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# Investigation of subgrains in directionally solidified cast mono-seeded silicon and their interactions with twin boundaries

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## Abstract

Directional solidification of a cast mono-seed and of a FZ-seed was performed and the grain and defect structures of the seeds as well as of the regrown parts are analyzed. *In situ* X-ray diffraction imaging enabled the observation of the dislocation arrangements. During the heating process, in the FZ-seed, mobile dislocations glide on {111} planes, whereas in the cast mono seed dislocations are arranged in a mainly immobile cellular structure. *Ex situ* grain orientation mappings reveal the presence of subgrains with misorientations up to 3° in the regrown part of the cast mono-seeded sample, which are not observed in the regrown part of the FZ-seeded sample. Subgrain boundaries characterized by misorientations around the [001] growth axis propagate roughly along the growth axis and increase their misorientation by merging with new subgrain boundaries appearing in their vicinity. Although the first inception of subgrain formation cannot be revealed, the comparison of the dislocation arrangements in the two seeds strongly suggests an influence of the latter on subgrain formation. In the regrown part, interactions between subgrain boundaries and twin boundaries show that they can follow  $\Sigma 3\{111\}$  and  $\Sigma 9\{221\}$  grain boundaries or cross  $\Sigma 3\{111\}$  grain boundaries. Whether  $\Sigma 3\{111\}$  GBs are crossed or not depends among other things on the orientation of the grains on either side of the twin. It demonstrates that the grain orientation relationship and not only the grain boundary character play an important role in the subgrain structure evolution and redistribution in a multicrystalline silicon ingot.

## 1. Introduction

Subgrain boundaries (SGBs) are known to be electrically active defects that decrease the photovoltaic (PV) efficiency and, therefore, should be limited during the manufacturing process [1]. SGBs consist of linear dislocation arrangements, as this configuration reduces the elastic energy in the crystal. In (100) grown crystals, SGBs can form during the directional solidification process by growing with the solid-liquid interface inducing a tilted subgrain structure with rotations parallel to the growth direction. The angular deviation of crystal orientations often increases with ingot height, as more dislocations of the same

46 character are incorporated in the SGBs [2, 3]. The orientation of the glide planes with respect to the growth  
47 direction, their activation due to equivalent resolved shear stresses during solidification, but also the  
48 generation and termination mechanisms determine the final dislocation distribution.

49 Subgrains are observed in all silicon ingots that are produced by casting processes for PV applications:  
50 in cast mono (cm-Si) [4-6], high-performance multicrystalline (HPmc-Si), [7] and conventional  
51 multicrystalline (mc-Si) silicon [8]. For mc-Si, either grains with <110> or <111> growth directions were  
52 reported to have the highest dislocation density [8-10]. In cast mono-Si, however, the <110> growth  
53 direction produced fewer dislocation clusters, i.e. narrower clusters originating at the seed junctions than  
54 the <100> growth direction [11].

55 The origin of dislocation clusters is found to be at grain boundaries [12, 13], with  $\Sigma 27a$  GBs [7, 9, 14]  
56 and SGBs [15] playing a dominant role. Besides, dislocations in cast mono-Si are mainly generated at the  
57 junctions of the seed pavement [16, 17]. Additionally, subgrains originate from dislocation alignments  
58 already present in the seed crystals that can propagate into the new grown crystal during growth [18-20].  
59 For the latter mechanism, the initial dislocation arrangement in cells in the seed is a main factor [19, 21]  
60 and depends on the stress in the seed [22-24].

61 Additionally, it was found that the dislocation density in the newly grown crystal is at first lower than  
62 in the un-melted seeds [17, 22]. On the one hand, it could indicate that the thermomechanical stresses  
63 imposed on the seeds during the heating process are higher than during crystallization. On the other hand,  
64 it must be considered that dislocation generation mechanisms in a seed and the newly grown crystal are  
65 different and that dislocation mobility is strongly dependent on temperature and impurities. Therefore, it is  
66 particularly important to control the crystal quality of the seeds in order to control the dislocations in the  
67 regrown part.

68 Just as important to understand the formation of subgrains is to find out the reason for their  
69 disappearance. A high amount of random angle GBs, like it exists in HPmc-Si, causes a decrease of the  
70 amount of subgrains in the ingot, because it is inferred that the dislocations are stopped at these boundaries  
71 [7]. It was also reported that neighbor grains with lower dislocation densities overgrow the grains with  
72 higher cluster densities [8]. These observations show that once formed, the subgrains can only be stopped  
73 by grain competition mechanisms, wherefore subgrain formation is highly undesirable in cast mono growth  
74 for which this mechanism is irrelevant due to the absence of grain boundaries.

75 In order to better understand the role of the seed in the formation of subgrains, small-scale directional  
76 solidification experiments using a float-zone (FZ) seed and a cast mono seed in [001] growth orientation  
77 were performed. We compare the developed grain structure by applying *in situ* X-ray diffraction imaging  
78 to monitor the solidification microstructure during growth and *ex situ* etch pit and electron backscatter  
79 diffraction (EBSD) analysis to reveal the grain structure and defect spatial distribution. Formation of SGBs  
80 in the cast mono-seeded sample is observed and discussed in terms of their formation, propagation and  
81 disappearance.

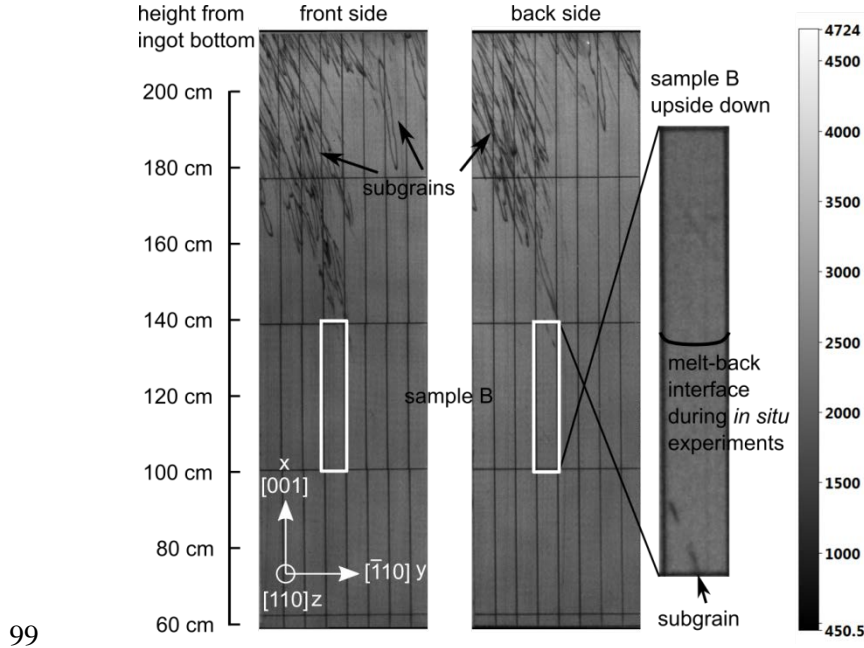
## 82 2. Experimental methods

### 83 2.1. Sample preparation and processing

84 Two samples originating from different processing techniques are used as seeds for directional  
85 solidification. Sample A-seed was produced by zone-melting using 9N material (SIL'TRONIX Silicon  
86 Technologies). Accordingly, the impurity concentrations of oxygen and carbon are below  $10^{15}$  at  $\text{cm}^{-3}$ . The  
87 sample was cut with a diamond wire saw to a size of  $38 \times 7$  mm and polished to a thickness of 0.3 mm. SiC  
88 abrasive paper followed by a  $6 \mu\text{m}$  and a  $3 \mu\text{m}$  diamond suspension was used for final polishing.

