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December 7, 2005 – June 6, 2008

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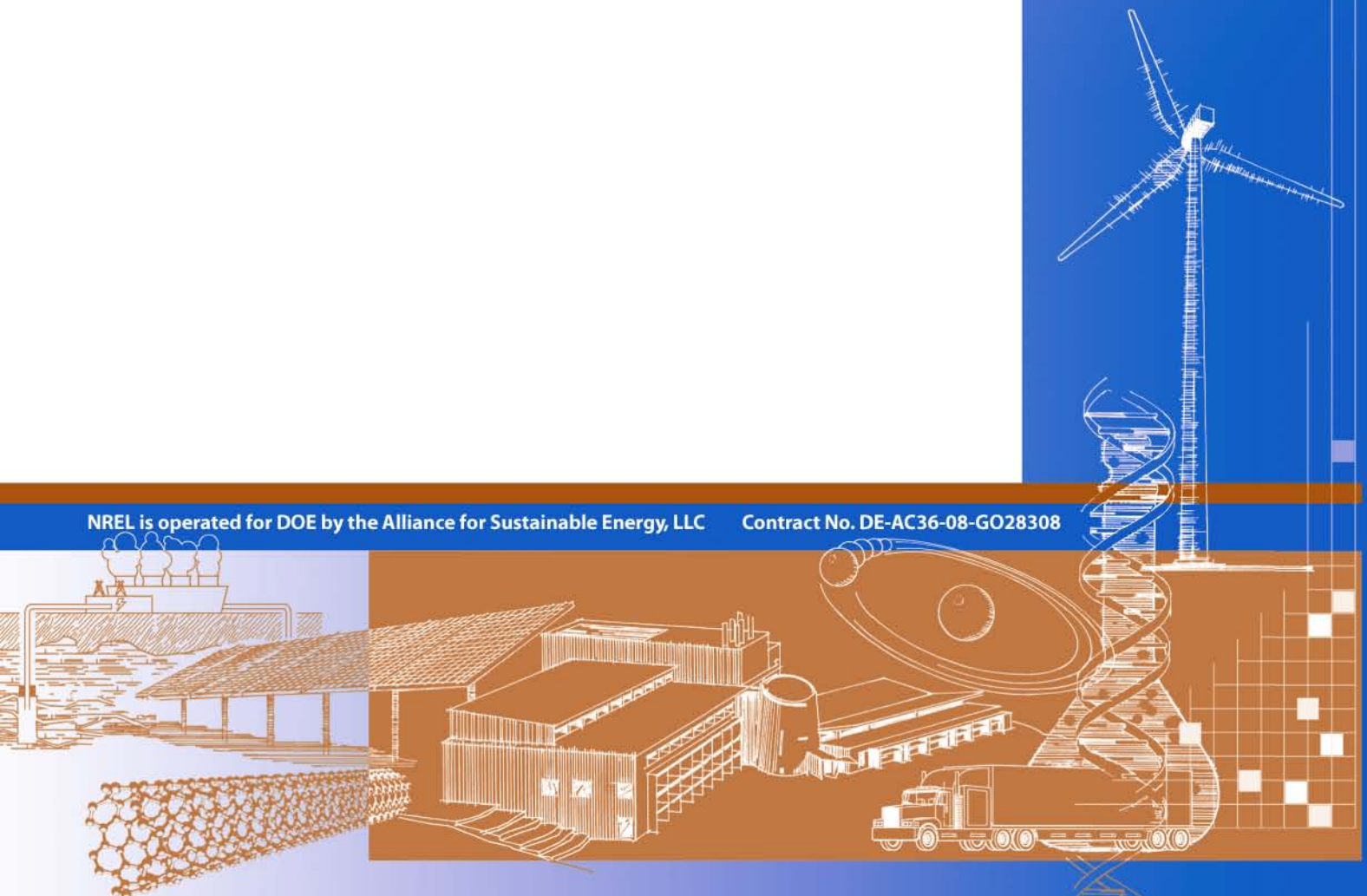

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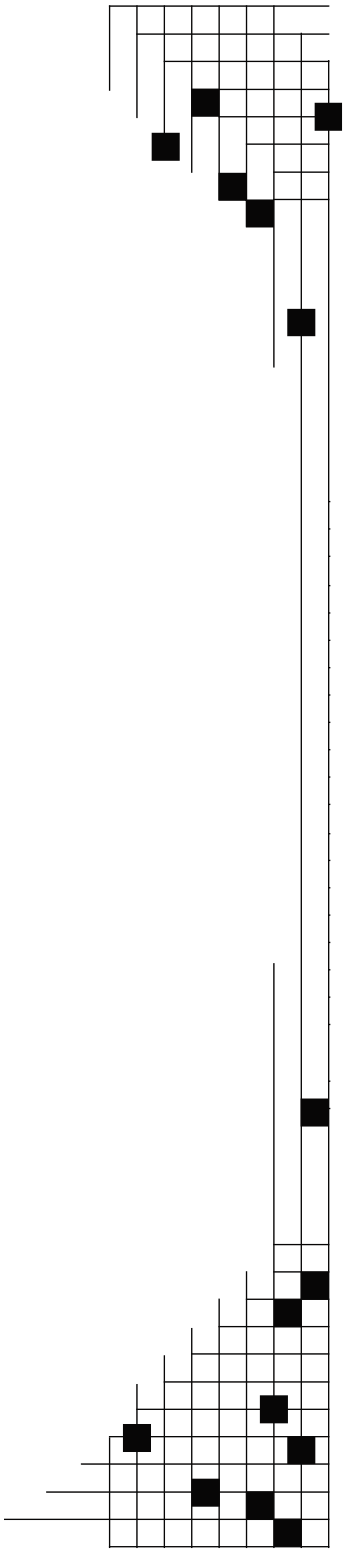

