HiPIMS-grown AlN buffer for threading dislocation reduction in DC-magnetron sputtered GaN epifilm on sapphire substrate

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Gallium nitride (GaN) epitaxial films on sapphire (Al 2 O 3 ) substrates have been grown using reactive magnetron sputter epitaxy with a liquid Ga target. Threading dislocations density (TDD) of sputtered GaN films was reduced by using an inserted high-quality aluminum nitride (AlN) buffer layer grown by reactive high power impulse magnetron sputtering (R-HiPIMS) in a gas mixture of Ar and N 2 . After optimizing the Ar/N 2 pressure ratio and deposition power, a high-quality AlN film exhibiting a narrow full-width at half-maximum (FWHM) value of the double-crystal x-ray rocking curve (DCXRC) of the AlN(0002) peak of 0.086° was obtained by R-HiPIMS. The mechanism giving rise the observed quality improvement is attributed to the enhancement of kinetic energy of the adatoms in the deposition process when operated in a transition mode. With the inserted HiPIMS-AlN as a buffer layer for direct current magnetron sputtering (DCMS) GaN growth, the FWHM values of GaN(0002) and (10 1 0) XRC decrease from 0.321° to 0.087° and from 0.596° to 0.562°, compared to the direct growth of GaN on sapphire, respectively. An order of magnitude reduction from 2.7 × 10 8 cm −2 to 2.0 × 10 6 cm −2 of screw-type TDD calculated from the FWHM of the XRC data using the inserted HiPIMS-AlN buffer layer demonstrates the improvement of crystal quality of GaN. The result of TDD reduction using the HiPIMS-AlN buffer was also verified by weak beam dark-field (WBD) cross-sectional transmission electron microscopy (TEM).

1. Introduction

III-nitride semiconductor layers, including AlN, GaN, InN, and their alloys, are widely used in high-power and high-frequency devices [1], high-brightness light-emitting diodes [2], and laser diodes [3]. These high-performance devices are mostly fabricated with multilayers on a high-quality GaN thick buffer layer prepared by metalorganic chemical vapor deposition (MOCVD) [4], hydride vapor phase epitaxy (HVPE) [5], or molecular beam epitaxy (MBE) [6]. Alternatively, magnetron sputter epitaxy (MSE), an inexpensive physical vapor deposition technique, is a promising technique for the growth of GaN with several advantages. For instance, the use of pure elemental sources, nitrogen (N 2 ), argon (Ar), and gallium (Ga), as precursors can avoid the incorporation of undesired impurities, growth at relatively low temperatures enables higher incorporation rate of magnesium (Mg) and indium (In) for heavier p-doping nitride and In-rich InGaN, respectively, as well as on temperature-sensitive substrates, and directional growth gives rise to growth of complex nanostructures [7–13]. In addition, MSE has the potential for large-scale growth required for industrial production. Although the low melting point (−29 °C) of Ga results in forming liquid target during sputtering and makes handling of sputtering difficult, the control of sputtering a Ga target in a stable condition has been studied previously [7,8] and growth of high-quality GaN layer was achieved [9]. Demonstrations of GaN-based electronic and optoelectronic devices produced by sputter deposition process at low temperature were reported [10,11,14–17]. Despite its impressive progress, the contained threading dislocation density (TDD) remains high in the GaN films, which may deteriorate device’s performance.

To reduce structural defects in the epitaxial film, along with optimizing growth parameters, the introduction of a buffer layer was often applied to further improve the film’s quality by decreasing the lattice and thermal-expansion-coefficient mismatches [18–22]. Such a strategy is commonly used to grow very high-quality epitaxial film in the III-nitride semiconductor community. For instance, AlN, Al x Ga 1−x N, and...
multilayer AlN/AlGaN thin films are promising buffer layers to GaN films due to each compound's wurtzite crystal structure and tunable lattice parameter [19–21]. As the study of sputtered GaN film is much behind other conventional growth techniques, most efforts were devoted to the process development. Reduction of TDD in sputtered GaN film was achieved when using a MOCVD-grown GaN/Al2O3 template, confirming the importance of a buffer for further improve the film's quality [23]. However, contamination during the sample transfer in air to a sputtering system may deteriorate the surface of the template, requiring additional pretreatment and regrowth to the template. Therefore, it would be good to grow a buffer layer by a sputter process to simplify the process.

