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InGaN Thin Films Grown by ENABLE and MBE Techniques on Silicon Substrates

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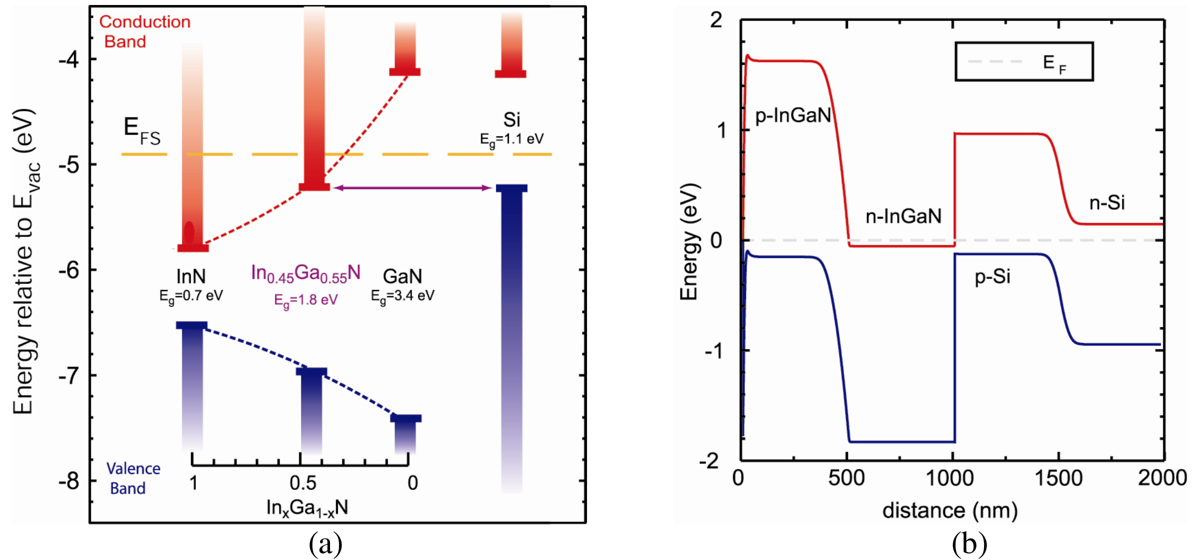
ABSTRACT

The prospect of developing electronic and optoelectronic devices, including solar cells, that utilize the wide range of energy gaps of InGaN has led to a considerable research interest in the electronic and optical properties of InN and In-rich nitride alloys. Recently, significant progress has been achieved in the growth and doping of InGaN over the entire composition range. In this paper we present structural, optical, and electrical characterization results from InGaN films grown on Si (111) wafers. The films were grown over a large composition range by both molecular beam epitaxy (MBE) and the newly developed “energetic neutral atomic-beam lithography & epitaxy” (ENABLE) techniques. ENABLE utilizes a collimated beam of ~2 eV nitrogen atoms as the active species which are reacted with thermally evaporated Ga and In metals. The technique provides a larger N atom flux compared to MBE and reduces the need for high substrate temperatures, making isothermal growth over the entire InGaN alloy composition range possible. Electrical characteristics of the junctions between n- and p-type InGaN films and n- and p-type Si substrates were measured and compared with theoretical predictions based on the band edge alignment between those two materials. The predicted existence of a low resistance tunnel junction between p-type Si and n-type InGaN was experimentally confirmed.

INTRODUCTION

The band gap of InGaN can be tuned from 0.65 eV to 3.4 eV [1]. This makes it a very attractive material for optoelectronic devices and, in particular, for solar cells, as this energy range is a perfect match to the part of the solar spectrum that reaches the surface of the Earth. InGaN is therefore a material of choice for a “full-spectrum-photovoltaic” development and there is a considerable research interest in the electronic and optical properties of InN and In-rich nitride alloys [2].

Growing InGaN on Si is interesting for various reasons. First of all, InGaN structures could be imbedded into the mature Si technology, and an obvious application in PV would be to improve the efficiency of a standard single junction Si solar cell by adding a matched InGaN top cell. The thermodynamic efficiency limit [3] of 29% for a Si solar cell under 1 sun standard illumination (AM1.5G) will increase to 42.5% already for only one additional cell with an optimum band gap of 1.8 eV [4]. To obtain this band gap, an InGaN film with a composition of 45% In is necessary.



