Chapter 5. Microstructural Evolution in Thin Films of Electronic Materials

Academic and Research Staff

Professor Carl V. Thompson, Dr. Craig Ballentine, Dr. Paul D. Bristowe, Dr. En Ma,[1] Dr. John Melngailis, Dr. Robert C. O'Handley

Visiting Scientists and Research Affiliates

Dr. Sergei Bogomolov,[2] Dr. Harold J. Frost,[3] Hideo Miura,[4] Takauki Uda,[4] Dr. A. Wagner,[5] Derek T. Walton[6]

Graduate Students

Roland Carel, Steven S. Cooperman, Anthony D. Della Ratta, Andrew D. Dubner, Jerrold A. Floro, Heather E. Inglefield, Young-Chang Joo, Harold Kahn, Brett D. Knowlton, Ya-Chin Liu, Hai P. Longworth, Jaesang Ro

Technical and Support Staff

Peter C. Heron

5.1 Electromigration and Microstructure

Electric-current-induced atomic self-diffusion, or electromigration, causes open and short circuit interconnect failures in integrated circuits. Electromigration occurs primarily by diffusion along grain boundaries, and interconnect failure initiates at grain boundaries, so that interconnect reliability is strongly dependent on the grain sizes and orientations in polycrystalline interconnects. The relationships between these microstructural characteristics and interconnect reliability have become more complex as interconnect dimensions (widths and thicknesses) have become comparable to grain sizes. We have studied the effects of microstructure on the reliability of submicron and larger interconnects as well as vias and contacts. We have done this by using a variety of techniques for controlling grain sizes and grain size distributions in thin films. We are also developing models for microstructural evolution in order to develop accurate models for electromigration in submicron interconnects.

5.1.1 Control of Microstructures in Aluminum Films

Sponsor

Joint Services Electronics Program
Contract DAAL03-89-C-0001

Project Staff

Hai P. Longworth, Professor Carl V. Thompson

In previous years, we reported the use of precipitate-induced abnormal grain growth to produce grains as large as 1 mm in 1 μm thick films. In the past year, we completed an extensive study in which we have established the requirements for precipitate-induced abnormal grain growth in thin films and have demonstrated similar

[1] Presently at the University of Michigan, Ann Arbor, Michigan.

[2] Ural Polytechnical Institute, Sverdlovsk, Russia.

[3] Dartmouth College, Hanover, New Hampshire.

[4] Hitachi Limited, Tokyo, Japan.

[5] IBM Corporation, Yorktown Heights, New York.

[6] Formerly of Dartmouth College, currently with Digital Equipment Corporation, Hudson, Massachusetts.

phenomena in Al-Cu-Cr, Al-Ag-Cr, Al-Mn-Cr, and Al-W alloys. We have found that to optimize the precipitate-induced abnormal grain growth process, the alloy additions should initially be present as pure layers in the middle of the Al film. In alloys containing Cr, the Cr serves to aid in the nucleation of precipitates, leading to localization of the precipitates in the middle of the film, and leading to a larger number of smaller, more closely spaced precipitates. Through controlled abnormal grain growth, the microstructure of thin films can be controllably varied over broad ranges.

5.1.2 Reliability of Vias

Sponsor

Semiconductor Research Corporation

Project Staff

Harold Kahn, Professor Carl V. Thompson

Over the past year, we have developed a via test structure that allows statistical characterization of via reliability by stressing multiple vias in *parallel*. We have used this process to characterize the reliability of vias with W plugs and have shown that under normal test conditions, joule heating of the plugs contributes to accelerated open failures of Al interconnects. We are developing simple thermal models to evaluate the effect of joule heating at service conditions. We have also demonstrated that the grain size in the Al interconnect greatly affects via reliabilities and also the statistics of failure. Large grains lead to bimodal failure distributions. We are developing tools for statistical analysis of via reliability.

