MEASUREMENTS OF RESIDUAL STRESSES IN MICRON REGIONS BY USING SYNCHROTRON EXCITED KOSSEL DIFFRACTION

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1 INTRODUCTION

Most of the common X-ray diffraction methods, e.g. the sin^2\(\Psi\) - method [1], for obtaining residual stresses from crystalline materials only provide integrated information about several square millimeters of the surface. Local deviations from the measured mean value of the stress component, especially within a single grain, cannot be identified (excepting extremely coarse-grained materials). It is well-known that the really acting local residual stresses are associated with local inhomogeneities (dislocations, inclusions / dispersoids, lattice vacancies, etc.) and with further material defects (extrusions, intrusions, microcracks, etc.). Hence, the knowledge of such local stresses additionally to other features of the real structure is supposed to be of great interest. In the present paper a new method to measure and determine the residual stresses in micron regions will be discussed in detail. Referring to a physical effect discovered by W. KOSSEL in 1934 [2] the high lateral resolution had been achieved. A fine focused electron beam incident on a crystal causes fluorescent radiation (characteristic X-rays) which may be diffracted in the target crystal itself. Both conductive and nonconductive materials can be investigated when the fluorescent radiation is excited by an external X-ray source. However, this interesting kind of the excitation of KOSSEL interferences, discovered by BORRMANN [3], has not been of great practical importance over many years. But nowadays the situation is completely different. When using synchrotron radiation for the excitation of KOSSEL diffraction lines, as shown by H.-J. ULLRICH and co-workers in 1992 for the first time [4], a further scaling down of the analyzed specimen area (in the range of \(\mu\text{m}^2\)) is not in conflict with an acceptable exposure time (a few seconds or minutes). More detailed information about the synchrotron excited KOSSEL technique and its application fields (overview) is presented by BAUCH et al. in the proceedings of the 47th Annual Denver X-ray Conference in 1998 (also see [5], [6], [7]).
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In the following chapter, a new method for the determination of the residual stresses from KOSSEL patterns is presented. Since all the X-ray measurements of residual stresses are based on the measurement of elastic strains in crystalline regions at first the interplanar spacings (d_{hkl} - spacings) have to be determined accurately. As a widespread range of procedures for obtaining lattice parameters from the KOSSEL lines already exists, this task seemed to be solvable.

![Diagram of KOSSEL cones and gnomonic projection center](image)

**Fig. 1:** Illustration of the spatial location of two KOSSEL cones relating to the gnomonic projection center (schematically)

However, the evaluation of individual d-spacings is much more complicated than the evaluation of lattice parameters. To reach a resolution of strains in the range of $10^{-6}$ to $10^{-5}$ a new method
was developed which refers to the high precision compensation method of KOSSEL. By modifying
the classical variant basically now for any crystal orientation individual d-spacings of different net
planes (instead of an averaged lattice parameter!) can be obtained from the KOSSEL lines. It is
worth mentioning that the original compensation method only allows to determine the lattice
parameter if a crystallographic reference pole nearby the gnomonic projection center (of the
KOSSEL pattern) is known (for special crystal orientations!). Now we are able to give up these
restricting conditions by calculating the coordinates \([x \ y \ z]_k\) of the gnomonic projection center
(see Fig. 1) relating to the crystal system. (The geometric location of the gnomonic projection
center on the detector [film] can be determined.)

Since the vector \([x \ y \ z]_k\) is the condition for evaluating d-spacings, strains and stresses as well as
it must reflect any deviation from the crystal symmetry by the acting stresses for the computation
an iteration procedure is advisable. Before presenting the iteration procedure, some basic
equations have to be established. The given theory can be easily transformed from the cubic
crystal symmetry group to another one when substituting the marked (*) equations.

\[
\cos \beta_i = \frac{h_i \cdot x + k_i \cdot y + l_i \cdot z}{\sqrt{(h_i^2 + k_i^2 + l_i^2) \cdot (x^2 + y^2 + z^2)}} \quad \Rightarrow \quad \beta_i \quad (*)
\]

