Microstructure dependent residual stress in reactively sputtered epitaxial Si-doped GaN films

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Abstract

Epitaxial Si-doped GaN films were grown on c-sapphire by rf magnetron reactive co-sputtering of GaAs and Si at different N\textsubscript{2} percentages in Ar-N\textsubscript{2} atmosphere. High-resolution x-ray diffraction and ϕ-scans reveal the mosaic growth of c-axis oriented GaN films. Energy dispersive x-ray spectroscopy reveal ~2 at.\% Si in all the films, while the N/Ga ratio decreased substantially with N\textsubscript{2} percentage in sputtering atmosphere. The micro-strain, screw and edge dislocation densities were respectively obtained from ω-2θ, ω, and in-plane ϕ-rocking scans. The films grown at 30%-100% N\textsubscript{2} reveal dominance of edge over screw dislocations, with both approaching similar densities at lower N\textsubscript{2} percentages. The c and a parameters were independently determined to obtain the out-of-plane and in-plane strain components. The strain data was analyzed to separate hydrostatic and biaxial contributions and their dependences on N\textsubscript{2} percentage. The film grown at 100% N\textsubscript{2} displays large hydrostatic strain and micro-strain, attributed to excess/interstitial nitrogen. Both hydrostatic strain and micro-strain decrease substantially with initial decrease of N\textsubscript{2} percentage, but increase slightly in the films grown below 30% N\textsubscript{2}, due to the incorporation of Ar. The films grown at \geq 75% N\textsubscript{2} reveal growth related, intrinsic biaxial stress, which is compressive and is attributed to the incorporation of excess nitrogen into grain boundaries and tensile side of edge dislocations. The films grown below 75% N\textsubscript{2} display stress reversal owing to the prevalence of coalescence related intrinsic tensile stress, which decreases in the films grown below 30% N\textsubscript{2}, due to the incorporation of Ar and their voided structure.

Keywords: GaN, Si-doping, epitaxial growth, reactive sputtering, residual stress

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1. Introduction

GaN and its alloys have emerged as important semiconductor materials due to the wide range of their current and potential applications, extending from short wavelength optoelectronics and white light sources to high power and high frequency transistors [1-4]. The ever-increasing scope of GaN calls for exploring non-conventional approaches of material growth and controlled in-situ doping, as well as the improved understanding of its structural properties, particularly those obtained by alternative methods. Hetero-epitaxial GaN films are traditionally grown on sapphire by molecular beam epitaxy (MBE), hydride vapour phase epitaxy (HVPE) and metal-organic chemical vapor deposition (MOCVD/MOVPE), in spite of the large lattice and thermal expansion coefficient mismatch between GaN and the substrate [5]. The GaN films grown on sapphire by MOCVD and MBE reveal high density of threading dislocations, in the range of $10^8 - 10^{10} \text{ cm}^{-2}$ [6-8], but unlike most other compound semiconductors, have been used to fabricate high quality optoelectronic devices [9, 10]. However, due to some limitations, such as, high temperature operation in MOCVD and the scalability and high cost of MBE, alternative methods, such as sputtering remain attractive and continue to be explored for the epitaxial growth of GaN [11-15]. In recent years, sputtering has demonstrated considerable promise for the growth of epitaxial GaN films, although with threading dislocations in the range of $10^9-10^{12} \text{ cm}^{-2}$ [16-18], and has been employed for obtaining undoped [16, 19] and doped (with Si, Ge and Mg) [17, 20-22] GaN films, as well as fabrication of GaN based LEDs [23].

