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Degradation study of InGaAsN PIN solar cell under 1 MeV electrons irradiation

M. Levillayer[1,2,3], S. Duzellier[1], I. Massiot[2], A. Arnoult[2], T. Nuns[1], C. Inguimbert[1], C. Aicardi[3], S. Parola[4], F. Olivié[2], R. Monflier[2], T. Le Cocq[1], R. Rey[1], C. Pons[1], G. Almuneau[2], L. Artola[1]

***Abstract*— The degradation of InGaAsN *pin* subcell under 1 MeV electrons irradiation was studied by characterizing solar cells and dilute nitride bulk layers before and after irradiation. Cells are measured to retain more than 94 % of their original photocurrent after 10^{15} cm^{-2} 1 MeV-electrons irradiation. Moreover, no significant degradation of the optoelectronic properties is observed after irradiation.**

***Index Terms*—Degradation, dilute nitrides, electrons, InGaAsN, irradiation, MJSC, solar cell, DLTS, PL, EQE**

I. Introduction

Multi-junction solar cells (MJSC) based on III-V materials offer today the highest conversion efficiency of all solar technologies, as their structure leads to the optimal usage of the solar spectrum. MJSC are then of prime interest for space applications where W/kg is the key parameter. Today's space solar cell market is dominated by the GaInP/(In)GaAs/Ge tri-junction cell lattice-matched to its substrate, with 28-30 % efficiency [1]. Both the architecture and the materials of this standard tri-junction cell have already been optimized and room for improvement is now extremely limited.

Changing or adding subcells appears then to be necessary in order to overpass the 30 % efficiency level. Theoretical calculations of the optimal bandgap combination point out the need to develop a 1 eV subcell to improve the solar spectrum harvesting [2]. One way for improving the structure is to replace the Ge with a 1 eV material in the bottom cell. Another possibility is to fabricate a 4-junction solar cell by introducing a 1 eV subcell between the GaAs and the Ge ones.

A major requirement for the 1 eV subcell is that the material needs to be lattice-matched on GaAs or Ge. Dilute nitride ($In_xGa_{1-x}As_{1-y}N_y$) alloys have been proposed as candidates for this application as both their bandgap and their lattice parameter can be tuned with varying indium and nitrogen content [3],[4],[5]. As a notable proof-of-concept, Solar Junction reported in 2011 a world record efficiency (at the time) of 43.5 % under AM1.5D concentrated light for a triple-junction incorporating a InGaAsN-based bottom cell [6].

Furthermore, space applications require the subcell to be radiation hard since constant irradiation commonly leads to cell degradation. Indeed, high-energy particles such as electrons or protons can create crystal defects through non-elastic nuclear interactions with the atoms of the lattice. The displacement damage dose (DDD) associated with these defects leads to a decrease in the minority carrier lifetime which reduces the collection efficiency [7]. This degradation can be particularly problematic for a MJSC since subcells are connected in series and thus are each required to generate the same amount of photocurrent.

Several degradation studies have already been reported for InGaAsN cells but the irradiations were carried out on non-optimized cells (regarding both the material and the architecture) grown by metal organic vapor phase epitaxy (MOVPE) [8],[9],[10]. More recent studies reported on quasi-optimized structures achieving short-circuit current density (J_{sc}) higher than 15 mA/cm² under integrated illumination condition (AM0 > 870 nm [11] and AM1.5 > 830 nm [12]). These cells are based on a *n-i-p* design which strongly relies on a field-aided collection regime. Moreover, the dilute nitride material quality was improved by using molecular beam epitaxy (MBE) instead of MOVPE, leading to lower background carrier concentration.

As lattice-matching and current-matching conditions are now fulfilled, InGaAsN subcell integration into MJSC is possible [6],[13]. It is now crucial to quantify and understand InGaAsN degradation under standard irradiation. To do so, we propose a study of the InGaAsN subcell degradation by combining characterization of *I-V*, external quantum efficiency (EQE), deep level transient spectroscopy (DLTS) and photoluminescence (PL) at beginning of life (BOL) and after irradiating samples with 1 MeV electrons.

