the advisability of working with bulk single crystals of gallium arsenide, which are known to be inhomogeneous, but Shaw, Solomon and Grubin maintained they had seen similar results on epitaxial gallium arsenide.

A number of papers had been presented on microwave generation in indium antimonide, and the conference closed with a panel session on this topic. The panel consisted of Betsy Ancker-Johnson, Bartelink, George Bekefi, David K. Ferry, Kino and Charles W. Turner, with Maurice Glicksman as chairman. A variety of views were given on the probable mechanism of the observed effects, ranging from acoustic-wave amplification to two-stream instabilities, and from surface effects to plasma breakdown. There certainly are many ways in which semiconductors can become unstable, and indium antimonide is one material in which most of these effects can occur and probably do. And on this note the conference ended.

* * *

The conference was jointly sponsored by the American Physical Society and IBM.

CYRIL HILSUM
Royal Radar Establishment
Malvern, England

Progress in Thin-Film Studies Discussed in Boston

The theme of the recent conference on thin films, and the main emphasis of the invited and contributed papers, was the structure-sensitive properties of films. More than one third of the 110 papers were addressed to problems of film formation or to structural and metallurgical properties.

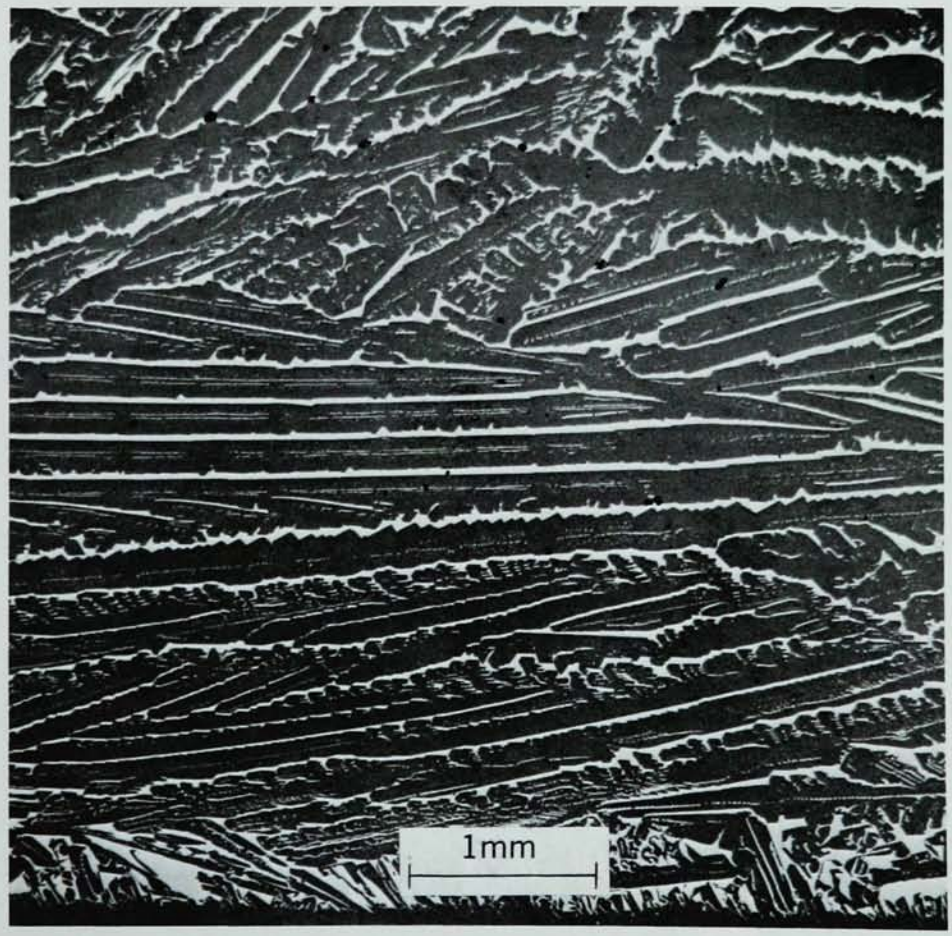
This International Conference on Thin Films, convened in Boston on 28 April, was the fourth of a series that started in 1959 with the meeting called by C. A. Neugebauer (General Electric Research Center) at Bolton's Landing, N.Y. Subsequent conferences held in 1961 at Louvain, Belgium and in 1965 at Clausthal-Göttingen continued to provide an international forum and an opportunity for reassessment of progress made towards a better understanding of the scientific and technological aspects of thin films.

