Towards Z-contrast in Electron Ptychography

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Abstract

In a purely elastic model of electron diffraction, we show that the scattering in the dark field region is concentrated primarily in the higher order Laue zone (HOLZ) rings but gets redistributed across the dark field region if thermal vibrations are added into the model using the frozen phonon approximation. This high angle region in diffraction space carries information about the atomic number (Z) of the scattering species. Since the forward model in ptychography uses a purely elastic calculation for diffraction patterns ptychographic reconstructions have poor Z-sensitivity. Using a correction term, we fit the azimuthally averaged intensities in the dark field region with an exponentially decaying function, and show that the fit parameters have a correlation with Z. Using this correction term to minimize the difference between the modeled elastic pattern and experimental pattern might enable us to extract Z information.

1. Introduction

Electron ptychography is a computational imaging technique used in scanning transmission electron microscopy (STEM) to retrieve phase information from diffraction patterns. Ptychography enables imaging at high spatial resolution and precision and is sensitive to light atoms, typically a challenge for conventional STEM methods. However, the technique has a poor sensitivity to atomic number (Z) making it difficult to distinguish different atoms from one another. This problem motivated us to explore two questions: How can we get information about the atomic number (Z-contrast) in ptychographic images? And where does this information about the atomic number lie in the diffraction patterns?

Purely elastic models do not include the effect of thermal vibrations making them less accurate for comparisons to experimental diffraction patterns. However, due to ease of computation, a pure elastic model is currently used in the forward model for ptychography. In contrast, the frozen phonon model approximates the effect of thermal vibrations and is a closer match to experimentally acquired diffraction patterns. However, calculation with the frozen phonons model is computationally more expensive, limiting its use in ptychographic forward models. We investigate whether we can use a correction term to a purely elastic calculation to bridge the differences to a

frozen phonon calculation, and whether the fit parameters in this correction term carry some Z-dependence.

2. Methods

We performed multislice simulations of electron diffraction patterns of a $SrTiO_3$ crystal, with the probe focused on top of i) Sr column, ii) Ti-O column, and iii) O column and for thickness ranging from 0 – 200 unit cells. The simulations were done using the abTEM simulation package. We compared diffraction patterns simulated with a pure elastic model as well as with the frozen phonon model, the latter used to include the effects of thermal vibrations.

3. Results

To obtain information about the atomic number from diffraction pattern, we examined the diffraction patterns calculated with the probe focused on each element and added up all the intensities in the dark field region between the two radii labeled in Figure 1(a) that includes the First order Laue zone (FOLZ) ring. The comparison of intensities obtained with the probe focused on different atomic columns and for different sample thicknesses is shown in Figure 1(b) and shows a clear trend of the integrated intensities with atomic number (Z).

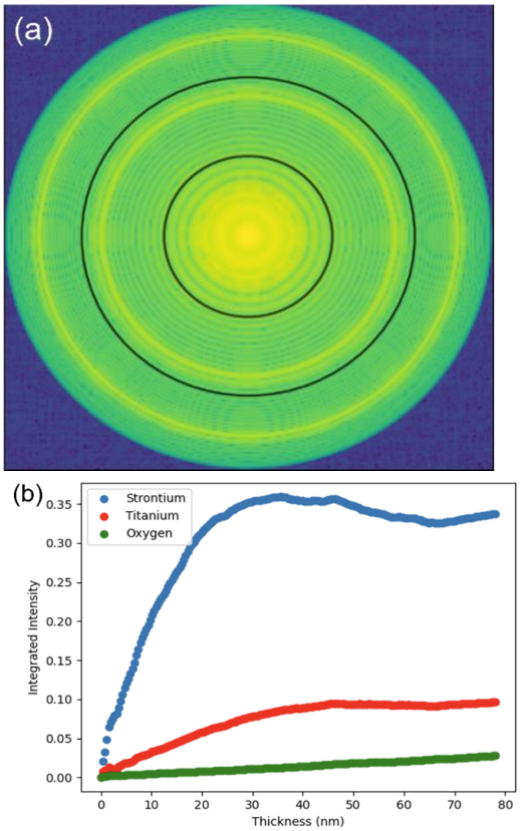


Figure 1: (a) Elastic diffraction pattern calculated with probe focused on Sr column in a SrTiO3 sample with a virtual annular detector marked with black lines that includes the First order Laue zone ring. (b) Intensities from virtual detectors with probe focused on different atomic columns plotted as a function of sample thickness.

Next, we excluded the FOLZ ring in the virtual detector area and compared the resulting integrated intensities obtained with a pure elastic model and frozen phonon model as shown in Figure 2. In a pure elastic model, the intensities drop almost to zero, indicating that most of the intensities in the dark field region is concentrated in the FOLZ ring. However, this is no longer the case for the frozen phonon calculation which shows considerable intensity in the selected dark field region, indicating an intensity

redistribution from the FOLZ ring into other dark field regions.