This paper was submitted the for review

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II. Experimental Method

A. *InGaAsN cell and diode fabrication for DLTS characterization*

The InGaAsN p-i-n cell structures were grown by MBE on 4-inch n-GaAs (001) substrates. The devices structure is composed of a p-i-n GaAs/InGaAsN/GaAs junction and includes a n-$Al_{0.4}Ga_{0.6}As$ back surface field (BSF), a p-$Al_{0.4}Ga_{0.6}As$ window layer and is capped with a p+ GaAs layer (Fig. 1a). TABLE I. summarizes the InGaAsN growth conditions of this series of studied cells, labelled as A, B, C and D. Other growth parameters were kept constant. The composition of the InGaAsN active layer was set to 4.5 % for In and ≈ 1.6 % for N corresponding to the lattice-matched condition. No rapid thermal processing (RTP) was performed on these wafers.

Furthermore, 1.5µm-thick non-intentionally doped (NID) InGaAsN bulk layers were grown on 4-inch n-GaAs (001) substrates with different As/III ratios for PL and DLTS characterizations. The growth conditions of these samples (E and F) can be found in TABLE I. Some of these samples were annealed at 750 °C for 30 s before being processed while others were processed as-grown. Electrochemical capacitance voltage (ECV) measurements showed that these InGaAsN layers are n-doped with concentrations ranging from 3.10^{16} to 1.10^{17} cm^{-3}.

AuGeNi/Au metallization was carried out on the whole backside of the substrate by sputter deposition and a Ti/Au front metallic grid was deposited by e-beam evaporation followed by a lift-off step. Wafers were then annealed at 350 °C for 90 s in order to achieve good ohmic contacts. Wet etching was performed using H_3PO_4/H_2O_2 to form mesa structures to isolate each cell. The GaAs cap layer was afterwards selectively removed using C_6H_8O7/H_2O_2. No anti-reflective coating (ARC) was deposited. Wafers were finally cleaved in 0.25 cm² and 1 cm² cells respectively for EQE and *I-V* measurements, with grid densities ranging from 0 to 10 %. A schematic of the cross-sectional view of the processed cell is shown in Fig. 1b.

The InGaAsN bulk layers were processed with the same technological steps as the solar cells in order to fabricate Schottky diodes (NID InGaAsN / TiAu) for DLTS measurements.

TABLE I. InGaAsN growth conditions

Wafer	Structure	Surfactant	Growth Temperature	As/III
A	*p-i-n* cells	Bi	465°C	12
B		Ø	465°C	12
C		Ø	445°C	10
D		Ø	485°C	10
E	InGaAsN bulk layer	Ø	465°C	11
F		Ø	465°C	8
G	GaAs cell			

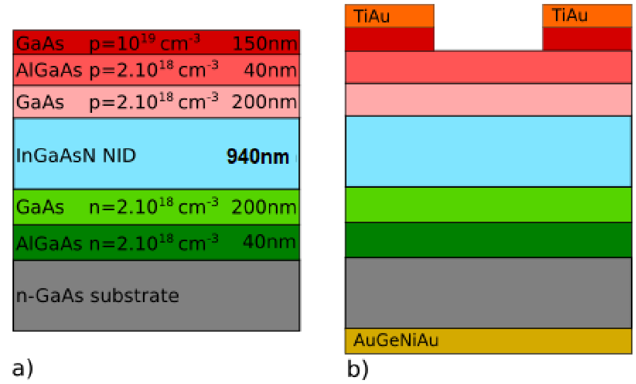

Fig. 1. Structure of the epitaxial stack of the InGaAsN cell (a) and after processing steps (b)

B. *Irradiation conditions*

Solar cells and samples dedicated to DLTS and PL measurements (A to G) were irradiated in the MIRAGE facility at ONERA with a Van de Graaff accelerator providing a 1 MeV electrons beam [14]. The electrons flux was maintained below 5.10^{10} $e^-/cm^2/s$ (≈ 8 nA/cm²) in order to avoid irradiation-induced overheating, as samples were not thermally regulated during irradiation. However, they were mounted on aluminum nitride support and copper plate (see Fig. 2), which both have high thermal conductivity. The irradiation chamber was at room temperature and the pressure was set below 10^{-6} mbar.

