1. Introduction

Due to its excellent electronic and optical properties, considerable research and development efforts have been, over the last decade, paid to exploring the potential of GaN for optoelectronic applications [1]. These efforts include improving growth technologies for the realization of high crystalline quality GaN, which can be used in high-brightness light emitting diodes, short wavelength laser diodes, UV and x-ray detectors, high electron mobility transistors, microwave radio frequency power amplifiers, etc [2]. Because of the high price of free-standing GaN wafers, the growth of epitaxial layers for device fabrication is mostly performed on Al₂O₃ and SiC. Due to the poor structural and thermal match between GaN and the substrate, thread dislocations are frequently formed in the epitaxial layers. These defects formed during epitaxial layer fabrication would deteriorate properties of GaN. The ‘smart-cut’ technology was firstly mentioned by Bruel [3]. This technology comprises H implantation into Si, then bonding them to a substrate stiffener. Finally, a splitting treatment is used to achieve layer transfer upon annealing at 673 K. The ‘smart-cut’ technology can provide templates with high structural quality. This technology has been widely used for fabrication of silicon-on-insulator (SOI) materials. On the basis of this technology, the free-standing wafer of GaN can be utilized to transfer multiple layers onto other foreign substrates such as Si, SiC, Ge, GaAs, InP, etc. Implantation dose, implantation...
temperature, and annealing temperature play important roles in the splitting process [4–7]. All these parameters depend strongly on the type of semiconductor used for splitting. As for the ion-cut technology of GaN, there are only a few studies of H implantation-induced GaN layer splitting [8–10]. Kucheyev et al [10] reported the H implantation-induced exfoliation in GaN. The exfoliation is observed in a narrow window of the implantation dose above 3 × 10^{17} \text{cm}^{-2}, which is more than one order of magnitude as high as the threshold dose of 2–3 × 10^{16} \text{cm}^{-2} for the appearance of exfoliation of Si and SiC [4]. Why is the threshold dose needed for exfoliation of GaN so high? Until now there has been no uniform explanation [8, 9]. The dependence of the H implantation-induced lattice disorder on implantation dose and temperature has been less investigated. Understanding the nature and formation of the defect clusters produced in H implanted-GaN will contribute to the interpretation of ion-cut mechanism of GaN.

In the present work, we have investigated the H-implantation-induced defects as a function of the implantation dose and temperature.

2. Experimental process

n-type wurtzite GaN layers 30 \mu m thick, epitaxially grown on a sapphire (0001) substrate by the metal organic chemical vapor deposition (MOCVD) technique were used for the experiment. GaN samples of 10 × 10 \text{mm}^2 in size were cut from a 2 in wafer and were carefully cleaned in alcohol solution before H implantation. The samples were implanted with \text{H}_2^+ ions with a kinetic energy of 134 keV to doses ranging from 3.75 × 10^{16} to 1.75 × 10^{17} \text{H}_2^+ \text{cm}^{-2} at room temperature (RT) in a chamber with a vacuum at 10^{-2} \text{mbar} in the ECR-320 KV High-voltage Platform in the Institute of Modern Physics, Chinese Academy of Sciences. The ion flux during irradiation was of the order of 10^{13} \text{ions cm}^{-2} \text{s}^{-1}. According to SRIM-96 simulation (the displacement energies of Ga = 20.5 eV and N = 10.8 eV, and density 6.1 g cm\(^{-3}\)), the mean projected range \(R_p\), the straggling \(\Delta R_p\), and the position of maximal damage from the surface are 440, 140, and 390 nm, respectively [11]. To investigate the effect of implantation temperature on structural damage, GaN samples were implanted to the same dose of 1.5 × 10^{17} \text{H}_2^+ \text{cm}^{-2} at three temperatures, RT, 573 and 723 K.

