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GaN Schottky diodes for proton beam monitoring

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Abstract

We have demonstrated that GaN Schottky diodes can be used for high energy (64.8 MeV) proton beam monotoring. Such proton beams are used for tumor treatment, for which accurate and radiation resistant detectors are needed. GaN Schottky diodes have been measured to be highly sensitive to protons, to have a linear response with beam intensity and fast enough for the application. Some photoconductive gain was found in the diode leading to a good compromise between responsivity and response time. The imaging capability of GaN diodes in proton detection is also demonstrated.

AlGaInN alloys are nowadays widely used for LEDs for lighting and displays, for laser in the Blu-ray technology, and start to emerge for electronic applications in the radio frequency and power domains. GaN is a wide band gap material with a strong chemical and mechanical stability. The displacement energy is as large as 45 eV for the Ga atom and 109 eV for the N atom [1]. As a result, GaN is expected to be more robust than many other semiconductors against degradation under ionizing radiations. As an example, GaN based High Electron Mobility Transistors are expected to stand 10 times higher doses than their GaAs cousins, partly due to the difference in displacement energies and partly due to the piezoelectric field in nitrides [2]. Ion implantation in GaN shows degradation thresholds in the range from 10^{14} cm^{-2} for heavy ions at an energy of 500 keV to 10^{16} cm^{-2} for light ions at 10 keV [3]. The GaN device resistance against radiation has been studied experimentally. Apart a few studies on LEDs [4], most studies were devoted to Schottky diodes and transistors with the motivation of using GaN electronics for space applications. For neutron irradiation, a degradation threshold was found [5-7] for doses in the range of 10^{15} cm^{-2}. For proton irradiation, the threshold dose was found to be above 10^{13} cm^{-2} for energies of 3 MeV [8] and degradation was clearly observed for a dose of 2×10^{15} cm^{-2} for energies of 5 MeV [9]. A more detailed analysis of structural defects created by an irradiation by 23 MeV protons at doses of 6×10^{14} cm^{-2} was reported [10, 11]. As another benefice of its resistance to irradiation, GaN can be used for fabricating radiation hard particle detectors [12] in harsh environments such as synchrotrons [13] or for fabricating X-ray detectors for medical applications [14-16]. In the medical area, high energy particles are commonly used for cancer treatment. In particular, proton-therapy is used in cases where the tumor to be irradiated is close to vital and sensitive organs. A typical example is the eye tumor. The irradiation must be limited to the tumor in the eye while sparing whenever possible macula and optic nerve. For that, one takes advantage of a specific property of protons, which present very steep stopping profile in matter (Bragg peak) [17]. The proton dose has to be very well

controlled, with an accuracy better than 3%, and detectors are thus needed both in vivo (to measure the dose during the patient irradiation) and on the beam (to monitor the beam prior to irradiation). Silicon detectors are used but present long term degradation. Wide band gap semiconductors have been proposed. Diamond is a good candidate due to its robustness and in vivo compatibility and has been proposed a long time ago [18]. More recently, high performance diamond based detectors have been reported for relative dosimetry application in clinical proton beams, both at low energy and high energy and for both stationary/scattered and scanned pencil beams. Diamond Schottky diodes [19, 20] and diamond photoconductors [21] have been successfully demonstrated. After a development phase by Tor Vergata university in Rome [19], diamond Schottky diodes are currently commercialized by the PTW Company (microDiamond 60019) for proton dosimetry (70-230 MeV). They are however limited to small sizes, well suited to dosimetry but less suited to external beam monitoring. On the contrary, GaN benefits from its large deployment for optics and electronics, and is available in large diameters and low costs, which allows fabricating larger detectors for beam monitoring. Let us add for the sake of completion that GaN has also been proposed for in vivo dose monitoring [22, 23], based on proton-luminescence. In this paper, we demonstrate for the first time that GaN Schottky diodes can be used for beam monitoring.