The high level of interest in mechanisms of nucleation and epitaxial growth that characterized the earlier conferences was still in evidence at this one. However, the usual skirmishes between the proponents of atomistic and capillarity models did not develop. Speakers tended, instead, to concentrate on critical tests of key relationships in the atomistic theory.

Atomistic Model. David Campbell (Plessey) reviewed some of the more controversial aspects of the atomistic model. Examples of these aspects are the relationships between saturation density of nuclei N_s and substrate temperature T for initially complete and incomplete condensation, the derivation of values for adsorption energy E_a and diffusion energy E_d and the roles of nucleation and island coalescence in epitaxial growth.

Hans Bethge (Deutsche Akademie der Wissenschaften) discussed the nature of nucleation sites in some detail and considered some of the factors influencing epitaxial growth. He demonstrated that the density of intrinsic defect sites on the face of an alkali-halide crystal is sufficiently high for one to assume always that it is higher than the number of nuclei formed. The number of such sites activated, and the size of the clusters formed, depends upon the deposition rates. He also speculated on the role of electrical double layers in enhancing epitaxial orientation and pointed out that an array of charged surface vacancies, generated for example by electron bombardment, could give rise to surface fields at the substrate with magnitudes of the order of 10^3–10^4 volts/cm.

The nature of such electron-induced surface vacancies, for example in potassium chloride substrates, was considered by Thor Rhodin (Cornell University) who showed that they were chlorine vacancies that could be stabilized by nucleating thin layers of silver or gold. Subsequent deposition of the same metal on such nucleated surfaces at higher temperatures promoted epitaxial growth, whereas deposition on untreated surfaces did not. Rhodin argued that bombardment-induced



TWO-PHASE FILM grown by electron-beam microzone crystallization on a glass substrate. Photograph shows an ordered array of indium filaments contained in a matrix of single-crystal indium antimonide dendrites; thickness of the film is about 5 microns. (From N. Davis and A. Clawson, to be published in J. Vac. Sci. Technol.)

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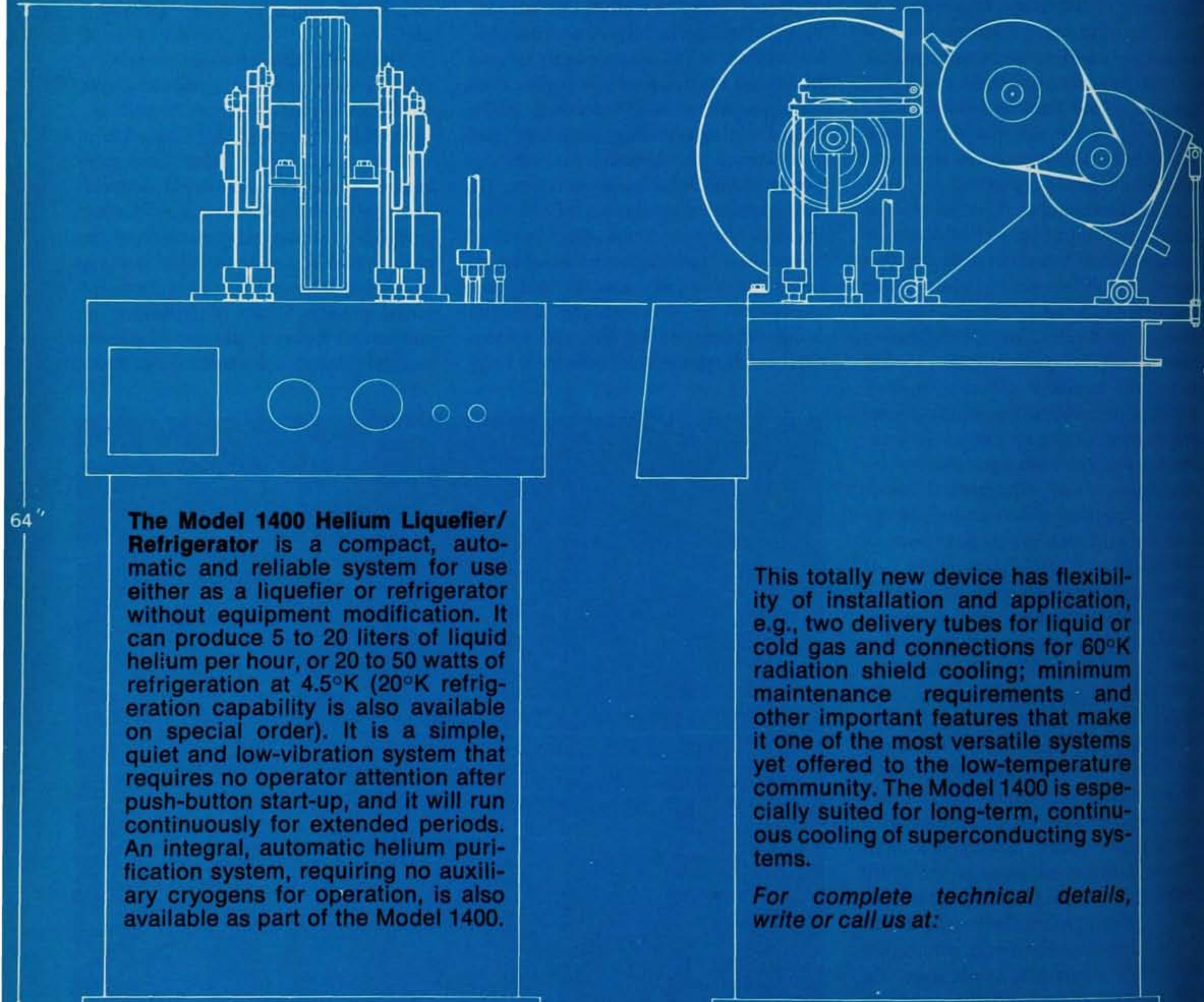

