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Reflector/Interconnect Applications in InGaAs Thermophotovoltaic Devices

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Growth and Properties of InGaAs/FeAl/InAlAs/InP Heterostructures for Buried Reflector/Interconnect Applications in InGaAs Thermophotovoltaic Devices

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Abstract

Thermophotovoltaic cells consisting of InGaAs active layers are of extreme promise for high efficiency, low bandgap TPV conversion. In the monolithic interconnected module configuration, the presence of the InGaAs lateral conduction layer (LCL) necessary for the series connection between TPV cells results in undesirable free carrier absorption, causing a tradeoff between series resistance and optical absorption losses in the infrared. A potential alternative is to replace the LCL with an epitaxial metal layer that would provide a low-resistance interconnect while not suffering from free carrier absorption. The internal metal layer would also serve as an efficient, panchromatic back surface reflector, providing the additional advantage of increased effective optical thickness of the InGaAs cell. In this paper, we present the first results on the growth and development of buried epitaxial metal layers for TPV applications. High quality, single crystal, epitaxial $\text{Fe}_x\text{Al}_{1-x}$ layers were grown on InAlAs/InP substrates, having compositions in the range $x = 0.40-0.80$. Epitaxial metal layers up to $1000\text{\AA}$ in thickness were achieved, with excellent uniformity over large areas and atomically smooth surfaces. X-ray diffraction studies indicate that all FeAl layers are strained with respect to the substrate, for the entire composition range studied and for all thicknesses. The FeAl layers exhibit excellent resistance characteristics, with resistivities from $60\mu\text{ohm-cm}$ to $100\mu\text{ohm-cm}$, indicating that interface scattering has a negligible effect on lateral conductivity. Reflectance measurements show that the FeAl thickness must be at least $1000\text{\AA}$ to achieve $>90\%$ reflection in the infrared.

1. Introduction

Thermophotovoltaic (TPV) cells have attracted great interest in recent years due to their ability to convert low energy radiation from thermal sources into useful electricity. TPV cells operate essentially as low energy (low bandgap) solar cells, and share many of the same performance tradeoff issues that characterize photovoltaic cells with regard to carrier collection efficiency, absorption and conversion efficiency. In addition, TPV cells are particularly susceptible to free carrier absorption (FCA) due to the intense infrared energy content of typical TPV thermal sources. In the monolithic interconnected module (MIM) configuration, FCA has been dealt with to a very large degree by the use of semi-insulating InP substrates on which low bandgap InGaAs TPV cells are grown.¹ The semi-insulating nature of the substrates mitigates FCA losses within the substrate; and, by coating the back surface of the substrate with Au, provides an efficient reflector of the infrared photons back to the heat source for recuperation. The MIM structure with a Au back surface reflector has already demonstrated outstanding TPV performance, which is now well documented.² One of the important issues remaining to be addressed is optimization of the lateral conduction layer (LCL), which is needed to provide the series interconnection between adjacent TPV cells. The typical LCL consists of heavily n-doped InGaAs, and its thickness must be such that resistance losses are minimized. Hence, the use of relatively thick (tenths of microns or more) n⁺ InGaAs as the LCL can lead to significant FCA. The ideal LCL would be one in which ultra-low resistance is possible and infrared radiation is either completely transmitted through to the backside

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of the wafer or is reflected back to the heat source by the LCL directly, with negligible absorption. With the latter in mind, we have been investigating buried epitaxial metal layers as possible LCL's for future MIM TPV cells.

