This article describes the historic development of the Erlangen Crystal Growth Laboratory CGL from its beginnings in 1974 at the chair of Materials of Electrical Engineering (Department of Material Science) of the University of Erlangen–Nuremberg until its current status as a large department “Materials” of the Erlangen Fraunhofer-Institute for Integrated Systems and Device Technology. Essential developments and scientific achievements in the various fields of crystal growth and epitaxy are presented from the early period until today.

1. History of Semiconductor Crystal Growth in the Erlangen Area from the Beginnings around 1950 Until Today

Very soon after the development of the first functional transistor in the year 1947 several research activities were started at different places in the Erlangen area to investigate semiconductor materials and their preparation. It turned out very quickly that these materials needed an extremely high purity and a very high crystallographic perfection, that is, it was necessary to develop new methods for purification of these materials and for growing single crystals.

The Siemens cooperation, which had installed its new headquarters and laboratories in Erlangen immediately after World War Two, started several research programs to develop the new semiconductor materials germanium, silicon, and semiconducting compounds. Especially, in a remote castle in the small village Pretzfeld near Erlangen a working group headed by Eberhard Spenke and Walter Schottky began to explore germanium and silicon. Around 1954 this group invented the process of the fabrication of highly pure semiconductor silicon which is used worldwide until today to produce annually more than 400,000 tons of silicon feedstock for the microelectronic and photovoltaic industry. Since that time this process is named “Siemens Process”[1]: Metallurgical silicon transformed by chemical reactions into chlorosilane compounds which are purified by multiple distillation steps and finally thermally decomposed to polycrystalline high purity silicon. The same group in Pretzfeld also invented the floating zone (FZ) technique and grew the first silicon single crystals by this method in 1954.[2] (It should be mentioned that nearly at the same time the floating zone method was also invented independently by Keck and Golay in the United States.)

In the year 1951, Heinrich Welker became a co-worker of Siemens with the position of the head of the solid-state physics research laboratory in Erlangen. He immediately started with the investigation of the III–V compound semiconductors, which he had also discovered.[3,4] Very soon the first single crystals of InSb, GaSb, AlSb, InAs, and GaAs were grown by Welkers’ co-workers. The growth of GaAs required the development of a new technique with an arsenic vapor source in order to prevent the decomposition of the melt and of the grown crystal. This problem was solved by Gremmelmeier and his colleagues[5] in a kind of hot-wall Czochralski set-up in 1956. The crucible, the crystal with the pulling rod, and the As source were arranged within a sealed silica glass ampoule and heated from the outside. Several years later in 1962 this growth technique was replaced by the liquid encapsulated Czochralski (LEC) method (invented by Mullin[6]), where the melt and crystal surface are covered by a layer of liquid boric oxide in order to prevent the decomposition by evaporation of As. Welkers’ group then developed such LEC pullers in-house and used them also for the high-pressure growth of InP and GaP crystals.

Parallel to the semiconductor activities of the Siemens engineers, specific research programs on semiconductor crystals were carried out at the Friedrich–Alexander University of Erlangen–Nuremberg (FAU). Since 1948, Erich Mollwo, the head of the Institute of Applied Physics, worked in the field of electronic and optical properties of II–VI compound semiconductors, especially ZnO. Mollwo, who was an earlier Ph.D. student and research assistant of the famous Robert Wichard Pohl in Göttingen, recognized very soon, that a prerequisite of his research was the...
availability of high-quality single crystals. Therefore, his research assistant Reinhard Helbig developed the growth of ZnO crystals from the vapor phase. Later on (from 1969 to 1974) Helbig introduced the Ph.D. student Georg Müller into the field of crystal growth.⁷

Erich Mollwo together with his colleagues from the physics department of the FAU, and especially Helmut Volz, a professor of theoretical physics, strongly promoted the idea of establishing an engineering science department (“Technische Fakultät”) at the university in Erlangen. This effort was successful and the study program “Materials Science” could be established in 1966, being the first of its kind in Germany. The department of Materials Science consisted of six chairs, each of them dedicated to a certain group of materials or material properties, respectively.

In 1973, Herbert Weiß, an earlier Ph.D. student of Erich Mollwo, was appointed as the first professor of the chair of “Materials of Electrical Engineering.” He worked for several years in the Siemens laboratory of Heinrich Welker. He was very familiar with III–V compound semiconductors, but also with silicon and related crystal growth problems. In 1974, Weiß established “Crystal Growth” as a field of research and appointed Georg Müller as a post doc to head this group named “Crystal Growth Laboratory” (CGL). The research concept of the new group CGL had its focus on the study of phenomena related to crystal growth processes, especially melt growth of semiconductor crystals. Initially, it was not intended that CGL should grow and supply crystals for other research groups. Instead, its working program should focus on studying basic phenomena in industrially relevant melt growth processes, for example, the occurrence of convection and its impact on the properties of the growing crystal.

At that time a critical industrial problem for the fabrication of semiconductor devices (both for microelectronics as well as power electronics) was the occurrence of so-called doping striations (see, e.g., ref. [8]) in silicon and compound semiconductor crystals. The correlation of unsteady melt convection and the formation of doping striations became an important research topic of CGL. Within this context the important role of gravity-driven “natural” convection (buoyancy) acted as the main motivation for CGL to participate in the German and European space program for materials research under microgravity conditions. These activities were strongly promoted by Herbert Weiß. Several series of experiments in various Spacelab missions (1. Spacelab Mission (1983), Spacelab D1 Mission (1985), Spacelab D2 Mission (1993)) as well as sounding rocket flights were prepared by CGL researchers, which will be described in more detail in Section 3.2.

Already in the early working period of CGL it was recognized that modeling and simulation of transport processes can be very helpful for understanding and improving crystal growth processes. Intensive studies on heat and mass transport, thermoelastic stress, plastic deformation, and their relations to the formation of crystal defects were carried out in the following years. Its nationally and internationally acknowledged results led to an increasing number of collaboration projects with industrial partners.

In this situation it turned out to be very advantageous that Heiner Ryssel, the director of the Erlangen Fraunhofer-Institute for Integrated Systems and Device Technology (IISB) (founded 1985) at that time, provided the opportunity for Georg Müller in 1996 to establish a working group “Crystal Growth” at Fraunhofer IISB. Already in 1999 this group received the status of a department. Since 2004, the department “Crystal Growth” is headed by Jochen Friedrich, an earlier Ph.D. student of Georg Müller. In 2005, he extended these activities via the foundation of a branch lab in Freiberg (Saxony) named Technology Centre Semiconductor Materials (THM). With his retirement in 2007, Georg Müller has completely transferred the crystal growth activities of CGL to the department at Fraunhofer IISB, which changed its name from “Crystal Growth” to “Materials” in 2014. At Fraunhofer IISB, the research in the area of crystal growth and epitaxy is not only continued until today, but has steadily grown. This may be underlined by the development of the number of co-workers, which is compiled in Table 1. The broad spectrum of materials, growth techniques, and research topics which were studied by CGL from the beginning until today is shown in Figure 1.
Table 1. Number of co-workers at CGL including students.

| Year | 1980 | 1990 | 2000 | 2010 | 2020 |
|------|------|------|------|------|------|
|      | 10   | 15   | 30   | 40   | 60   |

Figure 1. Materials, growth techniques, and research topics versus time studied by CGL and at Fraunhofer IISB (Copyright Fraunhofer IISB).

2. The Erlangen Research Concept of Crystal Growth

The fact that CGL was established as an independent unit without any obligations to supply other groups with crystal material turned out to be very advantageous because it allowed the definition of its own research program in the field of crystal growth. The new Erlangen research concept focused on the study of fundamental aspects of the techniques of growing crystals (“Kristallzüchtung”). This approach is different from the traditional topics of basic research in crystal growth which had its focus in the past mainly on phenomena related to growth kinetics (“Kristallwachstum”). Famous scientists like Kossel, Stranski, Vollmer, Weber developed this research field of crystal growth by their outstanding contributions in the first half of the previous century. Even Jan Czochralski, whose name was much later given to the crystal pulling technique, was primarily interested in the measurement of the growth kinetics (“kinetic velocity”) of metals in his famous work.[9]

Studying the technology of growing crystals was an approach which complemented the overall concept of the young Erlangen university department of materials science very well, where similar research strategies were developed for the manufacturing of metallic alloys, ceramics, glass, and polymer materials by other chairs. A dedicated application of a material requires certain specific properties, which are defined by its microstructure and depends therefore on the conditions of the production process. Only a precise knowledge of the physical-chemical mechanisms occurring during the production process and its relation to the materials’ microstructure allows tailoring of its properties. This general concept of materials engineering was now transferred to crystal growth by CGL.

The starting point is the use of the crystal or the substrate manufactured from the crystal, in Erlangen mostly a semiconductor for an electric or optoelectronic device. This application defines the specific crystal properties. They correlate with the microstructure of the material, that is, deleterious defects (typically dislocations, grain boundaries, impurities, etc.) and also beneficial defects (typically uniformly distributed doping atoms or sometimes precipitates). The formation of these defects depends on the thermo-physical conditions during the growth process and cooling down of the grown crystal. Further important issues of a crystal growth process are the crystal shape, its size, the yield, as well as other economic aspects.

This overall research strategy is shown in Figure 2. The profound study of the interrelations, which are sketched in this figure, resulted in the fields of research which will be discussed in the following sections in more detail:

- Fundamental aspects of heat and mass transfer in crystal growth configurations, especially convection phenomena in melts and their correlation to crystal non-uniformity
- Modeling of crystal growth processes and defect formation, for example, dislocations
- Development of crystal growth processes and growth apparatus, for example, vertical gradient freeze (VGF)
- Development of specific tools for process analysis and defect characterization, for example, in situ temperature measurement, in situ oxygen detection in Si-melts, mapping of dislocations, mapping of grain structure

Working on these topics was mainly carried out by students within the frame of their materials science study program, study theses (now bachelor), diploma theses (now master), Ph.D. theses, and by post docs. In this regard it was a big advantage that CGL was a part of the university department of material science and that crystal growth was fully embedded as a subject in this study program. In this respect Erlangen was one of the few universities worldwide which had lectures on crystal growth in the
study program of the main course. In total, up to now, about 300 theses (study/bachelor, diploma/master) and 50 Ph.D. theses were successfully carried out in the field of crystal growth in Erlangen. The contributions of Erlangen crystal growers were honored by a respectable number of research prizes and awards.

3. Fundamental Studies of Convective Phenomena in Melts and their Influence on Crystal Growth and Defect Formation

3.1. Melt Convection and Crystal Inhomogeneities (1974–Today)

The study of convective flow phenomena in semiconductor melts and their influence on crystal growth was one of the major research subjects in the first decade of working since the beginning in 1974. The motivation for this field of work came from the industrial device production. Yield problems in the fabrication of micro- as well as of power electronic devices were caused by periodic variations of the resistivity (so-called “doping striations”) in silicon as well as in compound semiconductor crystals.