5.1.3 Modeling of Microstructures in Interconnects

Sponsors

Semiconductor Research Corporation
National Science Foundation

Project Staff

Derek T. Walton, Dr. Harold J. Frost, Professor Carl V. Thompson

Over the years, we have developed and tested models for crystal nucleation and growth to form continuous polycrystalline films. We have also modeled grain growth in continuous polycrystalline films. In the past year we have modified these models to investigate the evolution of the microstructures of polycrystalline interconnects to "bamboo" structures, as illustrated in figure 1. We find that the approach to a fully bamboo structure is exponential in time and that the rate of transformation is inversely proportional to the square of the strip width. When the simulation is extended to model grain boundary pinning due to grooving at grain boundary-free surface intersections, we find that there exists a maximum strip width-to-thickness ratio (approximately 3) beyond which the transformation to the bamboo structure does not proceed to completion. In earlier work, we have shown that reliability of nearly-bamboo and fully-bamboo lines is greatly different. Our modeling work is now allowing us to identify the conditions under which it is reasonable to expect to obtain only fully-bamboo lines.

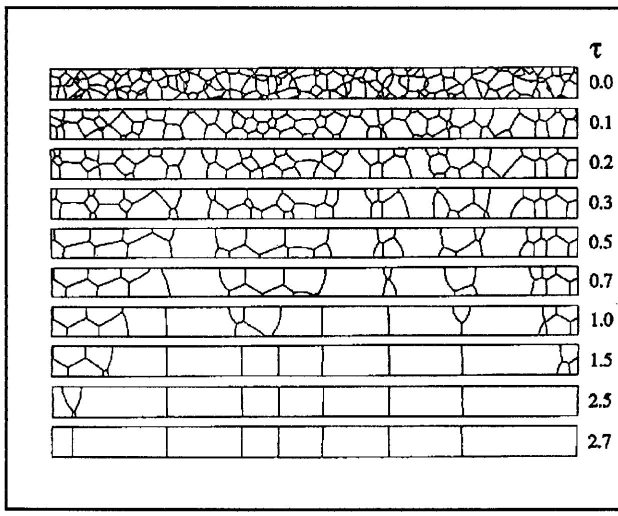


Figure 1. Evolution of the grain structure within a strip. The transformation to a "bamboo" structure is dominated by the growth of bamboo sections into regions of grain clusters. The rate of this transformation depends strongly on the strip width.

5.1.4 Electromigration in Bicrystals

Sponsors

Joint Services Electronics Program
Contract DAAL03-89-C-0001
Semiconductor Research Corporation

Project Staff

Hai P. Longworth, Young-Chang Joo, Brett D. Knowlton, Professor Carl V. Thompson

In previous work we have shown that individual grain boundaries greatly affect the reliability of an interconnect line. In order to understand the reliability of interconnects, it is therefore important to understand the reliability of individual grain boundaries. To do this we have developed a new technique that allows us to characterize the reliability of populations of lines with single, identical grain boundaries with controlled types, locations, and orientations. The technique is schematically shown in figure 2. We start by making NaCl bicrystals of the desired type, on which we grow epitaxial bicrystal Al films which are transferred and sintered to oxidized silicon wafers. We then use conventional lithographic techniques to pattern multiline test structures with the same controlled Al grain boundary running through all the lines. We can then investigate, as an example, the effect of the boundary orientation on the reliability of the line, as illustrated in figure 3.

We have shown that (1) failure times of lines containing single identical boundaries are lognormally distributed, (2) the medium time to failure depends more strongly on the boundary orientations than the types of grain boundaries, (3) the deviation in the time to failure has a large component not dependent on microstructure, and (4) both interfacial diffusion and grain boundary diffusion appear to contribute to failure in bicrystal lines and probably in bamboo and near-bamboo lines as well.