In this paper, we describe an initial investigation of $\text{Fe}_x\text{Al}_{1-x}$ layers grown epitaxially onto InP substrates and their suitability for eventual integration within InGaAs TPV cell structures. The use of such a metal layer may potentially demonstrate several advantages: (1) ultra-low resistance for the purpose of interconnections, (2) very high reflectivity throughout the infrared to reduce or eliminate absorption, and (3) very high reflectivity for above-bandgap photons which provides an effectively increased optical pathlength for the TPV cell. Hence, an epitaxial metal interconnect/reflector layer incorporated into a TPV cell above the substrate would theoretically behave as a near-ideal interconnect while having the added advantage of broad band reflectivity. Of course, there are some significant materials issues that must be overcome for this to become reality, as InGaAs/FeAl/InP-type structures represent a challenging integration of dissimilar materials. However, the notion of developing epitaxial semiconductor/metal heterostructures is not new, and has been explored recently for applications ranging from metal base transistors to magnetic devices.³ In general, the great potential of epitaxial semiconductor-metal heterostructures as the basis for new classes of devices has motivated significant efforts to identify the most promising metal compounds for pairing either with GaAs or InP due to the dominance of these semiconductors in optoelectronic and high speed device technologies.³ For InP (and also GaAs), the transition metal-group III alloys have been identified as particularly promising for the fabrication of buried metal layer structures. This is due to the thermodynamic stability of these compounds with InP and GaAs; the fact that these compounds have dominant cubic phases; and, since the lattice parameter of many transition metal-group III compounds are approximately half that of InP (and GaAs), the potential to achieve lattice matching at the metal/semiconductor interfaces ($2a_{\text{metal}} \sim a_{\text{semiconductor}}$). Specifically, the compound $\text{Fe}_x\text{Al}_{1-x}$ has been investigated for pairing with InP since the $\text{Fe}_x\text{Al}_{1-x}$ lattice constant is slightly less than half that of InP (for $x = 0.5$), resulting in a total misfit between $2a_{\text{FeAl}}$ and a_{InP} of 0.9%.³⁻⁶ However, in contrast to early work which focussed on the problem of achieving smooth, continuous, *thin* ($< 100\text{\AA}$) layers for applications such as metal base transistors, TPV cells will require much thicker (up to $1000\text{\AA}$ or more) layers to satisfy the requisite conductivity and reflectance properties. Here, we consider these issues and present the first report of $\text{Fe}_x\text{Al}_{1-x}$ epitaxial metal development for TPV applications.

2. Experimental details

$\text{Fe}_x\text{Al}_{1-x}$ /III-V heterostructures were grown using a multi-chamber MBE cluster, comprised of separate III-V MBE and electron-beam evaporation chambers connected by ultra high vacuum (UHV) transfer tubes. Hence, combinations of $\text{Fe}_x\text{Al}_{1-x}$ and III-V layers could be grown while maintaining a pristine surface for high quality heteroepitaxy. III-V MBE growth was performed in standard fashion using a modified Varian GEN II MBE chamber using conventional solid sources. $\text{Fe}_x\text{Al}_{1-x}$ deposition was achieved by co-evaporation using dual e-beam evaporators. Iron and aluminum fluxes were independently controlled by an EIES controller/monitor so that the FeAl composition could be precisely tuned and uniformity maintained in each run. A composition range of $x = 0.40 - 0.80$ was investigated so that a range of $\text{Fe}_x\text{Al}_{1-x}$ lattice constants could be explored for suitability with InGaAs/InP cell structures. Total deposition rates were adjusted to be within 0.8 to $1.0 \text{ \AA/sec}$. $\text{Fe}_x\text{Al}_{1-x}$ layers were grown to thicknesses between $200\text{\AA}$ and $1000\text{\AA}$, sufficient for evaluation of both optical reflectance and lateral conductivity. Based on earlier work, a range of substrate temperatures from 100° to 300°C for $\text{Fe}_x\text{Al}_{1-x}$ epitaxy was investigated, with best results achieved for a 200°C substrate temperature, consistent with earlier reports.⁶ A III-V buffer layer was grown on the (001) InP substrates in the III-V growth chamber prior to UHV transfer into the metallization chamber for $\text{Fe}_x\text{Al}_{1-x}$ epitaxy to provide a high quality surface for $\text{Fe}_x\text{Al}_{1-x}$ nucleation. For this purpose, $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ or $\text{Al}_{0.52}\text{In}_{0.48}\text{As}$ buffer layers lattice-matched to the underlying InP substrates were grown. Both were successful in providing a good surface for FeAl

epitaxy, but AlInAs was chosen as standard since it was anticipated that the presence of Al (instead of Ga) on the surface prior to $\text{Fe}_x\text{Al}_{1-x}$ deposition may encourage better $\text{Fe}_x\text{Al}_{1-x}$ nucleation. Hence, the nominal structure investigated was FeAl/AlInAs/InP. Reflection high energy electron diffraction (RHEED) was used extensively to monitor the quality of the growing films at various stages of growth, and Auger electron spectroscopy (AES) was performed *in-situ* on selected samples by transferring the wafer into an attached UHV analysis chamber equipped with AES and sputtering capabilities. *Ex-situ* measurements such as double crystal x-ray diffraction (DCXRD), triple axis XRD, 4-point probe resistivity measurements, and reflectance measurements were used to provide possible correlations between growth parameters and final film properties. For the remainder of the paper, we will refer to $\text{Fe}_x\text{Al}_{1-x}$ as FeAl.

