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Energy-weighted dynamical scattering simulations of electron diffraction modalities in the scanning electron microscope

Elena Pascal[a], Saransh Singh[b], Patrick G. Callahan[c], Ben Hourahine[a], Carol Trager-Cowan[a], Marc De Graef[b,*]

[a] Department of Physics, SUPA, University of Strathclyde, G4 0NG, Glasgow, UK
[b] Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, PA 15213-3890, USA
[c] Department of Materials, University of California at Santa Barbara, Santa Barbara, CA 93106, USA

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ABSTRACT

Transmission Kikuchi diffraction (TKD) has been gaining momentum as a high resolution alternative to electron back-scattered diffraction (EBSD), adding to the existing electron diffraction modalities in the scanning electron microscope (SEM). The image simulation of any of these measurement techniques requires an energy dependent diffraction model for which, in turn, knowledge of electron energies and diffraction distances distributions is required. We identify the sample-detector geometry and the effect of inelastic events on the diffracting electron beam as the important factors to be considered when predicting these distributions. However, tractable models taking into account inelastic scattering explicitly are lacking. In this study, we expand the Monte Carlo (MC) energy-weighting dynamical simulations models used for EBSD [1] and ECP [2] to the TKD case. We show that the foil thickness in TKD can be used as a means of energy filtering and compare band sharpness in the different modalities. The current model is shown to correctly predict TKD patterns and, through the dictionary indexing approach, to produce higher quality indexed TKD maps than conventional Hough transform approach, especially close to grain boundaries.

1. Introduction

Electron diffraction techniques in the scanning electron microscope (SEM) are established and versatile tools for microstructural investigation of crystalline materials. The strong and complex local interactions of electrons with crystalline matter offer a plethora of information about the crystal structure and material properties of a sample that can be recovered from the recorded signal. A review of these is given in ref. [3]. Kikuchi patterns are one representation of the diffracting behaviour of electrons in the form of a variation in the angular distribution of signal electrons. The geometry of these patterns is dictated by the unit cell of the crystal and its orientation. Other features, such as the width of the bands, for instance, are nevertheless influenced by the spatial distribution of electrons in the sample and their energy distribution (for a more complete discussion see ref. [4]).

We can distinguish a number of different SEM modalities employing the Kikuchi diffraction mechanism. If the recorded electrons are the backscattered ones (BSEs), then the technique is known as electron backscatter diffraction (EBSD) and the Kikuchi patterns obtained are called electron backscatter patterns (EBSP). Automated pattern indexing software established this diffraction modality as one of the conventional tools of orientation mapping, phase identification and/or relative lattice strain estimation in crystalline materials [3]. In order to increase the diffraction signal in this mode, the popular approach has been to tilt the sample to about 70° from horizontal towards the detector, which guarantees a maximum backscattered electron yield. However, the high tilt will also spread out the information volume (or interaction volume) of the electrons within the sample, resulting in limitation of the achievable spatial resolution.

Stimulated by the increased attention to nanostructured materials, which promise new and enhanced properties when compared to their larger scale counterparts, the interest in improving the resolution of established characterization techniques has also

* Corresponding author.
E-mail addresses: elena.pascal@strath.ac.uk (E. Pascal), ssaransh@andrew.cmu.edu (S. Singh), pcallahan@ucsb.edu (P.G. Callahan), benjamin.hourahine@strath.ac.uk (B. Hourahine), c.trager-cowan@strath.ac.uk (C. Trager-Cowan), degraef@cmu.edu (M.D. Graef).

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expanded. The use of forward-scattered electrons (FSEs) through a thin sample as diffraction signal collected from the bottom of the foil has been shown to improve the lateral spatial resolution to below 10 nm [5,6]; this technique is commonly known as transmission Kikuchi diffraction (TKD) or transmission EBSD (t-EBSD).

The modalities above are sometimes referred to as "channeling out" diffraction techniques [7] to suggest that the diffraction information has been sampled by electrons on their way out of the sample and that the volume from which the signal is collected is located close to the exit surface. The SEM can also be used in "channeling in" mode when electron channeling patterns (ECPs) are acquired [8,9]. In this case, Kikuchi-like diffraction patterns can also be obtained by varying the incident beam direction with respect to the crystal. Typically, those patterns have a smaller solid angle compared to their EBSD counterparts. Nevertheless, the physical scattering mechanisms that produce EBSPs and ECPs are related through the reciprocity principle [10].

Theoretical models have been developed and successfully applied to retrieve this wealth of information by taking into account the full dynamical behaviour of electron diffraction [11–13]. Electron diffraction calculations commonly handle inelastic scattering in a phenomenological way through the introduction of a complex optical crystal potential approximation. This assumption implies that inelastically scattered electrons, once they lose even a small amount of energy, will cease to contribute to the diffraction pattern. The predicted diffraction patterns based on this simplified model remain meaningful [14] but, understandably, are lacking quantitative precision. Due to the strong interaction of incident beam electrons at SEM energies with matter, the inelastic cross section is always comparable to the elastic one, and a portion of inelastically scattered electrons will reach the detector and contributes to the imaged pattern.

