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13. ABSTRACT (Maximum 200 words)

FeAl has been grown on psuedomorphic AlAs films on GaAs substrates. The growth was characterized by in situ reflection high energy electron diffraction, transmission electron diffraction, and electron channeling. Procedures for the growth of high quality films were developed and the defects analyzed. It was found that a low temperature nucleation followed by a high temperature anneal and low temperature growth gave strong layer by layer growth. RHEED intensity oscillations were observed for the first time in a metal aluminide system, allowing for precise control over ultrathin films. The first metal aluminides grown using metal organics were prepared. The results indicated that though there were advantages to this procedure, it was necessary to use full chemical beam epitaxy to avoid carbon contamination. The first measurement of critical thickness was made. GaAs was grown on top of FeAl films. The construction of a scanning tunneling microscope was begun.

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Contents

1 Objectives

2 Introduction

Based on the work reported here, we now have the opportunity to grow epitaxial metals on III-V semiconductors that will offer microelectronic structures with new levels of reliability and new classes of functionality. These metallic films grow one atomic layer at a time so that continuous, ultrathin, hyperabrupt structures can be fabricated. These thin films can be magnetic and they can be buried by subsequent semiconductor epitaxy. Fig. 1 shows a high resolution cross-sectional TEM photograph of an AlAs–FeAl–AlAs–GaAs structure that was grown in our laboratory under our current AFOSR program. This is the highest quality semiconductor ever grown on a metal. Fringes in each layer can be seen that correspond to columns of atoms. The modulated intensity indicates that the films are strained, as expected in this film that has only a few dislocations.

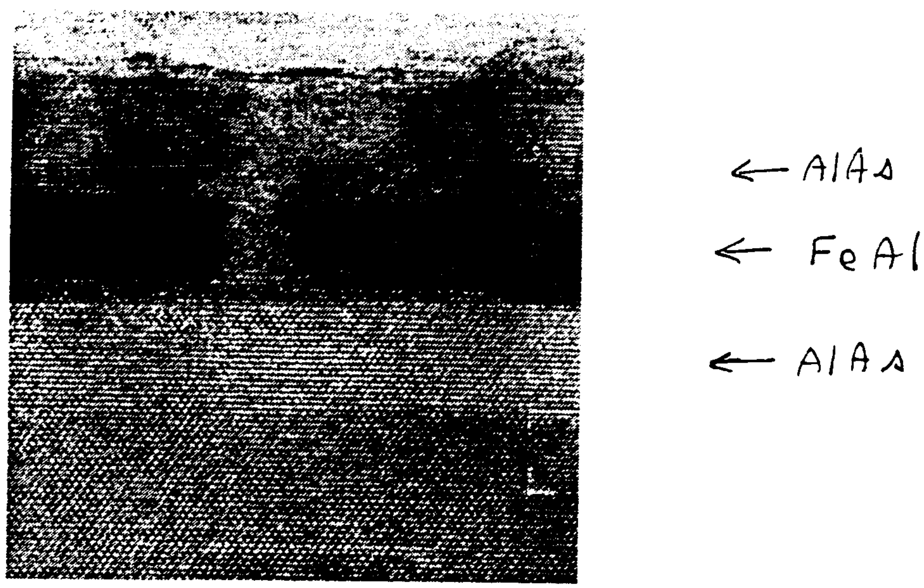

Figure 1: High resolution, cross-sectional TEM photograph of an AlAs/FeAl/AlAs grown by molecular beam epitaxy.