3. Results and Discussion

3.1. Epitaxial growth of FeAl on InP substrates

Figure 1 shows a representative series of RHEED patterns obtained on the surface of a typical AlInAs buffer immediately prior to FeAl deposition, and after FeAl deposition as a function of layer thickness for a single FeAl composition ($x = 0.45$). The substrate temperature in each case was 200°C. As seen in the figure, the AlInAs surface displays the expected two-fold reconstruction pattern. However, significant differences are seen after various stages of FeAl deposition. In general, FeAl exhibits dominant diffraction streaks with a much wider spacing than the AlInAs diffraction streaks, consistent with an FeAl lattice constant that is approximately half that of AlInAs. Occasionally we observe strong four-fold reconstruction on the FeAl surface, and this is seen clearly in the 500Å and 1000Å thick FeAl RHEED patterns in Figure 1. There has been very limited information published on the surface structure of FeAl layers to date, and this information is vital for successful III-V epitaxy on the FeAl surfaces. Nevertheless, the fact that all RHEED patterns display streaky features with only minimal degradation as the FeAl layer thickness is increased is indicative of high quality single crystal epitaxy. The RHEED results were further substantiated by AFM measurements from which we derived an RMS roughness of 3Å to 4Å for various FeAl layers, featureless Nomarski micrographs, and featureless SEM micrographs with no evidence of pinholes which were present and problematic in earlier reports.

One issue that did arise in evaluating growth properties of the FeAl layers came from Auger analysis performed *in-situ* in our attached UHV analysis chamber. Figure 2 shows an Auger spectrum of the surface of a 500Å FeAl layer grown on AlInAs/InP. A large indium peak is observed, likely due to the AlInAs buffer layer. To determine whether the indium was distributed within the FeAl layer or was "surface-riding" as a result of initial segregation on the FeAl growth front, Auger depth profiling was performed. Figure 2b shows that after a 30 second argon ion sputter to remove the first few monolayers of FeAl, the indium peak is no longer detected. This indicates that indium likely segregates to the FeAl surface initially and that subsequent growth of FeAl blocks any further solid-state diffusion after the initial surface segregation. Since it is not known what effect the presence of indium would have on subsequent InGaAs heteroepitaxy on the FeAl surface, we investigated methods to block indium from entering the FeAl layer. Figure 3 shows Auger spectra of an as-grown FeAl surface with and without the presence of a 10 monolayer AlAs barrier layer grown on the AlInAs prior to FeAl deposition.⁶ As seen, the In:Fe peak ratio is reduced by a factor of ~5 compared with the FeAl surface without an AlAs barrier. The indium peak was reduced to within the background noise by incorporating a 20 monolayer (ML) thick AlAs layer and also by reducing the FeAl growth temperature to less than 100°C. However, unlike the 10 ML AlAs layer, both of the latter approaches resulted in poor RHEED patterns and poor surface crystallinity. Hence, a 10 ML thick, strained AlAs layer was incorporated into the FeAl structures (FeAl/AlAs/AlInAs/InP) for subsequent studies.

3.2. Bulk crystalline properties of epitaxial FeAl/AlInAs/InP

High-resolution double crystal x-ray diffraction (DCXRD) measurements were used to assess the crystalline quality of the bulk FeAl layers and to examine how the lattice-mismatch between FeAl and AlInAs/InP varies with nominal changes in composition. Figure 4 shows a DCXRD measurement of a 1000Å thick $\text{Fe}_{0.75}\text{Al}_{0.25}$ layer grown on AlInAs/InP. Note that the FeAl peak position corresponds to a (002) reflection as compared with the (004) reflection for the substrate, due to the higher periodicity of the FeAl lattice constant. The presence of Pendellosung oscillations and the extremely narrow full width, half maximum (FWHM) of the FeAl diffraction peak are indicative of the excellent crystalline quality of the FeAl layer. The diffraction spectrum was simulated assuming a metal lattice constant of half that of InP, and, as seen in the figure, an excellent match is obtained for both peak height and width, indicating that high quality epitaxy of FeAl has been achieved. We obtained similar results for the entire composition range studied, and Figure 5 shows a collection of DCXRD scans for several FeAl compositions. Note that the FeAl peaks are broadened with respect to the scan in Figure 4 since in Figure 5 the FeAl layers were only 500Å thick.