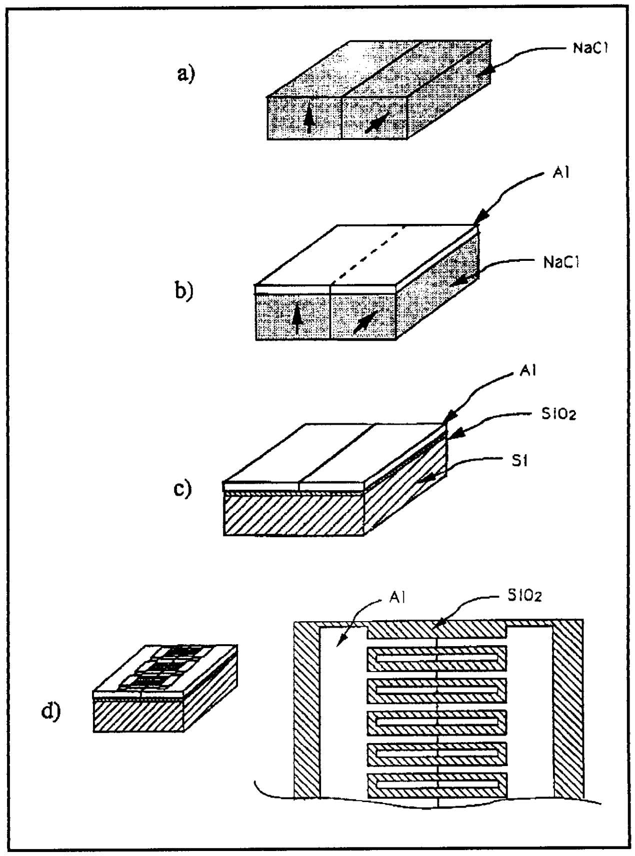


Figure 2. The fabrication process of bicrystal test patterns. (a) NaCl bicrystal substrate made from two NaCl single crystals; (b) Al bicrystal thin film epitaxially deposited on the NaCl substrate; (c) Al film transferred to an oxidized Si wafer; and (d) patterning.

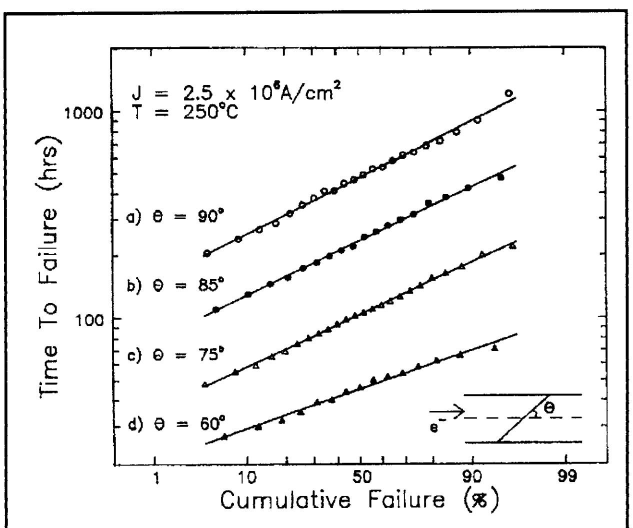


Figure 3. Failure distributions of $\Sigma13[100]$ bicrystal lines as a function of θ, the angle between the current direction and the grain boundary plane.

5.2 Microstructural Evolution in Polycrystalline Films

Polycrystalline thin films are used in a wide variety of applications, especially in electronics. In these applications the properties and performance of polycrystalline films depend strongly on the average grain size, the distribution of grain sizes, and the distribution of grain orientations. These are controlled by the deposition conditions and the conditions for post-deposition processing. In the past, we have carried out modeling and experiments on the effects of crystal nucleation and growth and grain growth on the microstructure of thin films. This has included work on semiconductors as well as metals, and has explored the effects of dopants, ion bombardment, and rapid thermal annealing, as well as other processing conditions. Our current activity is focused on modeling of grain growth in continuous and patterned films and experiments on epitaxial grain growth.

5.2.1 Modeling of Grain Growth in Thin Films

Sponsor

National Science Foundation

Project Staff

Jerrold A. Floro, Derek T. Walton, Roland Carel, Dr. Harold J. Frost, Professor Carl V. Thompson

We have developed a mean field model for grain growth in thin films which accounts for the effects of the energies of the free surfaces and the film/substrate interfaces of the films. Anistropies of these energies affect the evolution of the distribution of grain orientations, as well as grain sizes. With our analysis we can predict the differences in the rate of grain growth and film texture evolution that result from different specific interface or surface energy versus orientation relationships. These predictions are being compared with experimental results.

