Cu diffusion-induced vacancy-like defects in freestanding GaN

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Abstract. Positron annihilation spectroscopy was employed to elucidate the nature and thermal behavior of defects induced by Cu in freestanding GaN crystals. Cu atoms were intentionally introduced in GaN lattice through thermally activated diffusion from an ultrathin Cu capping layer. During isochronal annealing of the obtained Cu-doped GaN in the temperature range of 450–850 K, vacancy clusters were found to form, grow and finally vanish. Doppler broadening measurements demonstrate the presence of vacancy-like defects across the 600 nm-thick layer below the surface corresponding to the Cu-diffused layer as evidenced by secondary ion mass spectrometry. A more qualitative characterization of these defects was accomplished by positron lifetime measurements. We found that annealing at 450 K triggers the formation of divacancies, whereas further increase of the annealing temperature up to 550 K leads to the formation of large clusters of about 60 vacancies. Our observations

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suggest that the formation of these vacancy-like defects in bulk GaN is related to the out-diffusion of Cu.

1. Introduction

For about two decades gallium nitride (GaN) has attracted a great deal of attention due to its advantageous optical, thermal and electrical properties that can be exploited to implement a variety of novel or superior electronic and optoelectronic devices [1]. In particular, its wide and direct band gap of 3.4 eV enables the fabrication of unique devices such as blue laser diodes opening up new applications in optoelectronics, data storage and biophotonics [2]. These diodes are typically made of high-quality In-rich InGaN quantum wells grown on low dislocation density bulk or freestanding GaN (fs-GaN) substrates. The fs-GaN wafers needed for the fabrication of these emerging devices are currently produced by hydride vapor phase epitaxy (HVPE) at elevated temperatures as described in [3]. From a fundamental standpoint, exploring and elucidating the formation of defects and their behavior is of paramount importance for understanding the properties of GaN-based systems [4]–[14]. Theoretical calculations predict that Ga vacancies and related complexes are the most dominant point defects in n-type GaN [11]–[13]. These calculations show that both isolated and complexed Ga vacancies are plentifully generated in n-type GaN, whereas the dominating native defect in p-type GaN is the N vacancy. Moreover, it was demonstrated that Ga vacancies are electrically active behaving as acceptors, and are involved in the optical transition causing the emission of yellow luminescence light [11, 14]. In addition to the impact on understanding the basic properties of GaN, the elucidation of defect formation in GaN crystals is also crucial to optimize and control the emerging fabrication processes of advanced hybrid substrates [15].

Also of fundamental as well as practical importance is understanding the behavior of impurities and their role in shaping the electrical and structural properties of GaN. Copper (Cu) is among the impurities in GaN that has recently sparked a surge of interest because of the associated room temperature (RT) ferromagnetism, which could create new opportunities in GaN-based spintronics [16]–[22]. The extent of this Cu-induced ferromagnetism is, however, still under debate [17]. A possible role of vacancy-like defects was postulated as a key factor that should be considered for addressing this intriguing effect [19, 20]. In addition to intentional doping, the presence of Cu is also encountered during semiconductor device growth and processing [23]. Recently, the thermal stability of Cu gate AlGaN/GaN high electron mobility transistors was investigated [24, 25]. These reports do not show any obvious degradation in the device performance upon thermal annealing up to 500 °C. Moreover, no Cu diffusion up to this temperature is observed at the Cu and AlGaN interface as demonstrated by secondary ion

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mass spectroscopy (SIMS) [24, 25]. This illustrates that the Cu diffusivity in GaN is very low compared to other semiconductors such as GaAs [26, 27].