In addition, the quality of epitaxial films is strongly affected by the crystal quality of the used buffer layer [18]. In the past, the insertion of an AlN buffer layer resulted in enhancement in the crystalline quality of GaN, InN, and InAlN, that are grown using direct current magnetron sputtering (DCMS) [22]. However, the DCMS-AlN film was often grown in columnar structure, which results in films with high surface roughness [24]. Moreover, the tilted growth of the columns, owing to directionally inclined incoming flux, further generated more boundaries between columns and deteriorated the film quality [25]. To resolve this inherent issue that originates from the DCMS growth technique, deposition of AlN using high-power impulse magnetron sputtering (HiPIMS) [26,27] is proposed in this study. In HiPIMS, the plasma discharge is pulsed and operated at high peak powers and high peak discharge currents, but at a low duty cycle so that the average power remains below the maximum thermal load of the cathode target. Use of high discharge currents results in an increased electron density in the plasma, during the pulse, by two to three orders of magnitude compared to DCMS [26], which leads to significant ionization of the film-forming species [28,29]. The effect of such ionized flux onto the substrate is most notably exemplified by the growth of exceptionally smooth films, which exhibit mass densities close to the corresponding material bulk values, even when deposition is carried out at room temperature and on electrically grounded substrates [30]. This is attributed to an enhanced adatom mobility mediated by the energetic bombardment of the incoming ions [27,30].

Motivated by these previous reports, in this research, high-quality AlN grown by reactive magnetron sputtering was chosen as buffer layer for epitaxial GaN, thanks to a small lattice mismatch of ~3% to GaN. The growth of AlN buffer layer using HiPIMS was studied in terms of manipulating the nitrogen partial pressure (pN2) and average HiPIMS power under a proper pulsing condition. Samples were characterized by double-crystal x-ray rocking curve (DCXRC) and transmission electron microscopy (TEM) to study the structural defects. Furthermore, we investigate the effect of the HiPIMS grown AlN buffer layer on the crystalline quality of GaN grown by DCMS. The results are also compared with GaN deposited on DCMS-grown AlN buffer layer and GaN deposited on bare sapphire substrates. Reduction of TDD in sputtered GaN film with insertion a HiPIMS-AlN buffer demonstrates that the all-sputter process provides a possibility to improve the efficiency in the industrial scale due to the usage of same system for the high-quality III-nitride semiconductor film growth process.

2. Experiments

2.1. Sample growth

GaN and AlN films were grown in two different ultrahigh vacuum (UHV) magnetron sputter systems, which were connected via a transfer tube to avoid breaking vacuum between the subsequent deposition steps. The details of the deposition systems, including a photograph and schematic diagrams of the sputter chambers, can be seen in the supplementary information. Three different types of GaN samples were investigated: 1) GaN deposited directly on the c-plane of single crystal sapphire substrates, 2) GaN deposited on an AlN buffer layer, where the latter was grown by HiPIMS on single crystal sapphire substrates, and 3) GaN deposited on an AlN buffer layer, where the latter was grown by DCMS on single crystal sapphire substrates.

The UHV system used for GaN deposition was pumped down to a background pressure of 5 × 10^-9 Torr, after which a gas mixture of Ar and N2 was introduced. The working gas pressure was maintained at 10 mTorr, while keeping the Ar and N2 partial pressure at 6 and 4 mTorr, respectively. A high-purity 2-inch Ga target (99.9999%), facing upward, was used. Films were grown at a substrate temperature of 900°C using DCMS at an average power of 10 W and a deposition time of 60 min. More details on the deposition system are found in a previous report [7].