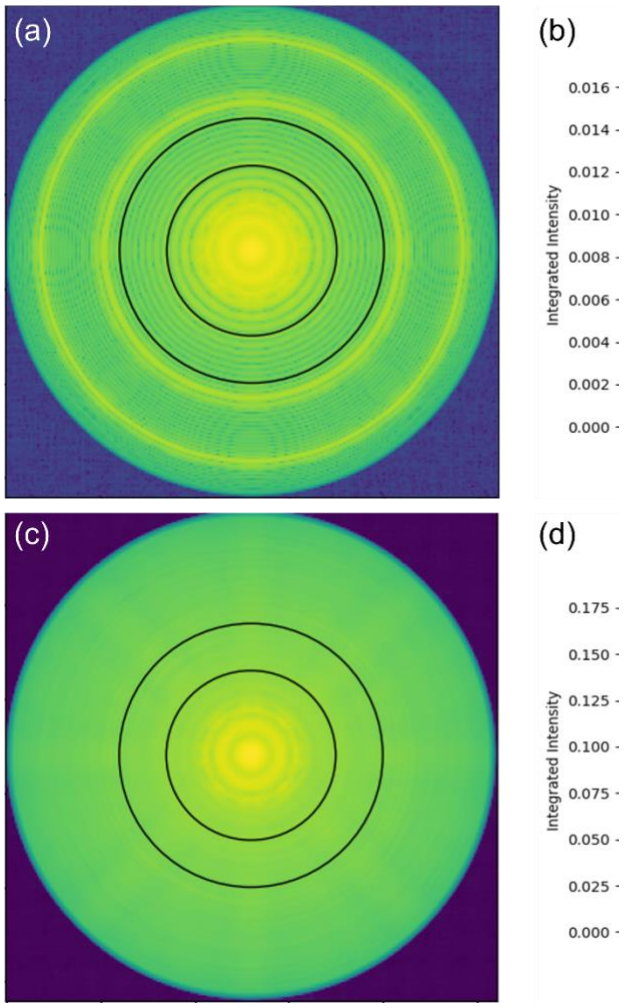
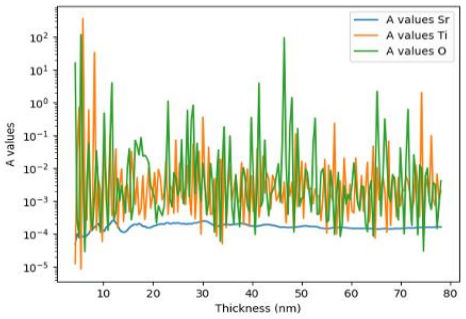


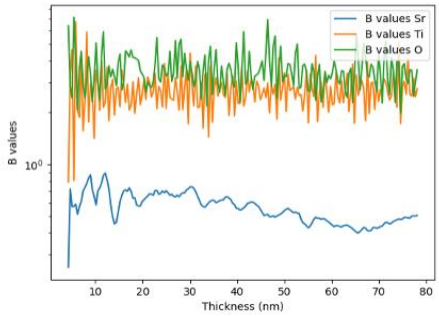
Figure 2: (a, c) – Diffraction patterns calculated with a pure elastic model and frozen phonon model respectively with the probe focused on the Sr column for a 75 unit cell thick sample. (b, d) Integrated intensities in the virtual detector marked with the black outline in (a, c) that excludes the First order Laue zone ring.

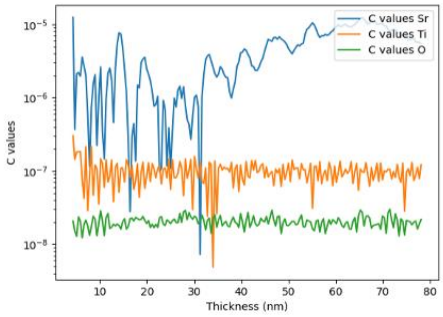
Hence, the elastic and frozen phonon calculations vary a lot in the dark field region indicated in Figure 2. We try to model the intensities in this dark field region in the frozen phonon calculation using an exponential correction term: $I(k) = A\, e^{-Bk} + c$ that currently considers variations only in the radial direction and averages azimuthal variations. The values of the fit parameters A, B, C obtained from the fits for different Z and thicknesses are shown in Figure 3, with the B and C parameters showing a trend with Z.

When we used the correction form along with the pure elastic model, we found that there was a correlation with the "B" and "C" value graphs where a higher atomic number yielded a lower "B" value, and a higher atomic number yielded a higher "C" value. This result also showed us that in future work when more diffraction patterns are used, we can use this same correction term to fit the diffraction patterns and extract relative Z information by comparing the B and C fit parameters.

Figure 3: Plots showing the fit parameters A, B, and C as a function of thickness for strontium, titanium, and oxygen columns.







In conclusion, we concluded that there is a direct relationship between the scattered dark field intensities and the atomic number, where a high atomic number yields higher intensity and vice versa with lower atomic numbers. We also found that most of the scattered intensity in a pure elastic calculation is concentrated in the FOLZ ring because we saw the key difference between the intensity percentages when the FOLZ ring was excluded. A simple exponential fit to model the dark field region in a frozen phonon calculation shows Z-dependence on some parameters, which could be useful in extracting Z information from ptychographic data.