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Accurate determination of low-symmetry Bravais unit cells by EBSD

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ABSTRACT

Unit cells lack of symmetry are difficult to determine accurately, compared to high-symmetry unit cells with many constraints. The electron backscatter diffraction (EBSD) technique in scanning electron microscopy (SEM) was considered inadequate for this task because of the highly defective band detections. We develop a new method for the Kikuchi-band detections, which can improve the accuracy of the EBSD technique in determining the lattice constants of totally unknown Bravais unit cells with low symmetry. The results show that, under ideal conditions (i.e., low-noise EBSD patterns and known projection center), the relative error of the unit-cell constants (*a, b, c*) is less than 0.3%, and that of the axial ratios (*a/b, b/c, c/a*) is less than 0.5%. The absolute errors of the inter-axial angles (*α, β, γ*) and crystal orientations are about 0.1°. Our method is perhaps not as accurate as the classical techniques such as X-ray diffraction, but is demonstrated as a practical tool for crystallographic characterization especially on low-fraction phases, and could be easily incorporated into an SEM to make the most of the SEM in the area of microanalysis.

1. Introduction

Crystals are built up by an orientational stacking of very small regular 'brick-like' unit cells. Edges of the unit cells are considered to be parallel to the three axial vectors ($\vec{a}$, $\vec{b}$, $\vec{c}$) of the seven crystal systems which are subdivided into 14 Bravais-lattice types according to different symmetries. In general, lattice constants are sensitive to a variety of external and internal conditions, such as temperature, stress, chemical composition, etc. Accurate determination of the Bravais unit cells (including their symmetries and lattice constants) is prerequisite to phase identification, calculating the atomic distance and bond energy, and analyzing the relations between lattice constants and various physical/chemical properties.

However, accurate determination of the Bravais unit cells is not trivial even for the cubic system with many symmetry constraints, and it becomes more difficult for the low-symmetry crystal systems, especially the triclinic one [1–3]. For the crystalline materials with low symmetries, their crystal structures are generally complex and often accompanied by relatively poor crystalline perfection with residual stress and fine grain size. These characteristics make their diffraction patterns complicated and any tiny error or mishandling in the data processing will severely impact the accuracy and reliability of the final determination results [4,5]. Take the X-ray diffraction (XRD) technique as an example, the best accuracy of measuring the lattice constants for cubic system is about 0.0005%, which is close to the accuracy of measuring the X-ray wavelength and thus is possibly so far the limit of measuring lattice constants [6,7]. In ordinary cases, the measurement accuracy for cubic system is about 0.002% [8,9]. For triclinic system, however, the best accuracy, under the condition of no systematic errors (e.g., instrumental misalignment, incident beam divergence, off-center specimen, specimen absorption, etc.), is 0.01% [10], one order of magnitude lower than that for cubic system. In practical measurement, it is, of course, not free from sources of the systematic errors. As a result, the measurement accuracy for triclinic system deteriorates even further to about 0.3% [10], two orders of magnitude lower than the ordinary cases for cubic system.

Given the above accuracies of the XRD technique, it is still challenging to characterize the phases with a low fraction because of their insufficient X-ray diffraction signals. In this case, the popular alternative technique is the transmission electron microscopy (TEM). But it requires time-consuming sample thinning and sometimes may not be readily available.

Nowadays, electron backscatter diffraction (EBSD) technique in scanning electron microscopy (SEM) is comparatively widespread for microanalysis. It is a locally resolving technique that enables a combination of the microstructural and orientational characterizations,

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such as morphology and texture based on orientational analysis of known crystals. It nonetheless has an unfavorable reputation of being too inaccurate to measure the lattice constants reliably. For lattice-constant determination, EBSD's best accuracy is considered to be 5%, and the error of the data directly extracted from EBSD patterns may be up to 20%, because of the normally vague diffraction patterns and the highly defective band detections [3,11].

In our previous works, a reliable three-dimensional reconstruction procedure was reported, which involved as many as possible Kikuchi bands that were visible in an EBSD pattern and built a system of equations (tens of thousands of) for a least-squares solution in order to over-determine the unknown unit cells by using a single EBSD pattern [12–16]. The corresponding determination error was confined mainly by building the overdetermined system of equations. Although the obtained accuracy (about 1% for lattice constants [17]) is reasonably high, the adopted band-detection method remains inherently unsatisfying and the accuracy was derived only from cubic and tetragonal unit cells. In this work, we propose an improved method for the Kikuchi-band detections, which utilizes the crystallographic information extracted from an EBSD pattern to further confine the determination error. Then we apply this new method to determine low-symmetry unit cells. The obtained accuracies for materials with triclinic unit cells are better than 0.3% for both the unit-cell constants (*a, b, c*) and the axial ratios (*a/b, b/c, c/a*), better than 0.3° for the inter-axial angles (*α, β,* γ), and 0.1° for crystal orientations. This is significantly better than the previous accuracy we achieved on high-symmetry cubic or tetragonal materials, demonstrating the great potential of our method for structure determination on SEM.

2. Geometric crystallography of EBSD

EBSD signals emitted from an effective point source inside a crystal sample are gnomonically projected onto a flat screen of the EBSD detector, recording a two-dimensional Kikuchi diffraction pattern. In this three-dimensional configuration of gnomonic projection, the ratio between the specimen-to-screen distance and the height of the Kikuchi pattern is defined as the detector distance (according to a definition by the Bruker Corporation), which affects the pattern's angular coverage φ (Fig. 1a). Typically, an EBSD pattern covers an angle φ in the range of 70–100°. A point in the two-dimensional pattern with the shortest distance to the emitting source (i.e., projection center) is usually called the pattern center.