At first, we assume that the crystal coordinates of the gnomonic projection center are known.
Furthermore, we set \([x \ y \ z]_k\) as a unit vector:

\[
x^2 + y^2 + z^2 := 1
\]

The quantities \(r_i\) and \(R\) have to be measured precisely to get \(\alpha_i\), whereby for the exact value of \(r_i\)
(distance between the projection center and the \(i^{th}\) KOSSEL line) a line profile analysis is
necessary. When using the iteration procedure a further KOSSEL cone is sufficient to eliminate the
distance \(R\) between the specimen and the film.

\[
\tan \alpha_i = \frac{r_i}{R} \quad \Rightarrow \quad \alpha_i
\]

The Bragg angle (diffraction angle) \(\Theta_i\) can be calculated as:

\[
\alpha_i + (90^\circ - \Theta_i) = \beta_i \quad \Rightarrow \quad \Theta_i = \alpha_i + 90^\circ - \beta_i
\]
and the net plane spacing $d_i$ is given by the BRAGG's law:

$$\lambda = 2d_i \cdot \sin \Theta_i \quad \Rightarrow \quad d_i = \frac{\lambda}{2 \cdot \sin \Theta_i} \quad (5)$$

When the lattice parameter $a$ is of interest, it can be obtained from:

$$a = \sqrt{h_i^2 + k_i^2 + l_i^2} \cdot d_i \quad (*) \quad (6)$$

The elastic strain $\varepsilon_i$ of the considered net plane group is given by:

$$\varepsilon_i := \frac{\Delta d_i}{d_0} = \frac{d_i - d_0}{d_0} = -\cot \Theta_0 \cdot (\Theta_i - \Theta_0) \quad (7)$$

($d_0$ ... net plane spacing of the stress-free / strain-free state
$\Theta_0$ ... BRAGG angle of the stress-free / strain-free state)

According to equation (7) which is the differentiated form of the BRAGG's equation (for a constant wavelength $\lambda$) any change of the net plane spacing (elastic strain) causes a measurable shift in the diffraction angle.

![Diagram showing net plane normal](image)

**Fig. 2:** Definition of the orientation angles $\Psi$ and $\phi$ of the net plane normal relating to an orthogonal coordinate system

(axis of the KOSSEL cone = net plane normal = measuring direction of the strain)

Since the value and the sign of the strain depend on both, the stresses and the orientation of the net plane, orientation angles ($\Psi$...tilt angle; $\phi$...azimuth angle) have been introduced (see Fig. 2).
Hence, we can set: \( \varepsilon_i = \varepsilon_{\psi \phi} \).

Now the basic equation for the X-ray analysis of residual stresses can be written as:

\[
\varepsilon_{\psi \phi} = \left( \frac{1}{2} s_2 \cdot \sin^2 \psi \cdot \cos^2 \phi + s_1 \right) \cdot \sigma_{11} + \left( \frac{1}{2} s_2 \cdot \sin \psi \cdot \sin^2 \phi + s_1 \right) \cdot \sigma_{22} + \right.
\]
\[
\left. + \left( \frac{1}{2} s_2 \cdot \cos^2 \psi + s_1 \right) \cdot \sigma_{33} + \left( \frac{1}{2} s_2 \cdot \sin \psi \cdot \sin \phi \right) \cdot \sigma_{12} + \right.
\]
\[
\left. + \left( \frac{1}{2} s_2 \cdot \sin \psi \cdot \cos \phi \right) \cdot \sigma_{13} + \left( \frac{1}{2} s_2 \cdot \sin \psi \cdot \sin \phi \right) \cdot \sigma_{23} \right) \quad (8)
\]

\( \frac{1}{2} s_2 \) and \( s_1 \): elastic constants of the considered \( \{h k l\} \) net plane group

When measuring the strains \( \varepsilon_{\psi \phi} \) of 6 independent (hkl) net planes of the crystal, the entire stress tensor can be determined by the solution of a system of 6 equations like equation (8).

