physica status solidi b
www.pss-b.com

Proton Energy Loss in GaN

Jean-Yves Duboz, Julie Zucchi, Eric Frayssinet, Sébastien Chenot, Maxime Hugues, Jean-Claude Grini, and Joël Hérault*

The proton energy loss in GaN in an energy range between 0 and 65 MeV is investigated. The energy of protons generated by a cyclotron at about 65 MeV is varied by inserting an energy-absorbing medium of varying thickness. The precise modeling of the GaN Schottky diode response as a function of the absorbing medium thickness allows us to demonstrate that the energy absorption loss in GaN precisely follows the Bethe theory. In addition, the region of the detector contributing to its response to a proton beam, which is of prime importance for proton detector optimization, can be identified.

1. Introduction

The energy loss of high energy particles in matter has been calculated a century ago and is well described by the Bethe formula.[1] This formula gives the average energy loss per unit distance or unit distance times density, but does not describe the random nature of the collisions and the related fluctuations in the energy loss. These fluctuations were first described by Landau[2] with a universal asymmetric probability density function, and refined by others.[3,4] Experimentally, stopping ranges and straggling ranges have been measured in silicon and germanium.[5,6] GaN is currently becoming the most important semiconductor after silicon. AlGaInN alloys are nowadays widely used in light emitting diodes (LEDs) for lighting and displays, for lasers in the Blu-ray technology, and start to emerge for electronic applications in the radio frequency and power domains. GaN is a wide bandgap material with a strong chemical and mechanical stability. As a result, GaN is expected to be more robust than many other semiconductors against degradation under ionizing radiations. High electron mobility transistors based on nitrides are expected to be more radiation resistant than Si or GaAs devices[7,8] and could replace them for space applications and in harsh environments. Schottky diodes based on GaN were also recently demonstrated to be sensitive, linear, and stable detectors for high energy protons with applications in proton therapy.[9] Hence, it becomes timely to investigate the energy loss of high energy particles in GaN. In this letter, we study the spectral absorption of protons in a GaN-based Schottky diode.

J.-Y. Duboz, J. Zucchi, E. Frayssinet, S. Chenot, M. Hugues
Université Côte d'Azur
CNRS
CRHEA
Valbonne 06560, France
E-mail: jyd@crhea.cnrs.fr

J.-C. Grini, J. Hérault
Fédération Claude Lalanne
Université Côte d'Azur
Nice 06200, France

J.-C. Grini, J. Hérault
Cyclotron Biomédical
Centre Antoine Lacassagne
Nice 06200, France

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/pssb.202100167.

DOI: 10.1002/pssb.202100167

2. Measurements

GaN Schottky diodes were fabricated. A 10 μm undoped active region was grown on a doped GaN substrate by metal organic chemical vapor deposition (MOCVD) in an Aixtron 6Closed Coupled Showerhead. A Schottky contact based on 10 nm of Pt followed by 100 nm of Au was deposited on the top while an ohmic contact was deposited on the back. All fabrication details can be found in the study by Duboz et al.[9] The diodes were tested in the Medicyc line (Lacassagne Proton Therapy Center, Nice), with 64.8 MeV protons and were found to respond linearly with proton beam current. The results suggested that the response was originating from the undoped GaN region,[9] whereas the 300 μm-thick GaN substrate was not or not noticeably contributing due to its high doping level. In fact, the carrier collection is much higher in the depletion region of the Schottky diode than in the remaining undoped GaN region. The depletion region is estimated to be a few μm thick, so that the contributing material thickness is likely to be even less than 10 μm. Some photoconductive gain was found, possibly related to the low field region in the undoped GaN layer, leading to a response larger than the pure photovoltaic response expected in a Schottky diode. The question of the contributions of the depleted region, of the undoped region, and of the substrate, will be discussed at the end of the article. Let us note also that the protons are incident on the front side of the diode, so that the proton energy distribution in the device is equal to the incident proton energy distribution.