From the crystallographic point of view, a single EBSD pattern provides abundant information about the crystalline phase (including its real and reciprocal unit cells). To be specific, in an EBSD pattern tens of Kikuchi bands represent the electron diffraction signals of lattice

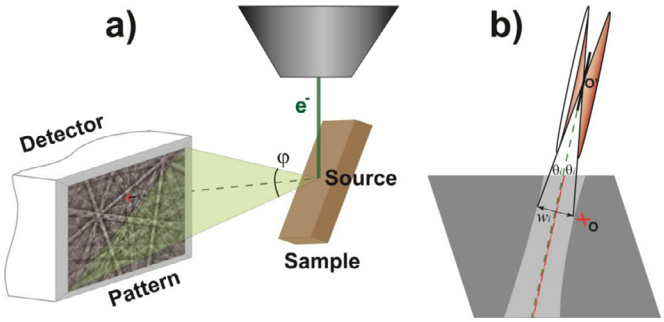

Fig. 1. Geometric crystallography of the EBSD: (a) Three-dimensional configuration of the gnomonic projection; (b) Discrepancy between the trace (green) of a diffracting lattice plane (*hkl*) and the center line (red) of a hyperbola-shaped Kikuchi band. The plane (*hkl*) bisects the angle $2\theta_i$. w_i is the band width. O and O' are the pattern center and the source, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

planes $(hkl)_i$. The widths of bands decide the absolute values of the unit-cell constants (*a, b, c*) according to Bragg's equation $2d_{(hkl)}\sin\theta_i = n\lambda$, where θ_i is approximately proportional to the width of a Kikuchi band (Fig. 1b). These Kikuchi bands intersect each other, forming hundreds of Kikuchi poles, namely, zone axes $[uvw]_m$: $(hkl)_i \times (hkl)_j = [uvw]_m$, with $hu + kv + lw = 0$. Here, the indices of *h, k, l,* and *u, v, w*, are all integers, which means that all bands and poles showing visible Bragg diffraction contrast within the angular coverage of an EBSD pattern have integer Miller indices which are free of errors. The positions of the diffracting-plane traces and zone axes together with the projection center depend on inter-axial angles (*α, β,* γ), axial ratios (*a/b, b/c, c/a*), and crystal orientations (e.g., the three Euler angles, Z, X, Z). The widths of the Kikuchi bands are associated with the magnitude of lattice constants. The success (including high accuracy and high reliability) of determining the Bravais unit cells based on the EBSD technique is therefore strongly dependent on the accuracy and precision of the adopted band-detection method. In particular, correctly locating the traces and describing the band widths of diffracting lattice planes are of critical importance.

Unfortunately, it is difficult to accurately determine the traces and widths in an EBSD pattern. Owing to the gnomonic projection (Fig. 1), the two edges of each band have a hyperbolic shape, and its width is approximately proportional to twice the Bragg angle ($2\theta_i$) of a diffracting lattice plane $(hkl)_i$. The trace of a diffracting lattice plane$(hkl)_i$, which is an invisible line of intersection between the $(hkl)_i$ plane and the Kikuchi pattern, is not coincident with the center line of corresponding hyperbola-shaped Kikuchi band, as illustrated in Fig. 1b. The maximum deviation between a center line and corresponding trace appears to be only few image pixels (about 0.1° within the angular coverage φ shown in Fig. 1a) and hence difficult to notice in an EBSD pattern. Such inaccuracy in trace positions may not affect the directions of the traces and the angles between them, but will cause considerable errors in the locations of zone axes as well as the following unit-cell determination.

With respect to the widths of Kikuchi bands, it is also difficult to perform an accurate measurement in an EBSD pattern because the intensity profiles of the diffraction bands are complex [18]. Thus the lattice spacing $d_{(hkl)}$ ($d^{-1}_{(hkl)} \propto \theta_i$) obtained from measurement of the band widths tends to be rather imprecise (with error up to 20%). In consequence, using the erroneously measured$d_{(hkl)}$, a poorly defined reciprocal primitive cell will be reconstructed, which eventually leads to an inaccurate and unreliable determination of the real unit cell.

In spite of the aforesaid unfavorable features of EBSD patterns, accurate determination of the traces and band widths is still achievable. For example, a large amount of zone axes in a single EBSD pattern can be accurately determined (which is why the EBSD technique is widely accepted for accurate crystal orientation analyses). Moreover, if gnomonic distortion of the EBSD pattern was corrected and also the pattern center, detector distance and crystal orientation were accurately known, all trace positions can be correctly calculated. The deviation between the trace and the center line of a hyperbola-shaped band becomes obvious for high-order Kikuchi bands.

Nevertheless, in the case of experimental pattern analysis, the pattern center and detector distance are difficult to determine with high accuracy [5,19], and not every Kikuchi band shows a high-order feature. In view of these practical reasons (e.g., inaccurate or even unknown projection center), our routine of determining the band edges (or measuring band widths) is mainly composed of two stages.

At the initial stage, we perform measurement with a recorded or estimated pattern center and detector distance (their detailed estimation process will be introduced in the next section). Hence, an approximation is made, i.e., the trace and center line are regarded as being coincident (only for initial Kikuchi band detection). Subsequently, we can use a pair of parallel lines and their center line to approximate the hyperbolic band edges and the corresponding trace, respectively. Here the band width w_i (marked by two arrows in Fig. 1b)