Determination of dislocation structure and vacancy concentration by in–situ synchrotron X-Ray diffraction

Theses of Ph.D. dissertation

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1. Introduction

In metals or alloys deforming by wavy glide, TEM observations have shown that in stage III, dislocations form a cell structure where thick and woolly cell walls of high dislocation density surround the cell-interior regions with considerably lower dislocation density [1]. At the beginning of stage III the misorientations between adjacent cell wall or cell interior regions are close to zero. Approaching the transition to stage IV, cell blocks appear which comprise many dislocation cells being separated by misorientations of tilt or twist. In stage IV, the cell walls get thin and often form lamellar structures, which are separated by marked misorientations. The strengthening of stage IV has been modeled by assuming that high and low dislocation density regions behave like a two-phase composite. In this work it is shown that a mechanical investigation with the simultaneous “in situ” X-ray diffraction study by synchrotron radiation enables a more detailed insight into the basically operating microstructural processes, especially the dislocation mechanisms governing the transition from stage III to stage IV during plastic deformation.

For the explanation of the macroscopic plastic flow constitutive dislocation models are considered. One of the basic assumptions of these models is that the material and its behavior is identical for all (micro) volumes, therefore averaging over the elementary processes of plastic deformation is appropriate. However, there are numerous experimental indications that the plastic flow is inhomogeneous and localized in certain regions of the material. Based on the results of an advanced strain measurement technique Zuef and co-workers suggest moving deformation fronts as the mechanism of single crystal deformation and explain it by a self-exited wave model. Moreover, it is known that the inhomogeneous distribution of the generated dislocations in the material leads to the formation of a deformation cell structure. This cell structure governs the
strength behavior on the microscopic level [7] and is the subject of numerous investigations. Additionally this microstructure is the precursor of the subgrain structure and can lead even to a nanocrystalline structure if deformed by the methods of severe plastic deformation (SPD). It is thus essential for the further progress of the understanding of the deformation process to find the relation between the heterogeneity of the flow behavior and the microstructure. This work tackles this task via in-situ X-ray Bragg Profile Analysis using synchrotron radiation yielding microstructural parameters with a high time and lateral resolution:

The production of point defects by plastic deformation is well known ever since different physical parameters, especially electrical resistivity have been measured as a function of deformation [2]. In 1978, Roennpagel and Schwink measured the stored energy during tensile deformation of copper single crystals in an in-situ deformation calorimeter [3]. Their dynamical measurements of the stored energy provided 3 to 4 times larger dislocation densities than observed by etch pitting or transmission electron microscopy (TEM). Wilkens suggested to measure the X-ray line broadening on the same bulk specimens, which were tested in the deformation calorimeter by Roennpagel. The result was rather sobering as for the correctness of the TEM results of dislocation densities, however, it did not clear the discrepancy. Weak-beam TEM micrographs of the deformed specimens have shown the presence of small defect clusters, most probably dislocation loops of vacancy type of an estimated diameter of about 3 nm or smaller. It was also observed that these defects were located preferentially in the dislocation cell-wall regions. Relaxed vacancies and/or vacancy clusters or small dislocation loops of vacancy type cause a lattice parameter increment in good correlation with the shifts of Bragg peaks to larger diffraction angles. The same lattice defects do not contribute to line broadening, since they contribute basically to the diffuse background scattering, often called Huang scattering. In the present work, it is shown that the diffuse background scattering in X-ray line profile analysis can be used for determining the rate of vacancy production during plastic deformation.
2. **Applied methods**

The measurements underlying the results of this work were carried out using 8 keV photons at the SAXS beamline of the ELETTRA synchrotron in Trieste which were provided by a 57-pole permanent magnet wiggler and a pair of asymmetrically-cut Si(1 1 1) double-crystal monochromator. After the monochromator, a toroidal mirror focuses the beam. A four-edge movable slit system enabled the reduction of the beam size. A second adjustable has been applied to reduce parasitic slit scattering induced by the first slits.

