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# FORMATION OF <sup>117</sup>In-H PAIRS AFTER ANNEALING OF GaN IMPLANTED WITH $$^{117}Cd(^{117}In)$$

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#### Abstract

We report on the annealing behaviour of GaN grown on sapphire after implantation of <sup>117</sup>Cd(<sup>117</sup>In). Isochronic annealing has been performed in order to remove the implantation induced lattice damage in evacuated quartz ampoules with additional Al. The extent of the lattice damage is investigated with perturbed  $\gamma\gamma$ -angular correlation spectroscopy (PAC). The implantation damage is annealed at temperatures above 1073 K to a large extent. 82(5) % of the probes are found on the Ga-site characterized by a quadrupole coupling constant of 20.9(1) MHz. After H loading, an additional interaction appears which is attributed to <sup>117</sup>Cd(<sup>117</sup>In)-H pairs.

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#### **1** INTRODUCTION

The current research on GaN is driven by its promising applications as material for LEDs and laser diodes in the blue and UV region [1]. One aspect to be investigated is the behavior of H in GaN. During different steps of device processing, like crystal growth or chemical etching, H can be introduced into the material unintentionally. It may change the electrical properties by saturating dangling bonds or passivating dopants.

Therefore the understanding of the behavior of H in GaN is essential for optimization of different processing steps. We have reported on studies of Cd-H pairs in GaN with perturbed  $\gamma\gamma$ -angular correlation spectroscopy (PAC) recently [2]. Implanted <sup>111m</sup>Cd substitutes the Ga-site and can act as an acceptor after annealing of the implantation damage. H is implanted with low energy and trapped by Cd. Cd-H pairs have been characterized [2] and should be formed with <sup>117</sup>Cd, too. H will be released after the nuclear disintegration <sup>117</sup>Cd  $\rightarrow$  <sup>117</sup>In because In is isoelectronic to the host element Ga. The initially captured H may then leave the probe and starts to diffuse. This onset of diffusion shall be investigated with PAC. A prerequisite is the annealing of the implantation induced lattice damage. This has been studied in detail for samples implanted with <sup>111m</sup>Cd. After treatment of the crystal at 1323 K under N<sub>2</sub>-atmosphere, 60 % of the probes substitute an undisturbed lattice site [3]. At higher temperatures GaN degrades. The possibility to reach temperatures up to 1573 K by adding a small amount of Al to the sample has been found recently [4]. We report on experiments that use this new technique to study the annealing of GaN after implantation of <sup>117</sup>Cd(<sup>117</sup>In).

## 2 EXPERIMENTAL

GaN grown on sapphire via MOVPE (Cree Research) has been implanted with <sup>117</sup>Ag that decays to <sup>117</sup>Cd with a half-live of 73 s at the on-line mass separator ISOLDE at CERN (60 keV,  $1.2 \times 10^{12}$  cm<sup>-2</sup>). The samples were annealed for 10 min in sealed quartz ampoules in vacuum together with elementary Al up to 1573 K. The PAC technique [5] is sensitive to electric field gradients (EFG) present at the site of the probe atom <sup>117</sup>Cd(<sup>117</sup>In). An EFG causes a hyperfine splitting of an excited state of the <sup>117</sup>In nuclei. The EFG is mainly described by the quadrupole coupling constant  $v_Q = eQV_{zz}/h$  (Q - nuclear quadrupole moment,  $V_{zz}$  - largest component of diagonalized EFG tensor). This quantity is measured by PAC and unique both for a lattice site in defect free material and for special defect complexes that include the probe atom.

The fraction of probe atoms involved in this complex can be determined from the characteristic modulation of the PAC spectrum R(t). A small damping of the observed modulation due to the superposition of slightly different EFG caused by defects relatively far away from the probe atom is described by the width  $\Delta v_Q$  assuming a Lorentzian distribution of these EFG. The PAC-measurements have been carried out at ambient temperature.

