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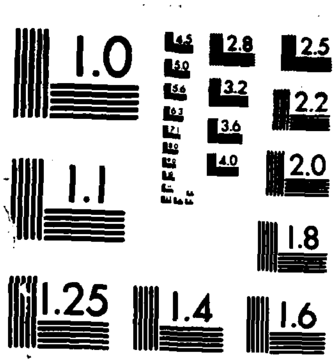

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"Single Crystal Films of Semiconductors on Amorphous Substrates Via a Low Temperature Graphoepitaxy"

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Massachusetts Institute of Technology
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The objective of this program is to carry out basic research in order to acquire fundamental understanding which will permit the development of a general, low temperature process for obtaining oriented, defect-free single-crystal semiconductor films on amorphous substrates.

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SUMMARY

Surface-energy-driven grain growth (SEDGG) has been investigated as a means of achieving device-quality semiconductor films on amorphous substrates. The time, temperature and film thickness dependence of grain growth was measured for Si and Ge, and the effect of P, As, and B doping in Si was studied. Grain growth rates are consistent with theory for early times. At longer times, a marked decrease in the rate of growth occurs which is attributed to thermal grooving at primary grain boundaries. Enhancement of grain growth by doping with electron donors is consistent with a Fermi-energy dependent increase in vacancy concentration. Ion bombardment dramatically enhances grain growth in Ge and Au at low temperature. Simultaneous ion bombardment and deposition appears to be a highly promising strategy for SEDGG-based graphoepitaxy. A deposition system to accomplish this has been procured.

I. INTRODUCTION

Future military electronic systems will require a wide range of device quality crystalline films. This research program is focussed on new low-temperature methods for forming device-quality films on amorphous substrates. Such a capability will permit greater flexibility and more highly integrated systems. At the present time, device-quality crystalline films can be formed from only a limited number of materials and on a limited number of substrates. Moreover, the defect density in such films is usually higher than desired. Novel means of eliminating or confining defects are also investigated in this research program.

II. BACKGROUND

Conventional heteroepitaxy requires a single-crystal substrate with a specific orientation and lattice-constant relationship to the film. Thus, there are severe constraints on the available film-substrate combinations. Similar constraints apply to various means of lateral epitaxy that are used to achieve crystalline films on amorphous substrates (XOA). The fundamental investigations pursued under this grant should lead to a much clearer understanding of crystalline film formation in general, and a practical low-temperature means of forming a wide variety of crystalline film on amorphous substrates.

Zone-melting recrystallization (ZMR) has been highly successful in producing Si films on SiO_2 of sufficient quality for MOS devices. In ZMR, however, Si must be melted and hence the process is intrinsically a high temperature one and of limited flexibility. In this work we have focussed on lower temperature solid phase processes.

III. SURFACE ENERGY DRIVEN GRAIN GROWTH (SEDGG)

The approach we have been pursuing under the AFOSR grant is called surface-energy-driven grain growth (SEDGG). It is depicted schematically in Fig. 1. In brief, an ultrathin film is formed on a substrate. The film is sufficiently thin that after normal grain growth, grains extend entirely through it, forming a columnar structure. Some grains are oriented such that their surfaces have minimum energy. As a result, a driving force exists which preferentially drives growth of grains with minimum surface energy. These grains consume their neighbors and become large secondary grains with a specific crystallographic texture. The driving force due to surface energy increases as the film thickness is decreased. Thus, our work is with films less than 1000 Å thick.

Secondary Grain Growth

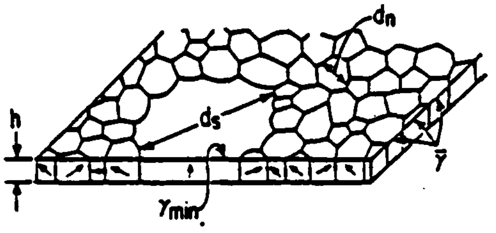


Figure 1. Schematic depiction of surface-energy-driven grain growth (SEDGG). A grain with minimum surface energy, $\gamma_{min.}$, is shown growing into a matrix of normal grains with average surface energy $\bar{\gamma}$.