Compared to vertical photoconductors with ohmic contacts, Schottky diodes offer the advantage of smaller dark currents and faster transients. Compared to horizontal Metal-Semiconductor-Metal detectors, vertical Schottky diodes offer a better charge collection in a larger volume, which is a key parameter for protons with a large absorption depth. Finally, pn junction offer similar advantages to Schottky diodes, with possible trapping issues in the Mg doped region. As a result, Schottky diodes were chosen for this study. They have been fabricated on a GaN layer grown by metal organic vapour phase epitaxy. A 20 µm thick non-intentionally doped GaN layer was grown on a conductive (n-type) GaN substrate (Lumilog).

A TiAl ohmic contact was deposited on the back side first, and annealed at 750°C for 4 min. Then large Schottky contacts were deposited on the front side, with an area varying between 1 and 2 mm^2, and based on 10 nm of Pt followed by 100 nm of Au. A SiO_2 passivation layer was deposited next by Plasma Enhanced Chemical Vapor Deposition at 340°C. This deposition lasted 2 hours and also acted as an annealing step for the Schottky contact. A thick metal layer was finally deposited on the contact for facilitating the wire bonding. The final device is shown in Fig.1. Samples with many diodes were mounted on ceramic chips with electrical connections. All measurements were made at room temperature. Electrical measurements were performed with a Keithley 2410-C Sourcemeter connected to the distant device through a 30 m coaxial cable.

Diodes were tested first outside the proton beam. We refer these measurements as "in the dark", although it was in the light of the room (we checked that the diode were insensitive to the light of the room). The diodes showed a rectifying behaviour, although not very strong, with some dispersion on the dark current (I_d) among diodes, between 0.2 and 10 nA at -2V. Then the diodes were measured in the proton beam of the MEDYCIC equipment, in the Lacassagne Proton-therapy Center [24]. The isochrone cyclotron delivers proton pulses with a duration of 7 ns and a frequency of 25 MHz. The beam is mono-energetic with a proton energy of 64.8 MeV. In the present study, the device was in the air along the beam without any medium in front of it, so that the incident proton energy is 64.8 MeV, which differs from the more complex set up used for in vivo type measurements. The beam size on the detector position (upstream from the clinic treatment room) is about 1×2 cm^2. Protons are incident on the front side of the sample. Although the proton beam is pulsed, all measurements are performed in CW as pulses are not resolved in our set up. The proton current can be varied from 10 pA to 100 nA. The total detector current in the beam (I_t) was measured for various proton currents and diode biases. It was found to be reproducible from diode to diode, within a factor of 2, and to be much larger than the dark

current. The response to the proton (called protocurrent I_p, not to be confused with the proton current) is defined as the total current under the beam minus the dark current, I_t-I_d. Figure 2 shows, in log scale, the dark current and the protocurrent for one representative diode under a proton beam current of 20 nA. We first observe that the protocurrent follows the same bias dependence as the dark current. Second, at zero bias, the protocurrent is positive (0.47 μA). It changes sign between 0 and -0.1 V and then is negative for negative biases below -0.1V. Both observations indicate that the diode is not working in the normal photovoltaic mode. Indeed, the signal at zero bias should be negative, if it would originate from the Schottky depletion region only. This result can be explained as follows. The penetration depth of 64.8 MeV proton in GaN is a few mm, while the depletion region below the Schottky contact is few μm thick, the un-doped region is 20 μm thick and the substrate is 350 μm thick. Hence most of the absorption is in the doped substrate, where carriers rapidly recombine, as the field remains small even under an applied bias. We assume that the substrate contribution remains negligible except close to zero volt. A very small absorption occurs in the depletion region, leading to a small negative signal, and which should increase as the depletion region width, i.e. as $(\Phi\text{-}e\times V)^{1/2}$, where Φ is the Schottky barrier height, e is the electron charge and V is the applied bias. An intermediately large absorption occurs in the un-doped layer, where the field is close to zero at zero bias, but becomes non negligible under bias. This leads to a contribution which changes sign with bias. Secondary electron emission from the Schottky metal contact may also contribute and lead to a positive signal at zero bias. Finally, a photovoltaic effect on the back side of the sample could also be considered, leading to a positive signal at zero bias. The signal at zero bias results from all contributions, and turns out to be positive. Under an applied voltage, the contribution of the un-doped region dominates and leads to a photoconductive behaviour. Photoconductive behaviour results from the capture of either the electron or the hole while the other carrier is collected [25]. In GaN, the hole is generally trapped [26]. In order to maintain

