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Gwenaëlle Hamon, Nicolas Vaissiere Vaissière, Romain Cariou, Raphaël Lachaume, José Alvarez, et al.. Plasma-enhanced chemical vapor deposition epitaxy of Si on GaAs for tunnel junction applications in tandem solar cells. *Journal of Photonics for Energy*, 2017, 7 (2), 10.1117/1.JPE.7.022504 . hal-01632906

HAL Id: hal-01632906

<https://centralesupelec.hal.science/hal-01632906>

Submitted on 11 Mar 2020

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Journal of Photonics for Energy

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Plasma-enhanced chemical vapor deposition epitaxy of Si on GaAs for tunnel junction applications in tandem solar cells

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Abstract. We fabricated (n) c-Si/ (p) GaAs heterojunctions, by combining low temperature ($\sim 175^\circ\text{C}$) RF-PECVD for Si and metal organic vapor phase epitaxy for GaAs, aiming at producing hybrid tunnel junctions for Si/III-V tandem solar cells. The electrical properties of these heterojunctions were measured and compared to that of a reference III-V tunnel junction. Several challenges in the fabrication of such heterostructures were identified and we especially focused in this study on the impact of atomic hydrogen present in the plasma used for the deposition of silicon on p-doped GaAs doping level. The obtained results show that hydrogenation by H_2 plasma strongly reduces the doping level at the surface of the GaAs: C grown film. Thirty seconds of H_2 plasma exposition at 175°C are sufficient to reduce the GaAs film doping level from $1 \times 10^{20} \text{ cm}^{-3}$ to $< 1 \times 10^{19} \text{ cm}^{-3}$ at the surface and over a depth of about 20 nm. Such strong reduction of the doping level is critical for the performance of the tunnel junction. However, the doping level can be fully recovered after annealing at 350°C . © 2017 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: [10.1117/1.JPE.7.022504](https://doi.org/10.1117/1.JPE.7.022504)]

Keywords: tandem solar cells; plasma-enhanced chemical vapor deposition; tunnel junctions; epitaxy; inverted metamorphic growth.

Paper 17006SS received Jan. 14, 2017; accepted for publication Mar. 13, 2017; published online Apr. 7, 2017.

1 Introduction

This work is part of the French IMPETUS project [innovative multijunction combining metal organic vapor phase epitaxy (MOVPE) and plasma-enhanced chemical vapor deposition (PECVD) epitaxy at low-temperature for solar applications]. Here we present an approach to produce tandem solar cells that is based on the inverse metamorphic growth of crystalline silicon on GaAs.¹⁻³ As shown in Fig. 1, the AlGaAs top cell is grown above 540°C by MOVPE on a lattice-matched GaAs substrate. Subsequently, the group IV cell (Si or SiGe) is obtained via heteroepitaxial deposition by low temperature PECVD (LT-PECVD). After the inverted metamorphic growth, the tandem solar cell is transferred to a low cost carrier. Following the removal of the GaAs substrate, the device process is finished on the low cost carrier involving metal contacts deposition, antireflection coating, and cell (mesa) definition. The main advantage of this approach is that the III-V subcell keeps its high quality as it is directly grown on a lattice-matched GaAs substrate. Moreover, at such a low growth temperature

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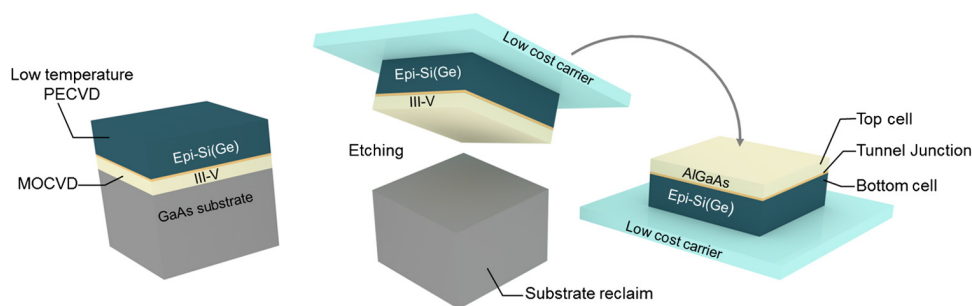


Fig. 1 Principle of the IMPETUS project approach by inverted metamorphic growth of Si(Ge) on III-V.

