

Spiral Crystal Growth in the Czochralski Process—Revisited, with New Interpretations

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Herein, the features of some Czochralski crystals with spiral morphology are investigated in detail to develop a model for their spiral growth. The case of a cylindrically growing crystal with a spontaneous transition to spiral growth is considered. At the onset of spiral growth, the development of a “foot” in the meniscus region is identified. It is shown that the growth of a foot takes place under “flaring growth conditions.” Insufficient axial heat transport through the crystal leads to a concave interface which is behind the flaring growth. The constraints of automatic weight control force the flaring crystal to grow as a spiral. The selection of a single foot is assisted by temperature fluctuations in the melt. Some symmetry breakings during spiral growth, for example, a meniscus deformation by rotation and the azimuthal growth in the meniscus, have been described already by the author earlier. Only recently spiral growth has been encountered with Czochralski silicon, grown at high pulling speed. These hypotheses on spiral growth hold for high-melting-point oxides as well as for the lower melting silicon.

meniscus diameter. Diameter changes will be therefore of ring-like nature under manual diameter control reflecting the crystal grower’s action.

1.2. Manual and Automatic Diameter Control

We have mentioned above the manual control of Czochralski crystal growth because all instabilities of the diameter growth can happen there; the crystal diameter can increase or decrease at any time because of changing temperature conditions (e.g., because of decreasing melt level by the pulling). The crystal diameter is in the state of labile equilibrium during the Czochralski process. For automatic diameter control of high-temperature oxide crystals, the weight of

the crucible is continuously monitored with high precision and electronically compared with a programmed weight decrease for growing a cylindrical crystal. Deviations between the measured and the programmed values will result automatically in appropriate power changes. Thus, cylindrical crystals with constant diameter can be grown. Another method is the continuous weighing of the crystal with comparison of its weight increase with the programmed one. For silicon, the position of the “bright ring” is taken as the indication of the crystal diameter. The bright ring is generated by reflection of light from the hotter crucible wall in the melt meniscus.

In spite of the cylindrical environment, spiral shape like diameter variations are observed at some high melting point crystals grown under automatic diameter control which have never been reported from manually controlled Cz-growth. We therefore consider the high precision of the automatic diameter control as fundamental in spiral growth. The transition from cylindrical to spiral growth, on the other hand, appears to be an instability. We are interested in this paper into this transition and into spiral growth. It is obvious that we need to produce cylindrical crystals and not spiral ones to cut wafers from them for scientific and industrial use. Any knowledge to avoid spiral growth is urgently needed though it is a fascinating phenomenon.

1.3. Flaring Growth as Special Diameter Instability in Czochralski Growth


The process of crystal pulling has been introduced by Jan Czochralski to measure the maximum solidification speed of

1. Introduction

1.1. The Czochralski Technique of Crystal Growth

It is the aim in Czochralski crystal pulling to grow a long part of the crystal in cylindrical shape from the cylindrical crucible in a cylindrical thermal environment. Growth is accomplished in manually controlled Czochralski growth by adjusting the pulling speed and the crucible temperature. The crystal grower must watch the crystal diameter continuously and must continuously regulate the electric power fed to the crucible to avoid diameter changes. This is not an easy job, lacking precision, because his man-decided action comes always late, namely only after the change of the crystal diameter shows up as a change of the melt

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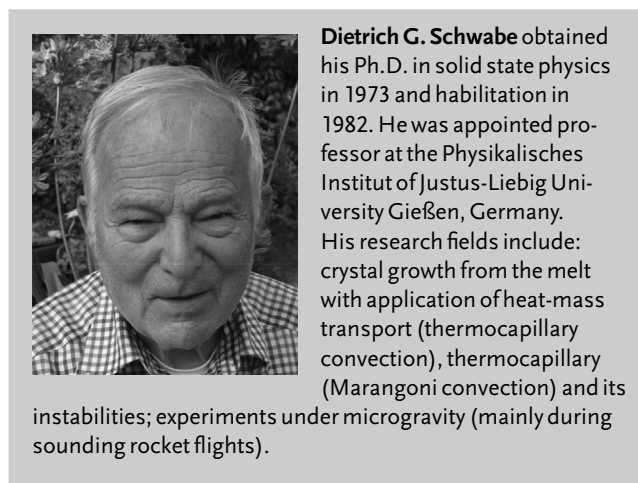
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metals.^[1] The crystal diameter decreased in his experiments at higher pulling speed until the loss of contact and detachment of the crystal from the melt (Figure 1a). One can call this diameter decrease of the crystal with final detachment from the melt an instability of the Czochralski process. We know on the other hand a growth instability in which the crystal diameter is increasing above that of the target diameter (Figure 1b). This kind of growth is called “flaring growth” and happens for growth from under-cooled melt, or for growth from mixed melts (which tend to under cool). Flaring is favored for high-melting point materials, where the radiant heat transport can dominate over conductive heat transport; the clod will act in this case as cooling plate, allowing increasing diameter growth by radiation of heat to the colder growth environment. In a sense, the diameter growth can accelerate itself by radiation cooling. Flaring growth can happen if the temperature conditions lead to a concave interface. It is obviously impossible to grow a cylindrical crystal under automatic diameter control with constant target diameter if the crystal has the tendency to flaring; any weight increase by flaring above the programmed one will be automatically stopped by extra heating. This forces the crystal to grow as a spiral, as outlined under Section 3.1. We will show there that spiral growth is driven by flaring growth under the action of automatic diameter control. As flaring is associated with an approximately concave crystal interface, the point of flaring and thus the point of onset of spiral growth can be influenced by crystal rotation. This important aspect, however, is beyond the scope of the present paper.

