

A New Initiative for Crystal Growth

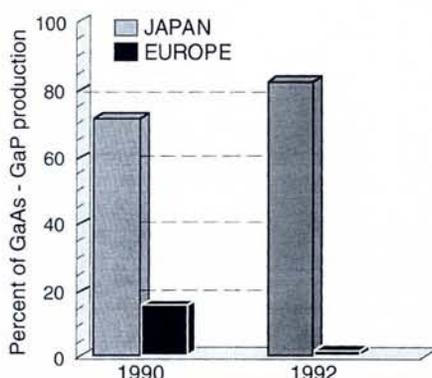
Emanuel Kaldis of the Laboratory for Solid-State Physics, ETH Zurich, describes a proposed multidisciplinary centre of excellence for technologically relevant crystal growth that aims to transform sophisticated empiricism into a science.

The importance of materials research has been generally recognized in the last 20 years. An major catalyst has been the success of electronic materials and the creation of the multibillion dollar electronics industry. The recent statement "who controls materials controls the technology" by the President of SONY Corp. testifies to this characteristic of the second half of the 20th century. The single crystal and its properties, especially its perfection, are of paramount importance, notably for electronic materials. This applies to the solid-state physics as well as to device technology: there is an ever increasing demand for improved crystal perfection, where progress controls advances in electronics to a great extent, and in solid-state physics to a certain extent.

Increasing crystal perfection has in most cases up to now been by trial and error. This can be understood to a certain degree from the complexity and the interdisciplinarity of the problem. However, the progress in, for example, molecular-beam epitaxy achieved in the last decades has shown clearly that a scientific approach towards growing single-crystal thin films can bring about major improvements in crystal perfection and technology, and also enable the realization of innovative ideas (e.g., quantum wells).

There is a widespread impression that the problems of crystal perfection are either solved, or can in principle be solved, with one of the advanced, thin-film deposition methods (MBE, MOCVD, atomic layer epitaxy, etc.). This goes somewhat against the principle of epitaxy because many experiments show that the crystalline perfection of a thin film cannot be appreciably better than that of its substrate despite using buffer layers and the like. An elemental semiconductor such as silicon can be grown from the melt with a high degree of perfection. So wafers cut from silicon single crystals for use as substrates for epitaxy have a perfection comparable to that of an epitaxial thin film. However, even for III-V compounds (the most investigated compounds after silicon), the situation becomes much more difficult owing to defect formation due to, for instance, nonstoichiometry. The types and properties of these defects depend strongly on the thermodynamic and kinetic conditions of bulk crystal growth. In most cases, these conditions have not been determined

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Comparison of GaAs-GaP production in Europe and Japan.

adequately up to now in spite of the fact that device performance depends strongly on eliminating defects.

Interdisciplinary Effort Needed

It seems imperative to make a concerted interdisciplinary effort to study the relationship between device performance, defect formation and crystal growth conditions. A real breakthrough in high-technology devices will be achieved if the growth conditions can be adjusted to avoid the formation of certain defects. For some materials (e.g., II-VI compounds, certain halides and organic materials), this may lead to abandoning an established growth method (e.g., melt growth) and trying to substitute it with a much less well-established method (e.g., vapour growth).

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EURO-CRYST

The EURO-CRYST initiative envisages an approximately 200-250 MECU multinational European centre of excellence for long-range studies of crystal growth. With an annual operating budget of 35-40 MECU, a staff of some 200 (25-30% permanent) and hosting 50 visiting scientists and engineers, it would carry out multidisciplinary projects decided on in collaboration with industry. Eventually occupying about 20000 m², the facility would take roughly four years to start up.

given compound in order to adjust the formation of defects. But while the mathematics of the general theory of crystal growth has been increasingly elaborated in the last decade, the gap between theory and experiment is still large. Numerical simulations are very useful, provided they are based on the precisely measured boundary conditions of the experimental set-up. Measuring the experimental parameters entering theory and simulation remains difficult so there is a general lack of sophisticated experimental crystal growth studies which permit large single crystals (>1.0 kg) with reduced defect concentrations to be grown. Some good work has been done for small crystals (weighing a few grams) under relatively easy experimental conditions (e.g., transparent aqueous solutions). However, the growth in the vapour phase of HgI₂ (see insert) has shown, for instance, that the scaling up of the size of a crystal is non-linear and can induce a different growth mechanism. The obvious conclusion is that scientific studies must be performed for large crystals, possibly for the growth conditions used in production. The first goal would be to monitor the main growth parameters with as high as possible sensitivity and accuracy (see insert).