(175°C), we limit the thermal expansion issues. Last but not least, growing the group IV material on the III-V material prevents the polarity issues encountered in direct growth of III-V on Si.^{4,5}

Previous work in the laboratory⁶ developed the growth of highly doped c-Si on c-Si substrate and on GaAs films, opening the path to the growth of III-V/Si hybrid tunnel junctions (TJ). The TJ is a crucial part of a multijunction solar cell, required to interconnect the subcells. While there is an abundant literature on III-V TJ,^{7,8} the use of hybrid TJ is limited to some works using a bonding approach.^{9,10} The direct deposition of Si on III-V has a huge potential and deserves to be considered for III-V/Si tandem solar cells. Indeed, possible defects or impurity incorporation at the III-V/Si heterointerface and therefore at the middle of the TJ could promote trap-assisted-tunneling. In our previous study, using numerical simulations,¹¹ it was shown that even without defects at the interface, both (n) Si/(p) GaAs and (p) Si/(n) GaAs heterostructures can tunnel if the layers have a sufficient doping level ($>2 \times 10^{19} \text{ cm}^{-3}$). Thus, in this study, we grew a hybrid (n) Si/(p) GaAs junction as well as reference (n) GaAs/(p) GaAs TJ, and investigated the current–voltage characteristics of the devices.

2 Experimental

In this work, it is proposed to grow and characterize heavily doped (n) GaAs/(p) GaAs junctions and (n) Si/(p) GaAs hybrid junctions, aiming at fabricating tunnel diodes. To grow the targeted device, MOVPE was used to grow highly doped GaAs on a (100) GaAs wafer. The used precursors are TMGa and AsH₃. SiH₄ and CBr₄ were added for the n-type and p-type doping, respectively. For each type of film, the highest doping level was achieved while maintaining high crystalline quality. Electrochemical capacitance–voltage (ECV) was used to calibrate the doping level in the p-type GaAs layers. This technique allows measuring the active dopant concentration and performing a depth profile of the carrier concentration. The highest doping level was $1 \times 10^{20} \text{ cm}^{-3}$ for the p-doped GaAs:C and $1.2 \times 10^{19} \text{ cm}^{-3}$ for the n-doped GaAs:Si. It is challenging to achieve a higher n doping level with silicon as a dopant. This is due to its amphoteric nature, which means that the carrier concentration self-compensates above a doping level around $2 \times 10^{19} \text{ cm}^{-3}$.¹² For the hybrid approach, doped Si films were deposited in a multiplasma chamber PECVD reactor operated at a frequency of 13.56 MHz.¹³ First, *in situ* cleaning of the GaAs oxide was performed using a SiF₄ plasma, followed by Si deposition at very low temperature (175°C) using silane (SiH₄) and hydrogen (H₂) gas mixtures to which phosphine (PH₃) was added, leading to an n-type doping level of $1 \times 10^{20} \text{ cm}^{-3}$. Diodes were then fabricated by standard technological steps in a clean room environment: photolithography, metallization, dry mesa etching of the epitaxial n-type silicon layer, and wet mesa etching of the GaAs.

Figure 2 shows schematic diagrams of the structures obtained after processing. Those structures were measured at room temperature in a four probes I-V bench, with a Keithley 2450 sourcemeter limited to 1A.