The implanted and unimplanted samples were analyzed by Rutherford backscattering spectrometry in a channeling geometry (RBS/C) using 2.022 MeV \text{He}^+ ions and a backscattering angle of 165°. The depth \(d\) is derived by the stopping power data according to SRIM-96 [12].

\[ d = (kE_0 - E_i)(kS_\text{th}/\cos \theta_1 + S_\text{out}/\cos \theta_2) \]  

The relative disorder was simply taken as a proportion

\[ n_d = \left[ Y - Y_c \right]/\left[ Y_c \right] \]  

where \(k\) is the kinetic factor for the element of interest, \(E_0\) is the incident ion energy, and \(E_i\) is the energy detected for the ion backscattered from the element at depth \(d\). The parameters \(\theta_1\) and \(\theta_2\) are the angles between the sample normal direction and the direction of the incident beam and scattered particles, respectively. \(S_\text{th}\) and \(S_\text{out}\) are ion stopping powers along the inward and outward paths in the materials, respectively, which depend on the ion energy. \(Y\) and \(Y_c\) are the ion backscattered yield in the channel for H-implanted GaN, virgin GaN and in random for virgin GaN, respectively. In the experiment, \(E_0 = \) 2.022 MeV; \(k = \) 0.798; \(\theta_1 = \) 0°; and \(\theta_2 = \) 165°.

The RBS/C measurements were carried out at the State Key Laboratory of Nuclear Physics and Technology, Peking University. The Raman spectrometry was carried out with a JY-HR80 spectrometer, in the backscattering configuration. In the experiment, the Raman spectra were obtained by using the 514.5 nm line of the Ar+ laser as the excitation source with a spectral resolution of ~0.5 cm\(^{-1}\). The laser beam was focused up to a spot size of 1 \mu m using a microscopic lens system. The scattered light was detected by a Jobin Yvon T64000 spectrometer with a LN\(_2\) cooled charge coupled device. The exciting wavelength should be able to penetrate the entire thickness of the GaN film [13]. The UV–visible optical transmittance spectrometer was performed using a Lambda 900 UV/VIS/NIR Spectroscope produced by Perkin Elmer Inc. Wolfram and deuterium lamps were used as incident light. The spectra resolution was 2 nm and the transmittance spectra were recorded in the range 200 to 850 nm wavelength region. To investigate the defect distribution with the sample depth, cross-sectional TEM samples were prepared. The samples were cut, glued and then thinned using mechanical polishing and final Ar-ion milling at RT. Cross-sectional TEM (XTEM) images were taken at 200 kV with a Tecnai G20 microscope equipped with a double tilt goniometer stage. XTEM samples were prepared by mechanical thinning up to approximately 40 \mu m in thickness and then ion milling with Ar ions by two steps. At the first step, ion milling energy was 5 kV with a glancing angle of ±5° until optically controlled perforation occurred in the middle of the XTEM sample. At the second step, ion milling energy was 2 kV with a glancing angle of ±3° for 1 h to minimize radiation damage induced by the Ar ions. The bubbles were imaged in under-focused and over-focused conditions to highlight the bubble edges with Fresnel contrast. The observed images of H implantation-induced stacking faults and dislocation loops were taken from cross sections of GaN along a \{01-10\} zone axis. The stacking faults and dislocation loops observed in the damaged layer were identified on cross sectional high resolution images taken along a \{11–20\} zone axis. The damaged layer was examined in conventional bright-filed (BF) mode and weak-beam dark-field (WBDF) mode with the image condition: \((g, 5g) = g = 0002\) near \(c = 11–20\). \(S\) in the TEM denotes the implanted surface of the sample.