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Introduction

The initial motivation for this project was the then-newly discovered bandgap of InN, re-measured at ~0.7 eV, rather than the previous 1.9 eV [1-3]. This makes InGaN a potential material for solar cell with the possibility of absorbing 99% of the solar irradiance. The advantages of the InGaN material system are the wide range of direct bandgap, high absorption coefficient, a low effective mass (high mobility), and strong polarization effects [4– 6]. However, the challenges include material quality, defect density, doping, substrates and growth issues. The current project led to the understanding of the above challenges; in particular, the material quality in terms of phase separation was studied and suppressed. The polarization property was modeled and solar cell design was developed with these new models.

The following section describes the issue of phase separation and its suppression. We then present a brief description of polarization and its modeling. The effect of phase separation and polarization on device operation follows. The next section presents the solar cell design and results achieved.

Phase Separation and Its Suppression

A solid-phase miscibility gap exists in the InGaN alloy due to the large difference in the lattice constants between GaN and InN, which is also the probable cause of multiple phases and consequent multi-peak luminescence observed in the material [7-8]. The equilibrium solubility of InN in the bulk GaN is about 6% at typical growth temperatures used in metal-organic chemical vapor deposition (MOCVD). However, the situation in thin InGaN films epitaxially deposited on GaN virtual substrates is significantly different. Theoretical calculations [9] based on a valence-force-field (VFF) model [7-10] predict that phase separation in InGaN strongly depends not only on the temperature and In composition, but also, on the strain state of the InGaN films. Thus, one or more indium-rich phases come into existence in the InGaN alloy layers during growth in an attempt to reach thermodynamic equilibrium during growth, as shown in Figure .

Phase separation is usually identified as secondary peaks in addition to the primary peak corresponding to the bulk material during photoluminescence; higher degrees of phase separation are also identified via X-ray diffraction (XRD), shown in Figure .

In addition to acting as a recombination channel, it can be correlated from fabricated quantum-well solar cells that the lower-bandgap phase-separated material will also tend to pin down the open-circuit voltage (V_{OC}) of the solar cell.

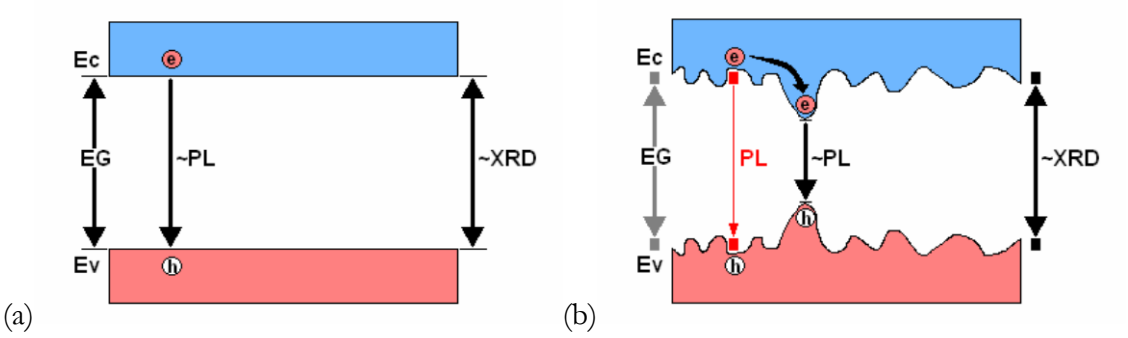


Figure 1: Schematic comparison of band structures of (a) an ideal material, and (b) a phase separated material.

