# Effects of sputtering pressure on the growth of AlGaN thin films using Co-sputtering technique

Nur Afiqah Othman<sup>a</sup>, Nafarizal Nayan<sup>a, \*</sup>, Mohd Kamarulzaki<sup>a</sup>, Zulkifli Azman<sup>a</sup>, Shuhaimi Abu Bakar<sup>b</sup>, Mohd Hafiz Mamat<sup>c</sup>, Mohd Zamri Mohd Yusop<sup>d</sup>, Mohd Yazid Ahmad<sup>e</sup>

<sup>a</sup> Universiti Tun Hussein Onn Malaysia (UTHM), 86400 Parit Raja, Batu Pahat, Johor, Malaysia

<sup>b</sup> University Malaya (UM), 50630 Kuala Lumpur, Malaysia

<sup>c</sup> Universiti Teknologi Mara (UiTM), 40450 Shah Alam, Selangor, Malaysia

<sup>d</sup> Universiti Teknologi Malaysia (UTM). 81310 Skudai, Johor, Malaysia

<sup>e</sup> Nanorian Technologies Sdn Bhd, 40 & 41-1, Jalan Kajang Perdana 3/2, Taman Kajang Perdana, 43000 Kajang, Selangor, Malaysia

\***Corresponding Author:** nafa@uthm.edu.my https://doi.org/10.55674/jmsae.v11i3.251506

Received: 29 January 2023 | Revised: 27 February 2023 | Accepted: 25 August 2023 | Available online: 1 January 2024

# Abstract

Aluminium gallium nitride (AlGaN) thin films provide promise for a variety of electronic devices due to their wide energy bandgap, which ranges from 3.11 - 6.40 eV. Here, a cosputtering approach utilising the RF and HiPIMS power supply of magnetron sputtering is used to deposit the AlGaN thin films. To examine their impact on the structural characteristics and morphology of the thin films, the AlGaN thin films were deposited under various sputtering pressures using the cosputtering technique. Following that, the films were examined using X-ray diffraction (XRD), atomic force microscopy (AFM), field emission scanning electron microscopy (FESEM), and surface profiling to determine their characteristics. XRD shows the polycrystalline AlGaN with (100), (101), (102), and (110) plane for the AlGaN deposited at a sputtering pressure of 3 mTorr and 5 mTorr with FWHM 0.622° and 0.732°, respectively. The increasing the sputtering pressure to 5 mTorr



is found to improve the crystallinity as well as the thickness of the AlGaN thin films from 234.27 - 1479.37 nm. AFM examination of the AlGaN film revealed a trend of increasing roughness and grain size together up to 3.25 nm and 47.22 nm respectively, with rising sputtering pressure from 1 - 7 mTorr. The co-sputtering of AlGaN can be successfully demonstrated in this study, and it is also shown that the sputtering pressure has a substantial impact on the development of AlGaN thin films produced using this technology.

Keywords: AlGaN; Co-sputtering; Room temperature; RFMS; HiPIMS

# Introduction

III-nitrides material has been widely used in fabrication laser diodes, photodetectors, biosensor, and etc. due to its superior properties, such as direct band gap, high electron mobility, high thermal conductivity, and etc [1]. Aluminium gallium nitride (AlGaN) is an alloy of aluminium nitride (AlN) and gallium nitride (GaN) that possesses useful semiconductor properties, as mentioned before. Due to Si's benefits, including its excellent thermal conductivity, huge size, low cost, and the tremendous potential of its compatibility with current processing methods created for Si integrated circuits, growing epitaxial films on Si substrates is a highly essential topic. However, the growth of epitaxial AlGaN on Si is usually with the presence of buffer layer of GaN or AlN due to the lattice mismatch between AlGaN and Si [2, 3]. Other than that, the growth of AlGaN epitaxial on

© 2024 Center of Excellence on Alternative Energy reserved



Si is usually deposited by a high temperature technique, more than 900 °C, which is metal organic chemical vapor deposition (MOCVD) [4], molecular beam epitaxy (MBE) [5, 6], and atomic laser deposition (ALD) [7, 8] to produce high crystallinity of AlGaN thin films.