In collaboration with Dr. H.J. Frost of Dartmouth College, we have also developed computer simulations of grain growth in thin films. We have included in our simulations the effects of grain boundary drag due to surface grooving. This leads to stagnation of normal grain growth at a point where the grain sizes are lognormally distributed and the average grain diameter is about three times the film thickness. This simulation result closely matches experimental results in a wide variety of systems. We can also include the effects of surface and interface energy anisotropy and determine the conditions which lead to surface-energy-driven abnormal or secondary grain growth. These simulations allow investigation of the effect of the surface and interface energy anisotropy on the evolution of texture in thin films. Our mean field models allow us to model and predict the behavior of large populations of grains, while our computer simulations allow us to investigate the behavior of smaller populations of grains as well as specific grain configurations. We have also recently modified our computer simulations to model grain growth in patterned films, as described in Section 5.1.3.

5.2.2 Epitaxial Grain Growth

Sponsor

National Science Foundation

Project Staff

Jerrold A. Floro, Roland Carel, Dr. Paul D. Bristowe, Professor Carl V. Thompson

When polycrystalline films on single crystal substrates undergo grain growth, interface-energy-anisotropy leads to the preferred growth of epitaxially oriented grains. We have demonstrated this phenomenon in metallic films deposited on single crystals of NaCl, mica, and MgO. We have also demonstrated epitaxial grain growth in polycrystalline Ag films on single crystals of Ni. We have chosen to study the latter experimental system because calculations of the structure and energies of Ag/Ni interfaces for a number of different Ag/Ni relative crystallographic orientations exist. These calculations have been shown to be consistent with results from rotating sphere experiments in which very small single crystal Ag spheres rotate into low energy orientations when annealed on single crystal Ni substrates. As-deposited epitaxial films of Ag on Ni are also readily attainable. We have found that the epitaxial orientations obtained are different in each of these three types of experiments (as-deposited epitaxy, rotating spheres, and epitaxial grain growth). These results suggest that the orientation obtained by conventional epitaxy may be metastable once a film is continuous. In exploring this issue, we are using embedded atom calculations to calculate interface and surface energies and using the results from these calculations in our grain growth models and simulations.

5.3 Magnetic Properties of Heteroepitaxial Thin Films

Sponsors

National Science Foundation
U.S. Army Research Office

Project Staff

Heather E. Inglefield, Dr. Sergei Bogomolov, Dr. Craig Ballentine, Dr. Robert C. O'Handley, Professor Carl V. Thompson

In this new program we are investigating the effect of strain and misfit dislocations on the magnetic properties of very thin ($\lesssim$10 nm) heteroepitaxial metallic films. In particular, we are studying Ni films deposited on Ni-Cu alloy films. In this and related systems, when overlayers are very thin, they will grow pseudomorphically, adopting the lattice parameter of the substrate. However, as the overlayer thickens, the strain energy will become sufficient to drive deformation and the formation of misfit dislocations. Grids of perpendicular arrays of misfit dislocations often result. These structural changes are expected to have profound effects on the magnetic properties of the films. We are preparing to use measurements of the magneto-optic Kerr effect (MOKE) and low energy electron spin polarization analysis (LEESPA) to analyze changes in magnetic properties *during* film deposition. We will correlate changes in magnetic properties with changes in structure as determined using *in situ* reflection high energy electron diffraction (RHEED), as well as *ex situ* x-ray diffraction analysis and transmission electron microscopy.

In addition to using *in situ* magnetic characterization of films to probe and monitor structural changes, we will also investigate the effects of misfit dislocations on the large-scale and small-scale magnetic properties of films. As part of this analysis we will use scanning electron microscopy and polarization analysis (SEMPA) to image magnetic domains with a spatial resolution smaller than the dimensions of the misfit dislocation grid.