$$\Delta\gamma = \gamma_{min} - \bar{\gamma}$$

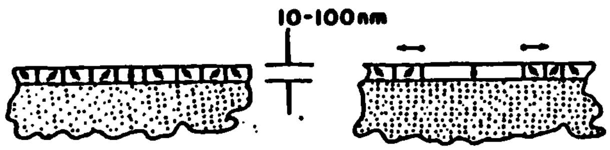
The driving force due to surface energy anisotropy

$$\Delta F = \frac{2A\Delta\gamma}{Ah} = \frac{2\Delta\gamma}{h}$$

To get large grains with uniform texture
decrease the film thickness

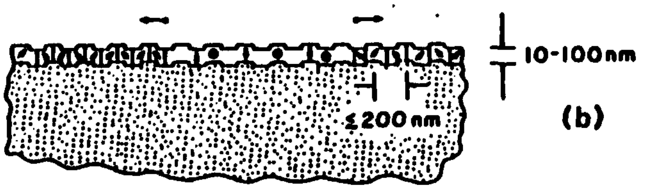
Figure 2 illustrates how surface-energy-driven grain growth (SEDGG) in conjunction with patterning of the substrate surface can lead to a film with a specific in-plane orientation as well as a specific axis perpendicular to the film. Growth to impingement of three-dimensionally oriented grains can lead to single crystal films.

SURFACE-ENERGY DRIVEN



(a)

Figure 2(a). Schematic cross section of a film undergoing SEDGG. The grain with minimum interfacial energy, by virtue of its orientation, grows by consuming grains with other orientations.



(b)

Figure 2(b). SEDGG in conjunction with surface patterning (solid-state graphoepitaxy). A grain of minimum interfacial energy is one that is oriented relative to the surface pattern as well as the substrate normal.

The key issues in achieving oriented single-crystal films via SEDGG and surface patterning are: 1) how to increase the mobility of grain boundaries so that the grain with minimum surface energy can grow rapidly and at low temperature while avoiding film beading; 2) how to increase the driving force for oriented secondary grain growth; 3) how to minimize the number of defects and control their location. To achieve the objectives of this program requires an emphasis on fundamental studies. We must understand interfacial energetics and the mechanisms of grain boundary motion in order to provide effective strategies for enhancing mobility, increasing the driving force, or controlling defect location.

IV. RESEARCH OBJECTIVEES

The following research objectives are aimed at acquiring fundamental knowledge about surface and interfacial energetics, and grain boundary motion in ultra-thin semiconductor films. This fundamental knowledge will help us to evaluate various methods of direct intervention aimed at promoting grain boundary motion and single-crystal film formation.

1. Investigate the kinetics of SEDGG including:
   a. the temperature, time and film-thickness dependence;
   b. the effect of stress.
2. Further develop a theory for SEDGG and related phenomena.
3. Experimentally investigate specific aspects of SEDGG in semiconductors, including:
   a. the origin of grain-growth slowing and saturation;
   b. the effect of encapsulation;

   c. the effect of dopants;
   d. the kinetics of grain boundary grooving and thermal beading.

4. Investigate means of enhancing grain boundary mobility, including:
   a. the use of ion bombardment;
   b. the use of dopants;
   c. the use of intense, rapid illumination.
5. Develop strategies for enhancing SEDGG and in-plane orientation, including:
   a. simultaneous ion bombardment and deposition;
   b. patterning films;
   c. use of ultrathin films, < 200 Å
6. Investigate the use of sub-100 nm lithography to pattern surfaces and induce in-plane orientation as well as control defect density and defect locations.

IV. STATUS OF THE RESEARCH

To date, the SEDGG phenomenon has been observed in all materials we have studied (Si, Ge, Au, and B_aF_2). We believe it is universal. In-plane orientation induced by fine surface-relief gratings (i.e., SEDGG-based grphoepitaxy) has also been demonstrated in those materials tried so far, Ge and Au. Thus, the basic feasibility of our approach is established.

In this Section, progress during the first year is described briefly with reference to the research objectives. More details are given in the publications listed in Section V.

