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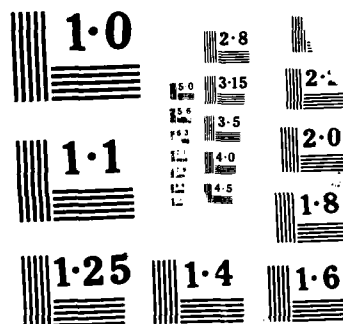
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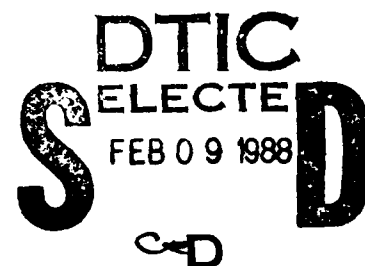
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Interim Report for
1 August 1986 through 30 September 1987

for

Contract N00014-84-K-0545

R&T No. NR 372-162



MICROSCOPIC CONTROL OF SEMICONDUCTOR INTERFACE REACTIVITY

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EXECUTIVE SUMMARY - The goal of our program is to control semiconductor surface and interface behavior by means of local modifications of the surface/interface chemical environment. Major thrust areas include the use of ultrathin catalyst layer to promote low temperature oxidation and nitridization of semiconductor surfaces, the search for passivating layers to reduce metallization corrosion, and the exploitation of diffusion barrier effects to control interdiffusion at metal-semiconductor junctions. Such studies have potential implications for low temperature synthesis of gate and field oxides, and for enhancing metallization stability against corrosion and electromigration.

Current technological trends emphasize the formation of passivating layers and the use of diffusion barriers in the interface region as methods of controlling corrosion and electromigration without imposing a radical change in materials or fabrication technology. Both methods modify the local environment of one of the two interfaces present in the system, i.e. the overlayer-atmosphere interface, and the semiconductor-overlayer interface. In fact, the two interfaces are thermodynamically coupled and the modification of one of the two may result in modification of the other. Recent studies of Au,^{1,2} Ag³⁻⁴ and Cr⁵ overlayers on Si(111), for example, have shown that exposure to oxygen yields growth of a Si-oxide layer on top of the metal, and that such a process is strongly dependent on the morphology of the silicon-metal interface. For different metals, such as Cr,^{6,7} the oxide is nucleated at the metal-semiconductor boundary, i.e. below the overlayer. When the oxide is nucleated underneath the metal, instead, as in the case of Cs and Na,^{6,7} ultrathin metal overlayers can be used as catalysts of semiconductor oxidation and allow low-temperature oxide growth.⁸ After growth of an oxide of suitable thickness, the metal catalyst can be removed with a number of etching techniques.⁸

Atomic interdiffusion at semiconductor-metal interfaces takes place during junction formation and processing for a large number of metals,^{9,10} even when no electric field is applied. Interdiffusion has been observed even at liquid nitrogen temperature,¹¹ it may be chemically activated,¹¹ and it determines the microscopic junction profile and the stoichiometry of the interface reaction products. For compound semiconductors, the preferential diffusion of one of the semiconductor constituents through the interface may affect the dominant type of electrically active defects that remain near the semiconductor surface. Such defects together with new localized interface states¹⁰ produced by the local reaction of metal and semiconductor atoms, determine the Schottky barrier and transport properties of the junction. To interpose suitable interlayers between the semiconductor and the metal has therefore the potential to control the junction profile and modify the Schottky barrier.^{12,13}

We are examining a number of potential catalysts for the oxidation of Si and GaAs surfaces, and exploring diffusion barrier effects at GaAs and Hg_{1-x}Cd_xTe/metal junctions. Our goals are to understand the microscopic mechanisms that determine catalytic and diffusion barrier effects, clarify the connection with the local chemical environment at the interface and identify the systems that offer the highest promise of technological applications. We use synchrotron radiation photoemission spectroscopy, supplemented by Auger and

XPS spectroscopies, on interfaces prepared *in situ* on cleaved single crystal substrates. Detail on the experimental procedures can be found in References 1-10.