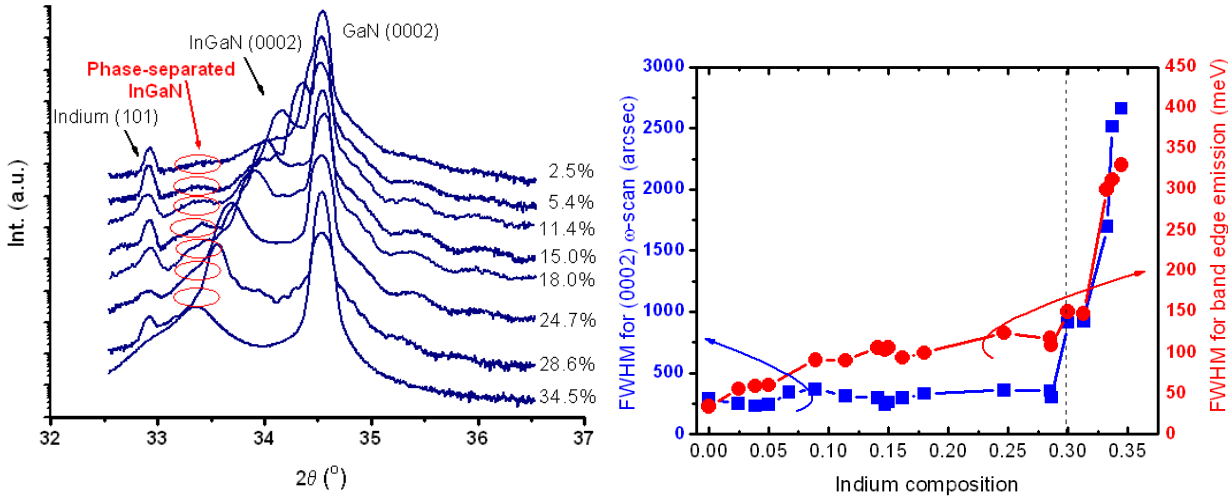


Figure 2: Summary of (a) photoluminescence (PL) and (b) PL and X-ray diffraction data for InGaN grown by MOCVD with indium composition ranging from 0% to 35%.

To minimize the effect of phase separation and increase the material quality of the grown layers, we examined and optimized the effect of TMIn flow, temperature, growth pressure, growth rate, and TEGa flow. The effect of TMIn flow is shown in Figure . Increasing the flow rate suppresses the secondary peak in PL signals, indicating that phase separation is suppressed.

The effect of growth temperature is shown in Figure . As the growth temperature is decreased, the surface morphology becomes rough and the growth mode changes from step growth to nucleation of discrete islands. However, the growth temperature, in conjunction with the TEGa flow rates, determines the growth rate, which also influences phase separation and the indium composition. As the TEGa flow rate increases, the full width at half maximum (FWHM) of XRD increases, indicating poorer material quality.

The final critical parameter in controlling phase separation is the thickness of the layer. The effect of thickness is indicated in Figure , which shows the appearance of a secondary phase of lower-bandgap InGaN with In ≈ 10% for thickness of 100 nm. Further optimization allows minimization of the phase separation for layers as thick as 200 nm.

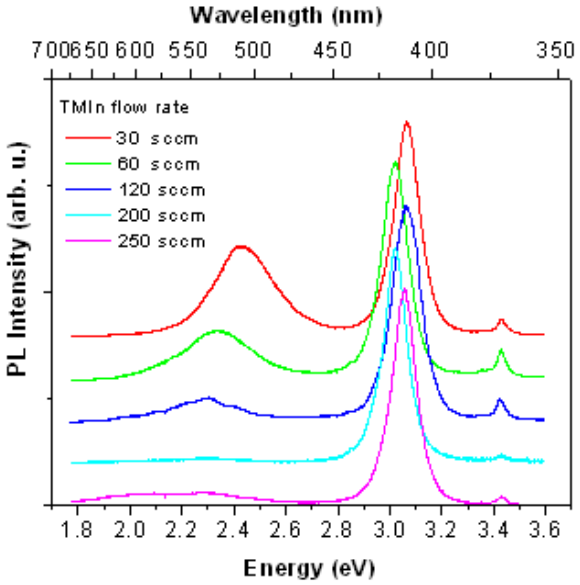


Figure 3: PL data summary of InGaN growth with variable TMIn flow rate.

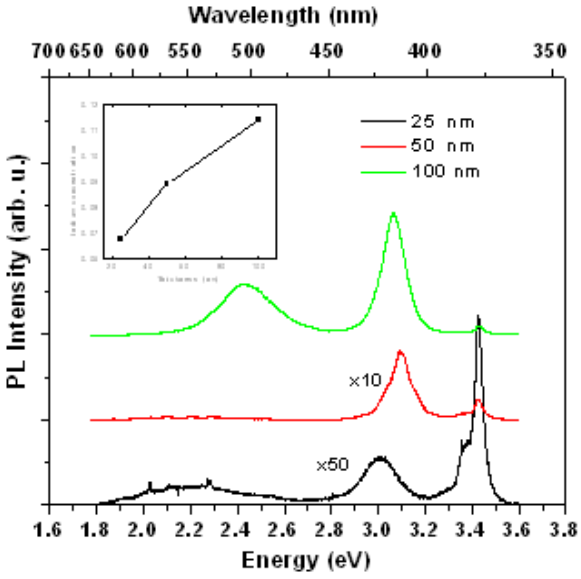


Figure 4: Photoluminescence data indicating increasing emission from phase-separated InGaN with increasing thickness.

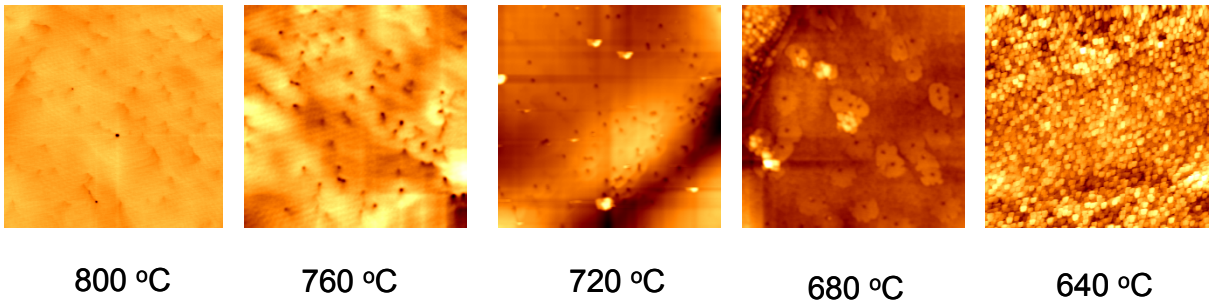


Figure 5: Effect of growth temperature on surface morphology, showing change of growth mode from step-like growth to nucleation of discrete islands.