\[
\begin{bmatrix}
\sigma_{11} & \sigma_{12} & \sigma_{13} \\
\sigma_{12} & \sigma_{22} & \sigma_{23} \\
\sigma_{13} & \sigma_{23} & \sigma_{33}
\end{bmatrix}
\Rightarrow
\end{bmatrix}
\]

\[
\begin{bmatrix}
\sigma_{11} & \sigma_{12} & \sigma_{13} \\
\sigma_{12} & \sigma_{22} & \sigma_{23} \\
\sigma_{13} & \sigma_{23} & \sigma_{33}
\end{bmatrix}
\]

\( (\sigma_{ij} \ldots \text{components of the stress tensor relating to a coordinate system} \)

with normal stresses \( [i = j] \) and shear stresses \( [i \neq j] \)

The transformation of the stress tensor \( [\sigma_{ij}] \) from the specimen coordinate system (s) into the crystal coordinate system (c) and vice versa is given by

\[
[\sigma_{ij}]_c = [G]_{cs} \cdot [\sigma_{ij}]_s \cdot [G]_{sc} \quad [\sigma_{ij}]_s = [G]_{sc} \cdot [\sigma_{ij}]_c \cdot [G]_{cs} \quad (10)
\]

\( ([G]_{cs} \text{ and } [G]_{sc} \ldots \text{orientation / rotation matrices with } [G]_{cs} = ([G]_{sc})^{-1}) \)

The basic equation for the X-ray stress analysis is generally valid in the presented form (8). Differences between the diffraction measurements of single crystals and polycrystals should be emphasized. Using a fine-grained polycrystal the orientation angles \( (\psi, \phi) \) can be set arbitrarily because the net planes of at least one grain are always in a reflecting position (compare with the \( \sin^2 \psi \) - method [1]). By contrast in the case of a single grain the angles \( \psi \) and \( \phi \) are only
predetermined by the crystallography. In Figure 3, the principle of the iteration procedure for evaluating the coordinates of the gnomonic projection center is presented. The condition for interrupting the procedure (convergence criterion) is that the calculated components of the stress (or strain) tensor result in minimized errors. Because by the use of (m+2) diffraction lines theoretically an unequivocal solution is enabled these errors express the deviation of the computed gnomonic projection axis from the real solution at the current iteration step (stress tensor with m components). Thus, there should be a greater number of diffraction lines than (m+2) in order to get information about the random errors.

When the total number of lines is n (n > m+2) there exist \( \binom{n}{m+2} \) combinations which can be used for the iteration procedure. Each of it yields both, the coordinates of the gnomonic projection center and the stress tensor. The deviations from the averaged results of all the combinations represent the random errors concerning the reliability of the measured quantities. It is recommended to set m = 6 (complete stress tensor) only in the case of a great number of evaluable KOSSEL lines (n > 9). Otherwise, proceeding from the assumption of a biaxial stress state (m = 3) relating to the specimen's coordinate system would be better. In other terms that means \( \sigma_{13} = \sigma_{23} = \sigma_{33} := 0 \) (z-axis perpendicular to the specimen's surface). It is well-known that X-rays because of their small penetration depth approximately provide information about the biaxial stress state of the surface. In this case, the exact value of the strain-free lattice parameter is not necessary for the calculations, too. However, it should always be proved carefully whether the influence of the third stress components (especially shear stresses!) can or cannot be neglected.

3 EXPERIMENTAL RESULTS

By example of measurements in a coarse-grained copper-manganese specimen (4 percent of Mn by weight) inhomogeneities of the stress state within one grain (residual stresses of third order) could be proved after cyclic deformation (push-pull-test: plastic strain amplitude = 2x10^-6, number of cycles = 5000). There exists a pronounced difference between the center and the grain boundary of the crystal which can be detected even with the naked eye by comparing the diffraction images.
1. input quantities:

- \( \lambda \) wavelength of the characteristic X-rays
- \( a_0 / d_0 \) strain-free lattice constant / net plane spacings
- \( R \) distance: specimen surface - film
- \( r_i \) distance: \( i^{th} \) Kossel line - projection center
- \( s \) elastic constants of the net plane groups

2. starting values for the coordinates of the gnomonic projection center (in the crystal system):

- \( x_k \)
- \( y_k \)
- \( z_k \)

