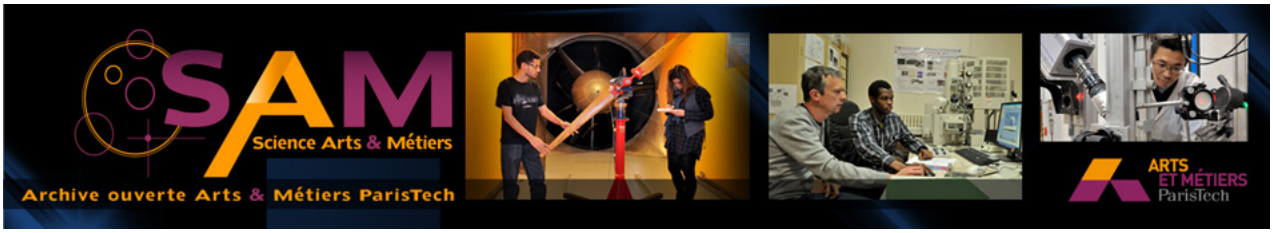

Stress analysis by Kossel microdiffraction on a nickel-based single crystal superalloy during an *in situ* tensile test – Comparison with classical X-Ray diffraction

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Abstract.
A Kossel microdiffraction experimental set up is under development inside a Scanning Electron Microscope (SEM) in order to determine the crystallographic orientation as well as the inter- and intragranular strains and stresses. An area of about one cubic micrometer can be analysed using the microscope probe, which enables to study different kinds of elements such as a grain boundary, a crack, a microelectronic component, etc. The diffraction pattern is recorded by a high resolution Charge-Coupled Device (CCD) camera. The crystallographic orientation, the lattice parameters and the elastic strain tensor of the probed area are deduced from the pattern indexation using a homemade software. The purpose of this paper is to report some results achieved up to now to estimate the reliability of the Kossel microdiffraction technique.

Introduction

Due to the growing complexity of new materials and their applications, it is increasingly necessary to know the strain and the stress state at a lower scale, a micrometer one, particularly in the case of micro- and nanosystems. Kossel microdiffraction is a tool that has been adapted for use in the Scanning Electron Microscopes (SEM), which enables to determine not only the crystallographic orientation, but also the inter- and intragranular strain and stress state while observing the microstructure and its possible evolution during *in situ* mechanical tests.
Different techniques are available to obtain residual stresses at the micrometer scale: for instance micro-Raman spectroscopy [1], X-Ray microdiffraction with a white synchrotron beam [2], convergent beam electron diffraction inside a transmission electron microscope [3]. Kossel microdiffraction is currently being developed because of some advantages comparing to these techniques [4]. Indeed, the use inside a SEM is easy, non destructive and can be applied to metallic components. Moreover, spatial and strain resolutions are competitive.
In the 1970s, Kossel microdiffraction was used for the determination of crystallographic orientations and lattice parameters [5,6,7]. Patterns were recorded on photographic films. In the 1990s, the development of high resolution CCD cameras began allowing easier and faster data collection. Lattice strain measurements finally began to be feasible [8].
The aim of this study is to check the reliability of the Kossel technique. Kossel line patterns on single crystals under loading have been recorded and the goal is to compare the stresses given by the technique with the stresses applied to the samples. We also compare results with those obtained by classical X-Ray Diffraction (XRD).

Kossel technique

In a SEM, when an electron beam is focused on a material, the latter is excited and thus an X-rays spherical wave is emitted. A part of these rays is diffracted by the crystallographic planes forming the so-called Kossel cones, in accordance with the Bragg's law: $\lambda = 2d \sin \theta$ (λ: wavelength of the

excited element, d: interreticular length, θ: Bragg's angle). A CCD camera intercepts a part of these cones, which are all emitted at the same time (one diffraction cone for each (hkl) diffracting plane). Kossel conics (lines or complete circles) are calculated from the orientation of the cones and the distance between the sample and the screen (Fig. 1).
It is possible to excite the material with different types of radiation. Conventionally, an electron beam is used to excite either the sample directly or a target placed close to the sample [9]. This latter technique is called pseudo-Kossel. Kossel microdiffraction can also be obtained by employing synchrotron radiation [10].