The initial proton energy is set by the cyclotron and cannot be varied. To tune the energy of protons incident on the GaN diode, we have inserted poly(methyl methacrylate) (PMMA) with a varying thickness in front of the GaN diode. This was done in two ways. First, we inserted PMMA plates with an increasing total thickness from 0 to 31 mm. The stopping range being about 30 mm in PMMA, we were able to cover the whole energy range from 64.8 MeV down to zero. The Schottky diodes were reverse biased from 0 to −1 V. The isochrone cyclotron delivers proton pulses with a duration of 7 ns and a frequency of 25 MHz. However, our setup was not able to time resolve the pulses and all measurements were made in CW. For a proton current of 20 nA, the current in the GaN diode was 0.18 μA at 0 V and increasing to 14 μA at −1 V. **Figure 1** shows the detector current at 0 V as a function of PMMA thickness, normalized by its value without PMMA (0.18 μA). We observe that the current is first constant for increasing PMMA thicknesses, then increases and finally abruptly decreases. This is in qualitative agreement with expectations and represents the Bragg peak in GaN. It was however impossible to measure the peak with a higher resolution with individual PMMA plates. Hence, we implemented a second way of measuring this peak with a higher resolution using a 90 steps PMMA wheel with an increasing thickness, from 23.7 to 30.3 mm. The wheel was rotating and its rotation angle was monitored so that the detector current could be continuously measured as a function of the wheel position or PMMA thickness. To shift the thickness range toward a zero transmission, a 2 mm PMMA plate could be added to the wheel. Figure 1 shows the current at 0 V as a function of PMMA thickness, including points measured with the additional 2 mm plate. We observe that the Bragg peak is much better resolved, with a larger dynamics, compared with the first method based on individual PMMA plates. This is explained by the lateral straggling of the beam in the PMMA. For practical reasons, individual plates could not be placed very close to the detector and the beam broadening was increased in the air gap between the plates and the detector. In addition, the broadening is even further increased due to the numerous PMMA/air interfaces. On the contrary, the wheel could be placed very close to the GaN diode and there were two interfaces only, both leading to a smaller impact of lateral straggling. We carried out the same measurements at different biases, and obtained similar results with however a tendency to reduce the peak dynamics (relative peak amplitude reduced by 13% at −1 V).

3. Interpretation and Modeling

To interpret the results, one must calculate the energy loss in the PMMA. The energy loss and stopping range in PMMA (density 1.18) are given by the Ziegler table (SRIM software).[10] The stopping range has been carefully verified experimentally as PMMA is routinely used in proton therapy to shift the penetration range in the tissues. The mean proton energy after crossing a given PMMA thickness can be calculated, and is found to first decrease linearly with PMMA thickness up to about 24 mm and then to decreases more and more rapidly between 24 and 30 mm. One can then plot the GaN diode response as a function of the mean proton energy. We find a broad spectral response (not shown), with a peak at 5.8 MeV and a full width at half maximum of 12 MeV, with a response extending to negative energies. This is absolutely different from the expected energy loss spectrum shown in **Figure 2** (note that the energy axis is in log scale), which is a narrow curve peaked at about 0.1 MeV and reaching zero for a proton energy equal to zero. The experimental curve must be understood in term of mean energy of the protons, and is strongly affected by the fact that protons are not monoenergetic anymore after passing through the PMMA. This broadening in energy has been described by Landau[2] and is calculated in the SRIM software by the straggling range, which increases along the proton path (roughly as the square root of the traveled distance) and reaches 1.2 mm for 30 mm of PMMA for 64.8 MeV protons. To consider the longitudinal straggling, we used a distribution function of the effective PMMA thickness with a full width at half maximum (FWHM) of 1.2 mm. We used either a Gaussian distribution or an asymmetric function close to the Landau distribution, with a broader width on the large PMMA thickness side which reflects the broader width on the high energy loss side in the Landau distribution.[2] Similar results were obtained in both cases and we will present results obtained with the Gaussian distribution. For each proton energy, we calculated the absorption in GaN using the absorption spectrum dE/dx given by the SRIM software. For a given PMMA thickness, the absorption in the GaN detector is given by

$$A(L) = \int_{L-\Delta L}^{L+\Delta L} R(E(x), T) \times g(x)dx \quad (1)$$

L and x are the nominal and effective PMMA thicknesses, respectively, $E(x)$ is the mean energy of protons of initial energy

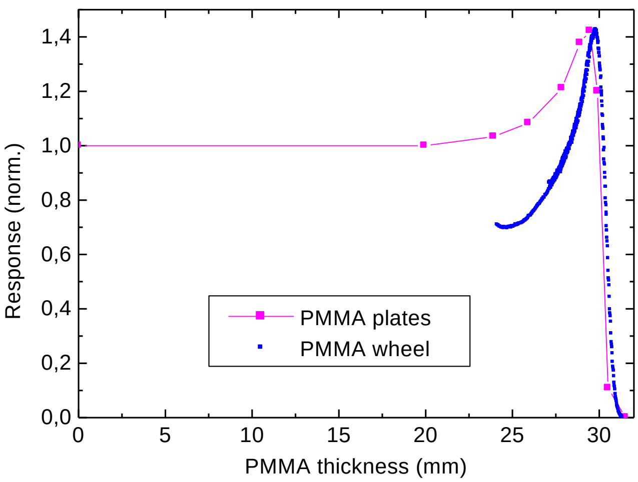

Figure 1. Current in the GaN diode at zero bias as a function of the PMMA thickness. The current is normalized by its value without PMMA. The PMMA is inserted in the form of individual plates (pink dots and line) or of a rotating wheel (blue dots) along the proton beam in front of the GaN diode.