Very soon a clear correlation was identified between these striation-like doping non-uniformities and temperature fluctuations in the melt during growth, as depicted in Figure 3. The temperature fluctuations are caused by unsteady convective flows in the melt which can have different origins such as buoyancy, capillary and forced convection or an interaction of them. The study of convective flow phenomena was carried out both experimentally and by theoretical considerations.

Model experiments with transparent fluids were used for a visualization of flow structures as a function of boundary conditions, which are similar to melt growth configurations. However, soon it became very clear that the study of the structure and behavior of flows in the melt of semiconductors (which behave like metallic liquids) strongly would benefit from the use of computational fluid dynamics. Two approaches were pursued:

i) The mathematical one, which means the solution of the differential equations describing the heat and mass transport by using numerical tools;
ii) The use of dimensionless numbers, which have been developed in engineering science to provide an order of magnitude analysis. In fact, such numbers can indicate the dependence of the flow behavior from a certain parameter of a crystal growth configuration.

At the beginning, the Erlangen crystal growers were not so familiar with these two approaches. Thus, collaborations with experts in the field of fluid dynamics like Franz Durst in Erlangen and the Munich mathematician Karl-Heinz Hofmann have been very beneficial and productive. Very soon the first scientific results could be published and presented at international conferences. Invitations to the famous Gordon Conferences (USA) and to Robert Brown (Massachusetts Institute of Technology (MIT), USA) followed, as well as an extended stay of Günter Neumann at MIT in the mid 1980s. The close relationship of Erlangen crystal growers to Jeffery Derby (University of Minnesota) dates back to this early collaboration when he was a Ph.D. student at MIT. Later on, the Bavarian network FORTWIHR provided a very strong support for collaborative projects and the development of modeling capability at CGL. Also, the first international meeting specifically on modeling of crystal growth processes was initiated and co-chaired by Georg Müller. The “International Workshop on Modeling in Crystal Growth (IWMCG)” held in 1989 in Parma, Italy. Meanwhile the ninth repetition took place in Hawaii in 2018.

Numerical modeling became an important issue in crystal growth in the period after 1980. Very soon, it was recognized that the available software for simulation of crystal growth processes was not well adapted for experimentally working crystal growers. Therefore, CGL decided to start with the development of its own software especially designed for crystal growth processes and user friendly for crystal growers. This work is related to several earlier Ph.D. students and CGL co-workers like Thomas Jung, Hans-Jörg Leister, Jakob Fainberg, Matthias Kurz, Michael Metzger, Marc Hainke, and Johannes Dagnor who contributed to the software development. In this relation also a close collaboration with the University of Illinois started, especially with Daniel Vizman and his former colleagues like Artur PusztaI and Gheorghe Ardelean. The resulting software packages STHAMAS and CrySVU and the later combined CrySMAS were licensed worldwide and used by companies and research institutes until today. Figure 4 shows the cover page of the simulation prospect of IISB from 2007 highlighting the software packages of IISB. Their use and important results will be shown in several of the following sections.

Let us come back to the application of dimensionless numbers in crystal growth, which was started in parallel to the development of numerical modeling. It can be useful for an order of magnitude analysis of transport phenomena in crystal growth, also with respect to unsteady behavior (“striation problem”). The first studies were carried out to identify which dimensionless numbers can be used adequately for certain geometry and boundary conditions of crystal growth. For example, an approach was developed which allowed for the first time to plot the thermal boundary conditions for various melt growth configurations like VGF, Czochralski, zone melting, casting within one diagram.

The influence of convective flow on the properties of a growing crystal is clearly related to the occurrence of non-uniformities of the distribution of doping elements. The related segregation phenomena are typically described by the distribution or segregation coefficient \( k \). Although \( k \) is only defined by phase diagrams, that is, equilibrium thermodynamics, crystal growers are...
Figure 4. Cover page of the simulation flyer of IISB from 2007. At that time three simulation tools were licensed: CrysVUn, STHAMAS, and STHAMAS 3D. (Copyright Fraunhofer IISB).

looking for a possibility to include the action of the growth velocity and convective flow on the segregation behavior. The most popular model at that time was that of Burton, Prim, and Slichter.\textsuperscript{[18]} They defined an effective segregation coefficient $k_{\text{eff}}$ which considers the growth rate. $k_{\text{eff}}$ has also some kind of fit parameter (convection-free boundary layer thickness). However, this model of $k_{\text{eff}}$ can only be used to describe existing segregation results by a fit of this boundary layer parameter. It is not successful on the prediction of segregation results in dependence of convective flow and the boundary layer thickness in front of the growth interface. This boundary layer topic was discussed in common publications of Franz Rosenberger and Georg Müller in 1983.\textsuperscript{[19]} Later, during a sabbatical stay Alexander Ostrogorsky developed at CGL in Erlangen in collaboration with Georg Müller a concept of $k_{\text{eff}}$ which quantitatively considers the influence of convective flows in the vicinity of the growth front.\textsuperscript{[20]}
The overall conclusion of the research efforts to control the uniformity of a growing crystal was the realization that it is necessary to control the flows in the various melt growth configurations in a quantitative manner. It was also recognized that semiconductor melts behave like liquid metals. This means a strong tendency for transition from steady to unsteady behavior. In the search for options to control the flows in such melts, various means were developed and studied at CGL:

i. Selection of crystal growth with advantageous hydrodynamic conditions like VGF with bottom seeding and axial heat flux (compare ref. [21] and see Sections 4.2–4.4 and 5.1)
ii. Crystal growth under microgravity conditions in space missions (see Section 3.2)
iii. Crystal growth under high gravity conditions on centrifuges (see Section 3.3)
iv. Crystal growth under the action of magnetic fields (see Section 3.4)

3.2. Crystal Growth under Microgravity Conditions (1974–Today)

Already in the year 1975, Herbert Weiß, the chair of “Materials of Electrical Engineering” at the Department Material Science of the University Erlangen–Nuremberg at that time, took on the task of a coordinator of the German program named “material research under space conditions.” He also contributed to this program by an own experiment proposal which should be elaborated by his new scientific co-worker Georg Müller. This space experiment which was scheduled to be carried out during the first Spacelab mission was concerned with the study of the eutectic texture during directional solidification of the semiconductor material InSb-NiSb. The average distance of the segregated NiSb needles should depend on the transport conditions in the melt and on the rate of solidification. But, the famous theory of Jackson and Hunt[22] was only existing for purely diffusive transport in the melt. Such conditions could only be provided under microgravity. Successful experiments were carried out during two space shuttle missions, Spacelab 1 (1983) and Spacelab D1 (1985), and in several rocket flights within the German TEXUS program. The microgravity results were used to calibrate the theory of Jackson and Hunt.

Even more spectacular crystal growth experiments from CGL were carried out in space onboard the German Spacelab Mission D2 in 1993. During this shuttle flight several GaAs crystals were grown by the floating zone method controlled via tele-science from ground. In the German Space Operation Center in Oberpfaffenhofen, Georg Müller and his Ph.D. student Frank Herrmann could observe on a monitor screen the free-floating melt zone and the diameter of the growing crystal and controlled the growth process via telecommand.

In advance of the D2-GaAs microgravity experiments, several preparatory investigations, which were mainly carried out within the Ph.D. thesis of Roland Rupp, were needed, especially with respect to the free surface of the GaAs melt and the related capillary convection (so-called Marangoni convection):

- Measurement of the surface tension of GaAs melt in dependence from the As vapor pressure by the sessile drop method[23]
- Study of Marangoni convection in GaAs melt zones and the determination of the critical Marangoni number for the transition from steady to unsteady capillary convection in dependence from thermal boundary conditions[24]
- Study of the impact of magnetic fields to suppress unsteady Marangoni convection[25]

As a result of these preparatory investigations, GaAs crystals were grown during the German Spacelab Mission D2 under microgravity conditions which had purity higher than any GaAs crystal grown on earth. Furthermore, the D2-GaAs crystals with a diameter of 20 mm set the world record with respect to the largest GaAs crystal grown by the floating zone method until today.[26] Under normal earth gravity it is not possible to stabilize a floating melt zone of more than 8–9 mm diameter due to the large density of the GaAs melt and its relatively small surface tension (see also Figure 5).
After the German Spacelab Mission D2 in 1993 the long ranging tradition of space experimentation with participation from Erlangen keeps on until today (see Figure 5). Around the year 2000, CGL was entering the field of solidification of metallic alloys in relation to materials research under microgravity conditions. CGL initiated an international European research project of academic and industrial partners, called MICAST\(^{[27,28]}\) (microstructure evolution in casting of technical aluminum alloys under diffusive and controlled convective conditions) which was also headed for several years by Georg Müller. An important goal of MICAST was to use the possibilities of microgravity conditions onboard the International Space Station for providing well-defined “diffusive transport conditions” during the solidification of alloys. The “controlled convective flows” should be generated by time-dependent external magnetic fields. The main task of CGL within the MICAST project consisted in a development of the modeling tool. For this purpose, the modeling capabilities of CGLs’ CrysMAS software had to be expanded. This means that in addition to the macroscopic transport and segregation mechanisms new phenomena had to be considered and implemented, like nucleation, formation of new phase, and grains. This work was mainly carried out within the frame of the Ph.D. theses of Marc Hainke and Johannes Dagner. The experimental verification of the simulations results occurred by comparison to data achieved by the group of Lorentz Rake (DLR Cologne) with the binary model system Al-Si. A major subject of CGLs’ studies was the analysis of the influence of time-dependent magnetic fields on the segregation during alloy solidification.\(^{[29,30]}\) The results were used to define the magnetic field experiments in the so-called ARTEX facility which was flown onboard the TEXUS 41 (2004) and MAXUS 7 (2007) sounding rocket missions. The simulation results of the solidification parameters agreed quite well with the experimental data of these space experiments.\(^{[31,32]}\) The successful simulation work resulted in the decision of ESA to commission CGL with the development of a Thermal Modeling Tool (TMT) for modeling of engineering furnaces and sample-cartridge assemblies of the Material Science Laboratory onboard the International Space Station.\(^{[33]}\) Until today, the TMT from CGL, which is basically CrysMAS, is used by several European research groups to support their solidification experiments onboard the International Space Station.