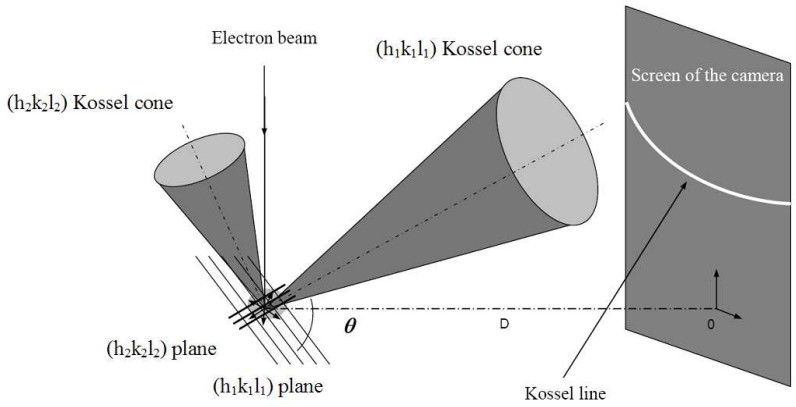


Fig. 1: The principle of the Kossel microdiffraction inside a SEM

Several materials have already been tested with this technique: copper, nickel and titanium alloys, steels and germanium. An example of a Kossel line pattern from a grain of interstitial free (IF) steel is shown in Fig. 2b. The grain that was selected is circled in Fig. 2a. The spatial resolution is estimated to be about $1\mu m^3$, for a 20kV microscope voltage.

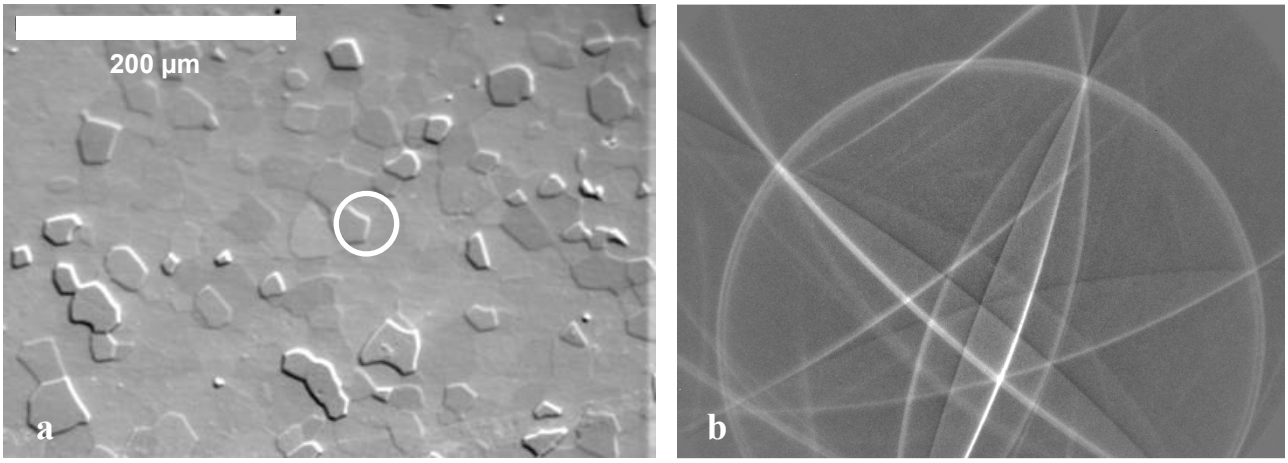


Fig. 2: a) SEM image of the microstructure showing the grain analysed by Kossel diffraction - b) Experimental Kossel line pattern from an IF steel grain

The obtained Kossel line pattern is then indexed using a semi-automatic program, KSLStrain, developed by Adam Morawiec [11]. Several parameters are fulfilled in the program: the radiation wavelength of the emitted X-rays that is known with relatively high accuracy, the unit cell of the material, the pixel size of the camera and approximate values of the camera geometry parameters (sample-to-detector distance and the location of the pattern centre). Then, conics on the pattern are marked manually selecting a number of points per each Kossel line (Fig.3a). Starting with fixed reference values of lattice parameters, a procedure is used to find the geometry of the pattern. Once

the crystal orientation is determined and the camera geometry parameters are tuned, the program can proceed with the refinement of lattice parameters. The program uses the so-called "K-line equation based scheme". Kossel conics manually marked by the operator are matched to corresponding conics in simulated patterns. At the end, the camera geometry parameters, the crystal orientation and the strain components are fitted together. Visual example of a result is shown in Fig.3b. The strain resolution is estimated to be about 3.10^{-4} [11].