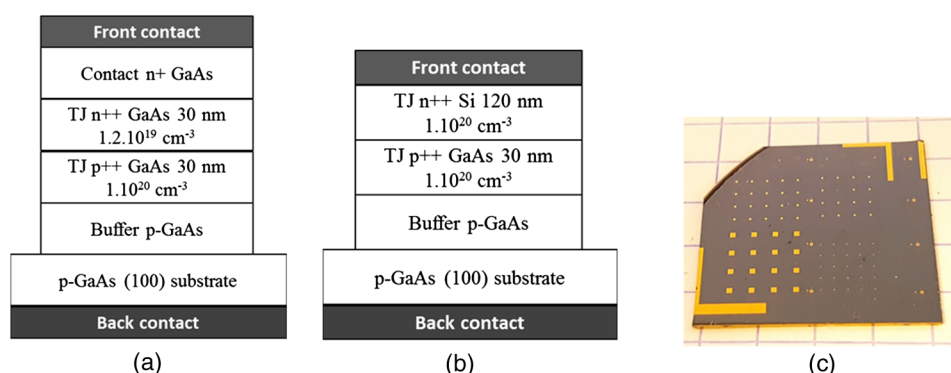


Fig. 2 Test structures of the heterojunctions: (a) reference GaAs TJ, (b) hybrid silicon on GaAs, and (c) picture of the device after processing several diode designs.

3 Results and Discussions

The structures were measured with a four-point probe technique to suppress the series resistance induced by the probes. To check reproducibility, the measurements were made on different diode designs (squares and circles of several sizes). The measured current density versus voltage curves, $J(V)$, are displayed in Fig. 3.

The red curve (circles) corresponds to the standard GaAs/GaAs TJ. As expected, the curve exhibits the characteristic $J(V)$ shape of a TJ with a peak current at low bias (~ 0.1 V), then a valley and finally the classical diode current. A low resistance of $1.4 \times 10^{-2} \Omega \cdot \text{cm}^2$ along with a peak tunneling current around 3 A/cm^2 are obtained. This performance is reasonable considering that we reached a carrier concentration of only $1.2 \times 10^{19} \text{ cm}^{-3}$ in the (n) GaAs, and it is compatible with its use in tandem solar cells working at low to medium concentration. The hybrid junction however (black curve, triangles) led to a normal diode behavior without tunneling effect. The simulations previously presented¹¹ predicted that with doping levels as high as $1 \times 10^{20} \text{ cm}^{-3}$ in both parts of the junction, a tunneling peak current above 100 A/cm^2 could be potentially achieved. Moreover, secondary ion mass spectrometry (SIMS) analysis of the device confirmed that we had a sufficient amount of C atoms in GaAs and P atoms in Si to provide the expected doping levels. However, the above characteristics do not meet the expectations. To explain these discrepancies, one can make the hypothesis that the doping level in the p-type GaAs is not as high as expected. Indeed, the doping level of the GaAs layer was calibrated separately on a test structure with thicker layers by using

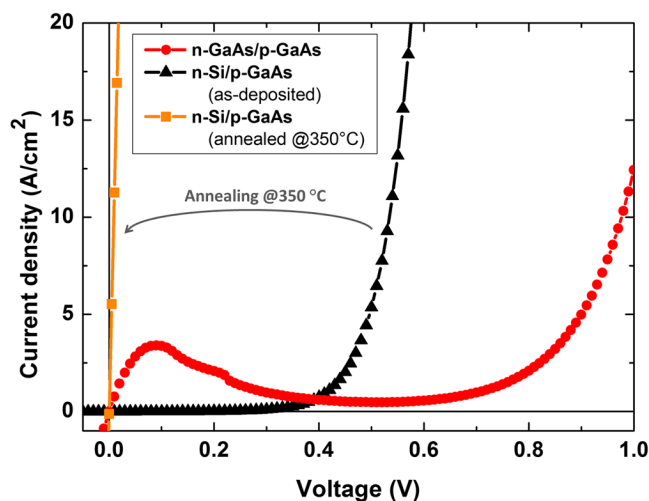


Fig. 3 J-V curves of (n) Si/(p) GaAs (black triangles) compared with a (n) GaAs/(p) GaAs TJ (red circles). Orange squares represent the (n) Si/(p) GaAs after annealing at 350°C .