3. calculation of the Bragg angles, net plane spacings and strains:

- \( \cos \beta_i = \frac{h_i \cdot x_k + k_i \cdot y_k + l_i \cdot z_k}{\sqrt{(h_i^2 + k_i^2 + l_i^2) \cdot (x_k^2 + y_k^2 + z_k^2)}} \rightarrow \beta_i \) (cubic lattice)

- \( \tan \alpha_i = \frac{r_i}{R} \Rightarrow \alpha_i \)

- \( \Theta_i = \alpha_i + 90^\circ - \beta_i \)

- \( \lambda = 2 \cdot d_i \cdot \sin \Theta_i \Rightarrow d_i = \frac{\lambda}{2 \cdot \sin \Theta_i} \)

- \( \varepsilon_i := \frac{\Delta d_i}{d_0} = \frac{d_i - d_0}{d_0} = -\cot \Theta_0 \cdot (\Theta_i - \Theta_0) \)

4. evaluation of the stress tensor (m independent components) from a system of m equations:

- \((m+2)\) lines are needed; \( m \) \( \sigma_{ij} \)-combinations

- stress tensor: \( [\sigma_{ij}] \)

- errors: \( [\Delta\sigma_{ij}] \)

5. iteration step (Newton or gradient procedure) until \( [\Delta\sigma_{ij}] \rightarrow \) minimum:

- \( x_{k+1} := x_k + \Delta x_k \)
- \( y_{k+1} := y_k + \Delta y_k \)
- \( z_{k+1} := z_k + \Delta z_k \)

- normalizing condition: \( (x_{k+1})^2 + (y_{k+1})^2 + (z_{k+1})^2 := 1 \)

**Fig. 3:** The principle of the iteration procedure for evaluating the precise coordinates of the gnomonic projection center (in the crystal system)
(Figs. 4 and 5). The sharp KOSSEL lines with the partially visible bright-dark structures (perpendicular to a line) and the small round LAUE spots are indicators for a low dislocation density and thus for negligible residual stresses in the center of the grain (Fig. 4). By contrast the situation changes nearby the grain boundary. Although the investigated grain is still the same, the KOSSEL lines become blurred and the LAUE spots turn their shapes from circles into rods (Fig. 5). The blurred KOSSEL lines contain information about the residual strains / stresses. Due to the calculated stress tensor it is obvious that the residual stresses nearby the grain boundary are not at all negligible.

The stress tensor corresponding to Fig. 5 is given as follows:

$$\begin{pmatrix}
-18 & -13 & 0 \\
-13 & -25 & 0 \\
0 & 0 & 0
\end{pmatrix} \pm \begin{pmatrix}
8 & 2 & - \\
2 & 9 & - \\
- & - & -
\end{pmatrix} \text{ MPa}
$$

(coordinate system of the specimen, biaxial stress state [assumption])

The small absolute errors of the stress components demonstrate the high precision of the stress analysis by using the KOSSEL technique. Furthermore, it should be mentioned that the spot diameter of the synchrotron radiation beam amounted to 50 μm which enabled the innergrain measurements in the present case. Investigations with higher lateral resolutions of 30 μm or better are already intended, e. g. by using a glass capillary for beam focusing.

In the submitted paper, a fast method (a few seconds / minutes) for the measurement of residual stresses in micron regions by using the KOSSEL diffraction has been represented. The measuring time as well as the measuring errors could be minimized extremely because of the simultaneous registration of differently orientated net planes in one KOSSEL pattern. Therefore, in comparison to the conventional X-ray measurements of stresses by diffractometers, not any movements of sample and detector are required when using the KOSSEL technique. A new procedure using iteration steps to calculate the residual stresses from the diffraction lines has been developed and applied successfully. The described results impressively demonstrate the necessity as well as the efficiency of microdiffraction methods like the KOSSEL technique allowing residual stress measurements with high local resolution.
Fig. 4:
KOSSEL diffraction image (section) of CuMn4 after slight plastic deformation, center of the investigated grain

Fig. 5:
KOSSEL diffraction image (section) of CuMn4 after slight plastic deformation, region near the boundary of the investigated grain


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