89 Sample B-seed was produced by the cast mono directional solidification technique from Cz seeds. The  
90 casted ingot had a laboratory scale 85 kg G2 and was provided by the Institut National de l'Energie Solaire

91 (INES). A photoluminescence (PL) image of a vertical wafer cut from which sample B was taken is shown  
 92 in Fig. 1. A white border marks the sample position (100 cm - 140 cm from the bottom). It is taken just  
 93 below the area where electrically active subgrains do appear on the PL maps. In order to study subgrain  
 94 influence during growth, the sample was put upside down in the furnace before partial melting. The final  
 95 melt-back interface during the experiments was still above the visible subgrains and is shown in black on  
 96 the magnified sample B image. The impurity content in interstitial oxygen  $[O_i]$  and substitutional carbon  
 97  $[C_s]$  at the height of the sample is  $(6 \pm 1) \times 10^{16} \text{ cm}^{-3}$  and  $(4.5 \pm 0.6) \times 10^{17} \text{ cm}^{-3}$ , respectively.  
 98



99  
 100 **Fig. 1** Photoluminescence (PL) image of a vertical cut of the source ingot of sample B. In the upper part, electrically  
 101 active SGBs are visible as dark lines. Sample B was taken just below this area.

102 Both samples A and B have the same crystallographic orientation. The two main surfaces are the (110)  
 103 and  $(\bar{1}\bar{1}0)$  planes, the vertical side planes are  $(1\bar{1}0)$  and  $(\bar{1}10)$  and the horizontal planes are (001) and  $(00\bar{1})$ .  
 104 The samples were placed in a boron nitride crucible and processed in a high temperature Bridgman  
 105 furnace known as GaTSBI (Growth at high Temperature observed by Synchrotron Beam Imaging) [e.g. 25,  
 106 26]. First, the temperature of both heating elements was slowly increased from room temperature up to a  
 107 temperature of 1100 °C. Then, a temperature gradient of 30 °C cm<sup>-1</sup> was applied to the heaters with  $T_{\text{Top}} >$   
 108  $T_{\text{Bottom}}$  and heating continued up to the melting point of silicon which is 1414 °C. The local vertical  
 109 temperature gradient  $G_{\text{local}}$  in the sample was always lower than the applied temperature gradient  $G_{\text{appl}}$  and  
 110 can be calculated with  $G_{\text{local}} = \dot{T}/v$  using the applied cooling rate  $\dot{T}$  and measuring the growth velocity of  
 111 the solid-liquid interface  $v$  [27, 28].  $G_{\text{local}}$  was approximately  $(14 \pm 3) \text{ °C cm}^{-1}$  during the experiments. When  
 112 partial melting of the sample was achieved, directional solidification was triggered, for sample B by pulling  
 113 the sample down with a speed of 0.03 cm min<sup>-1</sup> ( $5 \times 10^{-6} \text{ m s}^{-1}$ ), which corresponds to a cooling rate of 0.4  
 114 °C min<sup>-1</sup>, and for sample A by applying a cooling rate of 0.4 °C min<sup>-1</sup> to both heaters for the first half of  
 115 solidification followed by a cooling rate of 4 °C min<sup>-1</sup> for the second half of solidification. A total of five  
 116 and four heating/cooling cycles were carried out for samples A and B, leading to exposure times to  
 117 temperatures above 1100 °C of 20 and 8 hours, respectively. In this study the last heating/cooling cycle is  
 118 discussed, as the grain structure is preserved for *ex situ* analysis. Although the growth processes of sample  
 119 A and B differ, we do not expect significant differences during crystal growth due to the three following

120 reasons. First, for both analyzed experiments in samples A and B, the measured growth rate (measured  
121 directly thanks to the *in situ* imaging) is identical (within the measurement accuracy limits) and the applied  
122 temperature gradient is also the same. Second, the diffraction images recorded during the first experiments  
123 using cooling rate applied to both heaters in sample B show that the general solidification features are  
124 reproducible compared to the last experiment using pulling down to initiate solidification. Third, the grain  
125 structure observed in both samples A and B is comparable.

## 127 2.2. *In situ observation*

128 *In situ* and real-time observation of the melting and solidification processes was achieved by using X-  
129 ray synchrotron radiation at beamline ID 19 of the European Synchrotron (ESRF) in Grenoble, France. A  
130 combination of X-ray radiography and topography imaging allows studying the development of crystal  
131 defects [29]. Two individual camera-based detector systems were used to monitor the radiographic and  
132 topographic images with an image acquisition rate of  $2 \text{ s}^{-1}$ . The radiography image contrast results from the  
133 density difference between the solid and the liquid phases and provides information on the dynamics and  
134 the morphology of the solid-liquid interface. The topography image results from Bragg diffraction and can  
135 be considered as one extended Laue spot that provides information on the crystal structure and associated  
136 extended defects. Crystal imperfections such as deformations and dislocations change the diffraction angle  
137 of the X-rays, resulting in superimpositions or gaps that are visible in the image as a change in contrast.  
138 The observed contrast provides qualitative information on the strain level which is associated with the  
139 macroscopic crystal deformation.

## 140 2.3. *Ex situ analyses*

141 After the *in situ* solidification experiments both surfaces of the samples were first polished with SiC  
142 abrasive paper and then with a  $6 \mu\text{m}$  and a  $3 \mu\text{m}$  diamond suspension to remove the reaction layer of the  
143 boron nitride crucible and to obtain a flat surface for EBSD analysis. EBSD was performed using a FEG-  
144 SEM JEOL JSM 7001F operating at an acceleration voltage of 20 kV, a working distance of  $\sim 20.0 \text{ mm}$ , a  
145 tilt angle of  $70^\circ$  and several magnifications. The SEM was equipped with a HKL Nordlys camera driven  
146 by the 'Channel 5' softwares suite (comprising 'Flamenco' for acquisition, 'Map Stitcher' for stitching,  
147 'Tango' for mapping and 'Mambo' for pole figures) using either a  $7 \mu\text{m}$  or a  $1 \mu\text{m}$  step size depending on  
148 the studied area. Orientation maps parallel to the growth direction (x), perpendicular to the growth direction  
149 (y) or normal to the sample surface (z) are displayed using the conventional inverse pole figure (IPF)  
150 coloring. For both samples, the x direction corresponds to the growth direction. To obtain large-scale maps,  
151 the individually scanned images were stitched together. In some cases, the individual images do not fit  
152 together perfectly, which leads to visible stitching artefacts on the large-scale maps. Grain boundaries with  
153 a special character are shown in coincidence site lattice (CSL) maps.  $\Sigma 3 \langle 111 \rangle$  GBs are displayed in red,  
154  $\Sigma 9 \langle 110 \rangle$  GBs in blue and  $\Sigma 27a \langle 110 \rangle$  GBs in yellow.  $\Sigma$  (Sigma) is the ratio between the number of lattice  
155 points in the unit cell of the CSL lattice and the number of lattice points in the unit cell of the generating  
156 lattice. The rotations around the indicated  $\langle hkl \rangle$  directions satisfy the misorientation ranges given by the  
157 Brandon criterion [30], which are  $(60 \pm 8.66)^\circ$ ,  $(38.94 \pm 5)^\circ$  and  $(31.58 \pm 2.89)^\circ$  for  $\Sigma 3$ ,  $\Sigma 9$  and  $\Sigma 27a$  GBs,  
158 respectively. The range within special grain boundaries are defined as  $\Sigma$  grain boundaries is given by  $\theta =$   
159  $15^\circ/\sqrt{\Sigma}$ . Additionally, misorientation maps that show small angular deviations from a predefined  
160 crystallographic direction, are used to detect subgrains. The detection limit of misorientation angles is  
161 approximately  $0.5^\circ$  for the EBSD analysis with the highest spatial resolution of  $1 \mu\text{m}$  [31].

162 To reveal etch pits and grooves, the samples were etched for 5 min. with the chemical agent Sirtl (HF(40  
163 %):CrO<sub>3</sub>(5M) = 1:1). The etching revealed grain boundaries and emerging dislocations that were observed

164 using an optical microscope. The etching was done twice: the first etching after the EBSD measurements  
165 with 7  $\mu\text{m}$  resolution and the second etching after the EBSD measurements with 1  $\mu\text{m}$  resolution.