An important issue from the viewpoint of the completed TPV cell structure is the FeAl lattice constant. For the anticipated mismatch of $\sim 0.9\%$ between the FeAl ($x=0.50$) layer and the underlying substrate, we expected the FeAl layers to be at least partially relaxed due to the small critical thickness expected for this amount of misfit strain. However, triple axis XRD measurements indicated that the layers were completely strained, even at a thickness of 1000Å. To show what effect this has on the calculated perpendicular (out of the growth plane) lattice constant ($2a_{\text{FeAl}}$), Figure 6 summarizes the perpendicular FeAl lattice constant variation with FeAl composition assuming two cases, fully strained and fully relaxed. Also shown for comparison is the lattice constant of InP. The lattice constant, after correction for the tetragonal distortion associated with the tensile strain within the FeAl layer, is seen to depend slightly on iron content and is close to the reported value for bulk FeAl.³⁻⁵

3.3. Electrical and optical properties of epitaxial FeAl layers

For TPV applications, the FeAl layer must simultaneously demonstrate excellent conductivity and reflectivity, while maintaining high crystalline quality to support device-quality InGaAs TPV cell overgrowth. To date, there have been no reports on the effect of either FeAl composition or layer thickness on either of these critical properties for device applications, and it may be expected that both will have large impacts on these properties. Resistivity measurements were performed on a number of FeAl/AlAs/InAlAs/InP samples by four point probe measurements, where the Fe/Al ratio ranged from 0.4 to 0.8 and the FeAl layer thickness was varied from 200Å to 1000Å. Figure 7 shows the resistivity results as a function of FeAl layer thickness. As seen, the resistivity of all FeAl layers were in the range of $\sim 50\mu\text{ohm-cm}$ to $100\mu\text{ohm-cm}$ with no clear dependence on layer thickness, indicating that the lateral conductivity of the FeAl epilayers is not limited by interface scattering. This is further substantiated by the close agreement between these measurements and the resistivity values reported for bulk FeAl crystals ($80\mu\text{ohm-cm}$ to $90\mu\text{ohm-cm}$ at room temperature).⁷ Note that the resistivity of the FeAl is orders of magnitude lower than that of heavily doped InGaAs, demonstrating that FeAl layers have the potential of providing improved electrical interconnects.

It is of interest to determine how the FeAl resistivity may depend on composition, since adjusting the Fe:Al ratio allows some tuning of the lattice constant for improved lattice matching. As seen in Figure 8, there is only a slight dependence of FeAl layer resistivity on composition, with values increasing from $\sim 60\mu\text{ohm-cm}$ to $\sim 100\mu\text{ohm-cm}$ as the iron mole fraction is increased from 0.45 to 0.75. Such a trend is consistent with an expected increase in resistance for iron-rich layers. This weak dependence is clearly advantageous from the viewpoint of optimizing the lattice match with the

surrounding semiconductor layers since the lattice constant, which does depend on composition, can be essentially adjusted independently of the effect on resistivity. Thus, from the viewpoint of electrical properties, FeAl layers exhibit considerable promise for improved interconnects.

However, for TPV applications, the total reflectance of the FeAl layers is also critical. Figure 9 shows a comparison of reflectance data measured from the surface of bare $\text{Fe}_{0.50}\text{Al}_{0.50}$ layers having thicknesses of 200Å, 500Å and 1000Å, grown on AlAs/InAlAs/InP. As may be expected for very thin films, a significant improvement in reflectance is observed as the FeAl layer thickness is increased from 200Å to 1000Å, with no additional improvement beyond 1000Å in thickness. Reflectance values were found to maximize in excess of 90% for wavelength between 4μm and 20μm. However, all films exhibited a loss of reflectance at shorter wavelengths with the reflectance decreasing continuously with decreasing wavelength below 4μm. We attribute the reduced reflectance at shorter wavelengths to absorption by the iron transition metal component in the FeAl layers.⁸ This absorption can be mitigated by use of a front surface filter. These experimentally observed results are consistent with predictions of the optical performance modeled from band structure calculations for FeAl.^{8,9}

3.4. InGaAs overgrowth on FeAl/AlAs/AlInAs/InP: preliminary findings

Once a degree of confidence and reproducibility was established for the FeAl epitaxy, a series of samples were transferred back into the III-V growth chamber, after FeAl epitaxy, for InGaAs overgrowth. Since the FeAl lattice is strained with respect to the InP substrate, the in-plane lattice constant of the FeAl layer will be exactly matched to 0.74 eV bandgap InGaAs overgrowth. Initial overgrowth exhibited very spotty RHEED patterns, indicative of polycrystalline growth. It was determined that the ability to nucleate reasonable quality InGaAs on the FeAl surface depended strongly on the presence of arsenic. It was found that by reducing the arsenic exposure on the FeAl surface, streaky RHEED patterns could be obtained during nucleation. The reason for this is not yet clear and more work must be done, especially from the viewpoint of FeAl surface stability and reconstruction in the presence of a III-V flux at elevated growth temperatures. Some preliminary work did indicate that the FeAl surface stability improved significantly for higher iron compositions. Figure 10 shows a SIMS profile of an InGaAs/FeAl/AlAs/AlInAs/InP structure from which a few observations can be made. First, the AlAs layer forms a barrier to indium outdiffusion, as surmised by the Auger results discussed earlier. Second, the critical interface between the InGaAs and FeAl is quite abrupt, with insignificant iron and aluminum presence in the InGaAs overlayer. Interestingly, there is evidence for iron diffusion into the AlInAs buffer layer, although without SIMS standards it is not clear how significant this is or what effect this has on the overall structure.

