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Two-dimensional misorientation mapping by rocking dark-field transmission electron microscopy

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ABSTRACT

In this paper we introduce an approach for precise orientation mapping of crystalline specimens by means of transmission electron microscopy. We show that local orientation values can be reconstructed from experimental dark-field image data acquired at different specimen tilts and multiple Bragg reflections. By using the suggested method it is also possible to determine the orientation of the tilt axis with respect to the image or diffraction pattern. The method has been implemented to automatically acquire the necessary data and then map crystal orientation for a given region of interest. We have applied this technique to a specimen prepared from a Ni-based super-alloy CMSX-4. The functionality and limitations of our method are discussed and compared to those of other techniques available.

1. Introduction

Transmission electron microscopy (TEM) is a powerful and versatile tool for investigating the atomic structure and morphology of nano- and micro-objects. A very large fraction of the specimens investigated by electron microscopists working in the field of materials science are crystalline. Crystal defects break the translational symmetry of the crystal and lead to elastic strain and changes of the orientation of the unit cell of the crystal. Since specimens prepared for TEM should be reasonably thin in order to be transparent for the electron beam, they may also bend due to relaxation of strain present in the material, and in some cases also due to the specimen preparation procedure itself. Great efforts have been made to develop orientation mapping techniques of crystalline materials in TEM. Using orientation maps one can visualize variations in the local lattice orientation.

Routine crystal orientation mapping in TEM was first realized in the TEM by Dingley and Wright [1–3] and has initially been commercialized by TexSEM Laboratories (TSL) as *Automated Crystallography for TEM* (ACT). This method is referred to as "conical dark field scanning" because it uses a set of dark field (DF) images acquired at many different beam tilts and builds the absolute lattice orientation maps by constructing diffraction patterns (DP) from those DF images and indexing them. From then on many other TEM-based orientation mapping techniques have been developed. The "conical dark field scanning" technique was improved by employing a fast image matching algorithm [4]. In addition, a three-dimensional (3D) grain orientation mapping technique based on the same approach has been developed [5]. For that technique the crystal orientation within individual voxels (volume elements) are reconstructed from more than 10^5 DF images. Images are acquired for many different directions of the incident illumination and specimen tilts and the reconstruction is then carried out using algorithms originally developed for synchrotron diffraction data. There are also different mapping techniques based on the analysis of diffraction spot patterns or Kikuchi bands [6,7]. The individual diffraction patterns can be recorded with parallel (NBD [8]) or convergent (CBED [9,10]) illumination, or by combining these setups with precession of the illumination tilt angle [8]. The diffraction patterns are usually acquired by stepwise scanning of the specimen [11,12] and the orientation is then determined by matching each pattern to a large library of pre-generated template patterns [8,13] in combination with a grid search in orientation space [14] which is widely used in various areas of crystallography. It has also been shown that combining spot patterns with the analysis of Kikuchi patterns delivers more precise orientation maps [15,16].

In this paper, we report a technique which delivers precise orientation maps within a single grain of a crystal with high spatial resolution and a large field of view. In contrast to conventional orientation mapping in crystalline materials, our approach only maps small local deviations from a given zone axis, which we refer to as crystal lattice misorientation (CLM). However

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only the variation in CLM smaller than the tilt range can be reconstructed by this technique. In addition to lateral strain and in-plane rotation measurable by various techniques (varying the defocus at one of the tilt angles in the currently proposed method allows the 4 lateral components of the strain tensor to be mapped [17]), CLM completes two additional components of the 9-element strain tensor and is thus an important step towards mapping of the complete three-dimensional strain tensor by TEM. Moreover, we expect that this technique will be useful for quantitatively differentiating between plastic and elastic strain within the specimen. With the approach presented below, local orientation changes can be mapped with a precision of $<0.1^{\circ}$. The accuracy of the mapping should be better than 1 mrad. The spatial resolution of this approach is limited by the size of the unit cell, since this determines the size of objective aperture to be used for the experiment.

In the current work, we analyse one-dimensional rocking curves extracted from tilt series of DF images of multiple reflections. DF images are recorded at slightly different specimen tilts, so that the tilt range will cover only a few degrees, making use of the fact that the contrast in DF images is typically very sensitive to the specimen orientation. This method relies on the strong DF contrast between locations in the specimen whose local crystal orientation is slightly different, leading to the well-known bend contours (see Fig. 1). Since the proposed technique acquires a tilt series in image mode, any specimen shifts due to non-eucentric tilting or goniometer imperfections can be compensated easily.

In Section 2, we discuss in detail the specimen preparation, data acquisition, and the concept and different tools used for data analysis. In Section 3, we present the recovered orientation map and, then discuss possible applications in Section 4.