(a) (b)

Figure 1. (a) Absolute (relative to vacuum) valence and conduction band edges of $In_xGa_{1-x}N$ and Si. E_{FS} marks the Fermi stabilization energy at -4.9 eV (from [5]). (b) Band diagrams for an $In_{0.45}Ga_{0.55}N$/Si tandem solar cell, assuming $2x10^{17}cm^{-3}$(p,n) in InGaN and $2x10^{16}cm^{-3}$(p,n) in Si.

Series-connected multijunction solar cells require an efficient tunnel junction to provide electron hole recombination between the sub cells. Such tunnel junctions are usually formed by adding additional layers of highly doped material. The InGaN/Si hybrid cell offers a unique advantage here as a consequence of the band alignment between InGaN and Si. Figure 1(a) shows the absolute positions of the band edges for InGaN and Si. It can be seen that the conduction band of InGaN aligns with the valence band of Si for a 45% In composition. Figure 1(b) shows the calculated band diagram of a pn- $In_{0.45}Ga_{0.55}N$ /pn-Si combination [5]. Doping and composition can be chosen such that there is no band bending between n-type InGaN and p-type Si, and thus an ohmic contact is predicted and no heavily doped additional layer is required. Growth of high quality InN and GaN epitaxial films on Si (111) substrates using molecular beam epitaxy (MBE) technique has been demonstrated [6,7].

EXPERIMENT

For this study we investigated InGaN on Si structures that were grown by the newly developed "energetic neutral atomic-beam lithography & epitaxy" (ENABLE) [8] and by gas-source molecular beam epitaxy (MBE) [9]. ENABLE utilizes a collimated beam of ~2 eV nitrogen atoms as the active species which are reacted with thermally evaporated Ga and In metals. The technique provides a larger N atom flux compared to MBE and reduces the need for high substrate temperatures making isothermal growth over the entire InGaN alloy composition range possible. For growth on Si (111) typical substrate temperatures were ~600 ^{o}C and growth rates were approximately 2 to 3 µm/hr. InN buffer layers were used to facilitate InGaN lattice matching. MBE growth of $In_xGa_{1-x}N$ films with x = 0 to x = 0.5 on Si (111) was initiated by removal of residual oxide. This was achieved by a sequence of heating and Ga exposure steps. The cleaning procedure was followed by growth of a 30 to 50 nm AlN buffer layer. The InGaN layers were deposited at 630 ^{o}C.

Structural characterization of each film was performed using Rutherford backscattering spectrometry (RBS), ion channeling, x-ray diffraction (XRD), and transmission electron microscopy (TEM). Optical characterization was done with photoluminescence (PL) measurements, and electrical properties were studied with Hall effect. Surface morphology and metallic cluster defects were studied by atomic force microscopy (AFM) and optical microscopy. Electrical characteristics of the junctions between InGaN films and n- or p-type Si substrates were determined by I-V measurements in the dark and with simulated AM1.5 illumination.

RESULTS AND DISCUSSION

Figure 2 shows a TEM image of an InN film which illustrates the capabilities of the new ENABLE method. This micrograph shows a columnar growth of the InN film. Column diameters are in the range of 500 nm to 1000 nm. Some voids are observed at the InN/GaN interface, which help in stress relaxation and result in much better structural quality of the top layer compared to the area close to the interface. The sample surface show slight corrugation, not exceeding 50 nm. RBS channeling analysis (with channeling minimum yield < 4%) confirms a good structural perfection of this film compared to standard MBE grown InN grown on the same substrate. Residual donor concentrations below $10^{19}/cm^3$ were measured and electron mobilities higher than 1000 cm^2/Vs were achieved. It was noted that the mobility values were generally higher than in MBE grown films with comparable electron concentrations.

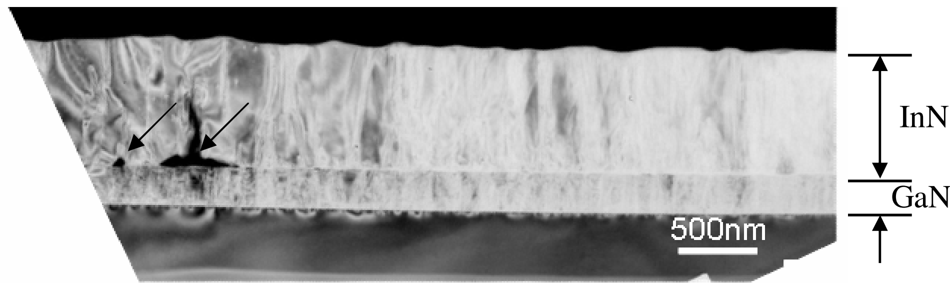


Figure 2. TEM image of an ENABLE grown InN film grown on a sapphire substrate with a GaN buffer layer. Arrows mark the formation of voids at the GaN/InN interface.