Four solar cells thereafter labelled A1, B1, D1 and G1 were irradiated four times with cumulative fluences of respectively 5.10^{13}, 1.10^{14}, 5.10^{14} and 1.10^{15} e^-/cm^2 allowing us to measure their *I-V* characteristics at various fluence levels. A2, B2, C2 and D2 cells were irradiated in one step up to 1.10^{15} e^-/cm^2. The rest of the samples was also directly irradiated to 1.10^{15} e^-/cm^2. GaAs *p-n* cells were also irradiated and used as degradation references (G cells).

III. Results and Discussion

A. *InGaAsN solar cells response to irradiation*

The I-V characteristics were measured with an Oriel solar simulator equipped with a Xenon lamp and a AM0 filter, preliminary calibrated with a reference tri-junction cell. Fig. 3a shows the J-V characteristics of the GaAs reference before and after the different steps of irradiation. As commonly reported, J_{sc} decreases because of defect introduction that shortens the minority carrier diffusion length leading to a reduction of the collection efficiency [15],[16]. The normalized J_{sc} values are plotted as a function of the fluence in Fig. 3b. A reduction of the J_{sc} can be observed as fluence increases, with a degradation rate in good agreement with the results reported in the literature [15],[16],[17]. Moreover, the J_{sc} degradation is found to be consistent with the EQE measurements (not shown here). Open-circuit voltage (V_{oc}) falls from 0.885 to 0.845 V accounting for 4.5 % degradation. This degradation correlates with an increase of one order of magnitude in the dark current density as depicted in Fig. 4 and is consistent with already published results [15],[17].

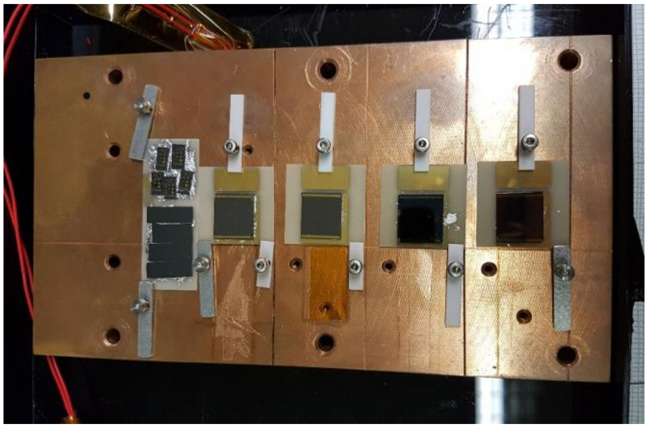

Fig. 2. Solar cells and samples for PL mounted on their holders for irradiation

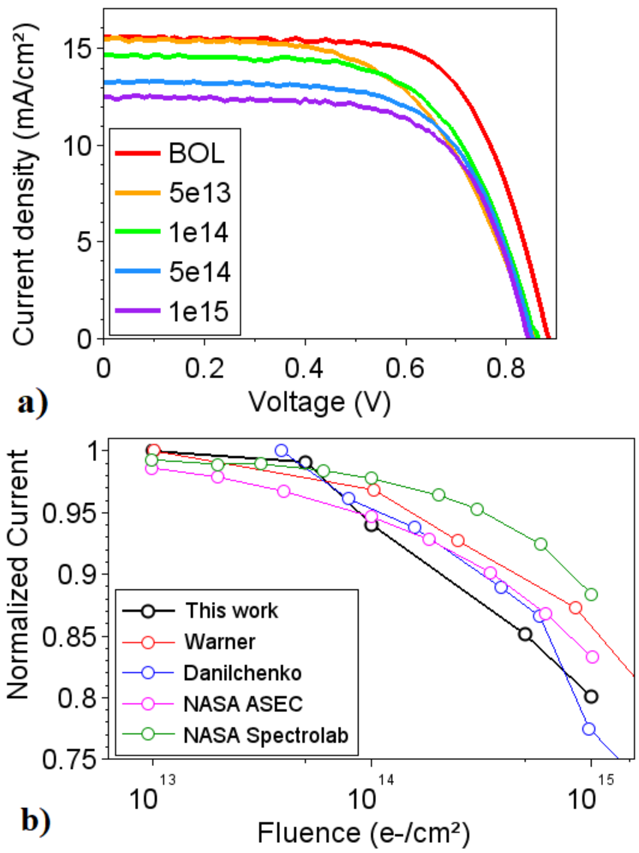

Fig. 3. a) *J-V* characteristics of GaAs G1 cell under AM0 light at 25°C and for different fluences b) Degradation of the photocurrent for G1 as a function of fluence, compared to literature data [15],16]