A compact compression test machine has been constructed to implement in-situ measurements. The compression machine is placed on a robust 5-axis Eulerian-cradle goniometer so that the specimen surface being hit by the X-ray beam is in the eucentric point of the goniometer. The irradiated specimen position was continuously monitored by a far-working-distance microscope. The peak profiles were measured in reflection by a gas filled linear position sensitive detector. Due to these experimental conditions, no instrumental corrections of the measured line profiles were necessary.

The evaluation procedure is based on a theoretically well-established function, which describes the shape of broadened line profiles. The parameters of this function were provided by Multiple Whole Profile fitting, which provided the average subgrain size, the size distribution, the dislocation density and the dislocation arrangement parameter [4].
3. New scientific results

3.1. A second-order phase-transformation of the dislocation structure during plastic deformation determined by in situ synchrotron X-ray diffraction [S2]

I have shown that in case of Cu single crystals:

- The Mecking-plot of compressed Cu single crystals show that the transition of III and IV stages of deformation occurs at $\sigma = 40 \pm 2$ MPa.
- Inhomogeneous distribution of the generated dislocations cause asymmetric diffraction peaks.
- The absolute values of internal stresses corresponding to both cell walls and interiors are increasing with the deformation. The values of the internal stresses corresponding to cell walls are negative.
- The values of dislocation density of cell walls and cell interiors increase by about one and a half orders of magnitude over the entire range of deformation. The dislocation density of cell walls increases faster in stage IV.
- The total internal stress acting between the cell wall and cell-interior regions increases too with deformation, but the speed of the increase was higher in stage III.
- Both local M values are decreasing up to the onset of stage IV. The Mw values remain nearly constant, while the Mc values increase again within stage IV.
- A well-pronounced strong maximum of the fluctuation square of the dislocation density can be seen at the transition from stage III to stage IV. The weighted flow-stress ratio increases monotonously up to about 40 MPa, the assumed transition from stage III to stage IV, and decreases monotonously above this stress level and finally almost saturates around the value of unity.
3.2. Microstructural Parameters In Large Strain Deformed Ni-Polycrystals As Investigated By Synchrotron Radiation [S1]

I have found that the dislocation density determined by residual electrical resistivity measurements show the same quantitative evolution like gathered from X-Ray measurements.

I have determined that while at $\varepsilon = 0.11$ all three parameters $|\Delta \sigma - \Delta \sigma_c|$, $\rho^*$ and $M$ show very homogeneous quantity distribution over the scanned path, the results at $\varepsilon = 0.62$ at one point the formal dislocation density is markedly higher than at the neighboring points but the values of $|\Delta \sigma - \Delta \sigma_c|$ and $M$ are lowered relative to their neighborhood. This inverted proportionality can also be observed at the scan with $\varepsilon = 0.91$ but contained in larger areas at general higher level of quantities.

3.3. Spatial Fluctuations of the Microstructure during Deformation of Cu Single Crystals [S4]

I found that dislocation density fluctuates over the whole scan path independent of the deformation degree. These fluctuations are well above the error of the method being about $1 \times 10^{13} \text{m}^{-2}$; The relative magnitude of the variations is about 30–40% for all scans.

I found that up to a deformation of $\varepsilon = 0.08$ the fluctuations of $\rho$ are definitely not stationary. However, for deformations $\varepsilon \geq 0.08$ the variation pattern seems to stabilize (especially if one looks at the most pronounced maxima and minima). A moderate growing of the dislocation density at small $\varepsilon$ ($\rho < 10^{14} \text{m}^{-2}$) is followed by a strong increase up to $3.8 \times 10^{14} \text{m}^{-2}$ from $\varepsilon = 0.12$ to $\varepsilon = 0.16$.

