

# Effect of Nitrogen Concentration on the Performance of $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$ Solar Cells

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# EFFECT OF NITROGEN CONCENTRATION ON THE PERFORMANCE OF $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$ SOLAR CELLS

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## ABSTRACT

A study of  $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$  solar cells shows that nitrogen degrades the solar cells even for very small nitrogen concentrations. By comparing the properties of p-on-n and n-on-p  $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$  cells as a function of  $y$ , we find that the n-on-p cells show a more abrupt decrease in the open-circuit voltage and greater decrease of the photocurrent. The asymmetry in the performance of the cells reflects the differences observed for electrons and holes in  $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$ . The electron mobility is degraded much more than the hole mobility when nitrogen is added to GaAs, implying that the electron diffusion length should be degraded more than the hole diffusion length. An electron trap (observed by deep-level transient spectroscopy) affects p-type  $\text{GaN}_y\text{As}_{1-y}$  more than n-type  $\text{GaN}_y\text{As}_{1-y}$ , consistent with the observation that the open-circuit voltage of n-on-p cells decreases more than that of p-on-n cells. The effect of nitrogen on GaNAs cells is shown to be much greater than expected for an isoelectronic impurity.

## BACKGROUND

A four-junction solar cell using  $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{Ga}(\text{In})\text{As}/\text{Ga}_{0.94}\text{In}_{0.06}\text{As}_{0.98}\text{N}_{0.02}/\text{Ge}$  has a theoretical efficiency of 40%-50% [1]. However, achievement of such high efficiencies has been elusive because of poor performance of the  $\text{Ga}_{1-x}\text{In}_x\text{N}_y\text{As}_{1-y}$  (hereafter, GaInNAs) subcell [2,3]. The cause of the poor performance is not yet understood. One solution to this problem is to reduce the photocurrent that is needed from the GaInNAs cell by dividing the photocurrent of the upper junctions, as in a 5- or 6-junction solar cell [4]. A better understanding of the effects of N on GaInNAs cells may lead to a method for improving the cells, or at least a determination of the best performance that can be expected.

The addition of nitrogen to GaAs has been shown to reduce the optoelectronic quality of GaAs, including reduction of the electron mobility [5], reduction of the photoluminescence intensity and lifetime, and an increase in the background carrier concentration [6]. The reduced electron mobility is at least partially caused by alloy scattering associated with the large bandgap bowing of the GaNAs alloy system [7].

Recently, we have reported the correlation between a degraded open-circuit voltage ( $V_{oc}$ ) and the observation of an electron trap using deep-level transient spectroscopy (DLTS) [8]. The structure of the electron trap has not yet

been identified, but its concentration correlates with the nitrogen concentration. The DLTS data is discussed in more detail elsewhere [9,10].

Improved device performance has been obtained by using a GaInNAs p-i-n structure with a wider depletion width. This approach is relatively difficult to implement because of high background carrier concentrations in GaInNAs, but excellent photocurrent can be obtained by growing GaInNAs by molecular-beam epitaxy to reduce the carbon and hydrogen contamination [11].

In this paper we explore the effect of variable amounts of N on the performance of GaInNAs solar cells. We compare results for n-on-p and p-on-n solar cells and report measurements of the dark current,  $V_{oc}$ , and photocurrent (quantum efficiency). The quantum-efficiency data are modeled to derive the minority-carrier diffusion lengths. The reasons for the greater degradation of the n-on-p cells are discussed.

## EXPERIMENTAL METHOD

The GaInNAs solar cells were grown by metalorganic chemical vapor deposition using triethylgallium, trimethylindium, arsine, and dimethylhydrazine or nitrogen trifluoride in a hydrogen carrier gas at atmospheric pressure. Diethylzinc and hydrogen selenide were used as the dopant sources. An arsine pressure of  $\sim 0.2$  torr and a growth rate of  $6.5 \mu\text{m/hr}$  were used. The dimethylhydrazine or nitrogen trifluoride flows were varied to adjust the nitrogen concentration. The growth temperature was also varied between  $600^\circ\text{C}$  and  $650^\circ\text{C}$  to obtain a wider range of nitrogen concentrations. GaAs cells were grown for each of the growth temperatures. The relatively high growth temperatures resulted in some annealing during growth of the devices, but no postgrowth annealing was applied to these samples. The effect of additional annealing is documented elsewhere [12].

