Investigation of Facetted Growth in Heavily Doped Silicon Crystals Grown in Mirror Furnaces

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Abstract: Herein, facets and related phenomena are studied for silicon crystals grown in the <100> and <111> directions, using the Zone Melting and Floating Zone techniques. Investigating the central facets of dislocation-free <111> crystals as a baseline allowed for the determination of the local temperature gradients. When comparing these results to dislocated <111> crystals, the presence of dislocations caused a clear reduction in the facet size, correlated with a reduction in the required local supercooling to estimated maximum values of around 0.6 K. Furthermore, for crystals grown on the rough {100} interface, attempts to provoke a morphological instability of the local phase boundary succeeded for crystallization velocities in the range of 10–16 mm/min, in good agreement with theory. Contrary to this observation, crystals grown in the <111> direction remained morphologically stable even at higher crystallization velocities due to the stabilizing effect of the atomically smooth interface. Additionally, crystals grown in the <111> direction with an oxygen skin by the Zone Melting technique reproducibly showed a non-periodic fluctuation of the central facet diameter at a certain translation velocity.

Keywords: silicon; floating zone; zone melting; facet; dislocations; growth kinetics

1. Introduction

While phenomena related to facets are a very common subject of studies in the field of oxides [1,2] and are a major concern for the growth of III-V materials (see, e.g., [3–7]), investigations on the impact of facets in silicon are rather sparse. The main activities in this field date back to the late 1960s to 1980s, including, for example, the exhausting work of Voronkov [8–12] and Brice [13]. Other activities deal with the correlation between twinning and facets in solar silicon [14,15], while recently some interest in edge facets in Czochralski and Floating Zone silicon has evolved [16–20]. Nevertheless, only very few publications can be found on the interplay of facets and defects such as dislocations [11,21,22] or the occurrence of morphological instability [1,23].

The aim of the present work was to study faceted growth in silicon and its related effects. For that purpose, we grew heavily As-doped silicon crystals with a diameter of 8 mm along the <100> and <111> crystal directions, using the Floating Zone (FZ) and Zone Melting (ZM) techniques. We investigated the behavior of the facets in dependence on the growth velocity and the presence of dislocations, and also studied the onset of morphological instability for growth on rough and atomically smooth interfaces.
2. Some Theoretical Considerations

2.1. Jackson Factor

It has been well known since the pioneering work of Kossel [24] and Stranski [25] that growth kinetics differ for atomically smooth and atomically rough crystal faces. The Jackson factor $\alpha$ was introduced to classify whether a crystal plane is atomically rough or smooth [26].

$$\alpha = \frac{\Delta S}{k} \xi_{hkl}$$  \hspace{1cm} (1)

$\Delta S$, $k$, and $\xi_{hkl}$ are the melting entropy, the Boltzmann constant, and a factor which is given by the number of bonds of growth units in the plane $\{hkl\}$ divided by the number of bonds of growth units in the crystal. In general, the interface is considered rough for $\alpha < 2$ and smooth for $\alpha > 2$.

In Table 1, the Jackson factor $\alpha$ of some semiconductors at their melting temperature is compiled for $\{100\}$ and $\{111\}$ crystal planes. In general, the $\{111\}$ planes of crystals with diamond and zincblende structures are atomically smooth, whereas most other faces such as the $\{100\}$ are atomically rough.

| Material | $\Delta S/k$ | $\eta/Z \{100\}$ | $\eta/Z \{111\}$ | $\alpha \{100\}$ | $\alpha \{111\}$ |
|----------|--------------|-------------------|-------------------|----------------|----------------|
| Si       | 3.6          | 1/2               | 3/4               | 1.8 (r)        | 2.7 (s)        |
| GaAs     | 7.4          | 1/2               | 3/4               | 3.7 (r)        | 5.6 (s)        |
| InP      | 5.6          | 1/2               | 3/4               | 2.8 (r)        | 4.2 (s)        |
| CdTe     | 4.4          | 1/2               | 3/4               | 2.2 (r)        | 3.3 (s)        |

2.2. Growth Laws

For an atomically rough interface, the normal growth rate $V$ is, in general, directly proportional to the supercooling $\Delta T$.

$$V \left[ \frac{m}{s} \right] = \beta \cdot \Delta T$$  \hspace{1cm} (2)

The proportionality factor $\beta$ is the so-called kinetic coefficient, which is about 0.5 m/sec K in the case of silicon [26]. The supercooling $\Delta T$ required for a rough interface to grow is in the range of $10^{-3}$ K [26]. Therefore, the shape and position of the solid–liquid interface for most crystal faces can be safely considered to be identical to the melting point isotherm $T_m$.

