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Backscatter and transmission Kikuchi diffraction for materials science

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Abstract

Over the last decades automated Kikuchi diffraction in transmission (TKD in the TEM) and in backscatter mode (known as EBSD in the SEM) has gradually evolved into an invaluable tool of materials characterization. It provides, at a sub-micron scale, a detailed quantitative description of grain orientations, material phases, state of deformation, local texture, and intuitively accessible visualizations of the microstructure in the form of orientation images. Recently interest in TKD has risen again, in particular as a supplementary facility to an EBSD system, driven by a wishful dream of improving resolution down to the nanoscale.

Experimental setups are briefly introduced. The limitations of TKD and EBSD as set by spatial resolution, grain size and bending of thin foils are discussed in more detail.

Keywords: Transmission Kikuchi diffraction TKD; backscatter Kikuchi diffraction BKD; EBSD; spatial resolution; bend contours.

1. Introduction

The first Transmission as well as Backscatter Kikuchi Patterns (later named TKP and BKP) were published 1928 in the same volume of Proceedings Imperial Academy of Japan [1, 2]. During the next decades physicists focused their experimental work preferably on Transmission Kikuchi Diffraction (TKD) because of instrument-based reasons. After the SEM became common place in the labs Backscatter Kikuchi Diffraction (BKD) in the SEM, then named Electron Backscatter Diffraction (EBSD), received attention of material scientists for texture analysis. Finally, since the end of last century, BKD is about to prevail thanks to the availability of commercial systems, the ease of operation, the convenient preparation of bulk samples, and the enablement of dynamic experiments [3, 4]. Recently interest in TKD has risen again, in particular as a supplementary facility to an EBSD system, driven by a wishful dream of improving resolution [5]. Current research focuses on nanoscale materials or severely plastically deformed materials, which may show drastic changes in properties with often unexpected results.

Most of natural as well as man-made materials (e.g. minerals, rocks, bones; metals, ceramics, crystalline polymers) have a polycrystalline structure as an intrinsic feature, but they are often assumed to behave uniformly with equal strength in all directions. The anisotropy of the individual crystals, however, is only balanced out in case of a large number of grains *and* a random distribution of the grain orientations [6]. In reality, a residual anisotropy is found which varies, depending on the actual statistical distribution of grain orientations – the crystal "texture", "preferred crystal lattice orientations" –, between the extreme values of the anisotropic physical property of interest with which a single crystal of the same material will respond to directional tests. *Local* variations in *crystal texture* as well as the arrangement and type of *grain/phase boundaries* may give rise to *inhomogeneous material properties* which may finally limit the performance of a workpiece. Texture forms during grain growth or deformation and is modified during recrystallization or phase transformations. Texture so bears information about the history of materials' processing and use. Therefore, the knowledge of texture is of great concern for quality control in a wide range of industrial applications and for basic materials research. Theories exist to roughly predict its origin, but in individual cases texture has to be verified precisely by experiment. A high spatial resolution in orientation measurement down to the subgrain scale is required for the study of fine grain and heavily deformed materials in general, of grain boundary characterization, of recrystallization and grain growth, in the study of coating processes, epitaxy and the impact effects of high-energy particles. Individual grain orientation measurement on a grid mesh lends itself to provide a database in elaborated Finite Element calculations tackling tensorial material properties in the nodal mesh points.

A full review of the entire state of the art in Kikuchi electron diffraction would go beyond the constraints of a short article, so it is intended here to look closer at two particular aspects, one being spatial resolution and the other practical limitations of the technique imposed by grain size and foil bending.

2. Experimental setup

In BKD mode the sample surface is steeply inclined by about 60º to 70º out of the horizontal to produce a sufficient backscatter intensity [7]. The EBSD detector is placed horizontally on the side (Fig. 2a). The distance between sample and screen is typically 2 cm, roughly like the diameter of the screen, so that a wide cone of about 45º of backscattered electrons is acquired. As a consequence of strong forward scattering of energetic electrons by matter a steep lobe of intensity is generated. The maximum is close to the direction of optical reflection that a light beam would have from an opaque surface. The height of the EBSD detector is therefore set such that the intensity maximum falls on the center of the phosphor screen.