3. Results and discussion

3.1. Rutherford backscattering-channeling spectrometry

The RBS/C technique gives a unique insight of the depth distribution of defects produced in the near surface layer of the samples. The RBS spectra were carried out along the \(<0001>\) direction of the \text{H}_2^+\)-implanted samples. Figure 1(a) illustrates the channeled spectra of the backscattered ions from
the H$_2$-implanted samples. In comparison, the random and channelled spectra of the un-implanted sample were also presented. Because Rutherford backscattering spectrometry along channels is sensitive to very small atomic displacements from the crystalline lattice sites and the main interaction is with interstitial-like defects, the emergence of a damage peak in the channeling spectra indicates the presence of interstitial-like defects in the lattice. The evaluation of the damage profile is more precise if one uses the Ga single due to the overlap of the surface N signal with the Ga bulk signal and the lower backscattering cross section of N. Figure 1(a) illustrates that there is an evident damage peak in the channeling spectra at the channel number of 315 for the samples implanted at RT and the relative Ga disorder at the damage peak is obtained using equation (2), as plotted in figure 1(b). The channel number of 315 corresponds to a depth of 380 nm determined using equation (1), which indicates that the peak damage induced by 134 keV H$_2^+$ implantation is located at the depth of 380 nm. It is consistent with the location of the maximum nuclear deposited energy simulated by SRIM-96. Therefore, the results imply that the H implantation-induced damage formation is determined by the energy deposition in nuclear processes. When the sample was implanted at 573 K, there is a damage peak in the channeling spectra at the channel number of 305, corresponding to a depth of 420 nm. A possible explanation for the observed behavior is that dynamic annealing causes the recovery of lattice defects at peak damage, while the recovery of lattice defects at the maximum H concentration is inhibited due to numerous H$_2$ bubbles in this region, consistent with the XTEM observations shown later.

Note that the change of the damage formation process at the damage peak as a function of the implantation dose can be simply divided into three steps. At the first stage of damage formation for displacements per atom (dpa) ≤ 0.5, the relative Ga disorder at the peak region increased slightly up to 0.05, which is consistent with the result of Wendler et al [13]. Because GaN shows very efficient dynamic annealing behavior due to the high diffusivity of the point defects in the bulk GaN lattice, in the first stage evident recombination effects within the primary collision cascades, the value of the relative Ga disorder at the peak region is very low. At the second stage of damage formation for doses between 3.75 × 10$^{16}$ and 5 × 10$^{16}$ H$_2^+$ cm$^{-2}$, corresponding to dpa between 0.5 and 0.72, the relative Ga disorder at the peak region increased rapidly up to 0.6. In this stage, we regard that the local defect concentration exceeds a critical value and, therefore, the types of lattice defects have changed from point defects to planar dislocation loops and stacking faults. These defects reduced the recombination efficiency, and thus the defect concentration increased rapidly with increasing dose. At the third stage of damage formation for doses between 5 × 10$^{16}$ and 1.75 × 10$^{17}$ H$_2^+$ cm$^{-2}$, the relative Ga disorder at the peak region increased slightly exhibiting a plateau-like behavior. The saturation value is about 0.75. In this stage, we regard that the types of lattice defects have not changed as compared to defects observed in the second stage. The microstructural evolution with implantation dose was investigated by XTEM and the results are discussed later.

It should be noted that three steps of damage formation in GaN implanted with Ar ions were reported by Wendler et al [14]. The authors regarded that at the first stage the defect concentration increases slightly with increasing ion doses corresponding to 0.18 to 1.3 dpa. There is a saturation value of 0.05 of the relative Ga disorder at the damage peak. At the second stage the defect concentration increases rapidly with increasing ion doses above 1 × 10$^{15}$ cm$^{-2}$ corresponding to dpa ≥ 2. The relative Ga disorder at the damage peak saturates at 0.7. During the third stage, the nucleation of amorphous zones is formed and these zones grow with increasing dose. The present results demonstrate that the evolution of defect concentration with implantation damage in the Ar-implanted GaN and H-implanted GaN is slightly different. On the basis of the defect density within the single collision cascades [11], the primary collision cascades are denser for the Ar ions and this can cause a more efficient defect agglomeration, resulting in an increment of the defect concentration at lower values of dpa. The present result is contrary to this explanation. The concentration of implanted ions can explain the present result. Because the dose for obtaining the same dpa for H ions is two orders of magnitude higher than Ar ions, therefore a high concentration, H reduces the recombination efficiency of lattice defects. It has been generally recognized that the impurity atoms can prevent the recovery of lattice defects in SiC [15]. Moreover, the chemical effect should be considered. H can react with N to form N–H in the H-implanted GaN, as reported by Mountainabir et al [8]. The dangling bonds of vacancies and vacancy clusters decorated by H atoms prevent the recombination of these vacancy-type defects with interstitial-type defects.

