A profile-based method of determining intragranular strains using Kossel diffraction patterns

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Abstract.
Kossel microdiffraction is one of a few experimental methods of investigating heterogeneities of elastic stresses within crystallites. With digitally recorded back-reflection Kossel patterns, one can determine absolute lattice parameters, and hence lattice strains and stresses, based on geometry of Kossel lines, but the strain resolution of this approach is limited by finite widths of the lines. A new method is proposed which considerably improves the resolution in cases when the patterns originate from areas with similar lattice orientations. The method is based on determination of differences between pattern geometries: lattice strains are calculated from mutual shifts of intensity profiles of Kossel lines. The strain accuracy of this profile-based approach was estimated. It is demonstrated that the limit of strain resolution reaches a few parts per hundred thousand, i.e., it is nearly one order of magnitude better than that of the conventional Kossel-based lattice parameter refinement. This improvement concerns the critical range of lattice strain, and it constitutes a qualitative leap in resolution. The paper describes main aspects of the new approach and strain resolution tests.

Introduction

There are a number of strain determination methods, but only some of them are adequate for investigating local stresses. For typical microstructures, such stresses can be determined only if a sufficient spatial resolution is achieved. In analysis of individual crystallites, an important role is played by anisotropy. Therefore, all components of the strain tensor need to be determined. Moreover, the accuracies of strain determination methods are of critical importance; in most applications, strain variations of about one part per ten thousand (1×10⁻⁴) are of interest, and consequently, only methods of at least this accuracy are of use. Last not least, a strain determination procedure should be easy to use or even automatic, as the automation of data collection and processing is important for its applicability.

The divergent X-ray beam technique is known to be suitable for investigating crystal lattices [1]. In particular, the Kossel technique with the source of divergent X-ray beam under the surface of the examined material [2] has been used for determining strains in individual crystals [3,4]. (The original Kossel technique needs to be distinguished from the pseudo-Kossel approach [5] with X-rays excited in an external source near the surface of the investigated material.) A Kossel diffraction pattern is built of a background of ‘Bremsstrahlung’ and traces of characteristic radiation. The traces have low signal-to-background ratio and shapes of conic sections. It needs to be mentioned that not all crystalline structures can be investigated with the Kossel technique (as some interplanar spacings must exceed half-wavelength of X-rays emitted by a strongly scattering element of the crystal, and the excitation energy must be sufficient to excite Kα lines), but the window of applicability covers many materials.

With Kossel patterns recorded on photographic films, any automation of data processing was a challenge. Modern-day variants of the Kossel technique, in particular, the laboratory-based microdiffraction of divergent X-rays brought about by electron beam in a scanning microscope and fluorescent radiation excited by microfocused synchrotron beam, make wide-ranging automation feasible. In both cases diffraction patterns are recorded using phosphorous screens and CCD digital...
cameras\textsuperscript{1} [6,7]. This allows for computer processing and analysis of the patterns. As the spatial resolution of these two methods is estimated to be a few micrometers, they are suitable for determination of second and third order residual stresses in a large class of polycrystals.

With the previous approach [8], local strains are calculated by relating sets of absolute lattice parameters obtained from Kossel patterns recorded at areas of interest. This allows not only for getting strains within one crystallite but also for comparing strains in crystallites of different orientations. Its strain resolution is directly linked to the resolution of the refinement of the parameters. The parameters are refined by minimization of differences between positions of corresponding diffraction lines in experimental and simulated patterns using the geometric theory of diffraction under the assumption of point-like radiation source. Actual Kossel conics are not geometrical lines but have finite widths. In result, the accuracy in determination of absolute lattice parameters is limited [8]. With the experimental set-up described in [9], the strain resolution of a few parts per ten thousand is achieved if a sufficient number of Kossel lines are used.

It is shown below that in measurements of intragranular strains the resolution can be considerably improved. Having diffraction patterns originating from distinct areas of similar lattice orientations, one can use small differences between roughly similar patterns of Kossel lines to calculate the strain at a given specimen area with respect to a reference area. Various approaches to matching the patterns can be envisioned. Here, a simple method based on fitting intensity profiles along directions perpendicular to Kossel lines is examined. We are not aware of any previous works on using Kossel line profiles for precise determination of all components of the strain tensor.