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Kelvin Park, 266 Second Avenue □ Waltham, Mass. 02154 □ (617) 899-8300 vacancies, not stabilized by nucleation, were annealed out at temperatures used for subsequent deposition and thus could not influence film growth.

Epitaxial growth. A lively interest was displayed in papers describing the initial stages of epitaxial growth and in phenomena such as pseudomorphism and misfit dislocations. It appears that, having chosen its mode of condensation, a film can during initial growth choose among a relatively large number of structural alternatives. Thus Shiro Ogawa (Tohoku University) demonstrated that during the "island" stage small particles formed around nucleation sites can adopt surprisingly complex, multiply twinned structures, in which the atomic distances differ from normal bulk values and approach those found for amorphous film structures.

An unusual example of atom shifts induced by substrate interactions, reported by Rolf Niedermayer (Physikalisches Institute, Clausthal), is observed in the early stages of epitaxy of silver islands on the (110) surface of single-crystal germanium. At low film thickness the silver layer exhibits, in the [001] direction of the substrate, an approximately 40 percent pseudomorphic strain, adopting approximately the spacing of germanium. With increasing thickness the strain is progressively reduced, until at about 40 monolayers the normal bulk spacing is assumed. Theory predicts that, at larger film thicknesses, interfacial stress arising from lattice-spacing differences between the epitaxial deposit and substrate should be relieved by the formation of regularly spaced misfit dislocations. Electron-microscope observations of such dislocations were illustrated for interface structures in the system germanium gallium-indium arsenide by Gunther Krause (Texas Instruments).

Internal stresses in thin films and the role of metastable modifications is promoting these stresses were reviewed by Werner Buckel (Universitat Karlsruhe). The subsequent paper of L. S. Palatnik (Lenin Polytechnic Institute, Kharkov) dealt at length with the chemical and structural changes involved in aging processes. In the absence of the author, unfortunately immobilized in Kharkov with a broken leg, this paper was read by the program chairman, Richard Hoffman (Case Western Reserve University); it yielded a lucid if somewhat gloomy picture of the complex oxidation, ordering, stress-relaxation, diffusion and precipitation phenomena encountered in thin films. It has been known for some time that thin films can sustain an unusually high internal stress, resulting apparently from pinning of dislocations. Siegfried Mader (IBM Research Center) reported studies performed with coworkers on the mechanisms of motion and pinning of dislocations, and he demonstrated that the stress needed to nucleate dislocations at grain boundaries is often much higher that the average intrinsic stress measured on the film.