The second UHV system, used for deposition of the AlN buffer layer, was pumped down to a background pressure of ~10^-8 Torr, after which a gas mixture of Ar and N2 was introduced. To optimize the growth conditions of the HiPIMS AlN layers, the Ar to N2 ratio was first varied in the range 4:1, 3.2, 2.5:2.5, to 2:3 to study the effects of gas composition, while keeping the total working gas pressure constant at 5 mTorr. A 3-inch high-purity Al target (99.9999%), facing upward, was used. Second, we fixed the average HiPIMS discharge power varied to either 150 W or 300 W to study the effects of deposition power. Increasing the average power allowed us to increase the peak current from approximately 40 A to 55 A and thereby increase ionization of the sputtered species [28,31] at a maintained HiPIMS pulse length and pulse frequency. The HiPIMS process was generated using a HIPSTER 1 HiPIMS unit (Ionautes AB) with the same deposition parameters as the DCMS process at a HiPIMS pulse length and pulse frequency of 30 µs and 1000 Hz (duty cycle = 3%), respectively, and at a substrate temperature of 1000°C with a deposition time of 5 min. The discharge current and voltage time characteristics were monitored and recorded on a Tektronix Keysight DSOX1240G digital oscilloscope, which was connected to the discharge voltage and current sensors of the HiPIMS unit. A corresponding DCMS reference process, used for benchmarking, was generated in the same deposition system using an Advanced Energy MDX-1 K DC power supply at an average power of 300 W, a deposition time of 5 min in a gas mixture of 3 mtorr Ar and 2 mtorr N2, and a substrate temperature of 1000°C. Further details on the deposition system are found elsewhere [18].

2.2. Characterisation

The crystal quality of the grown GaN films and AlN buffer layers were studied by double-crystal x-ray rocking curve (XRC) measurements using a PANalytical Empyrean x-ray diffractometer (XRD) with a 0.15406 nm x-ray wavelength Cu-Kα radiation. The voltage and current of the x-ray tube were set at 45 kV and 40 mA, respectively, and the step width was set to 0.05° with 2 s scanning time. A hybrid monochromator 2-bounce Ge (220) mirror is used as the incident beam optical module, and a 0.27° parallel plate collimator is used as the diffracted beam optical module. The curve data of the XRD and XRC measurement was fitted using a Voigt peak shape [32]. The function was derived by integrating over the Lorentz profile and weighting with a Gauss function using the numerical software. With the peak fitting calculation, information on peak position, area, and FWHM can be obtained.

Cross-sectional samples were prepared by traditional methods including mechanical grinding and 5–3 kV Ar + ion polishing (gradually reduced to minimize the damage). Weak beam dark field (WBDF) from 3 g condition was performed in transmission electron microscope (TEM) Linköping FEI Tecnai G2 operated at 200 kV (FEI Company, Hillsboro, Oregon, USA).

3. Results and discussion

3.1. AlN growth

To optimize the quality of AlN layers, the structural characteristics of HiPIMS deposited AlN thin films with different Ar to N2 gas partial...
pressure ratios, keeping the total working gas pressure constant at 5 mTorr, in the gas mixtures were first evaluated using XRC, presented in Fig. 1(a). At the lowest nitrogen partial pressure, Ar:N\textsubscript{2} = 4:1, i.e., p\textsubscript{N\textsubscript{2}} = 1 mTorr, the XRC of AlN(0002) reveals a broad peak with a FWHM of 1.296\degree, indicating a mosaic nature with a rather low crystal quality. After increasing the nitrogen content to p\textsubscript{N\textsubscript{2}} = 2 mTorr, the FWHM of the XRC dramatically decreases to 0.114\degree, indicating a massive improvement of the crystal quality. When further increasing the nitrogen partial pressure to p\textsubscript{N\textsubscript{2}} = 2.5 and 3 mTorr, the FWHM of the XRC instead increases to 0.688\degree and 0.757\degree. Note that it was not possible to increase the nitrogen partial pressure beyond this point, since the process became unstable and exhibited significant arcing.

The observed changes in crystal quality are connected to the amount of nitrogen available in the discharge, where the latter controls the fraction of compound formed on the target surface [33]. A too low supply of the reactive gas gives rise to the deposition process being operated in the metal mode and typically resulting in an under stoichiometric composition of the deposited film. A too high supply of the reactive gas will cause poisoning of the target surface leading to operation in the poisoned (compound) mode with significantly reduced deposition rates, and sometimes forming arc discharges leading to the incorporation of macroparticles in the film. The transition sputtering zone between these two modes is frequently unstable, and a hysteresis in the process parameters (i.e., discharge voltage, deposition rate, and reactive gas partial pressure) is commonly observed.