In the TEM the diffraction patterns are either acquired with a CCD camera 1 placed on-axis at the bottom of the microscope column or with a retractable CCD camera 2 above the viewing screen. A budget-friendly alternative is the acquisition of the patterns from a phosphor screen with a CCD camera 3 through a window from outside (Fig. 1) [8, 9].

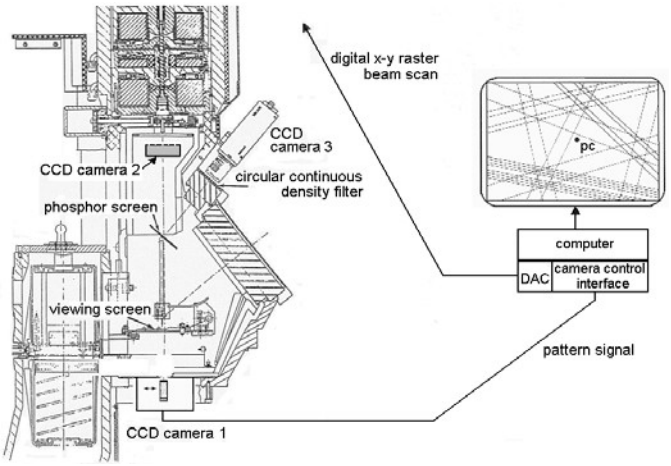

Figure 1 Projection chamber of a Philips 430 TEM with three positions of CCD cameras for the acquisition of transmission Kikuchi patterns.

A circular continuous density filter in the optical path is of advantage to equalize the steep drop of intensity from the blooming primary beam spot to the dim marginal area. Pattern intensity is of minor concern in TKD with a TEM. The patterns should cover a large angular range and be undistorted. These requirements limit TKD orientation mapping to the use of a TEM with a condenser objective lens and microbeam diffraction facility such as for instance the Philips CM series, respectively FEI TEM. For automated mapping [9] a computer controlled scan of the beam spot is implemented.

TKD in the SEM is applied to a thin foil at least parts of which have to be transparent to electrons. The foil sits horizontally in a special holder. Diffraction is either carried out with the EBSD screen positioned vertically beneath the sample so that about half the pattern cone is captured (Fig. 2b) [5]. A standard EBSD detector is placed in the same position as for BKD. An alternative setup (Fig. 2c) [10] uses a dedicated EBSD detector having the transparent phosphor screen horizontally about 1 cm beneath the sample. A mirror at 45° at the backside of the screen reflects the pattern to the CCD camera. The advantage of this setup is the acquisition of the whole cone of the Kikuchi pattern, as in the TEM equipped with a retractable CCD camera 2 on the side-entry flange. However, the intense central beam spot might overexpose the camera, as a consequence of limited dynamics, whereas the fringe of the pattern shows an intensity too low for evaluation.

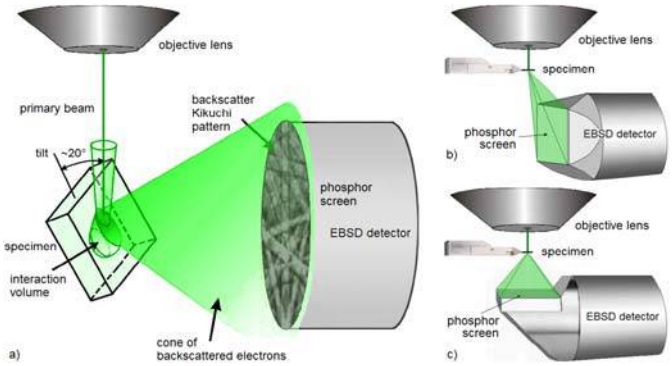

Figure 2 Kikuchi diffraction in the SEM. The cone sections of the acquired patterns are marked in green.
Fig. 2a shows the setup of BKD. In TKD the detector comes in from the side beneath the horizontal sample. Either the standard EBSD detector (Fig. 2b) is used, or (as in Fig. 2c) a dedicated detector with the transparent phosphor screen is placed concentrically to the primary beam spot beneath the sample.