Silicon (Si) is the most common n-type dopant in GaN, which has yielded carrier concentrations as high as $\sim 2 \times 10^{20} \text{ cm}^{-3}$ in the films grown by different techniques and is used in various device structures [20, 24-26]. As GaN based optoelectronic and electronic devices are widely fabricated on sapphire substrate, the presence of substantial strain in doped films influences their optical and electrical properties, often leading to device degradation [27]. The residual stress present in epitaxial GaN has been primarily attributed to the mismatch between thermal expansion coefficients, lattice parameters and elastic constants of the film and substrate, but the growth-related effects, including those due to point defects and impurities cannot be completely ignored [28]. The origin of stress in Si-doped GaN films grown by MBE and MOCVD has been investigated by several workers [29-37]. Ruvimov et al. [29] have shown that Si doping of MOVPE grown GaN improves the structural quality, displaying lesser compressive biaxial stress than the undoped film grown under similar conditions. On the other hand, Lee et al. [30, 31] have reported that Si doping induces overall defects in MOCVD grown GaN films having high carrier concentrations ($4 \times 10^{17} - 1.6 \times 10^{19} \text{ cm}^{-3}$), but causes stress
relaxation during the cool-down process, as inferred from the decrease of $E_2$ mode phonon frequency. Shmidt et al. [32] have also observed the relaxation in biaxial compressive stress and increase in the density of misfit dislocations parallel to the interface with increase of Si concentration, leading to increase in carrier mobility. Romano et al. [33] have investigated in detail, the effect of Si doping on the microstructure and strain behavior in GaN films grown by MOCVD and observed the strain reversal from compressive to tensile, with increase of Si concentration in the films. The increase in tensile stress caused by Si doping was proposed to be due to crystallite coalescence, in accordance with the Nix and Clemens model for polycrystalline films [38]. Cremades et al. [34] have also found that the incorporation of Si in GaN not only led to stress relaxation but also induced tensile stress in the films with higher carrier concentrations ($n > 10^{17}$ cm$^{-3}$). They have further shown that in comparison to fully relaxed GaN, there is an overall shift towards higher lattice constants in Si-doped films, which was ascribed to tensile hydrostatical pressure, probably caused by point defects [34]. The stress behaviour in Si and Ge doped GaN films has also been shown [35] to result in a larger increase of tensile strain with Si concentration. However, Xie et al. [36] have attributed the tensile strain in Si-doped GaN films to free carrier concentration, instead of Si concentration. Forghani et al. [37] have proposed that the tensile strain in Si-doped (Al)GaN epitaxial films may be attributed to the incorporation of Si at the compressive side of edge dislocations. It may be mentioned here that hetero-epitaxial GaN films grown on foreign substrates may also possess various types of defects, such as, threading dislocations and stacking faults, as well as point defects, which need to be considered, particularly in the films grown by alternative methods. It has been shown by Kisielowski et al. [39] that in the case of undoped GaN films grown by different techniques, both biaxial and hydrostatic strains are present. Joachim et al. [40] have also carried out a comparative strain–stress analysis of MBE and MOCVD grown undoped GaN films and related the hydrostatic strain to point defects and biaxial strain to buffer layer thickness. Harutyunyan et al. [41] have carried out high resolution x-ray diffraction studies of GaN films grown by MBE on Ga$_{1-x}$N$_x$/sapphire, with varying N content in the buffer layer and have also shown the presence of both hydrostatic and biaxial strains.

We have recently shown that hydrostatic and biaxial strains are simultaneously present in undoped epitaxial GaN films grown by reactive sputtering at different partial pressures of N$_2$ (10% - 100%) in Ar-N$_2$ sputtering atmosphere and are strongly influenced by the microstructure and defects present in the films [18]. The films grown at high N$_2$ percentages (~100%) displayed large hydrostatic strain and compressive biaxial stress due to the presence of interstitial nitrogen. With decrease of N$_2$ in sputtering atmosphere, the hydrostatic strain in
the films decreased substantially, along with a reversal of biaxial residual stress from compressive to tensile, which is determined primarily by growth-related effects during crystallite coalescence. It may be noted that although the presence of tensile stress has been widely reported in Si-doped GaN films, as described above, studies investigating its dependence on microstructure and defects and in particular, on growth-related effects have not been much explored, especially for the films grown by alternative methods, such as sputtering. Furthermore, the dependence of residual stress in sputtered Si-doped GaN films on growth atmosphere becomes important because sputtered films in general, are known to exhibit a strong dependence of residual stress on sputtering gas composition and pressure [42, 43]. A study relating the residual stress with microstructure therefore, assumes significance, considering the importance of epitaxial Si-doped GaN grown by alternative methods for diverse applications. The present study thus attempts to understand the nature and dependence of the residual stress in epitaxial Si-doped GaN films on the partial pressure of N₂ in sputtering atmosphere. Our results show that both hydrostatic and biaxial strains are present in these films and are significantly influenced by the composition of the sputtering atmosphere. Moreover, the intrinsic biaxial stress and its nature are strongly dependent on the microstructure and surface morphology of the films, as well as the incorporation of excess nitrogen or argon, depending upon the growth conditions.

2. Experimental details

Si-doped epitaxial GaN films were grown by reactive rf magnetron sputtering of GaAs and Si, as described earlier [21]. All the films were grown at 700 °C in Ar-N₂ atmosphere (total pressure, 0.86 Pa) over approximately 50 nm thick GaN buffer layers on c-sapphire substrate, which were grown in 100% N₂ at 300 °C. The films were grown at different nitrogen partial pressures in the range of 0.086 Pa to 0.86 Pa, by changing the corresponding N₂ percentage in sputtering atmosphere from 10% to 100%. Accordingly, the N₂ percentage during growth will be used to label the films grown at various partial pressures of N₂ in this work. The RF power was adjusted to maintain the growth rate of films at (1.0 ± 0.1) μm hr⁻¹, as the Ar percentage was increased. The thickness of all the films was in the range of 850 - 900 nm.