However, for an atomically smooth interface, such as the (111) faces in silicon (called facets), the growth mechanism is different. For the growth of a perfectly atomically smooth interface, a two-dimensional nucleus must form on the interface which then grows laterally along the (111) plane until the layer is completed. This process repeats periodically. In this case, the normal growth rate $V$ depends exponentially on the supercooling $\Delta T$ as expressed, e.g., by the following equation according to Voronkov [8,27]:

$$V \left[ \frac{m}{s} \right] = 0.63 \cdot (\Delta T)^{5/6} \exp \left( -\frac{42}{\Delta T} \right) \left( 1.66 \cdot 10^{-4} \cdot (1683 - \Delta T)^2 \right)^{1/6}$$  \hspace{1cm} (3)

The supercooling $\Delta T$ required for a stable 2D nucleus to form on a dislocation-free silicon (111) facet is 3.7 K according to Voronkov [9].

When dislocations whose Burgers vector is inclined towards the facet plane intersect with the atomically smooth face, steps will form on the facet plane. Therefore, a two-dimensional nucleus is no longer necessary for the facet to grow. Now, the growth rate $V$ depends quadratically on $\Delta T$, as proposed again by Voronkov [9].

$$V \left[ \frac{m}{s} \right] = 3 \cdot 10^{-4} \cdot \Delta T^2$$  \hspace{1cm} (4)
The supercooling $\Delta T$ theoretically required such that a (111) facet with dislocations will grow is reported to be $0.7 \, \text{K}$ [10].

The growth laws for the three cases according to Equations (2)–(4) are illustrated in Figure 1.

![Figure 1. Growth rate $V$ as a function of the supercooling $\Delta T$ for a rough interface (Equation (2), black), atomically smooth, dislocation-free (111) facet (Equation (3), blue), and a (111) facet with dislocations (Equation (4), red) for silicon together with some experimental data compiled in [27]. The supercooling $\Delta T$ determined in this work (see Section 4) for a faceted interface with dislocation is shown as a red square.](image)

### 2.3. Crystallographic Aspects

The occurrence of faceted growth during crystallization depends on the crystal structure and the thermal conditions. In the case of the growth of a <100>-oriented Si crystal, four (111) faces exist in the four <110> directions. However, only at the periphery of the crystal the conditions are typically fulfilled such that a supercooling $\Delta T$ of the melt in the vicinity of the (111)-planes can occur. Then, the so-called edge facets will form at the crystal periphery, which are inclined with a facet angle $\phi_{100}$ of $35^\circ$ from the vertical direction—in this case shown schematically in Figure 2. Due to their specific growth mechanism, the edge facets are visible on the outer crystal surface as protrusions which are also called growth ridges [19,20].

In <111>-oriented crystals, there are two sets of three (111) edge facets/growth ridges at the crystal periphery along the <211> direction. In this case, the edge facet is inclined with a facet angle $\phi_{111}$ of $20^\circ$ from the vertical direction. As the edge facet angle is different for the <100> and <111> crystal orientations, this will directly affect the geometry of the growth ridge because the growth ridge width $w$, which is visible on the crystal surface, is indirectly proportional to the $\tan \phi$ [20]. Assuming nominally the same thermal conditions during the growth of the <100> and <111> silicon crystals (Section 4.2), we expect therefore that the width of the growth ridge $w_{100}$ should be larger than the $w_{111}$ because $\phi_{100}$ is larger than $\phi_{111}$.
Figure 2. Upper part: Schematic representation of interface shapes during growth of Si by the Floating Zone or Zone Melting technique. (Note: For Cz configuration, the growth direction is opposite, and the whole sketch has to be mirrored at the horizontal axis). (left) Convex shape and rough interface for the <100> crystal orientation; (right) convex shape and central facet with a facet length $d$ for the <111> crystal orientation. The edge facets which form at the triple point (TPL) at the periphery of the crystal are shown for both cases. The distance $\Delta x$ of the central facet to the curved melting point isotherm $T_m$ at the symmetry axis is a measure of the maximum supercooling $\Delta T$.