1.4. Examples of Crystals with Spiral Morphology Grown Under Automatic Diameter Control

Figure 2a shows an extremely well developed spirally grown crystal from YVO_4 with a target diameter of 33 mm, having approximately six spiral turns, grown with 2 mm h^{-1} .^[2] YVO_4 melts at approximately $1810 \text{ }^\circ\text{C}$ with the consequence of dominating radiation heat transport during growth. The spiral growth of this crystal began spontaneously without any external disturbance after a 20 mm long cylindrical part. The spiral diameter (outer diameter of the crystal) is larger than that of the cylindrical part



indicating the transition to flaring growth. This small but important detail was not noted in the earlier paper^[2] and demands further discussion. Therefore, in the following growth of the crystal from YVO_4 , the growth conditions have not been changed at all,^[4] but we will observe and will interpret that the first distance of the spiral terraces is larger than that between the following terraces (see Figure 2a and Section 4.1).

Figure 2b shows a crystal from SmScO_3 with three spiral turns grown with 1.1 mm h^{-1} . The material melts at approximately $1850 \text{ }^\circ\text{C}$. The crystal shows beginning of spiral growth after half of its length. It was not possible to grow longer cylindrical crystal parts from this material as the one in Figure 2b. Figure 2c shows the beginning of the growth of a cone from the high melting point oxide gadolinium gallium garnet (GGG). It shows spiral growth during cone growth. Flaring growth is indicated by the sudden increase of the crystal diameter directly after dipping the seed (10 mm wide sheet from iridium) into the melt; spiral growth can occur as well during seeding on with diameter increase.

Figure 2d shows a silicon crystal of 1352 mm length with a mean diameter of 206 mm, pulled with an average speed of 1.03 mm min^{-1} . The growers have provoked spiral growth in the

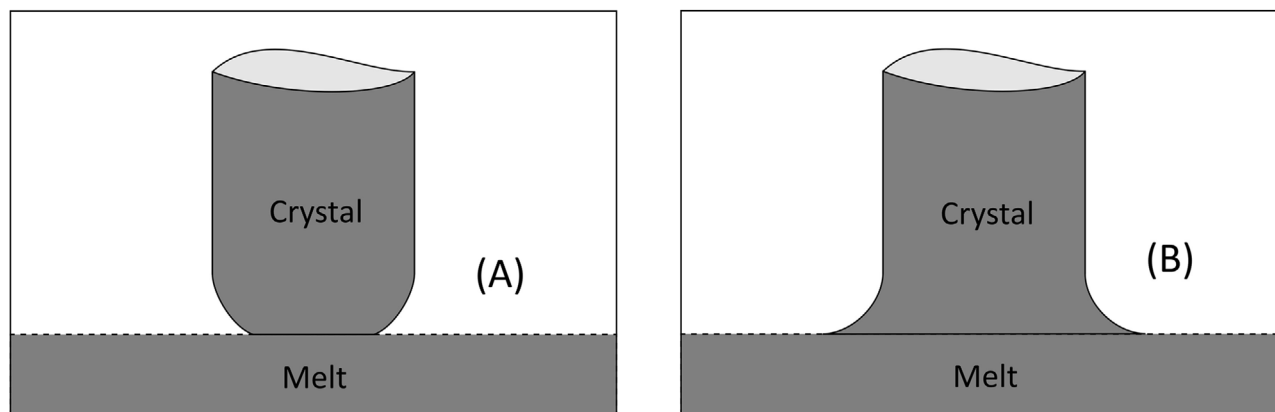


Figure 1. Common instabilities of the crystal diameter of a cubic material during manually controlled Czochralski growth (simplified). a) Diameter decrease until possible detachment from the melt. b) Flaring; relatively fast diameter increase until a new equilibrium diameter is reached. The much thicker crystal part, grown by flaring, is called a “clod.”