OXIDATION STUDIES - The deposition of most (but not all) metal overlayers on Si and GaAs surfaces yields an increase of the oxygen uptake rate relative to the clean semiconductor surface. Metals as diverse as Au¹⁻³, Ag¹⁻³, Cu,⁴ Pd,⁴ Cr,⁵ Cs,^{6,7} Na,⁷ and Sm^{6,7} have all been shown to give rise to such an effect. Our data show that the reaction products involve nonequivalent oxidation states for silicon, and are consistent with what could be expected for a disordered, substoichiometric SiO₂ phase. In the case of Cr, as well as Au,¹⁻³ Ag³ and possibly the other transition metal examined,⁴ the Si oxide appears to nucleate on top of the metal overlayer. Most models proposed to explain this effect suggest that the formation of a silicide-like interface reaction product breaks up the sp³ configuration of the Si substrate so that the Si atoms at the interface would be in a metallic environment with "disrupted bonds" and they could easily be oxidized. Abbati *et al.*⁴ proposed that a self-sustaining mechanism exists for which a flow of Si atoms is established through a silicon-metal silicide-like layer located between the growing oxide and the Si substrate. This layer would make Si atoms with broken sp³ configuration and weaker⁴ Si-Si bonds available for oxidation. The incoming oxygen would then react preferentially with Si and displace the metal so that the metal would become available for intermixing with the substrate and produce further silicide.

This model does not seem to apply in full to the low-electronegativity overlayers Sm,⁶ Yb, Cs,⁷ and Na.⁷

For Cs and Na (and possibly Sm) the Si-oxide appears to nucleate below the metallic layer. Furthermore, the alkali metals do not intermix with silicon, do not form silicide-like reaction products, and they do not break the sp³ hybridization of the Si surface atoms. We have proposed⁷ that for low-electronegativity overlayers the establishment of a large surface dipole is responsible for oxidation promotion. The chemisorption of low-electronegativity metals induces a modification of the electrostatic potential in the surface region. The metal valence electrons are polarized toward the substrate surface leading to a dipole layer which raises the electron levels of the surface relative to the vacuum level. The rigid shift of the Fermi level yields a measurable reduction of the work function. If a molecular species is adsorbed within the surface dipole layer, the energy shift of the molecular level is less pronounced than that of the substrate levels. This results in charge transfer to molecular antibonding orbitals, and formation of atomic oxygen that can react with Si atoms in the interface region. Similar phenomena, i.e. the nucleation of semiconductor oxides on top or below a metallic overlayer, have been observed on GaAs surfaces.

We are examining the validity of the different models proposed through systematic studies of the catalytic activity of alkali metals, alkali earth and rare earth metals on Si(111) and GaAs(110) surfaces. We are looking for a systematic connection between interface morphology, electronegativity, work function and specific catalytic activity of the overlayer. Overlayers to be examined include but

are not limited to K, Cs, Na, Ca, Eu, Gd, and cover a wide range of interface morphologies characteristic overlayer parameters.

Within the elements examined to date, the most promising appear the rare earth Sm and Yb. Both elements exhibit large catalytic activity, appear to promote the nucleation of semiconductor oxides below the overlayer, and are potentially less dangerous impurities than the alkali metals. The Patent Office of the University of Minnesota concurred in our assessment of the technological implications, and U.S. Patent applications have been filed (See Publications/Patents/Presentations/Honors Report) to cover the use of Sm and Yb catalysts during Si and GaAs oxidation. The first patent (U.S. Patent #4,684,541) was awarded on August 4, 1987.

DIFFUSION BARRIER STUDIES - Very few studies exist of the microscopy mechanisms that determine the properties of diffusion barriers. Al interlayers at GaAs(110)-metal interfaces have been shown to change dramatically atomic interdiffusion through the interface.¹⁰ Al interlayers at the CdS-Au interface have also been shown to change the Schottky barrier.¹² With increasing interlayer thickness the macroscopic junction behavior evolves from rectifying to ohmic. The origin of this impressive result is still subject of debate. On Si, Cr interlayers at the Si(111)-Au interface exhibit a striking non-monotonic effect related to the chemically activated character of the interdiffusion.¹³

In our study of diffusion barriers we have concentrated on compound semiconductors that exhibit critical metallization stability problems. The ternary alloy system $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ (MCT) offers a major challenge to standard contact technology. This important infrared detector material exhibits lattice and surface instabilities that derive from the coexistence of two different bonds (Cd-Te and Hg-Te) of widely different stability in the same matrix. In general, deposition of typical metallizations involving Al, In or refractory metals yields preferential metal/Te reaction at the surface, and formation of a Hg-depleted surface layer near the interface. Since the semiconductor optical and transport properties vary with the Hg-content, contact fabrication affects device performance and often gives unreliable or unreproducible results.

