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In-vacancies in Si-doped InN

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The introduction of vacancy type point defects by Si doping in InN grown by plasma-assisted molecular beam epitaxy was studied using a monoenergetic positron beam. With the combination of positron lifetime and Doppler broadening measurements, compensating In-vacancy (V_{In}) acceptors were identified in the material. For increasing Si doping an enhanced formation of V_{In} defects was observed, up to a concentration of $c_V = 7 \times 10^{17}\,cm^{-3}$ in the highest doped sample ($n_e = 6.6 \times 10^{20}\,cm^{-3}$). A strong inhomogeneity of the defect profile with a significant increase of the V_{In} concentration toward the layer/substrate interface could be detected. Additionally, larger vacancy clusters containing several V_{In} are formed in the proximity of the interface.

1 Introduction Despite a remarkable progress in crystal quality over the past years InN still exhibits considerably high defect concentrations and the fundamental understanding of the physics of defects in the material is of crucial importance in order to gain full control over the materials properties [1]. Due to a large lattice mismatch between InN and sapphire, which is commonly used as a substrate, a high concentration of strain relieving extended defects is formed especially at the interface [2]. InN has a strong affinity for the formation of donor type defects, leading to high electron concentrations and strong n-type conductivity. According to first-principle calculations [3] Si_{In} and O_N donors possess the lowest point defect formation energy in n-type InN and among intrinsic defects, N and In vacancies and vacancy complexes are created most favorably [3–6]. Nevertheless, the formation energies of intrinsic defects are predicted to be high compared to donor impurities.

Positron annihilation spectroscopy is a powerful method for the investigation of defects in semiconductors. Using a monoenergetic positron beam, positrons can be implanted into a defined depth of the sample. After the positron has thermalized it diffuses through the crystal and can get trapped at neutral and negatively charged open volume defects. Once the positron has annihilated with a crystal electron, the 0.511 MeV annihilation radiation can be used to get information about physical properties at the respective annihilation site. An analysis of the positron lifetime and the Doppler broadening of the annihilation radiation reveals the identity of the trapping defect and allows to determine its concentration. In previous studies of both MBE (molecular beam epitaxy) and MOVPE-grown InN using positron annihilation spectroscopy, negative V_{In} and vacancy clusters could be identified in the material [7, 8]. The vacancy concentration has been shown to depend highly on the layer thickness and structural quality of the material, rather than growth conditions and thermodynamic effects [9]. Studies of He-irradiated InN additionally proved that V_{In} provides one source of compensation in n-type material, although interstitial nitrogen may possibly be the dominating compensating center in the case of irradiated material [10].

2 Experimental details Two sets of InN samples with Si concentration of $1 \times 10^{18} - 6.6 \times 10^{20}\,cm^{-3}$ have

Table 1 Free electron concentration, mobility, and V_{In} concentration of the investigated selection of Si-doped InN samples. Increasing numbers in the sample ID signify an increase in Si doping. V_{In} concentrations were estimated from positron annihilation measurements.

samples	n_e (cm^{-3})	μ_e (cm^2/Vs)	V_{In} (cm^{-3})
Si-a1	1×10^{18}	1850	1×10^{16}
Si-a2	2×10^{18}	1850	$< 1 \times 10^{16}$
Si-a3	1.2×10^{19}	450	$< 1 \times 10^{16}$
Si-a4	4.8×10^{19}	100	1×10^{16}
Si-b1	4.5×10^{19}	600	2×10^{16}
Si-b2	1.3×10^{20}	150	1×10^{17}
Si-b3	4.0×10^{20}	80	2×10^{17}
Si-b4	6.6×10^{20}	38	7×10^{17}

been investigated (see Table 1). All samples have been grown by plasma-assisted molecular beam epitaxy (PAMBE) on c-plane Al_2O_3 substrates with a GaN buffer layer of 200 nm nominal width. For details of the growth of the samples labeled Si-a_x and Si-b_x, please see Refs. [11, 12], respectively. Carrier concentrations and mobilities were determined using single-field Hall measurements.