Lower part: cross sections of the Si crystals perpendicular to the growth direction. The edge facets are visible as protrusions on the crystal surface along the <011> direction for the <100> crystal orientation, and along the <211> direction for the <111> crystal orientation. The blue dotted line marks the target cross section for the sample preparation in this work.

In addition, in the case of a <111>-oriented crystal, a central (111) facet can easily form if the interface is curved with a convex shape towards the melt as shown in Figure 2. Analogous to the model of Brice [13], the axial temperature gradient $G$ can be determined from the distance $\Delta x$ between the melting point isotherm $T_m$ to the central facet at the symmetry axis, where the maximum supercooling $\Delta T$ is reached:

$$G = \frac{\Delta T}{\Delta x} \quad (5)$$

Thereby, the shape of the melting point isotherm in the center of the crystal can be determined by extrapolation of the solid–liquid interface from the atomically rough area to
the area with the central facet for the <111> crystals or alternatively from the interface shape obtained in the <100> crystals assuming the same thermal conditions (see also Section 4.2).

2.4. Morphological Stability at High Dopant Concentrations

If heavily doped crystals are grown, the melt contains at least one doping element in the concentration range of $10^{17}$–$10^{20}$ at/cm$^3$. Under such conditions, a morphological instability of the solid–liquid interface can take place. This originates from the strong enrichment of the dopant concentration in front of the solid–liquid interface (due to segregation), and the resulting reduction in the liquidus temperature by several degrees. If the actual melt temperature in front of the interface becomes smaller than the liquidus temperature, constitutional supercooling will occur and the interface becomes morphologically unstable because any fluctuation forms a protuberance of the interface which grows into the supercooled melt region. It has been theoretically and experimentally shown that a number of such projections will be formed in a close-packed array separated by distances which are determined by the local transport conditions [28–30]. The regions between the projections become progressively richer in solute and will finally be included in the crystal. This can be observed in the experiments by a breakdown of the regular interface.

The theoretical description of constitutional supercooling was developed by Tiller [31] and Hurle [32]. They derived a stability criterion for growth with an atomically rough interface. Chernov [1] extended the model in order to also consider atomically smooth interfaces. According to these authors, constitutional supercooling and the interface instability can be avoided if

$$
\frac{G}{\nabla} \geq \frac{(1-k)mC_l}{kD} \left(1-\Theta\right) - \frac{(\Delta H/\rho) \Theta}{a_\text{S} - a_\text{L}}
$$

where $G$ is the axial temperature gradient in the melt at the interface, $\nabla$ the growth rate, $m$ the slope of the liquidus line, $k$ the segregation coefficient, $D$ the diffusion coefficient, $C_l$ the dopant concentration in the melt, $\rho$ the specific heat in the melt, $\Delta H$ the latent heat, $a_{\text{LS}} = \lambda_{\text{L}}S/\lambda_{\text{S}}\rho_{\text{LS}}$ the thermal diffusivity in liquid $\text{L}$ and solid $\text{S}$. $\Theta$ is a kinetic factor which considers the reduced affinity of anisotropic growth for morphological instabilities.

For $\Theta = 0$ (atomically rough interface), Equation (6) is identical to the stability criterion derived by [31,32]. In this case ($\Theta = 0$), the maximum growth rate is 131 mm/min for $C_l = 1 \times 10^{19}$ at/cm$^3$, 44 mm/min for $C_l = 3 \times 10^{19}$ at/cm$^3$, and 13 mm/min for $C_l = 1 \times 10^{20}$ at/cm$^3$, assuming As-doped silicon with $k = 0.35$ and $G = 350$ K/cm. Thereby, $C_l = 1 \times 10^{19}$ at/cm$^3$ corresponds to the As concentration at the beginning of the growth process, $C_l = 3 \times 10^{19}$ at/cm$^3$ to the As concentration at the beginning of the last portion of the crystal growth process, and $C_l = 1 \times 10^{20}$ at/cm$^3$ to the As concentration during the final solidification of the last molten zone when taking into account the As concentration profile in the grown crystal based on Pfann’s theory [33]. It is obvious that constitutional supercooling is very unlikely in the present crystal growth experiments as the translation rate and therefore the actual growth rate is much lower than the critical growth rate except in the very last portion of the crystal growth process, when the lamp power is switched off and the crystal solidifies very quickly.

