MBE Growth and Characterization of ZnTe and Nitrogen-doped ZnTe on GaAs(100) Substrates

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0 Abstract

In this paper, we investigate the interfacial, electrical, and optical characteristics of ZnTe and p-type ZnTe:N thin films grown on lattice-mismatched GaAs(100) substrates by molecular beam epitaxy (MBE) to determine the suitability of GaAs as a substrate for a proposed integrated 6.1Å III-V/II-VI semiconductor solar cell. The interface between GaAs and ZnTe was examined with transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM). In addition, the hole concentrations and mobilities of the samples were determined with magneto-transport measurements, and the band structure was determined by photoluminescence (PL) spectroscopy. Despite a high lattice mismatch, the TEM images revealed a low density of defects, indicating that high quality ZnTe can be grown on GaAs, and the magneto-transport and photoluminescence data are generally consistent with heavily-doped ZnTe. We conclude that GaAs can be used as a substrate for the 6.1Å lattice solar cell project, instead of the more costly, lattice-matched GaSb, and that the nitrogen plasma used in the MBE can effectively dope ZnTe to sufficiently high concentrations. Further work on the solar cell project can now continue, with the next immediate goal being the n-type doping of ZnTe with Al donors.

1 Introduction

While high-efficiency solar cells are very desirable for both space and terrestrial applications, the current photovoltaic market is dominated by single-junction Si-based cells which have a maximum theoretical efficiency of just 31% under 1 sun. [1] Multi-junction solar cells can cover a much larger range of the solar spectrum, and can thus convert a much higher percentage of the incoming solar energy. The current record for solar cell efficiency is held by a

triple-junction GaInP/GaInAs/Ge cell at 40.7% achievable energy conversion efficiency under 240 suns. [2]

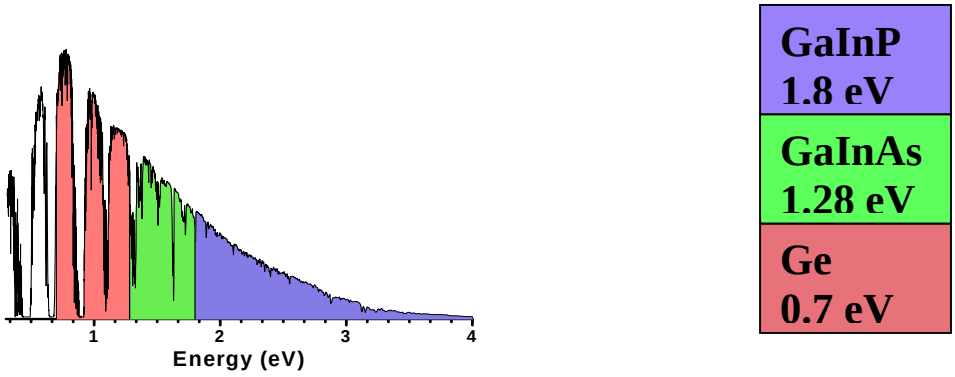

Figure 1: The energy spectrum of a 40.7% efficiency triple-junction solar cell.

For this cell, seen in Figure 1, the efficiency could be further increased by adding additional junctions with band gaps in the infrared part of the solar spectrum, but such an endeavor is complicated by the lack of appropriate lattice-matched materials. We propose that the ideal material system for a high- efficiency multi-junction solar cell is a series of monolithically integrated III-V and II-VI semiconductors with lattice constant around 6.1Å, as shown in Figure 2 below.

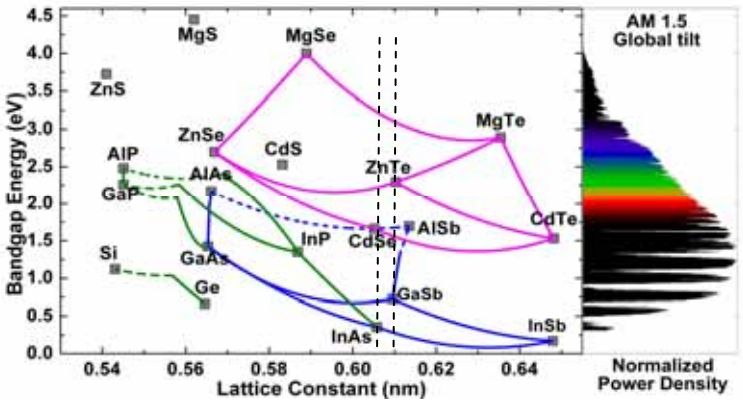

Figure 2: A comparison of band gap energy for the 6.1Å materials to the normalized solar spectrum.