Polarization Modeling

One of the unique characteristics about the wurtzite III-nitrides is its polarization effects. The net polarization, Spontaneous polarization ($\vec{P}_{sp}$) + Piezoelectric polarization ($\vec{P}_{pz}$), and consequent internal electric fields have been shown to be detrimental to the performance of optoelectronic devices [11-13]. Polarization discontinuities lead to potential barriers, band bending that can have undesired consequences on the device; electric fields with values as high as 1 MV/cm have been reported [14-15]. Polarization, however, can be used constructively by accommodating in the solar cell design to improve its performance. Thus, it becomes vital to successfully model this effect and incorporate it during device design.

Polarization is present in the III-nitrides as a consequence of the non-centrosymmetry of the wurtzite structure and the large ionicity of the covalent metal-nitrogen bonds. Spontaneous polarization ($\vec{P}_{sp}$) exists due to the non-centrosymmetry of the crystal, where the crystal behaves like an electric dipole. In the III-nitrides, the direction of spontaneous polarization is from the N-atom to the closest metal atom, i.e., along the $\pm$c direction, which is also the typical MOCVD or molecular beam epitaxy (MBE) growth direction. Hence, in Ga-face GaN, spontaneous polarization is opposite to the [0001] direction, i.e., in the downward direction pointing toward the substrate, as shown in Figure (a).

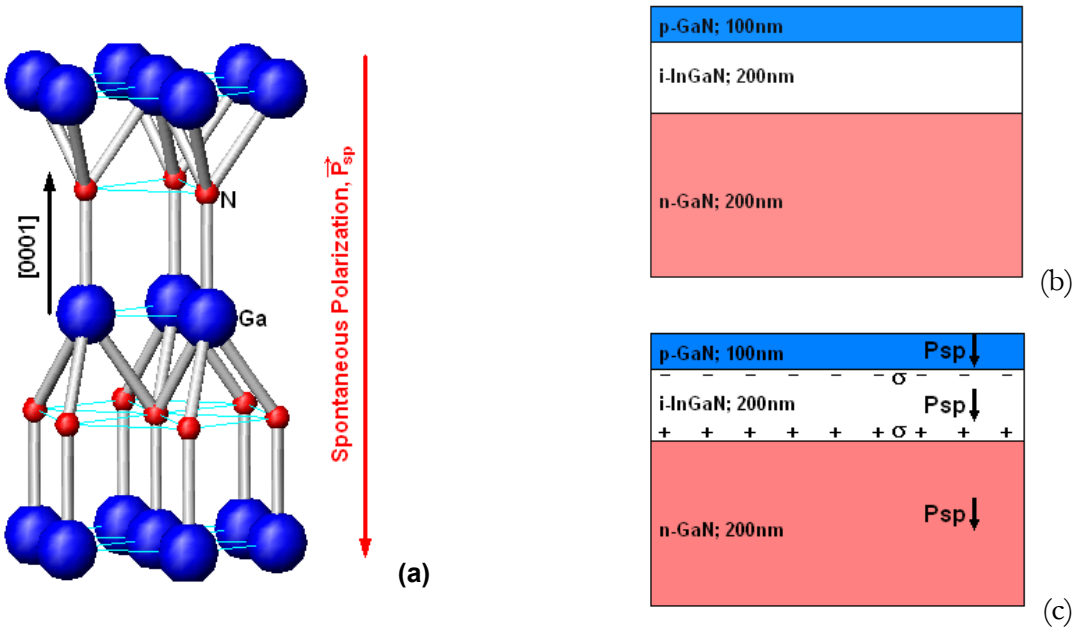


Figure 6: (a) Direction of spontaneous polarization jn Ga-face GaN and GaN/InGaN p-i-n test structure (b) without and (c) with spontaneous polarization effects.

Spontaneous polarization in ternary compounds can be calculated by interpolating the binary compounds using a bowing factor 'b' as shown in Equation 1.

$$P_{sp}^{ABN}(x) = P_{sp}^{AN}(x) + P_{sp}^{BN}(1-x) + bx(1-x) \qquad [\text{Units: C/m}^2] \tag{1}$$

The direction of piezoelectric polarization depends on the polarity of the material as well as on the strain, shown in Figure (b). Spontaneous and piezoelectric polarizations are parallel and add to each other when planar strain (perpendicular to c-axis) is tensile and are anti-parallel when planar strain is compressive.

The polarization-induced charge density with a gradient of polarization in space given by:

$$\sigma_P = -\nabla P \qquad [\text{Units: C/m}^2] \tag{2}$$

The polarization-induced charge density or electric field is used as the starting point for the modified PC1D solving routine at a given hetero-interface. If the charge density at a hetero-

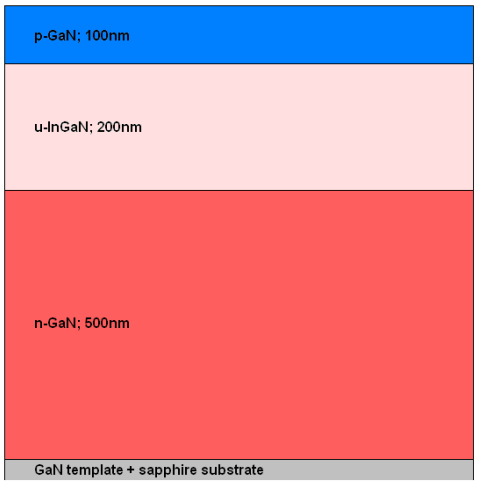
interface is positive, it will tend to accumulate a 2-Dimensional Electron Gas (2DEG); similarly, a negative hetero-interface charge density will tend to form a 2-Dimensional Hole Gas (2DHG).