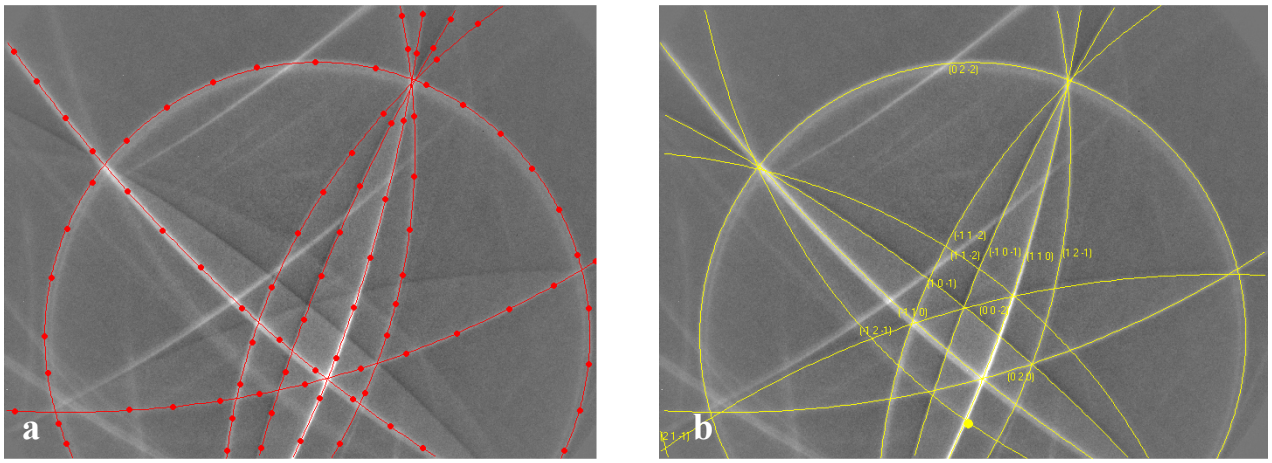


Fig. 3: a) IF steel pattern with some conics marked -
b) Simulated pattern after the fittings superimposed with the experimental pattern

Measurements on single crystals under loading

To check the consistency of the stress results given by the Kossel microdiffraction, we have performed measurements on a nickel-based single crystal superalloy during an *in situ* tensile test. The chemical composition (in weight %) of the material is the following: Base Ni - 8% Cr - 5% Co - 8% W - 2% Mo - 5% Al - 1.5% Ti - 6% Ta. Its main feature is a high yield strength (about 900MPa). In fact, it is made of two coherent phases: a Ni matrix γ and $((Ni, Co, Cr)_3 (Al, Ti, Ta))$ cubic precipitates γ' (Fig.4a). We used a small tensile testing device (Fig.4b) to apply loading to "dog bone" specimens up to about 700MPa.