For $\Theta > 0$ (atomically smooth interface), it can be seen from Equation (6) that facets turn out to be more stable against short-range perturbations even in highly supercooled melts in comparison to atomically rough interfaces. Figure 3 shows the maximum critical growth rate $V_{cr}$ for a stable interface in dependence on the kinetic factor $\Theta$ for As-doped silicon with $k = 0.35$, $C_l = 1 \times 10^{20}$ at/cm$^3$, and $G = 350$ K/cm calculated by Equation (6). Under these conditions, the maximum growth rate $V$ is 13 mm/min for an atomically rough interface ($\Theta = 0$) and almost doubles to 22 mm/min for faceted growth with $\Theta = 0.3$. 


Figure 3. Maximum critical growth rate \( V_{cr} \) for a stable interface in dependence on the kinetic factor \( \Theta \) for As-doped silicon with \( k = 0.35, C_L = 1 \times 10^{20} \text{ at/cm}^3 \), and \( G = 350 \text{ K/cm} \) calculated by Equation (6).

3. Experimental Details

In order to analyze phenomena related to faceted growth, we performed Floating Zone experiments in a double-ellipsoid mirror furnace [34] and Zone Melting experiments in a mono-ellipsoid mirror furnace [35]. For that purpose, single crystalline, heavily As-doped ([As] = 2.5 \times 10^{19} \text{ at/cm}^3; \text{ resistivity} = 2.5–3.0 \text{ m\Omega cm}), <100>- and <111>-oriented silicon rods with 8 mm diameter and a length of 80–100 mm were used. The silicon rods were positioned in such a way in the furnace that a molten zone with a zone height \( h \) of 10–12 mm is established in the lower part of the sample at the beginning of the crystal growth process. The parabolic temperature profile which was applied gives a maximum superheating \( T_{max} \) of around 50 K at half zone height \( h/2 \) [36]. After homogenization of the melt, the melt zone was moved through the silicon rod at constant translation speeds \( V_t \) between 1 mm/min and 8 mm/min while adjusting the heating power to keep the zone height nearly constant. In all experiments, the sample was continuously rotated at a rate of 8 rpm during the growth process in order to provide markers (rotational striations) for the determination of the actual growth velocity. At the end of the growth process, the power of the lamp system was switched off and the final molten zone solidified from both sides (top and bottom) to the center of the molten zone. For each translation velocity, the experiment was repeated at least twice to check the reproducibility of the results.

The experiments in the double-ellipsoid mirror furnace were carried out by the Floating Zone technique, i.e., with a free surface of the liquid zone, and therefore in the presence of Marangoni convection. Although under these conditions, the melt flow is three-dimensional and time-dependent [36], the advantage of using this configuration is that the silicon rod is not contained in a silica ampoule, and a Dash-neck [37] can be grown by adjusting the lamp power and the translation speed at the beginning of the growth process. The Dash-necking procedure in general allows for the elimination of dislocations and for the growth of dislocation-free silicon crystals with diameters up to 450 mm by the Czochralski method and up to 200 mm by the Floating Zone technique [38]. Additionally, in our case, we could successfully apply the necking procedure to grow crystals that are considered to be dislocation-free. XRT measurements of vertical cuts confirmed this, as they did not show any dislocations; moreover, no other indications for dislocations were observed for the samples discussed here. Figure 4a shows a photograph of a typical silicon crystal with a neck grown by the FZ technique in the present work.
Zone Melting experiments were performed in a mono-ellipsoid mirror furnace. In this case, the silicon rod was oxidized before the crystal growth experiment in order to form a 5 µm thick SiO$_2$ skin on the silicon surface, and then mounted into a silica ampoule. The ampoule was evacuated, and a defined oxygen atmosphere was established to ensure the stability of the SiO$_2$ skin during the whole growth duration. The purpose of the SiO$_2$ skin was to avoid Marangoni convection [39]. During the Zone Melting process, only weak buoyant convection is present, which is caused by the parabolic temperature profile [40]. However, the disadvantage of this approach is that no necking procedure can be carried out. Therefore, the grown crystals contain dislocations in the order of $10^2$–$10^3$ cm$^{-2}$ as measured by XRT on vertical cuts. A typical silicon crystal grown by the Zone Melting technique is shown in Figure 4b. More details about the experimental procedure can be found in [41].

For the investigation of the microstructure with a focus on the facetted growth of the silicon crystals grown by the procedures described above, three measurement techniques were used. In the case of the Floating Zone crystals, the so-called growth ridges are clearly visible on the crystal surface. Therefore, they were analyzed in a non-destructive, contact-less way by using surface topography measurements as described in [19,20]. In the case of the Zone Melting crystals, this was not possible because the growth ridges are less marked due to the presence of the SiO$_2$ skin on the surface during growth.