Due to its benefits, magnetron sputtering is frequently used to create III-nitrides materials. These benefits include the use of large area targets that provide uniform thickness across the wafer and the ability to control the thickness through deposition and other parameters; the fact that sputtered films typically have better adhesion to substrates and others. Magnetron sputtering consists of several power supplies that can used to growth the thin films, such as dc magnetron sputtering (dcMS), radio-frequency magnetron sputtering (RFMS), and high power impulse magnetron sputtering (HiPIMS). The sputtering is the process when a solid surface (target material) at cathode is bombarded with energetic ions, surface atoms of the target are scattered with high energy and deposited on substrate at anode to form thin films as shown in Fig. 1 [9]. RFMS technique has been used to deposit nitrides film such as AlN and GaN. Asim mantarci have demonstrated the GaN thin films using RF magnetron sputtering and study their structural properties by varies the parameters form RFMS and succeed to deposit GaN using RF magnetron sputtering at 300 °C [10, 11]. AlN have been successful deposited on Si (100) using HiPIMS and obtain (002) plane orientation of AlN, which is favourable for the application in microelectronics and optoelectronics [12]. Therefore, based on the findings of these previous literature, the AlGaN thin films will be conducted using these two magnetron sputtering, which are RF magnetron sputtering and HiPIMS, to obtain high crystalline quality and study the effects of the sputtering pressure on the growth of AlGaN on Si (111) using this technique at room temperature.



Fig. 1 Mechanism of the co-sputtering technique

#### **Materials and Methods**

#### Substrate preparation of AlGaN thin films

The substrate that is used in this experiment, which is Si (111), must be cleaned first before it undergoes the deposition process. This cleaning was conducted to remove any oxide layer that is present on the surface of silicon since silicon material to oxides with oxygen. Hydrofluoric acid (HF) was diluted, and silicon (111) was dipped into it for one minute in order to clean it. The Si wafer was then rinse using the deionised water and dry using the nitrogen gas. The cleaning process was done and the Si wafer was immediately put into the sputtering chamber to undergo deposition process.

#### Deposition process of AlGaN thin films

Once the Si substrate is in the sputtering chamber, a turbo molecular pump was switched on to achieve the base pressure at  $8 \times 10^{-6}$  Torr. When the desired base pressure was achieved, the power supply was then turned on to ignite the plasma. GaN target and Al target with 3-inch diameter (purity 99.999%) was used and connected to the RF and HiPIMS power supply, respectively. RF power supply with 13.65 MHz (Advance Energy, Cesar RF Power), in the meantime HiPIMS power consisted of DC power supply (PsPlasma, SDC1024) and impulse power supply (Starfire Industries, SF-Impulse-SH). 30 W of RF power and 50 W of HiPIMS was set and Ar/N2 flow rate was maintained at 100/25 sccm using the mass flow controller. The distance between the target and substrate was 10cm, and the substrate was set at 5 rpm. The deposition process takes place for 2h without additional temperature. The sputtering pressure was varied at 1, 3, 5, 7, and 10 mTorr and using the load lock chamber to take in and out the sample from the sputtering chamber. The experimental details are shown in Table 1.

<b>Table 1</b> shows experimental details of AlGaN thin films
---

Parameters	Details
Base pressure (Torr)	$8 \times 10^{-6}$
Pulse width (µs)	200
Ar/N <sub>2</sub> flow rate (sccm)	100/25
HiPIMS Power (W)	50
RF Power (W)	30
Working pressure (mTorr)	1, 3, 5, 7, 10

#### Characterization of AlGaN thin films

X-ray diffraction (XRD) PANalytical X-Pert Powder scan at 2 thetas of  $20^{\circ} - 80^{\circ}$  with <sup>1</sup>/<sub>2</sub> divergence slit and the

incident angle "omega" at 0.50 degrees was used to study the structural properties of AlGaN thin films. Atomic force microscopy (AFM5100N-DFM) for Hitachi, field emission scanning electron microscope (FESEM) (Jeol: JSM-7600F), and surface profilometer (DektakXT) were used to study the surface morphology and the thickness of the AlGaN thin films, respectively.

# **Results and Discussions**

### XRD phase study

Fig. 2 presents the XRD spectra of AlGaN thin films at different working pressure at room temperature. No AlGaN diffraction peak was established at 1 mTorr sputtering pressure, indicating that the AlGaN films were amorphous at that time. The diffraction peak of polycrystalline AlGaN began to appear at 3 mTorr sputtering pressure. At this sputtering pressure, the intensities of AlGaN (100), AlGaN (101), AlGaN (102), and AlGaN (110) peaks were the highest, which demonstrated that the AlGaN thin films have good crystallinity under that condition. The AlGaN peaks remain the same when the sputtering increased to 5 mTorr. However, the AlGaN diffraction peak disappeared when the pressure increased up to 7 mTorr, which suggests that the AlGaN film was amorphous. Furthermore, above the sputtering pressure of 7 mTorr, the low intensities of AlGaN (100) and AlGaN (102) peaks illustrated the decline in crystallinity of AlGaN films.