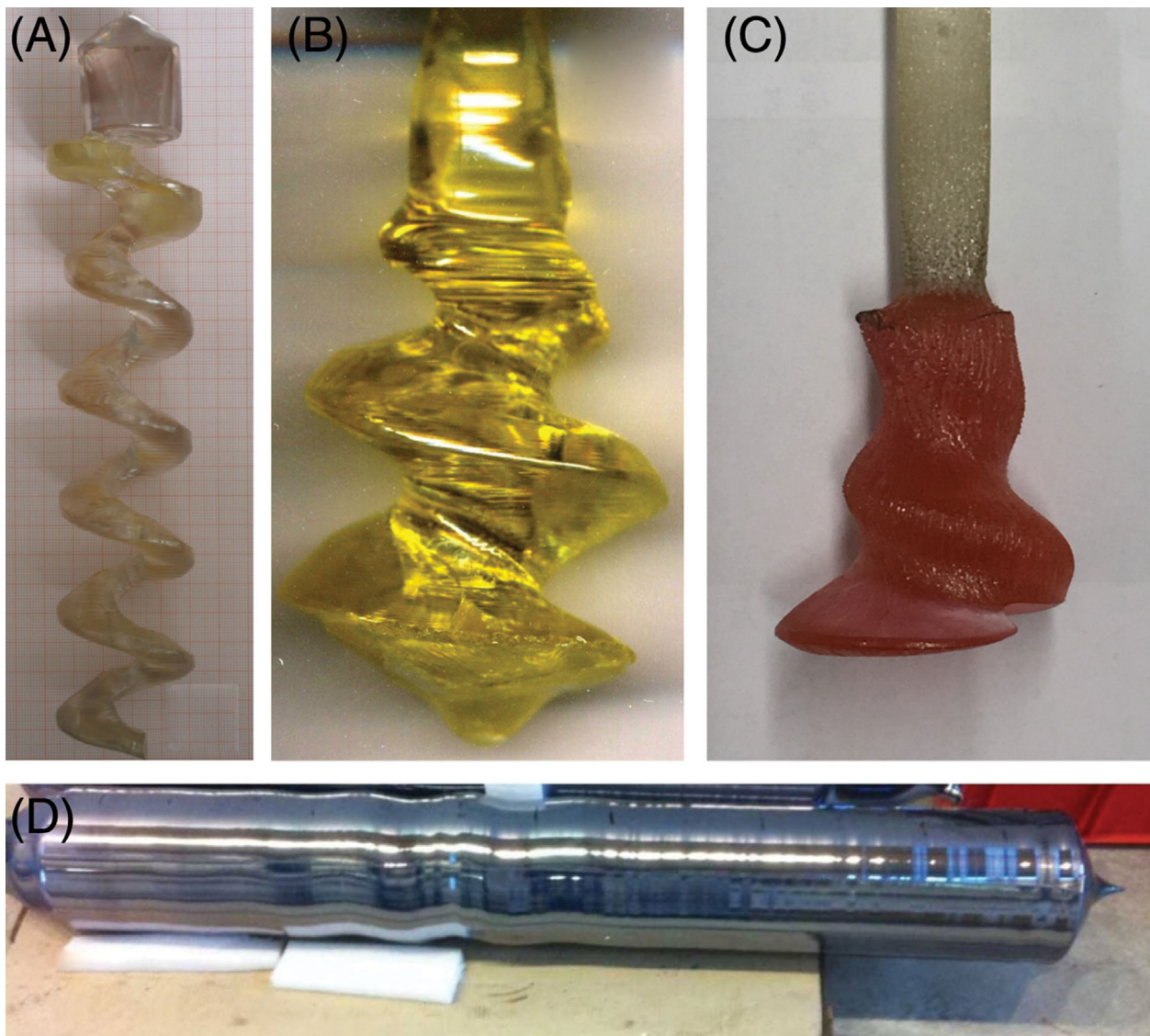


Figure 2. Various Czochralski crystals with spiral morphology grown under automatic control. First-grown part on top (for Si the first grown part is at the right). a) Crystal from YVO_4 . It shows six spiral turns. Reproduced with permission.^[2] Copyright 2011, Elsevier. b) Crystal from SmScO_3 . It has approximately three spiral turns. c) Upper conical part of a crystal from GGG, seeded on by a 10 mm wide sheet from iridium. d) Crystal from silicon, length of 1352 mm, with three spiral turns at approximately half length. Reproduced with permission.^[3] Copyright 2016, Elsevier; photo courtesy of Alexander Molchanov, PVATepla, Wettenberg, now with Siltronic. The spiral is “engraved” into the outer skin; thus, the crystal has the morphology of a spiral or that of a cork screw.

middle part by increasing the pulling speed for some hours while decreasing the melt temperature. The distance of the “spirals” is 180 mm which corresponds to 3 h of pulling time. This gives a turn-time of the spiral of 120° per hour. The spiral depth $R_o - R_i$ (with inner and outer radius R_i and R_o) of this Si-crystal is 10 mm. The outer crystal diameter is widened by 10% during the spiral part, indicating flaring growth.

Jochen Friedrich presented very recently a photo of a silicon crystal for photovoltaic use with spiral morphology and about the same size as the one in Figure 2d but with larger spiral depth.^[5] A larger growth rate was accompanied by a more convex interface, which led to flaring and this in turn led to spiral growth. This is the second silicon crystal with spiral morphol-

ogy which came to my sight and the authors presented for their growth exactly our proposed scenario for spiral growth; the cylindrical growth was limited by faster pulling and stronger cooling of the crystal periphery as already shown by Kaleev et al.^[6] I will try in this paper to find a common explanation of spiral growth for the high temperature oxides and for silicon pulled at high speeds.

1.5. Our Model for Spirally Growing Crystals

We are unable to observe what goes on at the crystal–melt interface during the onset of spiral growth and further during growth.

We can only take the outer shape of the crystal as the main source for interpretation.

Our model for the spiral growth is slightly simplified compared to reality and the simplifications are as follows:

- 1) The crystal is first growing fully cylindrical until the onset of spiral growth.
- 2) The crystal is growing without facets.
- 3) There is no end of the pulling process (the crystal is infinitely long, the melt-level in the crucible is constant).
- 4) The external thermal boundary conditions do not change during growth. An exception is in some cases the heat loss by radiation from the interface, which can become smaller for semitransparent crystal with increasing crystal length.
- 5) Convection by crystal rotation is constant because the diameter is constant, as is the rotation rate. The influence of rotation is constant to a first degree during the process.
- 6) Although some external triggers for the onset of spiral growth might exist, which result in a symmetrically lower growth of a spiral crystal, we discuss here an intrinsic reason for the onset of spiral growth, namely a spontaneous transition by flaring. We are forced to this assumption by observations.
- 7) The model of a transition to spiral growth based on flaring holds for oxides with high melting point and for Cz-silicon grown with high pulling speed.
- 8) The point of onset of spiral growth can be changed by the crystal rotation rate via the interface shape and its influence on flaring.

2. Influence of the Crystal Transparency and the Pulling Speed on the Interface Shape Which Can Result in Flaring Growth (Literature Research)

What drives the growth to become spiral? We will review selected literature, indicating the connection of spiral growth with a concave interface. It was already shown that the crystalline structure of the spiral crystal does not deviate from that of a cylindrical crystal and thus deviations from the structure are not driving spiral growth.^[7] The authors investigated a large spiral from GGG.^[7] They found no influence of doping on the X-ray structure.