For all samples, slices were cut parallel to the longitudinal axis of the rods as shown in Figure 2. The slices were subsequently prepared by grinding, polishing, and etching. The shape of the solid–liquid interface including the facetted areas was made visible by Nomarsky interference contrast images and analyzed as described in Section 4. The measurement principles and procedures are described in more detail in [41]. Furthermore, XRT measurements of the slices were carried out similar to those described in [22], in order to obtain information about the dislocation content in the crystals.

4. Results and Discussion

4.1. Growth Ridge Geometry for <100> and <111> Crystal Orientations

Figure 5 shows the growth ridge width $w$ measured by surface topography [19,20] in <100> (Figure 5a) and <111> (Figure 5b) dislocation-free silicon crystals grown by the FZ technique for various growth velocities $V$. It is obvious that the growth ridge width is on average 30% larger for the <100> orientation than for the <111> orientation, as we could expect from the geometrical location of the <111> edge facet and the corresponding edge facet angle with respect to the growth direction (see Section 2 and Figure 2). However, no clear influence of the growth velocity on the growth ridge geometry was found, as might be expected from the variation in growth velocity and temperature gradient [10,20]. This is caused by the timely non-constant temperature field causing periodic back-melting of the outer surface of the crystal as it passes through one of the two lamp foci. Therefore, the growth ridge cannot be measured in the as-grown state, strongly limiting the applicability of this analysis for this kind of sample.
Figure 5. Growth ridge width \( w \) measured by surface topography in \(<100>\) (a) and \(<111>\) (b) dislocation-free silicon crystals grown by the FZ technique for various growth velocities.

4.2. Diameter of the Central Facet

Figure 6a shows the diameter \( d \) of the central facet as a function of the growth velocity for dislocation-free \(<111>\) crystals grown by the FZ technique. As can be seen, the central facet becomes larger with increasing growth velocity. This is primarily a geometrical effect because the interface deflection \( \Delta x \) decreases with increasing growth velocity as shown in Figure 6b. From Figure 6b, it is also obvious that the thermal conditions are nominally identical during the growth of \(<100>\) and \(<111>\)-oriented crystals at identical translation velocities, because the interface deflection determined in the \(<100>\)-oriented crystals agrees very well with the deflection of the melting isotherm \( T_m \) of the \(<111>\)-oriented crystals. Thereby, the deflection of the melting isotherm was determined from the extrapolation of the interface shape from the rough interface in the outer crystal area to the inner area where the central facet occurs. The decreasing deflection in turn is a thermal effect. At higher translation velocities, more latent heat is released at the interface, and simultaneously, the crystal is pulled faster out of the lamp focus leading to a higher cooling rate.

Figure 6. (a) Diameter \( d \) of the central facet as a function of the growth velocity for dislocation-free \(<111>\) crystals grown by the FZ technique; (b) interface deflection as a function of the growth velocity for dislocation-free \(<100>\) and \(<111>\) crystals grown by the FZ technique. In the case of the \(<100>\) crystal, the interface deflection was directly determined from the microscope images, whereas for the \(<111>\) crystals, the deflection of the melting isotherm \( T_m \) is shown, which was obtained by the extrapolation of the interface shape from the rough interface in the outer crystal area to the inner area where the central facet occurs.

As a result, the temperature gradient \( G \) in the crystal increases, and therefore the interface deflection decreases due to both effects. This is confirmed in Figure 7, which
shows the temperature gradient $G$ in the silicon crystals obtained from experimental data, as well as from numerical simulations as function of the growth velocity. The experimental value for the temperature gradient $G$ was determined from the distance $\Delta x$ between the extrapolated melting point isotherm $T_m$ and the central facet at the symmetry axis as described in Section 2 (see also Figure 2). As all crystals grown within this experimental series are considered to be dislocation-free, the supercooling $\Delta T$ for the central facet to grow can be assumed to be constant and is set to $\Delta T = 3.7$ K according to Voronkov [9]. The temperature gradient $G$ increases from around 160 K/cm at $V = 1$ mm/min, over around 200 K/cm at 3 mm/min, to more than 300 K/cm at $V > 5$ mm/min. Similar behavior was found in our numerical simulations where the heat transport in the mirror furnace including the silicon rod was calculated using the Ansys software. More details about the modeling approach can be found in [42].