Because there are more lattice-matched materials to use, integrated III-V and II-VI

semiconductor solar cells have the potential to have many junctions. Moreover, the 6.1Å lattice constant materials have band gap energies which are more optimized to the solar spectrum. The result is that a III-V/II-VI cell can be made far more efficient than current solar cell technology.

A four-junction model of such a solar cell, with an achievable energy conversion efficiency of 40% under 1 sun and 48% under 1000 suns, is currently being developed at the University of Notre Dame in collaboration with Arizona State University. The cell was originally designed to be grown on lattice-matched GaSb substrates, but preliminary results indicated that far cheaper, but lattice-mismatched GaAs might be a suitable alternative. This paper concerns the growth and characterization of ZnTe and p-type ZnTe:N on GaAs(100) substrates to investigate that hypothesis.

2 Sample Growth

The samples for characterization were grown in the University of Notre Dame dual-chamber molecular beam epitaxy (MBE) laboratory. Epilayers of ZnTe were deposited onto GaAs(100) substrates, followed by epilayers of ZnTe:N, which were doped p-type by means of a nitrogen plasma, in the proportions shown in Figure 3.

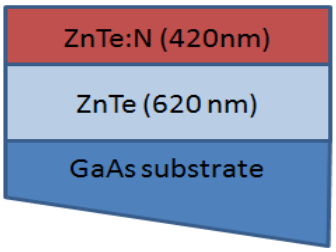

Figure 3: The dimensions of the GaAs/ZnTe/ZnTe:N samples

The flux of the nitrogen plasma during growth was varied to create a distribution of doping

concentrations in the samples.

3 Methods of Characterization

3.1 Transmission Electron Microscopy

To investigate the effects of the 8% lattice mismatch between GaAs and ZnTe, the interface between the two materials was imaged via transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) by the group at Arizona State University led by D.J.Smith. For TEM, beams of electrons are transmitted through an ultra-thin sheet of a material and focused, forming a magnified image of the sample being observed, much like in a conventional light microscope. Because the de Broglie wavelength of the electron is significantly smaller than the wavelength of visible light, the transmission electron microscope can resolve single atoms, providing a way to analyze the lattice structure and interfacial defects of the material directly.

3.2 Magneto-transport

The electrical characteristics of the samples -- in particular, the hole concentrations and mobilities of the ZnTe:N layers -- were determined with magneto-transport measurements, which are dependent on the Hall effect. Five gold wires were soldered to each sample in the configuration shown in Figure 4.

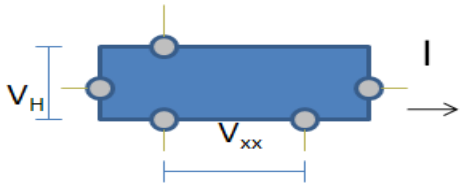

Figure 4: A sample with Ohmic contacts for magneto-transport measurements. The longitudinal and Hall voltages are marked.

The sample was mounted in a sample holder and wired into a current source and a series of

voltmeters. The sample assembly was then placed in a cryostat which allowed temperature control and application of an external magnetic field to the sample. A current was passed through the sample, and the magnetic field was swept from -0.75 T to 0.75 T. A voltage V_{xx} was applied across the longitudinal dimension of the sample (i.e., the direction of the current), and the motion of the charges due to V_{xx} in turn produced a voltage V_H along the transverse direction of the sample -- the phenomenon caused by the Lorentz force, known as the Hall effect. Using the voltmeters, we measured both V_{xx} and V_H, from which Ohm's Law yields the resistivities ρ_{xx} and ρ_H. These quantities allow us to calculate the hole concentration p and the hole mobility μ using equations (1) and (2) below:

$$\rho_{xx} \approx \frac{1}{pe\mu} \qquad (1)$$

$$\rho_H \approx \frac{1}{pe} B \qquad (2)$$

After determining the hole concentrations and mobilities at room temperature, the temperature of the cryostat was lowered, and these measurements were taken again at 250, 200, 150, 100, 50, and 25 K.

Photoluminescence

The optical characteristics of the samples -- and the band gap energy in particular -- were investigated by photoluminescence (PL) spectroscopy. For the PL measurements, light from a 45mW laser was directed onto the sample to excite electrons within the material from the valence band to the conduction band. The electrons are unstable in the conduction band states, and fall back to their equilibrium valence band state, releasing their excess energy by emitting photons equal to the energy difference between the bottom of the conduction band and the top of the

valence band. By detecting and analyzing the photons emitted from the sample, the band gap energy of the material can be determined.

4 Results,

4.1 TEM

The results of the transmission microscopy are shown in Figure 5 below.

