Crystal Growth

A.L. Gentile and D.F. Elwell, Guest Editors

Crystal growth is a vital and fundamental part of materials science and engineering, since crystals of suitable size and perfection are required for fundamental data acquisition and for practical devices such as integrated circuits.

The word "crystal" comes from the Greek and means "congealed by cold." The term was originally applied to ice crystals and to crystalline quartz found in such locations as the Alps and thought, at one time, to be some permanently frozen form of water. As a crystallization process, crystal growth extends throughout recorded history—the crystallization of salt from sea water may have preceded pottery-making as the oldest method for controlled phase transformation of materials.

The crystallization of salts such as sodium carbonate was known to the Egyptians, and the process of purification by recrystallization from aqueous solution was described by Greek and Roman writers before the Christian era. The "father" of crystal growth, however, is generally considered to be the English scientist, Robert Boyle, who in 1672 described experiments relating the form of crystals to impurities and changes in growth rate. Boyle believed in the healing powers of crystals, part of the mysticism which has had a strong revival in recent times. We understand that crystals are particularly valued for enhancing the ability of the third eye to perceive auras, but are unable to offer warranted samples.

Recognition as the "founder" of the theory of crystal growth is normally given to the American J. Willard Gibbs, who in 1878 published a seminal work on phase transformations, including a theory of the energy to generate a crystal nucleus and of the equilibrium form of crystals.

In more recent times, crystal growth became organized as an independent discipline. In 1966, the first in a series of

(now) triennial international conferences was held in Boston, Massachusetts. This initiated the beginning of the International Organization for Crystal Growth (IOCG), soon followed by numerous national organizations including the American Association for Crystal Growth (AACG). The first international specialist journal in this field, the *Journal of Crystal Growth*, originated in the same year, and there is now an extensive body of literature in books and journal articles, including a wide variety of technical publications.

This special issue of the MRS BULLETIN presents recent developments that illustrate something of the breadth of crystal growth. The first article, by Jeff Derby, describes the present state of computer modeling of crystal growth in the most popular configuration, the technique of pulling a crystal from the melt (named after J. Czochralski). The practice of crystal growth is still in transition from an art to a science, but both analytical and numerical modeling of this complex process have made major advances in recent years. Numerical modeling has particularly benefited

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from the techniques developed by Prof. Robert Brown and his group at the Massachusetts Institute of Technology, where Dr. Derby pursued his graduate studies. The crystal growth endeavor at Lawrence Livermore National Laboratory has been one of the relatively few in which a team of theoreticians and experimentalists have combined their

talents to improve the state of the art.

Since it is used for the growth of semiconductor crystals, including silicon and gallium arsenide, the technique of pulling from the melt is a core technology in the electronics industry. Today, the latest silicon pullers draw from melts weighing up to approximately 150 kg. Gallium arsenide presents more difficult challenges, which are addressed in the second article by Andrew Jordan of AT&T Bell Laboratories.

The growth of crystals of controlled shape, directly usable without (or with minimal) cutting and polishing, is a relatively recent development which could have great technological importance as applications develop. Sapphire sheet can now be seen replacing glass at the checkout counters of supermarkets, and great ingenuity has been used to develop inexpensive techniques for growing silicon sheets for solar cells. In his article, Robert Feigelson describes the growth of single crystals in the form of fibers, an area of particular interest at Stanford University. Optical materials including nonlinear and solid-state laser crystals as well as some of the new superconducting oxides have been grown in fiber form by techniques that include a "fiber" adaptation of the float-zone process.

In the final article, Hans Scheel of the University of Geneva, Switzerland, and Francesca Licci of MASPEC CNR, Parma, Italy, discuss the growth of single crystals of the new high temperature superconductors. These materials are an example of materials which cannot be grown directly from a melt of the same composition, so crystal-pulling techniques cannot be used. Such materials constitute the majority, and many materials with outstanding properties never become commercially developed because of the cost and complexity of the techniques necessary for crystal growth. The yttrium barium copper oxide described in this article has its unique set of challenges, which may have analogs in other areas of materials research. Scheel and Licci have been among the leaders in meeting these challenges and their crystals drew particular attention at the 1987 MRS Fall Meeting in Boston. □

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Anthony L. Gentile, Guest Editor for this issue of the MRS BULLETIN, is a senior scientist and program manager of the Photonic Materials Program, Chemical Physics Department, Hughes Research Laboratories, Malibu, California. He received his PhD from Ohio State University in thermochemical mineralogy, specializing in high temperature phase equilibrium studies. Gentile has worked in several materials science fields, with emphasis on crystal growth, including solid-state laser crystals, IR-transmitting electro-optic and nonlinear crystals involving II-VI compounds, and also ternary chalcogenide and pnictide compounds based on the diamondlike semiconductor structure, IR fibers, and IR detector materials. He is currently involved in crystal growth of

TiC by a high-pressure float zone technique and deposition of ferroelectric thin films by atomic layer epitaxy. Gentile is a member of the Materials Research Society, Sigma Xi (RESA), and the American Association for Crystal Growth (AACG). He served AACG as treasurer (1981-1984) and as president (1984-1987).