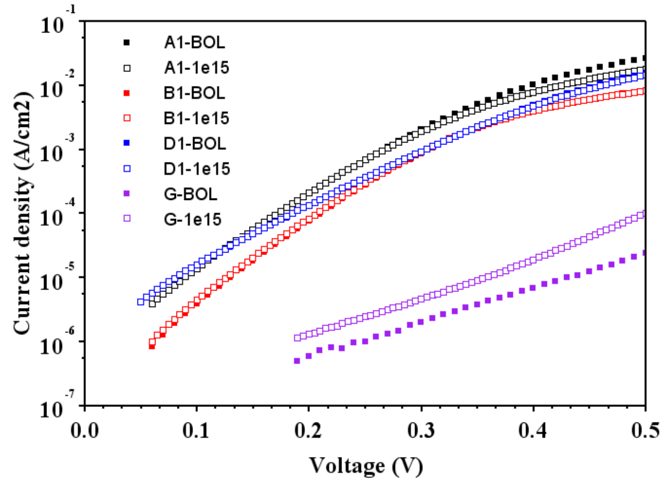

Fig. 4. Dark *J-V* characteristics of solar cells A1, B1, C1 and G before and after 1 MeV electrons irradiation

The *J-V* curves of the InGaAsN cells were measured under spectrally-filtered light in order to reproduce the absorption of upper subcells in a MJSC. For that purpose, a Newport high-pass filter ($\lambda > 870$ nm) was placed in the optical path. Fig. 5a presents the evolution of the *J-V* curves of the A1 cell for different irradiation levels. The B1 and D1 cells exhibit the same *J-V* variation vs fluence as the A1 cell. An electrons irradiation is found to be less detrimental to the InGaAsN cells than to the GaAs reference cell (G1), since the mean degradation of J_{sc} and V_{oc} after 10^{15} e^-/cm^2 are lower than 6 % and 2 %, respectively. Furthermore, the V_{oc} does not follow a monotonic degradation with the fluence which suggests that mechanisms with opposite effects on the recombination rate are simultaneously occurring during irradiation. The very low V_{oc} degradation of the irradiated InGaAsN cells correlates well with the non-degradation of their dark saturation current that can be observed in Fig. 4.

The degradations of J_{sc} for all dilute nitride cells are plotted in Fig. 5b. A clear difference in degradation rate can be observed between the cells irradiated directly at 1.10^{15} e^-/cm^2 and the cells subjected to successive irradiations. That could be due to defect annealing (as discussed below) enhanced by uninterrupted irradiation. Otherwise, epitaxial growth conditions are not found to affect the degradation behavior of the cells, as J_{sc} and V_{oc} degradations for A, B, C and D are found to be roughly the same.