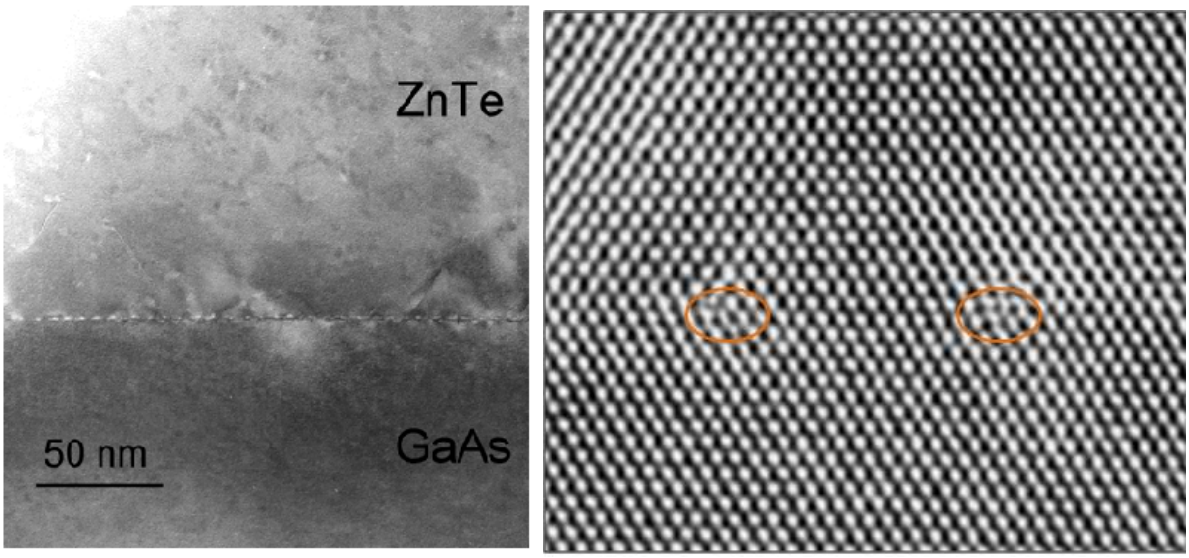

Figure 5: TEM images of the interface between GaAs and ZnTe

Despite the 8% lattice mismatch, the TEM images reveal a high-quality crystal growth with an unexpectedly low density of defects. In the lower resolution TEM, the defects which are present in the crystal appear in a nearly regular pattern -- an analysis of the HRTEM reveals that they occur as nucleation centers on the interface with a period of about 13 times the ZnTe lattice constant, or 14 times the GaAs lattice constant. The following calculation shows the effective reduction of the original 8% lattice mismatch: $13a_{ZnTe}$=79.348Å, $14a_{GaAs}$=79.146Å, resulting in a lattice mismatch of 0.255% (!) between the two crystalline layers.

Magneto-transport

The magneto-transport data for one sample are presented below.

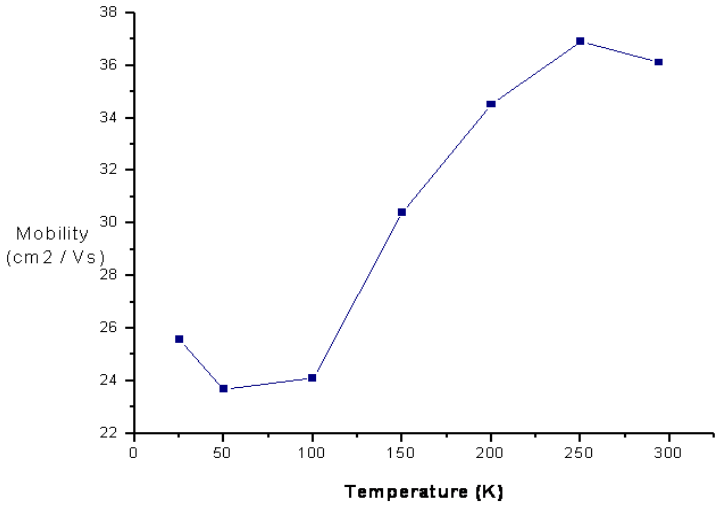

Figure 6: Hole mobility as a function of temperature

Hole mobility is related to scattering; as the amount of scattering within the lattice increases, the mean free path decreases, and the hole mobility similarly decreases. At low temperatures, the dominant form of scattering is ionized impurity scattering, which scales as $T^{-3/2}$. As the temperature increase, therefore, one would expect that the mobility would increase. The data shown in Figure 6 agree with this prediction.

