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Retrieving depth-direction information from TEM diffraction data under reciprocal-space sampling variation

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ABSTRACT

For full three-dimensional information retrieval from transmission electron microscope data, retrieving the third-dimension (beam-direction) information is an important challenge. Recently, we have developed an artificial-neural-network-based retrieval algorithm suitable for retrieving three-dimensional nanoscale crystal parameters like strain, including with noisy data (R.S. Pennington, W. Van den Broek, C.T. Koch, Phys. Rev. B 89 (20) (2014) 205409 [12]). In this work, we examine how reciprocal-space sampling conditions influence the retrieved crystal parameters, using crystal tilt as an example parameter, and demonstrate retrieval for 2.5 nm depth resolution. For noise-free data, we find that the total reciprocal-space area is the key parameter; however, when the data are noisy, the number of reciprocal-space points and the amount of noise are also influential. We also apply our algorithm to a simulated bent specimen, and recover the bending as expected. Guidelines for experimental applications are also discussed.

1. Introduction

In the transmission electron microscope (TEM), the electron beam passes through a three-dimensional specimen, forming a two-dimensional image or diffraction pattern on the other side. In this process, multiple scattering encodes some of the depth-direction, or "third-dimensional" information about the specimen in the beam [1,2]. Decoding this "third-dimensional" information carried by the electron beam is difficult because the effect of multiple scattering is highly nonlinear with specimen thickness [3]. Determining the TEM output data corresponding to a known specimen is a well-known procedure, with a number of algorithms that are suitable for different tasks [2–5]. However, for an unknown specimen, when multiple scattering is considered, the inverse problem of determining the specimen from the output data is far more difficult. For single-scattering-based measurements, where the depth information may be captured by using variations of specific experimental conditions that are considered to generate single-scattering data, the reconstruction algorithms are generally linear: electron tomography, either algebraic or geometric [6,7], is commonly limited to specimen density or shape information; focal-series annular dark field images [8] have been used to retrieve surface-layer strain, but only for the top 15 nm of the specimen; in high-angle annular dark-field scanning TEM or scanning confocal electron microscopy, depth-direction information can be recorded using focal variation [9–11]. Recently, we have developed an algorithm that enables efficient evaluation of this multiple-scattering inverse problem for nanoscale stacked-Bloch-wave simulations from a single convergent-beam electron diffraction (CBED) pattern, even if using only a few CBED disks and including noise [12]. This algorithm can be applied, in principle, to retrieving the "third-dimension" (depth-direction) variation of any specimen parameter affecting the Bloch-wave scattering matrix. Fig. 1 shows a CBED pattern of a centro-symmetric structure (silicon) with third-dimensional variation in crystal tilt, leading to left–right asymmetry in diffraction features. To solve this many-parameter optimization problem, our algorithm uses artificial neural network (ANN) optimization tools that are similar to those used to retrieve individual atomic positions in nanoparticles [13,14]; however, we apply these tools to a different optimization problem in a different context.

Our previous work [12] demonstrated that our algorithm *can* work; in contrast, this paper discusses *how well* our algorithm works and how to provide optimal input data. First, how does reciprocal space sampling affect our algorithm's effectiveness at retrieving specimen parameters? Second, how effective is our algorithm expected to be on noisy data, under different noise conditions? Third, what happens when the specimen has near-continuous orientation changes but is reconstructed using only a few slices? For the first two scenarios, we use the same target specimen as in our previous work, but we vary the experimental conditions in multiple different ways. For the third scenario, we use a simulated bent specimen generated using 0.5 Å

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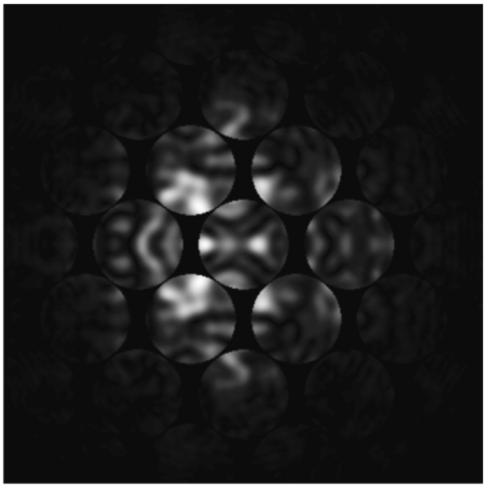

Fig. 1. Convergent-beam electron diffraction pattern, simulated using the asymmetric specimen crystal tilts given in Table 1 for a 100 nm specimen. The center of the features in each disk is shifted left due to the non-zero mean crystal tilt. Even including this effect, however, there is a left-right asymmetry in the diffraction features created by the third-dimension crystal tilt variation in the [001] direction. This pattern is simulated using the standard conditions listed in the text for Si[110] at 80 keV primary energy, a 0.01° dot pitch, a 6.5×10^{-3} radian aperture, and a 4°×4° field of view; except for the field of view, these are the same conditions used previously [12]. This larger field of view highlights the higher-order reflections, like the ⟨220⟩, which can appear both very weak and with obvious asymmetries. The display min/max is set to [0.0, 0.4] counts per pixel, and normalized so that, if no specimen were inserted, 1.0 counts would be incident on each pixel in the [000] beam.

slices, each with a slightly different orientation, and we apply our retrieval algorithm using 10 nm/slice.