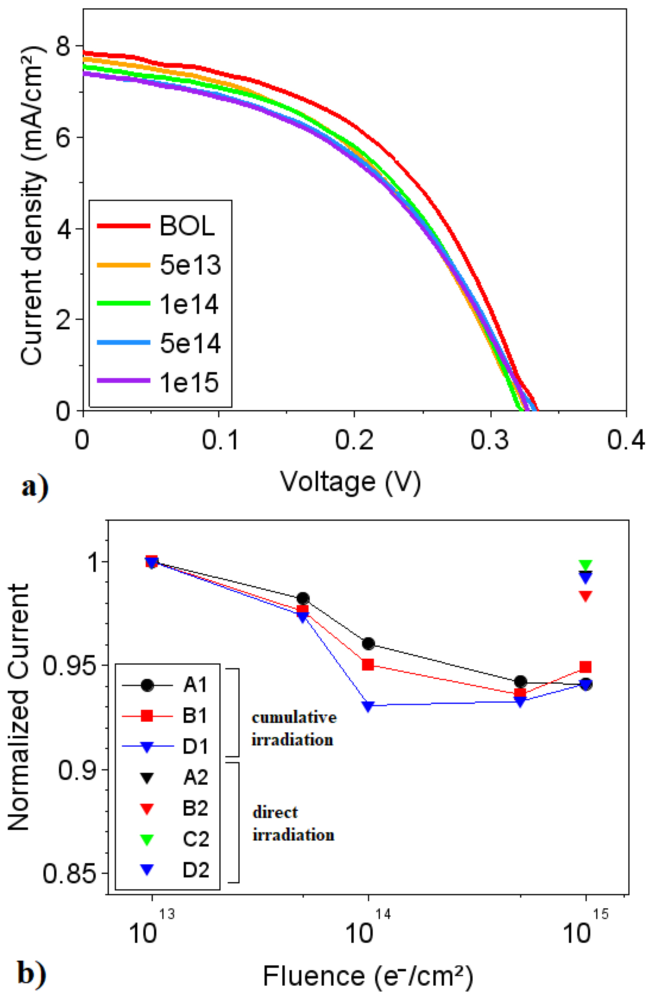

Fig. 5. a) *J-V* characteristics of InGaAsN A1 cell under AM0 > 870 nm light at 25°C and for different fluences b) Degradation of the photocurrent in InGaAsN cells as a function of fluence

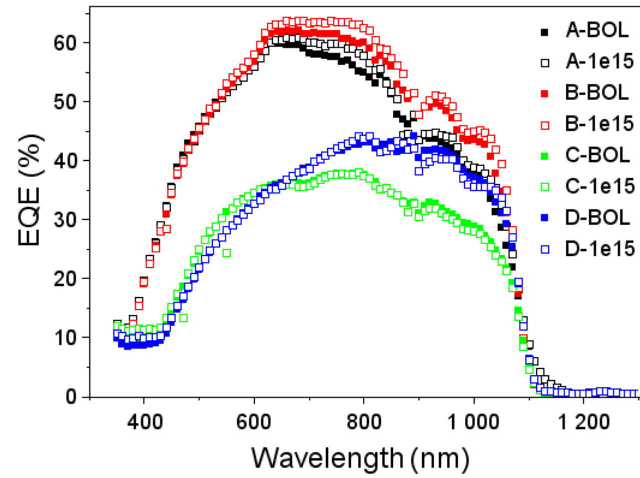

Fig. 6. EQE before and after 1 MeV electrons irradiation for 4 InGaAsN cells A, B, C and D

We performed EQE measurements on solar cells directly irradiated at 10^{15} e^-/cm^2. As it can be seen on the Fig. 6, irradiation has virtually no effect on the spectral response of InGaAsN cells and it can even be observed that the EQE in the range 650-900 nm slightly increases after irradiation for cells A and B. However, this improvement disappears for longer wavelength which explains why it does not lead to higher J_{sc} under filtered AM0 light.

To describe the radiation hardness exhibited by InGaAsN cells, we propose three hypotheses. First, defect annealing and defect introduction occur simultaneously under irradiation. Indeed, annealing of growth defects during irradiation has already been reported for GaAs [18] and InGaAsN [19]. This phenomenon is based on the recombination-enhanced annealing mechanism coupled with the creation of vacancies in the lattice. Irradiation induces multiple ionizations in the material which in turn lead to a high carrier recombination rate. Carrier recombination can then locally bring enough energy to the lattice to anneal defects. However, for the defect recombination to be possible, vacancies need to be present in the lattice as it greatly enhances atomic diffusion [20].

Furthermore, the atomic diffusion of defects increases with temperature, which could explain why direct irradiation leads to lower degradation, as the sample might heat up more than it does under cumulative irradiation.

The second hypothesis to explain the radiation hardness of the cells is the predominance of growth defects over the irradiation-induced ones. Indeed, if at BOL the defect concentration is already high, then the contribution of the defects created during irradiation on the minority carrier diffusion length would remain marginal – provided a low defect introduction rate – and would have a negligible impact on the cell's properties. This hypothesis is supported by the fact that a high density of N-related defects may be present in dilute nitride materials arising from the small size and the high electronegativity of the nitrogen anion [11],[21],[22]. However, our InGaAsN solar cells exhibit good *I-V* and EQE characteristics at BOL, which relatively temper this hypothesis.