Motivated by the research on the directional solidification of multicrystalline silicon (see Section 5.1) Jochen Friedrich and his co-workers started a research program in order to study the incorporation of foreign phase particles into silicon during crystal growth by experiments onboard the sounding rocket missions TEXUS 51 (2015), TEXUS 53 (2016), TEXUS 55 (2018). In these experiments it was found the melt flow has a strong impact on the engulfment of the foreign phase particles.\(^{[34,35]}\)

Now a new experiment series for analyzing defect formation in correlation with the growth of silicon crystals with faceted interface under microgravity conditions has recently been started as an outcome of the applied research on the defect formation during industrial production of heavily doped silicon crystal by the Czochralski technique (see Section 4.5).

3.3. Crystal Growth under High Gravity Conditions on Centrifuges (1974–1996)

In the early period of preparing the microgravity experiments (1975–1980), it turned out that the realization of the first flight experiments was delayed year by year due to technical problems of the space shuttle. In the absence of flight opportunities, the Erlangen crystal growers conceived the idea to study convection phenomena in crystal growth configurations under high gravity on centrifuges.\(^{[36]}\) The idea was that eventually microgravity results could be anticipated by extrapolation from higher gravity levels.

Two centrifuges were constructed and built by the university workshop as well as a crystal growth apparatus for high gravity conditions. The setups for zone melting in horizontal boats and VGF had a turnable mounting support, which caused the crystal growth setup always to be directed into the resulting acceleration (= vector sum of gravity and centrifugal acceleration).\(^{[37]}\) Figure 6 shows pictures of Erlangens’ large centrifuge with 3 m diameter with the turnable VGF furnace.

The action of “gravity”-driven convective flows and its transition to unsteady behavior could be studied systematically by the variable centrifugal force.\(^{[37–39]}\) Also, relations to microgravity conditions were possible.\(^{[8]}\) Furthermore, it was surprisingly found that bifurcations exist between convective flow states which are only observable on centrifuges.\(^{[11]}\)

The influence of convective flow by a higher centrifugal acceleration was also used to enhance the species transport in solution growth. The transport limited growth rate for the inclusion-free growth of GaSb crystals from a Ga-rich solution by the Traveling
Heater Method (THM) could be increased by one order of magnitude up to a rate of nearly 1 mm h\(^{-1}\) at an acceleration of 20 times earth gravity.[\textsuperscript{40}]

Furthermore, Jochen Friedrich could clarify in his Ph.D. thesis the occurrence of the so-called “magic-g-level”[\textsuperscript{41}] which had been reported in the literature some years before.[\textsuperscript{42}] The term “magic-g” refers to the observation of these authors that crystals which were grown at certain gravity levels had a higher uniformity than those which were grown at lower or higher gravity levels. Jochen Friedrich could show that, in fact, a minimum of the segregation-induced non-uniformity can occur at intermediate levels of centrifugal acceleration.[\textsuperscript{41}]

As the research of crystal growth phenomena under high gravity conditions on a centrifuge can be considered merely basic research these activities were not continued when CGL became part of Fraunhofer IISB.

3.4. Convection and Crystal Growth under the Action of Magnetic Fields (1980–Today)

The high electrical conductivity of semiconductor melts enables a strong interaction of convective flows and magnetic fields. The convective transport in crystal growth configurations can be controlled, reduced as well as enforced via the induced Lorentz force. These options were intensively studied in Erlangen from the very beginning until today.

Already in 1980, in one of the first study thesis experiments with a rotating magnetic field (RMF) were carried out to enhance the mixing in the solution and thus to increase the growth rate for inclusion-free growth of GaSb by the THM[\textsuperscript{39}] (compare Sections 3.3 and 4.1). Steady magnetic fields were known to damp convection and therefore to suppress temperature fluctuations. This is valid for buoyancy as well as for Marangoni convection. The latter plays an important role with respect to doping uniformity of, for example, GaAs crystals which were grown by the floating zone method under microgravity conditions (see Section 3.2). The transition to unsteady flow causes doping striations and occurs if the critical value of the Marangoni number \(M_a\) is exceeded. Roland Rupp calculated in his Ph.D. thesis by numerical simulation the dependence of \(M_a\) from the magnetic field strength applied to vertical GaAs melt zones with free surfaces. Based on these results a permanent magnet device was developed and used during the growth of GaAs crystals onboard the Spacelab mission D2.[\textsuperscript{35,76,77,44}]

Extended systematic studies of a variety of magnetic fields, both steady and time-dependent ones, were carried out within the frame of the Ph.D. theses of Johannes Baumgartl and Bernd Fischer. Baumgartl analyzed the mathematical treatment and solution strategies for the magnetohydrodynamic formalism. He studied the use of various levels of simplifications and approximations and their impact on the results for different crystal growth conditions. The calculated results were verified by model experiments with Ga melts in test cells. A fruitful collaboration was started at that time with the experts of the Research Center Rossendorf, Gunter Gerbeth and Andre Thess. The magnetohydrodynamic models MHD1 and MHD2[\textsuperscript{45,46}] proposed by Baumgartl were later implemented into the software STHAMAS (see Section 4.5) and extensively used by Bernd Fischer during his Ph.D. work. His profound analysis of the time-dependent magnetic fields became the basis of using rotating and traveling magnetic fields in crystal growth configurations. An important progress hereby was the possibility to calculate flows three dimensionally (STHAMAS3D). These studies resulted in quantitative correlations with experimental results of model configurations.[\textsuperscript{47–54}]

The application of steady and time-dependent magnetic fields to real crystal growth systems occurred within the frame of the development of the growth of InP by the LEC and VGF methods. For example, in the LEC growth of InP crystals, which will be presented in Section 4.2 in detail, a steady axial magnetic field was used in order to suppress convection-induced doping striations.[\textsuperscript{35}]

Another big issue during the growth of InP crystals is the formation of the so-called twin defect. It was found that the occurrence of temperature fluctuations in the melt correlates with a prevalent occurrence of these twin defects in agreement with the theory of Don Hurle.[\textsuperscript{56}] Therefore, a concept was developed to use a RMF in the VGF growth of InP to stabilize the melt flow against temperature fluctuations. The simulation results showed clearly that the necessary field strength of the RMF is much smaller than that of a static magnetic field with similar impact on the temperature fluctuations.[\textsuperscript{47,57,58}] However, it turned out, that the experimental realization of a magnet system outside of the high-pressure steel autoclave needed for InP crystal growth was too expensive. This technological problem was later circumvented very elegantly by generating the magnetic field inside of the growth vessel by using the so-called heater magnet module pioneered by Peter Rudolph at Institute for Crystal Growth (IKZ) in Berlin.[\textsuperscript{59}]. More or less in parallel to the developments in Berlin, Erlangen crystal growers transferred the idea to directional solidification of multicrystalline silicon,[\textsuperscript{60–64}] where the usage of time-dependent magnetic fields has meanwhile become state of the art to control the melt flow. More details are presented in Section 5.1.

In industrial growth of large diameter Si crystals by the Czochralski (Cz) method the use of magnetic fields has become standard technology as well. But, in the Cz technology, steady magnetic fields are commonly applied instead of time-dependent configurations. Since around 2000 they are used in production for silicon crystals with 300 mm diameter.[\textsuperscript{66,67}] During the development phase of the 300 mm Cz silicon process CGL studied systematically within the frame of collaborative industrial projects the influence of various types of magnetic fields on the behavior of melt flow, temperature, and oxygen distribution by numerical simulation and measurements in industrial facilities[\textsuperscript{66,68–77}] (see also Section 4.5).

4. Development of Industrial Singe Crystal Melt Growth Technologies

4.1. The Beginnings (1975–1980)

The entry into melt growth of semiconductors in the period of 1975–1980 was strongly supported by crystal growers of the Erlangen Siemens Research Laboratories, namely Dr. Gunter Raab,
Hans-Jochen Wolf, Siegfried Leibenzeder. Growth equipment for horizontal zone melting (ZM) and horizontal Bridgman (HB) was built in the workshops of the university according to the construction plans provided by the Siemens researchers. They also transferred the know-how to grow the first InSb and GaSb single crystals by the ZM and HB technique at CGL. These crystals were used as seed material for the development of a growth technique with a solution zone, called THM which was relatively new at that time. This method had the potential to improve the material quality by reduction of the dislocation density and the concentration of stoichiometric defects (in GaSb) due to the lower growth temperature. The details of the THM growth technique were developed in collaboration with the group of Klaus Benz who was at the University of Stuttgart during that time. Georg Müller presented the results of this work during his first participation in the International Conference on Crystal Growth ICCG-51977 in Boston (USA) and combined it with visits of Gus Witt, Harry Gatos, and Bob Brown at MIT. Also, the first InP crystals were grown at that time at CGL by THM. This opened a new working field, the growth of InP crystals for optoelectronic applications as shown in the following section.

4.2. Bulk Growth of InP Crystals by LEC and VGF (1980–2005)

Around the year 1980, the interest in InP crystals increased strongly in Germany when the concept of optical fiber communication was developed under the slogan “fiber to the home.” At that time, CGL started to explore the growth of InP bulk crystals by the LEC method. Again, the entry into the growth technique was supported by the Erlangen Siemens Research Laboratory by transferring a high-pressure LEC puller (Siemens homemade) including crystal growth know-how to CGL. This enabled a quick successful start into a research and development program on InP LEC growth with important results, which became soon internationally acknowledged. CGL researchers gave regular invited talks at the International Conference on InP and Related Materials. Of special importance for the whole research field including the later development of low-defect GaAs crystals were the systematic studies of the formation of dislocations during the crystal growth process within the Ph.D. work of Johannes Völkl. This study started already during Völkls’ diploma work in which he measured for the first time in situ the temperature distribution within a growing InP crystal by the use of grown-in thermocouples. A second important part of the work was the investigation of the plastic deformation of InP crystals by dynamical compression testing experiments at high temperatures in collaboration with the Erlangen group of Wolfgang Blum.

It could be shown that InP crystals do not behave strictly elastic (Hook’s law) even at low stress conditions, but are plastically deformed in the temperature range of \(0.6 T_m < T < T_m\) (\(T_m\) = melting temperature). The results also clearly showed that the so-called Critical Resolved Shear Stress model is very crude and not adequate for quantitative modeling. Therefore, it was replaced by a much more realistic quantitative model which was based on the material data of these deformation experiments and on the theory of plastic deformation of Alexander and Haasen.

This model describes the movement and multiplication of dislocations in the crystal glide systems and is directly related to the stress field which is caused by the temperature distribution during growth and cooling of the as-grown crystal. Based on this knowledge, the Erlangen InP crystal growers were able to grow the worldwide first InP crystal with 3" diameter by the LEC technique in 1987 (see Figure 7).

