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Formation of the spiral morphology of high melting point crystals pulled from the crucible ☆

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ABSTRACT

This paper presents a description and a model for the transition from cylindrical to spiral growth during Czochralski crystal pulling of high melting point materials. This transition comprises a number of symmetry breakings of the rotational symmetry of the crystal where the first one is ascribed to flaring growth. Flaring growth occurs when the crystal-melt interface has gained a supercritical concave shape. The concave shape arises in case of high melting point semitransparent oxides mainly when the axial radiative heat transport through the crystal becomes insufficient – e.g. when exceeding a certain grown length – and it arises for silicon crystals when the growth speed was chosen too large. The spiral growth starts with the growth of the crystal axis off the rotational axis. This is followed by two more symmetry breakings deforming the melt meniscus-crystal area. The azimuthal growth of the deformed meniscus, together with its axial growth by pulling, results in the spiral morphology. The transition from cylindrical to spiral growth discussed here is a genuine growth-instability – one which is not induced by external disturbances – and it is limiting the cylindrical growth in a fundamental way.

1. Introduction

The growth with spiral morphology (as well called cork-screw-or twisted-growth) is observed in Czochralski growth of high melting point materials like YVO_4, GGG, $DyScO_4$, $Te_3Ga_5O_{12}$ etc. and for silicon under special conditions. Spiral growth is interesting and appealing but very unwanted. It has to do with the dominating radiative heat transport for materials with melting point above 1600 °C. Fig. 1 shows two examples of spirally grown crystals.

A dozen of papers describe spiral growth of various materials and offer some insight into the conditions for spiral growth, as well as in parts of its processes. They are reviewed in [1,2]. The understanding of spiral growth of high melting point oxides is by far not complete though Uecker et al. [2] identified low transmission of the crystal as one reason, and thus low radiative axial heat transport. This finding was based on the measurements and comparison of the optical transmission in the near infrared of some crystals at temperatures up to 1300 °C by Polity et al. [3]. Schwabe et al. [1] gave a rather detailed description of spiral growth once it has started, however they enumerate a large number of possible triggers of the spiral growth, one of which is the inaccurate mounting of the seed crystal. However, they gave no definite preference for one of these triggers. This question is still open and I will answer it in the present paper.

It was shown in [1] by a striation-figure of a Czochralski-grown $Te_3Ga_50_{12}$-crystal (Figure 3 in [1]) that the solid liquid growth-interface is concave towards the melt when the transition from cylindrical to spiral growth takes place. This is an important observation to clarify the question after the trigger (onset of spiral growth).

Very recently, Kalaev et al. [4] published their numerical work on spiral growth as “Crystal twisting in Cz-Si growth” occurring for the high pulling speeds which are of interest in the growth of semi-single crystal material for photovoltaic application. They could correlate a kind of super-cooling of the melt surface near the crystal-melt meniscus with the start of crystal twisting. Their rather reliable match between the experimental data and numerical simulation of the temperature-field in the Cz-system allowed quantitative insight into the transition to twisted growth. This match is not as convincing for the growth of high temperature oxide crystals, mainly because of experimental difficulties. We note that the findings of Kalaev et al. [4] includes as well the concave interface of the crystal near the triple junction (Figure 6 from [4]) at the beginning of spiral growth which matches with the observation of the concave interface shape of $Te_3Ga_50_{12}$ reported in [1].

I will clarify the problem of “onset of spiral growth” by introducing the well-known term of “flaring growth” and discussing its consequences for the growth of high temperature oxide crystals

