New Laue Micro-diffraction Setup for Real-Time In Situ Microstructural Characterization of Materials Under External Stress

D. Popov, S. Sinogeikin, C. Park, E. Rod, J. Smith, R. Ferry, C. Kenney-Benson, N. Velisavljevic and G. Shen

Abstract Laue X-ray diffraction (XRD) is a powerful probe to characterize pressure-/strain-induced microstructural changes in materials. The use of brilliant synchrotron radiation allows Laue XRD to be measured in a fast manner, leading to microstructural characterization, such as two-dimensional maps of single-crystals, their texture, and deformation, to be made in time-resolved mode with temporal resolution down to seconds. This technique can be very efficient in the studies of mechanisms of deformation, grain growth, recrystallization, and phase transitions. A progress has been obtained to extend application of Laue diffraction to high-pressure area. Recent case studies of $\alpha \rightarrow \beta$ transition in Si and $\alpha \rightarrow \omega$ transitions in Zr are briefly reported. A new experimental setup specifically optimized for real-time in situ Laue measurements has been developed at the 16-BMB beamline at the Advanced Photon Source. Due to the large X-ray energy range, which is typically up to 70 keV, a polychromatic beam diffraction technique can be efficiently implemented despite some limits introduced to the scattering angle by strain generation devices. Currently, the X-ray beam is focused at the sample position down to ~$2.2 \times 2.2\ \mu m^2$ at the full width at the half maximum. Precision sample translation stages provide fast data collection with step sizes down to 0.5 μm.

Keywords Laue diffraction · Microstructure · High pressure Synchrotron radiation

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Introduction

Laue micro-diffraction using synchrotron radiation is an efficient tool to investigate mechanical behavior of materials in situ under external stress [1–3]. An exciting advantage of Laue diffraction, compared to the widely implemented diffraction with monochromatic beam, is that the single-crystal diffraction data can be collected several orders of magnitude faster, which enables fast microstructural mapping of materials with time resolution down to seconds. Changes in crystal morphology and orientation, twinning, strain, dislocations, and other defects can be observed, even if they are essentially controlled by kinetics and take place only in limited periods of time. At high-pressure collaborative access team (HP-CAT), considerable effort has been undertaken to implement this powerful technique to investigate mechanisms of materials transformation under quasi-hydrostatic high-pressure conditions. For instance, an irreversible $\alpha \rightarrow \beta$ transition in Si under hydrostatic compression in a diamond anvil cell (DAC) was characterized by D. Popov, C. Park, C. Kenney-Benson, and G. Shen [3]. By doing real-time mapping across the sample, gradual changes in morphology of the parental single-crystals and growth of the product phase were observed on the time scale of minutes. The morphology of the coexisting α- and β-phases is presented schematically in Fig. 1. Areas of the high-pressure phase were elongated parallel to a <100> direction of the parental single-crystal indicating that nucleation and growth of the β-phase may have been controlled by defects. The single-crystals of α-Si exhibited measurable deformation. Qualitatively this deformation can be interpreted as 'twisting' about their longest dimensions by angles less than 0.2°.

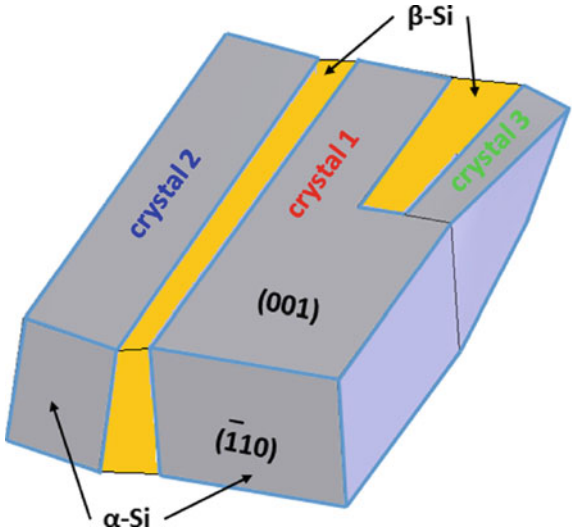

Fig. 1 Schematic of the appearance of the β-Si [3]. Crystals 2 and 3 are shown deformed assuming that they are twisted around their longest dimensions. Angles between the crystals are multiplied by a factor of 50

Grain enlargement is typically promoted by thermal energy to overcome activation barriers for diffusion, motion of dislocations, and migration of grain boundaries including newly formed ones. In some cases, however, pressure-induced phase transition results in grain enlargement in product phases at room temperature [4–7], and its cause may be fundamentally different from processes at high temperatures. Such pressure-induced grain enlargement has been reported in Zr based on comparison of X-ray diffraction patterns obtained with monochromatic beam on powdered α-Zr and on grainier ω-Zr [6, 7]. The first *in-operando* study of the grain enlargement process by real-time Laue diffraction mapping of the newly formed ω-Zr crystals has been conducted by D. Popov, N. Velisavljevic, C. Park, and G. Shen. The observed ω-Zr crystals nucleated locally, independently from each other, were growing gradually and continuously preserving their orientations during enlargement. The obtained results support a mechanism for the enlargement of the ω-Zr grains which are driven by energy stored during the deformation of the parental α-Zr. The widely known recrystallization process at high temperatures has the same driving force [8, 9]. The growth of ω-Zr crystals may be also driven by reduction of the total grain boundary area in a similar way as in the case of grain coarsening with 'abnormal' grain growth when selected grains grow up to much larger size than the average [8, 10].