![Temperature gradient G determined experimentally from the central facet geometry and melting isotherm and from numerical simulations as a function of the growth velocity.](image)

**Figure 7.** Temperature gradient $G$ determined experimentally from the central facet geometry and melting isotherm and from numerical simulations as a function of the growth velocity.

Figure 8 shows the diameter of the central facet along the growth direction for two $<111>$-oriented silicon crystals grown at a translation rate of $V = 3$ mm/min. One crystal was considered to be dislocation-free because it was grown with Dash-neck by the Floating Zone technique. The other was grown by the Zone Melting method without Dash-neck and thus contained dislocations. As expected (see Section 2), the diameter of the central facet of around 5 mm in most part of the dislocation-free sample is much larger than in the dislocation-containing one, where it is only 2 mm. The peak, which occurs in both crystals at the last crystal portion, is the result of a sudden increase in the growth velocity accompanied by a strong increase in the temperature gradient during the solidification of the final molten zone.
Figure 8. Diameter of the central facet along the growth direction for two <111>-oriented silicon crystals grown at a translation rate of $V = 3$ mm/min. One crystal was essentially dislocation-free (blue curve); in the other crystal, dislocation were present (red curve).

By using the Brice model [13] and assuming that the thermal conditions are nominally the same for both growth configurations, the supercooling $\Delta T$ required for the central facet to grow scales with the square of the diameter of the central facet. When the supercooling $\Delta T_{DF}$ for the dislocation-free sample is set to 3.7 K [9], then a supercooling $\Delta T_{WD}$ of only 0.6 K for the dislocation-containing sample is obtained. This value agrees nicely with the results of [21], as well as with the prediction of Voronkov [9] as shown in Figure 1.

4.3. Morphological Instability

Figure 9 shows microscope images of vertical cuts of two etched silicon samples to illustrate the evolution of the interface during the last portion of the crystal growth process. In the <100>-oriented crystal, the typical features of an interface instability due to constitutional supercooling are visible when the growth rate exceeds 10–16 mm/min (Figure 9a), whereas no morphological instability is observed in the <111>-oriented crystal due to the stabilizing effect of the central facet (Figure 9b). The experimental findings agree well with Chernov’s criterion (Equation (6)), where the atomically rough interface of the <100> crystal should become unstable when $V$ exceeds 13 mm/min and the atomically smooth interface of the <111>-oriented crystal is still stable up to a growth rate $V$ of 22 mm/min when $\Theta = 0.3$ is assumed (see Figure 3).
Figure 9. Microscope images of vertical cuts of two etched silicon samples to illustrate the evolution of the interface during the last portion of the crystal growth process. (a) <100>-oriented crystal exhibited interface instability when the growth rate exceeded 10–16 mm/min; (b) <111>-oriented crystal did not exhibit signs of morphological instability.

Figure 10 also shows the evolution of the interface of a <100>-oriented sample during the last portion of the crystal growth process. It can be seen that the interface instability due to constitutional supercooling can recover. In the instability region, the growth of the faceted columns is associated with the incorporation of large amounts of dopant species, which even leads to SiAs precipitates as reported by [23]. As a result, the dopant concentration in the melt in front of the interface decreases in this case even below the critical value such that the criterion for the avoidance of constitutional supercooling is met. Then, for a short time, the crystal is able to continue to grow with a stable interface until the dopant concentration in the melt violates the stability criterion and cellular growth sets in again.
4.4. Fluctuations of the Central Facet

Thus far, the experimental results agree very well with existing theories about facetted growth. However, unexpected phenomena were also observed which cannot be explained conclusively by existing theories. Figure 11 shows microscope images of vertical cuts of three etched silicon samples to illustrate the evolution of the central facet in the <111>-oriented crystals. In all cases, the translation velocity was 1 mm/min. In the dislocation-free sample, which was grown by the FZ method, the diameter of the central facet was very stable along the growth direction (Figure 11, left). In the dislocation-containing sample, which was grown by the ZM method, the diameter of the central facet fluctuated aperiodically but with distances between major amplitudes corresponding to around 0.025 Hz (Figure 11, middle). A repetition of the experiment reproduced the results (Figure 11, right). Such fluctuations were not observed in ZM crystals grown at higher translation velocities.
Figure 11. Microscope images of vertical cuts of three etched silicon samples to illustrate the evolution of the central facet in the $<111>$-oriented crystals. In all cases, the translation velocity was 1 mm/min. (left) Dislocation-free sample with a stable central facet diameter; (middle) dislocation-containing sample exhibiting fluctuations of the central facet diameter; (right) repetition of the experiment reproducing the results.

