

# TEM-based orientation mapping of polycrystalline materials

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## *Introduction*

Knowing the ‘topography’ of grain orientations is essential for investigation of polycrystalline materials. Maps of orientations are usually obtained by determining orientations of individual grains via diffraction. In practice, to ensure appropriate spatial resolution, *electron* diffraction is used. Automatic orientation mapping of polycrystalline materials by electron back-scattered diffraction (EBSD) and scanning electron microscopy (SEM) is already a well established technique, and commercial EBSD systems are widely used. There exist similar systems based on transmission electron microscopy (TEM). They are created in hope for them being used in applications, for which EBSD systems are not suitable. This particularly applies to mapping of microstructures requiring a high spatial resolution.

Automatic TEM-based mapping systems stem from computer supported indexing of diffraction patterns. Such approach with manually selected grains and manually detected positions of reflections is convenient for investigating small sets of grain orientations (see, e.g., Zaeferrer, 2000, Seyring et al., 2011). Automatic mapping is less tedious, and it eliminates subjective selection of grains.

There are several types of TEM-based mapping systems founded on somewhat different principles. Common to all of them is the use of analytical microscopes equipped with cameras. They have an ability to save automatically images or diffraction patterns, and to compute orientations for all pixels of a map. Thus, the main components of the systems are image/pattern acquisition procedures and the software for orientation calculation. For an example map obtained using TEM see Fig. 1.

## *Pattern acquisition*

The systems differ by the methods of scanning the specimen and by type of diffraction patterns used for calculating crystal orientations. Like in EBSD systems, TEM-based orientation maps can be created by a stepwise scanning of the specimen with the electron beam and by acquisition of diffraction patterns at each step of the scan (Fundenberger et al., 2003, Rauch and Dupuy, 2005). For this, the computer controlled pattern acquisition is coordinated with the repositioning of the beam (more specifically, with the current in beam deflection coils). Since the mapping may take a considerable time, an issue of the beam drift arises. Experimental practice shows that, ordinarily, with thermally stable environment (microscope and sample holder), the drift is negligible. The magnitude of the drift can be checked by moving the beam to the starting position after completing the scan. Moreover, in many cases, the mapping leaves a clearly visible grid of contamination spots, and the absence of the drift can be verified visually.

An alternative approach, characteristic only to the transmission technique, is known as a ‘dark field conical scanning’ (Wright and Dingley, 1999, Dingley, 2006). In this case, a series of dark-field images is collected for various incident beam directions. For certain directions, a given crystallite or its part, a pixel, will be in the diffraction condition, and it will be bright in the corresponding images. This allows for reconstruction of the (spot) diffraction pattern, and then, for calculation of the orientation at the considered pixel.

As for the types of patterns, so far either Kikuchi (Fig. 2) or spot diffraction patterns (Fig. 3) have been used. The Kikuchi patterns appear as a result of inelastic scattering of electrons (in relatively thick foils). These patterns are in a sense ‘fastened’ to crystals and, in consequence, they are sensitive to crystal orientation. The spot patterns used in mappings are acquired in nano-beam mode. The patterns have the same geometry as selected area diffraction patterns but instead of a parallel electron beam, convergent beam and a very narrow probe are used. Reflections in spot patterns blink on and off as the crystal orientation passes near the Bragg condition. The range of orientations producing detectable spots is determined by the excitation error and can be relatively large; in effect, the sensitivity of spot patterns to crystal orientations is limited.

## *Indexing and orientation determination*

The indexing, i.e., the assignment of Miller indices (hkl) to particular reflections of a diffraction pattern, is practically equivalent to orientation determination; an orientation can be easily calculated from known indices, and vice versa, with known orientation, one can easily assign the indices.

Two distinct approaches to pattern indexing are used. One of them is referred to as “template matching”. Briefly, orientations are determined by matching experimental patterns to simulated patterns (templates). With known crystal structure, the templates are simulated for a net of orientations. The orientation

corresponding to the template with the highest correlation to the experimental pattern is taken as the final result.

Since the computation of templates is time consuming, they are calculated once for a given material and measurement geometry and stored on a computer disk. The number of templates is determined by the distances between the orientations of the net. The average distance needs to be linked to the orientation resolution of the method. With relatively low sensitivity of spot patterns to crystal orientation, the number of templates needed to solve these patterns is relatively small.

