Instabilities of the Flow and the Temperature Field during Crystal Growth of Silicon from the Melt

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Summary

Silicon crystals have been grown from free melt zones. Radiation heating was used to establish the liquid zone. Specially prepared temperature sensors have been inserted into the melt to investigate the temperature field and the state of the convective regime. Strong convective flows are present, mainly forced by gradients of the surface tension and, to a lower degree, driven by buoyancy related effects. Rotating magnetic fields have been applied to improve the heat and mass transport in the melt and to reduce the strength of the convective flows. The flow regime was analyzed by three-dimensional numerical simulations (which mad the time-dependent, three-dimensional character of the convection clear), by the study of the temperature measurements (irregular temperature fluctuations up to 1 K have been measured), and by the characterization of the grown crystals (the convective flow causes irregular fluctuation of the composition and the electrical resistivity). Due to the rotating magnetic field, an axi-symmetric, quasi-steady flow was generated and the flow instabilities have been greatly reduced. The required magnetic field for the transition from time-dependent, irregular flow to the quasi-steady one is in the range of a few millitesla (at 50 Hz), the required strength depends on the melt diameter and aspect ratio.

Introduction

Molten silicon, like most semiconductor melts, is a low Prandtl number liquid. This means that it exhibits a high thermal conductivity (and a high electrical conductivity) and a low viscosity: The transition from laminar to oscillatory and to time-dependent flow takes already place for small temperature differences in the melt, if they destabilize the density gradient or act on a free melt surface. The high electrical conductivity favors the use of magnetic fields for the stabilizing or the relaminarization of convective flows [1]. For most semiconductor growth techniques from the melt, like Float Zone or Czochralski method, the melt is affected by strong, in most cases time-dependent, convective flows. As a consequence, instabilities of the flow, the temperature, and the concentration field occur, which result in instabilities of the solidification speed and the compositional homogeneity of the grown crystal.

Liquid silicon shows a melting point of 1420°C and is an opaque and chemically aggressive liquid. Direct measurements of the flow velocity are restricted to rather complicated x-ray techniques [2]. They can, due to the use of tracer particles, follow the

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flow only to a certain degree. Therefore, mainly the temperature field is accessible for direct measurements. Due to the high working temperatures and the reactivity of silicon, the temperature sensors have to be encapsulated (with the drawback of reduced sensitivity) or carbon coated optical thermometers might be used, although they have a lifetime of only a few minutes.

In the case of floating zone growth (the only commercial technique for the production of silicon single crystals beneath the Czochralski method), just a small part of the feed material is molten at a time; a free melt zone, held only by its surface tension, is floating between the growing crystal and the (polycrystalline) feed rod. This molten zone moves relative to the crystal / feed rod by pulling down the crystalline parts. The basic concept of the float zone technique is shown in Fig. 1, on the left hand side for a radiation or conduction heated configuration and on the right hand side for the "needle-eye" arrangement established in the commercial production of silicon.

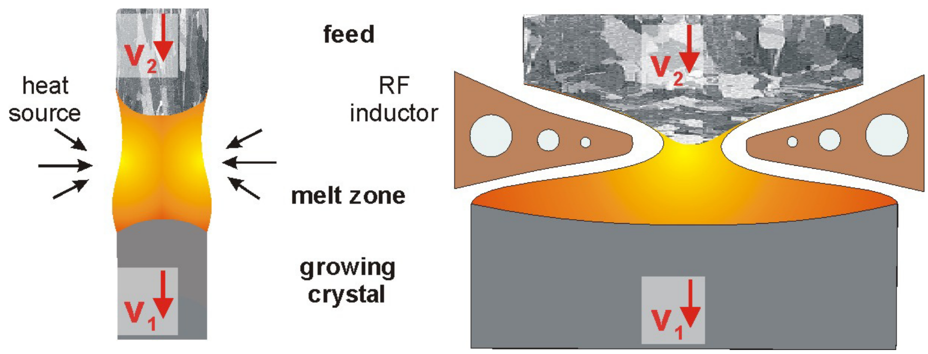

Fig. 1: Basic concept of the float zone process, on the left hand side for a conventional set-up heated by radiation or conduction and on the right hand side for the "needle-eye" technique. The growing crystal moves downward relative to the heater with a velocity v_1, which is in most cases independently controllable from the velocity v_2 of the feed rod.