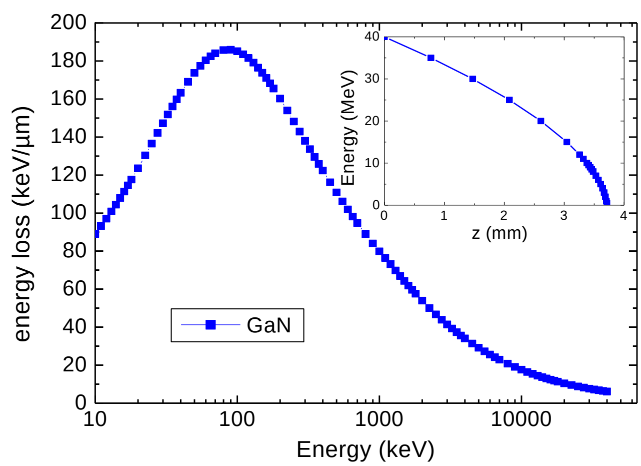

Figure 2. Energy loss in GaN as a function of proton energy, calculated from the SRIM software, and used to model the GaN response. Inset: variation of the proton energy along its path in GaN.

64.8 MeV after crossing a PMMA thickness of x, $R(E,T)$ is the energy absorbed in a GaN slab of thickness T by protons of energy E, and ΔL is the thickness range for the integration. ΔL varies as the longitudinal straggling $S(L)$ for protons of initial energy 64.8 MeV and after crossing a PMMA thickness of L. For instance, for a PMMA thickness of 30 mm, $S(30\text{ mm}) = 1.2$ mm as mentioned earlier. $g(x)$ is the Gaussian (or Landau) function with a FWHM equal to S(L). In practice, we took $\Delta L = S(L)$, so that we integrate for Gaussian values $g(x)$ larger then 1/16. Figure 2 shows the energy loss in GaN calculated by SRIM. Please note that the energy axis is in log scale to show the highly peaked function at small energies. The energy absorbed in a GaN layer is directly related to the energy loss shown in Figure 2.

However, it is not simply given by the product of the differential energy loss (dE/dx) by the thickness T as the absorption saturates at the Bragg peak. Such a saturation occurs when $dE/dx \times T \sim E$. Even for 1 µm-thick GaN layer, at 100 keV, the energy loss reaches 190 keV µm^{-1} leading to saturation and a complete absorption. To calculate the correct absorption, we must integrate the absorption loss along the proton path and find the variation along the path of the mean proton energy $E_{\text{GaN}}(x)$, as we did for PMMA. The inset of Figure 2 shows the variation of the proton energy along its path in GaN. The energy decreases first linearly with distance in GaN and then decreases more and more rapidly, as it does in PMMA. Then, we can calculate the absorption in a GaN layer of any thickness T at any position x by calculating $E_{\text{GaN}}(x) - E_{\text{GaN}}(x+T)$. We carried out the calculation of the integral in three cases: 1, 10 (undoped region), and 300 µm (substrate). **Figure 3** shows the result of the calculation and its comparison with experimental data (same experimental data as in Figure 1, for the wheel only). Note that all curves have been normalized to unity at the peak.

We observe that the best fits are obtained for absorption regions that are 1–10 µm thick. We cannot be more precise and cannot deduce the exact thickness of the contributing region in this 1–10 µm range. However, our results indicate that the GaN substrate (300 µm) does not contribute as the corresponding fit clearly deviates from experimental data, which confirms our initial assumption. The experimental peak is slightly broadened on the small PMMA thickness side compared with the theoretical one, which can be mostly explained by the lateral straggling in PMMA. Lateral straggling is smaller with the wheel than with the plates, but remains non negligible. Indeed, lateral straggling in 30 mm-thick PMMA reaches 740 µm, whereas the beam has a Gaussian shape with a full width at half maximum of 7 mm, and the GaN diode is 1 mm wide. Lateral straggling tends to reduce the signal when the thickness increases, which counterbalances the increase in energy loss. Other possible parasitic effects such as external photoemission from the GaN layers or from the metal contacts could also contribute. Another possible deviation could arise from the relation between absorption and current. It is commonly assumed that the number of electron hole pairs created by the absorption of an energy ΔE is given by $\Delta E/(3 \times E_g)$ where E_g is the bandgap energy. Any deviation from this law as a function of energy would modify the exact response shown in Figure 3. However, in this energy range, such a deviation from a constant ratio between absorption and protocurrent is very unlikely. Finally, let us add that longitudinal straggling in GaN was not considered. Indeed, for energies larger than 2 MeV, protons incident on GaN are passing through the 10 µm GaN region (see Figure 2), and longitudinal straggling in GaN will just affect their stopping range in the substrate, with no incidence on the detector response. For energies smaller than 2 MeV, longitudinal straggling in GaN is smaller than 1 µm, and can be neglected compared with the 10 µm GaN thickness. As explained in a previous publication,[9] the response at 0 V can be described by the photovoltaic response originating from the depletion layer which is 0.4 µm for a doping of 10^{16} cm^{-3}. The diffusion length is less than 1 µm in GaN,[11] so that a 1 µm thick region can be assumed to contribute to the response at 0 V. When the bias increases, the large increase in response cannot be explained from the increase in the depletion region only, and a contribution from the whole undoped region with a photoconductive gain is likely to explain the response. This information agrees with the conclusion of this study based on the spectral dependence of the response.