However, it was also clear from the results of the stress-dislocation model that InP crystals with low etch pit density (EPD) < 1000 cm\(^{-2}\) cannot be grown by the LEC technique. This conclusion was very important with respect to the further research program of CGL. Based on these results and encouraged by an article in the newsletter of the American Association of Crystal Growth from the year 1983 the opinion at CGL was clearly predominant, that low defect InP crystals and other compound semiconductors like GaAs (see following section 4.3) can only be grown by VGF, but not by LEC-type technology. Crystal growth by VGF means that a traveling temperature field controlled by the heater power should be provided by an arrangement of several separately controlled heaters.
From now on the work at CGL concentrated on the development of the VGF furnace technology, which is able to provide a temperature field with a constant axial temperature gradient. Deviations from this condition, that is, nonlinearity of the axial temperature profile, mathematically expressed the second derivative of the temperature field, had been identified as the source of thermo-elastic stress in the growing and cooling crystal.

However, the procedure of crystal growth itself changed totally its character when changing from Cz pulling to the VGF technique. The seeding process as well as the growth of the crystal itself is no longer visible. The crystal grower needs to anticipate the whole growth process in advance. The VGF process is totally controlled by the variation of the power of the heaters without the possibility to visualize the interface and the growing crystal (except mechanical dipping or sophisticated ultrasound methods which were developed and used by the Erlangen crystal growers\cite{58,88}). Thermal modeling enables the crystal grower now to transfer his ideas of the desired temperature field to the controllers of the heaters. This task fitted well with the modeling capabilities at CGL. Therefore, CGL could achieve very soon a good quantitative correlation between simulated and experimental growth rates.\cite{89}

In the first development phase of furnace concepts it was believed, that a large number of heaters would be necessary.\cite{89,90} Therefore, a furnace with 22 independently controlled heating zones was developed and used. However, it turned out, that a precise design and control of temperature field during VGF processing is also possible by only a few axially separated heating elements. In fact, the first low EPD twin-free InP crystal with 2″ diameter was grown by the VGF technique in a modified LEC facility under highly stabilized thermal boundary conditions and a flat bottom crucible (see Figure 8).\cite{91}

Despite this success the frequent occurrence of twins in the InP crystals remained a challenging research topic during the whole period of VGF InP growth at CGL until 2005. It became clear very soon\cite{92} that the origin of twin formation is related to facet formation at the periphery of the growing crystal as proposed by Don Hurle.\cite{56} But, the attempts to achieve a high yield in twin-free VGF growth of InP by avoiding temperature fluctuations near the growth interface failed. Finally, numerical simulation results suggested the use of time-dependent magnetic fields to improve the thermal boundary conditions near the growth interface.\cite{58} However, the experimental implementation of time-dependent magnetic fields around the VGF steel vessel was not possible. Later on, such fields were put into practice by other groups by using the so-called heater–magnet–module concept (see, e.g., ref. \[59\]).

Another big issue in the application of InP crystals for communication devices was the production of high-quality semi-insulating (s.i.) material (see, e.g., ref. \[94\]). Fe was used from the very beginning as a dopant with a near mid-gap acceptor-like electronic level to compensate the residual donor impurities in InP. However, Fe has several disadvantages, for example, a very low segregation coefficient leading to strong doping nonuniformities in the crystal and a high diffusivity which leads to a contamination of epitaxial layers. Hence, intensive research activities were undertaken to reduce the necessary Fe content or to avoid it at all. The hope existed that an intrinsic point defect could be found which would provide an electrical level similar to the famous EL2 level in GaAs.

In fact, in 1989 semi-insulating InP was achieved after annealing of undoped InP wafers in a phosphorous atmosphere and carefully interpreted as "nominally undoped s.i. InP."\cite{95} This result caused a strong international resonance. An extensive study program followed at CGL to clarify the origin of the electronic properties after wafer annealing of InP\cite{96–117} Even one of Japan’s market leaders in InP production, Nikko Kyodo Co., became interested in the CGL process and presented their results in a common publication.\cite{110} At that time, it was already clear that the relevant defect for the compensation mechanism is Fe which is introduced during annealing either unintentionally (by contamination) or intentionally (by Fe source material). However, the undoped annealed InP became semi-insulating at much lower Fe concentrations as conventionally Fe-doped InP.

However, as the twin problem in InP was not solved also by other research groups worldwide, the yield of twin-free crystals in industrial production remained low. Therefore, the prices for InP wafers were so high in the past, that market size for InP crystals remained small. Now, with the forthcoming introduction of the 5G communication technology there is a rising demand for InP again. Therefore, the research on the growth of InP crystal is revisited also at CGL.

4.3. VGF Growth of GaAs (1993–2003)

In the first working period of CGL, the material GaAs was not considered as a research topic as the LEC technology of GaAs seemed to be developed quite well. Later on, in the middle of the 1980s the company Wacker was interested in a collaborative project on the growth of GaAs. CGL proposed the development of VGF for GaAs according to the results of the profound analysis of the capabilities of LEC and VGF for the growth of InP (see Section
4.2. However, the management in charge of the compound semiconductor business at Wacker at that time insisted on a development of LEC in Erlangen. Consequently, the collaboration failed.

But history took a fortunate turn. In the course of the reunification of Germany, the company Freiberger Electronic Materials (since 1995 Freiberger Compound Materials [FCM]) became a new industrial partner of CGL. A quick start of the collaboration was facilitated because personal relations already existed since the middle of the 1980s by visits of Georg Müller in the former German Democratic Republic with Thilo Flade, the former CEO of FCM. The very fruitful and successful collaboration between Erlangen and Freiberg started with the modeling of the LEC growth process of GaAs. Most of Freiberg’s LEC equipment was transferred from Burghausen after Wacker had shut down its own III–V activities. The development of CGIs own efficient modeling software (later named STHAMAS, see Section 4.5) was strongly supported by FCM. The first common results were published in 1994.[118] In 1997, the first STHAMAS modeling results of the influence of Ar pressure on the heat transfer and thermelastic stress in the growing GaAs crystal (LEC) were released.[119] Also, the collaboration with the Institute for Crystal Growth IKZ in Berlin in the field of vapor controlled Czochralski (VCG) of GaAs was developing during that time. The modeling was already carried out by using the new software CrysVUn (see Section 4.5).[120] All the results about the study of LEC and VCG growth of GaAs confirmed the results obtained for the growth of InP which were presented in Section 4.2, that is, LEC (and VCG) are disadvantageous compared to VGF.

In 1994, CGL started the VGF growth of GaAs. Jürgen Amon and Josef Stenzenberger had the task within their Ph.D. works to develop an industrial VGF growth facility and a growth process for GaAs crystals with 3” and 4” diameter with potential to upscale to even 6” diameter. The development was based on the successful results and experiences of the VGF growth of InP. Starting with an ampoule in a multizone furnace, the hot zone design and concept was finally totally changed to graphite-based heaters and insulation with a reduced number of three independently controlled heaters. The whole set-up was designed on the basis of extended simulations by using the software package CrysVUn which has just been developed at that time. The development of the GaAs VGF furnace and the growth process demonstrated impressively the power of modeling. Further improvements in the VGF process optimization were achieved by the so-called “inverse modeling” strategies. In this approach several conditions and constraints of the temperature field are given, for example, the position and shape of the crystal melt interface and temperature gradients in the melt or crystal. The corresponding set of heating powers is then calculated by CrysVUn as a result of the mathematical “inverse problem.”[121–123]

In the following period high-quality GaAs crystals with 3” and 4” diameter could be reproducibly grown with very low dislocation densities (see Figure 8). This success was also based on a profound study of details of the growth process. For example, the formation of peripheral facets in relation to the cone angle[92] or the behavior of residual dislocations originating from the region of the seed channel[124] were analyzed quantitatively.

The close collaboration between CGL and FCM on the further improvement of VGF growth of GaAs continued until 2003 with a large series of publications.[121,125–134] The transfer of this VGF technology to FCM was very successful. The market share of the VGF GaAs crystals exceeded the one of LEC GaAs crystals by far in a short period of time after the VGF technology had been transferred to FCM, where it was continuously improved in-house and no further support of CGL was needed.

4.4. VGF Growth of CaF₂ (1996–2009)

The successful development of the GaAs VGF process and its transfer to an industrial partner is only one example out of many others of effective industrial collaboration. Such a co-operation with the company Schott in the development of the VGF growth of CaF₂ crystals also worked particularly well. This material was needed for the fabrication of lenses in so-called wafer steppers, which are used for microolithography in the fabrication of microelectronic devices. The progressive decrease of the device structures according to “Moore’s Law” required a reduction of the wavelength of the light which is used in the lithographic projection systems. Around the year 2000, a transition from the wavelength of 248 nm down to 193 nm and even further down to 157 nm was planned according to the technology roadmaps of semiconductor industry. In this range of the UV light, the so far used silica glass lenses were not transparent enough and should be partially replaced by CaF₂. This meant that the glass maker Schott had to become a crystal manufacturer. The requirements with respect to the optical properties, like absorption (purity) and refractive index homogeneity (residual birefringence) were so extremely high that only a very sophisticated gentle growth technology could be used.

In the years 1996–2009, CGL developed in close collaboration with Schott a CaF₂ VGF technology including furnace and growth process and transferred the results to the production site in Jena. Again, numerical modeling by using CrysVUn was used to support the construction of the production furnaces and the development of the production processes. Here a new important aspect was the radiative heat transfer within the (semi-)transparent CaF₂ and the whole facility because of the high temperatures.[135,136] In Erlangen a R&D VGF equipment was built for the VGF growth of CaF₂ crystals with 155 mm diameter within the frame of the Ph.D. work of Alexander Molchanov. Special challenges were the high temperature (up to 1700 °C), the low axial temperature gradient (1–5 K cm⁻¹), flat isotherms (low thermal stress), high purity (possibility for in situ purification), and extremely low growth rate (<1 mm h⁻¹). A distinctive feature of the VGF furnace was the possibility to measure in situ the position of the growth interface with the aid of a mechanical device which is introduced from above through the top of the growth vessel. Because of the aggressive atmosphere the thermocouples for controlling the heaters and for in situ temperature measurements in the melt and growing crystal needed the development of a special protection which was patented.[137,138] Further patented developments were devices to supply gases into the process chamber during growth.[139] This equipment enabled, for example, the controlled addition of oxygen as a dopant for CaF₂. In fact, oxygen contamination reduces the optical transmission of CaF₂ and should therefore be avoided. However, a well-defined doping in the range of 1–200 ppm enabled the quantitative determination of the optical absorption in dependence from the oxygen concentration in CaF₂.[140] This was
necessary to calibrate the optical absorption method for detecting residual oxygen contamination in the crystal production. A typical CaF\(_2\) crystal grown in Erlangen is shown in Figure 8. Due to the successful collaboration between CGL and Schott, already 9 months after the opening of the new CaF\(_2\)-production site in Jena, Schott Lithotec had delivered the 500st CaF\(_2\) single crystal to its customers worldwide. In 2003, the successful collaborative work was awarded by the “Wissenschaftspris des Stifterverbandes für die deutsche Wissenschaft.”