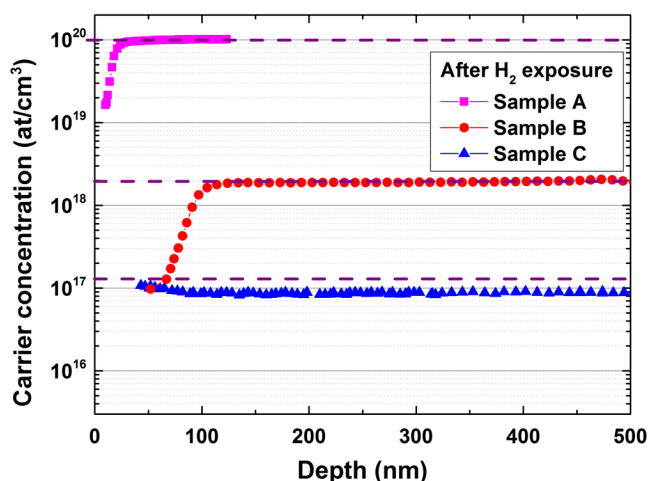


Fig. 4 Carrier concentration profiles of three GaAs(C) samples with different doping levels, after hydrogen plasma exposure. Dashed lines show the nominal carrier concentrations of each sample.

ECV measurements. However, in our final device, the GaAs layer was subject to the PECVD process of Si deposition, and therefore it was exposed to SiH_4 and H_2 plasma during the n-type silicon epitaxy. There are many reports on the effect of a H_2 plasma on doped GaAs. The diffusion of atomic hydrogen in the layer can lead to the formation of C-H complexes that neutralize the dopants. Rahbi et al.¹⁴ studied the acceptor passivation in p-type GaAs doped with different atoms: Zn, C, Si, and Ge. They exposed their samples to a hydrogen plasma at temperatures ranging from 150°C to 300°C for long durations (up to 8 h) and found a strong decrease in the doping level determined by Hall effect measurements. The decrease was about one-order of magnitude in the case of Si, Ge, and Zn doped GaAs. Their GaAs:C layers became highly resistive after plasma exposure. SIMS analysis confirmed that hydrogen had diffused in the p-type GaAs layers until a depth of 1 to 3 μm , depending on the hydrogen plasma exposure time and temperature.

In our case, the (p) GaAs layer was exposed to a H_2 plasma in the PECVD chamber for a much shorter time and lower temperature. It is thus important to verify if under our plasma conditions the dopants in the (p) GaAs are passivated, and to study how to recover from a drop in doping level. In our work, we investigate very thin layers and especially effects at the GaAs surface. This is not only important for understanding the behavior of hybrid p-n junctions, but it is also crucial for the final tandem device. Indeed, even for a standard III-V/III-V TJ in a III-V(MOCVD)/Si(LT-PECVD) tandem solar cell, the last layer before PECVD deposition will be highly doped and should be as thin as possible (below 30 nm).

To further examine the impact of the hydrogen plasma on (p) GaAs:C, three samples have been grown by MOVPE on (100) GaAs wafers with a buffer layer, followed by 500-nm-thick p-type layers with different nominal doping levels: $1 \times 10^{20} \text{ cm}^{-3}$, $2 \times 10^{18} \text{ cm}^{-3}$, and $1.3 \times 10^{17} \text{ cm}^{-3}$ for samples A, B, and C, respectively. The three samples were exposed to very short (30 s) hydrogen plasmas in our PECVD reactor under the standard conditions of epitaxy: substrate temperature of 175°C , pressure of 2.1 Torr, and RF power of 10 W. To characterize the doping level, ECV measurements were performed before and after hydrogenation. Figure 4 shows the resulting carrier concentration profiles after H_2 plasma exposure. The dashed lines show the nominal doping profile in each sample.

On samples A and B the doping activity of carbon was widely reduced close to the surface of GaAs. The doping level decreases by one-order of magnitude in both samples despite the two-orders of magnitude difference in their nominal doping level. For sample A, initially doped at $1 \times 10^{20} \text{ cm}^{-3}$, the 30 s hydrogen plasma exposure affected about 20 nm of the layer, while for sample B, initially doped at $2 \times 10^{18} \text{ cm}^{-3}$, the doping level was reduced over a thicker layer (about 100 nm). The thickness over which dopants are neutralized is inversely dependent on the initial C concentration. This result is consistent with the observation of Chevallier et al.¹⁵ in

Table 1 ECV analysis results on p-type GaAs:C doped at different doping levels.