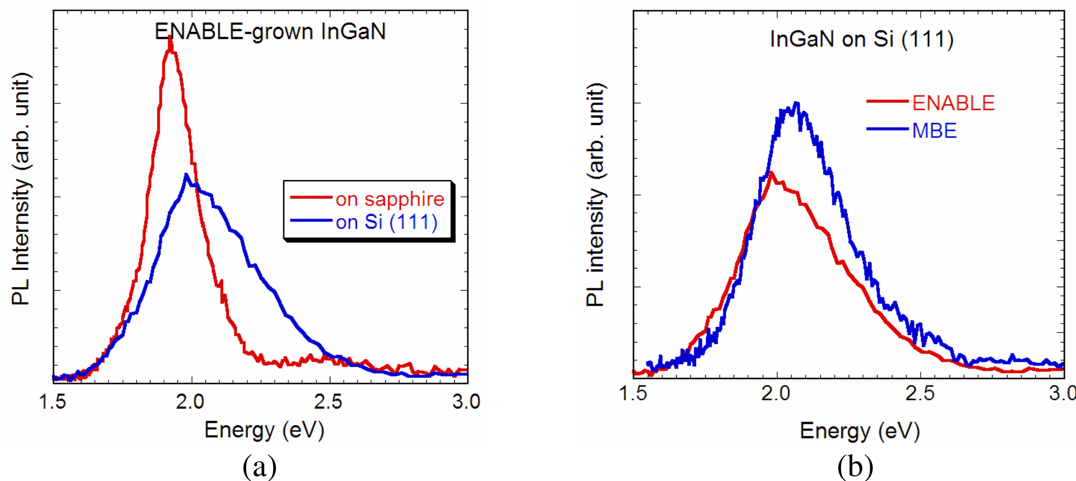


Figure 3. Room temperature PL spectra of ENABLE and MBE grown InGaN films.