The Si content in doped GaN films was investigated by energy dispersive x-ray spectroscopy (EDX) using JEOL JSM-7600F Field Emission Gun-Scanning Electron Microscope. High resolution x-ray diffraction (HRXRD) measurements were carried out with a Cu rotating anode (9 kW), RIGAKU-Smart Lab-X-Ray diffractometer, equipped with a 2-bounce Ge (220) monochromator, which collimated Cu Kα₁ to ~32 arcsec. The c-parameter
was obtained from $\omega$-2$\theta$ scans of symmetric reflections. The $a$-parameter was obtained from in-plane measurements as well as $\omega$-2$\theta$ scans of an asymmetric reflection. The latter was obtained by tilting the film about $\chi$ axis and optimizing the values of $\phi$, $\omega$, and $\chi$. The micro-structural parameters, namely, mosaic tilt and micro-strain were respectively obtained from out-of-plane measurements of symmetric (0002), (0004), and (0006) reflections in $\omega$ and $\omega$-2$\theta$ scan geometries. The mosaic twist was obtained in $\phi$-rocking curve geometry by in-plane measurement of (11$\overline{2}$0) reflection, using parallel slit collimator (PSC_1°) and analyzer (PSA_1°). Atomic force microscopy (AFM) was carried out to study the surface morphology of the films over an area of 2 μm × 2 μm by using a Nanoscope IV Multimode scanning probe microscope in contact mode, using silicon nitride probes.

3. Results and Discussion

The composition of Si-doped GaN films was investigated by EDX and the corresponding data (along with that of a commercially procured, MOCVD grown GaN film) is presented in figure S1 and Table S1 of supplementary information. It is seen that the measured N/Ga ratio in commercial GaN film is 0.70, while that for the Si-doped GaN films varies from 0.76 to 0.69, with decrease of N$_2$ in sputtering atmosphere from 100% to 10%. The EDX data also confirm the absence of arsenic and the presence of oxygen impurity (~3 at.%) in all the films. These results are in agreement with earlier XPS and SIMS studies of undoped GaN films [18, 44], which also indicated the possibility of excess nitrogen in the films grown at higher N$_2$ percentages, as in the case of Si-doped films. All the films reveal the presence of ~2 at.% Si, irrespective of N$_2$ percentage in sputtering atmosphere. Figure 1(a) shows $\omega$-2$\theta$ high-resolution scans for symmetric (0002), (0004) and (0006) reflections, corresponding to single-phase wurtzite GaN (JCPDS File 01-076-0703) in all the films, which display complete c-axis orientation of crystallites. The positions of (0002), (0004) and (0006) peaks were used to obtain the average values of c-parameter for the films, with an uncertainty of $\pm$ 0.001 Å. For the determination of $a$-parameter of the films, the $\omega$-2$\theta$ high-resolution (10$\overline{1}$1) asymmetric scans and the in-plane measurements of (11$\overline{2}$0) and (20$\overline{2}$0) reflections were carried out. Figure 1(b) shows the $\omega$-2$\theta$ asymmetric scans of (10$\overline{1}$1) reflection for all the films, while the corresponding in-plane data for (11$\overline{2}$0) and (20$\overline{2}$0) reflections are shown in figure S2 of supplementary information. These measurements were used to obtain the average values of $a$-parameter of the films, with an uncertainty of $\pm$ 0.002 Å. The reproducibility of measurements was confirmed by analyzing multiple films grown at each N$_2$ percentage. The average values of the lattice
parameters, \( c \) and \( a \) along with the corresponding error bars are plotted against Ar and N\(_2\) percentages in figure S3 of supplementary information.

