	Experiment title: ***In situ* and real-time X-ray radiography and diffraction imaging of the growth of silicon for photovoltaic applications.**	**Experiment number**: MA-4065
Beamline: **ID19**	**Date of experiment**: from: **06/06/2018** to: **11/06/2018**	**Date of report**: 20/12/2019
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Report:

Scientific Objectives:

The scientific objectives of the proposed experiments were to unveil fundamental mechanisms of the solidification of crystalline silicon (Si) for photovoltaic (PV) applications. This original experiment consists in applying synchrotron X-ray imaging (radiography and diffraction imaging / topography) during the solidification of Si. In particular, the main issue of the complex interaction and dependence between grain boundaries and structural defects namely dislocations need to be studied at high temperatures during heating, melting (1414°C) and solidification. Apart from this fundamental objective, the better understanding and control of defect development constitute major challenges for all most advanced crystalline Si fabrication processes for PV cells. The experiments proposed here were part of a larger project entitled CrySaLID (Crystallisation of seeded Si, Impact of Light Impurities and Defects) funded by the French National research agency (ANR) and coordinated by IM2NP.

Experimental method:

The experiments were carried out at the European Synchrotron Radiation Facility (ESRF) at beam line ID19. The beamline is ideally suited for this experiment because of the large field of view and of the excellent and uniform flux of photons. The unique IM2NP device named GaTSBI (Growth at high Temperature observed by Synchrotron Beam Imaging) was used for the experiments. This high temperature furnace allows Si melting / solidification and is compatible with X-ray synchrotron imaging methods: radiography and topography. In the radiography mode, the sample is illuminated by the white synchrotron X-ray beam. Radiography enables characterising *in situ* and in real-time the morphology and dynamic evolution of the solid/liquid interface during solidification, and measuring growth kinetics. In the diffraction imaging mode, topographs are collected to reveal grain orientation, twinning, crystal network deformation and dislocation related strain fields. We already showed the feasibility and the impact of such research from the scientific point of view applied to Si [1-4]. With this allocated beamtime, one of the main objective was to improve further the obtained results taking advantage of the unique systems and competencies at ID19 beamline. Indeed, so far, for all IM2NP X-ray imaging research activities concerning the solidification of Al-based alloys, quasicrystals and Si, radiography and diffraction imaging modes could only be used alternately during a particular experiment and sensitive photographic films were used to record diffraction images with a limited temporal resolution of about 0.04 Hz. The experimental objective of this experiment was to improve the temporal resolution of the topography images and to obtain a simultaneous recording of radiographs and topographs. This objective was attained during the June 2018 run thanks to the support and implication of the ID19 staff. The whole experimental setup is schematically shown in Figure 1.

The primary beam passing through the sample is used to record the radiography images. The beam is monochromatised after the sample in order to keep a constant heat load on the sample and the image is recorded by a camera. To this end, the white beam is turned monochromatic at a target energy of 17.5 keV using a vertically diffracting Si (111) double-crystal monochromator. The radiography images are recorded using a second detector (sCMOS lens-coupled to a LuAG scintillator) positioned around 7 m downstream to also benefit from propagation phase contrast imaging (edge detection mode). This camera records 2048 × 2048 pixels with a nominal pixel size of 6.5 μm^2 and a 16 bit dynamic range. Besides, a scientific CMOS camera lens-coupled to a LuAG scintillator (commercial Ce-doped $Lu_3Al_5O_{12}$, Crytur company – Czech

Republic) is used to record the images of one of the diffraction spots (topography) in transmission Laue geometry. The camera records 2048 × 2048 pixels with a nominal pixel size of 6.5 µm^2 and a dynamic range of 16 bit. It is coupled with a ×1.5 optic to decrease the pixel size to 4.3 µm^2. The camera is mounted to a manually on air pads moveable rack positioned around one meter away from the sample. To fine-tune the camera position relatively to the diffraction spots, the rack is equipped with two automatic motion systems that allows the camera to be moved horizontally (left and right) and vertically in increments as fines as 20 µm. Images recorded from both modes are fully synchronised. The image acquisition rate was at maximum 2 frames per second which is sufficient to follow the solidification front of the samples.