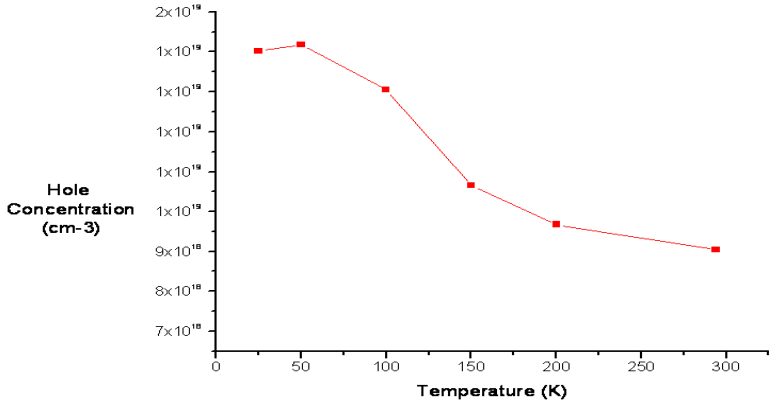

Figure 7: The hole concentration for one sample as a function of temperature

By contrast, the temperature dependence of the hole concentration, shown in Figure 7, does not agree with predictions. It takes very little energy for an electron to be trapped by an acceptor site, and thus as the temperature increases, more electrons will be thermally promoted to the impurity band. The exodus of electrons will leave more holes in its wake, and thus the hole concentration should increase with increasing temperature; however, the data suggest the opposite trend. The reason for this behavior is as yet a matter of speculation, and further investigation will be necessary to gain understanding of the physical process responsible for this unexpected behavior.

Photoluminescence

The photoluminescence data agrees well with prediction. The PL spectrum, seen in Figure 8, shows a maximum intensity at 2.32 eV, which is consistent with prior measurements of the ZnTe band gap energy.

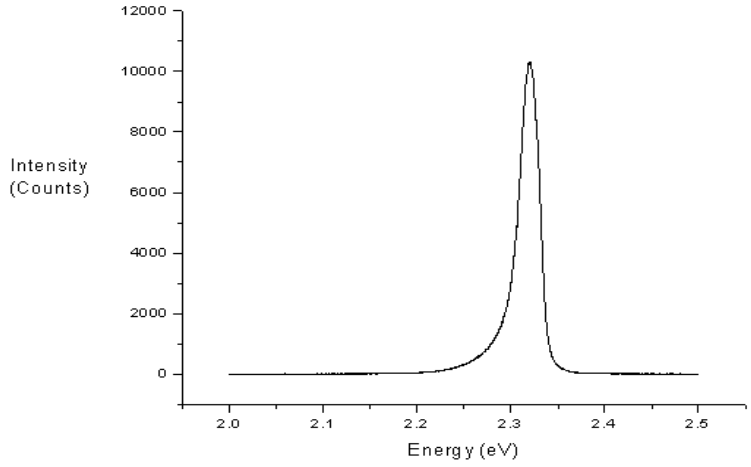

Figure 8: The photoluminescence spectrum for GaAs/ZnTe/ZnTe:N. The maximum intensity occurs at 2.32 eV.

The intensity of the ZnTe peaks is also rather high, with counts of over 100,000 in some samples. This is indicative of heavy doping, as interband transitions in intrinsic ZnTe are very difficult to detect via photoluminescence.

5 Conclusions

Although the full characterization of GaAs/ZnTe/ZnTe:N is not yet complete, the preliminary results are promising. Our results have demonstrated that – while a lattice mismatch of 8% should make high-quality growth of ZnTe on GaAs substrates infeasible – the 0.255% "effective lattice mismatch" existing between the two materials is small enough to make the use GaAs substrates a definite possibility. Moreover, the TEM imaging indicates that lattice relaxation occurs within a few monolayers of the interface, indicating that the rest of the ZnTe crystal has a very small density of defects. The use of GaAs as a substrate as opposed to GaSb would then very significantly reduce production costs of integrated multijunction solar cells, thus increasing the range of potential applications.

In addition to the question of substrates, the results for characterizing the doping in the ZnTe:N layer are also interesting. The photoluminescence and mobility data are suggestive of a heavy p-type doping, and thus an encouraging indication of the efficacy of the nitrogen plasma doping in the MBE. The hole concentration data, however, display a trend opposite to what would be expected for either an intrinsic or extrinsic semiconductor. While this last result merits further investigation, the rest of the data -- interfacial, electrical, structural, and optical -- supports the potential use of a GaAs/ZnTe junction in the proposed 6.1 Å III-V/II-VI solar cell project.

6 Future Work

Future work on this project will consist of two parts: further research on GaAs/ZnTe/ZnTe:N, and research for the next phase of solar cell design. With regards to the former, the focus will be on further magneto-transport data collection and electrical characterization. For the latter, the next materials development goals will be to achieve good n-type doping in ZnTe with Al donors, and the optimization of n- and p-type dopings in ternary II-VI materials.

7 Acknowledgments

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8 References

[1] W. Shockley and H. J. Queisser, JAP **32**, 510 (1961)
[2] R. R. King et al., Appl. Phys. Lett. **90**, 183516 (2007)