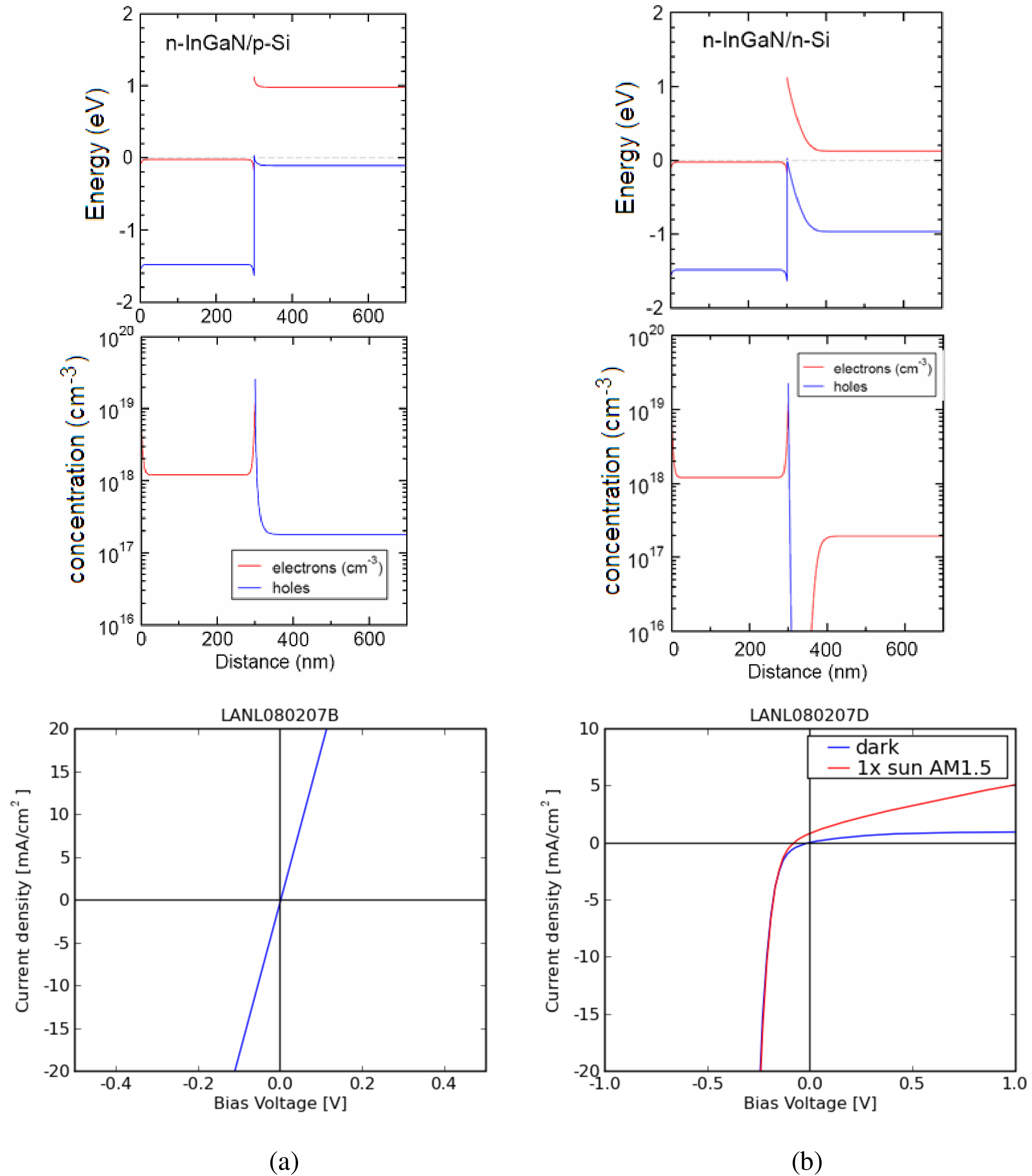


Figure 4. Calculated band diagrams, carrier distributions and measured I-V curves (top to bottom) of ENABLE grown InGaN/Si structures. In case (a), the n-type film was grown on p-type Si, (b) shows the data for the same film grown on n-type Si.

Strong band edge photoluminescence intensity is observed from InGaN films grown by ENABLE on sapphire and Si. Figure 3(a) shows PL spectra of ENABLE grown $In_{0.32}Ga_{0.68}N$ films on sapphire and (111) Si. Notice that the PL peak from the InGaN film grown on (111) Si is broader than from the film on sapphire. This suggests a better quality of the film on sapphire.

Figure 3(b) shows a comparison of the PL from InGaN films with similar composition grown by ENABLE and MBE on (111) Si. A very similar PL peak can be observed in both films.

Channeling-RBS and TEM investigations of the ENABLE-grown InGaN film reveal that the film is polycrystalline. This can be attributed to the direct growth of the film on Si without a AlN buffer layer. XRD measurement showed a single phase film with a (0001) preferred orientation. The polycrystalline nature of this film also results in a weaker and broader PL signal as well as a higher residual donor concentration of $6x10^{19}cm^{-}3$. The reduced film quality is also noticeable in the relatively large AFM surface roughness of 30 nm RMS. In case of the MBE samples, an AlN nucleation layer was first grown on the Si substrate and therefore an epitaxial InGaN film was obtained. MBE films typically showed an AFM roughness of less than 10 nm RMS.