The required number of templates is also influenced by the symmetry of the material. Due to the crystal symmetry, some orientation parameters are mutually equivalent, and only one set needs to be considered. In practice, this means that for *fcc* and *bcc* materials only 1/24 of the complete orientation space must be covered by the templates. Moreover, with the standard measurement geometry, for the crystallites differing in orientation by a rotation about the optical axis of the microscope, the corresponding diffraction patterns differ by a rotation about the pattern center. Therefore, it is sufficient to prepare just one template per zone axis, and to use rotations about the center in matching the experimental pattern to the templates. With this, the templates need to cover a region twice as large as the standard triangle.

The first to apply computerized 'template matching' for solving spot patterns were Rauch and Dupuy (2005). A similar approach was also used in combination with the 'dark field conical scanning'; orientations were obtained by matching templates to 'spot' patterns calculated from dark field images collected via conical scanning (Wu and Zaefferer, 2009). Template matching is not used for solving Kikuchi patterns. The reason is that with their sensitivity to crystal orientation, a prohibitive number of templates would be required. Therefore, Kikuchi patterns are solved via detection of positions of reflections.

#### *Detection of reflections*

In the case of Kikuchi patterns, the software needs to determine the positions of both individual (excess and deficiency) lines of a Kikuchi pair, and this must be done for a number of pairs in the pattern. (In the case of spot patterns, one would have to determine the positions of particular spots.) The detection of Kikuchi lines is carried out using the Hough transform – a standard algorithm for digital line detection. The reliability of the detection can be considerably improved by using the so-called 'backmapping' of Gerig and Klein (1986). With known positions of individual Kikuchi lines, pairs of lines need to be identified. This is done by using the requirements that the lines of a pair must be nearly parallel, and the distance between them must be within brackets calculated based on the camera length and the list of detectable reflections. To limit the detection time, all constant factors involved are calculated beforehand.

#### *Indexing of reflections*

After determining the locations of reflections, one can proceed to pattern indexing, and to the calculation of orientation. In a simplistic approach, a pattern can be indexed by comparison of angles calculated based on the positions of reflections with those obtained from the known crystal structure. Usually, these are inter-planar angles.

More formally, the coordinates of the reciprocal lattice vectors, say  $g_n$ , ( $n=1,\dots,N$ ) in an external (sample or microscope) coordinate system are calculated from the geometric positions of reflections. (At least three non-coplanar vectors are needed for unambiguous solution, and in current practice  $N$  does not exceed 10.) On the other hand, the reciprocal lattice vectors, say  $h_m$ , ( $m=1,\dots,M$ ) in the crystal coordinate system are obtained based on the known structure of the material and kinematically calculated intensities; typically, in the case of Kikuchi patterns of *fcc* materials, the list of vectors contains 14 to 20  $\{hkl\}$  families with Miller indices up to, say 5.

Now, in the general case, one has two sets of reciprocal lattice vectors:

- $g_n$  vectors in an external coordinate system, and
- $h_m$  vectors in the crystal coordinate system.

Some of the vectors  $g_n$ , may be spurious (false detection) but those corresponding to genuine lines, after some unknown rotation, must match some of the  $h_m$  vectors. The indexing is reduced to finding the assignment of the legitimate  $g_n$  vectors to some of the  $h_m$  vectors, and the orientation determination is reduced to finding the unknown rotation. These tasks are carried out by dedicated software.

#### *Reliability of orientations*

The reliability of orientations in TEM-based maps depends primarily on the quality of the diffraction patterns. The quality is determined by the quantity of defects in the crystallites. Thus, for instance, patterns originating from highly deformed materials (or highly deformed areas of the specimen) can be diffuse and, in effect, unsolvable.

Moreover, ambiguities arise for “overlapping” patterns acquired near grain boundaries. They originate from two (or more) differently oriented crystallites, and the indexing software may assign wrong orientations to them. This effect is similar in both SEM and TEM-based systems but the issue is more significant in the latter case. The TEM-based systems are applied to materials with very fine structures (small crystallites). On the other hand, a reasonable foil thickness is needed to see the structure element of interest (say, shear bands) or to get diffraction patterns (substantial thickness is necessary for getting Kikuchi patterns). With this, the pattern overlap may become a crucial factor determining the quality of the orientation maps.

There is an additional difficulty linked to the solid angle covered by patterns. If the angle is too small, the so-called 180deg ambiguity arises: two crystals differing in orientation by a 180deg rotation about the optical axis of the microscope may lead to identical diffraction patterns. A typical example of such ambiguity is the [112] zone axis pattern of *fcc* materials; it is symmetric with respect to a half-turn about its center (Fig. 4), whereas the half-turn about the [112] axis is not a symmetry operation of the crystal.