The epitaxial nature of Si-doped GaN films was ascertained by phi (\( \phi \)) scans of (10\( \overline{1} \)1) reflections (figure S4 of supplementary information), which shows six dominant peaks at intervals of 60° with each other, corresponding to (10\( \overline{1} \)1) reflections. The \( \phi \)-scan of (10\( \overline{1} \)4) reflections of sapphire, also shown in the figure, are rotated by 30°, which imply the in-plane epitaxial relationship of GaN[11\( \overline{2} \)0]∥\( \alpha \)-Al\(_2\)O\(_3\)[10\( \overline{1} \)0] and alignment of GaN with the oxygen sub-lattice of sapphire, thus confirming the epitaxial character of the films. A detailed microstructural characterization of Si-doped GaN films grown at different N\(_2\) percentages in sputtering atmosphere was carried out by HRXRD and these results are discussed below. The \( \omega \)-2\( \theta \) scans of symmetric (0002), (0004) and (0006) reflections are shown in figure 2(a), and the corresponding Williamson-Hall plots of \( \Delta q_z \) versus \( q \) are shown in figure 2(b). In this case, the film grown at 100% N\(_2\) shows relatively broad \( \omega \)-2\( \theta \) scans, revealing the presence of a large out-of-plane micro-strain. With the initial decrease of N\(_2\) percentage, the full width at half maximum (FWHM) of \( \omega \)-2\( \theta \) peak decreases, as shown for the typical case of the film grown at 50% N\(_2\). However, the films grown at lower N\(_2\) percentages again display slightly higher values of FWHM, indicating a small increase in micro-strain. Figure 3(a) shows the \( \omega \)-scans of symmetric (0002), (0004) and (0006) reflections and the corresponding Williamson-Hall plots of \( \Delta q_x \) versus \( q \), are shown in figure 3(b). The film grown at 100% N\(_2\) displays broad \( \omega \)-scans, which decrease substantially for the films grown at 50%-75% N\(_2\). With further decrease of N\(_2\) below 50%, the \( \omega \)-scans again display slight increase of FWHM, as shown typically for the films grown at 30% and 10% N\(_2\), which is indicative of a slight increase in mosaic tilt. The in-plane \( \phi \)-rocking curves of (11\( \overline{2} \)0) reflection of these films are shown in figure 4. The film grown at 100% N\(_2\) displays a large FWHM of the rocking curve, which decreases slightly with decrease of N\(_2\) and remains nearly the same for the films grown at 30% - 75% N\(_2\). However, with decrease of N\(_2\) below 30%, the FWHM becomes substantially smaller, as shown typically for the film grown at 10% N\(_2\), indicating a significant decrease in mosaic twist. The microstructural data obtained from HRXRD, i.e., the micro-strain, and densities of screw and edge dislocations, which were determined from tilt and twist values [46], are plotted in figure 5 for all the films against Ar and N\(_2\) percentages. It may be mentioned here that in hexagonal systems, including GaN, the density of edge dislocations dominates over the density of screw dislocations [47, 48], which was also observed in the case of undoped sputtered GaN epitaxial films [18]. Figure 5 shows that the film grown at 100% N\(_2\) displays high densities of edge (~3
× 10^{12} \text{ cm}^{-2}\) and screw (~5 × 10^{11} \text{ cm}^{-2}) dislocations, both of which decrease slightly with decrease of N₂ to 30%-50% in sputtering atmosphere. Interestingly, with further decrease of N₂ below 30%, the edge dislocation density decreases substantially, while the screw dislocation density increases, thus both attaining nearly similar values of (3 - 5) × 10^{11} \text{ cm}^{-2} for the film grown at 10% N₂. Figure 5 also shows that the film grown at 100% N₂ has large micro-strain (~1.5 × 10^{-2}), which decreases to ~2 × 10^{-3} with the initial decrease of N₂ to ~50%. However, with further decrease of N₂ percentage to ≤30%, a small overall increase in micro-strain is seen again. The substantial micro-strain in these films is attributed to the presence of the high density of screw dislocations [49]. However, as the sputtering of GaAs is carried out in Ar-N₂ atmosphere of a widely varying composition (10% - 100% N₂), there is also a possibility of the incorporation of excess nitrogen and/or argon in the films, depending on the N₂/Ar ratio, which may also contribute to micro-strain [50, 51], as will be discussed below.

The morphology of Si-doped GaN films grown at different N₂ percentages was also investigated and the results are shown for typical cases in figure S5 of supplementary information, along with the corresponding root mean square (rms) surface roughness data of the films. The films grown at 30%-100% N₂ display nearly similar morphologies, with evenly distributed and well-rounded lateral features, and with rms roughness in the range of 3-5 nm. Asymmetric and irregular lateral features with voided surface morphology and substantial increase of surface roughness are however seen in the films grown at 20%-30% N₂. Interestingly, the films grown at lower than 20% N₂, display symmetric and well-rounded lateral features, but with a much-voided surface morphology, resulting in a large rms roughness of 15-20 nm. In view of the nearly constant growth rate of the films, these variations in surface morphology are attributed to the progressively increasing Ar bombardment of the growing film [52].

The values of the measured out-of-plane strain (\(\varepsilon_c = (c - c_0)/c_0\)) and in-plane strain (\(\varepsilon_a = (a - a_0)/a_0\)) were obtained from the data in figure S3 and are plotted against Ar and N₂ percentages in figure 6, where, \(c_0\) (5.185 Å) and \(a_0\) (3.189 Å) represent the unstrained lattice parameters. The horizontal line represents the extrinsic in-plane (biaxial) strain (\(\varepsilon_c \approx -1.28 \times 10^{-3}\)), due to the mismatch between thermal expansion coefficients of GaN (5.59 × 10^{-6} \text{ K}^{-1}) and c-sapphire (7.5 × 10^{-6} \text{ K}^{-1}), corresponding to the cooling from growth temperature to room temperature (\(\Delta T = 670\) K), which is compressive in character. The film grown at 100% N₂ shows a large out-of-plane tensile strain (~5 × 10^{-3}), which becomes slightly compressive in those grown in the range of 20% - 50% N₂, but regains mildly tensile character at lower N₂.
percentages. On the other hand, a moderate in-plane compressive strain (-2.2 × 10^{-3}) is seen in the film grown at 100% N\textsubscript{2}. However, with decrease of N\textsubscript{2} to ~50%, the in-plane strain becomes tensile and attains the highest value of ~2.5 × 10^{-3} at 30% N\textsubscript{2}, but decreases substantially at lower N\textsubscript{2} percentages.