We laid a background to our diffusion barrier studies with photoemission investigations of binary MCT/metal and GaAs/metal junctions, involving metals as diverse as Al, Cr, Ag, Au, Sm, Yb, Cs, K and Na (see attached Publications/Patents/Presentations/Honor report and previous yearly reports). In a recent study we have examined the relationship between thermodynamic driving forces, and diffusion barrier effects for interfaces between the low stability narrow-gap $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ semiconductor alloys and typical reactive metals.¹⁴ Our semi-empirical calculations of the binary thermodynamic parameters following Miedema's model suggested that the rare earths should act as effective diffusion barriers at the interface between MCT and metals such as Al, In, or Cr, because of the superior thermodynamic stability of the MCT/rare earth reaction products and because of the chemically-driven nature of the intermixing. Our synchrotron radiation photoemission studies demonstrated that monolayers of Yb are effective in preventing Al-Te reaction and reducing the Al-induced mercury depletion of

the substrate.¹⁴

In those cases in which the driving force for interdiffusion is not only chemical, such as during high temperature processing or electromigration, a diffusion barrier may still be effective on the basis of two possible microscopic mechanisms. Chemical trapping of the moving species in the interface region may be possible by choosing an interlayer with large chemical affinity for the diffusing atoms. The ensuing chemical reaction could produce interface reaction products that structurally or electronically block the microscopic avenues for entropic and chemically activated diffusion. Alternatively, the establishment of an electrostatic dipole field at the semiconductor-metal interface may prevent diffusion of polar species without the formation of new reaction products.

One can distinguish between these two mechanisms on the basis of microscopic analysis of the interface region (depth profiling) or from the dependence of the diffusion barrier effect on interlayer thickness. For example, in the case of Al interlayers at the GaAs(110)-Cr interface, we have shown that the presence of the interlayer yields a trend towards Ga-rich outdiffusion in the overlayer that is maximum already at 1 Å interlayer thickness and remains relatively constant at higher thickness. This is in sharp contrast with the case of Al interlayers at the GaAs-Au interface,¹⁰ for which an increase of over one order of magnitude was observed in the relative Ga to As outdiffusion, but the effect was shown to increase monotonically with interlayer thickness. We proposed that in the Cr case the dipole field at the semiconductor-metal interface is the dominant mechanism in controlling interdiffusion, as opposed to the Au case, where chemical trapping of the As atoms in the interlayer is the main mechanism.¹⁵ Our results are in fact consistent with the evolution of the dipole layer, that is formed at low interlayer thickness and is largely unaffected by further increase in interlayer thickness.¹⁰

We are extending our studies of diffusion barriers at GaAs and MCT/metal interfaces following the same methodology. Namely, we are conducting synchrotron radiation photoemission studies of atomic interdiffusion across the interface as a function of interlayer thickness and interlayer electronegativity (local dipole). The measurements are supplemented by semiempirical calculations of the binary thermodynamic parameters following Miedema's model. Systems to be examined include, but are not limited to, GaAs/Yb/Cr, GaAs/Cr/Al, GaAs/Ag/Cr, and GaAs/Cr/Ag. For MCT we are examining a number of rare earth metal diffusion barrier and several reactive overlayer such as Al, In, Cr and Ti. The use of monolayer rare earth diffusion barrier to stabilize metal contacts on MCT seems to date the major technological application of our study. The Patent Office of the University of Minnesota concurred with our assessment, and on October 30, 1987, a U.S. Patent application has been filed to cover the use of rare earth diffusion barriers during MCT/metal contact fabrication (See Publication/Patents/Presentation/Honors Report).

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15. S. Chang, C. Caprile, E. Colavita, and A. Franciosi, *Solid State Commun.* (in press); see also attached publication list.

OFFICE OF NAVAL RESEARCH

PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS REPORT

for

1 August 1986 through 30 September 1987

for

Contract N00014-84-K-0545

R&T No. NR 372-162

MICROSCOPIC CONTROL OF SEMICONDUCTOR INTERFACE REACTIVITY

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a. Papers Submitted to Refereed Journals (and not yet published):

