direct imaging detector and accurate exit wave restoration algorithms allows nearly quantitative restoration of electron exit wave phase, which can be regarded as only qualitative for conventional indirect imaging cameras. This opens up a possibility of extracting accurate information on 3D atomic structure of the sample even from a single projection.

1. Introduction
Electron exit wave restoration in High Resolution Transmission Electron Microscopy (HRTEM) is a well established technique for quantitative investigation of structure at the atomic level. It requires a series of images recorded at different defocus levels, or a combined defocus series / tilt series, and is used widely to improve image interpretation and to increase resolution [1,2]. Recent developments include quantitative analysis of electron exit waves to obtain three-dimensional information about the sample at the atomic level [3]. Although images obtained in HRTEM are two-dimensional projections, the electron exit wave reconstructed from these images does contain encoded three-dimensional information about the sample due to the specimen thickness, relative position of the atomic layers and multiple scattering events as the electrons pass through the material.

The theoretical work presented here examines if it is possible to extract three-dimensional information about a crystalline Pt model, including the position of a single Co substituent atom, using realistic HRTEM image simulations and accurate electron exit wave restoration techniques. The influence of electron optics and detector parameters on the restoration is also studied and the optimal conditions for a successful experiment are discussed.

2. Experimental
Two cubic models of crystalline (001) Pt with 1.57 nm edge with a substituent Co atom at the top or bottom of the central column in the cell were prepared, as shown in Figure 1. The electron exit wave was simulated at 200 kV using a multislice method and weak phase-object approximation as implemented in E. Kirkland [4]. The simulated waves were used for subsequent simulations of a series of defocused images (as shown in Fig. 2). Typical microscope parameters for aberration corrected TEM were used, including a 2x10⁻⁵ electron beam energy spread, ΔE/E, and a 0.001 mm...
spherical aberration coefficient, $C_s$. The sampling interval was 0.0123 nm/pixel for 128 x 128 pixels array. Two different chromatic aberration coefficients were tested in the calculations, $C_c = 0.5$ and $C_c = 0.1$ mm. The chromatic aberration coefficient has an effect of reducing the information limit of the microscope as an incoherence envelope function.

Figure 1. Models of Pt cell with a substituent Co atom in high (a) and low (b) positions.

Accurate simulations of detector contributions to the simulated images were performed to account for Poisson noise, Detective Quantum Efficiency (DQE), Modulation Transfer Function (MTF) and any further noise introduced by the detector. These calculations included the contribution of each individual electron to the final image, where the input position into the detector corresponded to a probability function defined by the original intensity, which also provides the Poisson contribution to noise. The output pixel signals for each of these electrons were calculated using Monte Carlo simulations of their interaction with the sensor, following the standard approach with added relativistic corrections and generation of secondary electrons [5]. Two detector models for TEM were tested, an indirect scintillator-coupled charge-coupled device (CCD) camera [6] and a direct monolithic active pixel sensor (MAPS) [7]. CCD configuration is typical of conventional cameras. MAPS illustrate the much improved MTF and DQE of thinned direct detectors. Complete detector parameters, including detector dark noise, readout noise and dynamic range limitations, were set at typical values for such commercial cameras. To reflect typical experimental conditions, an average electron dose of 100 electrons/pixel and an integration time of 1 sec were used for both detector models. Examples of simulated defocused images, including the detector contribution, are presented in Figure 2.

The exit wave restorations were performed utilising two algorithms, a modified Fienup input-output algorithm [8] and an unconstrained trust region optimization algorithm [9]. Refinements in the wave restoration routine were carried out until the change in the sum of squared deviations between the input and the output images in two consecutive refinement steps was less than 0.1.

3. Results and discussion
The simulated electron exit wave phase shift shows clear differences between the two models, as illustrated by the line scan through the centre of exit waves in Figure 3a. There is also a considerable difference in the phase shift for the column containing the Co atom relative to the similar columns without the substituent atom (Figure 3a), as well as for the columns containing different number of Pt atoms.

As the restored exit wave phase can only be obtained up to the information limit of the microscope, the information limit plays an important role in determining the differences in phase shifts for the two tested models. It is found that a $C_c$ of 0.5 mm reduces these differences considerably (Figure 3c), and therefore only results with $C_c=0.1$ are used for further discussion.
Figure 2. Simulated images in a focal series of a PtCo model, with defocus (overfocus) ranging from 0 to 5 nm with step of 1 nm from left to right ($C_c = 0.1$ mm). Image intensity before detection (a); image intensity with indirect CCD (b) and direct MAPS (c) detector calculations. The substituent Co atom is located in the atomic column in the centre of the images.

Figure 3. Line scans of phase shifts in the simulated exit waves for models with low and high Co atom positions. Original exit wave (a); information transfer-limited exit wave with $C_c=0.1$ mm (b) and $C_c=0.5$ mm (c), showing the influence of the information limit.

Figure 4 presents a comparison between restorations using Fienup and trust region optimization algorithms, employing full derivatives of the exit wave and the performance of CCD and MAPS detectors. By comparison with the original exit wave in Figure 3b, it is found that the Fienup algorithm (Figures 4a,b), although fast, results in agreement only for the positions of peaks. The relative peak heights are not reproduced. This may be due to long periods of stagnation of this algorithm, which leads to early termination of the refinements. The trust region optimization algorithm with CCD detector shows a better performance, which however remains only qualitative (Figure 4c). The MAPS detector and the trust region optimization algorithm result in a considerably better agreement with the original exit wave phase shifts. For this case all columns of atoms show correct relative phase shifts and their absolute values are close to the original exit wave.

It is apparent, however, that all of the methods within the tested parameters failed to distinguish between different positions of Co substituent in the sample, because the difference in position results in a relatively small phase shift difference, beyond the detection limit. It is anticipated that such detection will become possible with parameters or techniques that allow higher information transfer.

4. Conclusions
It is found that direct detectors will allow quantitative restoration of electron exit waves and extraction of accurate 3D structural information. This shows that realistic modelling of the detector contribution...
to recorded scattering intensities is essential in assessing the applicability of novel approaches for electron exit wave analysis and for informing further development of such techniques.

Figure 4. Line scans of phase shifts in restored exit waves ($C_c=0.1\ mm$) for low and high Co atom position, including detector contribution for CCD camera (a,c) and MAPS detector (b,d) models. Restorations were performed using Fienup (a,b) and trust region optimization (c,d) algorithms.

For the sample model considered here, the use of a direct imaging camera together with an accurate phase restoration algorithm will allow close to quantitative determination of phase shifts in differently substituted columns. However, it was not possible to distinguish the position of a single Co substituent atom within a column from the restored phase shifts in a single projection condition. This task, nevertheless, seems amenable for the focal series/tilt series technique which allows higher information transfer for increased sensitivity in the direction of projection. It is envisaged that accuracy of this method will improve with further improvement in electron microscope lenses and imaging detectors.

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