the electrical neutrality, the device injects electrons until one electron recombines with the trapped hole. This electron injection leads to the photoconductive gain [25]. This process may happen in the bulk [26, 27] or close to the surface [28]. In other words, the Schottky diode can be modelled as a rectifying contact in series with a resistance (un-doped region). Due to processing problems, the Schottky contact is not strongly rectifying and the current is mostly limited by the resistive layer, both in the dark and under the proton beam. This leads to a photoconductive behaviour. Please note that the same diode may have a photovoltaic behaviour under UV illumination as the absorption would be in the depletion layer only. Hence, the observed photoconductive behaviour in the proton beam is largely due to the large penetration depth of protons. When turning the beam on, we observed that the protocurrent was rapidly increasing to 90% of its final value, and then increasing within a few seconds to its final value. This slow transient is typical for a photoconductive behaviour. A similar behaviour was observed during turn-off. Note that the fast transient could not be observed with a resolution better than a fraction of second, so that it may also reveal a photoconductive behavior, although with a faster component than the observed slow transient.

We will now discuss the absolute value of the protocurrent. Poly-methyl methacrylate (PMMA) is often used for proton dose calibration as its density is not too far the one of living tissues. The absorption depth in PMMA has been measured to be 29 mm. The density of GaN is 6.15 g/cm^3 while the PMMA one is 1.18g/cm^3, so that the absorption is stronger in GaN than in PMMA. The absorption depth calculated in GaN is supposed to be 8.6mm according to SRIM 2013 data tables [29]. This absorption depth is much larger than the device active region, we are in the plateau region of the energy deposition profile. Hence we can take a uniform absorption in depth so that the absorption in a GaN layer of thickness W (in μm) is W/8600. The power deposited by the beam of section S in the diode of section s is $W/8600 \times E \times I_p \times s/S$, where E is the proton energy and I_p the proton current. We then assume that protons obey the

following rule of thumb: the energy needed to create an electron hole pair is three times the gap energy, i.e. about 10 eV. The charge created per second in the diode is then $W/8600\times E\times I_p\times s/S/(3\times E_g)$. In a photovoltaic mode, without gain and with a unity collection efficiency, this charge created per second is equal to the protocurrent. Under a reverse bias of -2V, the depletion region W can be estimated to be about 4 µm. With s=1 mm^2 and S=2 cm^2, the calculation leads to a protocurrent of about 0.2µA. We have experimentally measured 100µA, which clearly shows that there is such photoconductive gain in the diode, confirming our assertion of a photoconductive behavior.

The diode response has been measured as a function of the proton beam current, from 10 pA up to 100 nA. Figure 3 shows the result (I_p) for various diode biases. We observe an excellent linearity over 5 decades.

One difficulty in the measurements was that the sample and sample holder became radioactive (^{15}O, ^{14}O, ^{68}Ge, ^{69}Ge, ^{71}Ge are the most likely produced isotopes) after exposure to the proton beam, and then impossible to handle. Hence, we tried to minimize the exposure time. In total, we can estimate that each sample remained at least 15 min in the beam, with an average current of 20 nA. This gives a cumulated dose larger than 0.5×10^{14} cm^{-2}. We sometimes observed that the dark current in reverse bias increased after a beam exposure, in particular when it was originally very small, but returned to its initial value after some time (less than one hour) or after application of a positive bias. Hence, the dark current change was mostly due to the proto-generation of charges and their subsequent trapping. No real degradation could be observed from electrical characteristics, which is a first indication of the GaN resistance to ionizing radiations. Longer-time tests remain however needed to have a qualitative idea of the radiation hardness and possible effects of radiation damage on device performance.