For samples grown at p\textsubscript{N\textsubscript{2}} = 1 mTorr, we are operating close to the metal mode, as indicated by a close to constant discharge voltage at around −600 V when changing from a pure Ar discharge to the desired Ar:N\textsubscript{2} mixture [34]. The absence of N\textsubscript{2} may result in an excess of Al, which causes the formation of local regions of pure Al in the AlN films. For example, Aita et al. [35] showed that Al regions were formed in a gas mixture of Ar (95%) and N\textsubscript{2} (5%). In addition, insufficient N\textsubscript{2} in the gas mixture increases the possibility of forming point defects and dislocations owing to implanted Ar and excess metal atom’s inclusion, which lead to lower crystal quality [35,36]. By increasing the nitrogen partial pressure to p\textsubscript{N\textsubscript{2}} = 2 mTorr, the recorded cathode discharge voltage strongly changes from about −588 V to −414 V, which shows a characteristic of transition mode of the hysteresis curve. The result indicates the grown films shift to more suitable and nitrogen-richer region in the coating. By further increasing the partial pressure to p\textsubscript{N\textsubscript{2}} = 2.5 and 3 mTorr, the discharge voltage only slightly changes from −414 to −388 and 386 V, respectively, which indicates operating the sputtering close to the compound mode [34]. We envisage that excessive N\textsubscript{2} in the gas mixture reacts with the Al target during the deposition process, forming a thin AlN\textsubscript{x} compound on the surface [37] leading to target poisoning. With target poisoning, both Al and Al–N precursors will sputter from the target and condense on the substrate surface [38]. As the Al–N precursor was formed at substantially lower temperature on the target site, they are less mobile than metallic precursors. Hence, the deposition of precursor on substrate leads to poor crystal quality of the AlN films.

A further evidence of target poisoning in HiPIMS can be seen in the discharge current evolution [39], where a transition from operation in the metal mode (low p\textsubscript{N\textsubscript{2}}) to the compound mode (high p\textsubscript{N\textsubscript{2}}) in reactive HiPIMS discharges are commonly associated with a change in the peak discharge current. For example, Shimizu et al. [40] reported a doubling of the peak discharge current at constant discharge voltage, pulse frequency, and pulse width between the metal mode and the poisoned mode in a hafnium nitride HiPIMS process. A similar change is also seen in Fig. 1(b), where the peak current increases from approximately 30 A to 51 A with increasing p\textsubscript{N\textsubscript{2}}. The behavior is connected to significant recycling of either ionized sputtered Al at low p\textsubscript{N\textsubscript{2}}, so-called self-sputter recycling, or primarily ionized working gas (Ar as well as N\textsubscript{2}) at high p\textsubscript{N\textsubscript{2}} flow rates, so-called working gas recycling, as described in detail by Gudmundsson et al. [39] and Brenning et al. [41]. These two recycling mechanisms lead to ions being drawn back to the cathode, which amplifies the discharge current. The large increase in peak discharge current with increasing nitrogen partial pressure in Fig. 1(b) is in a consequence of recombined gas ions that return from the target during the pulse and subsequently become ionized and drawn back to the target, which allows a very effective amplification of the discharge current through gas recycling [41].

From the literature, it is well-known that ionization of sputtered species in HiPIMS scales with peak discharge current [28,42]. To further explore a possible improvement of the crystal quality through the use of highly ionized deposition fluxes, the best Ar to N\textsubscript{2} gas ratio from the results above, i.e., Ar:N\textsubscript{2} = 3:2, was investigated at two different peak discharge currents. In the present case a peak current increase from approximately 40 A to 55 A was achieved by increasing the average power from 150 W, used in the previous experimental series, to 300 W at otherwise identical conditions. Fig. 2(a) shows the XRC measurements of the AlN buffer layers grown by HiPIMS at the two different average powers and peak currents, as shown in Fig. 2(b). We find that an increase in peak current results in a further decrease of the FWHM of the AlN(0002) XRCs from 0.114\degree to 0.086\degree, indicating a better crystal quality in the latter case.

