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Theory of dynamical electron channeling contrast images of near-surface crystal defects

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ABSTRACT

This paper describes the dynamical simulation of electron channeling contrast images (ECCIs) of dislocations. The approach utilizes both the Bloch wave formalism and the scattering matrix formalism to generate electron channeling patterns (ECPs). The latter formalism is then adapted to include the effect of lattice defects on the back-scattered electron yield, resulting in a computational algorithm for the simulation of ECCIs. Dislocations of known line direction and Burgers vector are imaged experimentally by ECCI and match well with simulated ECCIs for various channeling conditions. Experiment/simulation comparisons for ECPs and ECCIs are demonstrated for metals (Al), semiconductors (Si), and ceramics ($SrTiO_3$).

1. Introduction

Electron channeling, first observed as a manifestation of "Kikuchi-like" bands superimposed on a single-crystal surface imaged in a scanning electron microscope (SEM) by Coates in 1967 [1], has since been available as an SEM based imaging option for structural analysis and extended defect identification in crystals. *Electron channeling patterns* (ECPs), typically observed in the low magnification back-scattered electron (BSE) imaging mode, are caused by the angular variation in the BSE yield as a function of the orientation of the incoming coherent SEM electron beam with respect to the target's crystal lattice orientation; the width and symmetry of the channeling bands in an ECP are a direct consequence of the near-surface space group and averaged lattice parameters of the crystal itself. Thus, acquisition and indexing of ECPs provide a means for SEM-based crystal structure analysis [2].

In addition to crystal structure identification, electron channeling allows for direct imaging of extended defects (stacking faults, dislocations) in crystals. This approach, known as *electron channeling contrast imaging* (ECCI), relies on the localized lattice plane bending in the vicinity of individual extended defects to create BSE image contrast. High magnification imaging of a single-crystal surface should yield a constant BSE yield as the beam is scanned across a small area ($< 10\,\mu m$) since the incoming beam trajectories are essentially parallel to one another. Elastic strain around a defect will change the local orientation of the crystal lattice, producing a fluctuation in the BSE yield. BSE yield varies most strongly when the incoming beam trajectory is near the Bragg condition for a set of diffracting planes. These BSE intensity fluctuations manifest themselves as contrast features denoting the position of crystal defects in the resultant ECCI micrograph. More importantly, the intensity profile of an individual ECCI contrast feature can be related to the defect type [3,4].

Any experimental ECP and ECCI analysis approach requires reliable correlation to computed patterns or images. Quantitative assessment of BSE yield variations that give rise to electron channeling requires utilization of dynamical electron diffraction theory [3]. A number of studies have developed theoretical models based on dynamical effects using the superposition of multiple Bloch waves [5–9]. Initial studies by Hirsch and Humphreys [10] indicated that modeling the angular variations in the electron back-scattering probability requires consideration of multiple inelastic scattering events. Clarke and Howie [5] extended this approach to consider contrast variations surrounding crystal defects (screw dislocations and stacking faults). Spencer et al. [6] proposed the forward–backward approximation (FBA) to conveniently consider multiple inelastic scattering in terms of two groups of opposite trajectory electrons within the crystal. Spencer and Humphreys [11] later developed a general transport equation to account for multiple scattering. Dudarev et al. [12] extended this transport equation approach to a more efficient inhomogeneous transport equation based on the kinetic equation for the one-particle density matrix. Monte Carlo-based simulations have also been developed to replicate electron channeling phenomena [13]. More recent simulations have utilized the principle of reciprocity to relate incoming and outgoing electron beam intensities [4,9,14,15]. In these studies, quasi-elastic scattering is

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assumed to be sufficient to account for the energy losses by BSEs. As such, relatively good quantitative agreement to ECPs [14] and good qualitative agreement for individual defect images [4,15] have been demonstrated.

Nevertheless, a more comprehensive simulation approach with direct experimental validation is required to properly account for the dynamical diffraction effects that govern both ECP and ECCI generations. In transmission electron microscopy (TEM), it has been the standard approach for many decades to use a full dynamical electron scattering simulation of defects in materials with anisotropic elastic properties [16]. This simulation approach is usually based on the fact that one can orient a crystal in such a way that one set of lattice planes dominates in the scattering process; this is known as the systematic row case, which can be further simplified to the two-beam case for materials with small unit cells. One of two simulation paths is commonly used: Bloch waves [17] or direct solution of the Darwin–Howie–Whelan (DHW) differential [18,19]. The latter can be accomplished by means of standard Runge–Kutta-style algorithms, or, as described later in this paper, via the introduction of the scattering matrix [20]. Since the machinery of defect image simulations for TEM images is well developed, it appears logical to make use of this approach for the simulation of ECCIs as well.