1. Kinetics of SEDGG

We have developed the theory of SEDGG and use this theory to interpret experimental results. The temperature, time and thickness dependence of secondary grain growth has been measured experimentally for Si, Ge, Au and Al (the latter two under other sponsorship). Comparison with theory can be made, for example, by measuring the fraction of the film transformed into secondary grains as a function of time and temperature. For early times, growth rate is linear and is in agreement with the simplest model for grain boundary motion. Kinetic analysis of deviations from linear growth indicated that grain-boundary grooving was responsible for the slowing down or saturation in the grain growth rate. Thus, we now believe that grain growth saturation is due to grooving.

Encapsulation was shown to suppress themally-induced grooving and beading in Ge and Si. However, cross-sectioned TEM revealed that grooving still occurs even in the presence of a cap and thus may contribute to inhibiting grain growth.

2. Dopants

In the case of Si, dopants have a dramatic effect on grain growth. Heavy P or As doping enhances grain boundary mobility while a compensating doping with B can cancel the effect. Current evidence indicates that the enhancement effect is due to the increases in the total vacancy concentration associated with increases in the Fermi energy. These vacancies, which are negatively charged, are presumed to promote grain boundary dislocation climb, thereby increasing the grain boundary mobility. The precise mechanism has not yet been determined.

We have investigated and will continue to investigate techniques, such as ion bombardment, which lead to very high, non-equilibrium vacancy concentrations.

3. Grooving and Beading

Grain boundary grooving and film beading are driven by surface energy minimization and occur primarily through surface diffusion. Grain boundary grooving (the formation of a groove at the line of intersection of a grain boundary and the film surface) can lead to a drag on moving boundaries, thus retarding or even completely suppressing grain growth. Kinetic analysis of SEDGG indicates that grooving is responsible for decaying grain growth rates in Al and Au. Specifically, the growth rate becomes proportional to time to the 1/4 power. SEDGG in silicon capped with SiO_2 films is initially linear in time but also slows down at fairly short times. These results suggest that initial linear growth rates might be quite high but that grain boundary grooving must be eliminated for growth rates to remain high. Suppression of grooving will be one of the major foci of continuing work.

Grain boundary grooving can eventually lead to formation of holes in a film. Once these holes form, the film can begin to bead in order to minimize its total surface energy. This beading process occurs in solid films when they are very thin and therefore have high surface-to-volume ratios. The driving force for beading as a function of film thickness, surface energy, and grain size has been theoretically determined.

4. Ultrathin Films

Both SEDGG and beading are enhanced in ultrathin films and are therefore competing processes. However, preliminary experimental evidence in 150 Å-thick Ge films suggests the driving force for SEDGG is so high that it can occur even when holes have formed in the film and beading has initiated. We therefore plan to further investigate still thinner films and also to investigate means of suppressing beading.

5. Ion-Beam-Enhanced Grain Growth

We have demonstrated that ion bombardment of a thin polycrystalline film greatly enhances the motion of grain boundaries. This ion-beam-enhanced grain growth process has been demonstrated in thin Ge and Au films. Ge was deposited in the amorphous phase on SiO_2 and subsequently crystallized to form a polycrystalline film. This film was bombarded by a 50keV Ge ion beam while at a temperature of 500-700°C. The increase in grain size was proportional to the implanted ion dose, and was essentially independent of temperature in this temperature range. The enhancement of grain size was especially dramatic considering that the sample temperature was as much as 437°C below the melting temperature (T_m = 937°C for Ge). In order to achieve an equivalent grain growth rate by thermal annealing alone, the film would have to be annealed at approximately 100°C below the melting temperature. In addition to promoting grain boundary motion, ion-beam-enhanced grain growth also leads to a sharp reduction in the density of dislocations within grains, as compared to thermal annealing at an equivalent temperature. So far, enhanced growth of normal grains has been observed. It is anticipated that enhanced secondary grain growth will be observed when we implant with higher ion doses.

In work under other sponsorship, we have demonstrated ion beam-enhanced grain growth in 500 Å-thick Au films implanted with 200 keV Xe^+ ions. As in the work on Ge, the grain growth enhancement was proportional to ion dose. Large grains with unifrom {111} texture were observed, a strong indication that secondary grain growth occurred.