This work addresses the formation of vacancy-like defects and their thermal behavior upon diffusion of Cu in light-emitting diode quality fs-GaN crystals. Unlike GaAs and Si (see for example [27] and references therein), the influence of Cu on defect formation has never been investigated in GaN heretofore in spite of the crucial information it could provide regarding the fundamental properties of GaN. In the present study, we employed positron annihilation spectroscopy (PAS), which is an established non-destructive technique for the investigation of defects in semiconductors [28]. PAS can be used to detect vacancy-type defects, e.g. monovacancies and divacancies, with concentration above $10^{14}–10^{15} \text{cm}^{-3}$. In this technique, positrons are trapped in an open-volume defect (e.g. vacancies) due to the missing positive ion core (nucleus) at that site. The trapping can be experimentally observed either as an increase in the positron lifetime or as a narrowing of the Doppler-broadened 511 keV $\gamma$-annihilation peak. Vacancy-type defects in group III-nitride were previously investigated using this technique [29]–[37]. Those studies have demonstrated that PAS can clearly distinguish between vacancies and vacancy–impurity complexes. Recent lifetime PAS studies of H-implanted and annealed fs-GaN under the ion-cut conditions have provided evidence of the formation of positronium besides the detection of monovacancies, divacancies and vacancy clusters [38]. In the present study, the lifetime PAS probe of Cu-diffused GaN crystals shows that annealing is associated with the appearance of a relatively long-lifetime component attributed to vacancy clusters, whereas a small change in the average lifetime ($\tau_{\text{av}}$) is detected. To characterize the involved open-volume defects, we employed variable energy Doppler broadening spectroscopy. The thermal behavior of the observed vacancy-like defects suggests that their formation is related to Cu out-diffusion.

2. Experimental details

The investigated samples were cut from nominally undoped 300 $\mu$m-thick double side polished high-purity 2 inch fs-GaN wafers. The samples show $n$-type conductivity with a free electron concentration of about $N_e = 2 \times 10^{18} \text{cm}^{-3}$ at 300 K. This doping is most probably caused by the residual oxygen. The resistivity of initial material was about $10^6 \Omega \text{cm}$. The samples were covered by an 18 nm-thick Cu layer evaporated under ultrahigh vacuum (UHV) conditions. The deposited layer thickness was controlled by the frequency shift of a crystal oscillator, which was previously calibrated by atomic force microscopy. High-purity Cu-free quartz ampoules were employed for the annealing to induce the diffusion of Cu into fs-GaN samples. The samples were sealed in the quartz ampoules under argon. The annealing was performed for 96 h at 873 K. As demonstrated below, Cu diffuses into GaN lattice (in-diffusion) during this process. After annealing, the ampoules were quenched into water at RT. Cu introduced into the crystal is now, at this low temperature, expected to be oversaturated. Thus, the Cu atoms have the propensity to leave the lattice (e.g. forming precipitations) and hence start the out-diffusion. To study this process, the samples were isochronally annealed at different temperatures up to 850 K and slowly cooled down after each annealing step. Between the annealing steps, positron annihilation lifetime spectroscopy (PALS) measurements were carried out in the temperature range of 30–500 K using a conventional fast–fast coincidence system with a time resolution of 220 ps. A 30 $\mu$Ci $^{22}$Na positron source was sandwiched between two identical 1.5 $\mu$m-thick Al foils and placed between two identical samples. Typically about $4 \times 10^6$
annihilation events were collected in each positron lifetime spectrum. The lifetime spectrum \( n(t) = \sum_i I_i \exp(-t/\tau_i) \) was analyzed by a lifetime LT program [39] as a sum of exponential decay components convoluted with the Gaussian resolution function of the spectrometer after source and background correction. The positron in state \( i \) annihilates with a lifetime \( \tau_i \) with an intensity \( I_i \). The state in question can be either a delocalized state in the lattice or a localized state at a vacancy-type defect. An average lifetime above the defect-free bulk lifetime is indicative of the presence of vacancy-like defects in the material. This parameter can be experimentally determined with high accuracy and even as small a change as 1 ps in its value can be reliably measured. The analysis is insensitive to the decomposition procedure used. The trapping of positron in an open-volume defect is observed as an increase in \( \tau_{av} \). A vacancy-like defect can be identified by the characteristic lifetime component in the lifetime spectrum.