It is noted from figure 6 that in none of the Si-doped GaN films, the measured values of in-plane and out-of-plane strains display either purely hydrostatic character (\(\varepsilon_c = \varepsilon_a\)) or purely biaxial (elastic) character (\(\varepsilon_c/\varepsilon_a \simeq -0.53\) for GaN [53, 54]), indicating the strong possibility of their concurrent presence. Heteroepitaxial GaN films are known [55] to contain various point defects, such as, V\textsubscript{Ga}, V\textsubscript{N}, Ga\textsubscript{i}, N\textsubscript{i}, N\textsubscript{Ga}, Ga\textsubscript{N}, in addition to O interstitials [56] as impurities. As mentioned above, there is also a possibility of the substantial incorporation of excess nitrogen and/or argon in the films, grown at different N\textsubscript{2} percentages. The hydrostatic strain may originate from the above mentioned defects and impurities, which cause the hexagonal lattice of wurtzite GaN to contract or expand. On the other hand, the in-plane biaxial strain may be extrinsic, due to the large mismatch between the thermal expansion coefficients of GaN and sapphire or can be intrinsic due to coalescence [38] and other growth related effects. The complex behaviour of the strain and related stress in these films and their dependences on N\textsubscript{2}/Ar percentage have thus been analyzed by considering the simultaneous presence of hydrostatic and biaxial strains. The superposition of hydrostatic and biaxial strains results in the measured out-of-plane and in-plane strain components (\(\varepsilon_c\) and \(\varepsilon_a\)), which are given by [39, 41]

\[
\varepsilon_c = \varepsilon_c^b + \varepsilon^h \tag{1}
\]

\[
\varepsilon_a = \varepsilon_a^b + \varepsilon^h \tag{2}
\]

\[
\varepsilon^h = \frac{1-\nu}{1+\nu}(\varepsilon_c + \frac{2\nu}{1-\nu}\varepsilon_a) \tag{3}
\]

where, \(\varepsilon_c^b\) and \(\varepsilon_a^b\) are the biaxial strain components along \(c\) and \(a\) directions, respectively and \(\varepsilon^h\) is the hydrostatic strain. For GaN, the value of Poisson ratio \(\nu \{= c_{13}/(c_{13} + c_{33})\}\) is ~0.21, where \(c_{13} = 106\) GPa and \(c_{33} = 398\) GPa are the elastic constants [41, 53]. Equations (1)-(3) were used to obtain the hydrostatic and biaxial strains for all the films. In order to eliminate the effect of thermal strain contribution (\(\varepsilon_t\)) from the biaxial strain component (\(\varepsilon_a^b\)) along \(a\), which is elastically related to the biaxial strain component \(\varepsilon_c^b = -(2c_{13}/c_{33})\varepsilon_a^b\) [54], \(\varepsilon_t\) was subtracted from \(\varepsilon_a^b\) to obtain \(\varepsilon_a^{b-} \{= \varepsilon_a^b - \varepsilon_t\}\), which represents only the intrinsic contribution to in-plane biaxial strain. It may be mentioned that thermal strain contribution is not subtracted from the out-of-plane strain (\(\varepsilon_c^b\)). Accordingly, the intrinsic biaxial stress corresponding to \(\varepsilon_a^{b-}\), arising entirely from growth-related effects is
\[ \sigma_f = M_f \varepsilon^{b-}_a \]  \hspace{1cm} (4)

where, \( M_f \) \( ( = c_{11} + c_{12} - 2(c_{13}^2/c_{33}) ) \) is the biaxial elastic modulus, \( c_{11} = 390 \) GPa and \( c_{12} = 145 \) GPa are the elastic constants of GaN [41, 53].

Figure 7 shows the variations of \( \varepsilon^h \), \( \varepsilon^{b-}_a \), \( \varepsilon^b_c \) and \( \sigma_f \) with Ar and N\(_2\) percentages in sputtering atmosphere. The plot of \( \varepsilon^h \) in figure 7(a) shows that the film grown at 100% N\(_2\) displays a large hydrostatic strain \((\approx 2.5 \times 10^{-3})\). With the decrease of N\(_2\) to 50%, \( \varepsilon^h \) decreases substantially to less than \( 10^{-3} \) and remains at that level with further decrease of N\(_2\) percentage. The plots of elastic biaxial strain components \( \varepsilon^{b-}_a \) and \( \varepsilon^b_c \) are shown in figure 7(b). A large value of \( \varepsilon^b_c \) \((\approx 2.5 \times 10^{-3})\) is seen in the film grown at 100% N\(_2\), implying a tensile out-of-plane strain. Interestingly, with decrease of N\(_2\) to \( \approx 30\%\), the out-of-plane strain, \( \varepsilon^b_c \) changes from tensile to compressive \((\approx 3.5 \times 10^{-4})\) in the film grown at 10% N\(_2\). The intrinsic biaxial stress \( \sigma_f \) corresponding to growth related, intrinsic in-plane strain, \( \varepsilon^{b-}_a \) shown in figure 7(b), is plotted in figure 7(c). The film grown at 100% N\(_2\) displays a large compressive intrinsic stress, with \( \sigma_f \approx 1.6 \) GPa corresponding to \( \varepsilon^{b-}_a \approx 3.4 \times 10^{-3} \), which nearly relaxes in the film grown at 75% N\(_2\). Interestingly, the films grown at 30%-50% N\(_2\) display a strong tensile intrinsic stress, with \( \sigma_f \) in the range of 0.9 - 1.7 GPa, corresponding to \( \varepsilon^{b-}_a \) in the range of \((2 - 3) \times 10^{-3}\). However, with decrease of N\(_2\) below 30%, the tensile intrinsic stress tends to relax, as it decreases to \( \approx 0.3 \) GPa (corresponding to \( \varepsilon^{b-}_a \approx 6 \times 10^{-4} \) in the film grown at 10% N\(_2\)).

