Ultramicroscopy 160 (2016) 146–154

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Pattern matching approach to pseudosymmetry problems in electron backscatter diffraction

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ARTICLE INFO

Article history:
Received 3 May 2015
Received in revised form
9 October 2015
Accepted 11 October 2015
Available online 23 October 2015

Keywords:
Pyrite
EBSD
Pseudosymmetry
Cross-correlation
Pattern simulation
Hough transformation
Orientation precision

ABSTRACT

We demonstrate an approach to overcome Kikuchi pattern misindexing problems caused by crystallographic pseudosymmetry in electron backscatter diffraction (EBSD) measurements. Based on the quantitative comparison of experimentally measured Kikuchi patterns with dynamical electron diffraction simulations, the algorithm identifies the best-fit orientation from a set of pseudosymmetric candidates. Using measurements on framboidal pyrite (FeS_2) as an example, we also show the improvement of the orientation precision using this approach.

1. Introduction

The characterization of crystalline materials using electron backscatter diffraction (EBSD) in the scanning electron microscope (SEM) is a powerful tool for many different applications [1,2]. However, a fundamental problem in EBSD analyses concerns possible misinterpretations ("misindexing") of the measured Kikuchi patterns. Misindexing directly influences the qualitative detection of grains but also the quantitative characterization of grains and their boundaries. If misindexed data points are substituted in data-cleaning techniques, this can lead to questionable results [3]. Therefore, a correct orientation determination is a fundamental requirement for any subsequent microstructural characterization, and the prevention and the reliable correction of misindexing is clearly an important topic [4,5].

To a large extent, the symmetry of Kikuchi patterns is governed by the point group of a crystal. Crucial examples for misindexing problems are caused by phases that give rise to Kikuchi patterns with an apparently higher point group symmetry than is expected for the analyzed phase. This phenomenon is described by the term pseudosymmetry, which has been discussed in several EBSD-related publications [6–9,3]. In the presence of pseudosymmetry, an unequivocal orientation determination depends on the ability of the applied data analysis method to detect the significant deviations from the apparently higher symmetry. In this context, physics-based Kikuchi pattern simulations [10] can be helpful to quantify the potential symmetry breaking parts of a Kikuchi pattern, the scope of which can limit our actual ability to detect the corresponding changes in the experimental data.

Using framboidal pyrite (FeS_2) as an example, we show in the present paper how misindexing of pseudosymmetric Kikuchi patterns can be corrected by a pattern matching approach [11,10]. The comparison of experimentally measured Kikuchi patterns with dynamical electron diffraction simulations [12] makes it possible to identify the best-fit orientation from a set of pseudosymmetric candidates based on their cross-correlation coefficients. This procedure also quantifies the theoretical dissimilarity of the pseudosymmetric Kikuchi pattern candidates and thus allows estimation of the experimental limits of pseudosymmetric orientation discrimination. Additionally, the pattern matching approach also provides refined orientation parameters, which, in the investigation presented here, are at least two-times more precise than the values from the conventional orientation determination.

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http://dx.doi.org/10.1016/j.ultramic.2015.10.010
0304-3991/

2. Experimental details

2.1. Sample description

We investigate pseudosymmetry effects in framboidal pyrite from the Chattanooga Shale. Collection and sample preparation is given in detail in [7].

The pyrite framboids are typically 10 μm in diameter with individual microcrystals of pyrite being <1 μm in diameter, cf. Fig. 1 (a). Microcrystals within a given framboid vary little in size, and typically present idiomorphic crystals of cuboctahedral habit. The microcrystals may appear systematically arranged as shown in Fig. 1(a) or disordered [7]. The framboids tend to occur in clusters within the sample and are associated with marcasite, an orthorhombic polymorph of FeS_2, and quartz.

2.2. Data acquisition

The orientation maps have been generated using a field-emission SEM LEO1530 VP (ZEISS) with a Bruker EBSD system. The acceleration voltage was 20 kV and the beam current was ≈ 7 nA. The collected EBSD patterns were binned down on-chip to a size of 160 × 120 pixels. All collected raw Kikuchi patterns were stored in a data file for offline re-analysis and post-processing [13,14]. An isometric grid at spacing 60 nm was chosen for the orientation maps of framboids.

The maximal possible dwell time per frame was 15 ms. In order to improve the signal-to-noise ratio, 3 camera frames have been averaged for each individual Kikuchi pattern, see Fig. 1(b). For noise-reduced Kikuchi patterns derived from standard orientation maps, a pattern averaging algorithm has been applied [11]. This approach averages a number of Kikuchi patterns from a circular region of interest. The patterns are selected automatically on the basis that they show the best match to the reference pattern given by the central position of the selected region.