Depth-profiled Doppler broadening measurements of the as-grown and Cu-diffused GaN (GaN:Cu) samples were carried out using a variable energy positron beam to study the near-surface region. Monoenergetic positrons were produced by a 10 mCi \(^{22}\)Na source assembled in transmission with a 1 \( \mu \)m monocrystalline tungsten moderator and transported in a magnetic guidance system to the sample. The beam spot is characterized with a diameter of 4 mm and an intensity of \( 10^2 e^+ / s \). The samples were measured at RT under UHV. A high-purity Ge detector with an energy resolution of \( (1.09 \pm 0.01) \) keV at 511 keV was used to record the \( \gamma \)-annihilation spectra. The data were processed with a digital peak-stabilizing system integrated in the multi-channel analyser. A spectrum of about \( 5 \times 10^5 \) counts in the 511 keV peak is accumulated at each energy \( E \). The Doppler broadening spectrum of the 511 keV annihilation line is characterized by two parameters \( S \) and \( W \). The low momentum parameter \( S \) is defined as the fraction of counts in the central area of the peak divided by the total number of counts under the whole area. The high momentum parameter \( W \) is the fraction of counts in the wing areas of the peak, arising from the annihilations with high momentum core electrons. The trapping of positrons at open-volume defects is detected as an increase in \( S \) parameter and a decrease in \( W \) parameter. Using positron energies in the range of 0.03–35 keV it is possible to scan the GaN sample to a mean penetration depth of \( \sim 2 \mu \)m below the surface. At a fixed positron energy \( E \), the \( S \) parameter is described as \( S(E) = \eta_s(E)S_s + \eta_b(E)S_b + \sum_i \eta_d(E)S_{d,i} \), where \( S_s \) and \( S_b \) are the characteristic values of \( S \) parameter for positron annihilation at the surface and in defect-free bulk GaN lattice, respectively. \( \eta \) stands for the fraction of positron annihilations at each state. In the case of a sample containing defects that can trap positrons, the third term in the equation above should be taken into account. Here \( S_{d,i} \) characterizes the positron annihilations in a defect \( i \).

Elemental depth profiles were performed using time-of-flight SIMS (ToF-SIMS) using the Iontof TOF.SIMS 5 apparatus. The Cu depth profiles in GaN were acquired in positive ion polarity with the \( \text{O}_2^+ \) sputter source (energy of 2 keV) with a measuring area of 150 \( \times \) 150 \( \mu \)m\(^2 \) and a sputter area of 250 \( \times \) 250 \( \mu \)m\(^2 \) each time. An electron flood gun was used for charge compensation. Furthermore, the investigations were carried out in the non-interlaced mode for better peak separation and smaller charging of the sample surface. In this operation mode, the pulses of the sputter and analysis guns are used separately. The measured intensity (in counts \( s^{-1} \)) is a qualitative concentration value due to the different ionization probabilities of a certain element in the investigated matrix system (matrix effect). The ToF-SIMS time scale corresponds to the depth in dependence on the abrasion rate. The calibration of the depth scale was performed by using a profilometer.

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3. Results and discussion

SIMS is carried out on the as-quenched Cu-diffused samples. The results are displayed in figure 1. As is clearly seen in the figure, a considerable amount of Cu is detected in the region extending to $\sim 600$ nm below the surface. Figure 2 displays the positron lifetime spectra recorded for virgin fs-GaN and Cu-diffused fs-GaN and annealed at 550 K. In a positron lifetime experiment, positrons injected into the sample are thermalized at time $t = 0$. The vertical axis represents the number of positron–electron annihilation events at a time channel of 26 ps. In the reference sample, only a single exponential decay component with a lifetime of 154 ps is detected. This corresponds to the annihilation of positrons in the delocalized state of the nearly defect-free GaN lattice. This value is in good agreement with earlier experimental and theoretical studies [40, 41]. The annealing at 550 K of the Cu-diffused GaN sample induces an increase in the average positron lifetime by 4 ps. This increase is a direct evidence of
open-volume defects introduced in the sample presumably during the out-diffusion of Cu. Note that this increase is comparable to earlier observations of 2 and 5 ps increase in the average lifetime in GaN after electron irradiation at fluences of $3 \times 10^{17}$ and $1 \times 10^{18}$ cm$^{-2}$, respectively [29].