We have used the Schottky diodes to monitor the proton beam shape. The sample was mounted on a translation stage positioned in the center of the beam in the vertical direction

(beam size about 2 cm in the vertical direction), and was moved in the horizontal direction over 5 cm (beam size on the order of 1 cm) across the beam. The signal was recorded as a function of position, thus giving the beam shape. Two diodes separated by about 3 mm on the same sample have been used. Figure 4 shows the result recorded at -2V. The scan duration is about 1 min, which is much larger than the device response time. Hence, the device response time does not alter the profile, as demonstrated by the acquisition of the same profiles in both translation directions. The beam profile is slightly asymmetric, with a full width at half maximum of about 7.6 mm. The measured profile is the convolution of the actual beam profile and the detector width (1mm). The beam profile can be deduced from Fig.4 and is actually found to be close to 7.6mm. As an approximation, the convolution of two Gaussian profiles of width W_1 and W_2 gives a Gaussian profile with a width $W = \sqrt{W_1^2 + W_2^2}$. Hence, the beam profile is given by $\sqrt{7.6^2 - 1^2} = 7.53mm$. A similar profile was obtained at 0V but with a slightly larger width of 8.2mm. This shows that the beam profile is convoluted with a larger detector width of approximately 3mm. This may indicate that at 0V, the signal is mainly arising from the back side of the sample, which is about 3 to 4 mm wide. Finally, the beam profile was measured with a Si diode (pin BP 104F) mounted so as to detect protons from the edge of the diode, corresponding to an effective width equal to the depletion zone, id est less than 10 µm. The full width at half maximum was found to be about 7.5mm (Fig.4), with a clear asymmetry, which confirms the measurements based on GaN Schottky diodes. The peak positions measured by the two GaN diodes differ by about 3 mm, as expected from their separation on the sample. This shows that these Schottky diodes, if they are fabricated in an array, can be used for proton imaging. Typical sizes and periods for such an array of Schottky diodes would be about 100 µm for the proton-therapy application, and are easy to obtain from the technological point of view.

In conclusion, we have demonstrated that GaN Schottky diodes can be used to monitor proton beams at 64.8 MeV used for proton-therapy. Protons could be detected down to the smallest possible current, hence the sensitivity is high enough for the application. Diodes are linear in power, which is of prime importance for an accurate monitoring. They are resistant to degradation up to a dose of at least 0.5×10^{14} cm^{-2}. Time response is short enough (s) for the envisaged application. Some issues remain on elucidating the exact contribution of various parts of the device. Improvements in the processing and some changes in the epitaxial structure are likely to improve the device performance up to a commercial level. Best application of such devices fabricated in arrays is likely to be external beam monitoring.

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FIGURE CAPTIONS

FIGURE 1: Schematic of the GaN Schottky diode and the electrical circuit.

FIGURE 2: Current versus voltage for a Schottky diode in the dark (black square) and under proton irradiation (proton beam current is 20 nA) (red triangle).

FIGURE 3: Protocurrent in the Schottky diode under proton irradiation versus proton beam intensity, for various detector biases

FIGURE 4: Normalized protocurrent in two Schottky diodes, and in a Si diode as a function of the translation stage position. The currents at a bias of -2V were -5.3 and -7.3 μA for diodes A and B respectively. Proton current is 10 nA.

