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Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
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Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Structural disorder of refractory high-entropy alloys	**Experiment number**: MA-2019
Beamline: BM01A	**Date of experiment**: from: 06/11/2013 to: 09/11/2013	**Date of report**: 25/02/2014
Shifts: 9	**Local contact(s)**: Dmitry Chernyshov	*Received at ESRF:*

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Report:

Experiment

The main aim of the experiment was to investigate into the structural disorder of some of the refractory high entropy alloys (HEAs) with the help of diffuse scattering from single crystal X-ray diffractions. Single crystals of boby-centered cubic (BCC) HEA compositions like $Mo_{0.25}Nb_{0.25}Ta_{0.25}W_{0.25}$, $Mo_{0.20}Nb_{0.20}Ta_{0.20}W_{0.20}V_{0.20}$, $Ta_{0.25}Nb_{0.25}Hf_{0.25}Zr_{0.25}$ and $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ etc were measured at the Swiss Norwegian Beam Line (BM01A) using the PILATUS 2M detector. The refractory HEA materials were homogenized between 1600-1800 ^{o}C to achieve a uniform composition throughout the whole bulk material. Two datasets for each crystal of the above materials were measured, one for the diffuse scattering and another for the Bragg scattering intensities with an attenuator. Oscillation angle of 0.1^{o} and X-ray beam of wavelength 0.6966 Å were used for the measurements. Lanthanum hexaboride powder and a ruby crystal were used to determine the proper instrumental parameters. Crystals from HEA compositions like $Ta_{0.25}Nb_{0.25}Hf_{0.25}Zr_{0.25}$ and $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ show pronounced degree of diffuse scattering. For these materials the crystals were given three full rotations for 1 hour each (total 3 hours of diffuse scattering measurement for each crystal), and later the three datasets were merged together. All the diffraction measurements were carried out at ambient temperature conditions.

Results

All the measured crystals of the refractory HEAs form a single-phase BCC (spacegroup Im-3m) solid solution. The $Mo_{0.25}Nb_{0.25}Ta_{0.25}W_{0.25}$ and $Mo_{0.20}Nb_{0.20}Ta_{0.20}W_{0.20}V_{0.20}$ crystals did not show any detectable diffuse scattering, but the $Ta_{0.25}Nb_{0.25}Hf_{0.25}Zr_{0.25}$ and $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ crystals show significant amount of diffuse scattering near to the Bragg reflections. The datasets were integrated for their Bragg intensities with CrysAlisPro program. The reconstructed reciprocal lattice layers indicate an excellent quality of the crystals with no texture, mosaicity or crack present in it. On average the crystals have internal R value of around 15%. The experimental conditions were perfect in the sense that there was no fluorescence observed in the diffraction data frames. The

background intensity of the detector was almost zero other than little air-scattering around the direct beam. But the air-scattering did not affect the diffuse intensities around (110) reflections.

$Ta_{0.25}Nb_{0.25}Hf_{0.25}Zr_{0.25}$ and $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ crystals show two remarkable features in the reciprocal lattice layers. The first kind is the presence of streak-like diffuse scatterings extending towards lower diffraction vectors and adjacent to the main Bragg reflections (Fig. 1). These diffuse scatterings are almost parallel to the main crystallographic axes. This indicates presence of disorder perpendicular to the crystallographic axes and relaxation of the average structure due to the disorder.

The second kind of feature present in $Ta_{0.25}Nb_{0.25}Hf_{0.25}Zr_{0.25}$ and $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ crystals is the increasing Bragg-peak width broadening and a huge drop in the Bragg intensity with higher scattering vectors. From the Bragg peak width analysis it appears that the crystal has around 1% internal strain, even without any external damage to the structure. Also the pronounced drop in Bragg intensity with diffraction vector, indicate a high degree of local lattice distortion and presence of very high static atomic displacement parameters (ADPs). Presence of local lattice distortions have been already reported by us for the $Mo_{0.25}Nb_{0.25}Ta_{0.25}W_{0.25}$ alloy [1]. The diffuse scattering in Fig. 1 is arising both from chemical short range ordering (SRO) and local lattice distortion effects.

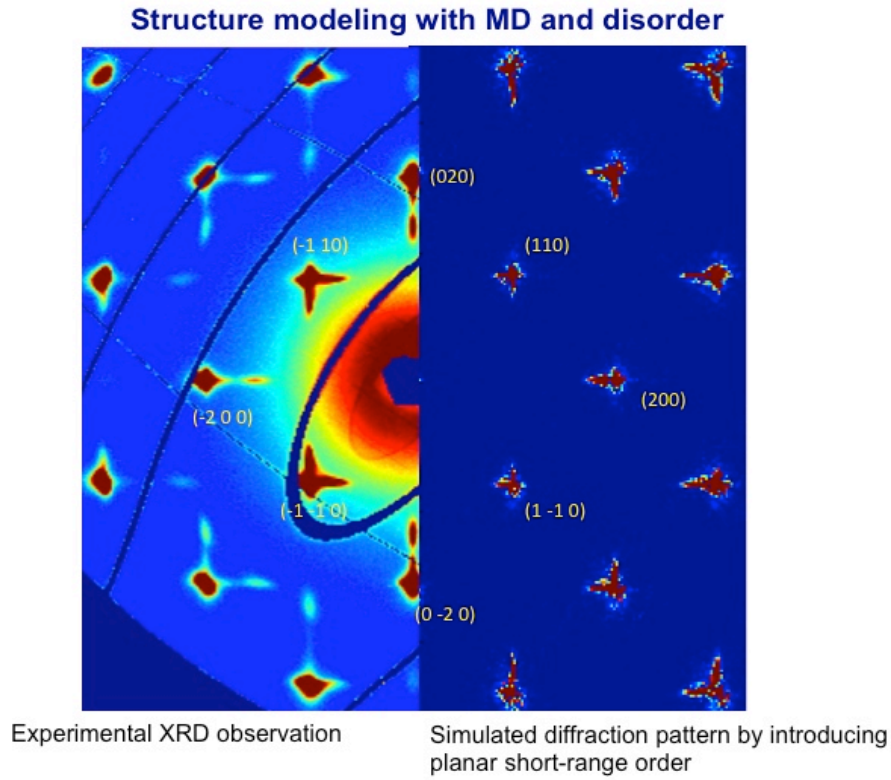

Fig. 1. Reconstructed hk0 layer of $Ta_{0.20}Nb_{0.20}Hf_{0.20}Zr_{0.20}Ti_{0.20}$ from synchrotron diffraction (left). Simulation of diffuse scattering (right) is done using molecular dynamics (MD) and atom probe tomography (APT).

Following the abovementioned results of refractory HEAs, a follow-up proposal to SNBL will be submitted in the running call (March 2014). In the next synchrotron experiments, the diffuse scattering HEAs will be annealed for different time periods such as 6 hrs, 12 hrs, 1-4 days, and will be measured for their diffuse scattering evolution. In this way the diffuse scattering due to chemical SRO and that from the local lattice distortions will be separated out. Modeling of SRO of Zr supported by atom probe tomography has already been performed (Fig. 1). The next synchrotron measurements will help reveal the local static lattice distortions, as suggested in the existing literatures [1, 2] and believed to be the reason behind the high mechanical strength of HEAs.

References

[1] Y. Zou, S. Maiti, W. Steurer, R. Spolenak, Acta Materialia (2014), 65, 85-97
[2] W. Guo, W. Domowski, et al., Metallurgical and Materials Trans, (2013), 44A, 1994-97