After Cu in-diffusion, the as-quenched sample showed an average lifetime very close to that of the as-grown one. The average positron lifetime as a function of the measurement temperature carried out after annealing of Cu-diffused GaN is shown in figure 3. The average lifetime was found to increase slightly by increasing the annealing temperature up to 550 K, indicating the creation of vacancy-type defects. With further increase of the annealing temperature, a decrease in the average positron lifetime was observed. A plausible reason for this decrease would be that at high temperatures vacancies migrate to the surface and anneal out, which decreases their concentration and hence the available positron traps. Additionally, the observed decrease can also be attributed to the agglomeration of vacancies to form clusters and the distance between them becomes longer than the positron diffusion length. Thus they become invisible for positrons. The same behavior was observed during Cu diffusion in the semi-insulating GaAs [27]. Nevertheless, the increase in the average lifetime, $\Delta \tau = \tau_{av} - \tau_b$, was found to be very large in GaAs, 61 ps.

Figure 4 represents the behavior of the average lifetime measured after different annealing temperatures. It is found that the $\tau_{av}$ measured at 333 K increases with increasing annealing temperature up to 550 K and then decreases upon annealing at higher temperatures. A defect-related lifetime ($\tau_d$) of 298 ps with an intensity of $1.9 \pm 0.1\%$ is observed at 450 K. This implies that positrons annihilate from a localized state at a vacancy-like defect. The open volume of the N vacancy ($V_N$) is much too small to explain the observed long lifetime of 298 ps, where the theoretically calculated lifetime associated with $V_N$ is 169 ps [14]. The observed 298 ps is also higher than that corresponding to the Ga vacancy ($V_{Ga}$), 235 ps [14, 29]. It can be inferred that the detected defects are possibly divacancies ($V_{Ga}$–$V_N$), which is consistent with the earlier assignment of 260–282 ps to this complex [38]. $\tau_d$ increases with increasing the annealing temperature to reach 433 ps at 550 K and then decreases further. Its intensity is

Figure 3. Average positron lifetime measured for fs-GaN samples at different temperatures. The samples are isochronally annealed. The temperature-dependent lifetime experiment was carried out after each annealing step as indicated in the figure.
Figure 4. Average positron lifetime of Cu-diffused fs-GaN samples after the isochronal annealing. The spectra were measured at a sample temperature of 333 K.

Figure 5. Average positron lifetime of the virgin fs-GaN sample as a function of the measurement temperature. The sample was annealed for 96 h at 873 K without deposition of Cu-cap layer. The sample is isochronally annealed at different temperatures. A reference sample before annealing is also shown for comparison.

also small, $1.5 \pm 0.04\%$. Note that this value of $\tau_d$ is much larger than that of the divacancy mentioned above. Thus it can be associated with the formation of larger vacancy clusters at this temperature. In fact, the lifetime of 433 ps corresponds to a cluster of 60 vacancies (30 GaN molecules are missing) according to calculations based on local density approximation for electron–positron correlation effects and the atomic superposition method [41]. $\tau_1$ is, however, found to be around the bulk lifetime.

To demonstrate that the observed vacancy-like defects are Cu related, an fs-GaN sample was annealed under similar conditions but without Cu treatment. The obtained lifetime results for this sample are shown in figure 5. As demonstrated in the figure and independently of annealing, the as-quenched sample shows an average lifetime very close to that of the reference
sample, which is 154.5 ps at RT. This behavior stands in sharp contrast to the data of Cu-diffused GaN samples (figure 3). In Cu-free samples, the average lifetime decreases on increasing the temperature (figure 5), but the change in the average lifetime remains below 1 ps over the entire temperature range. This indicates the presence of very low density defects. The formation of vacancies can be reasonably associated with the out-diffusion of Cu during the isochronal annealing of the sample. The out-diffusion process means the annealing of the Cu pre-introduced GaN samples in conditions without a Cu source, where the sample surface is not covered by any capping material. The diffusion mechanism is most likely the interstitial-substitutional mechanism (kick-out mechanism) [42]. In the case of the Frank–Turnbull mechanism [43], the diffusing atom does not replace a lattice atom at its site, but gets trapped in a vacancy. Thus, for the Frank–Turnbull mechanism the material must contain a large number of vacancies, as large as the number of Cu atoms to be placed into the lattice. Our positron lifetime investigations performed on initial (virgin) GaN material and Cu-diffused GaN in the as-quenched state did not show the presence of vacancies. This supports the suggestion of the occurrence of the kick-out mechanism. Nevertheless, we believe that more detailed studies are required for confirming the diffusion mechanism governing this process. In the kick-out mechanism [42], the interstitial-substitutional exchange can be described as 
\[ i_{\text{Cu}} \leftrightarrow \text{Cu}_{\text{Ga}} + I_{\text{Ga}} \],
where \( i_{\text{Cu}} \) stands for Cu atoms on interstitial sites, \( \text{Cu}_{\text{Ga}} \) for Cu on Ga sublattices and \( I_{\text{Ga}} \) for Ga self-interstitials. During the out-diffusion process upon thermal annealing, Cu atoms are likely to diffuse to possible sinks (surfaces, loops or dislocations) leaving behind Ga vacancies in the matrix, \( \text{Cu}_{\text{Ga}} \leftrightarrow V_{\text{Ga}} + \text{Cu}_{\text{sink}} \). These are most likely negatively charged vacancies.