Charge-carrier transport in thin films is affected not only by the same scattering processes present in corresponding bulk materials, but also by the more frequent collisions of drifting carriers with the film surfaces. The resultant surface scattering is a "size effect." It is a function of the mean free path l of the charge carriers relative to the film thickness d. Of particular interest is the degree of specularity of surface scattering. Richard Greene (US Naval Ordnance Laboratory) pointed out that a study of size effects can provide information about bulk and surface energy-bank structures as well as on charge-carrier scattering mechanisms in both normal and superconducting metals. Greene stated that size effects can affect the charge-carrier transport properties of solids through any or all of the following phenomena: surface mixing of bulk electronic energy states, surface effects on the electron–electron coupling in superconductors, surface effects on the phonon spectrum distribution and surface scattering.

A. V. Rzhanov (Institute of Semiconductor Physics, Novosibirsk) investigated charge-carrier scattering in thin germanium films grown by chemical vapor-phase transport. Rzhanov and his associates reported that surface scattering of electrons is diffuse from either the natural film surfaces or for surfaces prepared by polishing and etching. Scattering of holes is diffuse on the polished surfaces; on the virgin film surfaces it is only partially diffuse, with a temperature-independent probability for diffuse scattering of 0.3 to 0.4. The large surface-to-volume ratio enhances the importance of other surface-related phenomena in films. Such phenomena, the analogs of size effects, involve the relative size of l and the effective skin depth at rf frequencies, the distance l_w traversed by an electron during one cycle of a microwave field, the electrostatic shielding length L and the electron wavelength λ_e. Current topical interest concerns size-effect quantization in films. In degenerate semiconductors and semimetals, the size-effect energy levels depend on d and on the position of Fermi energy ϵ_F in the electronic band structure. As d decreases, the size-effect levels move past ϵ_F, which has an oscillatory character, producing in turn an oscillatory dependence in d of the measured resistivity, Hall coefficient and magnetoresistance as well as other related parameters.

Y. Katayama (Hitachi Central Research Laboratory) described the transport properties of n-type inversion layers of p-type indium antimonide at 4.2 K. He and his associates measured the magnetic-field dependence of the surface conductance increment $\Delta\sigma_s(H)$. He interpreted the angular and amplitude dependence between H and $\Delta\sigma_s$ in terms of the quantization of the magnetic Landau levels and the difference in electron scattering at the two surfaces of the inversion layer. Evidence of quantum size effects in tunneling experiments, using 0.08–0.2-micron thick bismuth films were described by V. N. Lutskyi and his associates (Institute of Radio Engineering and Electronics, Moscow). They found that the tunneling current has a nonmonotonic character as a function of voltage and interpreted this behavior as the result of dependence of the density of states on energy in quantum-size films.

Noncrystalline solids. The considerable current interest in noncrystalline solids was reflected in papers presented mainly in two sessions, one concerned with lattice disorder and diffusion, chaired by J. D. MacKenzie (Rensselaer Polytechnic Institute), and the other, on amorphous film, chaired by P. Duwez (California Institute of Technology). C. N. J. Wagner (Yale University) discussed small-angle scattering and large-angle x-ray and electron diffraction in terms of electron density fluctuations in highly disordered solids. Using diffraction studies as the sole criteria, he found difficulties in discriminating between amorphous films and randomly oriented microcrystals (about 3nm). Both yield quantitatively similar diffuse diffraction patterns. C. Breitling

(University of Tubingen), classified noncrystalline solids in terms of lattice-like and liquid-like amorphous substances.

Examples of the former are solid amorphous arsenic, germanium and silicon. Breitling stated that in amorphous germanium the interatomic distance between (111) double layers is greater than in crystalline germanium and suggested that the sigma bonding of electrons is changed into a pi configuration. H. Richter (Technische Hochschule, Stuttgart) described amorphous bismuth, iron and gallium as examples of liquid-like structures. The degree of order in amorphous films is, however, higher than that associated with the liquid state. Richter found evidence of both spherically close-packed atomic-chain structures as well as a layer lattice structure in the same amorphous films, and he suggested that this double structure stabilizes the noncrystalline state.