Dennis F. Elwell, Guest Editor for this issue of the MRS BULLETIN, is manager of technology, Microelectronics Circuits Division, Hughes Aircraft Company, Newport Beach, California. He received his BS from Imperial College, London and his PhD from Sheffield University, England, both in physics. Prior to joining Hughes, he was a faculty member at Portsmouth Polytechnic and held positions at the Stanford Univer-

sity Center for Materials Research and at J.C. Schumacher Company. He has a wide background in preparative materials research, especially crystal growth, and is currently working on high temperature superconductors. Elwell is a member of the Materials Research Society.

Jeffrey J. Derby, a new faculty member at the University of Minnesota, was formerly in the Physics Department at Lawrence Livermore National Laboratories, Livermore, California. There he worked on crystal growth modeling for the Laser Division. With a BS degree from the California Institute of Technology, and MSCEP and PhD degrees from the Massachusetts Institute of Technology (all in chemical engineering), Derby is now in the Depart-

ment of Chemical Engineering and Materials Science at Minnesota. His research has involved the development and application of numerical methods to describe crystal growth from the melt, computational fluid dynamics, transport phenomena, and large-scale simulation of materials processing operations.

Robert Feigelson, director of the Crystal Science Department in the Center for Materials Research at Stanford University, has specialized in the field of crystal growth for over 25 years. He received a BS in ceramic engineering from the Georgia Institute of Technology, an MS in ceramics from the Massachusetts Institute of Technology, and a PhD in materials science from Stanford University. He has worked with General Dynamics Corpora-

tion, Watertown Arsenal, Sperry Rand Research Center, and as a consultant. Founder of the Northern California Crystal Growth and Western Sections of the American Association for Crystal Growth (AACG), Feigelson is past president of that association, and editor of its newsletter. He is also a member of the Materials Research Society.

Andrew S. Jordan is supervisor of the Heterostructure Materials Group at AT&T Bell Laboratories, Murray Hill, New Jersey. He holds a BS degree in metallurgy from Pennsylvania State University and received his PhD in metallurgy from the University of Pennsylvania in 1965. He then joined Bell Laboratories and has been involved in the research and development of compound semiconductor materials. He has made significant contributions to the modeling of III-V and II-VI phase diagrams, and has studied defect chemistry and diffusion in some of these compounds. His work on the

thermal stress analysis of crystal growth has led to the preparation of GaAs and InP bulk materials with improved quality. His current interests include heteroepitaxial growth and characterization of III-V compounds. Jordan is the recipient of the Bell Laboratories Distinguished Member of Technical Staff Award. He is a member of the American Association for Crystal Growth and the Electrochemical Society, and is the author of over 60 papers.

Francesca Licci is with the Italian Research Council (CNR) at the MASPEC Institute, one of the main centers in Italy dealing with materials research in electronic and magnetic applications. In her own research as head of the Department of High Temperature Superconductor Materials, Licci focuses primarily on ceramic oxide, single-crystal preparation, and structural and magnetic characterization. She developed an original technique for producing submicron, high-perfection particles of

oxides for application in magnetism (recording/permanent magnets) and superconductivity (ceramics/thin layers). A member of the Materials Research Society and the Italian Association of Crystal Growth, she is author of approximately 60 scientific publications and international patents.

John M. Parsey, Jr. has a BS degree in electrical engineering from Michigan State University, and SM and PhD degrees in materials science from the Massachusetts Institute of Technology. Over the past nine years, while working at MIT and AT&T Bell Laboratories, Murray Hill, New Jersey, Parsey has been involved in the growth and characterization of bulk crystals of compound semiconductors. Currently he is supervisor of the GaAs IC Materials Development Group, AT&T Bell Laboratories, Reading, Pennsylvania, where he is responsible for MBE growth, ion implantation, and materials characterization. Parsey is a member of the Materials

Research Society.

Hans J. Scheel, Physics Department, University of Geneva, Switzerland, developed the accelerated crucible rotation technique (ACRT) and the slider-free multilayer LPE process. These contributions led to growth of larger, improved crystals and improved semiconductor crystal surfaces and flat p-n junctions. The recipient of numerous awards, Scheel has worked with IBM Zurich Research Laboratory, the Brazilian Institute for Space Research INPE, Sao Paulo University, and as a consultant on crystal growth and machining technologies of GaAs, Si, and oxide superconductors. A member of the American Association for Crystal Growth, the European Crystal Growth Association, and both the European and U.S. Materials Research Societies, Scheel has published a book and more than 50 technical papers.

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