\textbf{The method}

Briefly, in the proposed method, line profiles collected from a reference pattern (originating from an area which is assigned zero strain) are used to determine positions of corresponding lines in other patterns originating from strained areas of the same grain, and then – based on shifts of line positions – the strain tensor is calculated. The determined strains are relative to unknown strain state at the reference point.

Particular characteristics of diffraction patterns depend on the pattern generation and recording methods. Typical Kossel lines in CCD-recorded back-reflection Kossel patterns have widths in the range of 20-60\times10^{-4}\text{ rad} and various intensity profiles; some lines are bright, some are dark, but most have ‘Helldunkelstruktur’ of some degree (Fig. 1). In the classic (kinematic) description, the bright-dark structure of Kossel lines arises when deficiency (absorption) lines adhere to excess (reflection) lines; as the natural width of the X-ray emission lines is negligibly small, the structure is caused mainly by the finite size of the radiation ‘source’. A more complete understanding of the complex widths.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{kossel_lines.png}
\caption{Typical profiles of Kossel lines averaged over 20px long segments. They originate from a Kossel pattern of ferrite. The first profile consists of individual peaks of the K\textsubscript{α} doublet, whereas the second illustrates the bright-dark line structure. The units on the abscissa are pixels.}
\end{figure}

\textsuperscript{1} Although the resolution of photographic films is still better than that of CCD detectors, there is no return to the photographic technique because of tedious chemical processing and the need to digitize the patterns.
line profiles is developed by using dynamic theory of diffraction. For the discussion of the mechanisms leading to these profiles see [10,11] and references therein. The profiles of $K_{\alpha 1}$ lines are additionally affected by the proximity of the $K_{\alpha 2}$ lines. Individual peaks of the $K_{\alpha}$ doublet are discernible only for some lines.

Fig. 2. Schematic of the profile-based strain determination method. Patterns collected from different locations in the same crystallite are similar. Shifts of profiles of Kossel lines at corresponding points on the patterns are used to determine differences in geometry of the lines. Hence, one calculates the strain tensor (plus misorientation, pattern center and sample-to-detector distance).

To describe the profile-based strain determination method let us assume that a number of diffraction patterns collected from different locations in the same grain are available, and one of them is chosen as a reference pattern. The procedure includes the following steps: First, the reference pattern is processed conventionally, i.e., lattice parameters are refined [9, 12]. Then, profiles of the Kossel lines at suitably marked locations in the reference pattern are collected. To smooth the profiles, they are averaged over short segments of Kossel lines. Subsequently, each profile in the reference pattern is compared to profiles at corresponding locations in the remaining patterns. In general, these profiles are similar but mutually shifted, and the shift magnitude is obtained by matching the profiles (Fig. 2). Assuming that the shift was caused by strain, small orientation change, and the change of pattern center and pattern-to-detector distance, one finally calculates the components of the strain tensor. The calculation method is analogous to that used in lattice parameter refinement. The difference between these two approaches is that in the new one, the locations of markers on the Kossel lines are not chosen arbitrarily, but they are obtained by matching line profiles.

The profile-based approach gives the distortion of the strained lattice with respect to the reference lattice. Although the accuracy of the reference lattice parameters is relatively low, thanks to accurate shifts obtained by profile matching, the distortion is determined accurately. Thus, limiting the strain determination procedure to patterns of similar orientation allows for achieving...
high accuracy within the simple geometric description of diffraction without complicated dynamic simulation of line intensities.

Examination of the profile-based approach required dedicated software. An existing program (KSL\textit{Strain}) for Kossel-based refinement of lattice parameters was modified. The previous version (initially equipped with routines for manipulating markers of diffraction lines, a pattern indexing procedure, and procedures for lattice parameter refinement) was augmented with additional subroutines for detection of line profiles, checking suitability of the profiles, passing the profiles between different patterns, and for matching the corresponding profiles. The profiles can be aligned manually (using a computer mouse) or automatically via maximization of the profile correlation coefficient (Fig. 3). In the current version of the software, the resolution of individual profile matching is 1/10 of a pixel.