In this work, we explore how best to sample reciprocal space for optimal information retrieval. Our algorithm uses samples spread across reciprocal space, but our previous work did not fully explore optimal sampling conditions in reciprocal space. Different experimental techniques, such as CBED, large-angle rocking-beam electron diffraction (LARBED) [15], and dark-field-image tilt series (DFITS), have different reciprocal-space sampling conditions. DFITS is a series of bright- or dark-field images gathered at different tilt angles and from different diffraction reflections. Knowledge of optimal conditions – e.g. CBED disk size, LARBED precession angle, and the angle between successive DFITS images – can allow acquisition of the best possible experimental data. This represents a necessary and significant step forward for the practical application of this algorithm to experimental data.

In this work, we discuss the optimal sampling in reciprocal space for optimal information retrieval, including for different specimen thicknesses. We use the stacked-Bloch-wave (SBW) forward-simulation algorithm [16–18], implemented as described previously with an artificial-neural-network optimizer [19,12,20] as the basis for our retrieval algorithm. Our target parameter is the local crystal tilt in one direction for a 10 layer specimen. We consider first the optimal reciprocal-space sampling for noise-free data. Then, we demonstrate and discuss how our algorithm performs for data with noise. Finally, we consider how our algorithm would reconstruct a continuously bending specimen.

2. Results and analysis

In this section, we present the results of our third-dimension optimization algorithm under different conditions. In Section 2.1, we consider different reciprocal-space samplings without noise. These are all on the same specimen crystal-tilt geometry, but considering different thicknesses. In Section 2.2, we consider data with noise. Finally, in Section 2.3, we consider how our algorithm performs when the true object is realistically bent and has near-continuous parameter variation, but the retrieved parameters are for thick slices.

All of these simulations are run under the same conditions, unless otherwise stated. The TEM accelerating voltage is 80 kV, and the specimen is pure Si, with values from the literature for lattice parameter [21], Debye–Waller factor [22], and electron scattering factors [23]. Only zero-order-Laue-zone reflections with a distance to the Ewald sphere $s_z < 1.225\ \text{nm}^{-1}$ are simulated, thus including $K=197$ reflections [2]. In Sections 2.1 and 2.3, all 197 reflections are considered to be measured; in Section 2.2, only the five lowest-order reflections (the direct beam and the four ⟨111⟩ reflections) are considered to be measured. The absorption model of Bird and King is used [24], and the CBED patterns are considered to be zero-loss-filtered. All surfaces are assumed to be normal to the electron beam. We also assume that the CBED pattern is large enough on the detector that the intensity is slowly varying from one pixel to the next, so the effect of the camera modulation transfer function (MTF) is minimized and thus not considered here [25]. The specimen is in a [110] zone-axis orientation, and the crystal tilt basis vectors $\hat{\alpha}_x$ and $\hat{\alpha}_y$ are in the [001] and $[\bar{1}10]$ directions, respectively. The regions sampled are all squares with one corner in the middle of the pattern and extending along the [001] and $[\bar{1}10]$ directions.

We consider first the same "prototype" specimen used in previous work [12]. This specimen has third-dimensional variations in its local crystal tilt in the α_x direction, given in Table 1 as a function of percent-depth in the specimen. For specimens with different thicknesses, the same geometry is used, so doubling the total specimen thickness doubles the slice thickness, but keeps the same proportions and the same α_x values for each slice. All scenarios for this specimen are initialized with a uniform specimen with all α_x starting values as $\overline{\alpha}_x$, the average value of the true crystal tilt, given in Table 1. We consider $\overline{\Delta\alpha_x}$, the mean parameter mismatch over all ten layers, and $\overline{\Delta E}$, the mean intensity mismatch between the diffraction data. $\overline{\alpha}_x$ is close to the crystal tilt that one would be able to fit by assuming a crystal consisting of a single slice [12].

2.1. Noise-free reciprocal-space sampling

In this section, we consider how best to sample reciprocal space for optimal third-dimension information retrieval. This is necessary to determine a priori microscope parameters for data acquisition, such as how large the CBED disk should be on the camera and the

Table 1
Specimen crystal tilt (α_x) scenario used both in this work (Sections 2.1 and 2.2) and previous work [12]. d is in percent of total specimen thickness because four different specimen thicknesses are considered in Section 2.1: [25, 50, 100, 200] nm. Section 2.3 uses a different specimen, detailed in that section.

d (%)	α_x (deg)
0–10	0.00
10–20	0.00
20–30	−0.06
30–40	−0.15
40–50	−0.15
50–60	−0.20
60–70	−0.20
70–80	−0.15
80–90	−0.06
90–100	0.00
$\overline{\alpha}_x$	−0.097