### 166 3. Results

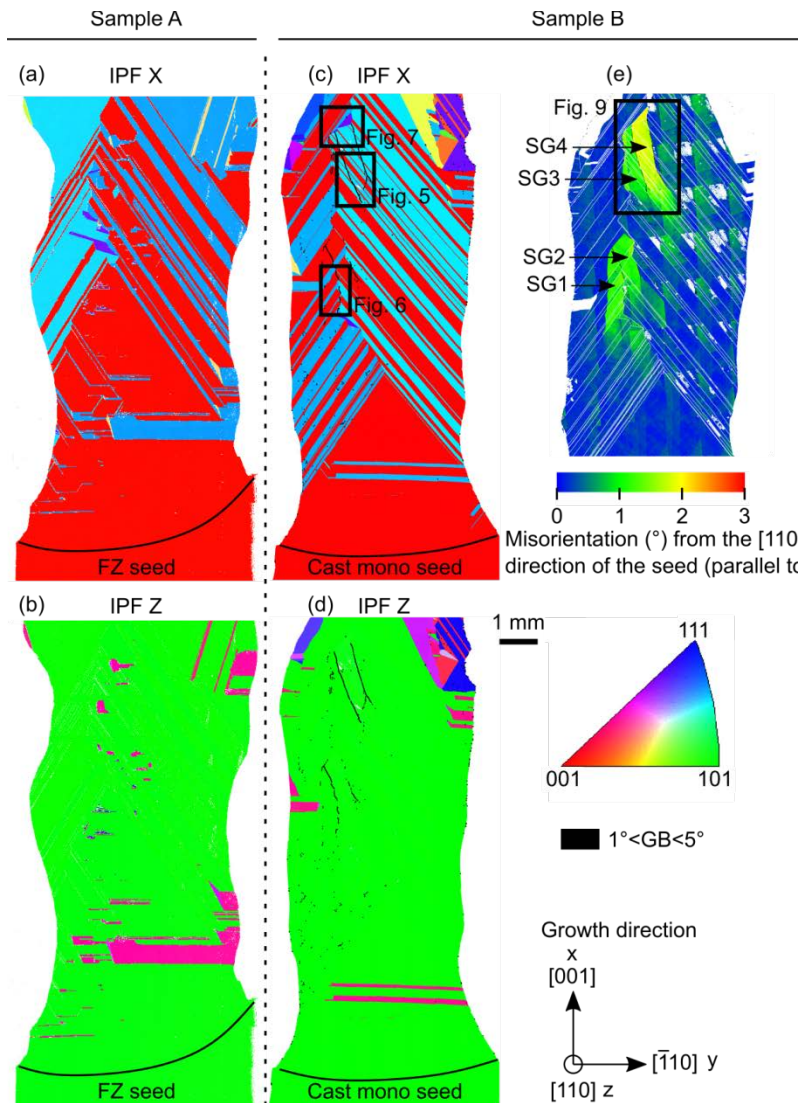
#### 167 3.1. Grain structure

168 The overall grain structures of samples A and B are dominated by diagonal successive twinning from  
169 both edges, represented in the IPF X images of Fig. 2(a) and (c), respectively.  $\Sigma 3$  twinning at the edges  
170 occurs because there exists a large undercooling [27] that facilitates the nucleation of twin grains [32]. The  
171 twin grains enter in competition at the center of the sample, though, the encounter is shifted towards the  
172 left side in both samples. From the radiographs (see Fig. 3(a) for sample A), it can be deduced that the twins  
173 nucleate earlier on the right side because the solid-liquid interface on the right side advances faster. This  
174 behavior was also reported and explained for this kind of experiments in [33]. The competition between the  
175 twins that propagate from both sides is accompanied by a significant amount of stress. This becomes clear  
176 by looking at the topography images (Fig. 3(b) and Fig. 4(a) and (b) for samples A and B, respectively),  
177 which show a very dark contrast area where the competition takes place. The dark contrast (stress) started  
178 to build up at the level of the initial solid(seed)-liquid interface. Additional dark contrast is observed on the  
179 left of Fig. 3(b), where several twinned grains nucleated as can be seen on the IPF X plot of Fig. 3(c).  
180 However, the higher stress (enhanced black contrast) is observed at the position of encounter and  
181 competition of the diagonal twins coming from the sides (Fig. 3(b)). It was also observed in our previous  
182 work [33] in comparable solidification conditions in a different sample and during another experimental  
183 campaign. In both samples the strain diminishes above in the crystal after the nucleation of a new grain  
184 (purple in IPF X map of Fig. 2(a) and (c)), which is assumed to nucleates inside a grain boundary groove  
185 created by the side twins in the sample center. It has a different crystallographic orientation than the side  
186 twins and the seed grains. Its growth direction is close to  $\langle 744 \rangle$ . In Fig. 3 and Fig. 4 these grains are  
187 encircled in black dotted lines. On top of them the global strain (dark contrast areas) reduces, as was  
188 previously observed in experiments with a FZ-seed of the same crystallographic orientation [33].

189 By looking at the IPF X and IPF Z images of both samples (Fig. 2(a)-(d)), it is apparent that the overall  
190 grain structure is the same. The [110] surface direction of the seed continues in the new grains almost over  
191 the entire sample height. The main difference in the structure of both samples is the development of high,  
192 up to  $3^\circ$ -misorientated subgrain domains in sample B, which are not present in sample A. The subgrain  
193 domains are detectable on the EBSD orientation maps because they have slightly different crystallographic  
194 orientations compared to the grain matrix. The black lines in Fig. 2(c) and (d) and the green areas in Fig.  
195 2(e) reveal the subgrains. Fig. 2(e) shows the degree of misorientation from the [110] direction of the seed  
196 crystal in different colors: dark blue means no misorientation with respect to the reference direction and  
197 green, yellow and red mean a misorientation of  $1^\circ$ ,  $2^\circ$  and  $3^\circ$  from the reference seed direction, respectively.  
198 The reference direction corresponds to a direction that is nearly parallel to the z-direction, i.e. normal to the  
199 sample surface.

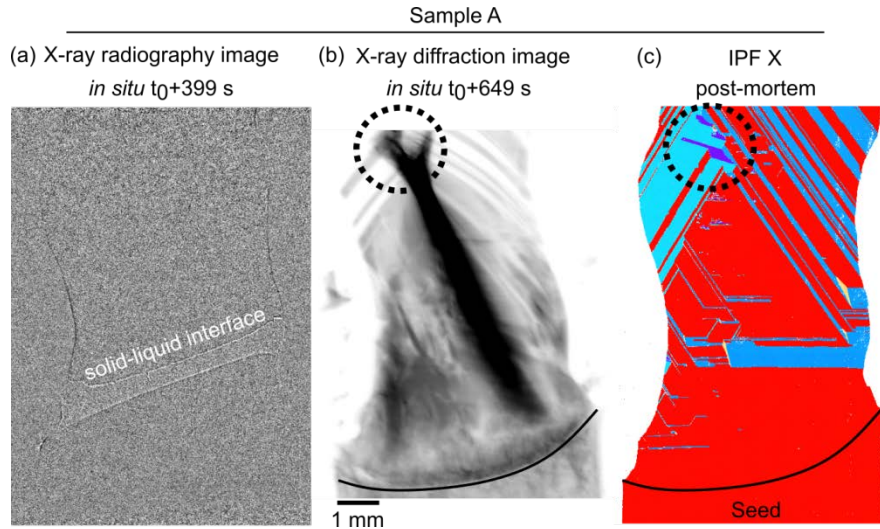
200 Two subgrain domains (1 and 2) are evidenced. Considering the detection limit, the first subgrain  
201 domain becomes visible ca. 5.8 mm above the seed-regrown silicon interface and consists of two subgrains  
202 (SG1 and SG2). The second subgrain domain becomes visible ca. 12.5 mm above the seed-regrown silicon  
203 interface and consists also of two subgrains (SG3 and SG4). The subgrains are finite and disappear again  
204 after a few millimeters.