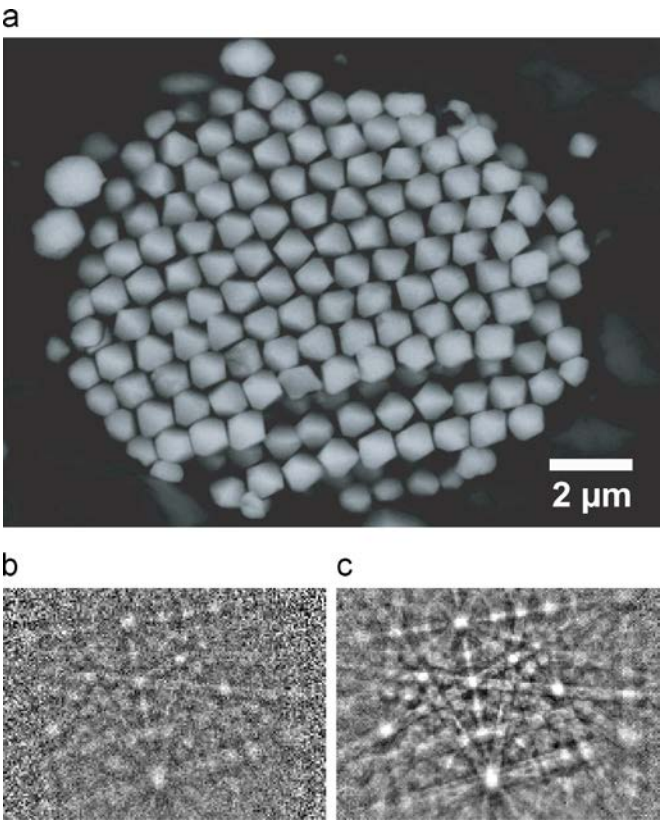

Fig. 1. A backscattered electron image of framboidal pyrite before EBSD preparation (a) shows a highly ordered arrangement of individually grown cuboctahedral microcrystals of pyrite [7]. Despite the comparatively long acquisition time a single, the background-corrected experimental Kikuchi pattern (160 × 120 pixels) in (b) is of low quality. The pattern in (c) represents the average of 32 single patterns collected in one microcrystal.

2.3. Data processing

EBSD data acquisition and indexing has been done using ESPRIT 1.94 (Bruker Nano). The Kikuchi pattern indexing was set up to work with between 12 and 6 identified Kikuchi bands. For the simulation of theoretical Kikuchi patterns, and for the discrimination between the pseudosymmetric orientations, we used ESPRIT *DynamicS*. The optimization of the orientational fit between the simulated and the experimental Kikuchi pattern in *ESPRIT DynamicS* involves reprojection of simulated test patterns from the theoretical master data, where the step size in orientation space is dynamically adapted according to the progress of the fit procedure. The removal of systematically misindexed data points by enforcement of consistent neighboring orientations (see below) was carried out in Tango (Channel5, Oxford Instr.).

All orientation and misorientation maps presented here have been created using the free and open-source Matlab toolbox MTEX [15]. MTEX offers inverse pole figure (IPF) color keys that suppress arbitrary color discontinuities for nearby orientations [16]. Moreover, MTEX colorizes EBSD orientation maps with respect to the, in general non-centrosymmetric, point-group of a phase instead of only the centrosymmetric Laue group [17,10]. This also includes coloring according to the proper rotation (enantiomorphic) groups [18], which are relevant for a correct description of crystal orientations.

3. Theoretical prerequisites

3.1. Pseudosymmetry

The symmetry of a crystal property or the phenomenon observed as result of a crystal property can be higher than the symmetry of the crystal itself and we thus may not be able to derive the full crystal symmetry from a measurement of the respective property (Neumann's principle) [19]. This can be due to *theoretically necessary* restrictions that the symmetry of the crystal structure places on a physical property tensor.

In this investigation, we use the term “pseudosymmetry” to describe an *experimental* limitation to detect deviations from a higher symmetry in a measured Kikuchi pattern (resulting from the interaction of electrons with the crystal structure). For instance, this can be caused by an inadequate analytical model for the interpretation of the experimental data [10], or by an insufficient signal-to-noise ratio. As the symmetry of Kikuchi patterns is governed to a large extent by the point group of a crystal, it is crucial to classify the patterns according to the presence and absence of characteristic symmetry elements. This can be experimentally challenging if the symmetry breaking is small and thus, effectively, a higher symmetry (“pseudosymmetry”) is assigned to the pattern. The resulting equalization of nonequivalent crystal orientations is central to the misindexing problem in the presence of pseudosymmetry.

3.2. Symmetry of the pyrite crystal structure

As indicated by the extended Hermann–Mauguin symbol $P2_1/a\bar{3}$, the crystal structure of pyrite does not contain any fourfold rotation or screw axis parallel to ⟨0 0 1⟩ [20]. The missing 90° rotation operation can be seen by the arrangement of the