A third way to explain this radiation tolerance comes from the *pin* structure and the collection regime of the cells. As we said previously, the InGaAsN *pin* cells rely on the built-in electric field extending across the unintentionally-doped layer to collect carriers, as a workaround for dilute nitride's low minority carrier diffusion length. The background carriers concentration (BGCC) of the InGaAsN layer measured by ECV and the calculated width of the space charge region (SCR) of each cell can be find in TABLE II. For the calculation of this latter value, we assumed the dielectric permittivity of InGaAsN to be equal to the one of GaAs ($\varepsilon_r = 12.9$) and we considered a built in voltage V_{bi} of 0.5 V. Even though the depletion region never exceeds the total width of the nitride layer, the electric field still plays an important role in the carrier collection as the majority of the incident light is absorbed in the SCR which is located at the p-GaAs / n^--InGaAsN interface.

Consequently, the decrease in the diffusion length through the introduction of defects during irradiation has a smaller impact on the InGaAsN cells performance than it has on GaAs ones. A low J_{sc} degradation for GaAs *pin* solar cells has already been reported and attributed to this drift regime [23].

TABLE II. INGAASN DOPING LEVEL AND SCR WIDTH

Wafer	BGCC (cm-3)	SCR width (nm)
A	7.10^{15}	320
B	8.10^{15}	300
C	3.10^{16}	150
D	$< 4.10^{15}$	> 420 nm

B. DLTS and PL evolution with irradiation

To assess the defect annealing explanation, i-DLTFS measurements were performed on InGaAsN samples before and after 1 MeV electrons irradiation. A Phystech DL8000 spectrometer and its analysis software were used for that matter. The reverse voltage was set to -1 V and we used a 0.5 ms filling pulse of 0 V. We measured the current transient between 10 and 50 ms after the end of the pulse. The DLTS spectra of as-grown and annealed samples from E and F wafers are shown in Fig. 7a. Although the crystal quality is different from one BOL sample to another - as depicted by the DLTS spectra - there is no significant difference between BOL and corresponding irradiated samples. Fig. 7b shows the deconvolution of two emission peaks that reveal the presence of two electron traps, with activation energy approximately equal to 0.6 and 0.85 eV (as determined with the Arrhenius plot in Fig. 7c). As the bandgap energy of the DLTS sample is equal to 1.05 eV, the 0.6 eV trap appears to be quite deep and probably acts as an effective non-radiative recombination center. This defect is conjectured to be a nitrogen split interstitial $(N\text{-}N)_{As}$ since it is a typical defect in dilute nitrides and as its activation energy was theoretically calculated to be 0.66 eV below the conduction band [24].

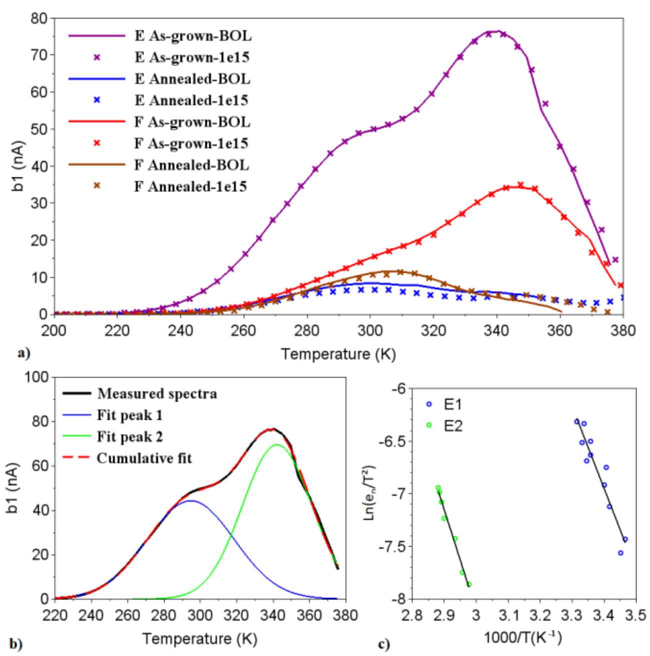

Fig. 7. a) DLTS spectra of samples before and after 10^{15} cm^{-2} 1 MeV electrons irradiation b) Deconvolution of the DLTS spectra of sample E As-grown-BOL in 2 emission peaks c) Arrhenius plot of E1 and E2 traps.

Interestingly, as the amplitude of the DLTS signal is proportional to the defect concentration, it can be deduced that the electrons irradiation does not lead to a change of any of these defects concentration [25].