Experimental Setup

A new experimental setup optimized for in situ high-pressure, time-resolved measurements using Laue micro-diffraction has been developed at the HP-CAT 16-BMB beamline of the Advanced Photon Source by the authors (Fig. 2). Different types of strain generation devices can be used in the setup, including deformation stages, indenters, other devices providing uniaxial compression/tensile, and DACs providing quasi-hydrostatic compression. For measurements with DAC, the setup is equipped with a membrane control system and a gearbox for remote tuning of pressure. A ruby fluorescence system is available on the side of the experimental table to adjust the DAC pressure before data collection.

All setup components are mounted on a granite table to maintain mechanical stability. A Perkin Elmer area detector (XRD1621) is used to record diffraction images. The angle of the area detector with respect to the incident X-ray beam can be changed in steps of 15° in the vertical plane using an adjustable detector arm, making it possible to collect Laue patterns in both transmitted and 90° geometries. The bending-magnet white X-ray beam is focused down to $2.2 \times 2.2\ \mu m^2$ at the sample position with KB mirrors. White X-rays with a 70 keV cutoff provide a reasonable number of reflections even in transmitted geometry, which is important since the 90° geometry is often difficult due to limitations on the scattering angle introduced by various strain generation devices. On demand, the incident flux can be increased by changing the focusing mirror angle (at a cost of lowering the upper X-ray energy cutoff).

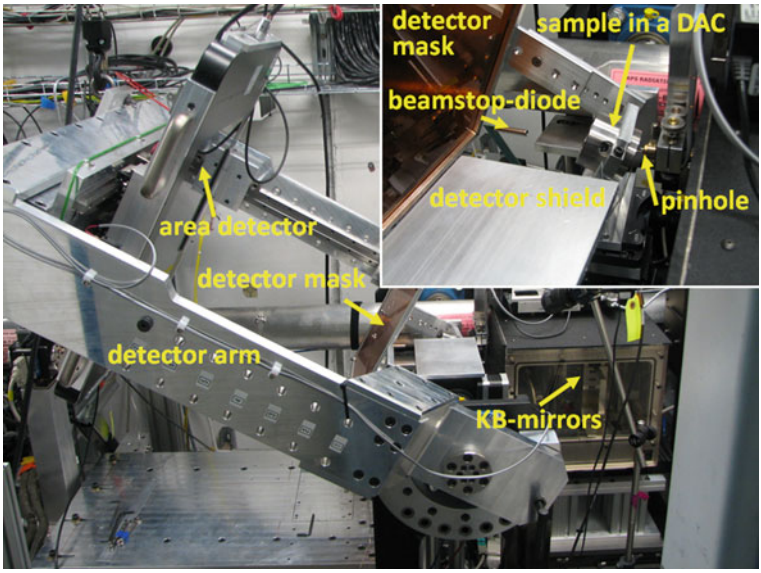

Fig. 2 Experimental setup at the 16-BMB beamline of Advanced Photon Source for in situ and in operando Laue micro-diffraction

Mutually perpendicular Newport horizontal translational stages and an Aerotech elevation stage with Newport XPS controllers are used for sample positioning. Samples mounted in strain devices with a mass up to 5 kg can be scanned in the two transverse dimensions with step sizes down to 0.5 μm. Multiple scans are collected in series in order to get sets of maps in real time. Strong reflections that can damage the area detector are blocked by a detector mask during data collection. A shield is introduced between the sample and area detector to protect the detector during the scanning process needed to align the sample to the X-ray beam. A Si111 channel-cut monochromator, switchable with the white beam in a matter of minutes, is used to calibrate detector geometry with the Dioptas [11] program, which is subsequently used for measurement of d-values of selected reflections and identification of powder-like phases.

Multiple data analysis programs are available at the beamline. Developed by D. Popov, the program PolyLaue, combined with Fit2d [12], can be used to identify single crystals by indexation of Laue reflections based on known unit cell parameters. The shape of the unit cell can be further refined based on the Laue data. In the case when multiple single crystals contribute to diffraction patterns simultaneously, orientation matrices of individual crystals are determined, and the predicted positions of diffraction spots are shown on diffraction images. PolyLaue can also generate maps of reflections. The XDI [13] program is also used to build maps of reflections but in a more efficient way by quantifying their intensities. The program LaueGo [14] maps lattice rotation and refines deviatoric strain. The current instrumentation

allows determination of deviatoric strain down to ~10^{-4} level. LaueGo is also useful to calibrate the position and orientation of the area detector in 90° geometry.

Conclusions

Synchrotron radiation Laue diffraction is a powerful approach to investigate materials in situ and *in-operando* under external stress. Substantial progress in application of this technique to the high-pressure area has been recently obtained at HP-CAT. New experimental setup specifically optimized for in situ real-time measurements at high pressures has been developed at the HP-CAT 16-BMB beamline of Advanced Photon Source.

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