One of the fascinating features of the FZ technique is that it is a completely containerless method; neither the growing crystal nor the melt are in contact with any other material. The advantages are manifold: high purity crystals can be achieved (silicon with low oxygen content is exclusively produced by the FZ process), stress is reduced, aggressive substances can be handled, or high melting point materials might be processed.

Experimental arrangement

The experiments have been performed with radiation heated silicon float zones, the diameter of the zones was in the range of 8 to 14 mm, their height between 8 and 12 mm. A 1000 W halogen lamp served as radiation source (so called "mirror furnace arrangement"; for details see e.g. [3]). The temperature difference within the melt amounts to 5 to 20 K, depending on the geometry of the zone. To avoid the oxidation of the molten silicon, an argon inert gas atmosphere was used. Typical solidification velocities for float zone silicon are in the range of 1 to 5 mm/min. After the growth experiments, the samples are cut lengthwise parallel to the (110) plane and prepared in a manner to exhibit the homogeneity of the compositional distribution.

For active control of the flow regime in the molten silicon, a rotating magnetic field device was mounted outside the growth chamber. It consists of three coil pairs capable to produce a magnetic induction of 7.5 mT at a fixed rotation rate of 50 Hz.

Flow configuration

Numerical simulations of radiation heated silicon float zones demonstrate the asymmetric, three-dimensional, time-dependent behavior of the flow regime, consisting of a large, diagonal flow roll in addition to some smaller ones (Fig. 2, left hand side) [4]. Flow velocities as high as 15 cm/sec are reached with the consequence of a symmetry break in the isotherms. The flow velocity as well as the spatial arrangement of the flow rolls and the temperature field fluctuate with time. It changes drastically when a rotating magnetic field is acting on the fluid (Fig. 2, right hand side): The radial and axial flow components are considerably reduced, now forming nearly axisymmetric tori, in favor of a high azimuthal flow with maximum velocities of u_φ=14 cm/s. The isotherms become axisymmetric

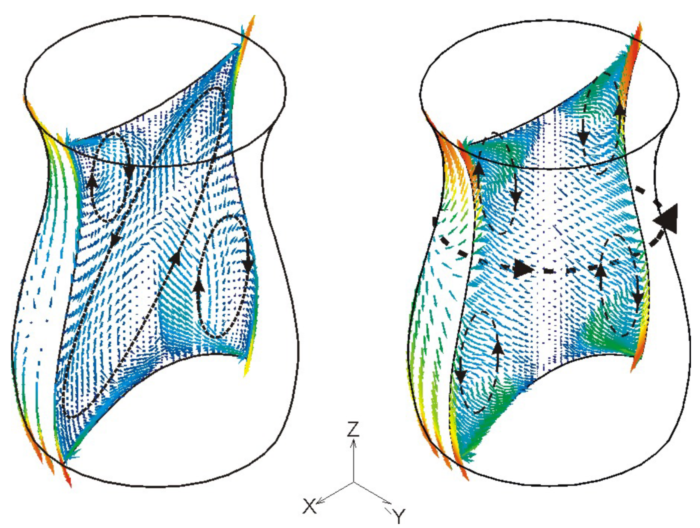

Fig. 2: The numerical simulations of silicon float zones reveal a three-dimensional, time-dependent flow structure with flow velocities as high as 15 cm/sec (left hand side). The isotherms are formed non-axisymmetric due to the irregular flow. 3D numerical simulation of a silicon float zone with aspect ratio = 1.5. Applying a rotating magnetic field, the flow field (as well as the isotherms) become axisymmetric with the main flow components pointing now in the azimuthal direction [5].