This work is based on the concept of growing ultrathin metal films that are thermodynamically stable. Normally if one deposits a metal on GaAs, say Ni, reactions take place forming nickel arsenides and nickel gallides. The principle here is to deposit the reaction product, removing the driving force for further reaction. From another point of view, one considers the ternary phase diagram of say Fe, Al, and As, shown in Fig. 2. This phase diagram is a plot showing the coexistence

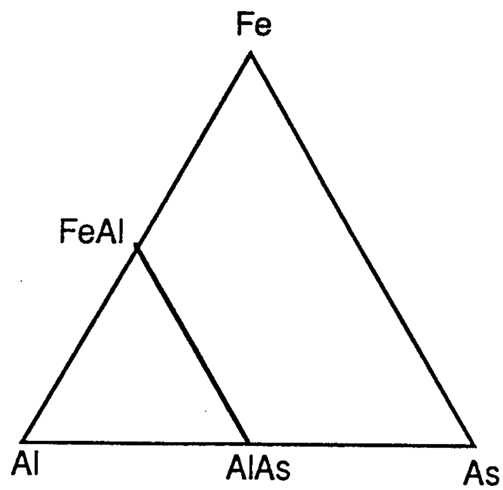

Figure 2: Portion of the ternary phase diagram showing phases that can coexist. Since there is not a tie line between Fe and AlAs, the pure metal cannot coexist with AlAs and will react in equilibrium. However, there is a tie line between FeAl and AlAs so that these two phases can coexist.

regions of Fe, Al, and As (similar ones can be drawn for other metals on III-V's). Since there is no tie line between Fe and AlAs, one cannot obtain a region in which elemental Fe and AlAs coexist. They should always (in equilibrium) react forming various compounds. This means that depositing a layer of pure Fe on AlAs is a fundamentally unstable structure. However, FeAl and AlAs can coexist; this suggests that there is no thermodynamic driving force for further reaction if, instead, FeAl is deposited on AlAs. It suggests that hyperabrupt interfaces of the intermetallic and AlAs could be obtained.

There are a number of important issues that need to be addressed if the technology of intermetallic/III-V interfaces is to be fully developed. The two main ones involve issues of strain and growth kinetics. In the case of strain, one expects that if dislocations are formed or if lattice planes deform, there will be a transition in the growth from smooth layer-by-layer to rough three dimensional growth. The latter will cause interfaces to be diffuse, playing the same role as interdiffusion processes. For our work we have examined FeAl on AlAs and InP substrates, with lattice mismatches of 2.9% and -0.9% , respectively. The results were that the quality of the growth was dramatically superior for the more lattice matched case, suggesting that further reduction in strain could aid the nucleation and growth process. Growth kinetics have only begun to be addressed, with some work comparing migration enhanced epitaxy (MEE) to molecular beam epitaxy (MBE) to metal-organic MBE (MOMBE). In general the level of experience in depositing these materials is still quite limited and an increased fundamental understanding is crucial.

3 Current Status and Progress

Remarkable progress [1, 3] has been made in preparing ultrathin intermetallic films on III-V's: a variety of materials have been explored, semiconductor–intermetallic–semiconductor (SMS) structures have been fabricated, some device characteristics have been measured, the character of the dislocations have been determined in a few systems, strain relaxation has begun to be studied, *in situ* growth monitors have been developed. Probably the most striking result the quality of the SMS structure that can be grown and illustrated in Fig. 1. No pure-metal/semiconductor combination has achieved such a high quality result. To my mind these structures are proof of concept. What is needed now is to understand the growth so that optimal kinetics and materials can be chosen and so that the results can be generalized to other semiconductor systems.

This is an expanding area with at least two groups in Japan, two groups in the U.S. and two groups in Europe now pursuing the growth of these films. Each of these groups is following different paths for growth of high quality films, depending upon the material system and their particular experience. For all groups, the usual difficulty of machine dependent parameters complicates direct transfer of growth conditions. What is needed is a further understanding of the fundamental growth processes so that we can modify materials and structures without having to repeat each others' tortuous path to attain the highest quality material.

Under our current grant we have addressed three main issues: (1) the growth of FeAl by MOMBE, (2) defect characterization and strain relaxation, and (3) material characterization. These were parallel efforts designed to improve the quality of the metal aluminide films. The MOMBE effort was begun to allow us to more closely match the surface mobilities of Fe and Al adsorbed on the surface. The study of defects and strain would allow us to design strategies to either preserve tetragonal distortion or to eliminate strain, as desired. The characterization efforts involved continued use of RHEED to study the growth mechanisms and a major effort to build a custom STM to give us the capability to study 3D growth modes and to obtain atomically resolved images of the surface structure. Each of these efforts is described in turn.