205



206

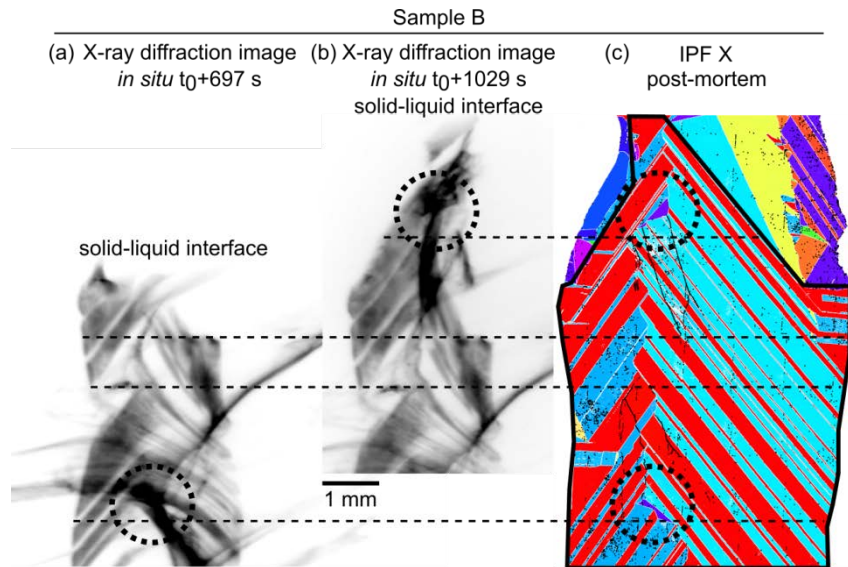
207 **Fig. 2** (a) and (c) IPF X and (b) and (d) IPF Z orientation maps obtained by EBSD measurements of the solidified  
 208 samples A (FZ-seeded) and B (cast mono-seeded), respectively. (e) The map shows the degree of misorientation from  
 209 the [110] direction of the seed crystal. In sample B, SGBs are observed that show misorientations up to 2° with respect  
 210 to the seed orientation (see also black-lines in (c) and (d)). The color gradations within the individual rectangles are  
 211 due to map stitching artefacts and due to deviations of the electron beam during large-scale map acquisition.



212

213 **Fig. 3** (a) *In situ* X-ray radiography image, (b) *in situ* X-ray topography image and (c) post-mortem IPF X orientation  
 214 map of the FZ-seeded sample A. The X-ray radiography image was post-processed to better visualize the solid-liquid  
 215 interface. The dark circle indicates a new grain nucleation (purple grain in (c)), which correlates to the reduction of  
 216 strain in (b).  $t_0 = 0$  corresponds to the start of solidification.

217



218

219 **Fig. 4** (c) IPF X orientation map with (a) and (b) corresponding to *in situ* X-ray diffraction snapshots at different  
 220 processing times of the cast mono-seeded sample B.  $t_0 = 0$  corresponds to the start of solidification. The red [001]  
 221 grains that lie in the black framed area of the orientation image are visible on the diffraction images. The circles  
 222 indicate the locations of new grain nucleations (purple grains) that contribute to recovery of lower strain levels in the  
 223 upper growing grains (reduction of the dark contrast area in the X-ray diffraction images). The dashed lines serve as  
 224 guides to the eyes to recognize corresponding regions.

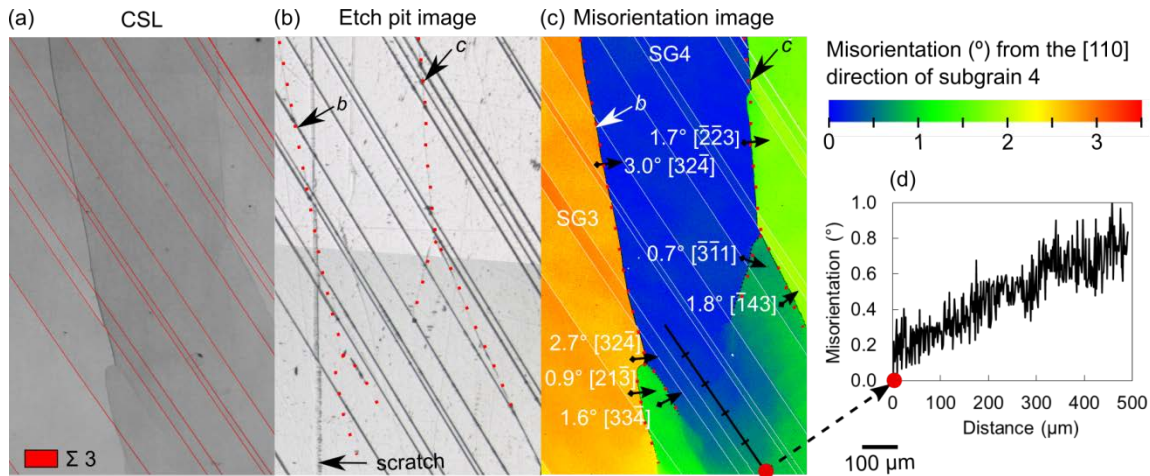
### 225 3.2. Formation of SGB domains

226 In Fig. 5 the formation area of subgrain 4 is shown. The CSL map (Fig. 5(a)) shows that the region  
 227 consists of diagonally arranged  $\Sigma 3$  twins. Figure 5(b) is an optical etch pit image on which the dark diagonal  
 228 lines represent the same twin boundaries. The red dotted lines show the traces of the SGBs that are only  
 229 weakly visible or in some places are not visible at all on the microscopic image after etching (Fig. 5(b)).



230 On the misorientation image of Fig. 5(c), the subgrains are clearly distinguishable because they exhibit  
 231 a misorientation up to 3°. The image shows the degree of misorientation from the [110] direction with  
 232 respect to subgrain 4. It can be seen that there is a misorientation of 3° between subgrains 3 and 4. The  
 233 misorientation between subgrain 4 and the grain matrix is 1.7°. Below the sample height at which the SGBs  
 234 become visible, a continuous increase of the misorientation can be measured along with the growth  
 235 direction (Fig. 5(c)). Furthermore, at the lower ends of the SGBs *b* and *c*, which delimit subgrain 4, two  
 236 SGB-branches initially merged into one SGB.

237 The misorientation angle of SGB *b* is the sum of the misorientation angles of the two initial branches.  
 238 A summation occurred because the two branches separate subgrains that have a similar misorientation axis  
 239 ( $[2\bar{1}3]$  compared to  $[3\bar{3}4]$ ). This supports the assumption that the two branches converged into one during  
 240 solidification to form a SGB with a higher misorientation. This suggests that the dislocations incorporated  
 241 in the SGB are of the same character. When the branches of SGB *c* merge, the misorientation is not the sum  
 242 of both. The reason is that the misorientation axis of both branches differ ( $[\bar{3}\bar{1}1]$  compared to  $[\bar{1}43]$ ). In this  
 243 case, the dislocations are probably of different character.  
 244



245  
 246 **Fig. 5** (a) CSL map, (b) etch pit image and (c) misorientation map of the formation area of subgrains 3 and 4. The  
 247 degree of misorientation from the [110] direction of subgrain 4 is shown. (d) The graph shows a continuous increase of  
 248 misorientation along the black line.