The semiconductor roadmap predicted a further reduction of lithography wavelengths which would have required more and larger lenses CaF\(_2\) lenses in a wafer stepper. However, a few years later semiconductor industry introduced the immersion lithography which allowed getting very small feature sizes without further decreasing the wavelength. This changed the general semiconductor technology roadmap and the need for a further development of the CaF\(_2\) technology. Consequently, the interest in research on the VGF growth of CaF\(_2\) decreased rapidly and the related activities at CGL were stopped.

4.5. Czochralski Growth of Silicon Crystals (1980–Today)

The work on Si crystal growth started at CGL already in the late 1970s with a Czochralski puller (Philips, RF heated) and a floating zone facility, both provided by Siemens. However, at that time it was not possible to start a collaboration with Wacker—the only potential industrial partner on Si crystal growth during that time. Besides several growth experiments, the demarcation of the growth interface by the Peltier effect using electric current pulses was studied for Si within the frame of diploma and study works. Without external support by funded projects these activities remained small. This situation changed around 1990 when CGL became involved in the European research project JESSI (Joint European Submicron Silicon) which had the goal to introduce Si wafers of 300 mm diameter and the corresponding device technology in European industry.

CGL contributed to this project by fundamental studies of convective transport phenomena in the Si Cz process, like study of melt flow structures, temperature, and oxygen distribution in the Si melt. These studies were carried out both by simulation and experimental work. The experimental approach concentrated on the development of sensor technology for temperature measurement and oxygen concentrations in situ during the Cz process.

The scaling of the industrial Cz pullers of Wacker Siltronic, which was the company name at that time, had achieved dimensions which did not allow to install and operate such a puller in Erlangen. Instead, the measurements were carried out at the production site in Burghausen. The large extent of the research program demanded additionally laboratory space and man power in Erlangen, which lead to the foundation of the department of “Crystal Growth” at the Erlangen Fraunhofer IISB (compare Section 1).

4.5.1. Oxygen in Cz Si

The first substantial research topic of CGL in the study of the Cz process of Si was the investigation of oxygen in the Si melt. The important features of oxygen in Si are the inevitable contamination of the melt by the dissolution of the silica crucible and the favorable effect of oxygen related precipitates in the Si wafers. The precipitates are formed during heat treatment in device processing and act as gettering centers for residual impurities. The crystal growers must provide a certain uniform oxygen concentration in the Si crystal in the range of 2–8 × 10\(^{17}\) atoms cm\(^{-3}\) to facilitate the gettering effect. Therefore, it is necessary to control the rate of dissolution of the silica crucible and to study the mechanisms of erosion. Furthermore, the convective transport of oxygen within the Si melt to the growth interface determines the oxygen content in the crystal as the segregation coefficient is close to one.

In the early 1990s, Albrecht Seidl and Georg Müller together with Rainer Marten started to develop an oxygen sensor which could measure in situ during Cz growth the oxygen concentration in the Si melt with a spatial resolution in the millimeter range. The sensor principle is based on the ionic conductivity of solid ionic materials like zirconia.\(^{[141,142]}\) With this sensor it was possible for the first time to show quantitatively the influence of certain Cz parameters, for example, crucible rotation on the oxygen transport within the melt.\(^{[143]}\)

The next important step was the measurement of the oxygen solubility in the Si melt in the relevant temperature range from the melting point up to 1836 K. The results showed clearly that this dependence cannot be neglected.\(^{[144]}\) This was of special importance for the numerical simulation of the oxygen transport.

However, the comparison of the oxygen distributions in the Si melt in a standard Cz process measured by the oxygen sensor and calculated by modelling did not provide a satisfactory correlation at that time (1999) which was attributed to weaknesses of the simulation models.\(^{[145]}\) Problems identified in the modeling concepts were the non-realistic 2D calculations, the non-appropriate k-ε turbulence model and the insufficient resolution of the solute boundary layers. These deficiencies were overcome later.

Another interesting experimental study was carried out in the frame of the Ph.D. work of Andreas Mühe. He developed a setup which could be used to measure in situ the dissolution rate of a silica Cz crucible by optical interferometry. A laser beam was transferred through the walls and the hot zone of a Cz puller and reflected at both surfaces of the silica crucible wall (see Figure 9). The interference pattern of the two beams was used to determine the change of the crucible wall thickness caused by the dissolution of the inner wall.\(^{[146,147]}\) The dissolution rates which were measured by this approach in the temperature range of 1700–1800 K were in the range of 5–20 µm h\(^{-1}\).

4.5.2. In Situ Temperature Measurements in Cz Si

The knowledge of the temperature distribution within the melt, the growing crystal, and the whole hot zone belongs to the most important features of controlling a crystal growth process (compare Section 2). The benefits of in situ temperature measurements during crystal growth have already been shown in the section on LEC growth of InP (see Section 4.2). In Cz growth of Si similar studies were carried out since the middle of the 1990s in industrial Cz pullers at the facilities of Wacker Siltronic in Burghausen by using sensor technology developed at CGL in
high or very low electrical resistivity are needed. For example, newable energies, etc. Therefore, silicon crystals with either very powerful, that is, smaller and smaller transistors in enormous numbers were needed for the personal computer market. In the last decade also the power electronic market has strongly increased due to the increasing importance of converters in power supplies, electrical vehicles, robots, motor controls, renewable energies, etc. Therefore, silicon crystals with either very high or very low electrical resistivity are needed. For example, a very low resistivity minimizes the resistance losses in power devices with vertical current flow. The low resistivity—below 5 mΩ cm—means a very high doping concentration in the crystal (>1 × 10^{19} atoms cm^{-3}) which needs the addition of very high concentrations of dopants to the silicon melt according to the segregation coefficients of B, As, P, etc. The high doping concentrations have several impacts on the Cz process as well as on the properties of the silicon crystals themselves. It results in a higher probability of so-called structure loss, that is, formation of dislocations which finally leads to polycrystalline material, which limits the crystal yield. Also, the formation of intrinsic point defects is influenced by the presence of high doping concentrations in the silicon crystal. CGL has analyzed these special features of growing heavily n-doped silicon in collaboration with Siltronic since 2012. In the frame of his Ph.D. thesis, Ludwig Stockmeier found that the structure loss occurs regularly, although the well-known criteria for constitutional supercooling are not yet fulfilled. By using different analytical methods, like defect selective etching or X-ray topography (XRT) he revealed that the origin of the structure loss can be attributed to instabilities of the growth process which are caused by temperature fluctuations in the melt. These instabilities influence the growth kinetics of the growth ridges at the crystal periphery which consist of the so-called (111) edge facets. As a result of these instabilities dislocations are formed in the vicinity of the border between the non-faceted and faceted solid–liquid interface. Based on these finding the industrial Cz process for growing heavily n-doped silicon can be optimized.

The in-depth analysis of the edge facets and the growth ridges leads to the idea whether the growth ridge geometry can be used to get direct information about the stability of the temperature field during growth. Therefore, a contactless, non-destructive approach to measure the geometrical parameters of the growth ridge, based on surface topography, was developed. Based on the measurement results, Voronkov’s theory of the shape of the growth ridge could be verified. By extending his theory it was possible to calculate the temperature gradient at the growth ridge from its geometrical parameters. Therefore, the measurement of the growth ridge geometry gives an easy, direct experimental access to the thermal conditions, both qualitative and quantitative, at the solid–liquid interface during the Cz but also FZ growth process.

4.5.3. Structure Loss in Heavily Doped Cz Si

The in situ temperature and oxygen measurements were carried out in a time period, where the Cz production was up-scaled from 200 to 300 mm diameter because of the strong growth rates in the “More than Moore” micro-electronic market, where more and more powerful, that is, smaller and smaller transistors in enormous numbers were needed for the personal computer market. In the last decade also the power electronic market has strongly increased due to the increasing importance of converters in power supplies, electrical vehicles, robots, motor controls, renewable energies, etc. Therefore, silicon crystals with either very high or very low electrical resistivity are needed. For example, a very low resistivity minimizes the resistance losses in power devices with vertical current flow. The low resistivity—below 5 mΩ cm—means a very high doping concentration in the crystal (>1 × 10^{19} atoms cm^{-3}) which needs the addition of very high concentrations of dopants to the silicon melt according to the segregation coefficients of B, As, P, etc. The high doping concentrations have several impacts on the Cz process as well as on the properties of the silicon crystals themselves. It results in a higher probability of so-called structure loss, that is, formation of dislocations which finally leads to polycrystalline material, which limits the crystal yield. Also, the formation of intrinsic point defects is influenced by the presence of high doping concentrations in the silicon crystal. CGL has analyzed these special features of growing heavily n-doped silicon in collaboration with Siltronic since 2012. In the frame of his Ph.D. thesis, Ludwig Stockmeier found that the structure loss occurs regularly, although the well-known criteria for constitutional supercooling are not yet fulfilled. By using different analytical methods, like defect selective etching or X-ray topography (XRT) he revealed that the origin of the structure loss can be attributed to instabilities of the growth process which are caused by temperature fluctuations in the melt. These instabilities influence the growth kinetics of the growth ridges at the crystal periphery which consist of the so-called (111) edge facets. As a result of these instabilities dislocations are formed in the vicinity of the border between the non-faceted and faceted solid–liquid interface. Based on these finding the industrial Cz process for growing heavily n-doped silicon can be optimized.

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**4.5.4. Modeling of Cz Growth of Si**

The development of software tools at CGL for modeling of Cz of Si started in 1995 with the Ph.D. work of Jakob Fainberg who collaborated with the post doc Hans-Jörg Leister. Their software, which was later called STHAMAS was based on the use of a structured grid and the finite volume method to solve the corresponding differential equations. These numerical techniques have been proven to be very efficient for the solution of the Navier–Stokes equations which describe convective flow.

However, the structured grid turned out to be also a weakness of STHAMAS, because it limits the consideration of complex geometries—like the hot zone of a Cz set-up. This dilemma
was solved at CGL in 1996 by starting the work on the development of the software package CrysVUn by Mathias Kurz supported by Artur Pusztai.\[161,162\] The “Un” in CrysVUn stands for the unstructured numerical mesh which makes it very appropriate for global simulations under consideration of complex geometric conditions. This includes the simulation of radiative heat transfer in cavities and complex hot zone geometries. CrysVUn offered a further advantage by providing the option of “inverse modeling,” an optimization strategy which had already been applied successfully in the development of VGF growth of GaAs (compare Section 4.3). This approach enables the crystal grower to set certain conditions or constraints of the growth process—and the software then determines corresponding solutions for heater powers, geometry, or even material properties of hot zone parts.\[120\] It was used, for example, to optimize the Cz growth of Si in order to meet the so-called V/G criterion (V pull speed, G axial temperature gradient; for details see, e.g., ref. [163]) and to create certain intrinsic point defect concentrations.\[164\] Another strategy was developed at CGL by Tim Fühner and Thomas Jung.\[165\] They used the concept of genetic algorithms for optimizing the geometry of heat shields in a Cz hot zone. Also, the selection of materials inside the furnace could be optimized by such an approach.