5.4 Interface Reactions in Multilayer Thin Films

Sponsors

Hitachi Limited
IBM Corporation

Project Staff

Dr. En Ma, Hideo Miura, Takauki Uda, Professor Carl V. Thompson

When two materials are brought into contact and heated, interdiffusion will lead to compound formation. This phenomena is important throughout technology, but is especially important in electronics in understanding unwanted reactions between films and substrates, as well as desired reactions that form silicides and diffusion barriers. Despite the importance of interface reactions, the initial processes that control which compound phase will form during a reaction (phase selection) is not well understood. We have investigated phase selection during interface reactions by using multilayer films in which we have alternately deposited pure layers with layer thicknesses as low as a few nanometers. By using multilayer films, we can focus on the earliest stages of interface reactions because these early reactions consume all or most of the volume of the overall films. This allows the use of tools such as transmission electron microscopy, x-ray analysis, and differential scanning calorimetry (DSC) to study formation of compound layers that are only a few nanometers thick. DSC is especially useful in studying the kinetics as well as the thermodynamics of these processes. We have used these techniques to study reactions in Ni/Si, Ti/Si, V/Si, Co/Si, and Ni/Al multilayer films. We have shown that interdiffusion always precedes nucleation of a compound and have argued that interdiffusion is in fact a necessary precursor to compound formation. Phase selection is therefore constrained by the rates of self-diffusion and interdiffusion. This insight has allowed us to explain our frequent observations that the initial phase to form is often metastable. We are also now able to make predictions about phase selection sequences in interface reactions.

5.5 Focused Ion Beam Induced Chemical Vapor Deposition

Sponsor

U.S. Army Research Office

Project Staff

Anthony D. Della Ratta, Jaesang Ro, Andrew D. Dubner, Dr. A. Wagner, Dr. John Melngailis, Professor Carl V. Thompson

A focused ion beam (FIB) can be used to cause local deposition of a film to write lines for x-ray mask repair or circuit restructuring. Ions are accelerated to the substrate where each ion can cause tens of decomposition events in a chemical vapor deposition process. We have carried out experiments focused on understanding the energy transfer mechanism which leads to FIB-induced chemical vapor deposition. The process is substrate mediated and scales with the ion energy and ion mass in such a way as to strongly suggest that decomposition is caused by collisions between knock-on substrate atoms and adsorbed monolayers of unreacted molecules. It also appears that removal of the reactant product can be rate limiting. It is found that film purities are improved when films are deposited at lower rates or higher temperatures. These insights will help in the development of processes for deposition of high purity lines with controlled widths and aspect ratios.

5.6 Publications

Dubner, A.D., A. Wagner, J. Melngailis, and C.V. Thompson. "The Role of the Ion/Solid Interaction in Ion Beam Induced Deposition of Gold." *J. Appl. Phys.* 70: 665 (1991).

Evans, P.V., D.A. Smith, and C.V. Thompson. "Absence of Electrical Activity at High Angle Grain Boundaries in Zone-Melt-Recrystallised Silicon-on-Insulator Films." *Appl. Phys. Lett.* 60: 439 (1992).

Floro, J.A., and C.V. Thompson. "Epitaxial Grain Growth and Orientation Metastability in Heteroepitaxial Thin Films." *Materials Research Society Symposium Proceedings* 187: 274 (1991).

Frost, H.J., C.V. Thompson, and D.T. Walton. "Simulation of Thin Film Grain Structures: II. Abnormal Grain Growth." *Acta Metall. Mat.* Forthcoming.

Frost, H.J., C.V. Thompson, and D.T. Walton. "Abnormal Grain Growth in Thin Films Due to Anisotropy of Free-Surface Energies." Proceedings of the First International Conference on Grain Growth in Polycrystalline Materials, Rome, 1991. Forthcoming.

Jiran, E., and C.V. Thompson. "Capillary Instabilities in Thin Continuous Films." *Thin Solid Films.* Forthcoming.

Kahn, H., and C.V. Thompson. "A Statistical Characterization of Electromigration-Induced Open Failures in 2-Level Metal Structures." *Materials Research Society Symposium Proceedings* 225: 15 (1991).