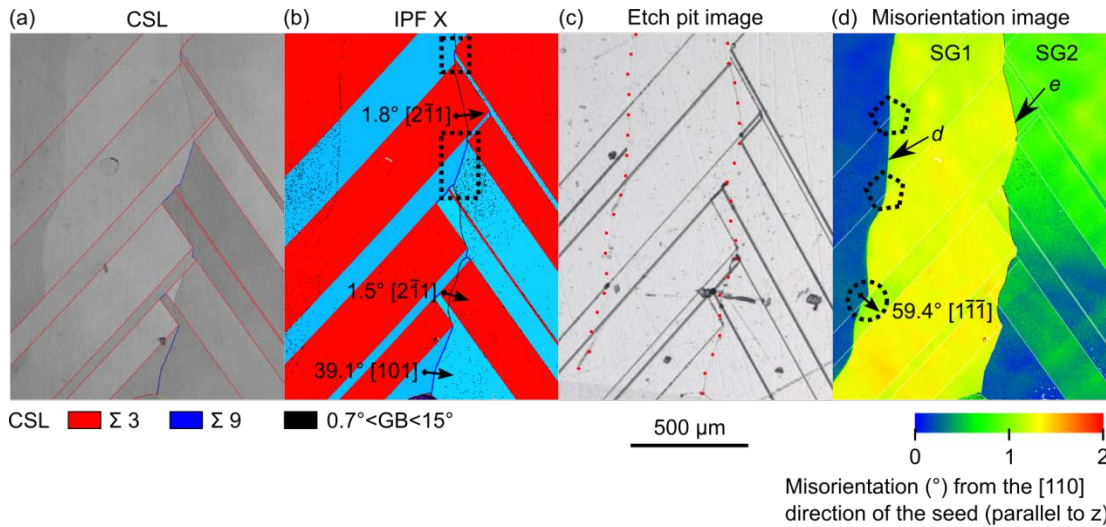
### 249 3.3. Propagation of SGBs

250 Once a SGB is formed, it evolves almost perpendicular to the solid-liquid interface. In Fig. 6 the  
 251 propagation of SGBs *d* and *e*, which separate subgrains 1 and 2, are shown. Figure 6(a) is the CSL map and  
 252 Fig. 6(b) an inverse pole figure map along the x-direction (corresponding to the growth direction). Figure  
 253 6(c) is an optical etch pit image, where the dotted red lines mark the trace of the SGBs observed with EBSD.  
 254 As was shown before, the SGBs are faintly visible on the etch pit image. Figure 6(d) shows the degree of  
 255 misorientation from the [110] direction with respect to the grain matrix.

256 SGB *d* follows the direction of growth and crosses  $\Sigma 3\{111\}$  GBs without producing noticeable changes  
 257 (see dotted pentagons in Fig. 6(c)). Along some short segments, SGBs also follow  $\Sigma 3\{111\}$  GBs (see dotted  
 258 circle in Fig. 6(c)). It causes the  $\Sigma 3$  GB $\{111\}$  to become an incoherent GB, which is expressed by the  
 259 misorientation angle that deviates from 60°.

260 SGB *e* is located in a region where grain competition among grains that grow from both sides takes  
 261 place. Either  $\Sigma 3$  or  $\Sigma 9$  GBs are formed depending on the twin relationship. A difference in the interaction  
 262 behavior between the SGB and  $\Sigma 3$  and  $\Sigma 9$  GBs can be seen. We find that SGBs never cross  $\Sigma 9$  GBs, but

263 follow them (see dotted rectangles in Fig. 6(b)). We also observe that the misorientation angle of SGB *e*  
 264 increases from 1.5° to 1.8° towards the top.  
 265



266  
 267 **Fig. 6** (a) CSL map, (b) IPF X orientation map, (c) etch pit image and (d) misorientation map showing the interaction  
 268 of SGBs with  $\Sigma 3$  and  $\Sigma 9$  grain boundaries.

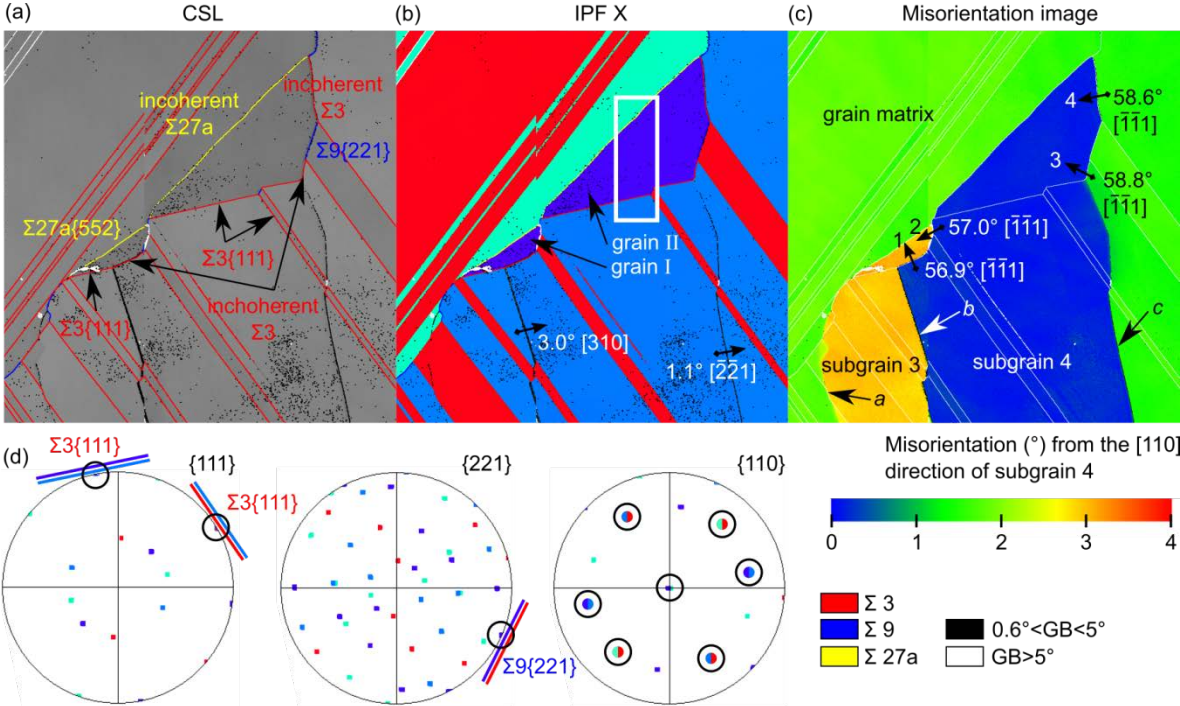
### 269 3.4. Disappearance of SGBs

270 The disappearance of both subgrains 3 and 4 is associated with the nucleation of new grains of the same  
 271  $\langle 744 \rangle$  type (grains I and II in Fig. 7). Nucleation of grains I and II takes place on  $\Sigma 3 \{111\}$  facets included  
 272 within the subgrains 3 and 4, respectively. This is why they have the same crystallographic misorientation  
 273 with respect to the matrix than the entire subgrain area (see Fig. 7(c)). The nucleation events of grains I and  
 274 II takes place in the subgrain areas 3 and 4 on the contrary to all other previous nucleation occurring during  
 275 growth. ~~It is the first time during growth that new grains nucleate inside the subgrain area.~~ Before these  
 276 nucleation events, the subgrains ~~increased their size~~ grew only by an upward propagation crossing grains  
 277 that nucleated far away from the subgrain area (at the sides of the sample). Although grain nucleation can  
 278 be triggered by the presence of dislocations and/or deformed areas [27, 33] and by extension possibly by  
 279 the presence of SGBs, it is not possible to conclude on this from our experiments. Moreover, it is worth  
 280 noting that the same kind of nucleation events (same crystallographic orientation and position) was  
 281 observed in sample A in which SGBs were absent.

282 In order to understand why the subgrains disappear, it is necessary to have a detailed look at the  
 283 development of the three SGBs *a*, *b* and *c*, which delimit subgrains 3 and 4 laterally. SGB *a* stops at the  
 284 encounter of grain I. SGBs *b* and *c* meet the new grains I and II at a  $\Sigma 3 \{111\}$  GB. In both cases, they do  
 285 not cross the GB but propagate along the GB inducing a direction change.

286 The  $\Sigma 3$  GB is modified from a coherent  $\Sigma 3 \{111\}$  to an incoherent  $\Sigma 3 \{111\}$  GB. This is reflected in the  
 287 misorientation angles of 56.9° (arrow 1 in Fig. 7(c)) and 58.8° (arrow 3 in Fig. 7(c)) between grains I and  
 288 II and their neighbor grains, respectively, to be compared to the value of 60° expected for a perfect  $\Sigma 3$  GB.  
 289 These grains have a  $\langle 744 \rangle$  orientation close to the growth direction. The misorientation of the incoherent  
 290  $\Sigma 3$  GB below grain I is higher than that on the right side of grain II. This is due to the fact that the  
 291 misorientation of SGB *b* is higher, too. Following the grain boundaries to the upper right, the misorientation  
 292 remains unchanged. Interrupted by  $\Sigma 9$  GBs, the next incoherent  $\Sigma 3$  GBs have similar misorientations of  
 293 57.0° (arrow 2 in Fig. 7(c)) and 58.6° (arrow 4 in Fig. 7(c)), respectively. This is consistent with the  
 294 misorientations of the SGBs measured before (3.0° and 1.1° for *b* and *c*, respectively). Then, the SGBs

295 encounter a triple junction between an incoherent  $\Sigma 3$ ,  $\Sigma 9$  and a  $\Sigma 27a$  GB. In fact, it is the first time during  
 296 their upward propagation that the SGBs meet a  $\Sigma 27a$  GB. After the encounter they do not continue their  
 297 way in the direction of growth. The formation of the  $\Sigma 27a$  GBs is associated with the nucleation of the new  
 298 grains I and II that form a  $\Sigma 27a$  GB on top. Since  $\Sigma 27a$  GBs are also observed in sample A, which has no  
 299 subgrains, these  $\Sigma 27a$  GBs are only the result of grain competition.  
 300



301  
 302 **Fig. 7** (a) CSL map, (b) IPF X orientation map, (c) misorientation map and (d)  $\{111\}$ ,  $\{221\}$  and  $\{110\}$  pole figures  
 303 of the grains lying inside the white rectangle of map (b). The dark circles encircle common crystallographic directions.