The effect of the implantation temperature on defect concentration produced by H implantation was also investigated. The evolution of the relative Ga disorder at the damage peak with the implantation temperature was shown in figure 1(c). The value of the relative Ga disorder at the damage peak decreased with increasing implantation temperature. This phenomenon is attributed to the temperature dependence of defect migration and agglomeration. Dynamic annealing becomes evident at elevated temperatures. One can see that it is a linear change of relative Ga disorder at the damage peak with the implantation temperature. It is consistent with previous reports [16].

3.2. Raman scattering using a 514.5 nm excitation

The Raman spectra of the GaN samples before and after 100 keV H$_2^+$ implantation to doses between 3.75 × 10$^{16}$ and 1.75 × 10$^{17}$ H$_2^+$ cm$^{-2}$ are shown in figure 2(a). The spectra covered a wave number range from 100 to 800 cm$^{-1}$ and were recorded in the z(x,x)z backscattering configuration with the z direction along the hexagonal c axis. The frequencies of the folded transversal and longitudinal phonon modes for the acoustic (FTA) and optical (FTO) branches of the dispersion curves of GaN have been investigated by Harima [17]. In the Raman spectrum of the as-grown sample, three sharp
lines were observed at 144, 570 and 736 cm\(^{-1}\). The 144 and 570 cm\(^{-1}\) lines are assigned to \(E_{2}^{\text{lo}}\) and \(E_{2}^{\text{hi}}\), respectively. The \(E_{2}\) modes are attributed to atomic movements normal to the \(c\)-axis in the Ga sublattice (\(E_{2}^{\text{lo}}\)) and in the N sublattice (\(E_{2}^{\text{hi}}\)). The 736 cm\(^{-1}\) line is assigned to the \(A_1\) (LO) vibration mode. Besides the three sharp bands, one weak band was also observed at 418 cm\(^{-1}\), which stems from the sapphire substrate [17]. One can see that the background intensity increased with increasing dose. It is caused by Rayleigh scattering from the implantation-induced defects. The energy positions of these features in the Raman spectra were obtained by fitting Lorentzian type line profiles to the experimental data. The spectrum of the \(E_{2}^{\text{hi}}\) phonon at 570 cm\(^{-1}\) monitored the crystalline quality of the sample because this band is very sharp, as shown in figure 2(a). It has a similar change of the \(E_2\) signal in the H-implanted GaN at elevated temperature. The integrated intensity \(A\) of \(E_2\) was normalized to the value \(A_{\text{cryst}}\) of the crystalline material, i.e. \(A_{\text{norm}} = A/A_{\text{cryst}}\). The value of \(A_{\text{norm}}\) decreased, while its linewidth increased with increasing dose. A saturated value of 0.14 at around 1.3 dpa was observed from the simulated curve, as shown in figure 2(b). Similarly, the value of \(A_{\text{norm}}\) increased exponentially with implantation temperature. The data were fitted by a function of an Arrhenius-type giving activation energy of 0.085 eV, as shown in figure 2(c). The presence of the weak \(E_2\) mode and the broad \(E_2\) mode in the spectra can be explained by disorder effects. Because of the phonon Raman scattering process, the wave vector selection rule and the polarization selection rule are strictly kept in perfect crystals. It can account for why sharp phonon Raman bands with different frequencies and wave vectors are observed in crystals. The implantation-induced defects produce a loss of the periodicity, reduction of the crystal symmetry and a topological disorder of crystals and therefore, they induce a breakdown of the selection rules for the wave vector, a reduction of the phonon lifetime and a relaxation of momentum conservation in the Raman scattering process. The reduction of the phonon lifetime and a relaxation of momentum conservation cause a broadening of phonon Raman bands. Stacking faults are typical defects in GaN crystals, and these defects lead to the partial breakdown of the wave vector selection rule, which causes the activation of phonon modes with wave vectors different from \(q = 0\), and thereby gives rise to an asymmetric band shape. As a consequence of the deterioration in the crystallinity of the
implanted layer, the absorption coefficient increases owing to accumulation of absorbing centers and the Raman polarizability decreases owing to breaking of bonds. Moreover, the changes in atomic forces and displacements lead to a decrease in intensity of sharp first-order Raman lines arising from the damaged layer. The change of the absorption coefficient in the H-implanted GaN was measured by UV–visible transmittance spectroscopy and the results are discussed later. Besides the change of $E_{2}$ signal, an asymmetric band shape of $A_{1}$ (LO) was clearly observed in the implanted sample. Katsikini et al. [18] observed a similar phenomenon of the asymmetric broad $A_{1}$ (LO) peak in the In-implanted GaN and they concluded the loss of periodicity and the large defect density have detrimental effects on the carrier transport and relax the plasmon-phonon coupling of LO modes.

