High dose Fe implantation of GaN: damage build-up and dopant redistribution

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Abstract. Undoped GaN epilayers implanted at room temperature with 50 – 325 keV Fe+ ions in the fluence range of $10^{15} – 10^{17}$ ions/cm² are studied by a combination of Rutherford backscattering/channeling spectrometry and time-of-flight elastic recoil detection analysis. Results show that for high ion fluences ($>$1×$10^{16}$ cm⁻²) enhanced Fe concentration closer to the surface is observed. The Fe redistribution towards the surface increases as the ion fluence increases. Our findings are attributed to radiation enhanced diffusion during ion implantation and increasing of Fe diffusivity in the implantation-induced amorphous phase near the surface.

1. Introduction
Implantation of GaN with transition metals, such as Fe, has attracted significant research interest in recent years [1-3]. A reason for such interest is that Fe doped GaN is a potential diluted magnetic semiconductor with a Curie temperature above room temperature (RT) [1]. Note that it is difficult to achieve highly doped samples during epitaxial growth without forming a secondary phase as magnetic ions typically have a low solubility in AIIIBVs semiconductors. Ion implantation provides an alternative way to introduce high concentration of magnetic ions in a semiconductor matrix.

However, it is well known, that bombardment by energetic ions inevitably produces radiation defects which often undesirably affect almost all properties of the material. It has been found that the ion beam-induced processes in GaN indicate strong dynamic annealing processes and a significant influence of the surface on defect accumulation [4-6]. Damage processes in GaN are also complicated by ion-beam-induced material dissociation at high ion fluences, where amorphization of GaN occurs [4,7].

Another major problem is dopant redistribution during ion implantation and subsequent annealing processes of GaN. Despite that numerous dopants in GaN exhibit excellent thermal stability up to 600 – 900 °C [8], a prediction of the final depth profile of implanted ions is very complicated. Furthermore, experimental results on ion ranges in GaN obtained by different research groups with different experimental techniques are somewhat contradictory [9-11]. This implies that more detailed studies are urgently needed to clarify the different effects of implantation conditions and damage build-up on the dopant distribution in GaN.

In this paper we have studied dopant redistribution in high fluence Fe implanted GaN. Our results
show that for heavily implanted GaN the Fe distribution in the near surface region strongly deviates from theoretically predicted. The possible reason for Fe pile-up near the surface may be an increased Fe diffusivity in the implantation-induced surface amorphous layer with disturbed stoichiometry.

2. Experimental
The wurtzite undoped (0001) GaN epilayers were grown on sapphire substrates by metal-organic vapor phase epitaxy (MOVPE). Implantation with 50-325 keV Fe\(^+\) ions was done at RT with fluences in the range of \(10^{15} - 10^{17}\) ions/cm\(^2\). During implantation, samples were tilted by 7\(^\circ\) relative to the incident ion beam to avoid channeling. The average scanned ion beam flux was kept constant at \(-3\times10^{12}\) cm\(^2\) s\(^{-1}\). Such a low beam flux was chosen to prevent heating of the samples during ion implantation.

Ion-beam produced structural disorder was characterized by Rutherford backscattering/channeling spectrometry (RBS/C) [12] with 2.0 MeV \(^4\)He\(^+\) ions incident along the [0001] direction and backscattered into a detector at 167\(^\circ\) relative to the incident beam direction. This geometry was used to provide enhanced mass resolution to separate signals from Fe and Ga atoms needed to accurately examine damage build-up near the surface in the Ga sublattice.

Time-of-flight elastic recoil detection analysis (ToF-ERDA) [13] using a 40 MeV \(^{127}\)I ion beam was applied to measure the depth distributions of implanted ions and main elemental depth profiles. The incoming ion beam and outgoing recoils impinged/exitd at 67.5\(^\circ\) relative to the sample surface normal, respectively, giving a recoil angle of 45\(^\circ\).

Both RBS/C and ToF-ERDA measurements were carried out using the 5 MV Pelletron accelerator at the National Tandem Accelerator Facility in Uppsala, Sweden.

3. Results and discussion
Fig. 1 shows RBS/C spectra of GaN irradiated at room temperature with 50 keV and 325 keV Fe\(^+\) ions at different ion fluences. Implantation fluences (in ions/cm\(^2\)) are indicated in the figure. The position of Fe atoms at the surface corresponds to channel 428 as indicated by arrows.