**Fig. 2** XRD pattern of AlGaN thin films at different working pressure

Table 2 shows the value of the FWHM and degree of crystallinity of samples deposited at different sputtering

pressure. From the table, AlGaN with 3 mTorr sputtering pressure has the lowest FWHM and the highest degree of crystallinity, which means that AlGaN films that deposited using that sputtering pressure have good crystal quality. Meanwhile, there is only a small difference in FWHM and degree of crystallinity between samples at 5 mTorr and 3 mTorr. AlGaN films at 10 mTorr have a huge value of FWHM and degree of crystallinity. Theoretically, when the number of the particles ejected from the target was greater, the higher the energy of each ejected particle, which causes the cooling rate of the ejected particles on the substrate to be slower and making it relatively easier for the particles to become crystalline [13].

**Table 2** shows the structural parameters of AlGaN films at different sputtering pressure

Sputtering pressure (mTorr)	hkl	Position [°]	FWH M (°)	Crystallinity (%)
1			NA	
3	AlGaN (100)	33.272	0.622	47.920
5	AlGaN (100)	33.272	0.732	45.486
7	NA			
10	AlGaN (100)	33.048	19.112	36.878

In such circumstances, the crystallinity of AlGaN films increase with the increase of sputtering pressure. However, at certain sputtering pressure, the crystallinity of the AlGaN films started to decrease. These phenomena occur when the quantity of ionised Ar<sup>+</sup> ions grows with increasing sputtering pressure, resulting in a significant increase in the number of sputtered target particles. On the other hand, high sputtering pressure increases the possibility of target particles bumping with gas particles inside the chamber, increasing the likelihood that the target particles will lose energy in the collision and reducing the energy of a single target particle that is deposited on the substrate surface [13]. This is apparent from the Fig. 3, which shows the thickness of the AlGaN films with the function of sputtering pressure. The thickness increases with the increase of the sputtering pressure. However, at 7 mTorr, the thickness started to decrease with the increase in the sputtering pressure. As a result, there must be a sputtering pressure in the range 3-5mTorr, where the crystallinity of the AlGaN layer on the



surface substrate is the maximum. The AlGaN film exhibited the best crystallisation performance under the 3 mTorr condition because the AlGaN atoms diffused and migrated adequately, as shown in Fig. 2 and Table 2.



**Fig. 3** Thickness of AlGaN thin films at different sputtering pressure.

# Topological analysis by AFM

Fig. 4 and Table 3 show the AFM images and the results of surface roughness and grain sizes of AlGaN films, respectively. The rise in gas pressure had a substantial impact on the microstructure of the films, specifically the size of the grain. The grain size of the AlGaN films increases with the increase of sputtering pressure. The average roughness of the AlGaN films sputtered at various sputtering pressure is 2.05 nm. The roughness value increases, accompanied by the increase of sputtering pressure until 7 mTorr. However, the roughness slightly drops when the sputtering pressure is at 10 mTorr. According to the Li Ling et al. [14], increasing the sputtering pressure causes the enhancement of the bombarded Ar<sup>+</sup> on the target atoms. As a result, a high number of ejected target atoms will increase the occurrence of sputtered species scattering, lowering incident ion kinetic energy. Thence, at greater sputtering pressures, the mobility of the ions on the substrate surface is reduced, which explained the condition of AlGaN film roughness at pressure of 10mTorr.



**Fig. 4** AFM images of AlGaN films at different sputtering pressure (a) 1 mTorr, (b) 3 mTorr, (c) 5 mTorr, (d) 7 mTorr, and (e) 10 mTorr.





**Fig. 5** FESEM images of AlGaN at different sputtering pressures (a) 1 mTorr, (b) 3 mTorr, (c) 5 mTorr, (d) 7mTorr, and (e) 10 mTorr.