3.1 Metal-Organic MBE of FeAl

We have just succeeded in growing the first FeAl by MOMBE. This means that we have achieved a much larger change in the growth kinetics than possible by variation of MBE growth parameters alone. This should allow the growth of FeAl at substrate temperatures for which the diffusion of both Fe and Al are more nearly matched. A RHEED pattern from the first FeAl film grown by this method is shown in Fig. 3. In the future we intend to use Fe carbonyl to be fully in the CBE mode. The main

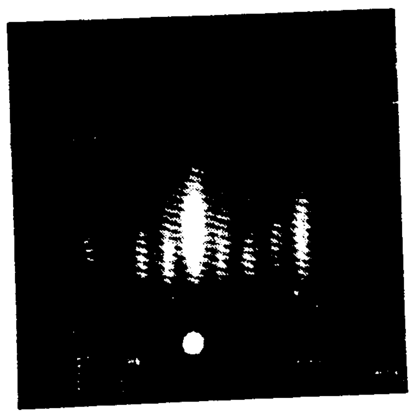

Figure 3: RHEED pattern from the first FeAl grown on GaAs(100) using DMEAA for the Al source at 250°C.

difficulty with MOMBE (elemental Fe and DMEAA) is that hot Fe tends to crack the DMEAA (Al source) in the gas phase. Currently we are examining the kinetics to see if atomic layer epitaxy will prove effective.

The advantages of this technique are that (1) we expect to be able to eliminate multiple domains that form in the growth of AlAs over FeAl by increasing the mobility of Al and (2) we will be able to take advantage of chemistry to eliminate point defects in the metal aluminide by ALE. The main difficulty to overcome has been to design a precursor that decomposes and does not leave carbon (that would form a carbide, for example) and which are reasonably convenient to work with in terms of their vapor pressure. We have collaborated with W.L. Gladfelter and A. Franciosi to design a chemical beam epitaxy (CBE) apparatus for the growth of these films. The initial work has been conducted in a custom, UHV facility that we jointly use as part of the NSF engineering research center at Minnesota. Prof. Gladfelter provided us with the technology to use dimethylethylamine alane (DMEAA) as the Al precursor. This is an adduct molecule – the Al is only weakly attached and when the molecule decomposes, no C bonds are broken. The result has been the first growth of AlAs and the first growth of FeAl using DMEAA. The latter is the first growth of FeAl by MOMBE.

3.2 Defects and Strain Relaxation

We have grown pseudomorphic films of FeAl on GaAs and InP(100) substrates by MBE. Using RHEED we have measured the lattice relaxation similar to that observed on III-V's – a Matthews- Blakeslee like variation in lattice constant as the film thickness increased. The surprise was that in every case, the measured relaxation occurred sooner than expected. The key issue was to determine the mechanism for the incorporation of dislocations, to understand how to inhibit dislocation formation, and how to drive the dislocations to the sample perimeter. We have made progress on the first of these, but the latter two remain.

The ultrathin films required for high speed electronic devices should be pseudomorphic – having the same structure as the substrate (and overlayer if any) without dislocations. For majority carrier devices, this requirement is relaxed but not eliminated. To create films with anisotropic magnetic properties, one would like to create films with as few dislocations as possible, to maximize the tetragonal distortion in the film. To understand and characterize the strain in the films the lattice parameter was measured during growth and then after growth the dislocations were studied by TEM and selected area electron channelling patterns (SAECP).