2. Method

2.1. Experiment

The specimen from which the experimental data was collected was prepared from a Ni-based super-alloy CMSX – 4® (Cannon Muskegon Corporation, Muskegon,USA). Due to its strength the material is used for high-temperature applications, e.g. in gas turbine engines. The material has an *fcc* crystal structure and consists of γ and γ' phases. The γ phase represents the ordered $L1_2$ structure, where *Al* and *Ti* atoms form alternating atomic planes. The specimen was prepared after stress was applied to it which leads to the deformation of the material. After applying stress, the interface between the γ and γ' phases appears wavy in cross-section, as can be seen in Fig. 1. This phenomenon is known as *rafting*. If no stress is applied, the γ' phase precipitates in cube-shaped domains surrounded by γ matrix channels. The specimen was prepared using electrochemical polishing according to [18], producing a geometry in which the specimen becomes thinner when approaching the edge. The average specimen thickness at the location investigated during the current investigation was approximately 200 nm. The crystal lattice is expected to be deformed due to the interface lattice misfit, lattice relaxation in thin parts and plastic strain. These deformations induce variations in the crystal orientation across the field of view (FOV). In the bright-field (BF) and DF images in Fig. 1 one can see very clearly the resulting bend contours. This makes this specimen very suitable for testing our technique. We also note visible in Fig. 1 the rapidly changing complex diffraction conditions across the interface occurring due to the lattice mismatch at the interface between the γ and γ' structures.

The experiment was carried out using the Sub-Ångstrom Sub-

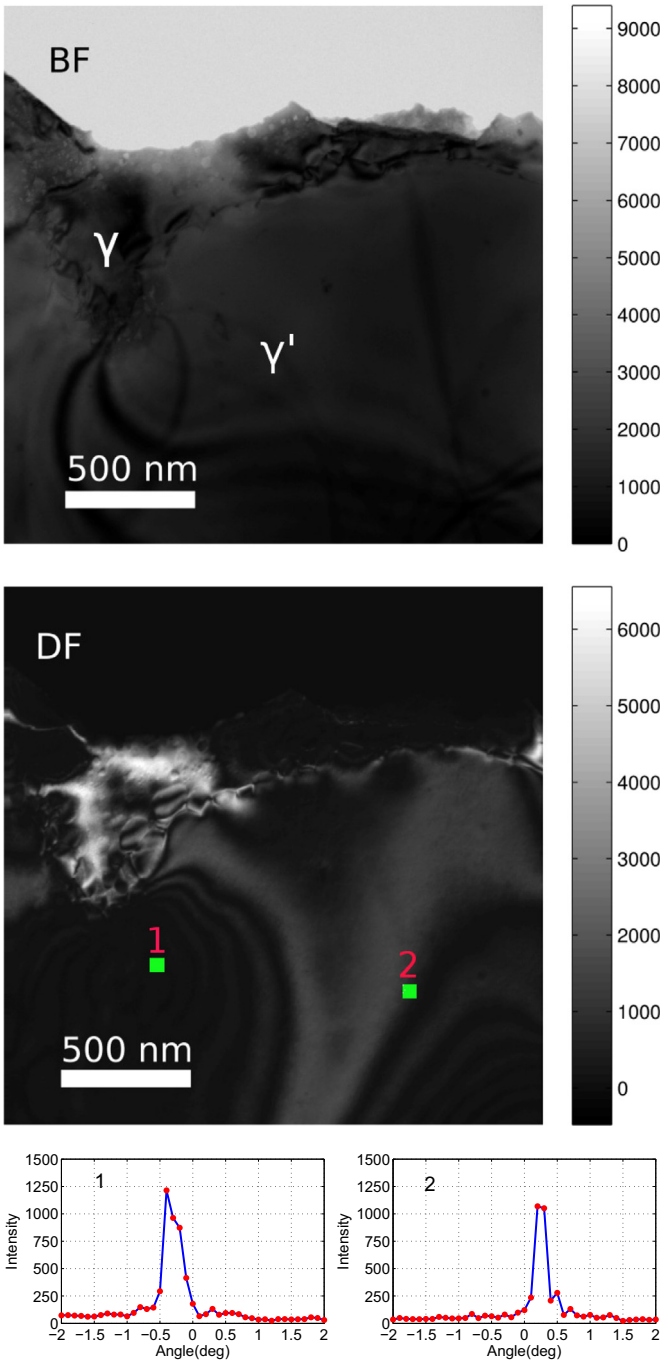

Fig. 1. *Top and middle*: one BF and one DF ($g = \bar{3}\bar{1}1$) image extracted from the respective tilt series used for this analysis. Regions composed of the γ and γ' phases are identified. *Bottom*: rocking curves extracted from the data at the two positions indicated in the image above.

Electron volt Microscope (SESAM) equipped with an in-column MANDOLINE filter (Carl Zeiss NTS, Oberkochen, Germany) [19]. The experimental DF tilt series was acquired by means of a fully automated acquisition procedure, implemented in-house as a script in DigitalMicrograph™ (Gatan Inc., Pleasanton, CA, USA) employing a $2k \times 2k$ charge-coupled device (CCD) camera (UltraScan™, Gatan, Inc.). Each gain-reference-corrected image was acquired at a nominal magnification of 10,000 × without hardware binning. These settings result in a pixel size of the acquired images of 1.01162 nm. The data stacks corresponding to different reflections were acquired for different exposure times determined using the strength of the reflection which in turn was judged by the