Electrical conduction in noncrystalline nonmetallic films was discussed by A. K. Jonscher (Chelsea College of Science and Technology). He described the conduction process in terms of a "mobility gap" and a highly localized high density of states in the effective bandgap of amorphous semiconductors with due attention to hopping motion of charge carriers. Jonscher emphasized the importance of making both ac and dc measurements, particularly on the frequency dispersion of the loss tangent of noncrystalline films. Considering experiments made on the anomalous field emission from amorphous semiconducting films, A. G. Zhdan and his associates (Institute of Radio Engineering and Electronics, Moscow) interpreted the "s"-shaped negative resistance observed in such films as the result of space-charge limited currents in the presence of high trap densities. Screening by free carriers causes the disappearance of localized levels, the formation of a high-conduction channel and an abrupt switching from a low-conductance to a high-conductance state. A. Witt and his associates (Drexel Institute of Technology) reported similar reversible switching phenomena in noncrystalline organic films of tetracene and copper phthalocyanine 0.1 – 4 microns thick.

Optical properties. The influence of a granular structure, crystalline defects and impurities on the optical properties of thin metallic films were reviewed by F. Abelès (Institut d'Optique, Université de Paris). He pointed out the various spectral ranges, from the infrared through the ultraviolet, where the film structure has a significant effect on measured optical parameters. Provided that single crystal films are available, information about the electronic energy-band structure of solids in spectral ranges well above their fundamental energy gap can be obtained readily from films. W. Paul (Harvard University), an eloquent exponent of this point of view, addressed two principal problems. The first was to obtain the most accurate spectrum of the complex dielectric constant $\epsilon^*(\omega)$ from optical transmission measurements on films. Paul's second problem was how to obtain the best resolved fine-structure in $\epsilon^*(\omega)$ irrespective of its absolute magnitude. A comparison of $\epsilon^*(\omega)$ data obtained by transmission measurements on films with those obtained from reflection measurements on corresponding bulk crystals shows good agreement, provided that the structural order of the films is high. Paul suggested that both single crystals and polycrystalline films are particularly useful for investigating critical points of the energy band structure. He also thought that films lend themselves to exploitation of differential methods (electroreflectance, piezoreflectance, temperature modulation) for even sharper resolution of the optical spectra.

For investigations concerning the electrical and optical properties of solids, it is often cheaper and faster to grow films rather than bulk crystals; this economy is of particular interest if a series of alloys with varying composition is under investigation. This type of work was reported by Jay Zemel (University of Pennsylvania) and his associates for the series $Cd_xPb_{1-x}S$. They found a systematic increase in the energy of the fundamental absorption edge as x is increased and an apparent shift from direct to indirect bandgap transitions.

Recrystallization. On the final morning of the conference, a complete session was devoted to recrystallization in films. Two types of transformation were considered — recrystallization of a polycrystalline structure from a molten zone, and crystallization from the amorphous state. The first effect was demonstrated in four papers dealing with recrystallization of indium antimonide films. Probably the most photogenic example was presented by Neil Davis (Corona Laboratories) who showed cine pictures of electron-beam zone recrystallized films (see figure). They demonstrated that, depending upon stoichiometry, zone temperature and rate of zone motion, crystallization could be made to proceed by dendritic or near-planar growth. David Brandon (The Technion, Israel) reported studies on electron-beam-induced crystallization of amorphous anodic oxides of aluminum, titanium, tantalum, niobium and tungsten, and showed that each oxide crystallized by a discrete nucleation and growth sequence. It is thought that such amorphous-to-crystalline transformations may be related to electrolytic breakdown and to changes in the mechanical properties of the films.

This report provides only a sampling of the many excellent papers that constituted the scientific program. It indicates, we hope, the variety of subjects and the range of subdisciplines involved in the study and research on films—filtered through the somewhat bemused perceptions of your reporters as we tried to keep up with the tightly paced double sessions over a five-day period. The large attendance, 97 from abroad and about 400 from the US, is an indication of the world-wide interest in the scientific and technological aspects of thin films. The conference chairman, K. H. Behrndt (NASA Electronics Research Center), drew attention to this fact in his opening remarks and stressed the significance of the feedback loop that connects fundamental research, technological application of thin films and the need for continued international coöperation and exchange of information on the many problems that remain as yet to be solved.

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The conference was organized by the thin-film division of the American Vacuum Society and sponsored by the International Union of Pure and Applied Physics and by the International Union for Vacuum Science Technology and Applications. The proceedings will be published as the July–August issue of the Journal of Vacuum Science Technology.

H. H. Wieder
Infrared Division, Corona Laboratories
Maurice Francombe
Westinghouse Research and Development Center