Figure 1. The random and channelling RBS spectra of GaN bombarded at RT with (a) 50 keV and (b) 325 keV Fe\(^+\) ions. Implantation fluences (in ions/cm\(^2\)) are indicated in the figure. The position of Fe atoms at the surface corresponds to channel 428 as indicated by arrows.

to different ion fluences. It is seen from Fig. 1b that for 325 keV Fe\(^+\) implantation the damage accumulates both in the crystal bulk, where the nuclear energy loss is maximum, and at the surface. Previous studies have shown that the GaN surface act as a nucleation site for amorphization and the surface damage peak in RBS/C spectra is due to an amorphous layer [4]. Fig. 1b also shows that bulk damage peak does not reach the random level even at highest fluence used, that indicates that sample are not fully amorphous in this region. Our estimations reveal that the damage in the bulk exhibits saturation at 45-50 % and moves deeper with increasing ion fluence. For the 50 keV Fe\(^+\) implantation.
regime (see Fig. 1a) the bulk and surface damage peaks are not clearly resolved. However, detailed analysis of the RBS/C data reveals the same tendencies in damage accumulation as for the 325 keV Fe\(^{+}\) implantation. Such damage accumulation behaviour is consistent with previous studies of GaN bombarded with intermediate to heavy mass ions at RT [4]. It should be noted that the increase of the RBS yield close to the 425 and 400 channels for 50 and 325 keV, respectively, are attributed to Fe atoms, for which the concentration reaches several at. % for the highest fluences used.

Another interesting feature seen from Fig. 1 is that both the height and the shape of the random spectra change with increasing ion fluence. In particular, as seen from Fig. 1, near the surface, the height of the spectra of GaN bombarded by 50 keV Fe\(^{+}\) ions with the highest fluence used \((2\times10^{16}\text{ cm}^{-2})\) is lower compared to the random spectrum of the unimplanted sample. This effect is even better illustrated by Fig. 1b, where a large dip is clearly seen near the 100 nm Ga depth for the random spectra of GaN bombarded by 325 keV Fe\(^{+}\) ions with a fluence of \(6.5\times10^{16}\text{ cm}^{-2}\). Such a decrease of RBS yield is due to a reduced Ga concentration in the region where the implanted atoms are located.

The changes in Ga concentration are also supported by the ToF-ERDA results as clearly seen from Fig. 2, which shows the depth profiles of Ga, N and Fe atoms in GaN implanted with 325 keV Fe\(^{+}\) ions. Interestingly to note that the N concentration is not changed in the region where Fe atoms are located in contrast to the Ga concentration. This observation was somewhat unexpected. However, at the surface a significant nitrogen loss is observed, while the Ga concentration is rather constant. This observation strongly indicates and supports the known [4,7] fact that material decomposition and nitrogen loss occur in GaN near the surface under high fluence irradiation.

The Fe depth profiles obtained by ToF-ERDA are shown in Fig. 3. It is seen that for the low ion fluences (up to \(1\times10^{16}\text{ cm}^{-2}\)) the measured implantation profiles of Fe are in good agreement with simulations using TRIM code [14] (version SRIM 2006). However, for the higher fluences the shape of the Fe concentration depth profile disagrees with the theoretically predicted. Indeed as seen in Fig.
3, an enhanced Fe concentration is observed close to the surface for ion fluences $> 1 \times 10^{16} \text{ cm}^{-2}$ for both ion energies and this pile-up of Fe near the surface increases as the ion fluence increases. Detailed analysis of RBS/C and ToF-ERDA data (represented in Figs. 1 and 3) reveals that the depth where significant Fe redistribution towards the surface occurs is comparable with the thickness of the implantation induced surface amorphous layer. This indicates that the Fe redistribution can be attributed to radiation enhanced diffusion during ion implantation and an increase of the Fe diffusivity in the amorphous phase near the surface, where the stoichiometry is disturbed. Such an increase of dopant diffusivity in an amorphous phase was also discussed to explain the near surface pile-up of Au atoms observed in Au implanted GaN [11].

Finally it should be noted that for high ion fluences the position of the maximum Fe concentration also deviates from the one predicted by TRIM simulations, as illustrated in Fig. 3. This effect can be attributed to a density reduction for very high-fluence implanted GaN (for the energy-depth conversion we use the material density value of 6.1 g/cm$^3$ which is typical for crystalline GaN). The resulting change in the energy loss of the analysing ions used in ERDA as well as the recoiling atoms would affect the depth scale.

### 4. Conclusions

Our results show that for all ion energies the damage in the bulk moves deeper and exhibits saturation with increasing ion fluence. Significant stoichiometric changes and material decomposition occurs in GaN for high-fluence Fe implantation. For low ion fluences (up to $1 \times 10^{16} \text{ cm}^{-2}$) the measured implantation profiles of Fe in the as-implanted samples are in good agreement with simulations using the TRIM code. However, for higher ion fluences we observe enhanced Fe concentration closer to the surface, and the Fe concentration increases as the ion fluence increases. This Fe redistribution can be attributed to radiation enhanced diffusion during ion implantation and an increased Fe diffusivity in the amorphous phase near the surface.

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