The structures for the n-on-p and p-on-n solar cells are shown schematically in Fig. 1. Most of the cells were grown without any indium. A few of the n-on-p cells included some indium. Heterojunction structures were intentionally chosen to help separate the effects of nitrogen on minority-carrier electrons (n-on-p devices) and holes (p-on-n devices). The device areas were  $0.25 \text{ cm}^2$ .

The dark-current-voltage (IV), light-IV, capacitance-voltage (CV), and quantum-efficiency (QE) measurements used standard techniques. The internal QE was calculated from the external QE and the reflectivity. The width of the depleted layer and the net acceptor or donor

concentration in the base were calculated from the 3-kHz CV data, assuming one-sided junctions. The N concentrations were estimated from x-ray diffraction data [6].

25 nm GaInP:Se	500 nm GaInP:Zn
100 nm GaAs:Se	100 nm GaAs:Zn
3 $\mu\text{m}$ Ga(In)NAs:Zn	3 $\mu\text{m}$ GaNAs:Se
150 nm GaInP:Zn	50 nm GaInP:Se

n-on-p device structure                      p-on-n device structure

Fig. 1. Device structures used for this study. Both were grown on GaAs substrates and used GaAs contacting layers, with electroplated gold contacts front and back (not shown). The emitter (front) layers were doped  $\sim 10^{18} \text{ cm}^{-3}$ . The nitrogen (and indium) concentrations were varied for the base layers.

### GaNAs CELL PERFORMANCE

The effect of a very small amount of N on the dark current of n-on-p and p-on-n GaNAs cells is shown in Fig. 2. Whereas, the addition of  $2 \times 10^{18} - 2 \times 10^{19} \text{ cm}^{-3}$  of N increases the dark current of an n-on-p cell by a factor of  $\sim 10$ , N has a relatively smaller effect on p-on-n cells.

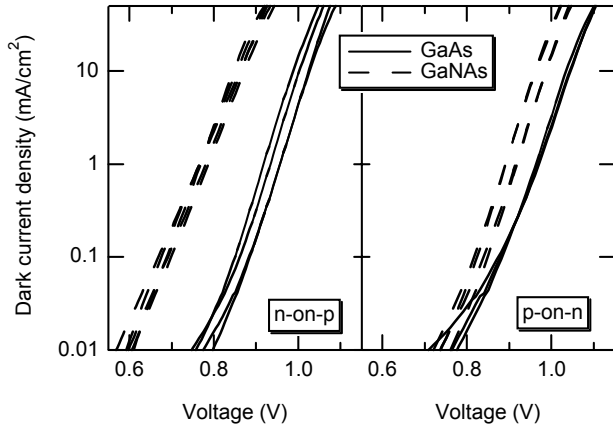


Fig. 2. The effect of  $< 0.1\%$  nitrogen on the dark current of n-on-p (left) and p-on-n (right) GaNAs cells. The bandgap change caused by the addition of the nitrogen was less than 20 meV.

The increase in dark current causes a decrease in the  $V_{oc}$  of the cells. To differentiate the unexpected (because of material degradation) from the expected (because of bandgap reduction) decrease in  $V_{oc}$  with addition of nitrogen, the  $V_{oc}$  is plotted as a function of bandgap in Fig. 3. Consistent with the data shown in Fig. 2, the  $V_{oc}$  decreases much more abruptly for the n-on-p cells than for the p-on-n cells. The addition of indium has very little effect on the  $V_{oc}$ .

The change in the photocurrent, as manifested by the internal QE, is shown in Fig. 4. Again, both types of cells show significant degradation, but the effect on the n-on-p

cells is more dramatic.