Doppler broadening measurements are described next. Figure 6 illustrates both \( S \) and \( W \) parameters measured as a function of the incident positron energy at RT in the virgin substrate and Cu-diffused GaN samples annealed up to 550 K, which shows the highest variation in lifetime data. \( S \) and \( W \) parameters are normalized to the corresponding values in bulk defect-free GaN, \( S_b \) and \( W_b \), respectively. As the positron energy increases, the \( S \) parameter in the virgin sample decreases from the surface-specific value towards a constant value (\( S/S_b \approx 1 \)) in the energy range 20–35 keV characteristic of bulk GaN. Conversely, the \( W \) parameter increases from the surface state to the bulk state. As mentioned above, the positron lifetime data for the virgin sample yield a single component lifetime of 154 ps. Thus, the Doppler broadening parameters recorded for this sample characterize the positron annihilation in defect-free GaN lattice. It is clearly shown that the annealed Cu-diffused GaN sample exhibits higher \( S(E) \) and lower \( W(E) \) curves than that of the reference in the positron energy range up to 16 keV (mean depth \( \sim 560 \text{ nm} \)). This behavior can be explained by the presence of open-volume defects in this layer. An increase of 6% in \( S \) at the positron energy of 3 keV is observed. This can be ascribed to the appearance of vacancy clusters [44, 45]. At positron energies higher than 16 keV, both \( S \) and \( W \) are almost the same as in the virgin sample. The inset in the lower panel of figure 6 presents the difference between the \( S \) parameter of the Cu-diffused sample annealed at 550 K and that of the virgin sample. It is evident that there is a correlation between the behavior of \( \Delta S \) and that of the Cu profile measured by SIMS (figure 1).

The number of different vacancy-type positron traps in the material can be identified by investigating the scaling relationship between \( S \) and \( W \) parameters. This is based on the fact that both \( S \) and \( W \) are sensitive to the defect concentration and defect types. If only a single type of vacancy is present, these parameters should vary linearly as a function of each other. Thus, the \( S–W \) linearity predominates only when the defect concentration (i.e. the fraction of positrons annihilating in the defect) changes [46, 47]. For instance, \( V_{\text{Ga}} \) was observed in

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Figure 6. Normalized Doppler broadening parameters as a function of the incident positron energy measured for the virgin and Cu-diffused GaN samples annealed up to 550 K. The low momentum parameter $S$ is shown in the lower panel and $W$ in the upper panel. The positron mean penetration depth is shown in the top axis. The inset of the lower panel displays the difference in $S$ parameter between the Cu-diffused and virgin samples. The mean penetration depth $\zeta$ is calculated from the positron energy ($E$) using $\zeta = E^n a / \rho$. $a$ and $n$ are empirical parameters; their values are $4 \mu g cm^{-2} keV^{-n}$ and 1.6, respectively [28]. $\rho (g cm^{-3})$ is the density of GaN.