![Fig. 3. Matching of Kossel line profiles of ferrite with KSL\textit{Strain}. The blue (partly dotted) profile comes from the reference pattern whereas the other profile was taken from the same reflection on a different pattern. The numbers below the profiles represent profile correlation coefficients. Ticks at the top of the figures mark individual pixels. As in Fig. 1, the total length of the profiles is 20px.](image)

With known shifts of individual line profiles, the association of the complete patterns of conic lines is carried out using \textit{KLEBS} [13]. Here, the \textit{KLEBS} procedure minimizes deviations between theoretical lines which would arise by straining the lattice obtained from the reference pattern and the actual markers on the pattern from the strained area. The objective function is based directly on the formal equation of the Kossel lines. Besides the strain tensor components, also the deviation from the reference orientation, the coordinates of the pattern centre and the sample-to-detector distance are parameters of the function. The computation of the strain tensor is carried out in Cartesian coordinate system linked to the crystal structure. Demo version of KSL\textit{Strain} is available at [14].

Differently than in the strain determination method based on lattice parameter refinement [9], in profile-based approach, lines with arbitrary (dark, bright or bright-dark) profile character can be used. Typically, 7 to 15 profiles per Kossel line are collected. This leads to 50-150 profiles per pattern. It must be taken into account that with automatically selected marking points, some profiles turn out to be not suitable (e.g., those at low angle intersections of Kossel lines) and they must be discarded; this issue can be easily resolved simply by increasing the number of collected profiles. When the difference in orientations of the reference and strained areas is large, some reference line markers arrive outside the pattern from strained area, and - consequently - some profiles are lost. One can partly alleviate this problem by using a small sample-to-detector distance; the smaller the distance, the larger the allowed orientation difference.

It is worth noting that if orientation differences between reference and strained areas are small, potential optical distortions of the pattern acquisition system will be roughly the same for all patterns. Consequently, they are expected to have much smaller impact than in the method based on lattice parameter refinement.
Strain resolution

Preliminary numerical tests were carried out to check the reliability of the new strain determination procedure and to get limits of its applicability. Real results cannot be more reliable than these limits. In the tests, besides all components of the strain tensor, also the misorientation, the location of the pattern center and the sample-to-detector distance were fitted. As in other diffraction experiments, strain resolution depends on the quality of the patterns and on the presence of conics corresponding to reciprocal lattice vectors of large magnitude (large Bragg angles).

Basic tests were performed on simulated patterns with the sought strain known beforehand. Diffraction patterns were generated numerically for assumed crystal structures and crystal orientations. Pattern size (~1Mpx), pixel size (27µm), sample-to-detector distance (30-40mm) and location of the pattern center were as in experimental set-up of [9]. In the artificial case of binary patterns with the value of 1 on one-pixel wide Kossel lines and 0 otherwise (and kinematical intensities used to decide about presence or absence of particular reflections), the strain resolution is limited only by the precision in marking the lines. Under such optimal conditions, in the presence of reflections with large Bragg angles, the accuracies of strain tensor components obtained by the profile-based approach were about 0.1×10⁻⁴. This is roughly 1/10 of the minimal angular step used for matching individual profiles (0.1px≈1×10⁻⁴rad). Broadening of lines in the reference pattern or the other patterns had little impact on the strain resolution.

More realistic tests were carried out on deliberately modified experimental patterns. A given pattern was enlarged and shrunk to test the impact of changing sample-to-detector distance. The pattern was also cropped in various ways to test the impact of changing pattern center. (These modifications led to a small change of the pattern size, which was neglected.) To give a concrete example, for such modified patterns of ferrite the deviations of the computation results from the true strain components did not exceed 1×10⁻⁴, and the mean of absolute values of the deviations was below 0.2×10⁻⁴. Also experimental patterns with artificial noise were investigated. The noise made the lines more diffuse. The deviations of the computation results from the true strain were small up to a certain level of the noise, and then the discrepancy grew rapidly.