304 **3.5. Dislocations in the seed crystals**

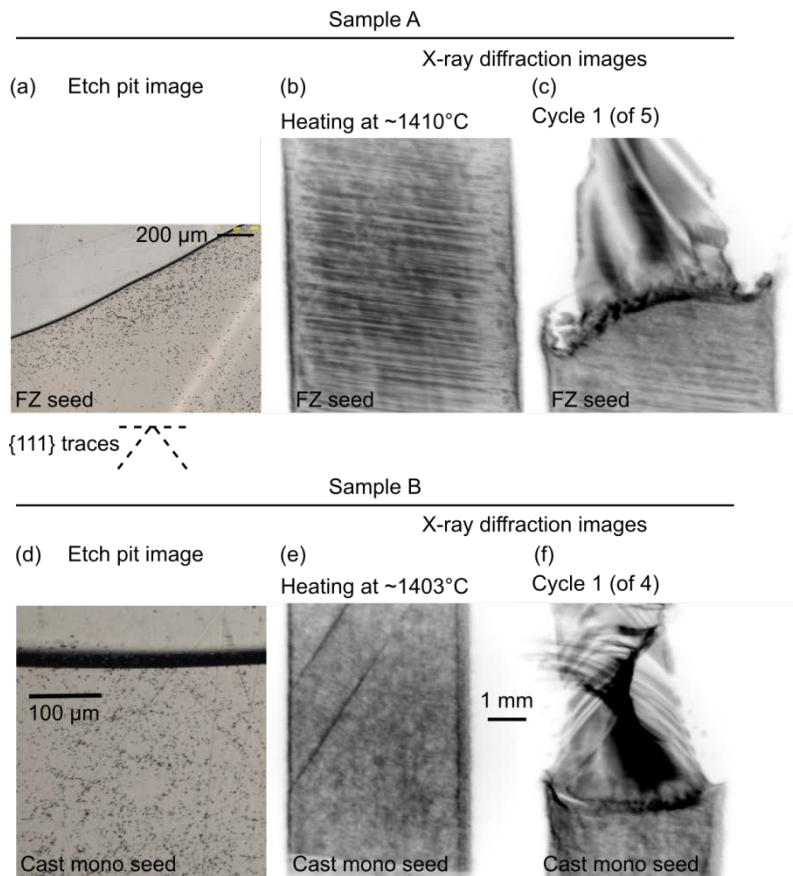
305 Dislocation distributions were observed by etching and by X-ray diffraction imaging. Dislocation  
 306 densities measured by etch pit counts only show dislocations that emerge at the surface. Dislocations that  
 307 stay in planes parallel to the sample surface are not revealed. In the diffraction images, dislocations are not  
 308 visible if they fulfill the extinction criterion  $\mathbf{b} \cdot \mathbf{g} = 0$  for the particular diffraction spot observed ( $\mathbf{b}$  is the  
 309 Burgers vector and  $\mathbf{g}$  is the diffraction vector). Therefore, both techniques do not represent the total amount  
 310 of dislocations, but give a qualitative indication of the amount.

311 Figures 8(a) and (d) show etch pit images of samples A and B at the position of the seed-regrown  
 312 interface. In Fig. 8(b) and (e) *in situ* diffraction images of both samples are presented that show the seed  
 313 crystals shortly before melting. The etch pits on the cast mono seed (sample B) form an array of dislocation  
 314 cells. In the FZ seed the dislocation are not arranged in cells and are more widely spread. The dislocations  
 315 are arranged in lines, which follow the traces of two crossing  $\{111\}$  family planes that are oriented  
 316 perpendicular to the surface. Finally, the dislocation density is higher in sample B-seed compared to sample  
 317 A-seed.

318 The diffraction images confirm the observation from the etch pits images. The cast mono seed shows a  
 319 dense network of dislocations arranged in cells compared to the FZ-seed. The time sequence of the *in situ*  
 320 X-ray images reveal that the dislocations are not very mobile during heating and up to the melting point in  
 321 sample B compared to sample A (videos that show the heating phases of sample A and B are available as

322 supplementary material). The two diagonal dark lines are probably micro-twins. In the FZ seed the  
 323 dislocations originating from sources at the sample edges are very mobile up to the silicon melting point  
 324 and move on different {111} planes. The almost horizontal lines form because dislocations propagate on  
 325 the [111]/[111] and [111]/[111] planes, whose projection traces at the surface plane are horizontal. As these  
 326 planes have an angle of 35.3° with the surface {110} planes, the movement of the dislocations on the planes  
 327 can be nicely observed on the projected images. The propagation of dislocations along these planes is  
 328 activated by the pressure exerted by the crucible on the main surface sides [34]. The reason for the slight  
 329 inclination of the “horizontal” lines by 8° is due to a geometric distortion during recording, which is  
 330 explained in [29]. Activation of the two {111} planes that are perpendicular to the surface can only be seen  
 331 close to the melting point. These are the diagonal lines ({111} traces) that are also observed on the etch pit  
 332 image.

333 What both samples have in common is that the etch pit density significantly reduces from the non-  
 334 molten seed to the new grown crystal. In Fig. 8(c) and (f), topography images of samples A and B shortly  
 335 after the first solidification cycle are shown. The seed-regrown interface is very dark and therefore distorted.  
 336 Inside the new grown grains dark areas are present as well. Although individual dislocations are poorly  
 337 visible, one can see that the dislocation structure is quite different in the new grown crystals: neither  
 338 horizontal dislocation lines for sample A, nor dislocation cells for sample B are visible.  
 339



340  
 341 **Fig. 8** (a) and (d) post-mortem etch pit images of the initial solid-liquid interface of the last cooling cycle of samples  
 342 A and B, respectively. (b) and (e) X-ray diffraction images of the unmolten seeds of samples A and B, respectively,  
 343 shortly before melting. (c) and (f) X-ray diffraction images after the first cooling cycles of samples A and B,  
 344 respectively, that show the seeds and the regrown crystals and their different dislocation arrangements.