After implantation to doses between $3.75 \times 10^{16}$ and $5 \times 10^{16}$ H$_2^+$ cm$^{-2}$, new weak peaks appeared at 300, 420 and 670 cm$^{-1}$. These spectra features were well reproduced on implanting other species, i.e. O$^+$, Ar$^+$, Xe$^+$ and Ca$^+$ [19, 20]. Thus, the signals are interpreted as defect-induced modes. The damaged GaN crystals can exhibit a different type of defect modes called the disorder-activated Raman scattering modes (DARS) [21]. A broad peak at 300 cm$^{-1}$ has been attributed to DARS of the highest acoustic-phonon branch and/or to scattering from vacancies in the Ga sublattice [18]. As for the two new peaks at 420 and 670 cm$^{-1}$, they have been attributed to scattering from vacancy-type defects most probably in the N sublattice [22]. It should be noted that the intensities of the peaks at 300, 420 and 670 cm$^{-1}$ decreased with increasing dose and nearly disappeared for doses in excess of $1.5 \times 10^{17}$ H$_2^+$ cm$^{-2}$. The increase of absorption coefficient with increasing dose causes a decrease in intensity of Raman scattering. Thus, these new modes caused by DARS are not clearly visible in a highly damaged sample.

3.3. UV–visible transmittance spectroscopy

The optical property of H-implanted GaN was investigated by optical transmittance in the wavelength range from 200 to 850 nm. The transmittance curves of GaN wafers implanted with 134 keV H$_2^+$ ions to doses of $3.75 \times 10^{16}$ to $1.75 \times 10^{17}$ H$_2^+$ cm$^{-2}$ are shown in figure 3(a), together with the spectrum of the un-implanted sample. The transmittance of the un-implanted sample closed to zero at $\lambda = 366$ nm, corresponding
to the bandgap of crystalline GaN, in agreement with previous reports [23]. The transmittance rapidly decreased due to the interband absorption at \( \lambda \leq 366 \text{ nm} \). After H implantation, the transmittance was reduced all over the wavelength range and the absorption edge was broader and shifted toward higher wavelength to decrease in the bandgap of crystalline GaN.

The absorption coefficient \( \alpha \) of crystalline GaN grown on the sapphire substrate is usually derived from the expression [24]

\[
\alpha = -d^{-1} \ln \left\{ \frac{A + [A^2 + 2BT(1 - R_3)]^{1/2}}{B} \right\}
\]

where \( A = -(1 - R_1)(1 - R_2)(1 - R_3) \), \( B = 2T(R_1R_2 + R_1R_3 - 2R_1R_2R_3) \), \( d \) = film thickness, \( T \) = transmittance, and \( R_1, R_2, \) and \( R_3 \) are reflectance of the air-film, film-substrate, and substrate-air interfaces, respectively. The reflectance from a boundary between two media, like \( a \) and \( b \), can be calculated with refractive indices \( n_a \) and \( n_b \) based on the relation \( R_{ab} = [(n_b - n_a)/(n_b + n_a)]^2 \). The values of \( n \) were 2.33 for the crystalline GaN and 1.765 for the sapphire substrate [25, 26].

After H implantation, the implanted GaN film is not a homogeneous medium. Thus, a two-layer model can be used [27]:

\[
T = (1 - R_v)(1 - R_e)(1 - R_i) \exp(-\alpha_i d_i) \exp(-\alpha_v d_v)
\]

where \( R_v, R_e, \) and \( R_i \) are the reflectance of the air-damaged layer, damaged layer-bulk GaN film and sapphire substrate-air interfaces, respectively. \( \alpha_i \) and \( \alpha_v \) are the absorption coefficient of the damaged and crystalline GaN films, respectively. \( d_i \) and \( d_v \) are the thickness of the damaged and crystalline GaN films, respectively. In the experiment, the value of \( d_i \) is 30 \( \mu \text{m} \), and the value of \( d_v \) depends on the implantation condition, which can be obtained via TEM results as shown later.