2.1. Reduced Transparency Leads to a Concave Interface and to Spiral Growth

Spiral growth is found in connection with a strongly concave interface because this makes flaring possible. The interface shape of high-temperature oxides depends to a high degree on radiation heat transport. Doping can therefore have indirect influence on spiral growth via optical absorption. Stronger optical absorption gives rise to a more concave interface because heat conduction by radiation is lower. A clear effect of the optically absorbing doping in dysprosium gallium garnet is described in ref. [8]. The authors found a decreasing tendency to spiral growth by adding Ca^{2+} . This increases the transparency of the crystal. In contrast to this, by adding Cr^{4+} , the transparency decreases with a

corresponding increase of the tendency for spiral growth. These authors substituted a part of gallium with aluminium, as reported in a following paper.^[9] This doping increases the transparency very much. They found a significant reduction of the tendency for spiral growth by substituting 40% of Ga with Al. We see that the tendency for spiral growth depends on the material and its composition, especially on factors influencing the transparency and the heat transport by radiation. The authors from the same group present a convincing influence of radiant heat transport on spiral growth.^[10] They grew DyVO_4 after a modified Czochralski technique and substituted Dy partly by Gd. It happens that GdVO_4 shows very low optical absorption in the range of 700–1500 nm, which is the range of a black body radiation at melting point temperature, whereas DyVO_4 has various strong absorption bands in this spectral range. In essence, the crystal of DyVO_4 cannot dissipate the heat from its interface by radiation axially upward as good as the crystal from GdVO_4 and its interface will therefore tend to a concave shape. This allows flaring growth and thus spiral growth. The mixed crystal from $\text{Dy}_{1-x}\text{Gd}_x\text{VO}_4$ has as well a concave interface and will show the tendency to spiral growth. The authors speak of “energy accumulated below the interface,” which comes near to our interpretation of “insufficient axial heat transport through the crystal.”

The authors in ref. [11] compare the tendency to spiral growth and the absorption of radiation of various scandates. Among the scandates, only GdScO_3 displays no spiral growth and it is the only one showing low optical absorption at elevated temperatures in the relevant range for radiation heat transport. This result supports the findings from refs. [8–10] and the authors of ref. [11] can interpret their result by including their studies of normally convex interface shapes: “The crystal melt interface becomes more planar (for absorbing crystals)”. Insufficient heat transport through the crystal–melt interface is therefore identified as a main condition for spiral growth.

In summary, we have a number of papers in the field of growth of high-temperature oxides indicating that semitransparent crystals tend to spiral growth if their optical transparency is low and if their length is exceeding a certain value; in this case, radiant heat transport away from the interface is insufficient and flaring might occur.

2.2. High Pulling Speed Leads to a Concave Interface of Silicon and to Spiral Growth

It was assumed for a long time that spiral growth would happen only for high-temperature melting materials until a numerical work showed up with “twisted growth”^[6] for silicon crystals for photovoltaic applications with industrial dimensions. Such Si crystals,^[3,5] as the one in Figure 2d, are grown with higher pulling speed (for a certain time) and under the application of cooling baffles for cooling the crystal periphery. Higher pulling rate means larger production of heat of fusion at the interface. These conditions generate a concave interface shape and possibly a region of under-cooled melt in the vicinity of the melt meniscus as calculated in ref. [6]. We identify this condition of a concave interface as the main cause for spiral growth which leads to flaring growth, both for high-temperature oxide crystals and for silicon.

3. A General Model for the Spiral Growth Based on Flaring Growth Under Automatic Diameter Control and on Selection by Growth Fluctuations

In the following, we will describe the spiral growth in detail. We assume for simplicity, that a cylindrical crystal has grown for a certain time until the onset of spiral growth occurs (Figure 2d). This scenario is observed during the growth of some high-temperature melting oxides. After this, we will then observe the emergence of various symmetry-breaking steps for the crystal shape. We distinguish two different phases of spiral growth, namely, i) the onset with the growth of a foot in Section 3.1 and ii) the subsequent following growth of the spiral crystal in Section 3.3.

3.1. Flaring Growth Initializes Spiral Growth Together with Temperature Fluctuations

We consider the unsolved question, How is the cylindrical growth mode transferred to the spiral one, how can a single foot develop under automatic growth control? Under manual growth control and under certain conditions, for example, with under-cooled melt, a clod is formed in the flaring mode. This experience can be encountered by all crystal growers during exploratory manually controlled growth. We can report on a mixed melt from $\text{Fe}_{(1-x)}\text{Mn}_x$ where deliberate seeding-on with a seed of 1–2 mm diameter was almost not possible because a clod formed immediately with a diameter of up to 40 mm within a few seconds. Such mixed melts are known for their tendency to under-cooling.

Centrosymmetric flaring growth is not possible under automatic diameter control because any formation of a clod would mean a weight increase above the programmed one and the control unit would increase the crucible temperature to melt the clod back. However, the tendency to form a very small “baby clod” will exist in the very beginning. Though we exclude crystals of lower symmetry from our considerations, we can assume that even such a baby clod will not grow in a perfectly cylindrical symmetric fashion because of imperfect rotational thermal symmetry of the set-up and fluctuations of the melt temperature. Instead it will show, already in “*statu nascendi*,” small extrusions together with small inlets (Figure 3). Fluctuations of the temperature in the

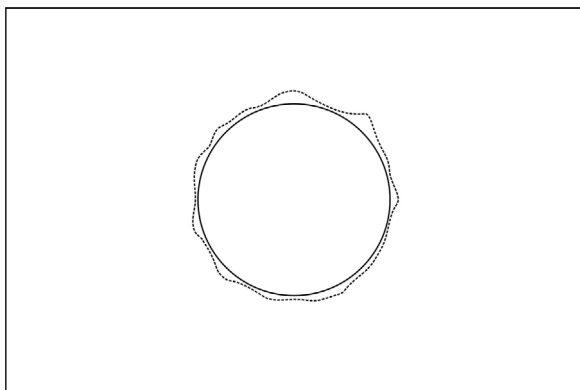


Figure 3. Schematic picture of diameter variations in radial and in azimuthal direction in *statu nascendi* of a clod (very much exaggerated). The three-phase line fluctuates under fluctuating temperature.