Kahn, H., and C.V. Thompson. "The Effect of Applied Mechanical Stress on the Electromigration Failure Times of Aluminum Interconnects." *Appl. Phys. Lett.* 56: 1308 (1991).

Longworth, H.P., and C.V. Thompson. "Abnormal Grain Growth in Aluminum Alloy Thin Films." *J. Appl. Phys.* 69: 3929 (1991).

Longworth, H.P., and C.V. Thompson. "An Experimental Study of Electromigration in Bicrystal Al Lines." *Appl. Phys. Lett.* Forthcoming.

Ma, E., and C.V. Thompson. "Nucleation and Growth During Reactions in Multilayer Al/Ni Films: The Early Stages of Al_3Ni Formation." *J. Appl. Phys.* 69: 2211 (1991).

Ma, E., L.A. Clevenger, C.V. Thompson, and K.N. Tu. "Kinetic and Thermodynamic Aspects of Phase Evolution." *Materials Research Society Symposium Proceedings* 187: 83 (1991).

Ma, E., C.V. Thompson, and L.A. Clevenger. "A Calorimetric Study of the Kinetics of Al_3Ni Nucleation and Growth During Reactions in Al/Ni Thin Films." *Materials Research Society Symposium Proceedings* 157: 455 (1991).

Miura, H., E. Ma, and C.V. Thompson. "Initial Sequence and Kinetics of Silicide Formation in the Cobalt/Amorphous-Silicon Multilayer Thin Films." *J. Appl. Phys.* 70: 4287 (1991).

Thompson, C.V., J.Y. Tsao, and D.J. Srolovitz, editors. "Evolution of Thin Film and Surface Microstructure." *Materials Research Society Symposium Proceedings* 202 (1991).

Thompson, C.V. "On the Role of Diffusion in Phase Selection During Reactions at Interfaces." *J. Mater. Res.* 7: 367 (1992).

Thompson, C.V., L.A. Clevenger, R. DeAvillez, E. Ma, and H. Miura. "Kinetics and Thermodynamics of Amorphous Silicide Formation in Metal/Amorphous Multilayer Thin Films." (Invited Paper) *Materials Research Society Symposium Proceedings* 187: 61 (1991).

Thompson, C.V. "Experimental and Theoretical Aspects of Grain Growth in Thin Films." (Invited Paper) Proceedings of the First International Conference on Grain Growth in Polycrystalline Materials, Rome, 1991. Forthcoming.

Walton, D.T., H.J. Frost, and C.V. Thompson. "The Development of Bamboo and Near-Bamboo Microstructures in Thin Film Strips." *Appl. Phys. Lett.* Forthcoming.

Walton, D.T., H.J. Frost, and C.V. Thompson. "Computer Simulation of Grain Growth in Thin-Film Interconnect Lines." *Materials Research Society Symposium Proceedings* 225: 219 (1991).

Walton, D.T., H.J. Frost, and C.V. Thompson. "Modeling of Grain Growth in Thin Film Strips." Proceedings of the First International Conference on Grain Growth in Polycrystalline Materials, Rome, 1991. Forthcoming.