The absorption coefficient \( \alpha_v \) was derived from the transmission spectra of the unimplanted sample using equation (3), and the result is shown in figure 3(b). After H implantation, the absorption coefficient \( \alpha_i \) was derived from the transmission spectra of the H-implanted sample using equation (4), and the results are also shown in figure 3(b). Note that the absorption coefficient increased obviously in the H-implanted sample compared to the unimplanted sample, due to the accumulation of optical absorbing H implantation-induced centers. Because the increase in absorption coefficient caused the increase in photon absorption, the Raman intensities decreased with increasing dose. We got the value of the absorption coefficient at \( \lambda = 514.5 \text{ nm} \), which was the excitation wavelength of Raman scattering. To investigate the change of the absorption coefficient at \( \lambda = 514.5 \text{ nm} \) with the implantation dose, the normalized value defined \( \alpha_i/\alpha_v \) was plotted in figure 3(c), together with the normalized Raman intensity. With increasing implantation dose, the normalized absorption coefficient increased,
while the normalized Raman intensity decreased. Similarly, due to dynamic annealing at elevated temperatures, the normalized absorption coefficient decreased, while the normalized Raman intensity increased, as illustrated in figure 3(d).

The results are reasonable to believe that the increase in absorption coefficient causes the decrease in Raman intensity.

3.4. Transmission electron microscopy

To understand the correlation between the disorder level and the microstructural evolution, the damaged profiles obtained by RBS/C analysis and the simulated damaged profiles via SRIM program were overlaid on the corresponding TEM images, as shown in figure 4. The images were taken along the GaN [0110] zone axis. No amorphous layer was detected anywhere in the damaged layer. High densities of black contrasts which are correlated to H implantation-induced dislocation loops, stacking faults and strain were observed in figure 4.

As shown in table 1, the width of the H implantation-induced damage band depends on the implantation condition. For the RT implanted sample, the width of the H implantation-induced damage band slightly increases with increasing dose. On the contrary, the width of the H implantation-induced

**Figure 4.** Damage profiles taken from the RBS/C analysis (the left scale, squares) and SRIM simulation (the right scale, dots) overlaid on the corresponding TEM images for the samples implanted at RT to a dose of (a) $3.75 \times 10^{16}$ H$_2$ cm$^{-2}$, (b) $5 \times 10^{16}$ H$_2$ cm$^{-2}$, (c) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$, (d) $1.75 \times 10^{17}$ H$_2$ cm$^{-2}$, (e) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$ at 573 K, (f) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$ at 723 K.

**Table 1.** The dependence of the damage band below the sample surface formed by H$_2$-implantation on the implantation condition.

| Dose (H$_2$ cm$^{-2}$) | Damage band (nm) |
|------------------------|------------------|
| $3.75 \times 10^{16}$   | 290–510          |
| $5 \times 10^{16}$     | 290–510          |
| $1.5 \times 10^{17}$ at RT | 230–580        |
| $1.75 \times 10^{17}$  | 220–580          |
| $1.5 \times 10^{17}$ at 573 K | 350–710         |
| $1.5 \times 10^{17}$ at 723 K | From surface to 710 |
Figure 5. XTEM bright field micrograph (a) of the damaged layer in GaN implanted with 134 keV H$_2$ ions to a dose of $1.75 \times 10^{17}$ H$_2$ cm$^{-2}$ at RT. Electron diffraction pattern taken from the damaged area is shown as an inset. (b) High-resolution TEM image showing the highly disordered regions and stacking faults in the damaged layer.

Figure 6. XTEM bright field micrographs of the bubble layer in GaN implanted with 134 keV H$_2$ ions at RT to a dose of (a) $5 \times 10^{16}$ H$_2$ cm$^{-2}$, (b) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$, (c) $1.75 \times 10^{17}$ H$_2$ cm$^{-2}$, (d) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$ at 573 K, (e) $1.5 \times 10^{17}$ H$_2$ cm$^{-2}$ at 723 K.
damage band significantly increases at elevated temperature implantation. It is caused by the diffusion of lattice defects at elevated temperature.