GaN [14, 48], where all the $(S, W)$ points were found to be on the same line in the $S$–$W$ plane. If more than one defect type is responsible for positron trapping, a deviation from the $S$–$W$ linear behavior would take place. The defect-free virgin sample shows only two positron annihilation states in the $S$–$W$ plot, one at the surface and the other in the bulk. This means that a straight line is obtained between the surface and the bulk. In the case of Cu-diffused GaN, there are three distinct contributions to the annihilation process: annihilation in the Cu-treated region, annihilation at the surface and annihilation in the bulk beyond the Cu-treated region. $S$ parameter is plotted versus $W$ parameter in figure 7. It emerges from this plot that the observed $S(E)$ and $W(E)$ curves in Cu-diffused GaN are due to the appearance of a defect type with larger open volume, since two different straight lines are found. The positron lifetime experiments on this sample demonstrate a relatively large component of 433 ps. Thus, one can conclude that the increase of $S$ and the decrease of $W$ correspond to vacancy clusters. Combining the results of SIMS and positron beam experiments, one can deduce that the defects are observed in the same layer where Cu is present. Therefore, it is reasonable to assume that Cu is responsible for the detected defects perhaps during its out-diffusion as suggested above. The observation of Cu at a depth of 600 nm after annealing for 96 h at 873 K shows a very low diffusion coefficient of Cu in...
GaN in contrast to other semiconductors, e.g. Si, Ge and GaAs. However, the mean penetration depth of positrons in GaN is \( \sim 40 \mu \text{m} \) [28], which is much larger than the 600 nm-thick Cu-diffused layer that contains the formed vacancy-like defects. This may explain the small intensity of the observed defect-related lifetime component, as mentioned above. Thus, the small effect observed in the lifetime is more likely due to the fact that the positrons penetrate much deeper than 600 nm, where only 1.5% of positrons are stopped. It is reasonable to assume that the value of \( \tau_1 \) is almost \( \tau_b \), where most of the positrons, 98.5%, are annihilating beyond the Cu-diffused layer, i.e. in the defect-free region. The defect concentration is roughly estimated to be higher than \( 10^{18} \) cm\(^{-3} \). Note that no significant amount of Cu was observed after annealing at the highest temperature as shown in figure 8 displaying the SIMS profiles of the Cu-diffused GaN sample after annealing at 850 K, which is in apparent contrast to the as-quenched sample that shows the existence of Cu up to 600 nm (figure 1). This difference between the Cu profiles measured by SIMS in the as-quenched sample and that isochronally annealed at 850 K provides clear evidence of Cu out-diffusion. This measurement supports our suggestion that Cu out-diffusion may create vacancy-like defects that can be observed by positron spectroscopy.

**Figure 7.** Plot of \( W \) versus \( S \) for the slow-positron beam measurement of the virgin and Cu-diffused GaN samples. The data were obtained from figure 6.

**Figure 8.** SIMS depth profiles of the Cu-diffused GaN sample after annealing at 850 K. Note that surface peaks are artifacts of SIMS measurements.
4. Conclusion

In summary, we have presented a first-of-its-kind study of Cu diffusion-induced vacancy-like defects in bulk GaN. We have applied PAS and secondary ion mass spectrometry for investigating defects generated during thermal treatment of Cu-doped fs-GaN samples. The average lifetime increases slowly during the subsequent post-annealing up to 550 K, suggestive of the creation of vacancy-type defects. At this annealing temperature, we observed a low-intensity and long-lifetime component of 433 ps attributed to vacancy clusters. With further increase of the annealing temperature, a decrease in the average positron lifetime was detected. This can be ascribed to defect healing or to larger vacancy clusters separated from each other by a distance longer than the positron diffusion length. The effect of Cu was evidenced by probing GaN samples annealed under similar conditions but without Cu treatment. In the absence of Cu, the average lifetime is found to decrease slightly (≤ 1 ps) on increasing the annealing temperature. The slow positron beam measurements showed that the vacancy clusters extend to a depth of 600 nm, which coincides with the Cu profile as shown by SIMS, which supports our claim that these defects are related to Cu out-diffusion. Unveiling the exact mechanistic picture of this phenomenon would, however, require extensive calculations to elucidate the atomic processes underlying the formation of vacancies and vacancy clusters on thermal treatment of Cu-treated GaN.

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