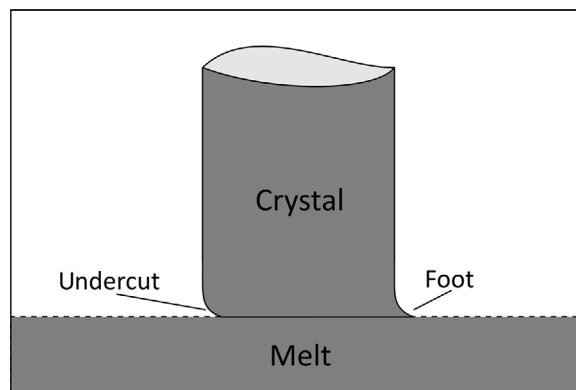


Figure 4. Sketch of the development of a foot and its undercut. The undercut compensates for the extra weight increase by the foot. The azimuthal motion of foot and undercut (discussed in Section 3.2), together with axial growth, results in a spiral crystal.

melt will generate small-diameter fluctuations of the three-phase line of the clod in its very early state, which vary in azimuthal position and in time (Figure 3). We assume again that these fluctuations (possibly together with those of the control unit) have the effect that the smaller extrusions will vanish and the largest extrusion will survive and will grow. This growth of the largest extrusion under fluctuating temperature conditions is supported by its better cooling from above-compared to the cooling of the smaller extrusions. Thus, the largest extrusion will grow and will become the foot (Figure 4). For compensation of the weight of the foot, an undercut must develop on the opposite side of the foot under the actions of the control unit.

The foot comes into existence by flaring and by the help of temperature fluctuations in the melt. Once a foot has formed, and if it travels azimuthally around (see Section 3.3), then the crystal will grow in a spiral shape. High working temperatures will assist the foot growth by radiation cooling of its surfaces pointing upward to the colder regions of the growth system. We can see in Figure 5 the connection of flaring growth (in this case of a three-fold spiral) with a very concave interface. This crystal starts to develop with a threefold spiral,^[3] which is a rare deviation from the common onefold spiral. We assume this threefold spiral to be due to strong under-cooling, with the growth overriding the foot-selection by temperature fluctuations.

We state that flaring growth, together with the automatic diameter control in combination with growth fluctuations, is leading to spiral growth with only one spiral.

The temperature fluctuations in the melt are not directly responsible for the formation of the spiral; they are not imprinting the spiral shape on the growing crystal; however these temperature fluctuations allow the selection process for one extrusion to become the foot.

The discussed temperature fluctuations in crystal growth melts are well known. They result in growth striations in oxides as well as in silicon. A recent example for striations and back melting was found by the investigation of the growth ridges of Czochralski silicon highly doped with arsenic.^[12] The authors report about a concave interface in the cylindrical part of the crystal, and further about morphological instabilities of the interface and about possible constitutional under-cooling in front of the

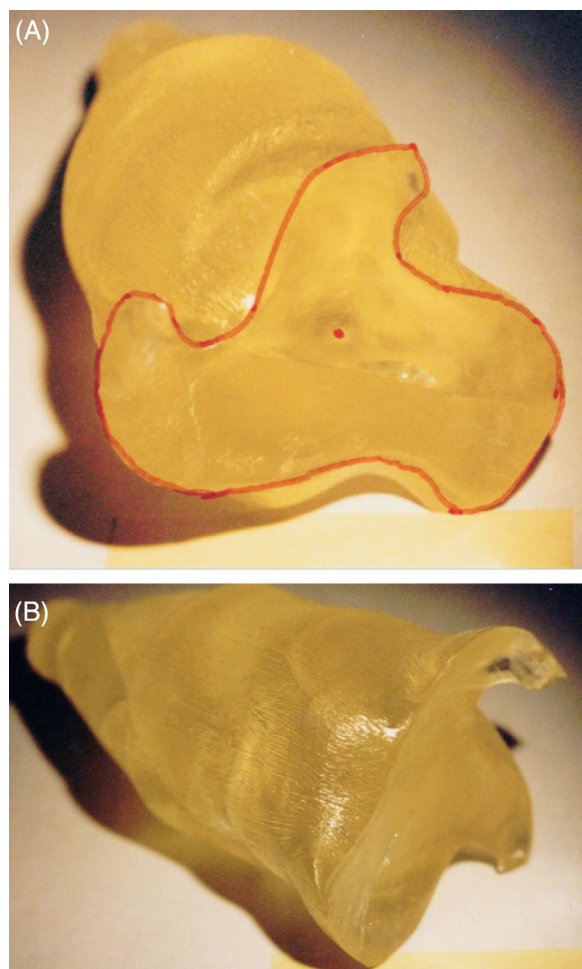


Figure 5. Photos of a 4 cm diameter TGG crystal detached from the melt. Reproduced with permission.^[3] Copyright 2016, Elsevier; photo courtesy by Lothar Ackermann, FEE, Idar-Oberstein. We can see the concave interface connected with flaring growth (in this exceptional case of a threefold spiral). The view on the pulled-off interface (a) shows threefold footing. The red line indicates the three-phase line during growth. The view (b), partly from the side, displays the very concave interface; the three feet could still grow because of radiation cooling of their surfaces, whereas the axial growth in the center of the crystal lags behind.

interface near the edge facets. One of their crystals was grown with cooling baffles with a pulling rate v of 1.2 mm min^{-1} , what is near the stability limit of cylindrical growth, $v_{\text{limit}} = 1.4 \text{ mm min}^{-1}$. These conditions are all pointing to possible flaring growth.