	Sample A	Sample B	Sample C
N° (cm^{-3})	1×10^{20}	2×10^{18}	1.3×10^{17}
$N^{\text{H}2}$ (cm^{-3})	2×10^{19}	1×10^{17}	8.5×10^{16}
d (nm)	20	100	500

GaAs doped with silicon. The neutralization of carbon is attributed to the fact that atomic hydrogen diffuses in the GaAs layer and forms complexes with carbon (C-H complexes) and thus deactivates it. The lower the concentration of carbon atoms, the deeper atomic hydrogen can diffuse. In the case of sample C, which has the lowest carbon concentration, the decrease in carrier concentration is smaller but happens over the whole 500-nm-thick layer. The effect of the H_2 plasma on lowly p-doped GaAs seems to be negligible at the surface. Table 1 summarizes the obtained nominal carrier concentration of the layer and its value at the surface of the GaAs after plasma exposure, where N° is the nominal carrier concentration, $N^{\text{H}2}$ is the carrier concentration at the surface after plasma exposure, and d is the depth to which the carrier concentration is modified.

This study on the effect of the H_2 plasma in our PECVD process shows that about one-order of magnitude of carrier concentration was lost at the surface of highly doped GaAs. In the case of our hybrid TJ, this happens at the interface between p-type GaAs and n-type Si, where the carrier concentration needs to be as high as possible to enable tunneling. With only $2 \times 10^{19} \text{ cm}^{-3}$ in the GaAs part of the TJ, no tunneling is expected,¹¹ which explains the diode behavior observed in Fig. 3 (black triangles).

Further experiments were designed to find out whether the electrical activity of acceptors can be restored by heat treatment. Thus, sample A has been annealed for 3 min at different temperatures from 250°C to 400°C after plasma exposure, and ECV measurements were performed after each annealing step. The results of this study are shown in Fig. 5.

Full symbols represent the active carrier concentration of the sample before (black squares) and after (blue circles) hydrogen plasma treatment. As explained earlier, the doping level decreases by one order of magnitude at the surface, over about 20 nm. After a 250°C annealing, a slight recovery of the carrier concentration was observed in the first 10 nm, but the dopant passivation extends over a deeper region. This suggests that hydrogen has migrated from carbon atoms located close to the surface to deeper carbon atoms into the bulk. Annealing at 300°C

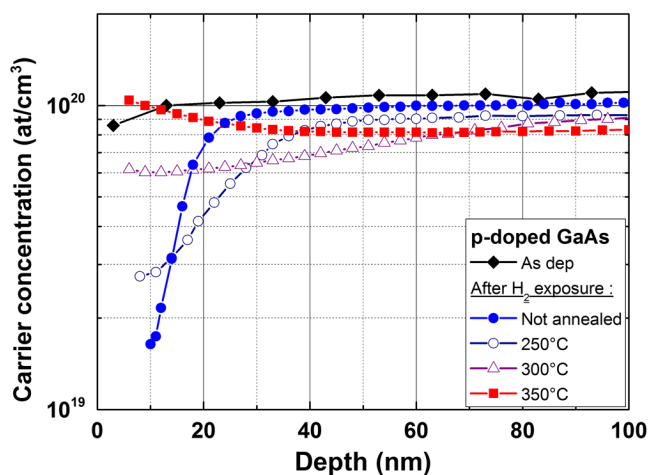


Fig. 5 ECV profiling of a p-doped GaAs layer grown by MOVPE ($1 \times 10^{20} \text{ cm}^{-3}$) as deposited (black diamonds) and after plasma exposure (full circles). The exposed samples have been annealed for 3 min at different temperatures: 250°C (open circles), 300°C (open triangles), and 350°C (red squares).

shows a better recovery of the doping level at the surface, which reaches $6 \times 10^{19} \text{ cm}^{-3}$. After a 350°C annealing, the doping level at the surface is almost fully restored. The same study has been performed on sample B (not shown here), and a complete recovery of carrier concentration was also obtained after heating at 350°C .