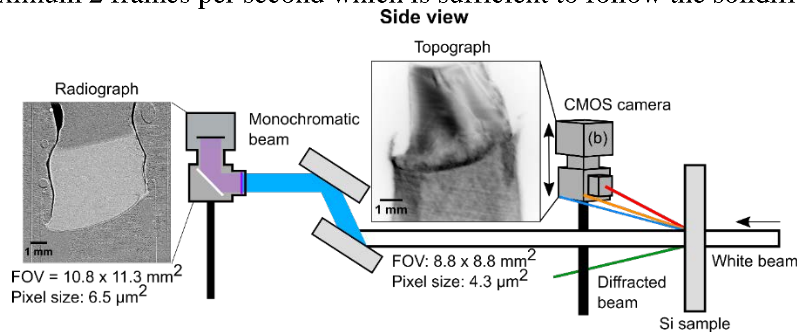

Figure 1. Schematic drawing of the experimental setup using X-ray radiography and diffraction topography modes simultaneously to monitor silicon solidification. The silicon sample is positioned in the solidification furnace not shown here.

Results:

These challenging, cutting-edge *in situ* experiments provided key information on the basic dynamic phenomena implied during Si growth. They were conducted at temperatures ranging from room temperature to the melting temperature of Si (1414 °C), temperatures at which dislocation mobility and interactions with GBs are not known due to the difficulty to work at such high temperatures. They allowed to unveil physical mechanisms during solidification processes and provided quantitative information on crystallographic defects, twinning, strains and dislocations.

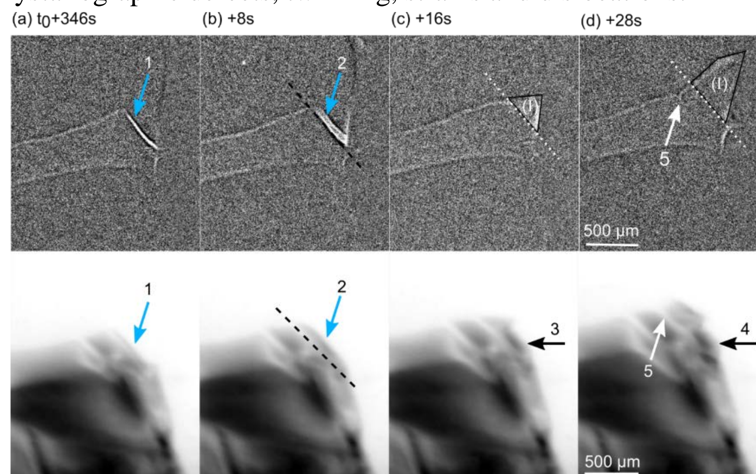

Figure 2. Time-resolved radiography (top row) and topography (bottom row) sequences of twinning and nucleation events [5].

Figure 2 shows a sequence of twinning with simultaneous recording of radiography and topography images. First results were published in *J. of Applied Crystallography* in 2019 [5], and were presented in several international conferences (*ICAPS-5 / CSSCR-5 in Austria in June 2019; DRIP XVIII- Conference on Defects-Recognition, Imaging and Physics in Semiconductors, in Germany in September 2019; MRS - Materials Research Society Conference in the USA in December 2019*). More detailed analysis is conducted and other publications are expected.

Moreover, this experimental development and these results benefited to the experiments performed in November 2018 (IN989) whithin the INSIDES (IN-SItu characterisation and SImulation of Defect Evolution in Silicon) project which is a project in collaboration between SINTEF in Norway and IM2NP.

Future work

The GaTSBI set-up coupled with the ID19 imaging environment together with the experience of both ID19 and IM2NP teams are unique tools to answer to the major issues of crystalline defect formation during growth of materials at high temperature.Following the success from the experimental point of view and the scientific progress in the understanding of defect formation during the solidification of silicon, we would like to improve further the experimental environment by allowing the recording of several diffraction spots with cameras in synchronisation with the radiography imaging. Several scientific subjects related to the defect (dislocations, sub-grains, twins..) formation in general are of current interest in industry and in research laboratories and will be studied.

References

1.M.G. Tsoutsouva *et al.*, *Acta Materialia* 115 (2016) 210-223; **2.** V. Stamelou *et al.*, *J. of Crystal Growth* 479 (2017) 1-8; **3.** M.G. Tsoutsouva *et al.*, *Physica status solidi (a)* (2018) 1700758; **4.** T. Riberi – Béridot *et al.*, *Acta Materialia* 177 (2019) 141-150; **5.** M. Becker *et al.*, *J. of Applied Crystallography* 52 (2019) 1312-1320.