The net ionized acceptor and donor concentrations were measured for the n-on-p and p-on-n devices, respectively, and are shown in Fig. 5. The systematic increase (decrease) in acceptor (donor) concentrations with increasing nitrogen implies that the nitrogen is causing point defects that act as acceptors. These defects may be gallium vacancy complexes [13]. Nitrogen-related point defects can also be donor-like [14].

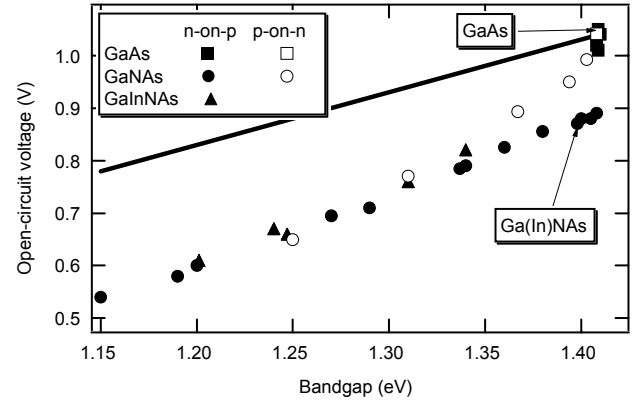


Fig. 3. The open-circuit voltage as a function of bandgap for GaInNAs cells. The line has unity slope, indicating a linear change in  $V_{oc}$  with bandgap.

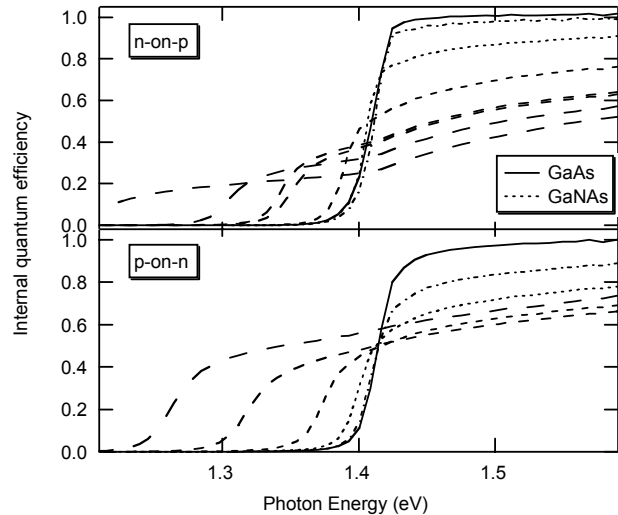


Fig. 4. Internal quantum efficiency for n-on-p (top) and p-on-n (bottom) GaAs (solid lines) and GaNAs (broken lines) cells with variable amounts of nitrogen. The shift to lower bandgap gives an indication of the nitrogen concentration.

### MODELING OF QUANTUM EFFICIENCY

The decrease in the quantum efficiency is dependent both on the device structure and on the material quality. Modeling is necessary to separate these two effects. We chose an emitter design that is independent of nitrogen concentration, simplifying the modeling. The use of doped active layers reduces the depletion layer width; we have assumed that a photocarrier is collected for every photon

absorbed in the depleted layer. The intentional doping gives some control over the base doping. In contrast, if no doping is added to GaNAs, the base doping is highly variable and may be nonuniform, especially if the sample is annealed after growth [14], complicating the modeling. The photocarrier collection in the base and emitter were calculated using Hovel's equations [15].

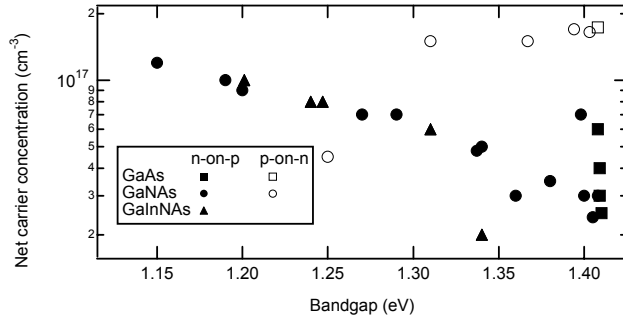


Fig. 5. Base doping of GaInNAs cells as measured by CV at the short-circuit condition.