Other reasons for foot growth, such as local disturbances mentioned in ref. [2] cannot be excluded but are unlikely; we see such disturbances to grow out and vanish after some time as long as this local disturbance is not continuously in action. In contrast to such an external reason for foot growth, a crystal growing in the flaring mode can continuously provide material for spiral growth with larger outer diameter due to its tendency to grow with a diameter larger than the target diameter. Flaring is an intrinsic reason and it is essential for foot growth. This process is assisted by the temperature fluctuations in the melt. Growth of a foot with its undercut is the first symmetry breaking in the Czochralski pro-

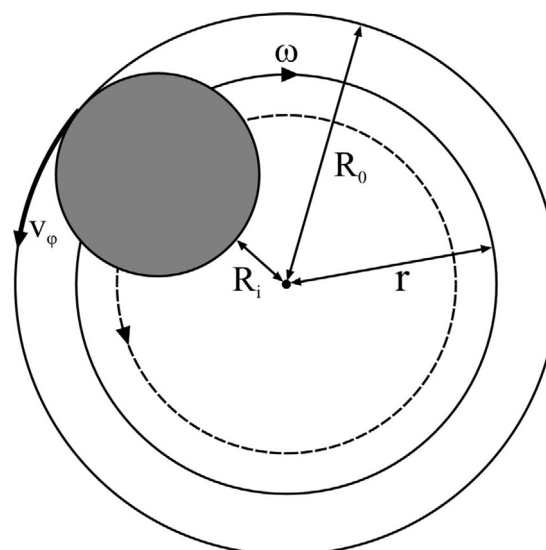


Figure 6. Schematic drawing of a horizontal cut through the liquid meniscus. It shows an idealized (circular) form of the foot, growing with v_ϕ antiwise to the crystal rotation direction ω . The foot is not circular in reality. It can have the shape of the projection of a kidney (see Figure 5 of ref. [2]). Because of the eccentric motion of the foot, its meniscus will experience a larger shear and larger deformation on its outside compared to its inner side. As well the cooling of the outer meniscus will be larger. We thus expect a higher azimuthal growth velocity on the outside of the foot. $R_0 - R_i$ = diameter of the foot; r = target radius of the crystal.

cess leading to spiral growth. It should be clear that foot growth of crystals with lower symmetry do not need foot selection by temperature oscillations.

3.2. Symmetry Breakings during the Growth of a Complete Spiral Crystal Leading to Azimuthal Growth

The growth of a spiral crystal, with its series of symmetry breakings is complicated and was described in large parts for the first time in ref. [2]. It will be repeated here in a concentrated way. The formation of a foot is the first symmetry breaking of the original cylinder symmetry of the crystal, brought about by thermal fluctuations during flaring growth. The melt meniscus of the foot will be deformed by the crystal rotation because the foot is no longer positioned symmetrically in the center (**Figure 6**). The foot is ploughing along the melt surface where its meniscus becomes steeper in the sense of crystal rotation and becomes flatter counter to the sense of the crystal rotation (like a boat with bow and stern). This is a second breaking of the cylinder symmetry, in this case by hydrodynamic forces. The flatter part of the meniscus will be better cooled because it points more to the colder upper part of the growth system and it will therefore grow faster in azimuthal direction than the opposite steeper part of the meniscus. The foot is therefore growing slowly in counter direction to the sense of crystal rotation, already known for some time.^[13] This is the third symmetry breaking, this time by growth, reacting on asymmetric radiation cooling. The superposition of the azimuthal growth of the foot with the axial growth by pulling results in the spiral morphology of the crystal.

The azimuthal growth velocity v_ϕ should be proportional to the azimuthal temperature gradient in the melt meniscus due to the difference in cooling of the flat and the steep meniscus side. An influence of the rotation speed on v_ϕ should be visible but is not reported until now. The azimuthal growth velocity v_ϕ of the foot in case of silicon (Figure 2d) was approximately 3.6 mm min^{-1} and was thus larger than the pulling speed.^[3] In summary, during spiral growth, the crystal is growing in axial direction by pulling and its bulge is growing in azimuthal direction driven by itself (by off-centering and by asymmetric cooling of an asymmetrically deformed melt-meniscus).

3.3. The Role of the Automatic Diameter Control for Spiral Growth

We cannot imagine that one could grow with manual control a foot with its undercut and further grow a spiral crystal because the grower would have to observe the change of the crystal shape and would have to react on it in an uncommon way. What could his reaction look like? In contrast to this problem in manual control, the automatic weight control unit will react only on the weight with rather high precision. This slavish way of the control unit to follow a programmed weight increase of the crystal is the basis for the development of perfect spiral morphology; the azimuthal growth instability travels around the crystal, driven by azimuthal temperature gradients in the melt meniscus. A growth instability is behind spiral growth. This growth instability is an intrinsic natural phenomenon. The automatic diameter control paves the way for perfect spiral growth and is thus attached to it.

4. Interpreting the Shape of the Spiral

We ask for the symmetry properties of spiral crystals (see YVO_4 in Figure 2a and the middle part of Si in Figure 2d). These crystals have an almost constant inner and outer radius of their spiral. For each of them, the distance of the spiral turns is almost constant.

4.1. The Scale of the Spiral Crystal and Fine Details

What rules the width of the spiral? Under width of the spiral or “spirality,” we understand the normalized difference between the outer and the inner diameter of the spiral crystal (Figure 6). The outer diameter of the spiral crystal will be proportional to the depth of its growth conditions in the flaring range, in other words, “how much will the outer diameter of the spiral deviate from the target diameter.” The spirality of the YVO_4 -crystal in Figure 2a is rather large directly in the beginning of spiral growth (top of Figure 2a). The spiral part of this crystal grew deeply in the flaring range. The spiral diameter is smaller for the SmScO_3 -crystal in Figure 2b and even smaller for the Si-crystal in Figure 2d. These two crystals grew less deep in the flaring range.