3. Indexing and representation of orientation data

The geometry of a Kikuchi pattern can be interpreted as a gnomonic projection of the crystal lattice on a flat fluorescence screen. The point of impingement of the primary beam on the specimen surface is the center of projection. In particular, the angles between the center lines of Kikuchi bands correspond to interplanar angles, and the widths of Kikuchi bands correspond, according to Bragg's law, to interplanar spacings. The "stars" correspond to zone axes of the crystal. The same methods can be used for indexing transmission and backscatter Kikuchi patterns [7, 8] since their geometry is quite similar.

The geometry of a Kikuchi pattern is unique for the crystal structure and the crystal lattice orientation of the grain under the beam. It is sufficient for indexing to know the *positions and widths* of some bands in the pattern. The geometrical features of a pattern are extracted by automated pattern recognition applying a modified Hough [11] or more precisely a Radon [12] transform. Typically the positions of the 5 to 10 smallest and most intense bands of a pattern are passed to the indexing routine. Reference directions in the specimen space (e.g. specimen normal direction and transverse directions in the specimen surface) are finally indexed, and the crystallographic orientation of the grain is described either in (hkl)[uvw] notation, by three Euler angles, (φ_1, Φ, φ_2), or by the rotation matrix, g, which transform the specimen coordinate system under consideration into the crystal-fixed coordinate system. Angular resolution is limited by the precision the bands can be located in the pattern. Details of the indexing routine can be found for instance in [7]. Several commercial EBSD systems are on the market.

The spatial distributions of grain orientations are visualized by assigning orientation-specific pseudo-colors to the measured points on the scanning grid. Thus crystal orientation maps of the microstructure are constructed with the advantage of providing quantitative orientation contrast (Figure 3). In a similar way, misorientations across grain boundaries, Σ values of grain boundaries, or other microstructural characteristics and derived entities such as dislocation densities, activated glide systems, types of twins or resolved shear stress are visualized by staining the grains in the micrograph with specific colors [13]. All grain boundaries exceeding some tenth of a degree are identified. Stereological as well as orientation data, as sensitive indicators of the production process, are readily available. Automated Kikuchi diffraction and the interpretation of data in the form of crystal orientation maps are known as Orientation Microscopy (OM). It enables a major progress in the quantitative characterization of microstructure.

Figure 3 shows inverse pole figures, the crystal orientation maps and the pattern quality map of a laser weld seam between dissimilar Al sheet metals [14]. The distinct grain shapes and textures clearly mark the melting seam in the center, the heat affected zones and the original sheet metals. It is worth noting that the maps are graphical representations of the microstructure to ease imagination. They are backed by the complete information about the grain orientations in every measured point such providing means of a quantitative description of the microstructure on a grain-specific scale.

4. Spatial resolution

Kikuchi diffraction is a useful analytical tool for characterizing the arrangement of crystalline volumes. But why does spatial resolution in Kikuchi diffraction fall more than one order of magnitude behind spatial resolution in conventional SEM imaging, and still further behind when compared to the spatial resolution of a TEM?

The inherent resolution limit of Kikuchi diffraction is set primarily by the excitation volume rather than by the diameter of the beam spot at the point of impact on the sample surface. The excitation volume is the fraction of the interaction volume of the primaries with the sample from where the pattern forming electrons are diffracted and can leave the crystal with no further scattering. Shading in Fig. 4 indicates this volume fraction. The footprint of the beam spot is marked in bright.

The TEM is operated at a significantly higher accelerating voltage than the SEM. In contrast to EBSD, spatial resolution, δx, in microbeam TKD in the TEM as well as in the SEM is still approximately limited by the diameter of the beam size. The reason is that the sample has to be thinned to a thickness in the range of the mean free path of the energetic electrons so that only a small interaction volume can develop (Fig. 4) [15]. Actually some multiple scattering is tolerable since it contributes to the background. Foil thickness may so approach almost half a micrometer, depending on the accelerating voltage and density of the particular material.