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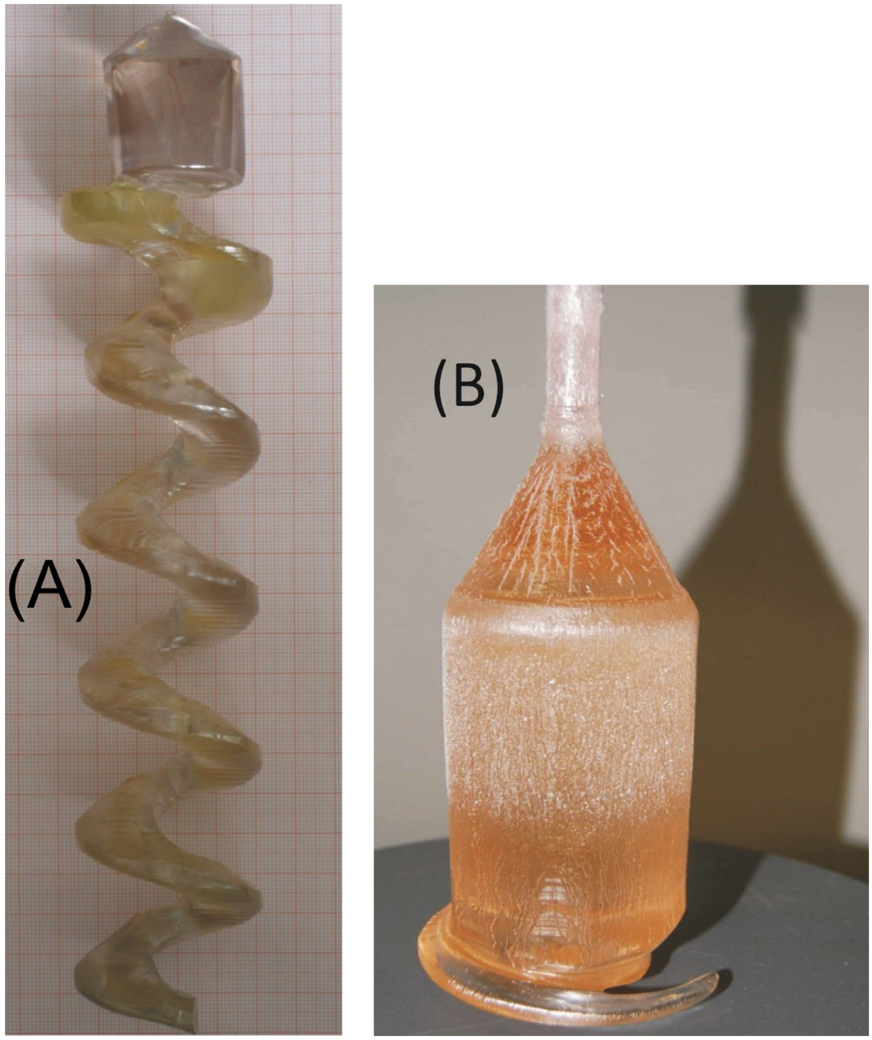

Fig. 1. The figure shows photographs of two spirally grown Cz-crystals of high melting point material which is semitransparent for the light emitted at temperatures near the melting point. (A) A long spiral from Yttrium Vanadate with extremely early beginning of spiral growth (From [1]). (B) crystal from Terbium Gallium Garnet with a diameter of 50 mm and a total length of 147 mm, showing a transition to spiral growth at the end when a certain length was reached (courtesy of Klaus Duprée, FEE, Idar-Oberstein, Germany).

under automatic control. Flaring growth without automatic control is defined as the growth with fast increase of the crystal diameter with the formation of clod (almost not controllable diameter-increase). I will point out that the transition of cylindrical growth to spiral growth is an instability in the sense of mechanical systems or hydrodynamic systems, occurring even under else perfect boundary conditions. This instability is limiting the cylindrical growth mode and it will become obvious that the material properties of the grown crystal limit our possibilities to avoid the instability.

2. Description of spiral growth

In this chapter I will summarise along [1] what is known about spiral growth until now before introducing a new piece of understanding.

During the transition of cylindrical Cz-growth to spiral Cz-growth the following symmetry breakings take place:

1. The crystal axis grows out of the rotational axis to a certain degree where Δr is the radial displacement between the two axes. This phenomenon is sometimes called “foot growth” in the literature. The cause of this foot growth was not known until now. A great number of possible causes have been discussed in the literature, summarised in [1], of which I mention only one as example, namely miss-orientation of the seed axis with respect to the rotational axis. All of the 17 “triggers” for spiral growth mentioned in [1] (and possibly more) might be operative in the one or the other case. We call them “external reasons for symmetry-breaking”. All of them could be avoided by a careful set-up of the growth system and one could assume that the spiral growth could be avoided by careful work. This is not true for all conditions. I will later present a symmetry breaking phenomenon which is fundamental (“internal reason”) and which will generate foot growth as the first step to spiral growth. This symmetry breaking occurs under certain conditions and constitutes an absolute limit of cylindrical growth.
2. It is clear that the asymmetry of excessive foot-growth must be compensated under automatic control by less crystallisation (e.g. on the opposite side of the foot). The crystal grows there with a partial necking-in (undercut), and this undercut is “trapped” by itself, because the necessary cooling by radiation is smaller in the undercut compared to that of the foot. The large radiative heat transfer of the high melting point materials is important for this behaviour.