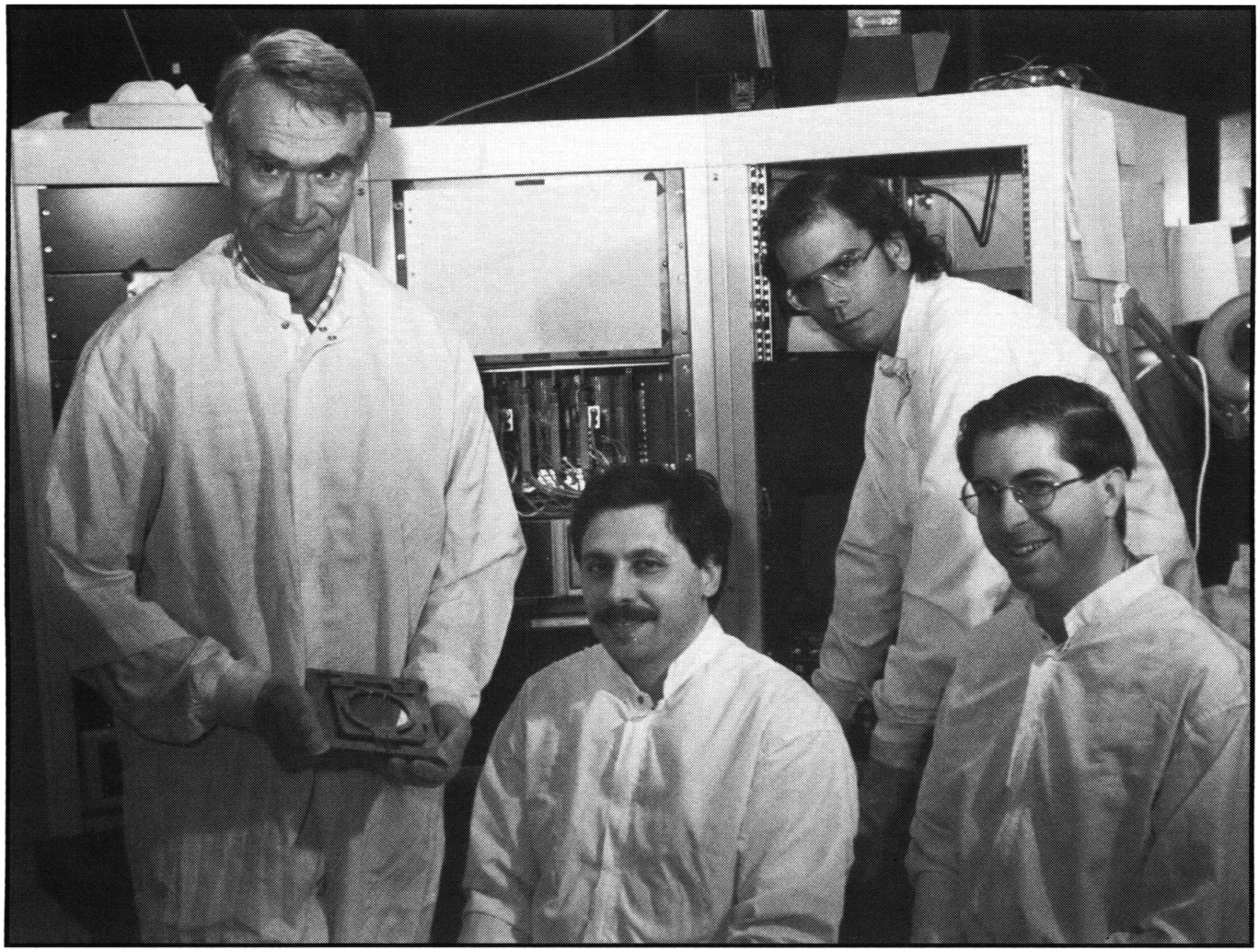
5.7 Theses

Cooperman, S. *Electromigration in Submicron-Wide Interconnects.* S.M thesis. Dept. of Mater. Sci. and Eng., MIT, 1992.

Liu, Y. *Coplanar-Spherical-Particle Coarsening in Discontinuous Ultrathin Polycrystalline Gold Films on Amorphous Silicon-Nitride Substrates.* Ph.D. diss. Dept. of Mater. Sci. and Eng., MIT, 1991.

Longworth, H. *Electromigration in Aluminum and Aluminum Alloy Thin Films with Controlled Microstructures.* Ph.D. diss. Dept. of Mater. Sci. and Eng., MIT, 1992.

Ro, J. *Microstructural Evolution During Focused Ion Beam Induced Deposition.* Ph.D. diss. Co-supervised with Dr. J. Melngailis. Dept. of Mater. Sci. and Eng., MIT, 1991.



From left: Senior Research Scientist Dr. John Melngailis with Research Engineer Sergey Etchin of MIT's Microsystems Technology Laboratories, and Research Assistants Christian R. Musil and Henri J. Lezec