Using existing experimental techniques, such measurements are very difficult for some materials so new experimental methods are required. The underlying physical principles and the corresponding measurement techniques clearly have to be developed by an interdisciplinary team of physicists, crystal growers, applied mathematicians (fluid dynamics; numerical simulations), and electrical and mechanical engineers. The scientific and technological de-

Scaling up α -HgI₂ Crystals

Crystals of mercury iodide (α -HgI₂; see cover illustration) seem promising for use as room-temperature semiconducting x- and γ -ray detectors of spectrometric quality and with a high radiation hardness (they can tolerate up to 10⁸ protons with an energy of 1 GeV). At the same time, α -HgI₂ is a model substance for studying mechanisms arising in the vapour-phase growth of large single crystals. These can be grown, at present, by physical vapour transport with weights of up to 1000 g. However, they contain traps for positively charged carriers. Striations shown on the cover are generated during the early stages of growth by a mechanism that is different to one found in crystals grown from the melt. They comprise narrow zones of high dislocation density resulting from the interference of curved growth macrosteps propagating along the growing {110} facets of the crystal [M. Piechotka, *J. Crystal Growth* **146** (1995) 1]. The steps are probably triggered by minute convective currents that arise under the conditions of normal gravity. Scattering of a laser beam allows the *in situ* study of the formation of striations, and the motion of the macrosteps can be monitored *in situ* using reflected light. Fundamental studies of these phenomena should lead to crystals with higher perfection, allowing more sophisticated applications.

velopments of the last decade have made such an approach possible.

Such interdisciplinary working groups have never been active in the field of crystal growth, and small university laboratories cannot afford them. Indeed, the lack of academic interest has led industry to adopt empiricism to find practical solutions for the most urgent problems. Of course, neither an understanding nor final solutions could be obtained, so problems were perpetuated albeit in slightly different forms. In the best cases, the problems were circumvented instead of solved.

It is ironic that this policy aimed to keep the low running costs of certain device fabrication processes, by avoiding investment in the fundamental research that was needed to be in a position to control the growth process. It has resulted instead in exorbitant prices for some electronic materials, and their exclusion from civil applications. Thus, according to A. Witt of MIT [1] the price breakdown of the most important infrared detector material CdHgTe (\approx \$US 2000.- per gram of single crystalline wafer) is \approx \$US 100.- for materials, services and energy, and \approx \$US 1900.- for the empiricism. This situation naturally leads to production by trial-and-error with low yield.

A New Initiative

EURO-CRYST, a new initiative for a centre for crystal growth and technology, is based on the premise that only an institute of above-critical size will have the potential to form the interdisciplinary groups needed to study systematically the crystal growth of important materials. Some academic readers may consider such an initiative as involving routine, applied research which can be performed by the electronics industry. However, the tasks at hand are very complex and difficult and the electronics industry cannot tackle them by itself. To expect it to do so is similar to asking it 30 years ago to develop by itself the solid-state physics which is the foundation of modern electronics. Clearly, industrial laboratories played a very important role by discovering the transistor, but the largest part of the scientific foundation came from universities.

Unfortunately, universities do not provide an alternative to industry for achieving breakthroughs in understanding and controlling crystal perfection. Crystal growth groups of subcritical size do exist in a few European universities, but they are clearly too small and they do not have the necessary interdisciplinarity.

CRYSTAL GROWING IN EUROPE

The EURO-CRYST International Advisory Board estimates that at least about 115 small university groups and research institutes together with 35 companies investigate single crystals in Europe.

Accurate Crystal Growth Measurements

The problem of measuring accurate experimental parameters during crystal growth is illustrated by considering two parameters whose measurement is often considered trivial, namely temperature and growth rate.