Note, however, that the improvement in crystal quality is, relatively speaking, smaller compared with the changes observed when varying the gas composition (compare Fig. 1(a) and 2(a)). It should also be pointed out that the increase in average power for the same Ar to N\textsubscript{2} gas ratio may also lead to a slight shift reactive process mode where a higher average power increases compound removal on the target and the process might thereby shift somewhat in the direction toward the metal mode [34].

In addition, as a benchmark, an AlN film was grown on sapphire

![Fig. 1. (a) XRCs of AlN (0002) peak measured from samples grown with 150 W average power in the 5 mTorr total working pressure with different Ar to N\textsubscript{2} partial pressure ratio range from 4:1, 3:2, 2.5:2.5, to 2:3. (b) Discharge current of HiPIMS power supply with 150 W average power in the 5 mTorr total working pressure with different Ar to N\textsubscript{2} partial pressure ratio range from 4:1, 3:2, to 2:3.](image-url)
substrate by DCMS using the same condition at 300 W. The XRC of AlN (0002) shows a slightly broad peak with a FWHM of 0.143° in comparison with the HiPIMS-grown AlN film (FWHM = 0.086°), see Fig. 3 (a–b). In comparison, previously reported values of the FWHM from XRC of AlN(0002) are in the range of 1° to 0.1° [43,44]. Our DCMS- and HiPIMS-grown AlN films thus exhibit a relatively high crystal quality. With a multiple-peaks-fitting analysis of the XRC peak using the Voigt peak-shape, the deconvolution reveals two different types of features in both cases. We observed that the peaks in the low-intensity region are relatively broad while towards the high-intensity region, they are very narrow. The broad background has been observed previously in GaN [45], AlN [46], and GaN/AlN heterostructure systems [47]. The work of Miceli et al. [48] demonstrated the superposition of a narrow peak and a broad background peak due to the coexistence of the long-range and short-range scattering order from epitaxial growth. The narrow peak is mainly due to higher crystal quality domains with less lattice relaxation from the layer below. The broad background is attributed to the migration of the generated defects to the relaxed region and the broadness is increasing with increasing amount of the disordered region, defects, or boundaries. By extracting the peak position of HiPIMS- and DCMS-grown AlN films, we found that the position of the broader background peak is closer to the fully relaxed AlN(0002) peak position at ω = 18.014° [47], which is in agreement with Miceli et al. [48]. For convenience, we termed the narrow peak and broad peak as high- and low-quality AlN phases, respectively. In the case of DCMS-grown AlN film, shown in Fig. 3(a), the (0002) XRC curve was deconvoluted into two peaks with FWHM of 0.07° and 0.30°, indicating a possible coexistence of two different quality AlN phases. The amount of high- and low-quality AlN phases are equally presented in the DCMS-grown film. The results suggest a competitive coexistence of a low-quality and a high-quality region of the AlN films. In contrast, the deconvolution of AlN(0002)’s XRC of the HiPIMS-grown film shows a much higher portion of the narrow peak, with FWHM of 0.06°, than the broader peak, with FWHM of 0.24°, showed in Fig. 3 (b), indicating that the high-quality AlN phase is dominant in the film. In previous studies [49], it was observed that the highly energetic adatoms (adions) that are generated from HiPIMS process lead to formation of larger grains or columns in grown films and less internal voids. Thus, here we propose that this high quality AlN will provide a good template for further growth of GaN with low defects density and low strain.

3.2. GaN growth

In this section, we further evaluate the crystalline quality of GaN thin films deposited (i) directly onto c-plane of sapphire substrate (ii) on a DCMS grown AlN buffer layer, and (iii) on a HiPIMS deposited AlN buffer. In all cases, the GaN films were deposited with a DCMS technique with an identical growth condition. From the previous study of GaN using magnetron sputter, increasing N:\textsubscript{2}/Ar pressure ratio results in lower density of Ga droplets on surface of films and rough surface with hexagonal features. To obtain a balance between two competitive changes, a N:\textsubscript{2}/Ar pressure ratio of 4/6, was chosen.