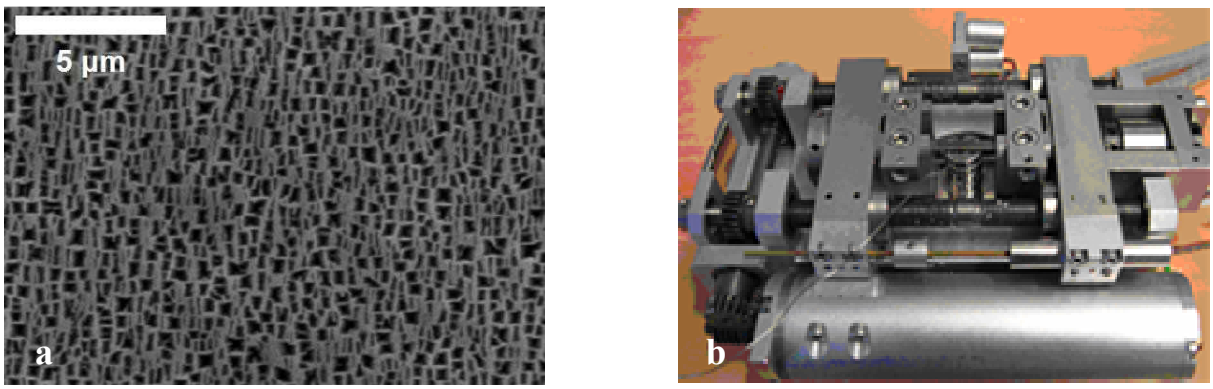
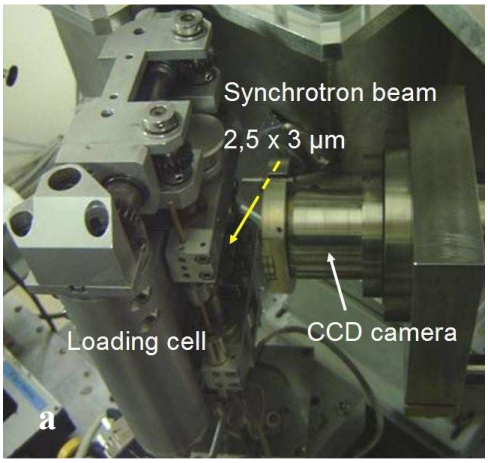


Fig. 4: a) SEM image of the nickel-based superalloy microstructure -
b) *In situ* uniaxial tensile/compressive testing device

Kossel experiments were performed by two different ways. First, a monochromatic synchrotron beam (European Synchrotron Radiation Facility, ID13 beamline, Fig.5a) was used to excite the material fluorescence. The beam size was about 3 micrometers. Photon energy of 9.4 keV was chosen to be just above the K-edge of the nickel. Data was collected at different loadings and stress

tensors were determined assuming a biaxial stress state, due to the low emission depth of X-rays (Fig.5b).



Initial state	400MPa loading	590MPa loading
$\begin{pmatrix} 10 & 55 & 0 \\ & -25 & 0 \\ & & 0 \end{pmatrix}$	$\begin{pmatrix} 400 & -10 & 0 \\ & -135 & 0 \\ & & 0 \end{pmatrix}$	$\begin{pmatrix} 500 & -20 & 0 \\ & -200 & 0 \\ & & 0 \end{pmatrix}$

b

Fig.5: a) Experimental set up used at ID13 (ESRF) to obtain Kossel line patterns from a monochromatic synchrotron beam - b) Stress tensors (MPa) obtained by Kossel diffraction with a synchrotron beam at different stress states. σ_{11} according to the tensile direction. Assumption: biaxial stress sate.

The same experiments were performed inside the SEM. Results are shown in Table 1 and an example of a Kossel line pattern is shown in Fig.6.

Table 1: Stress tensors (in MPa) obtained by Kossel diffraction inside the SEM at different stress states. σ_{11} according to the tensile direction. Assumption: biaxial stress sate.

Initial state	400MPa loading	680MPa loading
$\begin{pmatrix} -90 & 25 & 0 \\ & 0 & 0 \\ & & 0 \end{pmatrix}$	$\begin{pmatrix} 400 & 25 & 0 \\ & -25 & 0 \\ & & 0 \end{pmatrix}$	$\begin{pmatrix} 715 & -10 & 0 \\ & 10 & 0 \\ & & 0 \end{pmatrix}$