The absorption coefficient was approximated by  $4.5 \times (E - E_g)^{0.5} \mu\text{m}^{-1}$  where  $E$  is the photon energy and  $E_g$  is the bandgap. The absorption coefficient of GaAs varies slightly with doping, but this effect was not included.

The photocarrier collection in the base was calculated assuming a back-surface recombination velocity of 100 cm/s. Changing the assumptions about the back-surface recombination or absorption coefficient changed the modeled results slightly, but the trends remained the same. The uncertainty increases for the cases when the modeled diffusion length,  $L$ , is longer than the thickness of the base ( $3 \mu\text{m}$ ) or shorter than the depletion width ( $0.1 \mu\text{m}$ ). Depending on the assumptions made, the n-on-p GaAs cell data could be fit only with  $L > 30 \mu\text{m}$ . The  $30\text{-}\mu\text{m}$  value was selected based on literature data [16].

The data in Fig. 4 and QE data from other cells were fit with the model, and the resulting  $L$  values are shown in Fig. 6. Both the hole and electron  $L$  values decrease dramatically with the addition of a small amount of nitrogen. The degradation of  $L$  for the n-on-p cells is much greater (from  $\gg 3 \mu\text{m}$  to  $< 0.1 \mu\text{m}$ , or, according to our best estimate, by a factor of  $\sim 1000$ ) than for the p-on-n cells (from  $\sim 5 \mu\text{m}$  to  $\sim 0.5 \mu\text{m}$ , or a factor of  $\sim 10$ ).

## DISCUSSION

The  $V_{oc}$  of a solar cell is a complicated function of a number of material and device parameters. It is degraded primarily when the minority-carrier lifetime is reduced, but is relatively independent of the mobility. The exact cause of the decreased lifetime in GaNAs alloys is unknown, but our observation that the  $V_{oc}$  of n-on-p GaNAs cells is correlated with an electron trap observed by DLTS may imply that this trap is the problem. This correlation is also observed for the p-on-n cells for which the trap concentration and change in  $V_{oc}$  are both smaller. Because of the higher trap concentration and larger expected degradation of electron lifetime in p-type GaInNAs, the n-on-p cell that relies on collection of

electrons from the p-type base is more strongly affected. We plan to make a more quantitative treatment in a future publication, including quantifying the electron trap concentration and the photocarrier lifetime as a function of bandgap for n- and p-type GaInNAs.

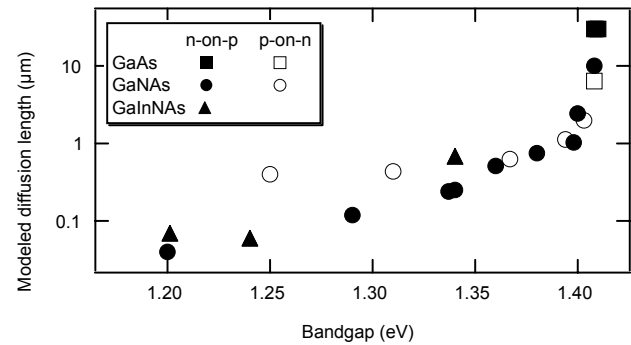


Fig. 6. Modeled electron and hole diffusion lengths.

In contrast to the  $V_{oc}$ , which is primarily dependent on lifetime, the minority-carrier diffusion length is proportional to the square root of both the minority-carrier lifetime and the mobility. Based on the  $V_{oc}$  data, the electron diffusion length should be shortened more than the hole diffusion length because of the relative changes in the lifetime, but changes in mobility should also be important. In high-purity GaAs, the electron diffusion length is longer than the hole diffusion length because of the higher mobility of the electrons. The addition of nitrogen is known to decrease the electron mobility so that it is similar to the hole mobility in GaNAs. The observation that nitrogen degrades the electron diffusion length more than the hole diffusion length is qualitatively consistent with the relative degradation of the majority-carrier transport properties.