The influence of the introduced AlN buffer layer, grown by DCMS and HiPIMS, in the process of GaN films growth by DCMS was further evaluated using XRC technique again. Fig. 4(a) shows that the GaN film directly grown on sapphire substrate has a wide GaN(0002)’s rocking curve with a FWHM of 0.321° (1155 arcsec). With introducing a DCMS AlN buffer layer, the FWHM is dramatically reduced to 0.122° (439 arcsec). With an AlN buffer layer grown using HiPIMS, the FWHM is further improved to 0.087° (313 arcsec), which is comparable to μm-thick GaN films directly grown onto c-plane sapphire substrate and/or...

Fig. 2. (a) XRC measurement of AlN (0002) peak grown with different average power at 150 and 300 W in a gas mixture of Ar (3 mTorr) and N:\textsubscript{2} (2 mTorr). (b) Discharge current of HIPIMS power supply with different average power at 150 and 300 W in a gas mixture of Ar (3 mTorr) and N:\textsubscript{2} (2 mTorr).

Fig. 3. XRC of AlN(0002) measured from (a) DCMS- and (b) HPIIM-grown AlN films. The red, green, and blue curves represent to low quality AlN, high quality AlN and fitted curved, respectively.
with an AlN buffer layer assistance using conventional MOCVD technique \cite{50}. In addition, the XRC's width of GaN films is similar to the AlN buffer layers, implying that the growth behavior of GaN is guided by the structure of the buffer layer \cite{22}. With a multiple-peaks-fitting analysis, the GaN film grown on DCMS AlN buffer can still be deconvoluted into two peaks with FWHMs of 0.30° and 0.09°, indicating that the film still is composed of two phases with long- and short-range orders. However, the short-range order is completely suppressed in the case that GaN film grown on HiPIMS AlN buffer.

Nevertheless, in Fig. 4(b), the XRC of GaN (1011) shows a similar FWHM value around 0.8° (2880 arsec) of Direct growth, and HiPIMS-AlN buffer samples. With DCMS-AlN buffer layer, the FWHM of GaN film even increases to 0.954° (3434 arsec). All these films have broader XRC in comparison with MOCVD-grown GaN films, which normally have a narrow FWHM around 0.2° \cite{50}.

To quantitatively analyze the crystal quality, the screw- and edge-type threading dislocation densities (TDDs), \( N_s \) and \( N_e \), respectively, can be calculated using the full width at half maximum (FWHM) of GaN(0002) and (1010) XRCs, represented as \( \Gamma_S \) and \( \Gamma_E \), respectively, using Eqs. (1) and (2), derived by Kobayashi et al. \cite{51},

\[
N_s = \frac{\Gamma_S^2}{4.35 |b_s|^2},
\]

\[
N_e = \frac{\Gamma_E^2}{4.35 |b_e|^2},
\]

where the \( b_s \) and \( b_e \) represent to burgers vector of screw- and edge-type dislocation with the value of 2.7 \( \times \) \( 10^{-15} \) and 1.015 \( \times \) \( 10^{-15} \) cm\(^2\) for hexagonal GaN, respectively.

Here, the \( \Gamma_S \) can be extracted from the FWHM value of XRC result of GaN(0002). The \( \Gamma_E \) was extracted using a fitted nonlinear curve formula \cite{52},

\[
\Gamma(x) = \left( F_e \cos \chi \right)^2 + \left( F_e \sin \chi \right)^2,
\]

where \( \chi \) is the angle between the pyramidal plane and basal plane, (0002), in a hexagonal crystal. \( \Gamma \) is the FWHM of the XRC measured from the pyramidal plane. Here, several XRCs measured from pyramidal planes, including (10\( \bar{1} \)3), (10\( \bar{2} \)), (10\( \bar{1} \)), and (20\( \bar{2} \)), and basal plane are used for the fitting to extract \( \Gamma_E \).