I have shown that the plots of the outer cut-off radius $R_e$ of the dislocation-strain-field and of the dislocation-arrangement-parameter ($M = R_e (\rho)^{0.5}$) are over all parallel to each other but antiparallel when compared to the graph of the dislocation density. At $\varepsilon = 0.16$ the concurrent behavior of $M$ and $R_e$ starts to disappear: At locations with
similar values of $R_e$ the values of the dislocation density differ. Neither has it been possible up to now to measure the microstructural evolution during deformation nor to obtain quantitative information on the dislocation density and particularly the strain field of dislocation arrangements as a function of deformation and lateral position.

3.4. **Vacancy concentrations determined from the diffuse background scattering of X-rays in plastically deformed copper [S2, S3, S5]**

I found that in the polycrystalline specimen, the dislocation density increases considerably faster at the beginning of deformation and remains much larger throughout the entire deformation range than in the single crystalline specimen. For the polycrystalline specimen, the ratio of the background to peak scattering increases considerably faster at small $\varepsilon$ values and remains much larger throughout the entire deformation range than the ratio corresponding to the single crystalline specimen.

I have shown that a fairly good proportionality between the $R$ values and the dislocation densities can be observed. The rates of the increase of the diffuse background in the two different specimens are obtained to be $R_0^{sx}=0.002$ and $R_0^{px}=0.026$, respectively, where the dislocation density is measured in units of $10^{14} \text{m}^{-2}$.

4. **Conclusions**

The fluctuations of the parameters of the microstructure reach a maximum during the process of deformation, and then decrease with notable degree. In the stage IV of deformation the microstructure are homogenized because of the form of extra vacancies causing heavy diffusion activity at high deformation values.
The experimental results indicate that when the deformation process passes from stage III to stage IV the dislocation structure reveals a second-order phase transition in [100] single crystals being oriented for ideal multiple slip. The fluctuations of the dislocation density and the weighted flow-stress ratio go through a maximum as a function of the applied stress in a similar way as, for example, the magnetic permeability in a magnetic phase transition. The experiments show that $M_w$, the dislocation arrangement parameter within the cell-wall material, plays the role of the order parameter of the second order phase transformation in the dislocation structure. At the transition to stage IV the PDW structure collapses while the more densely populated PTW walls produce tilts (and twists) where the adjacent cells are usually alternately misoriented with the dislocation arrangement in the cell walls being typical of small angle grain boundaries.

The cell wall transformation model [5, 6] can also be applied to large strain deformed Ni, where the microstructural changes set in at the transition range from deformation stage III to stage IV.

Observing copper single crystals during compression a discontinuous behavior of the dislocation density was observed in spatial scans. This can be interpreted as a consequence of the propagation of deformation waves in contrast to the homogeneous deformation observed in polycrystals. This spatial distribution of the dislocations seems to get stationary at higher deformation of $\varepsilon = 0.12$ when dislocation dipoles become sessile by forming a cell structure. At higher deformation, the PDW structure transforms to PTW structure, but not at once in the whole sample.

The background intensity of X-ray diffraction patterns is analyzed in terms of point defects, especially vacancies, in poly- and single-crystalline copper specimens. The samples have been deformed by compression in-situ in a synchrotron peak profile experiment. Systematic comparative analysis of X-ray, electrical resistivity, and calorimetric measurements indicate that

(i) point defects, especially vacancies, are produced during plastic deformation and
(ii) that the point defect concentration is increasing concomitantly with the actual dislocation density [7].

The evolution of X-ray diffuse background scattering indicates that the vacancy production rate in polycrystalline specimens is more then an order of magnitude larger then in single crystal samples. This result is interpreted by assuming that the vacancy accumulation in the grain boundary region is larger than in the grain interior or matrix regions. The evaluated vacancy concentration values approach vacancy concentration values corresponding to those prevailing the melting temperature of the material. This result would indicate that the state of the grain boundary region in strongly deformed metals is somewhat similar to the state at the melting temperature.
5. **Own publications**


6. References