It is clear from above that the Si-doped GaN films grown at high N\(_2\) percentage \((75\% - 100\%)\) display large hydrostatic strain \((>10^{-3})\), which relaxes substantially with decrease of N\(_2\) percentage and tends to increase again in the films grown at 10% - 20% N\(_2\). Interestingly, this behaviour is quite similar to that of the micro-strain, as shown in Fig 5. The hydrostatic strain in GaN films, which is characterized by the concurrent increase of lattice parameters \( c \) and \( a \) has been attributed to the presence of self-interstitials [57], as was also recently observed in undoped GaN films grown by sputtering [18]. As the EDX measurements have indicated above, the films grown at high N\(_2\) percentages \((\approx 75\%)\) are likely to contain excess nitrogen as interstitials, which decreases with decrease of N\(_2\) percentage in sputtering atmosphere. Thus, the large hydrostatic strain as well as large micro-strain in the film grown at 100% N\(_2\), having the highest N/Ga ratio is attributed to excess/interstitial nitrogen and their subsequent decrease with N\(_2\) percentage is seen to be due to the decrease of N/Ga ratio in the films. Figure 7(a) shows a slight increase of hydrostatic strain in the films grown at 10% - 20% N\(_2\), as was also
seen in the case of micro-strain in figure 5, and is attributed to the incorporation of Ar in these films, which are grown at 80% - 90% Ar in sputtering atmosphere.

It is seen from figure 7(c) that there is a substantial intrinsic biaxial stress in Si-doped GaN films, which strongly depends on the N\textsubscript{2} percentage during growth. Interestingly, in most of the films, the intrinsic stress is tensile in character, except for those grown at high N\textsubscript{2} percentages (75%-100%). The growth-related intrinsic tensile stress in polycrystalline thin films has been attributed to the attractive forces between crystallites and explained initially by the grain boundary relaxation model [58]. Nix and Clemens [38] have proposed a quantitative model for the evolution of tensile stress in polycrystalline thin films, based on crystallite coalescence and have asserted that under conditions of low adatom mobility, the tensile stress generated during crystallite coalescence is maintained in the course of the growth of film. It has also been reported [59, 60] that edge type threading dislocations of high density (~10\textsuperscript{10} cm\textsuperscript{-2}) are formed at the coalescence stage during the mosaic growth of epitaxial GaN films, which subsequently lead to low-angle grain boundaries that are rotated about the c-axis. Potin et al. [60] have also observed high-angle grain boundaries extending throughout the film, which are attributed to the local arrangements of threading dislocations. It is therefore likely that the large density of edge dislocations in sputtered Si-doped GaN films originates at the coalescence stage, resulting in the formation of low and high angle grain boundaries and hence a mosaic structure with considerable twist and tilt. It is thus inferred that the tensile stress seen in most of the Si-doped GaN films is related to crystallite coalescence during the initial stages of growth, in line with the Nix and Clemens model [38].

Interesting features are however seen in the behaviour of intrinsic biaxial stress at the higher and lower ends of N\textsubscript{2} percentage in sputtering atmosphere. Firstly, a reversal of intrinsic stress from tensile to compressive is clearly seen in the film grown at ~75% N\textsubscript{2}, which becomes substantially high (-1.6 GPa) at 100% N\textsubscript{2}. Secondly, in the films grown at low N\textsubscript{2} percentages (10% - 20%), the tensile intrinsic stress, tends to decrease substantially. The transition from tensile to compressive stress has been extensively reported in the early literature on sputtered polycrystalline metal films. It has been observed in a large number of metallic films grown at low sputtering pressure [42, 43, 61, 62] and high negative substrate bias [61, 63], and has been attributed to the incorporation of inert gas atoms [42, 43] as well as impurities, such as oxygen [64]. In the present work, all the Si-doped GaN films were grown at nearly the same sputtering pressure, but as mentioned above, the films grown at higher N\textsubscript{2} percentages display significantly larger N/Ga ratios compared to those grown at lower N\textsubscript{2} percentages, while the Si and O contents of the films remain practically unchanged. It is likely that the reason for intrinsic
stress reversal seen in these films is the same as that invoked for explaining the large hydrostatic strain in these films. Hence, it is inferred that the presence of excess nitrogen, and its incorporation into the grain boundaries, results in the relaxation of coalescence related tensile stress and generation of compressive stress in these films [38, 65]. Another mechanism that may also contribute to the stress reversal in these films is the attraction of interstitial nitrogen towards the tensile side of edge dislocations, leading to the dominance of compressive stress [66]. Thus, the stress reversal leading to high compressive stress in Si-doped GaN films grown at higher N\textsubscript{2} percentages (~100%), is primarily attributed to the presence of excess/interstitial nitrogen, which is also in agreement with our earlier studies on undoped GaN films [18]. With the decrease of N\textsubscript{2} percentage (30% - 50%) in sputtering atmosphere and the consequent decrease in N/Ga ratio in the films, these effects are suppressed, resulting in the prevalence of growth-related tensile stress. Finally, the decrease of intrinsic tensile stress seen in the films grown below 30% N\textsubscript{2} may be explained by the possible incorporation of Ar in the films grown at high Ar percentages, since the incorporation of excess atoms, particularly Ar at grain boundaries, is known [43] to relax the growth-related tensile stress, as mentioned above. Thus, the possible incorporation of Ar in the films grown at low N\textsubscript{2} percentages consistently explains the increase in hydrostatic strain and micro-strain, as well as the decrease of intrinsic tensile stress. It may also be noted these films display a much-voided structure along with a substantial increase in their surface roughness, which may also contribute to the decrease in intrinsic tensile stress, as reported in the case of zone 1 type sputtered metal films [67, 68].