The 180deg ambiguity may arise in the case of Kikuchi diffraction only if the camera length is relatively large. The problem is more serious in the case of spot patterns (Morawiec and Bouzy, 2006). It cannot be removed by decreasing the camera length because intensities of high order reflections are too low for the reflections to be used in solving the patterns. In the commercial system described by Rauch et al. (2010), the problem is resolved by application of the precession method of Vincent and Midgley (1994). In the precession method, the beam is deflected prior to the interaction with the material, and then moved back after the interaction. The number of reflections excited for a given deflection is not large but with the precession, the beam forms a cone of illumination, and in effect, spots of significant intensities cover a solid angle much larger than in the conventional case. The precession of the beam is equivalent to rocking the crystal but the precession angle is small, and the determined crystal orientation is affected negligibly.

#### *Orientation resolution*

The accuracy of orientations in orientation maps is influenced by a number of factors. In practice, it is usually dominated by the inaccuracies in keeping the right orientations during foil preparation and while mounting the specimen in the specimen holder. However, there is frequently more interest in *precision* of orientations than in their accuracy.

The precision of the Kikuchi technique is different for different orientation parameters. The parameters describing rotations about axes perpendicular to the optical axis can be determined with very high precision below 0.1deg (depending on camera length). The precision of the angle of the rotation about the optical axis is much lower; with currently used cameras, it is about 0.2deg.

The precision of orientations obtained from spot patterns depends on the experimental setup and software. Experiments show that about 90% of results fall within 1.5deg from the orientations determined via Kikuchi patterns. In this case, the angle of the rotation about the optical axis is expected to be the most precise of orientation parameters.

#### *Spatial resolution*

The spatial resolution of the TEM-based mappings is primarily influenced by the probe geometry and foil thickness. With nano-beam diffraction, the usual probe size is 5nm, and the convergence angle is about 0.5-1mrad. The same probe diameter is usually used for Kikuchi patterns. The step size needs to be adequate to the investigated microstructure; typically, it is about 20nm. As for the foil thickness, it limits the resolution of the systems making use of Kikuchi diffraction because it requires foils of a considerable thickness (about 200-300nm in typical conditions).

#### *Applications*

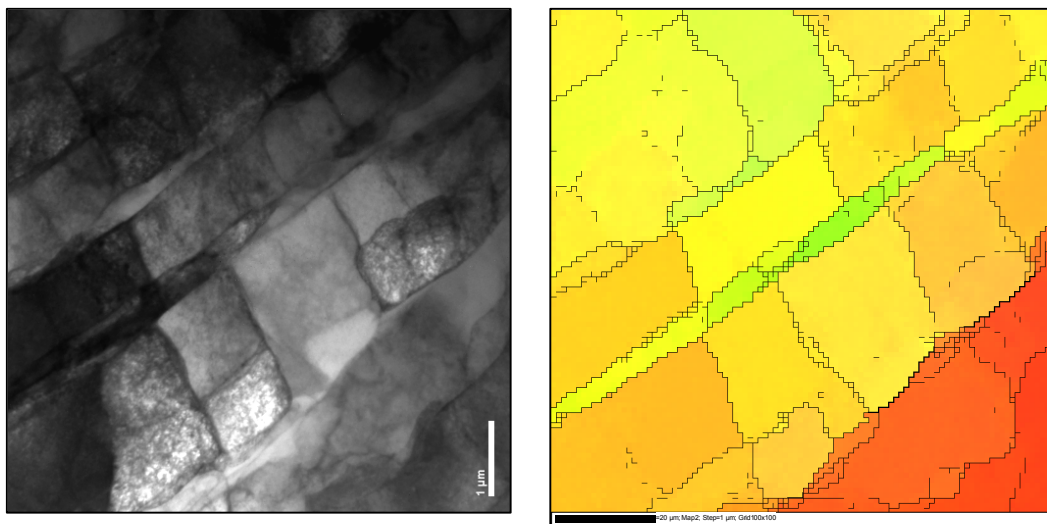
Although the TEM-based orientation mapping systems are still *in statu nascendi*, and most papers on the technique concern its development, there exist a number of examples of their applications. One of them is the explanation of shear band formation and the impact shear bands on textures of *fcc* materials of various stacking fault energies given by Paul et al., (2004, 2009).