In Fig. 5, the fitted curves of Direct growth, DCMS-AlN buffer, and HIPIMS-AlN buffer samples were shown with the dashed curve. Those \( \chi \) angles calculated from hexagonal GaN samples are used as x-axis in Fig. 5. Using Eqs. (1)-(3), measured \( \Gamma_S \) and extracted \( \Gamma_E \), both screw- and edge-type dislocation densities can be calculated. The direct growth GaN film contains highest screw dislocation density, 2.7 \( \times \) \( 10^9 \) cm\(^{-2}\). After introducing a DCMS-AlN buffer layer, the screw-type TDD decreases to 3.8 \( \times \) \( 10^8 \) cm\(^{-2}\). Furthermore, the screw-type TDD decreases to 2.0 \( \times \) \( 10^8 \) cm\(^{-2}\) using HiPIMS-AlN buffer layer.

The edge-type TDDs measured from HiPIMS-AlN buffer GaN film and direct growth GaN film are very close to each other, i.e., 4.2 \( \times \) \( 10^{10} \) to 4.1 \( \times \) \( 10^{10} \) cm\(^{-2}\) respectively. The DCMS-AlN buffer GaN film shows a higher density of 6.3 \( \times \) \( 10^{10} \) cm\(^{-2}\), which is around one order higher than the MOCVD-grown GaN films. However, the edge-type dislocation dominated the generated defects in GaN films can be attributed to a small film thickness, around 500 nm. In the report of Chierchia et al. \cite{53}, the edge-type TDD drops with increasing layer thickness, whereas the screw-type TDD remains constant. Hence, to further suppress the formation of edge-type TDDs in GaN film, an increase of deposition rate to grow thick film in a reasonable time will be in demand.

The reason of the crystal quality improvement of the grown GaN films after introducing the AlN buffer layer is assumed to be the same crystal structure and small lattice mismatch to decrease the strain and deformation between the surface between GaN film and substrate. The improvement of crystal quality after changing from DCMS to HiPIMS is due to a smoother surface between AlN and GaN. Compared with DCMS-AlN buffer layer, the HIPIMS-AlN buffer layer can provide smoother surface (see cross-section TEM measurement in next section), which further decreases the dislocation density of the grown GaN films. Also, as the results shown in Fig. 5, the HIPIMS-AlN buffer layer contains larger...
3.3. Transmission electron microscopy of films

Fig. 6 shows the weak beam dark-field (WBDF) cross-sectional TEM image of three different as-grown GaN films, which were grown directly onto sapphire substrate (left panel), with an assistance of a DCMS-AlN buffer layer (middle panel) and a HiPIMS-AlN buffer layer (right panel). The weak beam dark-field (WBDF) method is a technique based on the formation of a diffraction-contrast from the weakly excited beam. Dark field images of weak beam are normally taken at high order (3 g) of Bragg settings. Dislocations could be differed in screw-, edge- or combined-type and visualized as sharp bright lines in WBDF [54].

In Fig. 6(a–c), the WBDF images were taken along the zone axis of [0002], and 0002 g-3g condition was used, where only screw- and mixed-type threading dislocations (TDs) can be seen by the visibility criterion $\mathbf{g} \cdot \mathbf{b} \neq 0$. As can be seen in these images, most TDs are generated at interface and propagate toward to the film surface. Some TDs merge into one dislocation through bending, resulting in annihilation with thickness. The annihilation of dislocation is attributed to a preferable lateral growth under a Ga-rich conditions, which was reported in a previous study [55]. For the Direct growth sample, seen in Fig. 6(a), an obviously distorted region can be observed in the film at the GaN/AlN interface. Such small islands can lead to not only the formation of TDs (white lines in the film), but also to lattice distortion, which gives rise to the broadening of GaN(0002) XRC’s peak width. After introducing the DCMS-AlN buffer layer, the GaN film contains a large number of dislocations near the GaN/AlN interface (under the blue dashed line region) and a dramatic decrease of dislocation’s density near the film surface (above yellow dashed line region), see Fig. 6(b). However, these dislocations may not be all counted as screw-type TD because many of them may be contributed from grain boundary of columns, which is in agreement with the XRC result that the film contains two different quality components. These grain boundaries disappear and only a few TDs exist at film surface. In opposite, the formation of high-density dislocation at GaN/AlN interface is eliminated when using the HiPIMS-grown AlN as a buffer layer, as shown in Fig. 6(c). Only threading-type dislocations are seen in the GaN film. Also, these dislocations are mostly generated as an extension of existing dislocations in the AlN buffer layer rather than newly formed dislocations from the buffer’s surface.