Resolution in depth, δz, simply equals foil thickness. In practical work, the smallest probe size and hence spatial resolution is limited to about 10 nm with TKD, due to the decrease in image brightness as the probe becomes smaller, and due to the spread of the scattered beam on its way through the specimen foil provided the foil is thin enough for the applied accelerating voltage. If neighboring grains are studied, orientation differences down to 0.1° can be measured with TKD in the TEM and about 0.5° in the SEM.

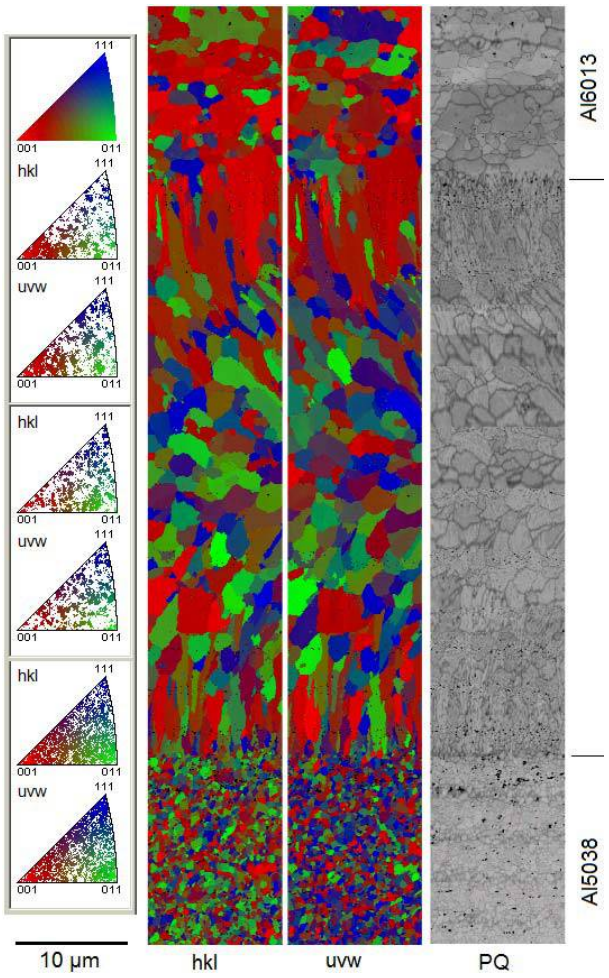

Figure 3 Inverse pole figures, the crystal orientation maps, hkl and uvw, and the pattern quality map, PQ, of a laser weld seam.

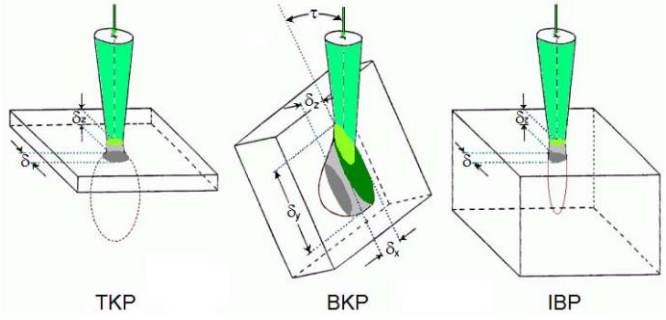

Figure 4 Excitation volume and spatial resolution in TKP, BKP and IBP.

On bulk samples a pear-like interaction volume develops beneath the surface due to strong forward scattering of energetic electrons. For the purpose of sufficient pattern intensity in BKD the sample surface is steeply tilted from normal incidence to the side the detector is looking at. Thus the sample surface cuts out a large area from the interaction volume from where back-diffracted electrons can emanate to

form the pattern. Furthermore, as a consequence of the steep sample tilt, the footprint of the beam is elongated by projection. Hence spatial resolution is reduced but intensity increased by the tilt, and a compromise at about 60º to 70º is made. Resolution along the beam direction on the sample surface, δy, is hence about three times less than δx. The escape depth, δz, is limited by the mean free depth of penetration of the back-diffracted electrons in the sampled material at the actual beam voltage.