CrysVUn proved to be very powerful for simulations of crystal growth processes which were not influenced too strongly by convective flow like VGF of GaAs or Cz of GaAs in the complex vapor-controlled configuration VCZ (compare the results achieved by ref. [166]). At that time there was a strong scientific competition between different approaches to simulate the Cz configurations for industrial Si growth adequately. The competition took place within CGL between the STHAMAS and CrysVUn groups as well as within the University Erlangen–Nuremberg between CGL and the group of Franz Durst at the chair of fluid dynamics,\[152\] and finally within the international community, for example, François Duprets’ group in Belgium and Koichi Kakimoto’s group in Japan. The results were intensively discussed within the crystal growth conferences, especially during meetings of the “International Workshop on Modelling in Crystal Growth” (IWMCG). In 1997, a comparative test of three different codes with benchmark temperature measurements in Cz facilities of Wacker Siltronic resulted in a “draw,” that is, similar results of all tested software packages.\[149\]

In 1997, also a 3D version of STHAMAS was presented at the PAMIR conference in Paris.\[167\] The further development of STHAMAS3D was strongly supported by the collaboration with Daniel Vizman from the university of Timisoara (Romania).\[168\]

A fully 3D model of a Cz growth process is very challenging—and not yet solved satisfactory until today. However, several attempts were made to combine a local 3D model, which considers convective flows in the melt with a global model of the whole Cz set-up. Firstly, the calculations for the melt and crucible region were carried out separately by using boundary conditions at the crucible wall which were provided by (also separate) calculations with a global model. This approach had two inherent disadvantages. The boundary conditions were 2D and fixed, that is, the impact of the convective heat transport on the temperature distribution of the crucible wall was not considered. Several attempts were made to achieve an exchange of heat transfer data between the melt and the external region, that is, a direct coupling of both models.

The collaborative work with Jeff Derby from the university of Minnesota resulted in several approaches to solve this problem, see, for example, refs. [169,170]. Around 2006, the hybrid code CrysMAS was presented for the first time.\[171\] It was developed to combine the advantages of the unstructured grid of CrysVUn and the structured grid of STHAMAS. The structured grid is used in all parts of the Cz facility where convection occurs (melt and gas). The unstructured mesh is used for all other parts (crystal, crucible, heaters, shields, insulation, etc.). The solid–liquid interface is solved as a free boundary problem between the structured and the unstructured grid. Also, a first attempt of a fully 3D global model was presented in 2006 using a 3D version of CrysVUn.\[172\] However, this development was not continued, instead the 2D–3D coupling of CrysMAS was further improved. In 2013, Jung and co-workers\[175\] presented a work which was a global axisymmetric thermal model of a Cz growth facility which is coupled to an external local 3D time-dependent flow model of the Si melt. The boundary conditions of the 3D model were updated by using the results from the 2D global model. In the 3D model the boundary layers were resolved by aggressive mesh refinement toward the walls. The turbulent melt flow was calculated by the so-called large Eddy simulation approach. The comparison to experimental results gave a fairly good agreement for interface shapes and oxygen concentration in the crystal. This model is currently used to support the development of the next generation Cz Si pullers.

5. Growth of Photovoltaic Materials

5.1. Growth of Silicon for Photovoltaic Applications (2004–Today)

In the year 2000, the “Erneuerbare Energie Gesetz EEG” (renewable energy law) became effective in Germany. As a result, several companies wanted a piece of the pie and started to grow silicon crystals for photovoltaic (PV) applications in Germany. In parallel, a supplier industry began to develop which produced equipment and consumables for the manufacturers of PV silicon crystals. Mainly three crystal growth techniques were pursued and the Erlangen crystal growers at IISB were involved as collaborative partners in these activities:

i. Growth of thin walled multicrystalline silicon by ribbon technologies:

The edge-defined film-fed growth (EFG) was used mainly by RWE Solar in Alzenau (later Wacker Schott Solar after several company take-overs). Hollow silicon tubes were grown with 12 faces of 125 mm width and a length of 7 m. The wall thickness was less than 200 μm. This EFG production was shut down in 2009.

The string ribbon (SR) technology was used by Sovello in Bitterfeld-Wolfen to produce in a quasi-continuous process silicon ribbons with 150 mm width and a thickness of only 120–150 μm. This SR production was shut down in 2012.

ii. Directional solidification of multicrystalline (mc) silicon ingots:

The directional solidification (DS) which was initially also named ingot casting was pioneered by Bayer Solar in Freiberg, Saxony, which became later Solarworld. In Freiberg mc silicon ingots were produced until 2016 when the
production was stopped. A few years earlier in 2012 Schott Solar shut down its silicon ingot production in Jena, which was started only a few years before, when furnace capacities became available due to falling demand for CaF₂ crystals (see Section 4.5; note CaF₂ and silicon have almost the same melting point). In Arnstadt, Thuringia the production of mc silicon by directional solidification started already in 1997 at Ersol company, but later on around 2005 the company switched to the production of single crystalline silicon by the Cz technique.

### iii. Czochralski technique to grow single crystalline silicon crystals:

The Cz activities for PV applications had its root at Siemens Solar in Munich, where they had grown so-called tri-crystalline silicon crystals until the beginning of the 21st century. The growth of Cz solar silicon was continued in Arnstadt by Ersol where a production line was built up and operated by the help of Russian crystal growers and German equipment. The production was in operation until 2012 when Bosch Solar Energy, which took over Ersol in 2008, stopped its solar business. In 2013 the Cz production was ramped up again, when Solarworld acquired the former Bosch site, but closed in 2017 when Solarworld became bankrupt.

Interestingly Siltronic, the leading German silicon crystal growth company, did not enter the solar business, although they carried out the pioneering work for (i) and (ii) in the 1980s when photovoltaics was in its infancy state. Furthermore, the mother company Wacker became one of the leading feedstock supplier for both microelectronic and photovoltaic Si.

The research activities of IISB in the field of silicon crystal growth for photovoltaic application started in the year 2000. First the Cz growth of tri-crystalline silicon was supported by numerical simulations, next a global model of the edge-defined film fed growth of photovoltaic silicon was developed. This work included several detailed considerations, for example, the feeding process, the stability of the melt meniscus, and chemical reactions. Furthermore, the formation of SiC particles in the silicon melt caused by its reaction with the graphite crucible material was studied experimentally. The systematic investigations of the interaction of liquid silicon with different graphite materials and coatings brought the conclusion, that in the EFG Si process the formation of SiC particles cannot really be avoided. Also, the occurrence of the inherent special grain structure of EFG and SR silicon could be explained by a growth model which considers the surface energies of the growing grains and the necessary undercooling in front of the phase boundary. This special grain structure is finally also responsible for the strongly non-uniform distribution of the Fe contamination and the dislocations. Although there are areas on a wafer with low Fe content and low dislocation density, a large portion of the wafer contains a high amount of Fe and a high dislocation density. This problem cannot be avoided because of the special growth process and the resulting grain structure. Therefore, the ribbon silicon turned out to be disadvantageous compared to standard mc silicon where growth conditions had been found to reduce both the dislocation density and the Fe impurity in the whole wafer.

In 2004, IISB began a collaboration with Solarworld on the directional solidification of mc silicon which had also triggered the foundation of the Fraunhofer Technology Center for Semiconductor Materials in Freiberg 1 year later. The first research topic was on the formation and avoidance of C and N related precipitates during the directional solidification process. A fundamental scientific understanding on the formation of C and N related precipitates was achieved by profound experimental analysis and numerical modeling of the convective heat and mass transport processes in the Ph.D. work of Christian Reimann. A series of scientific papers was published on this topic. Based on these findings technical solutions, which were partially patented, how to avoid the harmful crystal defects were proposed. Relatively quickly, it became clear that a thorough mixing of the melt would be beneficial to prevent particle formation. Therefore, research was started on the usage of time-dependent magnetic fields to enforce the melt flow (see Section 3.4). As a result, innovative R&D and production furnaces were built, where the arrangement of the heaters was interconnected in an intelligent way to generate a certain magnetic field. This field was able to control the melt flow in such way that the particle formation could be prevented. This approach has become state of the art today. Although technical solutions were found to avoid the particle formation, the scientific question remained how the particles are incorporated into the crystal. To clarify this open question a series of space experiments were carried out (see Section 3.2). The outcome of these experiments was that the lift force which acts on the particles due to the melt flow could push the particles away from the solid–liquid interface, and thus would result in higher critical growth velocities under convective conditions.

The second big research topic was the systematic study of the principles of the directional solidification process of quasi-mono silicon ingots within the framework of the Ph.D. thesis of Matthias Trempa. The principle of the quasi-mono technology in comparison to other important seeding variants, which are used to control the formation of the grain structure in the DS of Si, is shown in Figure 10. At that time around 2010 the quasi-mono technology was believed to have a great potential for PV industry. However, the yield of single crystalline material was strongly affected by twin formation at the edge of the crystal close to the crucible wall. The experience on the twin formation in compound semiconductors helped IISB to understand quickly the twinning process and to propose methods to overcome the twinning problem. Another focus of the work was on the defect formation which occurs at seed joints and the influence of the grain boundary types on the defect formation. IISB was one of the first who proposed the so-called “grain boundary engineering” for silicon. It could be shown that a proper arrangement of the seeds with different orientations results in a reduction of the dislocation density in the quasi-mono ingots.

Other important problems occurring in the quasi-mono technology were also investigated, for example, the indentation of the seeds by the silicon feedstock or the contamination of the seeds by iron during the crystallization process. These scientific results contributed considerably to the success of the industrial partner to achieve cell efficiencies over 20% by using the quasi-mono technology on G5 (880 × 880 mm²) ingots.

Simultaneously to the development of the quasi-mono technology the technology of solidifying so-called high performance
multicrystalline (HPM) silicon began in 2011. Actually, it was by chance a spin-off of the quasi-mono technology, when a granular feedstock was used in test experiments in order to find the proper conditions for positioning of the solid–liquid interface during the seeding process. The grain structure of the resulting mc ingots differed considerably from that of conventional mc silicon (see Figure 10). The grains were much smaller and consequently more grain boundaries were present on the wafer surface. Surprisingly solar cells processed on such HPM wafers had better efficiencies than those on conventional coarse-grained wafers with a lot of Σ3 grain boundaries [191, 192]. IISB contributed much to unravel the mystery of the HPM material and how to achieve it reproducibly [193]. A characterization tool called “Laue scanner” was developed which was extremely useful to study grain orientations and types of grain boundaries of mc silicon on the full wafer scale [194]. This tool was extensively used within the Ph.D. thesis of Toni Lehmann to study and compare the evolution of the grain structure in the DS of Si: a) classic mc, b) quasi-mono, c) high performance mc, and d) high performance 2.0. (Adapted with permission [195]. Copyright 2018, Springer.)