6. Illumination

Experiments were carried out on the use of intense illumination to enhance grain boundary mobility in P-doped Si. We obtained extremely rapid grain growth and believe this is due to the fact that grain growth occurs very rapidly before grooving acts to suppress it. Initial temperature calibrations led us to infer that an optical effect was at work i.e., preferential bond breaking at grain boundaries. However, it is now believed that the effect is purely thermal, and that rapid initial growth is entirely consistent with the theoretical analysis. That is, it reflects the character of the growth prior to grooving.

Based on our research to date we believe the optimal strategy for achieving SEDGG and SEDGG-based graphoepitaxy is to employ ion bombardment to stimulate grain growth at low temperatures while simultaneously depositing new material to prevent grooving and to compensate for material lost by sputtering. In this way the film will remain thin and smooth, while under a constant flux of energetic ions and arriving atoms. Independent evidence in experiments with Au indicated that grain growth is more rapid during deposition than afterwards. We have purchased and installed a new ultra-high vacuum dual e-beam deposition system in which this work will be done.

Earlier we found that patterning films into stripes a few micrometers wide greatly promotes secondary grain growth. Enhancement may be due to stress relief or edge effects, or perhaps other effects. Investigation of this phenomenon will be made in the second year.

As mentioned earlier, in ultrathin (~15 nm) films of Ge a very dramatic grain size enhancement is observed. This may be due entirely to an enhanced driving force, but it may also be related to the presence of a large number of small voids in the film. The edges of these voids may serve as sites for grain boundary defect formation and hence promote growth. For example, some researchers believe that grain boundaries move by mechanisms which can be described as ledge nucleation and growth. Voids might be sites for enhanced ledge formation. We plan to investigate films which are intentionally made discontinuous.

In order to induce in-plane orientation by surface-relief patterning (i.e., SEDGG-based graphoepitaxy illustrated in Fig. 2) the relief sidewalls should be smooth and straight, and the ratio of groove depth to linewidth should be maximized. Since groove depth should not also be greater than perhaps 1/5th of the film thickness (i.e., $\lesssim$ 10 nm) it is important to reduce the linewidth as far below 100 nm as possible while retaining smooth walls. At present our relief structures have very rough sidewalls, corresponding to angular deviations exceeding 10 degrees. It is perhaps somewhat surprising that the graphoepitaxial alignment achieved to date is as good as it is , which may be an indicator of improvements to be achieved with better patterning. We have conceived new techniques for achieving sidewalls that may be smooth on the nanometer scale and have linewidths of 40 nm. Preliminary results look promising.

V. LIST OF PUBLICATIONS

1. Journal Articles

C.V. Thompson, "Secondary Grain Growth in Thin Films of Semiconductors: Theoretical Aspects", J. Appl. Phys. 58, 763 (1985).

H.-J. Kim, C.V. Thompson, "Compensation of Grain Growth Enhancement in Doped Silicon Films", Appl. Phys. Lett., 48, 399 (1986).

C.C. Wong, H.I. Smith and C.V. Thompson "Surface-Energy-Driven Secondary Grain Growth in Thin Au Films", Appl. Phys. Lett., 48, 335 (1986).

H.J. Frost and C.V. Thompson "The Effect of Nucleation Conditions on the Topology and Geometry of Two-Dimensional Grain Structures," to be published in Acta. Metallurgica.

2. Published Conference Proceedings

H.I. Smith, M.W. Geis, C.K. Chen, and C.V. Thompson,"Crystalline Films on Amorphous Substrates by Zone Melting and Surface-Energy-Driven Grain Growth in Conjunction with Patterning." Proc. Fall Meeting of the Materials Research Society, Symposium C, Semiconductor-on-Insulator and Thin Film Transistor Technology, Boston Marriott Hotel, Boston, MA., Dec. (1985).

H.A. Atwater, H.I. Smith and C.V. Thompson "Enhancement of Grain Growth in Ultra Thin Germanium Films by Ion Bombardment", Proc. Mat. Res. Soc., fall meeting, Boston, MA, Dec. 2-6, 1985.