345

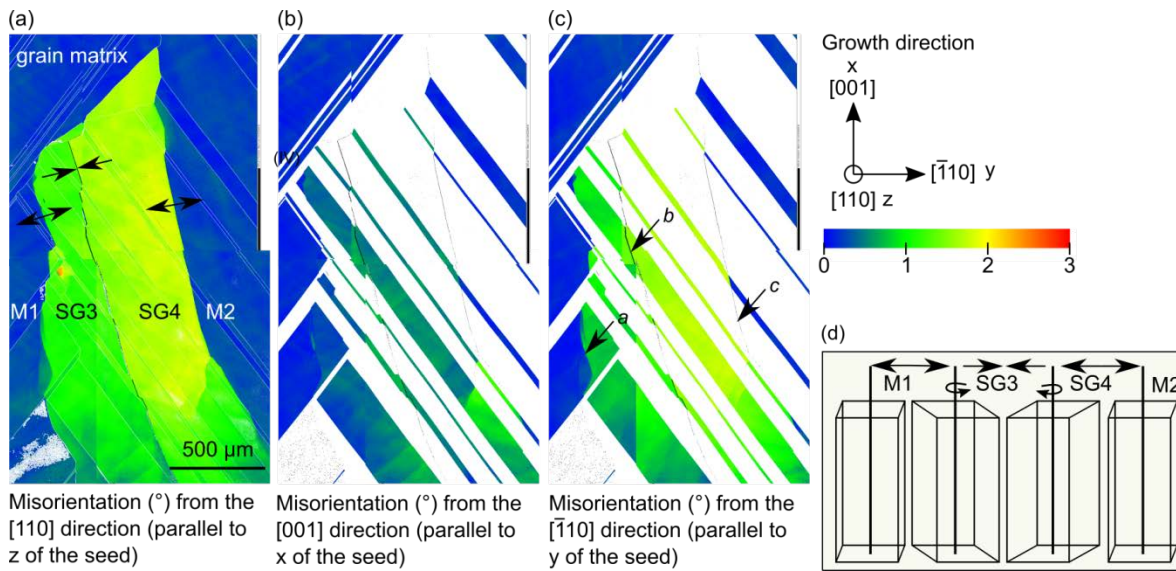
346 **4. Discussion**

347 *4.1. SBG structure*

348 *4.1.1. Orientation relationship of the subgrains*

349 The SGBs in sample B are rotated towards each other mainly by a rotation around the [001] axis, i.e.  
 350 the growth direction. This is illustrated in Fig. 9, where the degrees of misorientation from the [110], the  
 351 [001] and the  $\bar{1}10$  directions with respect to the seed orientation are shown from (a) to (c), respectively.  
 352 In the x- and y-representations only the grains that have an orientation close to the original orientation of  
 353 the seed can be seen in color because the other grains have a higher misorientation than  $3^\circ$ . The subgrains  
 354 show a low misorientation along the x-direction but a similar high misorientation along the y- and z-  
 355 directions consistent with tilt GBs. In Fig. 9(d) the inclination of the subgrains 3 and 4 relative to the grain  
 356 matrix on both sides is schematically shown.

357 The fact that almost no etch pits are visible along the SGBs means that the dislocations do not emerge  
 358 at the surface. Hence, it supports the point that the dislocation lines of the SGBs are mainly oriented along  
 359 the growth direction. Since the rotation axis is principally along the growth direction, many dislocations  
 360 have an edge character. This is consistent with the analysis of Lantreibecq et al. [18] who found that the  
 361 SGBs in the source cast mono ingot are mainly composed of dislocations with a [001] line character and a  
 362 Burgers vector  $a/2[1\bar{1}0]$ . They proposed that these dislocations grow by epitaxy on the solid-liquid  
 363 interface. They also found two other families of dislocations that have Burgers vectors at  $45^\circ$  from the  
 364 growth axis. These dislocations could be responsible for deviations of the tilt direction of the subgrains also  
 365 observed in this study.  
 366



367 **Fig. 9** Misorientation maps show the degree of misorientation from the (a) [110], (b) [001] and (c)  $\bar{1}10$  direction  
 368 with respect to the seed orientation. (d) Schematic representaiton of the tilt directions of the two subgrains SG3 and  
 369 SG4 and the matrix grains on the left and right side. SGB3 and SG4 are inclined towards each other.  
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#### 4.1.2. SGB misorientations and their dislocation density

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For SGB  $b$  we observe a continuous increase of the misorientation angle in the direction of growth, which is consistent with an accumulation of dislocations in a dislocation wall. This is also supported by our observation that lower misoriented branches merge to form a higher misoriented SGB in the direction of growth (cf. Fig. 5). A similar branched structure was earlier reported by Chuang et al. who observed SGB formation in silicon *in situ* suggesting that dislocations aggregate at the solid-liquid interface and increase the misorientation angle through the continual incorporation of new dislocations [2]. The branched dislocation arrays that merge into one SGB, the increase of the SGB misorientation and their principle orientation and tilt along the growth axis suggest a SGB formation during growth. SGBs aligned along the growth direction in mc-ingots have also been reported by other authors, who also assumed a generation of dislocations during growth at the solid-liquid interface [8, 13].

The distance between individual dislocations  $d$  assuming that the SGB is constituted of perfect edge dislocations aligned along the SGB can be evaluated by

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$$d = \mathbf{b}/\sin(\theta).$$

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Using  $\mathbf{b} = 0.384$  nm for the Burgers' vector magnitude in silicon along a  $\langle 110 \rangle$  direction, the distance between each dislocation is  $d_1 = 7.3$  nm and  $d_2 = 22$  nm for  $\theta$  of  $3^\circ$  and  $1^\circ$ , respectively. These misorientation angles are in the same order of magnitude as what was observed by Lantreibecq et al. in the source cast mono ingot at a height of about 160 mm from the melt-back surface [18], which is above the area where electrically active subgrains do appear on the PL maps (cf. Fig. 1). The rotation axis is also similar.

The spacing of the dislocations can be used to estimate the number of dislocations within the SGBs. With a sample thickness  $t = 3 \times 10^{-2}$  cm and a dislocation spacing in the order of  $d_1 = 1 \times 10^{-6}$  cm,  $3 \times 10^4$  dislocations exist within SGB  $b$  assuming a Burgers vector of 0.384 nm standard for hypothetical edge dislocations aligned vertically constituting the boundary. Accordingly, approximately  $1.5 \times 10^4$  dislocations exist in SGB  $a$  and  $c$ , respectively. This makes a total of  $6 \times 10^4$  dislocations that should intersect a horizontal virtual surface to create the observed boundary misorientations.

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#### 4.2. SGB origin and implications of the crystal quality of the seeds

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Oriwol et al. [12] performed a detailed study on the origin of dislocation clusters and found that 97% originate at grain boundaries. Different from this report and from other studies [e.g. 9, 35], SGBs start in the bulk crystal and not at a specific GB in our experiment. One reason for the high number of dislocations in sample B could be that dislocations propagate from the seed in the regrown crystal. Dislocation density was measured in the range  $1 \times 10^4$  cm<sup>-2</sup> to  $3 \times 10^4$  cm<sup>-2</sup> in another brick of the source ingot at a similar height to the one at which the sample was extracted for our experiments. Such a dislocation density results in a maximum of 540 dislocations crossing each sample horizontal surface. Compared to a number of  $6 \times 10^4$  dislocations for the regrown part of sample B, the density in the seed crystal is too low to be the origin of all dislocations associated with the subgrains. Another argument against the idea that all dislocations originate from the seed is that the etch pit structure changes above the seed-regrown interface, which indicates that there is either a reduction of the total number of dislocations or a change in dislocation character and orientation or both. Hence, there are not enough dislocations in the seed to create the observed misorientations and dislocation multiplication and not only accumulation must have taken place in the regrown part.

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The question arises about the reason for the dislocation generation in sample B and not in sample A. That FZ-seeded samples do not develop subgrains has been confirmed several times by similar experiments using FZ seeds [33, 36]. When considering the two seeds A and B, it is obvious that a main difference lies in the initial dislocation density and structuration. The initial dislocation network in the cast mono seed at

416 the melting point shows a low mobility and is arranged in a cellular structure, whereas the dislocation  
417 network in the FZ seed is very mobile and aligned on  $\{111\}$  glide planes.

418 In comparison to the source cast mono ingot of sample B, very similar subgrains are observed indicating  
419 a similar origin. Since sample B was taken from a domain just below SGB multiplication started in the  
420 source ingot, we can conclude that subgrain formation occurs at a similar height in both cases. As only the  
421 cellular dislocation structure and microtwins are initially characterized in sample B (see Fig. 8(e)), one of  
422 those defects might carry some deformation in relation with the SGB generation. Additionally, as  
423 microtwins are not present anymore after our melting step, we hypothesize that the origin of the SGB  
424 development could be due to the cellular dislocation structure present in the seed.