- a1. A. Franciosi, A. Raisanen, A. Wall, S. Chang, P. Philip, N. Troullier, and D.J. Peterman, "Ytterbium Monolayer Diffusion Barriers at $\text{Hg}_{1-x}\text{Cd}_x\text{Te}/\text{Al}$ Junction", Applied Physics Letters (in press). Supported in part by McDonnell Douglas Independent Research and Development Program.
- a2. S. Chang, P. Philip, A. Wall, A. Raisanen, N. Troullier, G. Haugstad, and A. Franciosi, "Yb-Induced versus Sm-Induced Oxidation Promotion at Si(111) and GaAs(110) Surfaces", submitted to Physical Review B. Supported in part by the Minnesota Microelectronics and Information Science Center.
- a3. A. Raisanen, A. Wall, S. Chang, P. Philip, N. Troullier, A. Franciosi, and D.J. Peterman, "Diffusion Barriers at Mercury-Cadmium-Telluride/Metal Interfaces: The Case of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}(110)/\text{Yb}/\text{Al}$ ", submitted to J. of Vacuum Science and Technology. Supported in part by McDonnell Douglas Independent Research Program.
- a4. S. Chang, C. Caprile, E. Colavita, and A. Franciosi, "Trapping Mechanisms and Diffusion Barriers: Al Interlayers at the GaAs(110)-Cr Interface", submitted to Solid State Communications. Supported in part by the Minnesota Microelectronics and Information Science Center.

b. Papers Published in Refereed Journals:

- b1. A. Franciosi, P. Soukiassian, P. Philip, S. Chang, A. Wall, A. Raisanen, and N. Troullier, "Electronic Promoters and Semiconductor Oxidation: Alkali Metals on Si(111) Surfaces", Physical Review B35, 910 (1987). Supported in part by the Minnesota Microelectronics and Information Science Center.
- b2. E. Colavita, M. DeCrescenzi, S. Nannarone, P. Philip, S. Chang, A. Wall, and A. Franciosi, "Surface Electron Energy Loss Investigation of Cr 2p Core Edge: Bulk Cr and Si-Cr Interfaces", *Vuoto* (Italy) 16, 38 (1986). Supported in part by the Italian National Research Council International Collaboration Fund.
- b3. S. Chang, P. Philip, A. Wall, A. Raisanen, N. Troullier, and A. Franciosi, "Low-electronegativity Overlayers and Enhanced Semiconductor Oxidation: Sm on Si(111) and GaAs(110) Surfaces", Physical Review B35, 3013 (1987). Supported in part by the Minnesota Microelectronics and Information Science Center.
- b4. A. Wall, A. Raisanen, S. Chang, P. Philip, N. Troullier, A. Franciosi, and D.J. Peterman, "Semiconductor Composition During Metal/Mercury-Cadmium-Telluride Interface Formation", J. of Vacuum Science and Technology A5, 1535 (1987). Supported in part by the McDonnell Douglas

Independent Research and Development Program.

- b5. A. Franciosi, P. Philip, S. Chang, A. Wall, A. Raisanen, N. Troullier, and P. Soukiassian, "Catalysis of Semiconductor Surface Reactions", in *Proceedings of 18th Int. Conference on the Physics of Semiconductors*, O. Engstrom, ed., World Scientific Publ. Co., Singapore, 1987, pp. 141-144. Supported in part by the Minnesota Microelectronics and Information Science Center.
- b6. A. Wall, A. Raisanen, S. Chang, P. Philip, N. Troullier, A. Franciosi, and D.J. Peterman, "Local Stoichiometry and Atomic Interdiffusion During Reactive Metal/Mercury-Cadmium-Telluride Junction Formation", *J. of Vacuum Science and Technology*, A5, 3193 (1987). Supported in part by the McDonnell Douglas Independent Research and Development Program.
- b7. S. Chang, P. Philip, C. Caprile, A. Wall, and A. Franciosi, "Microscopic Control of Metallization Stability", in *Proceedings 3rd Annual ASM International Conference on Electronic Packaging*, ASM International, Metals Park, Ohio, 1987, pp. 35-40.
- b8. A. Wall, A. Raisanen, S. Chang, N. Troullier, A. Franciosi, and R. Reifenberger. "CdTe(110)-Mn Interface Reaction and the Formation of Non-equilibrium Ternary Semimagnetic Semiconductor Alloys", *J. of Vacuum Science and Technology* A May/June (1988).
- b9. D.J. Peterman, A. Raisanen, S. Chang, P. Philip, A. Wall, and A. Franciosi, "Nonmonotonic Evolution and Thermodynamic Trends at Metal/(Hg,Cd)Te Interfaces: Yb/Hg_{0.78}Cd_{0.22}Te". *J. of Vacuum Science and Technology* A May/June (1988).