Fig. 4 shows the lattice parameter of an FeAl film grown on a GaAs(100) substrate. The open symbols are the RHEED data and the solid curve is a calculation based on the classic Matthews-Blakeslee (MB) theory. The MB result is equivalent to assuming either force balance or an array of dislocations somehow forming at the interface. In the calculation we assumed known elastic moduli, which in reality is a substantial source of error. Two measurements are shown for each point. The unannealed point corresponds to the initial growth at a given thickness. The annealed is the lattice parameter measurement after heating to 550°C. There are two important features. First, the lack of relaxation upon annealing serves to illustrate the stability of the films. Second, the MB relaxation occurs more slowly than expected and the initial critical thickness is very sensitive to the assumed elastic parameters. This is

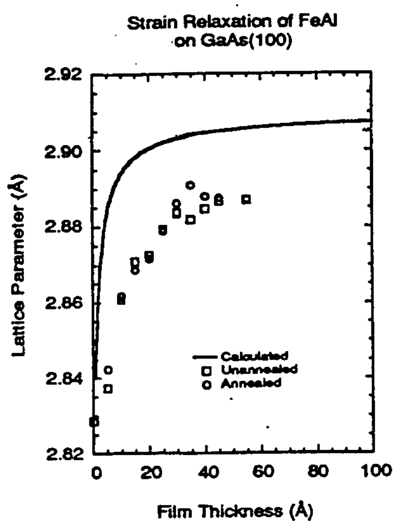

Figure 4: Lattice parameter of FeAl grown on GaAs(100) as a function of film thickness.

the first measurement of lattice relaxation of these films.

To understand the detailed mechanism of the defect generation and to compare to the RHEED measurements of these slight changes in lattice parameter, TEM and SAECP measurements were made on the same films. These were in collaboration with W.W. Gerberich, J.E. Angelo, and R.R. Keller. An important result was that the SAECP measurements of the thin film strain confirmed the RHEED measurement of surface lattice parameter. I should note that this is perhaps one of the most important questions currently being argued in understanding the relaxation mechanisms of thin epitaxial films. We found that the Burgers vector of the dislocations was $a[100]$. Considering the slip system of FeAl and GaAs, this means that the dislocations were not due to threading dislocations from the substrate and that dislocations nucleated at the surface did not glide down to the interface to relieve the strain. We need to choose between dislocations climbing to the interface, dislocations forming at 3D island perimeter, or determine which other slip system is operating. The problem is that RHEED measurements indicate that the films grow layer-by-layer, so that 3D island formation was not observed. Hence we are forced to conclude that dislocations come in by climb or slip via a secondary system. This relaxation mechanism needs to be elucidated. It determines the strategies to be followed to maintain full tetragonal distortion or obtain complete relief.

3.3 Material Characterization

Though only three components, FeAl/AlAs is still quite a complicated system. FeAl appears to admit dislocations into the interface by slip (cf. Sec. 4.2) or, less likely (as is claimed for CoAl [2]), at the edges of flat, 3D islands that form during growth. FeAl has a CsCl structure that admits a variety of order and disorders – in addition to grain boundaries and stacking faults, as Fe is added to the lattice, Al is displaced, until at Fe_3Al a bulk superlattice is formed. In the Al planes of this structure, every second Al is replaced by an Fe atom. We know that Fe_3Al is antiferromagnetic and when grown shows striking changes in the time evolution of the RHEED intensity. We suspect that the kinetics of strain relaxation will change as the structure changes from FeAl to Fe_3Al. But, because of the complexity of the growth and our still modest understanding of RHEED methods, with diffraction alone we are unable to conclusively follow the growth and formation of these films. What is needed is *in situ* imaging capability, such as could be provided by scanning tunnelling microscopy (STM).

To examine spatially inhomogenous features, growth by 3D islands, and the structure of FeAl surfaces, we have constructed a UHV scanning tunnelling microscope to attach to our MBE apparatus. A photograph of the STM that we have constructed is shown in Fig. 5. The design of an STM is complicated when attaching to an MBE due to the severe vibration environment. We followed the successful MBE/STM design of Biegelsen at Xerox and incorporated a double spring system with long springs. The internal mechanism is a modification of the successful STM used by Lagally's group at Wisconsin. Currently this STM is being attached to the MBE, with vibration coupling still impeding atomic resolution. Our goal is to transfer an FeAl sample from the MBE growth chamber to the STM for imaging. With the image we will be able to understand the RHEED pattern more clearly, allowing us to interpret the subtle changes observed during growth at and growth temperatures.