4. Conclusion

The residual stress in hetero-epitaxial Si-doped GaN films on sapphire grown by reactive co-sputtering of GaAs and Si at different N\textsubscript{2} percentages (10% -100%) in sputtering atmosphere have been comprehensively investigated and correlated with their composition and microstructural parameters. EDX results show that all the films contain ~2 at.% Si and ~3 at.% oxygen as impurity, while the N/Ga ratio in the films decreases with decrease of N\textsubscript{2} percentage in sputtering atmosphere. The lattice parameters (c and a) were independently measured to obtain the out-of-plane and in-plane strain components, respectively, which show strong dependences on N\textsubscript{2} percentage and reveal the presence of both hydrostatic and biaxial strain contributions. The films grown at 30%-100% N\textsubscript{2} show the dominant presence of edge dislocations (density ~10\textsuperscript{12} cm\textsuperscript{-2}), which decrease to ~5 x 10\textsuperscript{11} cm\textsuperscript{-2} at ~10% N\textsubscript{2}. However, in the films grown below 30% N\textsubscript{2}, the density of screw dislocations increases and approaches values comparable to those of edge dislocations. The film grown at 100% N\textsubscript{2} displays large
micro-strain (~1.5 × 10^{-2}), hydrostatic strain (~2.5 × 10^{-3}) and growth related compressive biaxial stress (-1.6 GPa), which are attributed to the presence of excess nitrogen in interstitial locations, grain boundaries and possibly the tensile side of edge dislocations. With decrease of N\textsubscript{2} in sputtering atmosphere to ~30% and the consequent decrease of N/Ga ratio in the films, the micro-strain and hydrostatic strain decrease substantially, and a reversal of biaxial stress from compressive to tensile is seen due the decrease of excess nitrogen as well as the prevalence of tensile stress generated during the coalescence stage. The films grown below 30% N\textsubscript{2} show a slight increase in micro-strain and hydrostatic strain as well as decrease in coalescence related tensile stress, which are attributed primarily to the incorporation of Ar in the films and possibly their voided structure.

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**AUTHOR DECLARATIONS**

**Conflict of Interest**

The author has no conflicts to disclose.

**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.
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**Figures**

**Figure 1.** The out-of-plane $\omega$-$2\theta$ scans of (a) symmetric (0002), (0004) and (0006) reflections (plotted in log scale) and (b) asymmetric (1011) reflection for Si-doped GaN films grown on c-sapphire at different $N_2$ percentages in sputtering atmosphere (as indicated).

**Figure 2.** (a) $\omega$-$2\theta$ scans of (0002), (0004), and (0006) reflections and (b) the broadening ($\Delta q_z$) in reciprocal space along $\omega$-$2\theta$ axis for (0002), (0004), and (0006) reflections plotted against the magnitude of reciprocal lattice vector (q) for typical Si-doped GaN films grown on c-sapphire at different $N_2$ percentages in sputtering atmosphere (as indicated).
Figure 3. (a) $\omega$ scans of (0002), (0004), and (0006) reflections and (b) the broadening ($\Delta q_x$) in reciprocal space along $\omega$ axis for (0002), (0004), and (0006) reflections plotted against the magnitude of reciprocal lattice vector ($q$) for typical Si-doped GaN films grown on $c$-sapphire at different N$_2$ percentages in sputtering atmosphere (as indicated).

Figure 4. In-plane $\phi$-rocking scans of (1120) reflection for typical Si-doped GaN films grown at different N$_2$ percentages in sputtering atmosphere (as indicated).

Figure 5. Micro-strain (▲), density of screw dislocations (●) and density of edge dislocations (■) in Si-doped GaN films, plotted against Ar and N$_2$ percentages in sputtering atmosphere.
Figure 6. The measured out-of-plane strain, $\epsilon_c$ (▲) and in-plane strain, $\epsilon_a$ (■) in Si-doped GaN films, plotted against Ar and N$_2$ percentages in sputtering atmosphere. The dashed horizontal line represents the in-plane compressive strain due to the mismatch of thermal expansion coefficient between GaN and c-sapphire.