c/d. Books Submitted for Publication/Published:

None.

e. Patents Filed:

- e1. A. Franciosi, "Ytterbium-promoted oxidation of silicon and gallium arsenide surfaces", U.S. Patent application No. 114,773 filed on October 30, 1987.
- e2. A. Franciosi, "Method to stabilize metal contacts on mercury-cadmium-telluride alloys", U.S. Patent application filed on October 30, 1987. Supported in part by the McDonnell Douglas Independent Research and Development Program.

f. Patents Granted:

- f1. A. Franciosi, "Samarium-promoted oxidation of silicon and gallium arsenide surfaces", U.S. Patent application filed on June 11, 1986, U.S. Patent #4,684,541 granted on August 4, 1987. Supported in part by the Minnesota Microelectronics and Information Science Center.

g. Invited Presentations at Topical or Scientific/Technical Society Conferences:

- g1. A. Franciosi, "Catalytic Control of Semiconductor Surface Oxidation", Annual Meeting of the American Institute of Chemical Engineers, Miami Beach, Florida, November 2-7, 1986.
- g2. A. Franciosi, "Semiconductor and Metal Interfaces", series of four invited lectures to the GNSM-CISM Summer School *Physics of Surfaces, Interfaces, Superlattices*, Villa S. Giovanni, Italy, September 14-26, 1986.

h. Contributed Presentations at Topical or Scientific/Technical Society Conferences:

- h1. A. Franciosi, S. Chang, P. Philip, A. Wall, A. Raisanen, N. Troullier, and P. Soukiassian, "Catalysis of Semiconductor Surface Reactions", *18th Int. Conference on the Physics of Semiconductors*, Stockholm (Sweden), August 11-15, 1986.
- h2. A. Wall S. Chang, A. Rizzi, C. Caprile, A. Franciosi, and D.J. Peterman, "Composition Control During Reactive Metal/Mercury-Cadmium Telluride Interface Formation", *33rd National Symposium AVS*, Baltimore, Maryland, October 27-31, 1986.
- h3. S. Chang, P. Philip, A. Wall, A. Raisanen, N. Troullier, A. Franciosi, and P. Soukiassian, "Electronic Promoters and Semiconductor Oxidation", *19th Annual Synchrotron Radiation Center Users Group Conference*, Stoughton, Wisconsin, October 20-21, 1986.
- h4. A. Wall, S. Chang, P. Philip, A. Raisanen, N. Troullier, A. Franciosi, and D.J. Peterman, "Local Semiconductor Stoichiometry During Reactive Metal/HgCdTe Junction Formation", *1986 Workshop on the Physics and Chemistry of Mercury-Cadmium-Telluride*, Dallas, Texas, October 7-9, 1986.
- h5. S. Chang, P. Philip, A. Wall, C. Caprile, A. Raisanen, N. Troullier, A. Franciosi, and P. Soukiassian, "Metallic Promoters of Semiconductor Oxidation", *14th Annual Conference on the Physics and Chemistry of Semiconductor Interfaces*, Salt Lake City, Utah, January 26-29, 1987.

- h6. S. Chang, P. Philip, C. Caprile, A. Wall, A. Franciosi, "Microscopic Control of Interface Stability", *3rd Ann. Am. Society for Metals Int. Conf. on Electronic Packaging*, Minneapolis, Minnesota, April 28-30, 1987.
- h7. A. Raisanen, A. Wall, S. Chang, P. Philip, N. Troullier, A. Franciosi, and D.J. Peterman, "Diffusion Barriers at MCT/Reactive Metal Junctions: Yb Interlayers at the $\text{Hg}_{1-x}\text{Cd}_x\text{Te}(110)/\text{Al}$ Interface", *1987 U.S. Workshop on the Physics and Chemistry of Mercury-Cadmium-Telluride*, New Orleans, Louisiana, October 6-8, 1987.
- i. **Honors/Awards/Prizes:**

None.
- j. **Graduate Students and Postdoctorals Supported Under the CRP for the Year Ending 1 October 1987:**
 - j1. P. Philip, graduate student in Materials Science, Ph.D. expected in 1989.
 - j2. A. Raisanen, graduate student in Materials Science, Ph.D. expected in 1990.
 - j3. Dr. M. Pedio, postdoctoral associate, now at Frei Universitat, Berlin.
 - j4. Professor S. Nannarone, visiting investigator, Department of Physics University of Rome.

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