Thermal instabilities in the melt zone

A graphite coated quartz glass sensor (sensor diameter: 300 µm) was inserted into the melt to measure directly the temperature and the convectively induced temperature fluctuations in the molten silicon. The sample rate was 20 Hz. Without an external magnetic field, convective temperature fluctuations reach peak-to-peak values of 1K, mainly situated in the frequency range of 0.05 to 0.3 Hz. Frequencies above 0.5 Hz are present only with weak intensities. The intensity of the convectively induced temperature fluctuations drops dramatically under the influence of 7 mT / 50 Hz, the fluctuation rate is reduced by more than an order of magnitude. The frequency of the fluctuations is changed from 0.05-0.3 Hz (no field) to 2.6 Hz (7 mT / 50 Hz). There is an intermediate flow state between the initial one and the one dominated by the rotating magnetic field. This state exhibits fluctuations with frequencies higher than the original ones but comparable amplitude: The three-dimensional flow character is still present, the large flow roll seen in fig. 2 on the left hand side is now rotating in azimuthal direction under the influence of the magnetic field but it is still intact. Above the threshold value of 4 mT, the large diagonal roll breaks and only small, axisymmetric poloidal tori remain which rotate with a high azimuthal velocity.

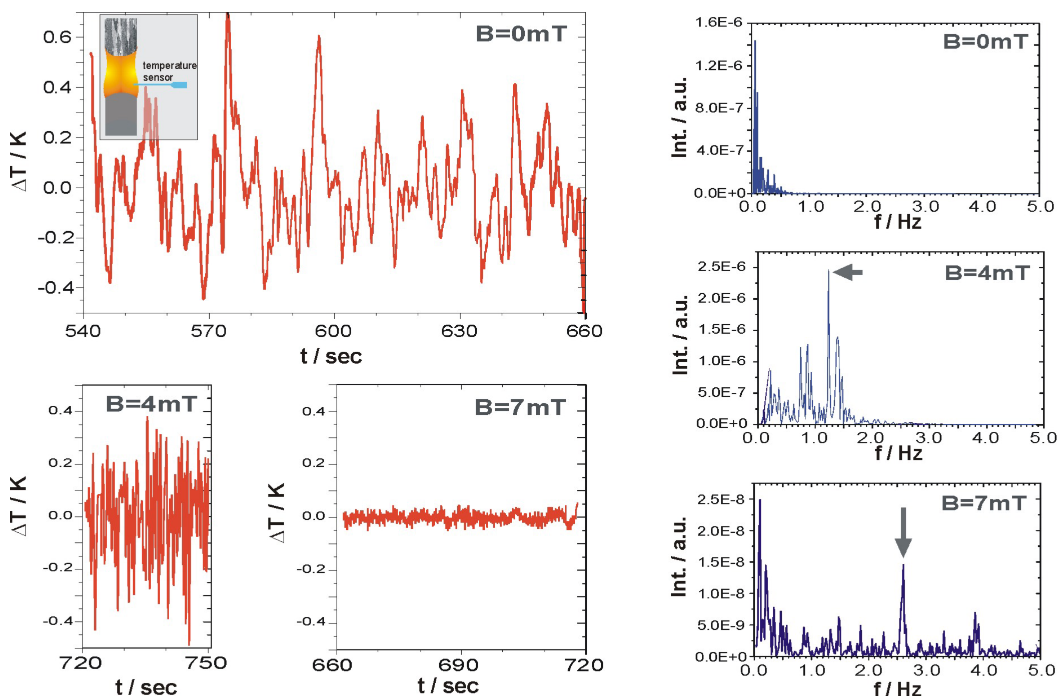

Fig. 3: Performing temperature measurements in the silicon melt (melt diameter: 10 mm), the threedimensional, time-dependent character of the flow field becomes obvious. The flow instabilities are strongly reduced under the influence of a rotating magnetic field.

Impact of the flow instabilities on the concentration field of the crystal

Flow irregularities are reflected in the grown crystal by variations of the composition on the microscale. These concentrational inhomogeneities are bands (so called striations) formed perpendicular to the growth direction. The width of these irregularities ranges from a few micrometer up to some hundred micrometer, depending of the flow regime. As a function of the segregation coefficient, the peak to peak values of the concentration fluctuation can reach values of 50% and higher. These irregularities of the composition are visualized by chemical etching of polished crystal slices. Fig. 4 shows four sections of a crystal grown under different magnetic field strengths. The magnetic induction was altered several times during the growth run which had no effect on the zone shape or the diameter control of the crystal. The aspect ratio (height to diameter ratio) of the floating zone was 1.