The proposed method is based on the assumption of similarity of the investigated and reference patterns. The differences are expected to grow with growing strain and with growing distances between the investigated areas. Both may be linked with a considerable orientation change and a change of line-width. These differences in diffraction patterns and resulting dissimilarities in profiles of lines will affect the strain accuracy. Therefore, before applying the new method and software to real strain determination problems, one needs to carry out additional tests checking the reproducibility of results from experimental patterns.

Conclusions

A new relatively accurate approach to strain measurement using digitally recorded back-reflection Kossel diffraction patterns is proposed. With Kossel patterns corresponding to similar lattice orientations, lattice strains are obtained by matching intensity profiles of Kossel lines. The new approach is applicable to determination of intragranular strains. It can also be used for investigation of the mechanisms of (poly)crystal deformation by in situ experiments with Kossel patterns collected from a single location in a crystal(lite) during straining. Numerical tests indicate that, in favorable experimental conditions, strains can be measured with the accuracy of about 1×10⁻⁴. This is nearly one order of magnitude better than the resolution of strain obtained by refinement of lattice parameters. The improvement concerns the range of strains critical from the viewpoint of practical applications.

The proposed method is also a step toward automation of intragranular strain determination. The best solution would be a fully automatic mapping of local elastic strains. Such a capability can only be considered if ‘manual’ approaches with complete control over all steps give reliable results.
Results of tests of the particular profile-based approach show the potential of a more general class of methods based on matching fine details of Kossel patterns for improving resolution of strain determination. Such improvements will advance research on residual stresses and create new opportunities for exploration of material properties.

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References

[1] W. Linnik, Über eine Abänderung der Drehkristallmethode zur Untersuchung der Kristallstruktur mit Röntgenstrahlen, Z. Phys. 61 (1930) 220 – 226.
[2] W. Kossel, V. Loeck, H. Voges, Die Richtungsverteilung der in einem Kristall entstandenen charakteristischen Röntgenstrahlung, Z. Physik 94 (1935) 139 – 144.
[3] R. Tixier, C. Waché, Kossel patterns, J. Appl. Cryst. 3 (1970) 466 – 485.
[4] V.V. Lider, X-ray divergent-beam (Kossel) technique: A review, Crystallogr. Rep. 56 (2011). 169 – 189.
[5] K. Lonsdale, Divergent-beam X-ray photography of crystals. Philos. Trans. R. Soc. London A240 (1947) 219 – 250.
[6] H.J. Ullrich, J. Brechbühl, J. Bauch, H. Lin, Detektion von Kossel-Linien mittels marCCD-Detektor, In: HASYLAB annual report (1998) 887 – 888.
[7] S. Däbritz, E. Langer and W. Hauffe, New observation method for divergent beam X-ray diffraction patterns, J. Anal. At. Spectrom. 14 (1999) 409 – 412.
[8] E. Langer, S. Däbritz, C. Schurig, W. Hauffe, Lattice constant determination from Kossel patterns observed by CCD camera, Appl. Surf. Sci. 179 (2001) 45 – 48.
[9] D. Bouscaud, A. Morawiec, R.Pesci, S.Berveiller, E.Patoor, Strain resolution of SEM-based Kossel microdiffraction, submitted.
[10] D. Stephan and V. Geist, Theoretische Intensitätsprofile von Kossel-Linien und Vergleich mit experimentellen Linienprofilen ({111} PKα-Interferenzen von GaP-Einkristallen bei Protonenstossanregung), Exp. Techn. Phys. 34 (1986) 153 – 167.
[11] G. Nolze and V. Geist, On the intensity of Kossel reflections, Cryst. Res. Technol. 27 (1992) 421 – 430.
[12] A. Morawiec, R. Pesci, J.S. Lecomte, Semiautomatic determination of orientations and elastic strain from Kossel micro-diffraction, Ceram. Trans. 201 (2008) 163 – 169.
[13] A. Morawiec, A program for refinement of lattice parameters based on multiple convergent-beam electron diffraction patterns, J. Appl. Cryst. 40 (2007) 618 – 622.
[14] http://personal.imim.pl/adam.morawiec/A_Morawiec_Web_Page/downloads.html