In the CBE chamber used to grow the FeAl using DMEAA, we have a commercial STM that is being used in a similar fashion. It has just achieved atomic resolution and application to FeAl is proceeding. Fig. 6 shows an image of a GaAs(311) surface prepared in UHV merely by annealing in an As flux. This was chosen as a test structure for the STM since we expected a simple, larger scale structural variation. As expected, following work of Ploog [4] we find an anisotropic, microscopic ripple along the $\langle 0\bar{1}1 \rangle$ direction. The is the first STM image of this structure; the surprise is that the ripple was seen to depend on growth conditions and were different than those shown by Ploog. One question now is whether we can deposit FeAl into these channels. The STM will be used to characterize the success of the effort. It could also be used to make contact, allowing measurement of the IV characteristic of

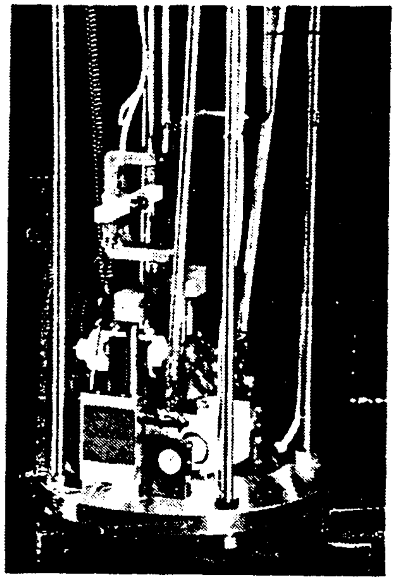

Figure 5: Photograph of custom STM to be attached to our MBE system. For clarity only one of the two sets of vibration isolation spring supports is shown.

metallic, quantum wires.

3.4 Quantum Well Structures

We have grown AlAs/FeAl/AlAs/GaAs structures and characterized the resulting quantum well with RHEED and cross-sectional and plan view TEM. Fig. 1 shows the cross-sectional TEM of one such quantum well. The interface is exceedingly abrupt; there is excellent epitaxy; the contrast variations are due to strain; there is a tetragonal distortion of 1.2% , and lattice fringes are observed in both films. The top layer exhibits roughness that we believe is due to the low mobility of Al at the growth temperatures. One of the goals of the MOMBE program is to improve on the abruptness of this upper interface so that a high quality superlattice could be grown. The evidence is that the H that is present during the growth of AlAs using DMEAA induces islands to break up and aids the mobility of the Al adatoms.

A second difficulty is that we need to improve the nucleation process. Though it is clear from the Fig. 1 that high quality interfaces and films can be obtained, it is not yet possible to do this over a very large area. We believe that there is some rotational disorder, about the normal that weakens the RHEED pattern from the

Figure 6: STM image of GaAs(311) surface in which the long channels are in the $\langle 0\bar{1}1 \rangle$ direction. We will attempt to deposit FeAl into these channels to fabricate ordered arrays of quantum wires of controllable separation and length.

overlayer in comparison to that from the substrate. In the proposed work we will grow on vicinal surfaces to eliminate this nucleation uncertainty while maintaining the excellent epitaxy.

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5 Publications and Presentations

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1. "Epitaxy of FeAl Films on GaAs(100) by Molecular Beam Epitaxy," (with J.N. Kuznia and A.M. Wowchak), J. Elect. Mat. v19 (1990) 561.

2. "A Study of FeAl/AlAs/GaAs interfaces using Moiré-fringe contrast in a transmission electron microscope," (with J.E. Angelo, J.N. Kuznia, A.M. Wowchak, and W.W. Gerberich), in "Evolution of Thin Film and Surface Microstructure," C.V. Thompson et al., eds., Mater. Res. Soc., 585 (1991).