The modified PC1D, which incorporates polarization, is used in the design of InGaN solar cells. The design of pn-junction solar cells starts with a p-i-n structure and evolves to the current status of using an n-GaN capping layer. Along with the design and fabrication of pn-junction solar cells, alternative approaches in designing solar cells are also considered. The pn junctions and alternative approaches are presented in the next section.

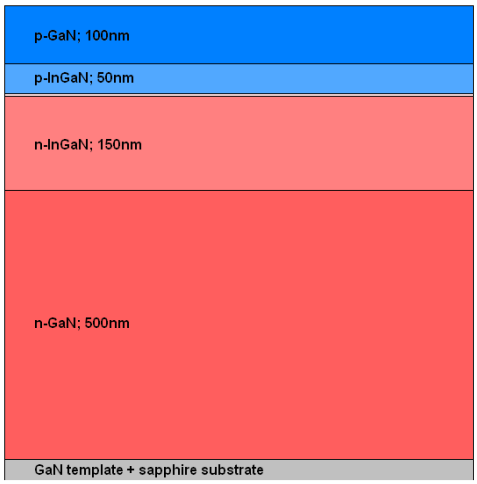
Device Design and Results

Two approaches have been attempted in the design of InGaN solar cells, pn-junction solar cells and Schottky-barrier solar cells. This section presents the evolution of InGaN pn-junction solar cell designs fabricated and tested, summarized in Figure , followed by results of Schottky-barrier solar cells.

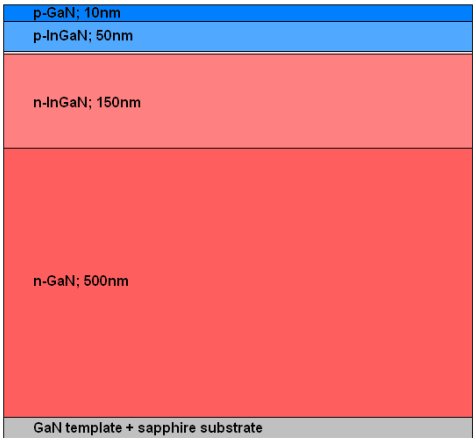
The solar cells are designed, grown by MOCVD/MBE, fabricated, characterized, and analyzed before designing the next generation of devices. The p-i-n device structure (Figure (a)) is taken as the starting point to design subsequent InGaN solar cells due to a detailed understanding obtained from preliminary experiments. The indium composition in the test devices is fixed at 12% (bandgap [E_g] – 2.9 eV) to ensure successful p-type and n-type doping. Here, the p- and n-GaN regions are fixed at 100-nm and 500-nm thicknesses, respectively, and doped at maximum concentrations without reaching a level of degeneracy. The thickness of the i-region is fixed at 200 nm.



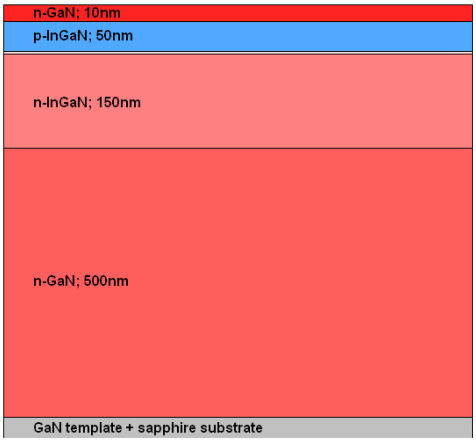
(a) Gen 1: p-i-n structure.



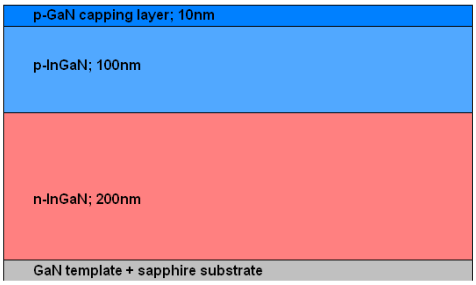
(b) Gen 2: i-InGaN replaced by p-n InGaN.



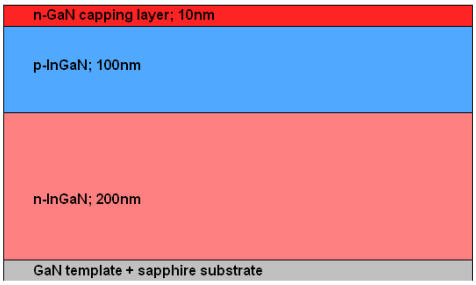
(c) Gen 3: Thickness of p-GaN reduced.



(d) Gen 4: p-GaN cap replaced by n-GaN.



(e) Gen 5: Thickness of InGaN increased.