This result implies that the radiation hardness exhibited by the InGaAsN cells cannot be explained by the annealing of deep level growth defects such as $(N\text{-}N)_{As}$. However, it also shows that there is no sensible introduction of deep-level defects through the irradiation process, which supports our second hypothesis. Moreover, this observation is consistent with the non-degradation of the saturation current and the open-circuit voltage.

PL measurements were then made to assess the impact of irradiation on optical properties, as depicted in Fig. 8. The PL spectra were obtained at room temperature with a 15 mW 488 nm laser diode from Oxxius acting as the excitation source. From Fig. 8 it can be seen that the PL signal increases after irradiation for as-grown samples whereas it remains constant or degrades for annealed ones. The fact that the PL increases after irradiation for as-grown samples means that there is less non-radiative recombination after irradiation. This effect points out the presence of growth-related shallow defects (not detectable with DLTS) and their curing during the electrons irradiation process. That result correlates well with the slight increase in the EQE of unannealed cells after irradiation (Fig. 6). Such a correlation can be explained by the fact that shallow defects can change the doping concentration through compensation mechanisms. Curing those defects through irradiation can then lower the unintentional doping and enhance the drift collection, leading to higher quantum efficiency. Additionally, we can see that the PL intensity of unannealed samples after irradiation gets closer to the intensity of annealed samples at BOL. This suggests that the irradiation acts as the 750 °C RTP for curing shallow defects without however leading to the characteristic bandgap blueshift associated with thermal annealing of dilute nitrides. The plateau-like degradation of the InGaAsN cells between 10^{14} and 10^{15} e^-/cm^2 as well as the small J_{sc} recovery of B1 and D1 cells (Fig. 5b) could then results from the annealing of these shallow defects.

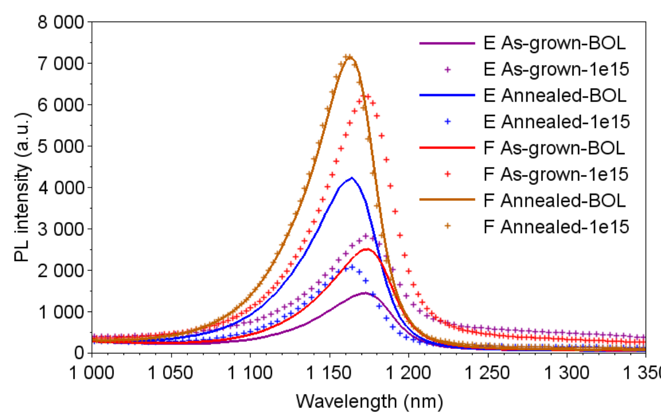

Fig. 8. PL spectra of samples before and after 10^{15} cm^{-2} 1 MeV electrons irradiation obtained at room temperature

Moreover, the fact that the PL signal of the annealed samples does not increase (and even decreases in the case of wafer E) after irradiation can be explained by the change of InGaAsN preferential atomic bonding after RTP. Indeed, nitrogen atoms are reported to go from a Ga-N bonding configuration to a In-N one after annealing [26],[27], which could affect the defect energy formation and consequently the radiation hardness of the material.

IV. Conclusion

InGaAsN *pin* subcells and bulk layers were fabricated, electron-irradiated and characterized, showing a better resistance to 1 MeV electrons than GaAs cells. We have suggested three physical interpretations to explain this degree of tolerance based on both the specific material properties and the junction structure. DLTS and PL analysis on InGaAsN bulk layers show that the radiation hardness cannot be explained by the introduction and the annealing of deep-level defects. Indeed, the growth-defect density may be too important compared to the density of irradiation-induced defects. The PL enhancement and the slight EQE increase are then attributed to the annealing of growth shallow defects during irradiation.

The electrons radiation tolerance exhibited by the InGaAsN cells is an encouraging result for the integration of 1 eV dilute nitride subcell within MJSC for space applications. Indeed, a low degradation rate of the current-limiting subcell is a great asset for high EOL-efficiency MJSC. As a perspective, the degradation of InGaAsN cells through protons irradiation should be studied in order to confirm the radiation hardness of those cells. Multi-junction structures such as GaAs/InGaAsN tandem or GaInP/GaAs/InGaAsN triple-junction solar cells should also be characterized before and after irradiation to validate the degradation studies of individual subcells.

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