Figure 7. The calculated (a) hydrostatic strain, $\epsilon^h$ (♦), (b) biaxial strains, $\epsilon^{b-}_a$ (●) and $\epsilon^{b+}_c$ (▲), and (c) the growth-related intrinsic biaxial stress, $\sigma_f$ (●) in Si-doped GaN films, plotted against Ar and N$_2$ percentages in sputtering atmosphere.
Supplementary Information

Microstructure dependent residual stress in reactively sputtered epitaxial Si-doped GaN films

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1. Energy dispersive X-ray spectroscopy

Energy dispersive X-ray spectroscopy (EDX) was carried out to study the composition of Si-doped GaN films, in particular, to evaluate their Si content and confirm the absence of arsenic, as shown in figure S1. The results of EDX analysis are presented in Table S1.

Figure S1. EDX results of Si-doped GaN films grown on c-sapphire at different N₂ percentages in sputtering atmosphere (as indicated), which also include the data for a commercially procured, MOCVD grown undoped GaN film.

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Table S1. Summary of EDX analysis of Si-doped GaN films grown at different N₂ percentages in sputtering atmosphere along with the data for MOCVD grown undoped GaN.

| N₂ (%) | Ga (at. %) | N (at. %) | Si (at. %) | O (at. %) | N/Ga |
|--------|------------|-----------|------------|-----------|------|
| 100    | 54.33      | 41.03     | 1.92       | 2.72      | 0.76 |
| 75     | 54.78      | 41.17     | 1.47       | 2.58      | 0.75 |
| 50     | 54.50      | 40.97     | 1.65       | 2.88      | 0.75 |
| 20     | 56.04      | 39.45     | 1.54       | 2.97      | 0.70 |
| 10     | 55.73      | 38.88     | 1.85       | 3.54      | 0.69 |
| MOCVD grown GaN | 58.36 | 40.87 | -           | 0.77      | 0.70 |

2. In-plane x-ray diffraction

The in-plane measurements of (1120) and (2020) reflections were used to obtain the a -parameter of Si-doped GaN films grown at different N₂ percentages in sputtering atmosphere. These results are shown in figure S2.

Figure S2. In-plane measurement of (a) (1120) and (b) (2020) reflections for Si-doped GaN films grown on c-sapphire at different N₂ percentages in sputtering atmosphere (as indicated).
3. Lattice parameters

The $\omega$-2$\theta$ high resolution scans of symmetric and asymmetric reflections as well as in-plane measurements were carried out to independently obtain the $c$ and $a$ parameters of epitaxial Si-doped GaN films grown at different N$_2$ percentages. The average values of $c$ and $a$ parameters along with the error bars are plotted against Ar and N$_2$ percentages in sputtering atmosphere in figure S3. The film grown at 100% N$_2$ shows a significantly larger $c$-parameter ($5.211 \pm 0.01$ Å) and a marginally smaller $a$-parameter ($3.182 \pm 0.0002$ Å), compared to the corresponding standard values for bulk GaN ($c = 5.185$ Å, $a = 3.189$ Å). With decrease of N$_2$ percentage in sputtering atmosphere, the $c$-parameter decreases monotonically and becomes ($5.181 \pm 0.001$ Å) for the film grown at 30% N$_2$, which is slightly smaller than the standard value. However, with further decrease of N$_2$ percentage, the $c$-parameter increases again, becoming slightly larger than the standard value. On the other hand, the $a$-parameter increases initially with decrease of N$_2$ percentage and becomes substantially larger than the standard value for the films grown at 30% N$_2$. However, with further decrease of N$_2$ percentage (<30%), the $a$-parameter decreases substantially, showing values comparable to the standard value.

Figure S3. $c$-parameter (▲) and $a$-parameter (■) of Si-doped GaN films plotted against Ar and N$_2$ percentages in sputtering atmosphere. The horizontal lines (—) and (——) represent the standard values of $c$ and $a$ parameters, respectively.
4. Phi (ϕ) scans

The phi (ϕ) scans of (10̅11) reflections, including ϕ-scan of (10̅14) reflections of sapphire are shown in figure S4, which show six dominant peaks at intervals of 60° with each other, corresponding to (10̅11) reflections. The ϕ-scan of (10̅14) reflections of sapphire are rotated by 30°, which imply the in-plane epitaxial relationship of GaN[11̅20]∥α-Al₂O₃[10̅10].

![Figure S4. ϕ-scans of (10̅11) reflections of Si-doped GaN films grown on c-sapphire at different N₂ percentages in sputtering atmosphere (as indicated), along with the ϕ-scan of (10̅14) reflections of c-sapphire.](image-url)
5. Surface morphology

The surface morphology of Si-doped GaN films grown at different N\textsubscript{2} percentage was investigated and results are shown in figure S5 for typical cases, along with corresponding root mean square (rms) surface roughness data plotted against Ar and N\textsubscript{2} percentages in sputtering atmosphere.

**Figure S5.** Top view AFM images of typical Si-doped GaN films grown at (a) 100%, (b) 50%, (c) 30%, (d) 20%, and (e) 10% N\textsubscript{2} in sputtering atmosphere, along with (f) the rms surface roughness (●) of Si-doped GaN films plotted against Ar and N\textsubscript{2} percentages in sputtering atmosphere.