Positron lifetime spectra have been measured using a pulsed positron beam. For each spectrum 4×10^6 counts have been recorded and the peak-to-background ratio was better than 10^4. The overall time resolution was 270 ps and lifetimes of down to 100 ps can be resolved by using decomposition techniques.

Doppler broadening measurements were performed using a monoenergetic slow positron beam. The positron implantation energy was varied from 0 to 38 keV and the Doppler broadening of the annihilation radiation was recorded with two Ge detectors, with an energy resolution of 1.24 eV at 0.511 MeV. The data was analyzed using the conventional S and W parameters, that represent the fraction of annihilations with low momentum electrons ($|p_L| < 0.4$ a.u.) and high momentum electrons (1.5 a.u. $< |p_L| < 0.4$ a.u.), respectively. More information about the experimental setup can be found elsewhere [13].

3 Results and discussion

Doppler broadening spectra have been measured at room temperature to investigate the role of open volume point defects in Si-doped InN. Figure 1 shows the S and W parameters as a function of the positron implantation energy, which corresponds to a mean implantation depth in the material. At low implantation energies most positrons annihilate at the surface of the sample. For energies above 3–4 keV the surface effects become negligible and the S and W parameters represent the characteristic value for the InN layer. It can be seen from Fig. 1 that for all samples the S (W) parameter of the layer is higher (lower) than the measured InN reference [8] (red dotted line), where positrons are not trapped by vacancy defects and annihilate in the defect-free state. At a vacancy

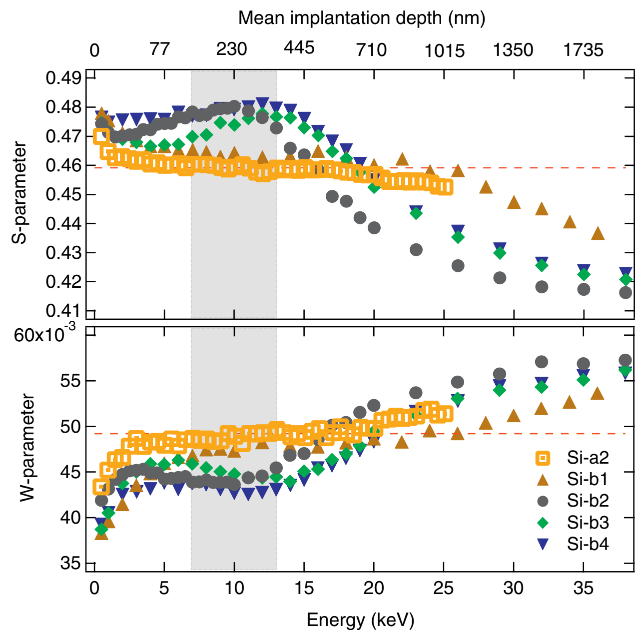

Figure 1 (online color at: www.pss-a.com) S and W parameters of various Si-doped InN samples as a function of the positron implantation energy, which corresponds to a mean implantation (and hence information) depth in the material. The red dotted lines indicate the measured S and W parameters of the InN lattice for a reference sample.

the electron momentum distribution is narrower compared to the (vacancy) defect-free state. This leads in the annihilation spectrum to an increase (decrease) of the S (W) parameter compared to the defect-free situation. Hence, the presence of vacancy type defects is clearly visible in all the measured layers. Additionally, the vacancy signal increases strongly in samples with higher electron concentration, i.e., enhanced Si doping. A linearity analysis of the annihilation parameters measured in different samples proves that in all the samples the dominant type of positron trapping defect is similar. For implantation energies between 4 and 13 keV a strong profile of the annihilation parameters is observable in the samples with high Si concentrations. The S (W) parameter increases (decreases) with increasing implantation energies and reaches a maximum (minimum) for around 10–13 keV at the InN/GaN/sapphire interface. The interface is visible here as a turning point in the annihilation spectra, when positrons are implanted at a mean depth of around half of the layer thickness and at the same time start to annihilate in the substrate [14].