C.V. Thompson and H.I. Smith, "Secondary Grain Growth in Thin Films", Proc. fall meeting of the Mat. Res. Soc., Boston, MA, Dec. 2-6, 1985.

H.J. Kim and C.V. Thompson, "The Effects of Dopants on Surface-Energy-Driven Grain Growth in Ultrathin Si Films", Proc. fall meeting of the Mat. Res. Soc., Boston, MA, Dec. 2-6, 1985.

C.V. Thompson, "Secondary Grain Growth in Ultra-Thin (<100 nm) Films of Silicon and Germanium" (invited), Interface Migration and Control of Microstructure Symposium of the Annual American Society of Metals Congress, Detroit, Michigan, Sept. 1984.

H.J. Frost and C.V. Thompson, "Microstructural Evolution in Thin Films," presented at the Symposium on Computer Simulation of Microstructural Evolution, fall TMS-AIME meeting, Toronto Canada, October (1985).

H.-J. Kim and C.V. Thompson, "The Effect of Dopants on Grain Boundary Mobility in Silicon". Presented at the Japanese Institute of Metals International Symposium on Grain Boundary Structure and Related Phenomena, November (1985).

H.J. Frost and C.V. Thompson, "Modelling of Thin Film Grain Structures and Grain Growth," Proceedings of Computer Based Microscopic Description of the Structure and Properties of Materials Symposium of the fall meeting of the Materials Research Society, December (1985).

3. Theses

J.E. Palmer, "Secondary Grain Growth in Ultra Thin Germanium Films on Silicon Dioxide," M.S. Thesis, Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, August, 1985.

C.C. Wong, "Secondary Grain Growth and Graphoepitaxy in Thin Au Films," Ph.D Thesis, Department of Materials Science & Engineering," Massachusetts Institute of Technology, February, 1986.

4. Miscellaneous Presentations

H.A. Atwater, H.I. Smith, and C.V. Thompson, "Enhancement of Grain Growth in Ultra-Thin Germanium Films by Ion Bombardment", VLSI Review, M.I.T., December 1985.

H.A. Atwater, "Device Quality Crystalline Films on Amorphous Substrates by Zone-Melting Recrystallization and Grain Growth Processes," presented at the Sohio Research Ctr. Cleveland, OH.

VI. RELATED WORK

We have continued research on the theory of normal and secondary grain growth in thin films. Both analytical and computational modelling has been carried out. We have recently demonstrated (using several analytical techniques) that secondary grain growth cannot be driven by reduction of grain boundary energy alone. (It should be noted that this result is contrary to popular belief.) This confirms our expectation that other forces, such as surface energy anisotropy, are required for secondary grain growth. This encourages us in our belief that, when properly controlled, surface-energy-driven grain growth can be highly selective.

We have also developed computer models for microstructural evolution during film formation and during grain growth. These models will eventually allow simulation of the effects of modified film formation conditions and of the early as well as late stages of secondary grain growth. Analytical modeling of film formation has also been improved.

As an outgrowth of the work sponsored by AFOSR we feel that we have gained new insights into the mechanisms of conventional epitaxy. When the surface energy for the film/substrate interface is sufficiently high (as is usually the case for couples forming incommensurate or discommensurate interfaces) film growth initiates through formation of discrete, three-dimensional clusters on the substrate surface. This is known as Volmer-Weber growth. It may be that these clusters or nuclei are oriented with respect to the substrate as soon as they form. However, there is evidence that in some systems nuclei are not oriented and remain misaligned until a continuous film forms. It is at this point that surface-energy-driven grain boundary motion can lead to formation of an epitaxial, single crystal film.

In our AFOSR sponsored work we use artificial surface topography to produce in-plane anisotropy which leads to growth of three-dimensionally oriented grains. In heteroepitaxy, film/substrate lattice registry leads to in-plane surface energy anisotropy and can lead to epitaxy via grain growth. We have begun experimental investigation of this mode of epitaxy and are seeking additional support from other agencies for this work.

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