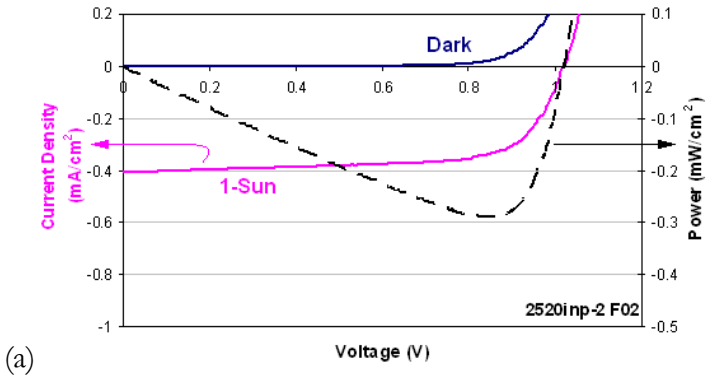


(f) Gen 6: Thickness of InGaN increased.

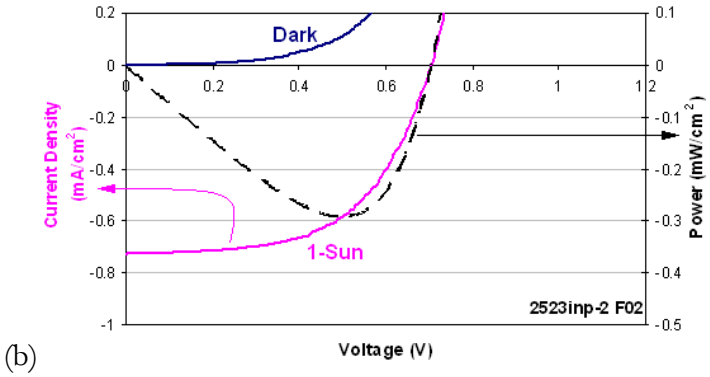
Figure 7: Generations of InGaN solar cell evolution.

As the i-InGaN region adds to the series resistance of the solar cell, this region is replaced by a p-n InGaN junction in second-generation solar cells as shown in Figure (b). In this case, the additional electric field generated by the InGaN p-n junction enhances the quantum efficiency (QE) of the solar cell. Moreover, the p-GaN layer not only acts as a window layer for the underlying InGaN p-n junction, but the GaN junctions also induce front-and back-surface fields for this device. The thicknesses of p- and n-InGaN are set to 50 nm and 150 nm, respectively, to maintain a net InGaN thickness of 200 nm.

The current-voltage (I-V) curves for the second-generation InGaN devices show a substantial improvement in short-circuit current of the solar cell compared to the first-generation devices, as shown in Figure (a) and (b). The increase in short-circuit current is supported by the increase in internal quantum efficiency of the second-generation devices up to 50%, as seen in Figure . However, there is a decrease in the open-circuit voltage due to reduction in quasi-Fermi-level energy difference caused by additional doping in the InGaN junctions and/or formation of phase-separated defects. The third-generation InGaN devices are designed to minimize the voltage loss caused by the p-GaN window layer due to its high resistivity. Hence, the thickness of the p-GaN window layer is reduced from 100 nm to 10 nm, as shown in Figure (c). However, the 50-nm p-InGaN layer is not thick enough to provide charge to the depletion region, which results in incomplete junction formation. Hence, these solar cells measure very low open-circuit voltages and quantum efficiencies.



(a)



(b)

Figure 8: Sample I-V characteristics of (a) first-generation, and (b) second generation test solar cells.

The fourth-generation devices are designed to enhance the window layer by replacing the 10-nm p-GaN with n-GaN. The strained window layer forms a 2DEG at the n-GaN/p-InGaN interface, as explained in the previous section, thus improving the lateral conduction in the material, whereas the n-GaN window enhances tunneling to reduce the top-contact resistance of the device. However, these devices also fail to provide substantial open-circuit voltage and quantum efficiency due to incomplete charge provision by the thin p-junctions.

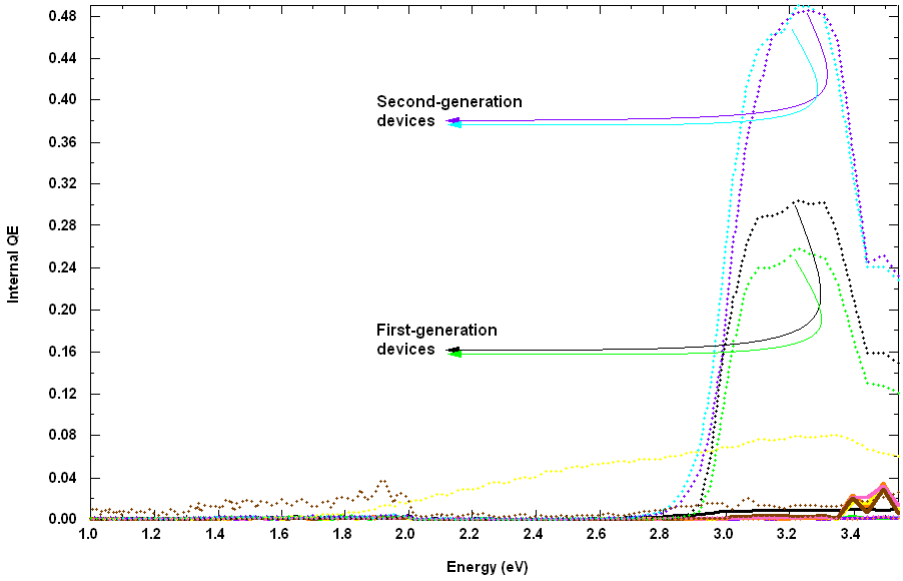


Figure 9: Internal quantum efficiency comparison of first- and second-generation test solar cells.