## **3 RESULTS AND DISCUSSION**

Fig. 1 shows the fraction of <sup>117</sup>Cd(<sup>117</sup>In) atoms at identical lattice sites in GaN and the width of the distribution of the quadrupole coupling constant  $\Delta v_Q$  in dependence on the annealing temperature. After thermal treatment at 773 K, 51(1) % of the probes are exposed to an unique EFG



**Figure. 1:** Fraction f of <sup>117</sup>Cd(<sup>117</sup>In) atoms at identical lattice sites in GaN (left) and width of the distribution of the quadrupole coupling constant  $\Delta v_Q$  (right) in dependence on the annealing temperature T<sub>A</sub>. The samples where annealed in evacuated quartz ampoules with additional Al for 10 min.

determined by the non-cubic arrangement of the host atoms in Wurtzite structure GaN. The fraction increases with rising annealing temperatures to 82(5) % at 1073 K and remains mainly constant to the final temperature of 1558 K. Thus, about 80 % of the probe atoms are at sites without defects in the nearest neighborhood. Nevertheless, there are defects further away that are not directly correlated with the probe atoms. These are responsible for the distribution of EFG represented by  $\Delta v_Q$ . The remaining 18(5) % of probes experience correlated defects in their nearest neighborhood creating strong non-unique EFG.

The quadrupole coupling constant after annealing at 1073 K has a value of  $v_Q = 20.9(1)$  MHz. The comparison of this value with the EFG determined with <sup>71</sup>Ga at the Ga-site with help of NMR [6] proves once more that Cd substitutes the Ga-site. The width of the distribution  $\Delta v_Q$  of the quadrupole coupling constant decreases steeply between 773 K and 973 K. At higher temperatures one observes a spread of the values. Nevertheless,  $\Delta v_Q$  exhibits the tendency to become smaller until the final temperature of 1573 K is reached.

Summarizing the slope of the fraction of probes and that of the distribution of the quadrupole coupling constant, we get the following result: Annealing at temperatures above 1073 K does not brake the bonds between probes and the remaining correlated defects in their nearest neighborhood. On the other hand, the density of uncorrelated defects can be decreased by annealing at higher temperatures. The annealing of GaN under N<sub>2</sub>-atmosphere after implantation of <sup>117m</sup>Cd shows similar behavior. The saturation of the fraction of 60 % is already reached after annealing at 900 K [3]%.

We have implanted H into annealed samples. Fig. 2 shows the PAC spectrum of <sup>117</sup>Cd(<sup>117</sup>In) in GaN recorded at 10 K after annealing at 1523 K and low energy implantation of H (300 eV,  $1 \times 10^{15}$  cm<sup>-2</sup>, 423 K). Besides the dominating interaction of the probe atoms in undisturbed environments, a fraction of 6(2) % is exposed to an additional EFG with  $v_Q = 344(8)$  MHz. This interaction is caused by free H atoms that are close to the <sup>117</sup>In probe atoms after the nuclear disintegration. The fact that the fraction is so small is caused by the incompleteness of the annealing. Both the correlated and uncorrelated defects that influence the PAC spectrum can be electronically active. They may provide a high density of traps for H with higher trapping probability than our dopant <sup>117</sup>Cd. Furthermore these defects may cause an incomplete electrical



**Figure. 2:** PAC spectrum of  ${}^{117}$ Cd( ${}^{117}$ In) in GaN after additional implantation of H. The measurement has been carried out at 10 K. Besides the slow modulation caused by the EFG of the Wurtzite lattice, a high frequency is visible which represents 6(2) % of the probe atoms and will be attributed to a  ${}^{117}$ Cd( ${}^{117}$ In)-H pair.

activation of the probe atoms and the observed H-related fraction reflects the electrically active dopants.

Our results demonstrate that it is not sufficient to increase the annealing temperature in order to create a high fraction of <sup>117</sup>Cd-H pairs for the study of H-diffusion in GaN. Another approach could be either to protect the GaN surface during annealing or perform the annealing under high external N pressure.

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