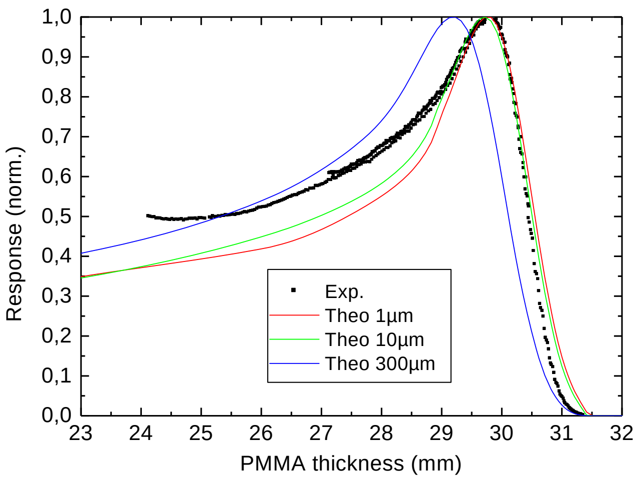

Figure 3. Experimental response of the GaN diode as a function of PMMA thickness (same as in Figure 1 for the wheel) and corresponding calculated responses for three GaN layer thicknesses (1, 10, and 300 µm).

4. Conclusion

In conclusion, we have experimentally measured the proton energy loss in GaN and we have demonstrated that it precisely follows the theoretical spectral variation given by the Bethe formula[1] and shown in Figure 2, at least up to 65 MeV. We have also shown that this spectral response can be used to identify the region contributing to the current in a device where many regions are absorbing, which will strongly help in developing proton detectors and improving their performance.

Acknowledgements

The authors acknowledge support from GANEX (ANR-11-LABX-0014). GANEX belongs to the public funded "Investissements d'Avenir" program managed by the French ANR agency.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Keywords

Bethe theory, energy losses, GaN, protons

Received: April 20, 2021
Revised: June 14, 2021
Published online: July 11, 2021

[1] H. Bethe, *Ann. Phys. (Leipzig)* **1930**, *397*, 325.
[2] L. Landau, *J. Phys. USSR* **1944**, *8*, 201.
[3] P. V. Vavilov, *Sov. Phys. JETP* **1957**, *5*, 49.
[4] H. Bichsel, *Phys. Rev.* **1970**, *B1*, 2854.
[5] S. Meroli, D. Passeri, L. Servoli, *JINST* **2011**, *6*, P06013.
[6] H. Esbensen, O. Fich, J. A. Golovchenko, S. Madsen, H. Nielsen, H. E. Schiøtt, E. Uggerhøj, C. Vraast-Thomsen, G. Charpak, S. Majewski, G. Odyniec, G. Petersen, F. Sauli, J. P. Ponpon, P. Siffert, *Phys. Rev. B* **1978**, *18*, 1039.
[7] B. D. Weaver, T. J. Anderson, A. D. Koehler, J. D. Greenlee, J. K. Hite, D. I. Shahin, F. J. Kub, K. D. Hobart, *ECS J. Solid State Sci. Technol.* **2016**, *5*, Q208.
[8] A. Stocco, S. Gerardin, D. Bisi, S. Dalcanale, F. Rampazzo, M. Meneghini, G. Meneghesso, J. Grünenpütt, B. Lambert, H. Blanck, E. Zanoni, *Microelectron. Reliab.* **2014**, *54*, 2213.
[9] J.-Y. Duboz, J. Zucchi, E. Frayssinet, P. Chalbet, S. Chenot, M. Hugues, J.-C. Grini, R. Trimaud, M. Vidal, J. Hérault, *Biomed. Phys. Eng. Express* **2019**, *5*, 025015.
[10] J. F. Ziegler, *The Stopping and Range of Ions in Matter*, http://www.srim.org
[11] J.-Y. Duboz, F. Binet, D. Dolfi, N. Laurent, F. Scholz, J. Off, A. Sohmer, O. Briot, B. Gil, *Mater. Sci. Eng. B* **1997**, *50*, 289.