In Fig. 6(d–f), the WBDF TEM images were taken along the zone axis of [1120], and 1120 g-3g condition was used, where only edge- and mixed-type threading dislocation are visible. Compared with WBDF image along [0002], all the grown GaN films contain higher edge-type TDD than screw-type TDD near the film surface region, approximately $10^{11}$ cm$^{-2}$. This result indicates that number of TDDs is dominated by the edge-type dislocation in GaN films, which is consistent with the XRC results presented in Fig. 4(b). Furthermore, the formation of edge-type TDs in these films also starts from the beginning of growth at interface, GaN/sapphire or GaN/AlN, rather than during the growth. A part of dislocation annihilates during growth is more obvious when the GaN film was deposited on an AlN buffer layer. Note that decrease of dislocation density from interface to film surface was partly resulted from the gradient thickness from the sample preparation process.

The reason for the generation of dislocations in the cases that GaN directly grown on sapphire can be associated with the formation of a thin distorted layer with tiny islands at interface due to a very large in-plane lattice mismatch of ~16% between GaN and sapphire substrate. While the dislocations formed in the GaN film on AlN buffer are mostly from the extension of dislocations that already existed in the buffer layer. The very high dislocation density near the interface in the case of GaN grown DCMS-AlN buffer is assumed to be the columnized growth of the AlN layer, owing to limited adatom mobility as reported in a previous study [56]. The boundaries between the columns and the rough surface provide the condition to generate extra dislocations in the GaN film [56]. The HiPIMS-AlN buffer layer contains larger grain sizes and less low-quality region and thus reduce the number of dislocations extended from the buffer to GaN film, which provides the suitable conditions to the formation of high quality GaN with low defects and deformation.
deformation. The increase in AlN grain size is due to enhanced adatom mobility mediated by the ionized film-forming flux, in line with other reports comparing HiPIMS and DCMS [57]. The result establishes that very high crystal quality GaN films can be achieved with this technique.

4. Summary

We have demonstrated that high-quality GaN films can be grown in an all-sputtering process. Reduction of screw-type threading dislocation density with an order of magnitude in DCMS-grown GaN film was achieved by inserting an HiPIMS-grown AlN buffer layer. It is found that the quality of HiPIMS-grown AlN layer is highly dependent on the partial pressure ratio of the used Ar and N2 gas mixture. Increasing average magnetron power can improve the crystal quality slightly. The XRD measurement of AlN(0002) shows that the HiPIMS-grown AlN layer has a smaller FWHM value of 0.086°, which indicates the highest crystal quality of the AlN layers. Applying the same condition to grow AlN by HiPIMS demonstrates an improvement of GaN film crystallinity by the HiPIMS process is attributed to an improvement of crystallinity by the HiPIMS process.

Furthermore, GaN films deposited on c-sapphire substrate inserted with an AlN buffer layer demonstrate a decrease in mosaicity of GaN(0002) plane. The screw-type TDD obtained from XRC FWHM values for GaN films deposited with bare sapphire, DCMS-AIN, and HiPIMS-AIN buffer layer are 2.7 × 105, 3.8 × 105, and 2.0 × 105 cm−2, respectively. The results of TEM cross-section images in weak beam condition also demonstrate an improvement of GaN film crystallinity when introducing an AlN buffer layer grown by HiPIMS. The formation of dislocations in the GaN film directly grown on sapphire is associated with a thin distorted layer with tiny islands formed at the interface, while the dislocations formed in the GaN film on AlN buffer are mostly from the extension of dislocations already existing in the buffer layer.

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CRediT authorship contribution statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.vacuum.2023.112553.

Appendix B. Supplementary data

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References


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