It is worth mentioning that besides crystallite orientations, the maps may discriminate between phases present in the investigated material; for an application of the phase identification, see (Góral et al., 2005).

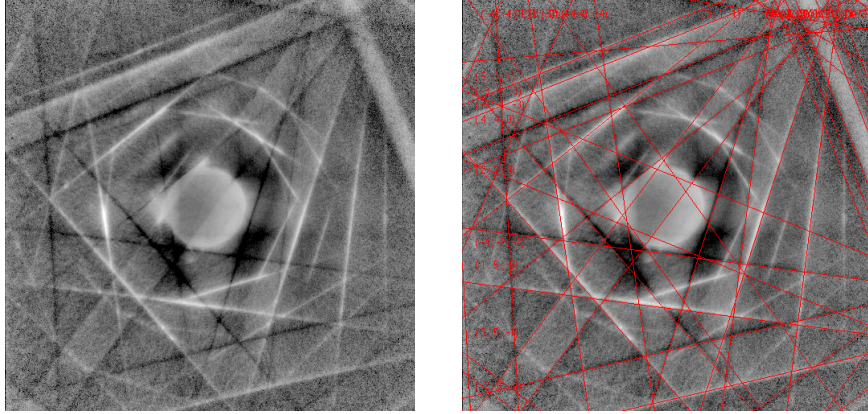
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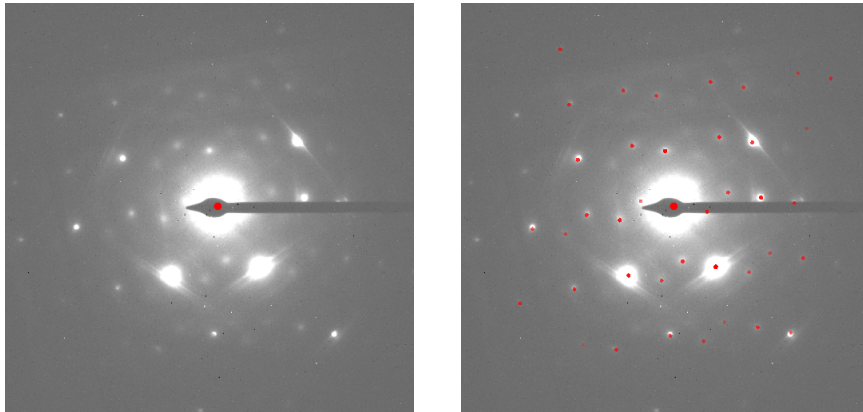
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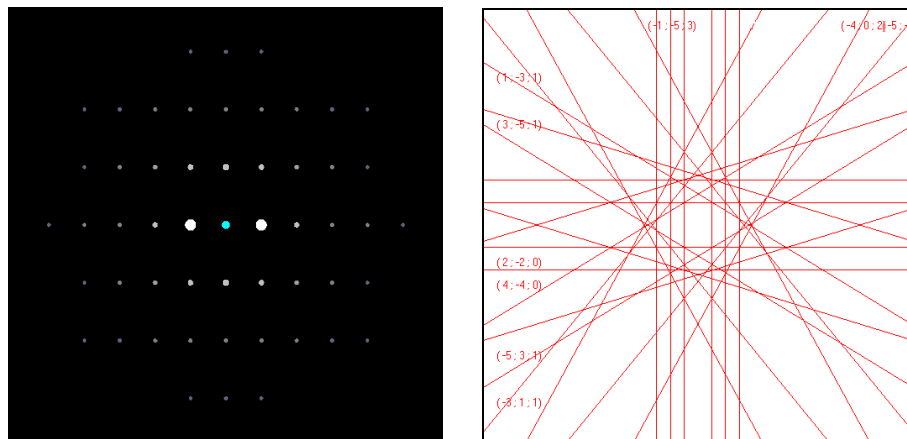
**Figure 1.** A TEM image of microstructure of Al and an example map created using a system based on Kikuchi patterns. Courtesy of H. Paul.



**Figure 2.** A Kikuchi pattern of Al and the same pattern with an automatically obtained solution. The zone axis is about 0.5deg from  $[5\ 4\ 7]$ .



**Figure 3.** A spot pattern of Al and the same pattern with an automatically obtained solution. The zone axis is about 2.4deg from  $[1\ 4\ 9]$ .



**Figure 4.** Schematics of  $[112]$  spot and Kikuchi patterns of Al (200kV). The patterns are invariant with respect to 180deg rotation about their centers.