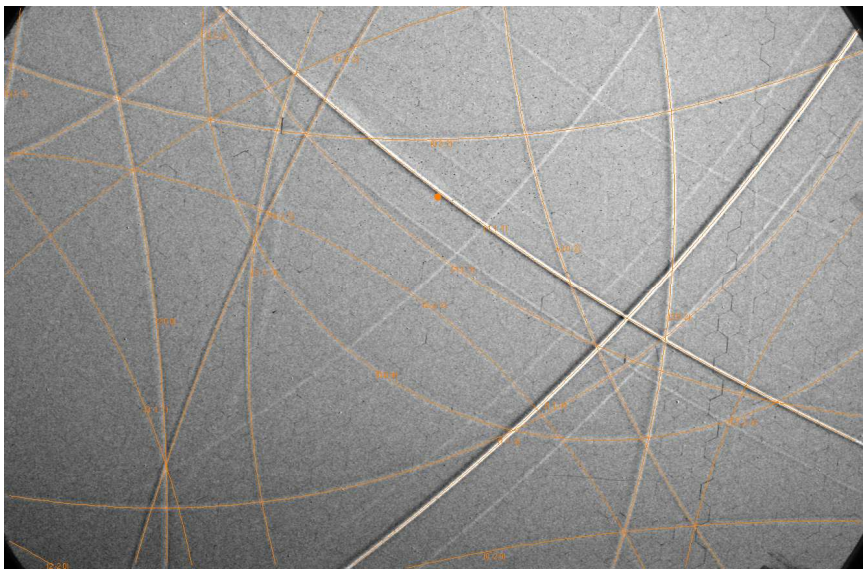


Fig.6: Simulated pattern after the fittings superimposed with the experimental one (400MPa loading)

Single crystal XRD stress analyses, using the Ortner's method [12], were carried out within a four-circle goniometer to compare with Kossel experiments (Table 2). The probed area was about a few mm².

Table 2: Stress tensors (MPa) obtained by single crystal XRD.
σ_{11} according to the tensile direction. Assumption: σ_{33}=0.

Initial state		**650MPa loading**
$\begin{pmatrix} 0 & 45 & -10 \\ & -120 & 10 \\ & & 0 \end{pmatrix} \pm \begin{pmatrix} 40 & 20 & 10 \\ & 35 & 10 \\ & & 25 \end{pmatrix}$	→	$\begin{pmatrix} 600 & 40 & -20 \\ & -135 & 5 \\ & & 0 \end{pmatrix} \pm \begin{pmatrix} 45 & 20 & 10 \\ & 40 & 10 \\ & & 30 \end{pmatrix}$

With the assumption of a biaxial stress state for the stress calculation from Kossel patterns, *in situ* stress results given by the three techniques and the macroscopic stress applied by the machine were compared, showing a good agreement.
XRD measurements in one grain take several hours, so a slight relaxation of the tensile device can explain why the σ_{11} obtained is a little below the applied stress. The σ_{22} compressive values, which are the same for the two loadings, are probably due to the surface preparation.
In synchrotron experiments, only one Kossel line pattern was obtained per loading. The synchrotron beam was focused at a random location on the single crystal surface. On the other hand, in the SEM, three patterns were recorded per loading and stress tensors shown in Table 1 correspond to an average of three measurements. Three electronic spot locations were chosen a few hundreds of micrometers apart. The results are consistent with the loading applied but we noticed high standard deviations. The σ_{11} deviations are shown in Table 3. For the initial state, the non-uniform surface preparation could explain these deviations. Once the material is loaded, the standard deviations decrease.

Table 3: σ_{11} standard deviations ($\Delta\sigma_{11}$) calculated from three different Kossel analyses.

	Initial state	**400MPa loading**	**680MPa loading**
$\Delta\sigma_{11}$ [MPa]	135	75	70

These deviations are probably due to the stress distribution between the two phases, the lattice misfit or local deviations of residual stresses induced by the surface preparation. The last value (about 70MPa) can also give information about the stress resolution of the Kossel microdiffraction, when such a pattern as shown in Fig.6 is obtained.

Conclusion

This paper reports on microscale stress measurements using a non-destructive method, Kossel microdiffraction. Kossel patterns were obtained during *in situ* tensile experiments on single crystals in the Scanning Electron Microscope and with a monochromatic synchrotron beam. The patterns, recorded by a high resolution CCD camera, contained enough lines to determine the crystallographic orientation of the crystal and the elastic deformation of the lattice. Nickel-based single crystal superalloys were analyzed: different loadings up to 700MPa were applied on these specimens with a tensile micromachine and compared with the values obtained using the Kossel technique. A good agreement was observed. The results show how Kossel microdiffraction can be used as a precise tool to obtain residual stresses in polycrystalline samples. This technique could be

used, for example, to analyse residual stresses in microelectronic components like Cu-MEMS (Micro Electro Mechanical Systems).

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