The next-generation solar cells are designed to overcome the limitations caused by the thin p-type InGaN junctions. Here, the thickness of InGaN p-n junction is increased to 300 nm, where the thickness of p- and n- InGaN are designed at 100 nm and 200 nm, respectively. The fifth- and sixth-generation solar cells employ a 10-nm-thick p- and n-GaN window layer, respectively. These devices yield a substantially higher performance compared to the fourth- and fifth-generation devices. Highest open-circuit voltages are obtained from the sixth-generation solar cells, which employ a thin n-GaN tunneling window layer, as shown in Figure .

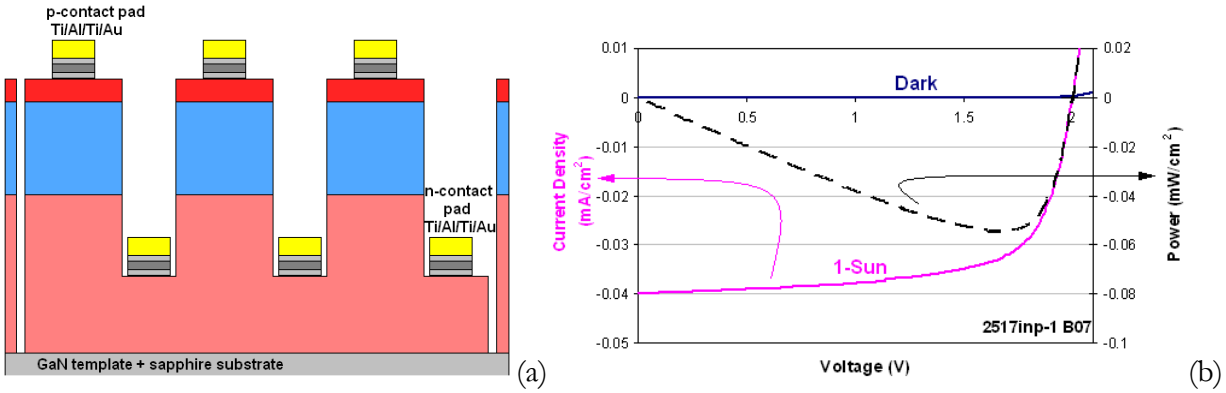


Figure 10: Sixth-generation (a) test solar cells structure and (b) I-V photo response.

The pn-junction InGaN solar cells have a good response, but the understanding of the electrical and optical issues originating in each layer are better studied with Schottky-barrier devices. The Schottky device structures play an important role in understanding the electrical properties of the material, along with being good alternative approaches for making solar cells. Two types of Schottky-barrier solar cells can be formed: (a) Metal-Semiconductor and (b) Metal-Insulator-Semiconductor (MIS). Each of these is presented below.

The difference between an ohmic contact and Schottky contact is the difference in the work function between metal and semiconductor. In the present work, we are studying metal-InGaN. Figure pictorially demonstrates the formation of ohmic and Schottky junctions.

Initial experiments on *p*-GaN were conducted with Ti/Au as the Schottky metal contact. The metal contacts were annealed in the temperature range of 550° and 700°C. Below 550°C, no Schottky diodes were formed. At annealing temperatures greater than 650°C, the metal transformed into small clusters on the surface. Forming gas was used for gas flow, and no significant effect of gas flow rate was observed. The initial results are shown in Figure 12(a) for a Ti/Au metal contact to *p*-GaN annealed at 650°C for 2 min in forming gas, highlighting the photovoltaic effect.

The Schottky-barrier heights are calculated based on Richardson plots, as in Figure 12(b). The barrier height of the device is calculated using the following formula [16]:

$$\phi_B = \frac{V_1}{n} - \frac{k}{q}\frac{d[\ln(I/T^2)]}{d(1/T)}$$,where V_1 is a reference voltage, and n is the ideality factor.

The Schottky-barrier height is calculated to be 0.48 eV.