Positron lifetime measurements have been performed in order to get further information about the identity of the dominant positron trapping defects in the samples and investigate the origin of the strong profile in the Doppler spectra. Figure 2 shows the average lifetime of sample Si-b3 for implantation energies between 7 and 13 keV (for comparison, see shaded area in Fig. 1). The average lifetime shows a similar behavior as observed for the Doppler

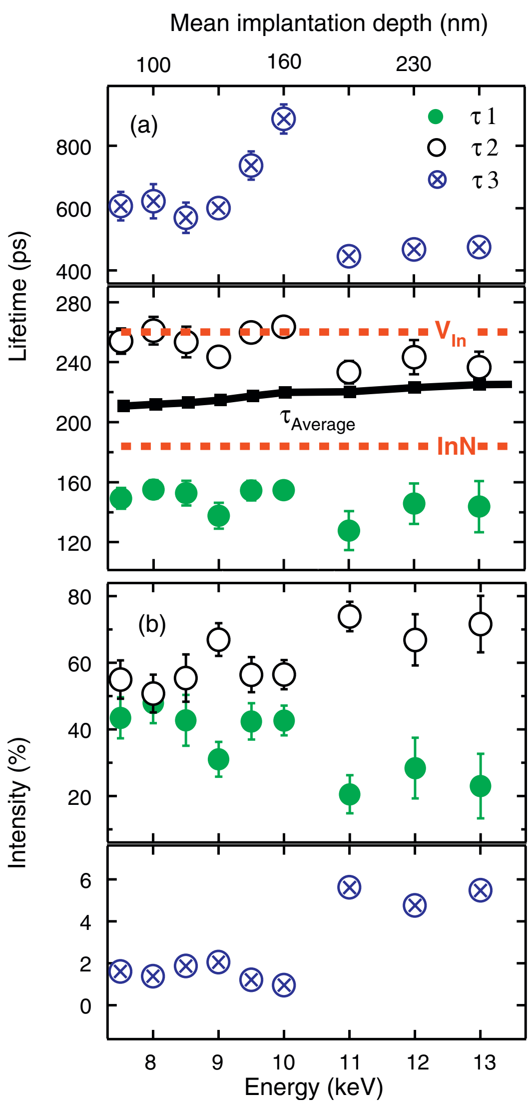

Figure 2 (online color at: www.pss-a.com) Average positron lifetime of sample Si-b3 as well as decomposed lifetime components (a) and their respective intensities (b) as a function of the positron acceleration energy.

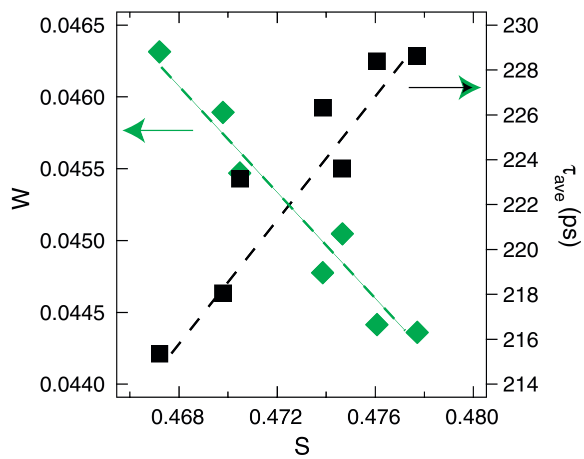

Figure 3 (online color at: www.pss-a.com) The S and W parameters and the average positron lifetime of sample Si-b3 in the layer specific region. The linearity of W, S, and τ_{ave} proves the presence of only one dominant vacancy trap in the sample.