Figure 10. Schematic representation of the four important seeding variants which are used to control the formation of the grain structure in the DS of Si: a) classic mc, b) quasi-mono, c) high performance mc, and d) high performance 2.0. (Adapted with permission [195]. Copyright 2018, Springer.)

Therefore, in the last few years IISB investigated together with the supplier industry new approaches for the production of HPM silicon, which are named HPM2.0 (see Figure 10). The main aspect was to provide foreign nucleating agents at the crucible bottom and to solidify the silicon melt directly on them to achieve the fine-grained HPM structure. It turned out that the alternative nucleation methods developed at IISB provide good perspectives to replace the classical seeding on a silicon feedstock particle layer in order to reduce the production costs and to increase the yield of high-quality HPM silicon wafers [191, 196–200].

The current research topic at IISB in the field of directional solidification of silicon is to reduce the contamination of the silicon ingots by metallic impurities coming from feedstock, crucible coating, and the crucible itself by utilizing functional diffusion barrier coatings.

5.2. Study of Copper–Indium–Selenide (CIS)-Based Solar Cell Materials (1993–2007)

The so-called CIS material group is very interesting with respect to the applications for solar cells because of its semiconducting properties and bandgaps in the optimum range for solar radiation. The advantage of the direct bandgap (in contrast to silicon) means that light is absorbed to a high percentage within a few micrometers of materials layer. This enables a solar cell concept which is based on a thin film of the semiconductor CIS. The bandgap can be adjusted by the composition of the mixed crystal system Cu(In$_x$Ga$_{1-x}$)(Se$_y$S$_{1-y}$)$_2$. The CIS film is typically deposited on a glass substrate by various techniques like sputtering, evaporation, and chemical vapor deposition of the elements. The formation of the CIS compounds takes place during an annealing step in a temperature range up to about 500 °C.

CGL became involved in the study of the CIS material system around the year 1993 as a collaboration partner of Siemens. The cooperation was organized and supported within the frame of the project FORSOL of the Bavarian Science Foundation. The first studies were concerned with solution growth of bulk crystals and epitaxial layers of CuInS$_2$ and CuInSe$_2$ [201–203]. The idea was to study the electronic material properties with respect to the composition (intrinsic point defects), doping, etc. in order to improve the knowledge of the solar cell properties of the polycrystalline thin films (Ph.D. thesis of Christina Hack). Later on, the work concentrated on the study of the CIS formation mechanisms within the Siemens “stacked elemental layer” process. Dietrich Wolf developed during his Ph.D. work the use of the method of thin film calorimetry to analyze quantitatively the kinetics of the CIS formation. The differential scanning calorimetry could be used for ex situ phase analysis as well as in situ analysis of the sequential formation of the various phases in the Cu-In layers and the subsequent selenization [204–208]. It could be shown for the first time that the formation of CIS is the final step of a complex sequence of coupled reactions with increasing temperature (see Figure 11). A certain positive influence on the kinetics of CIS formation is caused by the admixture of a small amount of Na which came originally by out-diffusion process. Later on, it was a well-defined addition in the process.
In the following period of work until 2007, the analysis of details of the formation kinetics of Cu(In$_{x}$Ga$_{1-x}$)$_{2}$Se$_{2}$ and the impact of Na was continued successfully within the frame of the Ph.D. works of Patrick Berwien and Michael Purwins. After the retirement of Georg Müller, the running CIS project was handed over to Peter Wellmann, who continued these research activities at the Institute of Material Science of the University Erlangen–Nuremberg.

6. Development of Wide Bandgap Semiconductors (SiC, GaN, AlN)—Crystal Growth and Device Studies

6.1. Defect Engineering in SiC Epitaxy (2004–Today)

R&D of power electronic devices and the corresponding material basis has a strong tradition at Siemens in Erlangen as it was already described in the Section 1 of this article (e.g., invention of the silicon “Siemens process”). In the late 1980s the growth of SiC crystals by a sublimation process and its application for power electronic devices were developed. Later the growth technology was transferred to the Institute of Material Science of the University Erlangen–Nuremberg, where it was further improved within the framework of several Ph.D. theses. During this time fundamental new insight was achieved into the relevant growth mechanisms of SiC and the formation of defects, critical for SiC device production. The results were then successfully transferred to the spinoff company SiCrystal in the mid 1990s, which became one of the leading suppliers of SiC wafers. Simultaneously at IISB the basic SiC semiconductor device technology was developed, again in close collaboration with Siemens and later on with the Erlangen SICED company, a joint venture between Siemens and Infineon. Around 2002, IISB installed a SiC epitaxial growth reactor which was the starting point of the research activities of IISB in the field of SiC epitaxy. As history repeats itself, SICED researchers were very gracious and open to transfer their know-how on SiC epitaxy to IISB. Based on this knowledge and on the long-standing experience of CGL in GaN epitaxy, as well as crystal growth and crystal defects in general, IISB took a quick start in developing the SiC epitaxy process. In the beginning the influence of the epi-parameters like growth temperature, C/Si ratio, doping atoms and their concentrations, and the off-cut angle of the substrate on the formation of dislocations in the SiC epilayer and on the surface morphology were the focus of the investigations. The main target was the improvement of the observed degradation of bipolar SiC devices with high blocking voltages due to so-called “drift of the forward voltage” caused by basal plane dislocations (BPDs) in the epilayer. In the frame of her Ph.D. work Birgit Kallinger, who was the last Ph.D. student of Georg Müller, could clarify how the different kinds of dislocations propagate from the substrate into the epilayer. The lattice misfit in the SiC epilayer which is induced by growing a low N or low Al doped layer on a highly N doped substrate was analyzed and the critical epilayer thickness in dependence on the doping concentration was determined. It could be shown that the harmful BPDs can be converted into non-dangerous threading edge dislocations (TED) during the epi-process. The experimental findings could be explained by the so-called Klapper model, showing that the off-cut angle of the vicinal substrates is one of the most important parameters for improving BPD conversion. Based on experimental and theoretical results from different groups worldwide the off-cut angle of the vicinal substrates was changed during that time in the whole SiC industry from 8° to 4° because of the related increase of the conversion probability for BPDs into TEDs during the epitaxial growth. As a result, a much higher yield of bipolar devices on a SiC wafer, which do not show degradation of the forward voltage, was achieved.

At that time, defect selective etching of substrates and epilayers in molten KOH and subsequent analysis of the etched samples by using light microscopy was the method of choice to analyze the dislocation content in substrates and epilayers. To calibrate the etching procedure and to gain further insight into the dislocation propagation, it was necessary to directly measure the different kinds of dislocations by using the synchrotron white beam X-ray topography (SWXRT) and to correlate the SWXRT results to etch pit geometries, which depend on the dislocation type as well as on the doping atoms and their concentration.

Although the etching method is a very robust and reliable method, it is time consuming and destructive. Therefore, IISB has found the ultraviolet-photoluminescence (UVPL) imaging technique to be a well-suited alternative, because it was known from the literature that extended defects in the indirect semiconductor SiC, like BPDs and stacking faults, show radiative recombination at certain wavelengths when excited by UV light. These bright spots can serve as a fingerprint for defects. In collaboration with an Erlangen metrology company a tool for fast structural characterization of SiC wafers based on UVPL imaging was developed.
developed. With this tool SiC wafers up to 200 mm in diameter can be studied in-line, non-destructively and without preparatory effort within minutes.\textsuperscript{226,227} By scanning the substrate directly after the epi-process with the UVPL scanner it was possible to predict which devices would later drift due to bipolar degradation, and which devices would exhibit reliable behavior. This procedure was validated by forward bias electrical stress tests performed on the fabricated 6.5 kV pn diodes.\textsuperscript{228} Nowadays, UVPL augmented by surface imaging has become an indispensable tool for SiC R&D and industrial production.

Although UVPL imaging works very well for epi layer and partially processed substrates, its application to the heavily n-doped SiC substrates is limited because of the very short minority carrier life time. Therefore, IISB is currently utilizing an advanced XRT tool. Like classical synchrotron-based topography measurements the XRT tool enables investigation of crystallographic defects, but in opposite to synchrotron it can be used in the lab with high speed and highest resolution on full wafer scale.

Besides the extended defects also point defects in SiC are especially crucial for bipolar devices. For such devices with high blocking voltages a high minority carrier lifetime is needed. However, the lifetime is mainly limited by the so-called Z1/2 defect which is correlated with carbon vacancies. IISB is conducting lifetime measurements at the electron beam facility BETA at PSI. Therefore, no principal advantage exists compared to the mostly used hydride vapor phase epitaxy (HVPE) technique.

Motivated by promising results from Japanese and Polish researchers on the solution growth of GaN from Ga containing solutions IISB started to explore the LPSG technique. In the LPSG method a seed is placed at the bottom of a crucible, which is filled with a Ga containing solution. In the gas atmosphere NH\textsubscript{3} is used which allows working at ambient pressure. The nitrogen from the growth atmosphere is dissolved at the surface of the solution and transported by convection and diffusion to the seed where crystallization occurs.\textsuperscript{235,236} Although the principle sounds rather easy, several problems had to be overcome. First, proper additives to the solvent had to be identified in order to increase the solubility of nitrogen in the Ga containing solution.\textsuperscript{237} Simultaneously to the gain of the fundamental results on the mechanism of the LPSG technique, the technology was scaled up quickly with respect to the crystal size. Whereas the GaN crystals were only 1 mm in size grown by the LPSG technique in 2001, three GaN crystals with a thickness of a few micrometers were grown simultaneously on sapphire substrates with 3” diameter in 2007.\textsuperscript{241} It was also demonstrated, that the LPSG method allows for the reduction of the dislocation density by at least one order of magnitude in comparison to metal-organic chemical vapour deposition (MOCVD) GaN on sapphire substrates.\textsuperscript{242} Despite all the scientific and technological achievements, which were obtained mainly during the Ph.D. theses of Stephan Hussy and Guoli Sun, the LPSG technique finally turned out to be not well suited for the industrial production of GaN bulk crystals because of the very low growth rate which is only in the order of 1 \(\mu\)m h\(^{-1}\).\textsuperscript{234} Therefore, the activities on the development of the LPSG technique were stopped at IISB in 2007.