Compared to the damage distribution obtained by RBS/C, the damage distribution obtained by TEM is wider. One possible reason that may explain the observed phenomenon is the volume swelling induced by H implantation. Ion implantation-induced volume swelling in GaN has been well studied. Bae et al. [28] measured the atomic-level volume changes in the amorphous region produced by Au irradiation in GaN and they found an atomic-level volume expansion of 18.9% after Au-irradiation. In the present study, the selected electron diffraction pattern shows no amorphous layer observed in the GaN film as the sample was implanted at the maximum dose [see the inset of figure 5(a)]. However, many highly disordered zones in the damaged layer were observed by high-resolution TEM, as shown in figure 5(b). The highly disordered regions appear to be inhomogenous with distinct defect zones on the order of 1–5 nm in size, as indicated by circles in figure 5(b). In addition, some mottled contrast due to lattice stress induced by lattice defects were observed in figure 5(b). At some zones indicated by white arrows, the fringes are wavy, indicating that these zones contain stacking faults. These stacking faults located in the basal plane were formed by intrinsic or extrinsic type defects. Thus, the H implantation-induced volume swelling in GaN is smaller than 18.9%. Zhang et al. [29] measured the atomic-level volume in the damaged region with a relative Ga disorder level of 0.55 and they found an atomic-level volume expansion of 3%. In the study performed by Zhang et al. [29], they used Au ions to implant GaN films and the effect of bubbles is excluded when the damaged region has a relative Ga disorder level of 0.55. In the present research, we used H ions to implant GaN films and numerous nano-size bubbles which contain hydrogen were formed in the damaged layer. Figure 6 shows under-focused bright field TEM images of GaN implanted to doses of 5 × 10^{16} to 1.75 × 10^{17} cm^{-2}. The distribution of bubbles is related to the implantation condition. No bubbles were found in the damaged layer of the GaN sample that was implanted to a dose of 3.75 × 10^{16} cm^{-2} (no shown). A homogenous

Figure 7. XTEM weak-beam dark field micrographs of GaN implanted with 134 keV H^{+} ions at RT to a dose of 5 × 10^{16} cm^{-2} using (a) \( g = 0002 \), (b) \( g = −2110 \), (c) \( g = 1−100 \), and (d) \( g = 10-10 \). (e) High-resolution TEM image showing stacking faults taken from the damaged region.
distribution of bubbles was observed in the damaged layer when the sample was implanted to a dose of $5 \times 10^{16} \text{H}_2 \text{cm}^{-2}$, as shown in figure 6(a). An inhomogenous distribution of bubbles was observed in the damaged layer when the samples were implanted to doses between $1.5 \times 10^{17}$ and $1.75 \times 10^{17} \text{H}_2 \text{cm}^{-2}$, as shown in figures 6(b) and (c). The majority of the bubbles are distributed around $R_p$, which corresponds to the zone of the maximum H concentration. For the elevated temperature implantation, a homogenous distribution of bubbles was observed in the damaged layer as shown in figures 6(d) and (e), respectively. There is no significant change of bubble diameter in the H-implanted GaN. Therefore, it is reasonable to assume that the atomic-level volume expansion in the GaN samples implanted to doses of $5 \times 10^{16}$ to $1.75 \times 10^{17} \text{cm}^{-2}$ is larger than 3%.

To investigate geometry of the dislocation loops produced by H implantation, Burgers vector analysis was carried out using $g, b$ criteria. Figure 7 is a montage of the same region.
imaged under different diffraction conditions: \( g = 0002 \), \( g = 1−100 \), \( g = −2110 \) and \( g = 10−10 \) in the \( \text{H}_2 \)-implanted GaN to a dose of \( 5 \times 10^{16} \text{H}_2^+ \text{cm}^{-2} \). When imaged with \( g = 0002 \) [see figure 7(a)], the loops can be divided into three regions. First, the region close to the surface contains many small loops with weak contrast. Second, at the region between the peak damage and the projected range, there are many large loops with high contrast. Third, beyond the projected range, there are many small loops with weak contrast. Defects with Burgers vector 1/2\[0001\]. It is reasonable that the (0001) plane is the most densely packed plane for the wurtzite-type structured GaN. To elucidate the microstructural feature of the observed basal loops in the damaged layer, a high-resolution XTEM image was shown in figure 7(e). One can see that crystalline GaN has a…ABABAB…stacking sequence (where A refers to a pair of atom planes). In the damaged zone, where contains the extrinsic fault, becomes a …ACBAABAAB…structure (where C refers to the extra layer), as shown in figure 7(e). Thus, the basal loops are of an interstitial type. This is consistent with the report of Wang et al [30].