In the discussion above, we have contrasted the differences in the effects of nitrogen on n-on-p and p-on-n cells. Understanding these differences are useful toward diagnosing the problem, but the more important observation may be that nitrogen dramatically reduces the performance of all of the GaNAs cells.

Some impurities degrade the performance of solar cells more than others. Impurities forming mid-gap recombination centers are the most problematic. Dopants are associated with shallow levels and can be tolerated in small quantities, but dopant concentrations  $> 10^{18} \text{cm}^{-3}$  also reduce the photovoltaic performance. In contrast, addition of indium (an isoelectronic impurity) has very little effect on GaAs, and can actually improve solar cell performance [17].

Nitrogen in GaAs is an isoelectronic impurity. One might expect the addition of nitrogen to affect GaAs in a way similar to the addition of indium. In Fig. 7 we plot the electron diffusion length data from Fig. 6 as a function of nitrogen concentration and as a function of the electron trap concentration measured by DLTS and corrected by a factor of 20 to compensate for the small penetration of the minority carriers into the depleted layer [10]. We also compare this data to diffusion lengths expected for an isoelectronic impurity such as indium (negligible effect in this concentration range) and shallow dopant such as

selenium. The effect of a deep level is expected to be much stronger. The data presented here show that N degrades GaAs much faster than In. The DLTS data suggest that the electron trap concentration is  $< 10\%$  of the  $[N]$ . This raises the question of whether N can be added without forming the problematic defect. The literature shows that annealing can reduce the electron trap concentration [12,18,19] as well as improve the performance of the solar cells [3,12]. Thus, it may be possible to remove the electron traps without removing the nitrogen, giving hope that GaNAs may yet be used in high-performance devices.

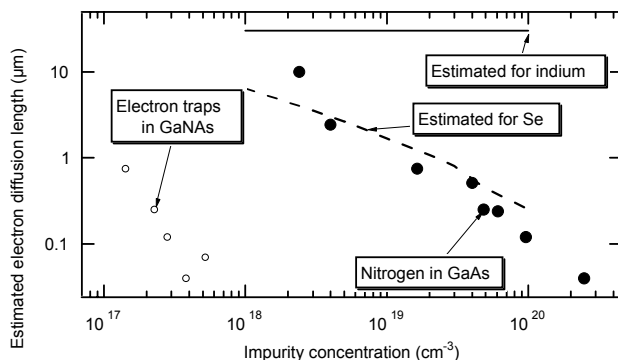


Fig. 7. Electron diffusion lengths as a function of impurity concentration in GaAs. The nitrogen data are from Fig. 6. These data are plotted both as a function of the nitrogen concentration and as a function of the electron trap concentration. The data for In and Se are estimated from the literature [17,20].

### SUMMARY

In summary, we observe a dramatic change in the dark current of n-on-p GaNAs solar cells with the addition of doping levels of N. Even with N concentrations too low to affect the band gap, the N increases the dark current by about a factor of 10, also reducing the open-circuit voltage (Voc). Addition of more N causes a change that is less dramatic and is more consistent with the observed change in band gap. The reduction in Voc is less abrupt, but of a similar size, for p-on-p GaNAs cells. The n-on-p cells exhibit higher electron-trap concentrations than the p-on-p cells, possibly explaining why the Voc (and lifetime) are degraded more for the n-on-p cells.

The minority-carrier diffusion length also decreases very quickly with the addition of nitrogen, with the electron diffusion length changing more than the hole diffusion length. The larger change in electron diffusion length likely reflects the larger change in the electron mobility as well as the larger change in electron lifetime.

Substantially more work will be needed to fully understand why nitrogen is degrading GaAs so much more than is expected for an isoelectronic alloy and whether the material quality can be improved.

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