3. "The identification of the misfit dislocations at an FeAl/AlAs/GaAs interface using Moiré-fringe contrast in a TEM," (with J.E. Angelo, J.N. Kuznia, A.M. Wowchak, and W.W. Gerberich), Appl. Phys. Lett., v59, 63 (1991).

4. "Electron Channelling analysis of strained iron aluminide films," (with R.R. Keller, J.E. Angelo, A.M. Wowchak, and W. W. Gerberich),in "Advances in Surface and Thin Film Diffraction." T.C. Huang, P.I. Cohen, and D.J. Eaglesham, eds., Mater. Res. Soc., 205 (1991).

5. "Growth and Characterization of Iron Aluminide Films on Compound Semiconductors," (with R.R. Keller, A.M. Wowchak, J.E. Angelo, J.N. Kuznia, and W.W. Gerberich) J. Elect. Materials, v20 (1991) 319.

6. "Strains and relaxations near metal aluminide/semiconductor interfaces," (with W.W. Gerberich, J.E. Angelo, R.R. Keller, and A.M. Wowchak), proc. of the Symposium on Mechanical Behavior of Materials and Structures in Microelectronics, E. Suhir et al. eds., Mater. Res. Soc., (1991), 279.

7. "Heteroepitaxial growth of AlAs using dimethylethylamine alane as an Al precursor," (with K.M. Chen, T. Castro, A. Franciosi, and W.L. Gladfelter), Appl. Phys. Lett., v60, 2132 (1992).

Presentations

1. "Layer by layer growth of FeAl lattice matched to InP(100)," (with J.N. Kuznia and A.M. Wowchak) Phys. and Chem. of Semicon. Interfaces, Bozeman (1989).
2. "Nucleation and Growth of FeAl on InP(100)," (with A.M. Wowchak and J.N. Kuznia) Electronic Materials Conference, Boston, June, 1989.
3. "Layer by layer growth of NiAl on GaAs," (with W.-H. Liu and A.M. Wowchak), Electronic Materials Conference, Santa Barbara, June, 1990.
4. "Channelling Analysis of Strained Metal Aluminide Films," (with R. Keller, A.M. Wowchak, and W.W. Gerberich) presented at the Fall Symposium of the MRS, Boston, 1990.
5. "Characterization of Strained Epitaxial Films by Electron Channelling," (with R.R. Keller, W.W. Gerberich, and A.M. Wowchak), TMS, Detroit, 1990
6. "Growth and Characterization of Iron Aluminide Fiolms on compound semiconductors," Annual Meeting of the Minerals, metals,a nd Materials Society, Anaheim, 1990. (invited) (W.W. Gerberich)
7. "Dynamics of MBE Growth: in situ studies of the role of defects,", Gordon Conf, New Hampshire, July 1991. (invited)

6 Personnel and Collaborations

Faculty

1. P.I. cohen is the principal investigator leading the project
2. W.W. Gerberich (Mat Sci Dept) led the microscopy effort and assisted in understanding the role of defects, unfunded.
3. W. Gladfelter (Chemistry dept) developed DMEAA for the metal organic work, unfunded.

Research Associates

1. H.D. He led a portion of the effort on the MBE growth of the metal aluminides
2. K.M. Chen ed the effort on metal organics

Graduate Students

1. A.M. Wowchak developed the growth of intermetallics and received a Ph.D. in EE in Dec. 1990. "The growth of Ultrathin Intermetallic films by MBE"
2. J.N. Kuznia developed the measurement of strain relaxatioon and received an M.S. EE in Dec. 1989 "The nucleation of Iron and Iron Alumindes on Compound Semiconductors."
3. J.E. Angelo did the electron microscopy and received a Ph.D. in Mat Sci, "Studies of dislocation structure at metal-semiconductor interfaces,", 1990.
4. R.R. Keller did the electron channelling and received a Ph.D. in Mat. Sci. "Electron channeling analysis of local strain distributions associated with interfaces"
5. Feng Wang is a graduate student in EE and is extending the work to superlattices.