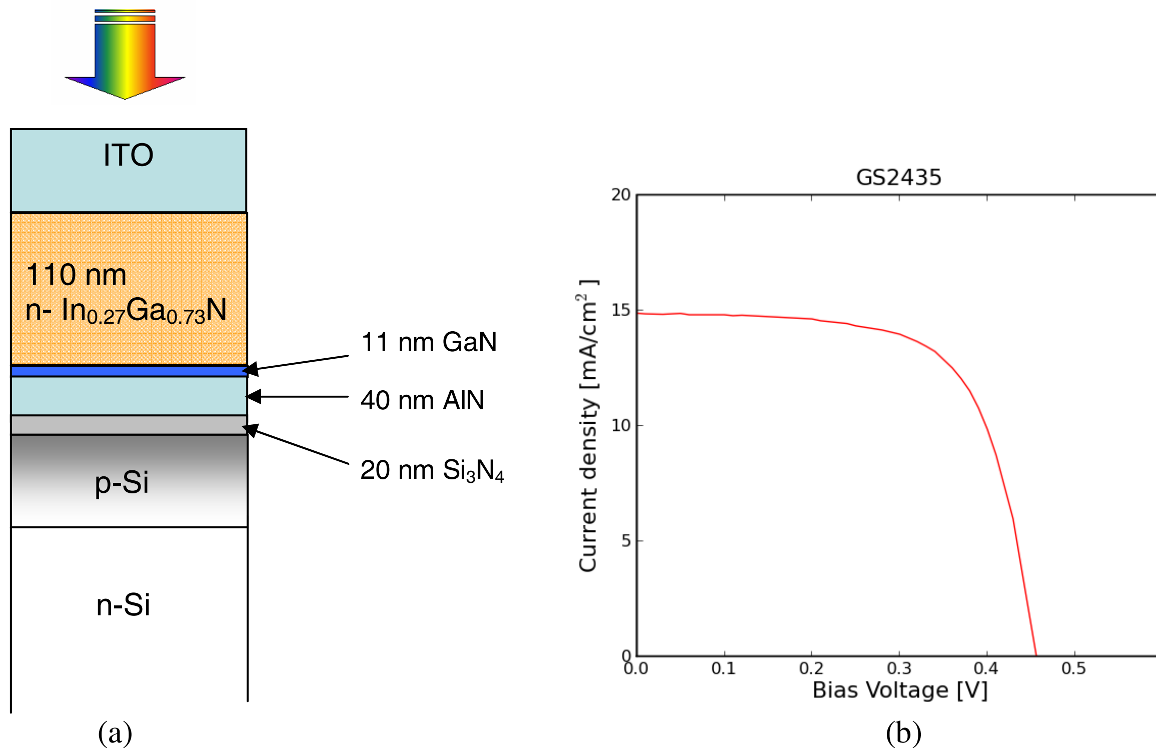


Figure 5. (a) MBE grown InGaN film on an n-type Si wafer with a diffused p-type top layer and (b) corresponding I-V curve under 1x sun (AM1.5) illumination.

In order to test the predicted electrical transport between InGaN and Si, undoped n-type InGaN of 1 μm thickness was simultaneously grown on p-type and n-type Si substrates. The results of the current-voltage tests are shown in figure 4. The InGaN/Si heterojunctions behave electrically as predicted by the calculated band diagrams in figure 4. On p-type Si, a linear I-V curve is measured (figure 4(a)), demonstrating the perfectly ohmic character of the tunnel junction. Figure 4(b) shows for comparison the same ENABLE film grown on n-type Si. The InGaN acts like a metal in this case because of the very low position of the conduction band and depletes the n-type Si. A Schottky type junction is predicted and a rectifying behavior is indeed observed. The light sensitivity to 1x sun illumination is rather small in this case. We attribute this to light absorption mostly outside the rather small and buried active region and to imperfect crystallinity.

Finally, as a first stage towards the realization of an InGaN/Si double junction tandem solar cell, we fabricated a n-InGaN/p-Si/n-Si test structure as shown in the schematics in figure 5(a). A 110 nm thick n-type InGaN layer was grown on a pn-Si-structure by MBE. This example demonstrates the effectiveness of the tunnel junction. Figure 5(b) shows the I-V characteristics of this structure under 1x sun AM1.5 illumination. Photocurrent is generated at the pn-junction in the Si and transported to the ITO top contact. PV properties of this structure are comparable to a reference pn-Si sample.

CONCLUSIONS AND FUTURE WORK

InGaN films with a wide range of In contents were grown on Si by ENABLE and MBE and their structural and optical properties compared. Junctions between n-type InGaN films and n- and p-type Si substrates behave as theoretically predicted based on the band edge alignment between those two materials. We have demonstrated the formation of an ohmic tunnel junction at the heterointerface between n-type InGaN and p-type Si. ENABLE growth will be improved in the near future with an additional AlN buffer layer. In order to complete an InGaN/Si hybrid solar cell, p-type doping of InGaN is required. We recently achieved MBE grown Mg doped p-InGaN films on Si. Mg doping for the ENABLE technique is currently under development. Both techniques are very promising for InGaN/Si photovoltaic.

ACKNOWLEDGMENTS

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