425 Besides the dislocation structure, another difference is the chemistry of the cast mono compared to the  
426 FZ seed. The oxygen concentration in both seeds is very low, but the cast mono seed contains more carbon  
427 than the FZ seed. However, the sample is not oversaturated in carbon so that only a small number of  
428 precipitates should be present. It was reported that precipitates in the source cast mono seed do not  
429 immobilize dislocations, but that they can create dislocation cross-slip, if they are exposed to sufficient  
430 stress [18]. A strong influence of C- and N-based impurities on subgrain formation is therefore not  
431 suspected. In addition, both samples have boron concentrations  $>10^{17}$  at.  $\text{cm}^{-3}$  due to contact with the boron  
432 nitride crucible. Since both samples are equally exposed to the boron nitride crucible, a different influence  
433 behavior of boron on the formation of subgrains is improbable.

434 The clearly different dislocation structures of the two seeds A and B indicates that it is probably the  
435 decisive factor causing the formation of subgrains. Unfortunately, it is not possible to give more information  
436 on the origin of the subgrains, because subgrains with smaller misorientations that probably exist in the  
437 lower part of the sample as it was reported in [18] could not be detected during this study due to  
438 experimental limitations. In a next step, the recording multiple diffraction spots providing complementary  
439 information is foreseen to clarify further details about the generation of subgrains.

#### 440 4.3. SGB propagation and termination

441 The grain orientation relationship and the GB character play an important role for the propagation of  
442 the SGBs. Once the SGBs are generated, they cannot leave the sample because they grow with the solid-  
443 liquid interface and are arranged parallel to the growth direction. When meeting a grain boundary, the  
444 existence of a common slip system on both sides of the grain boundary is necessary for dislocations to cross  
445 the twin boundary.  $\Sigma 3\{111\}$  GBs that separate  $\{110\}\langle 001\rangle$  (red) and  $\{110\}\langle 221\rangle$  (blue) oriented grains  
446 are crossed by the SGBs without inducing any visible changes (colors refer to the IPFs).  $\Sigma 3\{111\}$  GBs that  
447 separate  $\{110\}\langle 221\rangle$  (blue) and  $\{110\}\langle 744\rangle$  (purple) oriented grains are not crossed, but change the  
448 direction of the SGBs. SGBs propagate rather along the  $\Sigma 3$  and  $\Sigma 9$  GBs limiting the purple grains than  
449 through the purple grains. This is plausible as dislocations can be absorbed in a GB, splitting into grain  
450 boundary dislocations (GBD) with smaller Burgers vectors, and move in the boundary [37, 38]. We observe  
451 that  $\Sigma 3\{111\}$  GBs become incoherent GBs when SGBs merge with them and follow them. This observation  
452 is different from the *in situ* observations of Chuang et al., who detected only cases where SGBs crossed  
453  $\Sigma 3\{111\}$  GBs that remain coherent according to EBSD detection limits [39].

454 Notably, the orientation relationship of the blue and purple grains is not favorable for the dislocations  
455 in the SGB to cross. It was reported by Schmid et al. that a common slip system is not enough to explain  
456 the correlation between dislocations and grain boundaries, but that the structure of the grain boundaries has  
457 to be taken into account [40]. From a macroscopic point of view, there is no difference in the character of  
458 the crossed and uncrossed GBs. From a microscopic point of view, the GB structure is unknown and TEM  
459 measurements would be required to obtain this information. In addition, it depends on the local stress and  
460 on the type of dislocation whether it can pass through a grain boundary [41, 42]. It has been observed [41]  
461 and simulated [43] that even  $\Sigma 3\{111\}$  GBs can act as accumulation obstacles for dislocations. The

462 stereographic projection in Fig. 7(d) shows that the common  $\langle 110 \rangle$  direction of the red/blue grains is  
463 roughly oriented in the direction of the SGB path. The common  $\langle 110 \rangle$  direction of the blue/purple grains,  
464 however, is almost perpendicular to the SGB direction, i.e. parallel to the solid-liquid interface. The highest  
465 probability of crossing for dislocations and thus for dislocations arranged in a SGB is obtained if there is a  
466 common glide plane (common  $\langle 110 \rangle$  direction) if other conditions of stress and dislocation character are  
467 fulfilled. Consequently, as the growth direction is forced upon the grains by the temperature gradient, the  
468 unsuitable orientation of the common  $\langle 110 \rangle$  direction might be the reason for the SGB change, especially  
469 if there is no Burgers vector common to both slip systems.

470 Above the purple grains, the SGBs do not continue their way. Either dislocations rearrange at the level  
471 of high disorder twin boundaries, like  $\Sigma 27a$  GBs that limit the purple grains on top and emit new  
472 dislocations, or the dislocations leave the sample by cross slipping at the  $\Sigma 3$  or  $\Sigma 9$  GBs.  $\Sigma 27a$  GBs have a  
473 higher boundary energy than  $\Sigma 3$  and  $\Sigma 9$  GBs and were found to be the source of new dislocations [13, 14,  
474 35] probably as the result of atomic faceting [44, 45]. We do not observe any new dislocation emissions  
475 originating at these  $\Sigma 27a$  GBs, but first, the *in situ* X-ray diffraction image is very dark at this position,  
476 which conceals the observation of dislocations, and second, new generated dislocations could grow out of  
477 the sample surface due to the thin sample geometry.

478 It is also important to note that these purple grains were identified as the grains whose nucleation  
479 releases strain previously accumulated as a result of grain competition when no SGBs are observed [33].  
480 The new grain nucleation leads to a subsequent grain structure reorganization, which contributes to a  
481 reduction of the strain in the growing ingot. Although the disappearance of the subgrain domain is probably  
482 affected by the thin sample geometry, it means that the purple grains have an unambiguous influence on  
483 the SGB evolution. In a large-scale casting it could be a mechanism to stop or spread SGBs.

484

## 485 5. Conclusion

486 *In situ* growth experiments and *ex situ* analyses of two seeded wafers were performed. The initial seed  
487 orientation along the growth direction was [001] for both the FZ and the cast mono seeds. Several  
488 millimeters long subgrain boundaries, elongated in the growth direction and crossing a number of twin  
489 boundaries are formed in the cast mono-seeded sample, but not in the FZ-seeded sample. The subgrains  
490 have a main rotation axis along the [001] growth direction and increase their misorientation by merging  
491 with new SGBs appearing in their vicinity.

492 *In situ* X-ray topography imaging during the heating and solidification experiments showed that the  
493 dislocations are very mobile in the FZ seed and propagate on  $\{111\}$  slip planes, whereas the dislocations in  
494 the cast mono seed are less mobile and are arranged in cells. The significantly smaller number of  
495 dislocations in the cast mono seed compared to the number of dislocations contained in the SGBs shows  
496 that dislocation multiplication mechanisms must have taken place in the regrown part. Although the source  
497 of the dislocations could not be revealed, the observations suggest that subgrain formation is related to the  
498 initial cellular dislocation structure in the cast mono seed.

499 Once formed, the SGBs either follow  $\Sigma 9\{221\}$  and  $\Sigma 3\{111\}$  GBs or cross  $\Sigma 3\{111\}$  GBs. In the situation  
500 where SGBs follow twin boundaries, the coherency of the  $\Sigma 3\{111\}$  GBs is degraded due to the presence of  
501 the SGBs. A particular situation is observed where a  $\Sigma 3\{111\}$  GB that separates  $\langle 221 \rangle$  and  $\langle 744 \rangle$  oriented  
502 grains constitutes an obstacle and is not crossed by the SGBs. Whether the dislocations stop or cross-slip  
503 cannot be determined, because the sample is very thin and a change in direction of the dislocation lines  
504 would cause the dislocations to escape at the surface. However, in either of these cases the vertical SGB  
505 propagation in the growth direction is disturbed. Transferring this finding to a real size casting, the  
506 nucleation of  $\langle 744 \rangle$  oriented grains that are known to release strain could be responsible for the stopping  
507 or lateral spreading of SGBs.



508  
509

### Credit author statement

510 **Maïke Becker:** Conceptualization, Investigation, Writing - Original Draft, Visualization, Funding  
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512 **Guillon:** Methodology, Investigation, Resources, Data Curation, Writing - Review & Editing  
513 **Laurent Barrallier:** Methodology, Resources, Writing - Review & Editing **Gabrielle Regula:**  
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515 Investigation **Guillaume Reinhart:** Investigation, Methodology **Nathalie Mangelinck-Noël:**  
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517 Project administration, Funding acquisition

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