Depending on the types of inelastic channels allowed, these electrons can suffer diffraction after losing a small amount of energy, contributing then to the diffuseness of the Kikuchi patterns. This process is especially relevant for "channelling out" modalities where electrons with energies lower than the incident energy can still contribute to the diffraction pattern. Alternatively, if electrons are scattered at a large angle multiple times such that memory of their original direction is lost, they will also contribute to the background intensity. This is the case for both channeling modalities. We call the later type of inelastically (back/forward-)scattered electrons (B/F)SE2 in order to differentiate them from (B/F)SE1 electrons carrying diffraction information.

It is therefore essential to explicitly consider inelastic scattering and its effects on the signal contributing electrons, such as their energy and spatial distributions [1,15]. This is especially important if finer features of the Kikuchi bands (size, absolute intensity relative to background, band edges) are to be correctly predicted. A full account of the inelastic channels in electron diffraction poses a challenging problem. While general Schrödinger equation solutions for inelastic scattering in perfect crystals have been proposed by Yoshioka [16] and solved for various electron microscopy applications (see Howie [14] for small angle plasmon scattering and Forbes et al. [17] for single thermal diffuse scattering events), to our knowledge, readily implementable solutions relevant for SEM electron energies have yet to be proposed.

In this work, we assume inelastic scattering events to be stochastic and that Monte Carlo (MC) techniques can estimate both the trajectories of electrons that suffered such events as well as their energy distribution. Such models have been proposed and widely used to correctly predict distributions of backscattered electrons [18]. The assumption that the distributions of escape energies and trajectories of electrons carrying diffraction information can be estimated from the last elastic event predicted by MC models has already been successfully applied for EBSPs [1] and ECPs [2]. The electron energy at the last elastic event, prior to leaving the sample, is regarded as the diffraction energy (energy at which the diffraction event occurs), and the distance to the exit surface from the elastic event (escape or exit distance) is used as the diffraction distance (electron path length over which coherence is not lost). Dynamical diffraction modelling is then applied for the full MC predicted electron energy and path distributions. Here, we extend this model to TKD patterns by considering the geometry of a thin film sample where the entry (top) and escape (bottom) surfaces are different such that the incoherent events acting as sources of diffracting electrons are scattering in a forward direction.

While this approach may not take into account the full extent of inelastic scattering effects on diffracted electrons proposed by the Yoshioka equations, it leads to a model of manageable complexity which is straightforward to implement and whose predictions are easily understood. Most importantly, it represents a step forward in taking into account the full physics of electron diffraction in matter by considering the full distribution of energies of channeling electrons and produces accurate predictions when compared to experimental patterns, as shown in Section 3.2.

In Section 2 we describe the typical geometries for EBSD, TKD and ECP data acquisition and formulate a general expression for the thickness integrated back-scattered electron intensity that is applicable to all three diffraction modalities. We describe the particulars of the Monte Carlo trajectory simulations in Section 2.2, along with the resulting differences between the modalities. Master patterns for the three modalities are described and compared in Section 3.1. In Section 3.2 we compare experimental and simulated TKD patterns, and Section 3.3 illustrates how the recently developed dictionary indexing technique [19] can be applied to TKD patterns. We conclude the paper with a brief discussion and summary in Section 4.

2. Theoretical model

2.1. Energy and diffraction distance integrated electron intensity

The simulation of the (back/forward-)scattered electron distribution emerging from a sample illuminated with a fine, nearly-parallel, electron probe can be achieved in general by integrating over both the energy range of the exiting electrons and the distance traveled in the sample between the scattering site and the sample surface. The probability of a (B/F)SE emerging from the sample in the direction $\hat{\mathbf{k}}$ (the hat indicates a unit vector) can be written as follows:

$$P(\hat{\mathbf{k}}) = \sum_{n \in \text{A.U.}} P_n(\hat{\mathbf{k}}), \tag{1}$$

where A.U. stands for asymmetric (primitive) unit and the index n runs over all positions in the asymmetric unit. The probability P_n is defined as:

$$P_n(\hat{\mathbf{k}}) = \sum_{j \in \mathcal{S}_n} \sigma_j \int_{E_{\min}}^{E_{\max}} \mathrm{d}E \int_0^{t_0(E)} \mathrm{d}t\, \bar{\lambda}_{\hat{\mathbf{k}}}(E,t) |\Psi_{\hat{\mathbf{k}}}(\mathbf{r}_j; E, t)|^2. \tag{2}$$

Here, $\sigma_j = Z_j^2 D_j$ (with Z the atomic number and D the Debye–Waller factor) is the Rutherford scattering cross section for atom j in the set of equivalent positions $\mathcal{S}_n$; $E_{\max}$ is the maximum energy (potentially the incident beam energy E_0) and $E_{\min}$ the lowest energy considered in the calculation; t is the distance between the scattering site and the sample surface, measured along the exit direction; $t_0(E)$ is the maximum distance to be considered; $\bar{\lambda}_{\hat{\mathbf{k}}}(E,t)$ is a weighting function describing the fraction of incident electrons (per unit energy and per unit length) of energy E, originating a distance t from the sample surface and traveling in the direction $\hat{\mathbf{k}}$; the wave function $\Psi_{\hat{\mathbf{k}}}$ is evaluated for the equivalent atom positions $\mathbf{r}_j$ and the parameters E and t. For the latter, one can use ei-