4. Conclusions

This paper presents the first experimental report of buried epitaxial metal layers on InP for potential applications in TPV devices. Based on its thermodynamic stability with InP and a favorable lattice constant, FeAl compounds were explored to investigate their potential as buried interconnect/reflector layers in InGaAs/InP TPV structures. FeAl epitaxy was successfully achieved up to 1000Å in thickness on AlInAs/InP and AlAs/AlInAs/InP substrates. RHEED and DCXRD indicate high crystalline quality for compositions of $x = 0.40 - 0.80$. From triple axis diffraction measurements, the FeAl layers were fully strained with respect to the InP substrate. AFM and SEM measurements indicated that the FeAl surfaces had monolayer smoothness, enhancing the chances for good quality InGaAs heteroepitaxy on these layers. Auger indicated that indium from the AlInAs buffer layer rides the surface of the FeAl growth front, and by inserting an AlAs layer prior to FeAl epitaxy, the presence of indium is dramatically reduced. The FeAl layers were found to exhibit outstanding electrical properties, with resistivities in the 50μohm-cm to 100μohm-cm range. A flat optical reflectance spectrum in excess of

90% was achieved from 4 μ m to 20 μ m at only 1000Å thickness. This, combined with the resistivity results, implies that a 1000Å thick FeAl layer would be optimum for eventual TPV applications. Unfortunately, the presence of iron in the FeAl layer was explained to be responsible for a roll off of reflectance below 4 μ m. To mitigate this effect, a front surface filter could be employed. Initial InGaAs epitaxy on the FeAl/AlAs/AlInAs/InP structures was successfully demonstrated, and abrupt interfaces were achieved. These findings provide an excellent starting point for continued optimization of InGaAs overgrowth on epitaxial metal layers.

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AllnAs
surface

200 Å FeAl

500 Å
FeAl

1000 Å
FeAl

Figure 1. Series of RHEED patterns obtained for A) an AllnAs buffer surface, B) the same after 200 Å of FeAl growth, C) after 500 Å of FeAl growth and D) after 1000 Å of FeAl growth. The composition of these layers was $x = 0.45$.

as-grown FeAl surface after air exposure

Fig. 1(a) No Sputtering

After 30 sec. sputter etch

Fig. 1(b) After 30 sec Sputtering

Figure 2. Auger electron spectra for A) the FeAl surface showing a large indium peak (the oxygen peak is due to ambient exposure prior to obtaining AES measurements on this sample and B) after a 30 second argon ion sputter etch, which removes the indium signal.

Figure 3. In-situ Auger Electron spectra of the FeAl surface A) without an ALAs barrier layer and B) with a 10 monolayer ALAs barrier layer grown immediately prior to FeAl deposition. The indium:iron Auger signal ratio is reduced by a factor of five by this barrier layer.

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Figure 4. Double crystal x-ray diffraction measurements and simulation of 1000Å thick Fe_{0.75}Al_{0.25} layer grown on AlInAs/InP

500Å FeAl/(In,Al)As/InP:
Lattice constant vs. stoichiometry

Figure 5. Series of double crystal x-ray diffraction scans of 500Å thick FeAl layers for the compositions indicated. The broad appearance of the FeAl peak is due to the thinness of the layers.

Figure 6. Summary of the perpendicular lattice constant for FeAl ($2a_0$) as a function of layer composition. The InP lattice constant and the bulk FeAl value from Reference 5 are shown for comparison.

Figure 7. FeAl measured resistivity as a function of FeAl layer thickness.

Figure 8. FeAl measured resistivity as a function of A) substrate temperature during growth and B) FeAl layer composition.

500 Å FeAl on AlInAs/InP

1000 Å FeAl on AlInAs/InP

Figure 9. Front and backside reflectances for FeAl/AlAs/AlInAs/InP structures with the metal thicknesses indicated.

Figure 10. SIMS depth profile of an InGaAs/FeAl/AlAs/AlInAs/InP structure, indicating very abrupt InGaAs/FeAl interfaces are achieved.