The excitation volume increases for light materials and high beam voltages. Spatial resolution in BKD can be improved to some extent by lowering the beam voltage from typically 20 keV down to a few keV in the SEM. Intense small spots at low accelerating voltages are the domain of Field Emission (FE) SEM. Drawbacks of low accelerating voltages are the susceptibility of the beam to magnetic stray fields (hence a small working distance is mandatory which, however, may conflict with the design of current pattern acquisition systems), the low efficiency of present aluminum-coated phosphor screens and the high liability of pattern quality to preparation artifacts or foreign surface layers

Spatial resolution in BKD with copper is better than 0.05 µm at 20 kV using a tungsten filament and presently about 0.03 µm with a FE gun thanks to the higher beam current in small probes. Backscatter Kikuchi patterns have been found to disappear when a thin foreign surface layer is present. Depth resolution is assumed as being about twice the Rutherford elastic mean free path at the given beam energy. That means about 100 nm for Al, 20 nm for Ni and 10 nm for Au at 40 kV accelerating voltage and 20º angle of incidence to the surface [16]. Theoretical and experimental values of mean free path, however, relate to amorphous material, but can be significantly larger and orientation dependent in crystals, as a consequence of the channeling effect. So real escape depths in backscatter Kikuchi diffraction are supposed to be larger than these estimates or predicted by Monte Carlo simulations based on mean free path data.

Since spatial resolution is limited by the size of the excitation volume – respectively the diameter of the beam spot – rather than on the actual magnification of the SEM, a high spatial resolution can only be obtained by correct focus settings across the tilted sample surface. The accessible specimen area is then limited by the largest field of view of the SEM at the lowest magnification and largest working distance. Hence a large specimen area or distant surface regions may be studied not only by employing a mechanical translation stage or by stitching together small subsections of scanned fields, but by digital beam scan in one single sequence. Provision has to be made for a sufficiently high and ripple-free digitization of x-y scan positioning.

A further limitation of spatial and depth resolution in Kikuchi diffraction is set by the smallest grain size. To exclude overlapping Kikuchi patterns, grain size should be larger than the excitation volume. This means in particular that grain size in x and y has to be at least larger than the footprint of the beam spot, and grain size in depth beneath the surface has to be larger than the escape depth in z of the pattern forming electrons in backscatter diffraction. In TKD the extension of grains in foil normal direction should be larger than the foil is thick, which means that columnar grains are optimum. In general it is not wise to focus the primary beam spot to less than the size of the smallest grains in the sample. The adverse effect would be less intense patterns or, as an alternative, to turn up beam density and possibly accept a strong increase in contamination rate by polymerization of hydrocarbons under the beam.

While resolution within a grain is of lower significance, it becomes quite critical when the beam spot approaches a grain boundary. A smart pattern indexing software can then improve resolution in TKD and EBSD by taking account of the intensity levels of superimposed patterns, rejecting less likely orientation solutions and comparing orientations in neighboring pixels.

A promising alternative to EBSD will be crystal orientation mapping using Ion Blocking Patterns (IBP) [15]. These patterns consist of a bright even background superposed by a regular system of very sharp black lines. An IBP is similar to quasi a negative image of a BKP. A black line is formed by ions which impinge on the surface parallel with a low-indexed lattice plane, channel deep into the crystal, but cannot leave the crystal unless they are backscattered again in the channel of an angular width of less than twice the Bragg angle to the lattice plane. As a consequence of the small wavelength of ions the lines are significantly narrower than the corresponding bands in BKP. A thin foreign amorphous layer fans out the incoming beam by scattering, thus suppresses channeling into the crystal and lastly the formation of black lines. The requirements for a gentle sample preparation and a clean vacuum are very high. At present commercial ion scanning microscopes with high current densities in small probes in the nanometer range and a clean vacuum in the specimen chamber are not yet available. Spatial resolution equals the footprint of the ion probe on the surface.