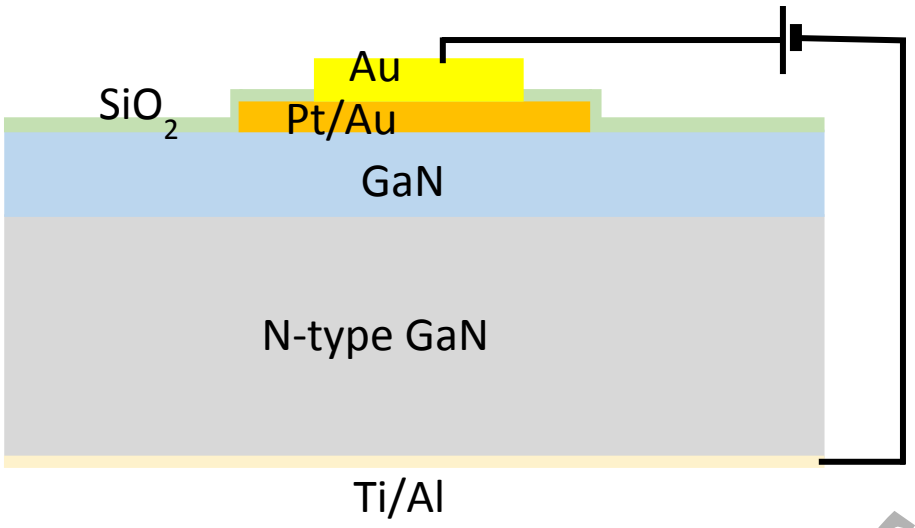

FIGURE 1

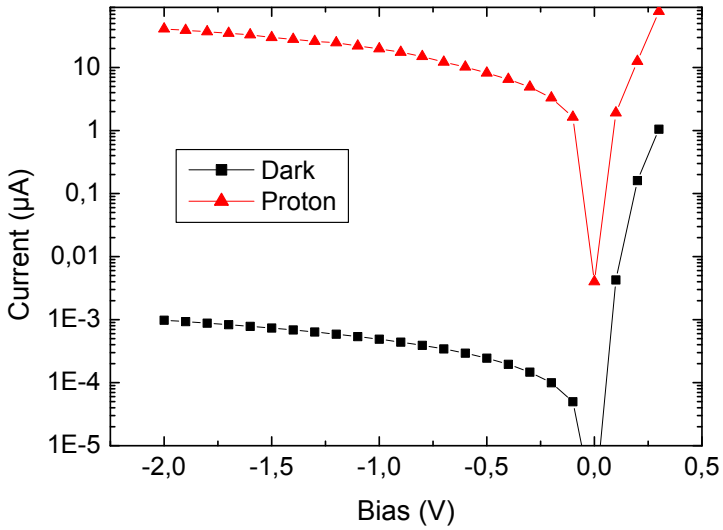

FIGURE 2

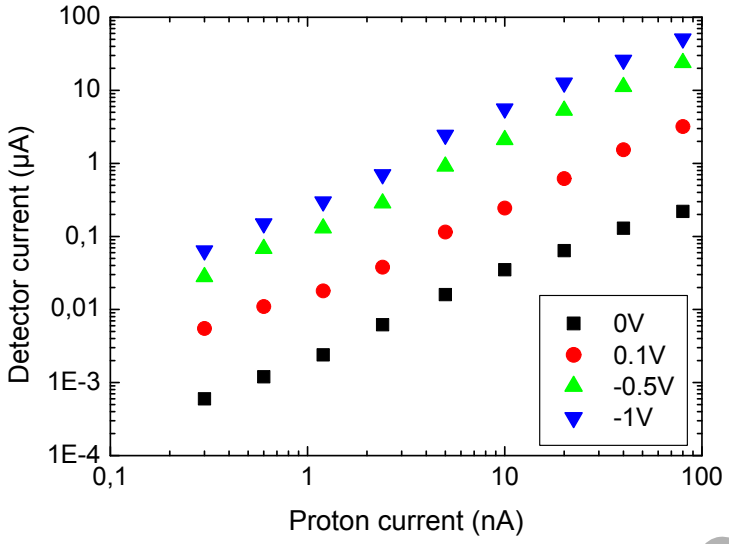

FIGURE 3

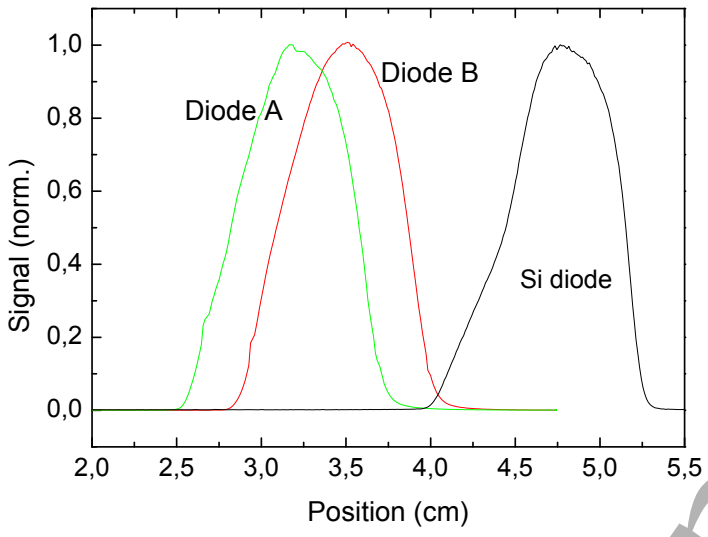

FIGURE 4