The growth conditions for a cylindrical crystal of a certain diameter can be assumed to be fixed with regard to a certain pulling speed. They are unique in the sense that the growth of a cylindrical crystal is only possible with the mentioned parameters in the envisaged growth system. The same is true for growth in the

flaring regime and thus holds for spiral growth. We can therefore assume the continuation of spiral growth in its regime as it holds for cylindrical growth in its regime. This explains the equidistant spirals of spiral crystals; well performed spiral growth under constant growth conditions (constant flaring conditions). This seems to be only possible under automatic growth control which imposes strictly the condition of constant mass increase for the growth of a “cylindrical” crystal.

We will now investigate the spiral crystal in Figure 2a in more detail. The crystal grows after the onset of flaring on its left (in Figure 2a) side in radial direction until the diameter of the flaring clod is reached. This diameter of the spiral is considerably larger than the target diameter of the cylindrical crystal. This shows that spiral growth takes place under flaring conditions. The diameter of the spiral of YVO_4 in Figure 2a is significantly larger in the very beginning compared to later turns, where it is constant or slightly decreasing. This happened without any changes of the growth parameters.^[4] We can interpret the overshooting diameter growth in the beginning growth of this YVO_4 -crystal; it indicates stronger cooling from above this first part of the spiral compared to the following spiral turns.

Can we influence the onset of spiral growth? The only free parameter to achieve this is the rotation rate which deforms the meniscus. After my understanding, the meniscus deformation tends to zero for zero rotation rate. With it could the azimuthal growth velocity go to zero. This dependence gives the possibility to suppress spiral growth. The decrease of the rotation rate must be applied early enough before a foot has formed and a large spiral crystal has developed, which then would distort the thermal field above the crystal meniscus. One could as well try to change the rotation rate of the crystal with a sufficiently large frequency, always changing the asymmetry of the melt meniscus.

4.2. The Distance of the Spiral Turns

The distance A of the spiral turns is given by the axial growth during one spiral turn. The latter is given by the azimuthal growth velocity in the meniscus. We are following the idea that the azimuthal growth is driven by the azimuthal left-right asymmetry of the cooling of the meniscus. The asymmetric cooling is due to a meniscus deformation by the rotation of the off-centered crystal foot. The asymmetry of the cooling will depend on the eccentricity of the position of the meniscus surfaces and on the strength of the cooling. This will be considered now; the surface of the liquid meniscus is pointing upward with partly radiation heat exchange with the lower side of the spiral terrace above (Figure 7).

The azimuthal growth velocity increases with the cooling from above the liquid meniscus. Thus, the distance A of the spiral turns will be partly governed by the temperature of the crystal terrace just grown before. Because the cooling and growth of the lowest terrace is partly driven by heat exchange with the terrace above, spiral growth can be considered as a kind of self-organization, as already proposed by the author.^[2] We can observe such an influence of the upper spiral turn on the one below at the YVO_4 crystal in Figure 2a; the first spiral terrace is considerably less inclined (larger azimuthal growth velocity v_ϕ) than the following terraces. The distance A of the terraces on the left side of the YVO_4 crystal at the top is significantly smaller than the

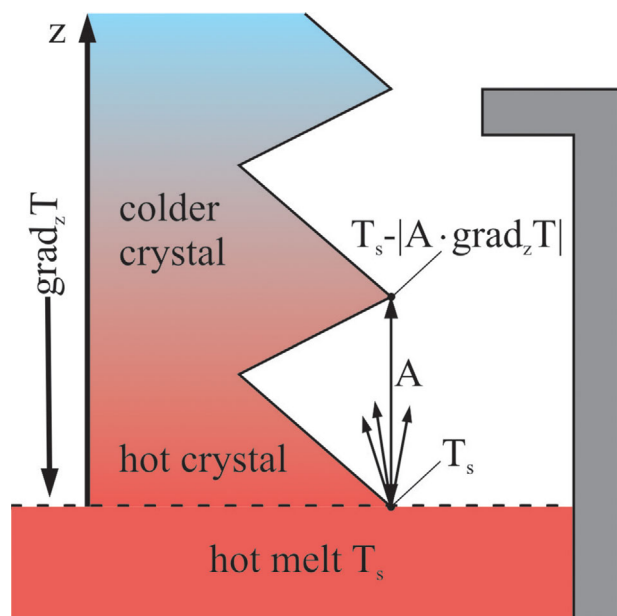


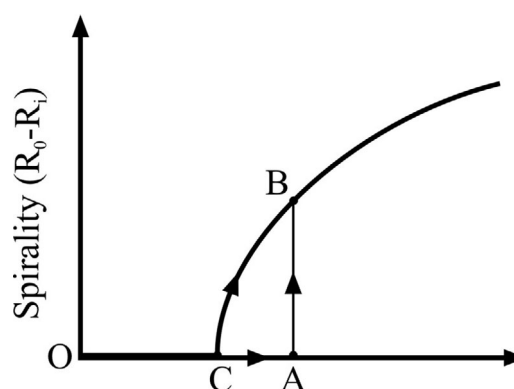
Figure 7. Cooling (effective radiation heat flux) of the growing spiral terrace by heat exchange of its meniscus region with the colder terrace above it (schematically). The azimuthal growth speed of the spiral increases with cooling from above.

following ones. The larger v_ϕ can be understood as the influence of stronger heat flux to cooling from above as long as the spiral does not shield the meniscus against radiant heat loss to above (as long as the hole in the lid, covering the crucible, is not yet closed by the spiral crystal). Moreover, one can see that the spiral distance of the later grown spiral turns is smaller and constant because it is governed by constant and less heat exchange with spiral turns above.