In the case of back-scattered electrons in the SEM, one distinguishes between BSE1 and BSE2 scattering events (e.g., [21]). In a BSE1-type event, an incident beam electron is back-scattered immediately after entering the sample, i.e., as the first scattering event, whereas for a BSE2 event, the electron does not leave the sample until several (i.e., more than one) inelastic scattering events have occurred, at which point the electron may have traveled some lateral distance away from the entrance point. The BSE1 electron emerges close to the entrance point, and has an energy very close to the incident beam energy; for the BSE2 electron, the exit energy depends on the number and type of inelastic events that occur between entrance and exit events. In general, BSE2 electrons display a wide range of energies. An estimate of the ratio of BSE1 to BSE2 electrons can be obtained by means of a simple Monte Carlo simulation. We take $SrTiO_3$ as an example, since several of the results reported in this paper deal with this compound. For an incident beam energy of 20 keV, 1.5×10^9 electron trajectories were simulated using the continuous slowing down approximation (CSDA) [22]; a total of 2.41×10^8 (16.1%) electrons were back-scattered. Of these electrons, 419,921 were BSE1-type, the others were BSE2 electrons; in other words, the BSE1 electrons make up about 0.17% of the total number of BSEs. The majority of BSE1 electrons leave the sample at a relatively large angle with respect to the (normal) incident beam direction, with a maximum between 60° and 70°. For a realistic BSE detector size (annular, with inner and outer radii of 2.75 mm and 8.25 mm, respectively), and a working distance that maximizes the total detected BSE signal, the total integrated BSE signal has about 1 BSE1 electron for every 1000 BSE2s.

It is important to realize that only BSE1 electrons carry diffraction contrast information. As the incident electron enters the sample, it will channel through the crystal lattice, so that the probability of undergoing a back-scatter event will be modulated by orientation-dependent dynamical interactions. If the electron is back-scattered out of the sample, i.e., a BSE1 event, then its contribution to the overall back-scatter signal will be modulated by this orientation-dependence. For BSE2 events, which also channel through the lattice in between repeated inelastic scattering events, the effects of channeling average out and the BSE2 contribution to the total BSE signal (which accounts for 99.9% of the signal in the case of $SrTiO_3$) does not contain any diffraction information, i.e., the BSE2s provide a large background signal. To detect the weak BSE1 signal on top of this large background, one typically must adjust the brightness and contrast controls of the SEM close to the edge of the available range.

In this paper, we will focus exclusively on BSE1-type electrons, since they are the only electrons that carry diffraction information. In view of the comments in the preceding paragraph, in the absence of absolute intensity measurements, we can always scale the contrast and brightness of the simulated images to match the experimental observations as closely as possible; agreement between simulated and experimental images will hence only be of a qualitative nature. As is common in TEM scattering simulations, inelastic scattering events along the incident electron trajectory will be incorporated in terms of a phenomenological imaginary absorptive potential added to the standard electrostatic lattice potential [18]. The goal of the present study is to extend these well-established dynamical diffraction formalisms (Bloch waves, scattering matrix) from TEM in order to realize a realistic method for simulating near-surface crystal defect images obtainable in a modern SEM via electron channeling. The structure of this paper is then as follows: in Section 2 we begin with a description of the theory of ECPs (Section 2.1) which we then augment in Section 2.3 to derive a model for ECCI defect image contrast. In Section 3 we provide example simulations for ECPs (Section 3.1) and ECCI defect images (Section 3.2) and compare them to experimental observations. We conclude the paper with a brief discussion and comments in Section 4.

2. Theoretical development

An ECCI is formed by BSEs that experienced the back-scatter event as their first scattering event upon entering the sample. Along the trajectory from the entrance point to the back-scatter event, the electron experiences the lattice potential and channels along the atomic columns. This channeling modifies the probability that the electron will be found at a given depth inside the crystal, which, in turn, modifies the BSE yield as a function of depth. The total BSE1 signal is then obtained by integrating the channeling-modified yield over an appropriate depth range. While one can obtain an estimate of the relevant depth range by means of Monte Carlo simulations (see [23] for an example of how to combine Monte Carlo and dynamical simulations), in the present study we will assume that the correct integration depth, z_0, is known.

As described in the next section, the computation of an electron channeling pattern is typically performed using the Bloch wave approach. For defect image simulations, however, it turns out that it is useful to express the ECP model in terms of a superposition of plane waves traveling in the directions predicted by the Bragg equation, and incorporate the effect of lattice defects in the resulting dynamical and scattering matrices. This new derivation is presented in Sections 2.2 and 2.3.

2.1. Bloch wave electron channeling pattern model

We begin with a crystal structure with N_a atoms per unit cell, distributed over n positions in the asymmetric unit; for each such position, there is a set $\mathcal{S}_n$ of equivalent atom positions. In the single scattering approximation, neglecting multiple incoherent scattering, the probability that an incident electron with wave vector $\mathbf{k}_0$ will be back-scattered at any depth z in the range $[0, z_0]$ below the surface is proportional to the integral:

$$\mathcal{P}(\mathbf{k}_0) = \sum_n \sum_{i\in\mathcal{S}_n} \frac{Z_n^2 D_n}{z_0} \int_0^{z_0} \mathrm{d}z\, |\Psi_{\mathbf{k}_0}(\mathbf{r}_i)|^2, \tag{1}$$

where Z_n is the atomic number for atom type n, D_n is the Debye–Waller factor for atom type n, and $\mathbf{r}_i$ is the fractional atom position