Nevertheless, the Erlangen crystal growers continued the GaN material development until today. Three research directions are pursued. i) the development of the HVPE technology for industrial production of GaN bulk crystals, ii) correlation of structural properties of GaN with performance and reliability of GaN opto- and power devices, iii) fundamental study of the ammonothermal synthesis of nitride compound crystals.
i. After it had become clear that there is not really an alternative to the HVPE technology for growing GaN, IISB started to support its strategic partner FCM in developing this technique. For that purpose, IISB developed a new HVPE reactor concept. Among others one special feature of the new reactor was the possibility to measure optically in situ growth rate, layer thickness, and curvature of the GaN crystal\textsuperscript{243} for the first time. Such possibility had turned out to be very important for the development and control of the HVPE process. Today, 2” and 3” GaN crystals are reproducibly grown in a pilot production line in Freiberg. A typical 3” GaN crystal is shown in Figure 12.

ii. Around the year 2010, it became clear that a commercial market with high annual growth rates will develop for GaN high electron mobility transistors (HEMTs) fabricated on silicon substrates to be used in power electronic industry. However, GaN heteroepitaxially grown on silicon has some problems with respect to the performance and reliability of the devices which are caused by different lattice constants and thermal expansion coefficients. In order to extend the application range of the GaN-on-Si HEMTs toward higher blocking voltages it is therefore of utmost importance to understand the role of dislocations and of other defects such as V-pits or point defects on the leakage currents and therefore on the electrical breakdown of these devices. To answer these questions, IISB is using a combination of different analytical techniques\textsuperscript{244–247} namely defect selective etching, cathodoluminescence imaging, SEM, and TEM analysis as well as different variants of the atomic force microscopy (AFM) technique (e.g., C-AFM). It was found that the presence of facets around the tip of dislocations at the growth interface influences the kinetics of the incorporation of impurities and therefore locally the electrical properties\textsuperscript{248,249}. As a consequence, local leakage current paths can occur which are responsible for failures of the HEMT devices. This knowledge is an important feedback for optimizing the epitaxy process and will help to extend the application range of industrially produced GaN HEMTs toward blocking voltages beyond 600 V\textsuperscript{250,251}.

iii. The topics (i) and (ii) are very application oriented and therefore fully in the scope of Fraunhofer, whereas in topic (iii) the fundamental study of the ammonothermal synthesis of nitride crystals is mainly pure basic research, which does not fit the Fraunhofer model of applied research and therefore was carried out at the University Erlangen–Nuremberg. Thanks to the strong engagement of Elke Meissner the research group “Chemistry and Technology of the Ammonothermal Synthesis of Nitrides” (Ammonofor) was founded in 2011. The research group which was funded by the German Research Foundation DFG was focused on the development of a fundamental understanding of the ammonothermal synthesis, which takes place at pressures of up to 300 MPa and temperatures of up to 600 °C. A detailed insight in the mass transport processes was gained both experimentally\textsuperscript{252–254} as well as by modelling\textsuperscript{255}. IISB contributed to the development of novel in situ monitoring technologies, which enabled the determination of thermodynamic and crystallographic data\textsuperscript{256,257}. The investigations allowed the implementation of specific crystal growth processes for novel, high-quality nitrides.

In 2001, the team of Boris Epelbaum and Mathias Bickermann started to pioneer the growth of AlN crystals by the PVT technique at the University Erlangen–Nuremberg. Their scientific results were published in a series of articles (see, e.g., refs. [258–278]). In 2010 the company CrystAl-N was founded out of their research activities. However, CrystAl-N became bankrupt a few years later in 2015, because the AlN wafer demand developed slower than anticipated. Also the research activities on AlN crystal growth at the university ended. Despite this regrettable development, it is believed that AlN crystals are needed for the UVC LED market, which is forecasted to grow strongly over the next decade. Theoretically, AlN would also be an ideal material for power electronic applications. Therefore, IISB continued the research on AlN crystal growth with the know-how of the PVT experts Boris Epelbaum and Stephan Müller, which became IISB co-workers. Figure 12 shows a typical AlN crystal grown at IISB by the PVT technique. However, the approach of IISB to tap the potential of AlN differs from the former strategy of the University Erlangen–Nuremberg and of CrystAl-N, which were focused on crystal growth only. IISB targets the full value chain, that is, from crystal growth over wafering to epitaxy and device demonstrators (see, e.g., ref. [279]), to accelerate the AlN material development and to promote the commercialization of AlN. This approach was possible only by faithful collaboration with strategical partners having complementary know-how along the whole value chain and by the BMBF initiative “Forschungsfabrik Mikroelektronik Deutschland,” which allowed IISB to invest in equipment specifically tailored to the AlN material and device development. Therefore, the prerequisites are excellent for IISB to develop the AlN technology further in such way, that AlN substrates from IISB will become market-ready within the next 5 years.

Figure 12. Gallery of wide bandgap semiconductors (Copyright Fraunhofer IISB): a) SiC epiwafer with 100 mm diameter; b) GaN crystal with 3” diameter grown by HVPE; c) AlN crystal with 1” diameter grown by PVT.
7. Conclusions and Outlook

Research and development of semiconductor materials and their related crystal growth processes started in the Erlangen area already after World War Two. The pioneers during that time developed the research strategy of a systematic correlation of the properties of semiconductor devices with the properties of the crystals and their production conditions. In the 1970s Georg Müller took up this strategy when he founded the Crystal Growth Laboratory at the University Erlangen–Nuremberg and made it to his working principle. In the 21st century, Jochen Friedrich continued the approach until today to do successful applied research at Fraunhofer IISB.

The main achievements of the crystal growers from CGL and later on from IISB are

- the scientific investigations of the influence of melt flow on the heat and species transport in various crystal growth configurations
- the effects of steady and time-dependent magnetic fields on the heat and species transport in various melt growth configurations
- the pioneering work in the field of the development of user-friendly simulation programs for usage in the area of crystal growth
- the development of technological fundamentals of the VGF technology, especially for GaAs, InP, CaF$_2$, and Si, of the vapor phase crystal growth methods for GaN and AlN, and of the epitaxial growth of SiC
- the correlation of the performance and reliability of power electronic devices with extended and point defects in wide bandgap semiconductor crystals and epilayers

It is believed, that this strategy will be successful in the future to promote and accelerate the commercialization of the wide bandgap semiconductor materials. They also pose a great potential in the emerging field of quantum technology. Therefore, the development of semiconductor materials by tailored crystal growth and epitaxy processes will remain a big issue in the future. These future prospects let us conclude that research and development in the field of crystal growth and epitaxy will be carried out successful in Erlangen also over the next decade.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

convection, epitaxy, melt growth, semiconductors, vapor growth

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[1] See especially F. Bischoff, Patents 1102117, 1140549, 1954 and H. Schweikert, K. Reuschel, H. Gutsche, Patent 1061593, 1954.
[2] R. Emeis, Z. Naturforsch., A 1954, 9, 67.
[3] H. Welker, Z. Naturforsch., A 1952, 7, 744.
[4] H. Welker, Z. Naturforsch., A 1953, 8, 245.
[5] R. Gremmelmaier, Z. Naturforsch., A 1956, 11, 511.
[6] B. Mullin, W. B. Straughan, W. S. Brickell, J. Phys. Chem. Solids 1965, 26, 782.
[7] G. Müller, R. Helbig, J. Phys. Chem. Solids 1971, 32, 1971.
[8] G. Müller, J. Cryst. Growth 1993, 128, 26.
[9] J. Czychalski, J. Phys. Chem. 1918, 81, 219.
[10] G. Müller, Adv. Space Res. 1983, 3, 51.
[11] G. Müller, in Crystal Growth from the Melt. Crystals (Growth, Properties, and Applications), Vol. 12, Springer, Berlin 1988, p. 1.
[12] G. Müller, G. Neumann, W. Weber, J. Cryst. Growth 1984, 70, 78.
[13] G. Müller, G. Neumann, H. Matz, J. Cryst. Growth 1987, 84, 36.
[14] G. Neumann, J. Fluid Mech. 1990, 214, 559.
[15] J. Friedrich, H.-J. Leister, A. Seidl, G. Müller, Overview of Research Projects on the Cray Y-MP at the Leibniz-Rechenzentrum München, München 1996, p. 125.
[16] J. Derby, P. Daoutidis, Y. Kwon, A. Pandy, P. Sonda, B. Vartak, A. Yeckel, M. Hainke, G. Müller, in High Performance Scientific and Engineering Computing (Eds: M. Breuer, F. Durst, C. Zenger), Springer, Heidelberg 2002, p. 185.
[17] J. Baumgartl, W. Budweiser, G. Müller, G. Neumann, J. Cryst. Growth 1989, 97, 9.
[18] J. A. Burton, R. C. Prim, W. P. Slichter, J. Chem. Phys. 1953, 21, 1987.
[19] F. Rosenberger, G. Müller, J. Cryst. Growth 1983, 65, 91.
[20] A. G. Ostrogorsky, G. Müller, J. Cryst. Growth 1992, 121, 587.
[21] W. A. Gault, E. M. Monberg, J. E. Clemans, J. Cryst. Growth 1986, 74, 491.
[22] K. A Jackson, J. D Hunt, Trans. Metall. Soc. ASME 1966, 236, 1129.
[23] R. Rupp, G. Müller, J. Cryst. Growth 1991, 113, 131.
[24] G. Müller, R. Rupp, Cryst. Prop. Prep. 1991, 35, 138.
[25] F. M. Herrmann, J. Baumgartl, T. Feulner, G. Müller, in Proc. VIIth European Symp. Materials and Fluid Science in Microgravity, ESA SP-333, Brussels 1992, p.57.
[26] F. Herrmann, G. Müller, J. Cryst. Growth 1995, 156, 350.
[27] L. Ratke, S. Steinbach, G. Müller, M. Hainke, J. Friedrich, A. Roosz, Y. Fautrelle, M. Dupuyot, G. Zimmermann, A. Weiss, J. Lacaze, R. Valdes, G. Grün, H. Nicolai, H. Gerke-Cantow, Microgravity Sci. Technol. 2005, 16, 99.
[28] L. Ratke, S. Steinbach, G. Müller, M. Hainke, A. Roosz, Y. Fautrelle, M. D. Dupuyot, G. Zimmermann, A. Weiβ, H. Diepers, J. Lacaze, R. Valdes, G. U. Grün, H.-P. Nicolai, H. Gerke-Cantow, Mater. Sci. Forum 2006, 508, 131.
[29] M. Hainke, J. Friedrich, G. Müller, J. Mater. Sci. 2004, 39, 111.
[30] M. Hainke, J. Dagner, J. Friedrich, G. Müller, Microgravity Sci. Technol. 2005, 16, 59.