5. Foil thickness and buckling set limits to TKD

Kikuchi diffraction not only puts particular requirements on the microscope hardware and the system software, but the user is also often faced with difficulties arising from sample preparation. This may be a cause of an “unknown” phase to be analyzed, or measurement conditions have to be reconsidered when pattern quality is poor. Further challenges are samples of low electrical conductivity, electron-beam sensitivity, chemical instability, heavy plastic deformation and the need for *in-situ* dynamic investigations.

Most notably are thin foils often difficult to prepare from the bulk. It is in many cases questionable whether a thin foil truly represents the structure of the interior of the material. The grains relax on both surfaces when the matrix is removed. Due to different grain orientations in a polycrystal the foil contains local residual stress. Therefore, surfaces of free-standing foils are often not really flat but show a grain-specific surface relief and bulging. This foil bending and puckering cannot be easily excluded. The result is a change in local orientations of up to several degrees with respect to the microscope-specific reference directions.

A large grain or a single crystalline foil is bent, but roughly retains its intrinsic lattice orientation relative to the local foil normal. Pattern indexing, however, is related to the microscope-specific reference directions (x, y and z) that

likewise deviate from the local grain-specific reference directions. This is visualized in the grain orientation map by color shading for low deviations in orientation. At larger surface slopes pattern indexing may jump to another solution, which is not correct for the orientation if it could have been measured in the bulk material. Such jumps in orientation are reproduced by distinct color skips in the orientation map. Irregular bending and wrinkles are readily perceived in orientation maps of coarse grain and single crystal foils at large area scans since the resulting damask pattern appears factitious.

The tiling of fine grains in a wrinkled foil, however, follows the local inclination of the foil without bending the lattice. The orientation of a thus tilted grain is rotated as a whole against the microscope-specific reference directions and hence measured with a consequential error. The superposition of the surface topography and the rotated orientations of the individual grains results in a distinct color, presumable without shading, for each grain in the orientation map. If texture is not extremely sharp the map looks like a map composed of a usual fine grain microstructure. Foil wrinkling is in the end disguised.

The slopes of foil bending are in the range of several degrees. As a consequence measured local orientations scatter by an error of this range about the orientations one would obtain if the sample were flat. The ODF and pole figures are slurred by widening sharp peaks of orientation densities by several degrees as well. Ideal thin samples with bulging inclinations below the angular resolution of a Kikuchi system are very hard to realize and require special preparation techniques [17]. This is true in particular for transmission studies in the SEM where foil thickness has to be much smaller, to be transparent at the lower accelerating voltage, than in the TEM.

Foil buckling is nicely displayed in “bend contours”, also named “extinction contours”, in TEM micrographs. They become visible preferably at lower magnifications when a small selector aperture is used. They are dark in brightfield images, respectively light in darkfield images, at crystal regions where the lattice planes are in Bragg position. Contours of equal intensity represent lines of equal inclination to the electron beam. The angular deviation between contour lines is thus a multiple of the Bragg angle, see TEM textbooks for details, e.g. Edington [18].

6. Conclusion

Preparation of freestanding, flat and transparent thin samples for TKD in the TEM and SEM is often tedious. It is a difficult task in particular when the sample is non-noble, heavily deformed, tends to reactions and etch pitting or contains several phases.

Buckled foils are in many cases not a real problem for the study of polycrystals or single crystals with the TEM. The metallurgist uses a TEM typically to investigate very small sample areas, e.g. for dislocation or phase analysis, special grain boundaries, lattice misfits, high-resolution lattice imaging and Burgers vector analysis [19]. The operator can thus select a small area with no bend contours and other flaws in the images to produce trustworthy crystal orientation maps.

With TKD in the SEM we usually want to study large sample areas containing many grains. Then bent foils may be a problem since the orientation data are spoiled by the overlay of geometrical wrinkling. With fine grain material the false orientations are not obviously apparent by deviating colors or shades in the crystal orientation maps. However, pole figures and ODF are unduly smeared up by topography. Interpretation of texture may be misleading. Misorientation data across abutting grain boundaries are not seriously affected by foil buckling. EBSD in the SEM is carried out on bulk samples. The main requirements are a flat and clean surface free from preparation artifacts such as undue deformation, reaction layers or phase-selective etch.

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