The root cause for these fluctuations is unclear. Since the effect occurred only in the dislocation-containing crystals, a correlation with the dislocation density might seem natural. If the dislocation density becomes small enough, such that the distance between the leading dislocation and the wafer center is not always close to zero simply due to statistics, one expects the supercooling and thus the facet diameter to depend on this distance. The leading dislocation is the source point that has the highest supercooling “and therefore creates the largest lateral flux of steps” according to Voronkov’s definition [43]. However, XRT measurements have not shown clear evidence thus far, as such fluctuations are also observed in regions with moderately high dislocation densities.

Other explanations would be an aperiodic change in the interface shape [44], which in turn would be linked to a corresponding oscillation in the heat transport. Mechanical vibrations of the translation unit, as well as fluctuations of the lamp power in this frequency range as other obvious explanations for fluctuations of the heat transport are very unlikely. Particularly, tests with deliberately varied or pulsed lamp power had essentially no effect on the central facet diameter. There is only an indirect effect of the crystal diameter on the facet length. A diameter increase is connected with a stronger convex deflection of the interface. As a result, the diameter of the central facet becomes smaller, even for a constant temperature gradient and thus identical distance between the smooth interface and the isotherm. The opposite happens in the case of a diameter decrease. However, the facet fluctuations described above do not coincide with this effect, i.e., they also occur when no variations in the crystal diameter and/or the deflection are observed. Furthermore, it can be speculated whether these fluctuations are somehow linked to flow instabilities. It is known from the literature [45] that in Zone Melting configurations, several flow transitions will occur in dependence on the so-called Wall–Raleigh number $R_{w}$, which is defined as:

$$R_{w} = \frac{2g\beta h^4 (T_{\text{max}} - T_{m})}{\nu \kappa}$$

with gravity $g$, volumetric expansion coefficient $\beta$, zone height $h$, maximum temperature $T_{\text{max}}$ at half zone height $h/2$, zone diameter $d$, kinematic viscosity $\nu$, and thermal diffusivity $\kappa$. 
In the experiments, the zone height $h$ could not be kept completely constant. Therefore, another possible explanation for our observations might be a fluctuation of the zone height $h$ around a critical value which separates two flow regimes according to the stability diagram shown in [45]. However, the $Ra_w$ number estimated for the present configuration should be well in the stationary regime and far away from critical $Ra_w$ numbers where flow transitions are expected to occur. In order to gain further insight into whether the fluctuations of the central facet are correlated with flow instabilities, a corresponding microgravity experiment is planned onboard the TEXUS59 mission. If no facet fluctuations occur in the microgravity experiment, then flow instabilities are most likely to be the root cause for the observed phenomena. On the other hand, if the facet length also fluctuates under microgravity conditions, other phenomena must be responsible. In both cases, further investigations including numerical simulations of the growth conditions are needed.

5. Conclusions

Dislocation-free, small-diameter silicon crystals were grown using the Floating Zone technique. The temperature gradient at the interface was determined ex situ from etched cross-sections and varied between 200 K/cm and 400 K/cm, depending on crystallization velocity. These samples were used to study the occurrence of constitutional supercooling. For the rough interface of <100>-oriented crystals, a critical growth velocity of $10^{-16}$ mm/min for a temperature gradient of 300–400 K/cm was found, in good agreement with predictions from established theory. In contrast, the crystals were much more stable against morphological instabilities when grown in the <111>-direction due to the faceted interface, again in agreement with theory. Dislocated crystals grown by the Zone Melting technique exhibited smaller central facet diameters. The maximum supercooling at the facet interface was estimated to 0.6 K/cm from our experimental data. Additionally, at low growth velocities, these Zone Melting crystals exhibit unexpected, non-periodic fluctuations of the central facet which are not yet understood and are the subject of further investigations.

Author Contributions: Conceptualization, J.F. and C.K.; methodology, C.K.; investigation, S.G., T.S. and T.J.; writing—original draft preparation, J.F. and C.K.; writing—review and editing, J.F. and C.K.; supervision, J.F.; project administration, C.R.; funding acquisition, C.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the DLR/BMWK under FKZ 50WM1845.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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