Weak-beam dark field with \( g = 0002 \), \( g = 10−10 \) near \( z = 11−20 \), was chosen to investigate the microstructural evolution of the damaged layer with increasing dose or temperature. In this way the local atomic displacement along the [0001] direction can be observed. Figure 8 shows cross-sectional WBDF TEM images of the microstructures in the \( \text{H}_2 \)-implanted GaN at different doses. The dislocation loops and stacking faults are visible in the weak-beam images appearing as white dots. It can be seen that very dense defects were formed in the damaged layer. The width of the damaged layer increased with increasing dose. The sizes of observed defects did not change markedly. Figure 9 shows the microstructural evolution of the \( \text{H}_2 \)-implanted GaN at different temperatures. There is a similar defect contrast observed in the \( \text{H}_2 \)-implanted GaN at different doses. Note that the width of the damaged layer increased with increasing temperature. Moreover, with increasing temperature, the sizes of observed defects increased slightly, while the number density of these defects decreased.

4. Conclusions

The formation of lattice disorder in H-implanted GaN epitaxial films was investigated. The evolution of the damage peak measured by RBS/C with the implantation dose or implantation temperature illustrates that a three-step damage process depending on the change of the relative Ga disorder with the ion dose can be found. At doses \( \leq 3.75 \times 10^{16} \text{H}_2^+ \text{cm}^{-2} \), the relative Ga disorder at the peak region is only 0.05. At doses between 3.75 \times 10^{16} and 5 \times 10^{16} \text{H}_2^+ \text{cm}^{-2} \), the relative Ga
disorder at the peak region increases rapidly up to 0.6, because numerous hydrogen bubbles and extended defects, i.e. stacking faults and basal dislocation loops, are formed in the damaged layer. These defects inhibit the recombination of the primarily produced defects within the collision cascades. At doses $\geq 5 \times 10^{16} \text{H}^+ \text{cm}^{-2}$, the relative Ga disorder at the peak region approaches saturation. There is no amorphous layer in the sample implanted to a dose of $1.75 \times 10^{16} \text{H}^+ \text{cm}^{-2}$ at RT. The increase in optical absorbing H implantation-induced centers with increasing dose leads to the increase in the absorption coefficient of the samples, and thus the decrease in the intensities of Raman scattering is identified. Implantation dose and temperature dependence of the normalized intensity of $E_2^\text{LO}$ phonon at 570 cm$^{-1}$ can be simulated by exponential curves. Three new weak peaks at 300, 420 and 670 cm$^{-1}$ are found in the GaN epitaxial films implanted to doses between $5 \times 10^{16}$ and $5 \times 10^{17} \text{H}^+ \text{cm}^{-2}$ at RT. These new peaks are attributed to the disorder-activated Raman scattering modes. A volume expansion over 3% is estimated in the damaged region after H$^+$/implantation. Most of the lattice defects induced by H implantation are of interstitial-type basal loops with Burgers vector $1/6 [0001]$, which do not appear to be correlated to both the implantation dose and temperature.

Acknowledgments

The work was supported by National Nature Science Foundation of China (Grant Nos. 11005130, 11475229) and West Light Foundation of the Chinese Academy of Sciences. The authors appreciate the Laboratory of 320 kV High-voltage Light Foundation of the Chinese Academy of Sciences. The work was supported by National Nature Science Foundation of China (Grant Nos. 11005130, 11475229) and West Light Foundation of the Chinese Academy of Sciences. The authors appreciate the Laboratory of 320 kV High-voltage Light Foundation of the Chinese Academy of Sciences.

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