The previous experiments performed on p-type GaAs doped with carbon helped us to understand what happened at the interface of our hybrid device. Thirty seconds of H_2 plasma exposure are enough to reduce the doping level by one-order of magnitude, but this effect can be recovered by annealing. Thus, we propose to anneal the device at a temperature of 350°C to restore the doping level of the GaAs layer. The $J(V)$ curve of the hybrid device after annealing is plotted in Fig. 3 (orange squares). It exhibits a linear behavior, with a very low resistance of $9 \times 10^{-4} \Omega\cdot\text{cm}^2$. The best III-V TJ reported in the literature shows resistances below $10^{-4} \Omega\cdot\text{cm}^2$ for high concentration applications.¹⁶ The resistance of our hybrid devices is thus very promising for our low concentration application. As a matter of fact, as judged by the slopes at the origin, the resistance of the hybrid device (orange squares) is smaller than that of the GaAs:Si/GaAs:C TJ grown in our laboratory (black triangles, $1.4 \times 10^{-2} \Omega\cdot\text{cm}^2$). However, the typical negative region of a TJ is not visible, which could be caused by a high dislocation density in the layers.¹⁷

4 Conclusion

In summary, we have presented a first hybrid PECVD Si on MOVPE GaAs device aiming at obtaining a hybrid TJ. Heavily n-doped crystalline silicon was grown successfully by PECVD at 175°C on a p-doped GaAs layer. The electrical results of the fabricated device unexpectedly showed a diode behavior instead of a TJ behavior, meaning that the actual doping level in the device was not as high as the nominal one. We highlighted the strong effect of the hydrogen plasma during the PECVD process: 30 s of H_2 plasma at 175°C are sufficient to reduce the doping level of a heavily doped p-type GaAs by a factor of 10, due to the neutralization of carbon dopants after formation of C-H complexes, thus preventing the hybrid device from tunneling. This neutralization can be recovered after a 350°C annealing. Highly doped GaAs:Si/GaAs:C junctions were also processed and showed good tunneling properties, suitable for a III-V/Si tandem solar cell.

Acknowledgments

This work was supported by the French ANR research program IMPETUS (ANR-13-PRGE-0009) and by a CIFRE convention of the ANRT (no. 2014/1031). The author would like to thank Christoph Sachs and Linda Assam for the attentive reading of this paper.

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Nicolas Vaissiere was qualified to PhD level in 2014 from ENS Cachan. He made the first x-ray detector based on epitaxial diamond by BEN-MPCVD at CEA. During one year at GEMaC/CNRS as a postdoc researcher, he succeeded in adapting MOCVD technology on a commercial MPCVD reactor for n-type doping of diamond. Currently, he is at LPICM/Ecole Polytechnique working on the epitaxial integration of Si(Ge) on III-V at low temperature PECVD for tandem solar cells.

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Jean-Paul Kleider holds his engineering degree from Ecole Supérieure d'Electricité in 1984 and received his PhD from the University Pierre et Marie Curie (Paris VI) in 1987. He is leading the physics and electronics of materials, devices, interfaces, and contacts division at GeePs, where he works as a CNRS director of research. He has extensive experience in the field of electrical and optoelectronic characterization and has published over 240 papers.

Jean Decobert is epitaxy team leader at III-V Lab. He has been working on epitaxial growth since 1987. He received his PhD in microelectronics from the University of Lille and joined France Telecom in 1993. In 2004, he joined the III-V lab, where he was in charge of MOVPE for optoelectronic applications, with research activities on photonic integrated circuits. Actually, he focused on selective area growth technique, III-V integration on silicon and photovoltaic tandem solar cells.

Pere Roca i Cabarrocas received his PhD from the University Paris VII in 1988. After a post-doc at Princeton University, he joined the LPICM, where he holds a position as a CNRS director of research. He has a long experience in the plasma deposition of silicon based thin films. He was the recipient of the silver medal from CNRS in 2011. He has over 450 papers in peer-reviewed journals and holds 34 patents.