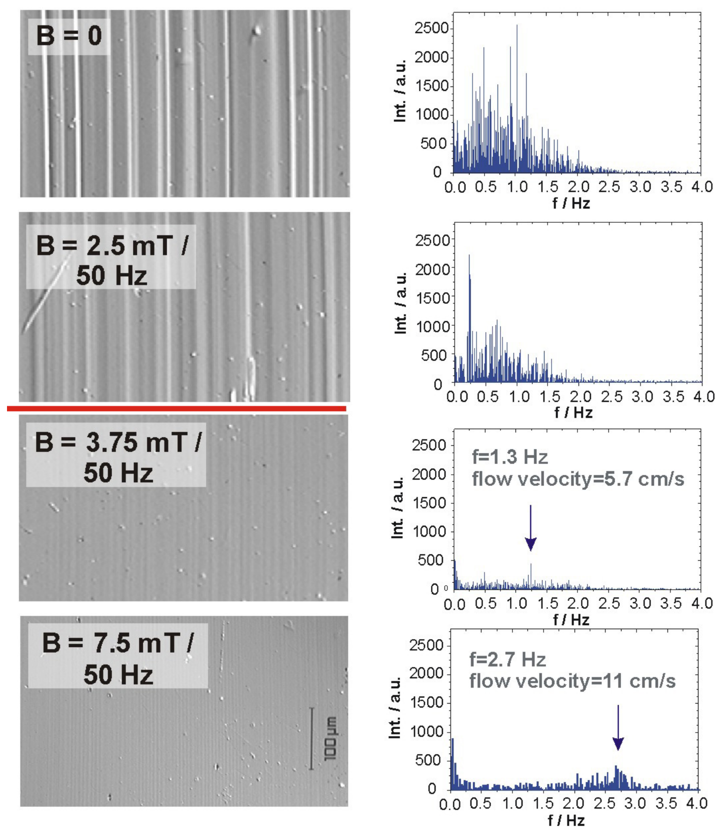

Fig. 4: Striation patterns (left column) and corresponding frequency distribution (right column) of an antimony doped silicon crystal grown under different rotating magnetic field strengths. The most significant change in the intensity of the dopant non-uniformities was found between 2.5 and 3.75 mT / 50 Hz (sample diameter: 14 mm) and shifts to higher values for smaller samples.

Without magnetic field (Fig. 4, top), the distribution of the dopant non-uniformities is irregular. The frequency spectra as shown in Fig. 4 were calculated from the brightness signal of a trace taken perpendicular to the striations and transformed into the time-domain by using the average translation rate of 1mm/min. They reveal no specific frequencies but a broad distribution between approximately 0.1 and 2 Hz. This corresponds to a thickness of the dopant striations between approximately 8 and 170 μm. A strong reduction is evident in the parts grown with a rotating magnetic field, as can be seen in the sections 3 and 4 from the top in Fig. 4. The signal intensity and the frequency spectra change only slightly by applying a RMF of 2.5 mT, but a strong transition in the characteristic microstructure of the dopant distribution is found between 2.5 and 3.75 mT. A drastic reduction of the intensity of the nonuniformities is observed and the remaining striations show a spacing of approx. 12 μm, corresponding to a characteristic frequency of 1.3 Hz. Increasing the magnetic induction to 7.5 mT, the characteristic frequency shifts to ≈ 2.7 Hz (equivalent to 6 μm). Assuming that these quasi-periodic, high frequency striations result from the flow induced by the RMF, i.e. each dopant striation corresponds to one azimuthal revolution of the fluid driven by the rotating Lorentz force, one can easily calculate the maximum azimuthal flow velocity. The maximum velocity is reached at the free surface (because of the slip condition) resulting in a flow velocity of approximately 5.7 cm/s in the case of 3.75 mT and 11 cm/sec for 7.5 mT. The frequency of the dopant striations is in a very good agreement with the temperature measurements shown in Fig. 3 taking into account the different melt diameters.

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