4.3. Scenario for the Transition from Cylindrical to Spiral Growth

We add a generalized description of the transition of an idealized crystal from cylindrical to spiral growth. The knowledge of a diagram for the onset of spiral growth as in **Figure 8** would be helpful for the crystal grower to know the limits of cylindrical growth in his set-up. PVATePla (Wettenberg near Giessen, Germany) tries to measure point C in Figure 8 for predicting the highest possible pulling speed but to avoid spiraling crystals.^[5]

Figure 8 shows such a transition in the framework of “transitions of state.” The control parameter for such a transition to spiral growth could be the pulling rate or the length of a semi-transparent crystal, or the cooling of the crystal periphery by a baffle (Figure 8). The concavity of the interface could be taken as a suitable parameter, which reflects the sum of all influencing parameters just mentioned. Unfortunately, it is not known during growth, but only afterward from a vertical cut of the grown crystal. We assume cylindrical growth in the beginning of growth, which means “spirality = zero.” The spirality starts to develop at the bifurcation point C (Figure 8) from the value zero, possibly as to the power of $1/2$. The growing crystal could follow the path either toward point B, or it could happen that the transition to spiral growth is hindered (the flaring growth is kinetically suppressed).



Concavity of interface or pulling speed or cooling of crystal periphery

Figure 8. Bifurcating transitions from cylindrical to spiral growth at the critical point C. The growth system can develop either continuously from C to B or from C to A, and then jump spontaneously with its spirality to B (as for TGG in Figure 9).

The crystal then stays with spirality zero until it reaches point A, from where it jumps from zero spirality to the large value B. Suppressed flaring with a jump to a larger spirality could have been the case at the beginning of spiral growth of YVO_4 in Figure 2a and for the TGG-crystal in **Figure 9**. The sudden diameter increase of the TGG-crystal near its end in Figure 9 supports the interpretation given in Figure 8. In support, we cite our colleague Klaus Dupré^[14] who grew high temperature oxide crystal for many decades: “The spirals form always spontaneously without interference of the grower and as well without any indication in the weighing-signal nor in the error-signal nor in the power-signal. The formation of a spiral can be detected only visually (one has to look into the growth vessel). After a growth time of the spiral of approximately 5–6 h (approximately 5 mm), one can see more noise in the diameter control signal and some hours later one can see significant indications in the weight signal.” The spontaneous onset of spiral growth with large spirality, shown in Figure 9, indicates suppressed crystallization. The Si-crystal and the crystal from SmScO_3 in Figure 2 seem to follow the smooth way from C to B in Figure 9 with no jump of the spirality. Counteracting the spiral growth of silicon was possible by reducing the pulling speed (Figure 2d). This silicon crystal has not been grown as deeply in the flaring range and its spirality was consequently comparatively small.

5. Summary

We give a complete picture of the spiral growth of Czochralski crystals. For spiral growth, we propose that the thermal boundary conditions must allow a concave interface which is leading to flaring growth. Flaring growth together with the constraints by automatic diameter control is behind spiral growth. Flaring growth leads to the formation of a clod. However, the growth of a clod is hindered under automatic weight control. The weight



Figure 9. Crystal from Terbium Gallium Garnet with target diameter showing a sudden transition to spiral growth because it has the tendency to a larger diameter by flaring. Reproduced with permission.^[3] Copyright 2016, Elsevier.

increase by flaring growth will be counteracted instantaneously by the automatic control unit by an increase of the crucible temperature. The fluctuations of the melt temperature will induce fine fluctuations of the clod diameter during the beginning of flaring growth. Small-diameter fluctuations will occur with extrusions at various and changing azimuthal positions, which are counteracted by temperature changes by the growth control unit. One can well assume that the largest extrusion of the crystal will grow under these fluctuating growth conditions because its growth is favored against that of the smaller ones by its stronger radiation cooling. The largest extrusion will survive in this selection process and will become the “foot” of the crystal, whereas the smaller extrusions will fade away. Less material will crystallize onto the opposite side of the foot compensating for the mass of the foot. An undercut will develop in this place. The formation of a foot together with its undercut represents the first symmetry breaking of the former cylindrical symmetry of the crystal. Other symmetry breakings follow during the course of growth; the crystal foot is no longer in the rotational center and drives as a boat on a circle on the melt surface. Because of this eccentric rotation, the meniscus of the foot will be deformed. It will be steeper in the direction of motion and flatter in the opposite direction. This is the second symmetry breaking. The flatter side of the meniscus points to a larger degree upward to the colder environment than the steeper meniscus side and so it is better cooled. The flatter side of the crystal will therefore grow faster in azimuthal direc-

tion than the steeper side of the meniscus. The foot will thus grow against the rotational sense of the crystal. The breaking of the rotational temperature distribution in the crystal meniscus is the third symmetry breaking. The left–right growth anisotropy is the fourth symmetry breaking. The superposition of the azimuthal growth of the foot with the axial growth by crystal pulling finally results in the spiral morphology of the crystal. This picture of spiral growth is confirmed by many details exhibited by the investigated crystals. It holds for high-temperature melting oxides and for silicon pulled with high speed.

A spiral can be characterized by its azimuthal width (spiral-ity) and the distance of its spiral turns. Both are influenced by the degree of cooling from above; the next spiral turn is always influenced by its predecessor. This can be called “partly self-organization.” Finally, I describe the retarded flaring growth observed at some high-temperature oxide crystal and its influence on the spiral shape.

The spiral shape of Czochralski crystals is not due to a new kind of instability but it goes back to the well-known phenomenon of flaring growth, which is forced into spiral growth by the automatic diameter control. Thermal fluctuations in the melt allow the selection of only one foot to grow as the beginning of a spiral.

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Conflict of Interest

The author declares no conflict of interest.

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