parameters and increases toward the interface from 215 to 230 ps. The spectrum can be decomposed into three lifetime components τ_1, τ_2, and τ_3 (Fig. 2a) with respective intensities (Fig. 2b). In earlier studies the positron lifetime in the InN lattice and the V_{In} have been determined as 184 and 260 ps, respectively [7]. The lifetime component τ_2 can be clearly identified as coming from V_{In}, while τ_1 is shorter than τ_b and hence represents annihilations in the InN lattice. The third lifetime component τ_3 has a mean lifetime above 400 ps, which is a clear footprint of vacancy clusters with higher open volume and lower electron density compared to the single V_{In}. The large scatter in τ_3 below 10 keV and its comparably low intensity suggest that in fact there are no vacancy clusters in this region, while the increase of I_3 to 6% is a clear indication of the presence of such a lifetime component in the interface area of the spectra. Nevertheless, the increase of I_3 is too small to be solely responsible for the observed increase of the average lifetime. A much bigger contribution originates from the intensity increase of I_2, which grows from around 55 to 75% at the interface. This means that an increasing concentration of V_{In} can be identified as the major cause of the observed profile in the average lifetime and also Doppler broadening. This is additionally reflected by the relationship of the layer annihilation parameters τ_{ave}, S, and W (see Fig. 3) between 7 and 13 keV. All parameters show a linear dependence and prove that the vast majority of positrons are trapped by only one dominant vacancy type defect, that increases in concentration with increasing implantation depth.

The combination of both results from the positron lifetime and Doppler broadening measurements leads to the following final picture. The pure or decorated V_{In} can be identified as the dominant defect type in the samples from the point of view of positrons. The V_{In} concentration in the layer increases strongly, when approaching the interface. Additionally, a small contribution from clusters of several In vacancies (and presumably a similar number of N vacancies) is observed at the interface. It has to be noted that our results do not distinguish between the situation of a gradual increase in the vacancy concentration and a two-layer model with a fixed concentration at the layer and the interface. Additionally, talking of V_{In} does not exclude the possibility of a decoration of the vacancy with other defects. A decoration is only visible as a minor effect in the spectra and the exact chemical identity of the V_{In} defect could not be identified so far. Nevertheless, the decrease of τ_2 at the interface possibly suggests a (different) decoration of the vacancy near the interface.

Based on the results from lifetime and Doppler broadening measurements, it is possible to estimate the V_{In} concentration in the investigated layers. In the standard positron trapping model, the vacancy concentration can be estimated from Ref. [13]

$$c_{\mathrm{V}} = \frac{N_{\mathrm{at}}}{\mu_{\mathrm{V}}\tau_{\mathrm{b}}}\frac{S - S_{\mathrm{B}}}{S_{\mathrm{V}} - S}, \quad (1)$$

where μ_{V} is the positron trapping coefficient, N_{at} the atomic density of InN, and τ_{b} the positron lifetime in vacancy-free InN [7]. S_{V} represents the characteristic S parameter for the V_{In} that has been determined in earlier studies with $S_{\mathrm{V}} = 1.049 \times S_{\mathrm{B}}$ [10]. The resulting V_{In} concentrations are shown in Table 1 and based on experiments performed at temperatures from 100 K–300 K, also taking into account the effect of positron trapping at negative ions [15]. The given concentrations have to be regarded as a lower limit until experiments above room temperature are performed and the full effect of additional acceptors in the samples is determined. For low doped samples the V_{In} concentration remains close to the detection limit, i.e., $1 \times 10^{16}\,\mathrm{cm}^{-3}$. Starting from carrier concentrations of mid-$10^{19}\,\mathrm{cm}^{-3}$ the vacancy concentration increases significantly up to $7 \times 10^{17}\,\mathrm{cm}^{-3}$ in the highest doped sample. This behavior can be understood well from V_{In} acting as a compensating defect in the material, trying to prevent an increase of the free electron concentration above the value that is defined by the Fermi-stabilization energy [16]. Nevertheless, it is important to note that the experimentally determined vacancy concentrations are orders of magnitudes higher than what could be expected based on first-principle calculations of the formation energy of V_{In} in InN [3]. It is possible that strain and a high concentration of extended defects could promote the formation of vacancies, which is especially true for the InN/GaN/sapphire interface.

4 Summary We have used positron annihilation spectroscopy to study the effects of Si doping on the introduction of vacancy type defects in InN grown by PAMBE. V_{In} could be identified as the dominant positron trapping defect in the material, with concentrations up to $7 \times 10^{17}\,\mathrm{cm}^{-3}$. Additionally, a small contribution from vacancy clusters containing several V_{In} could be observed at the interface. The vacancy concentration shows a strong increase toward the sample/substrate interface and is of orders of magnitudes higher than the value that could be expected based on first-principle calculations of the formation energy of V_{In} in InN.

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