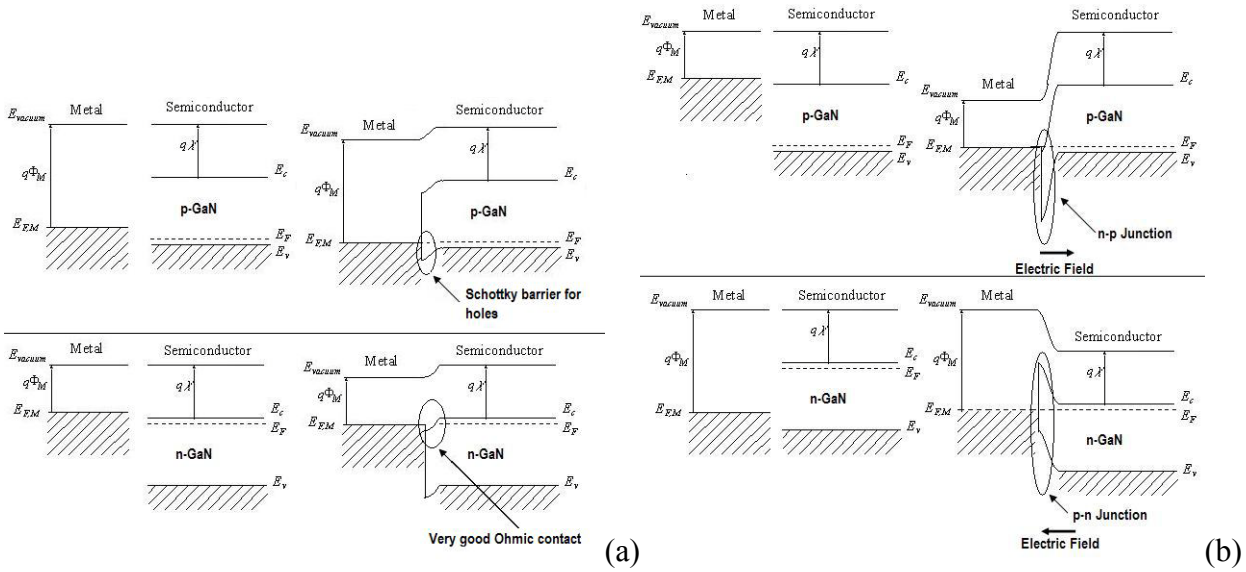


Figure 11: Metal-Semiconductor junction formation.

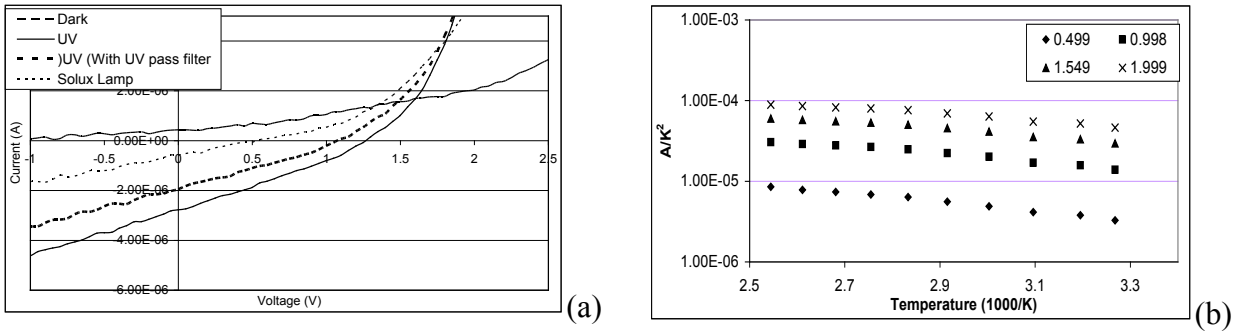


Figure 12: (a) I-V results of Ti/Au metal-*p*-GaN Schottky junction; (b) Richardson plot.

Conclusions

The current research expanded our understanding of InGaN as the potential material for photovoltaics. Phase separation was identified as a key loss mechanism. Phase-separated material (a lower bandgap) controls the open-circuit voltage (Voc), whereas the non-phase-separated material (higher bandgap) controls the absorption. Phase separation in InGaN layers was controlled via optimization of growth conditions and device thickness.

InGaN solar cells with Voc ≈ Eg – 0.4 V and using 2.5-eV bandgap have been demonstrated. This is the first report of (a) an InGaN solar cell with such a low bandgap and (b) a Voc with about 0.4 V of the bandgap, for any material composition in the InGaN material system. The realization of a Voc close to its practical limit for a new material system in such a short timeframe is a major accomplishment. Internal quantum efficiency > 60% for near-bandgap light in GaN solar cells have been demonstrated. The ability to make photovoltaic devices without

requiring a pn junction by using Schottky/MIS approaches has been demonstrated. These are the first reports of Schottky/MIS solar cells.

Modeling of polarization effects in InGaN solar cells has been implemented in PC1D. This is the first photovoltaic modeling to include peizoelectric and polarization effects in a solar cell model. Device design rules for InGaN solar cells, including the impact of p-i-n structures and the impact of polarization and piezoelectric properties of InGaN, have been set forth.

In addition to material quality and growth, modeling, and fundamental device design issues, we have also investigated fabrication and processing approaches. Of particular importance is the demonstration of ohmic contacts to InGaN solar cells using metal contacts, and tunneling contacts have been demonstrated. Further, a standard processing sequence has been developed for fabricating InGaN solar cells.

Overall, the research shows that the InGaN material system can be used to realize high-efficiency solar cells, making contributions to growth, modeling, understanding of loss mechanisms, and process optimization.

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The initial motivation for the University of Delaware/Georgia Institute of Technology project was the then-newly discovered bandgap of InN, which was re-measured at ~0.7 eV, rather than the previous 1.9 eV. This made InGaN a potential material for solar cells with the possibility of absorbing 99% of the solar irradiance. The advantages of the InGaN material system are the wide range of direct bandgap, high absorption coefficient, a low effective mass (high mobility), and strong polarization effects. However, the challenges include material quality, defect density, doping, substrates, and growth issues. Our project led to the understanding of the above challenges; in particular, we studied the material quality in terms of phase separation and how to suppress it. We modeled polarization and developed a solar cell design with these new models.

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