Growth temperature (*i.e.*, the actual temperature of the growing interface) has rarely, if ever, been measured during crystal growth. The temperature is measured in the best cases inside the growth chamber, and in most cases outside the chamber. An accurate measurement is obviously very important in order to adjust the supersaturation — the driving force for crystal growth — where an accuracy of the order of 1-10 mK may be necessary. Monitoring routinely a crystal growth process without interfering with the crystal at a sensitivity (let alone an accuracy) of this order is a formidable experimental task which only a few crystal growth laboratories can hope to master.

Growth rate in the various lattice orientations. This is the most sensitive crystal growth parameter as it registers all the perturbations of the crystal growth process. Once again, it is difficult to measure without interfering with the growth process. Moreover, it is not very specific so interpreting data needs a strong theoretical understanding as well as additional measurements made as close as possible to the microscopic scale (*i.e.*, one needs to measure the velocity of a growth step). Access problems are formidable. For instance, a direct optical link (transparent media; optical windows) and a minimum working distance are necessary for many high-resolution optical methods so it is difficult to avoid perturbing the temperature fields around the crystal.

EURO-CRYST aims to close this gap: it would be a European institute interacting with industry and academia, dedicated to the fundamental and applied research in crystal growth which is the basis for developing high-technology devices.

The strong interdisciplinarity envisioned for EURO-CRYST means that most of the academic personnel of this institute will be drawn from physics. So the support of the physics community is important, beside that of larger European companies.

Europe, although heavily involved in the discovery of important electronic materials such as the III-V compounds, has not transferred effectively academic knowledge to technology. Thus the technological revolution of the last two decades took place mainly in USA and then in Japan. This is an important problem for Europe since losing the battle for high technology would make Europe technologically dependent. A comparison of the number of patent applications in microelectronics in Europe and USA (according to the Deutsches Patentamt) shows that a dramatic change has taken place between 1987 and 1993: the US number has doubled (to about 1800 in 1993) while the number for Europe has almost been halved (to about 350). Strategies that help to close the gap are very important, and EURO-CRYST could represent an essential component.

Consider, as an example, silicon carbide which is at present one of the most interesting materials for the semiconductor industry, being not only suitable for power electronics but also for high-frequency applications up to 5 GHz. It could be the breakthrough material for the semiconductor industry in the next century provided that crystal perfection in the bulk can be drastically increased. But introducing SiC will not be easy in view of the extreme growth conditions (2500°C) and a number of difficult problems

(micro- and macro-pipe defects, unknown growth mechanism, increase of graphite purity, *etc.*). Following the approach of the Japan's Ministry for Industrial Technology and Innovation, a European research consortium should attack the crystal growth of SiC to allow Europe to take the lead. EURO-CRYST would be the ideal institution to carry out this project.

Attention so far has been given mainly to the electronics industry. However, progress in several other fields can be achieved by studying and understanding crystal growth. Interesting examples include single-crystal turbine blades which function at higher temperatures than polycrystalline types, and therefore at a higher level of efficiency, owing to a superior corrosion resistance.

A Phase II feasibility study of the EURO-CRYST Project carried out on behalf of the Austrian Government ended recently. The reports of both Phases I [1] and II of the feasibility study will be submitted to the Austrian Government in April 1995. If the electronics industry agrees to participate, it is expected that the Austrian government will propose EURO-CRYST for support by the European Union.

INFORMATION

[1] *EURO-CRYST Feasibility Study: Phase I* (March, 1994).

[2] For further information about EURO-CRYST and its scientific programme (bulk growth, thin films and characterisation), contact the Project Director (Prof. A. Preisinger, TU Vienna), the Chairman of the Feasibility Board (Prof. A. Witt, MIT, Cambridge, USA) or one of its members (Dr. P. Glasow, Siemens AG; Prof. R. Kern, University of Marseilles/CNRS; Prof. B. Mullin, Malvern; Prof. E. Kaldis, ETH, Zurich) preferably via the EURO-CRYST Secretariat, Vienna (Fax : +43-1-586 81 36).