Further, the thickness of the films may also affect the roughness of the thin films [15, 16]. Fig. 3 shows the thickness of AlGaN film increases until 7 mTorr, where the film thickness started to decrease. As previously mentioned, as the sputtering pressure rises, more  $Ar^+$  is ionised to bombard the target as a result of a decrease in the average mean free path of Ar molecules and an increase in the probability of an electron-Ar molecule collision. Additionally, as the number of sputtered atoms rises, more target atoms are deposited on the substrate and their thickness rises, increasing surface roughness. This was consistent with the literature; Safdar et al. [17] investigated how thickness affected surface roughness and discovered

that when layer thickness increased, surface roughness increased as well.

Table 3	shows	results	of	AFM	analy	ysis
						/

Working pressure (mTorr)	Roughness (nm)	Grain size (nm)
1	0.29	19.90
3	1.54	56.57
5	2.75	69.58
7	3.25	47.22
10	2.42	66.52





Fig. 6 Mechanism of sputtering pressure (a) low sputtering pressure, and (b) high sputtering pressure.

# Morphological of AlGaN by FESEM

Fig. 5 shows the FESEM images of AlGaN thin films grown Si (111) substrate depending on the growth pressure. AlGaN on silicon substrate has pebble like structures although they have different growth conditions. It is obvious that granules have grown on the surface, especially when the sputtering pressure rises. The fact that increasing sputtering pressure causes columnar growth with enormous feature sizes as the sputtering pressure increases is a matter that is particularly evident in FESEM images, as discussed. The FESEM results are in agreement with the AFM results, which demonstrate that the AlGaN films generated under low sputtering pressure seemed to be more closely packed and had a larger surface covered than the film coated at strong sputtering pressure. This is due to the energy loss of Al and GaN particles when the sputtering pressure increases to form low dense of AlGaN films [18].

Theoretically, fluctuations in the number and energy of Ar+ ions as well as the energy of ejected Al and Ga atoms caused the sputtering working pressure to have an effect on the crystalline quality of AlGaN ejected atoms. At low sputtering pressure, the mean free path of the electrons increasing causes the reduction in collision with gas particles, such as Ar and N<sub>2</sub>. Higher energy of electrons can be restored in the long mean free path before colliding with Ar gas molecules to produce energetic  $Ar^+$  ions form the collision. Many positive  $Ar^+$  ions will be produced and bombarded into Al and GaN target molecules before eacting with N to form AlGaN films (Fig. 6(a)). Vice versa, as the sputtering pressure increase, Al and Ga sputtered atoms

struck the Ar<sup>+</sup> ions and radicals N in the plasma frequently, leading to energy loss of Al and Ga sputtered atoms. The mobility of Al and Ga sputtered atoms on the substrate surface decreased, which led to the formation of the low crystal quality of AlGaN films, as shown in Fig. 6(b). The crystalline quality of AlGaN was optimised by the deposition at poor sputtering working pressure and the accessibility of Al and Ga sputtered atoms on the substrate continued to increase with high energy. [19].

# Conclusion

In the current study, it was examined how sputtering pressure affected the morphological characteristics and crystalline quality of AlGaN films that were co-sputtered using the RFMS and HiPIMS techniques on Si (111). The polycrystalline of AlGaN films were grown on Si (111) substrate at sputtering pressure 3 - 5 mTorr. Surface of AlGaN thin films densely deposited at low sputtering pressure. Because of the variations in the amount of Ar+ ions and Al/GaN sputtered atoms with different energies, the sputtering pressure had an impact on the crystalline quality and structure of AlGaN sputtered films. The long mean free path of electrons at low sputtering pressure also affected the higher number of energy of Al/GaN sputtered atom to deposit on the AlGaN films. Since the collision frequency was low at low sputtering pressure, the energy of Al/GaN sputtered atoms rose. At low sputtering pressures of 3-5mTorr, the mobility of Al and GaN ejected atoms on the substrate surface increased, and the crystalline quality of AlGaN improved.



# References

- [1] Q. Hua, B. Ma, and W. Hu, Aluminum, Gallium, and Indium Nitrides, Encyclopedia of Material: Science and Technology. 3 (2021) 1 – 10.
- [2] Y. Li, W. Wang, Y. Lin, X. Li, L. Huang, Y. Zheng, Z. Zhang, G. Li, Growth of high-quality AlGaN epitaxial films on Si substrates. Materials Letters, 207 (2017), 133 136.
- [3] A. Jarndal, AlGaN/GaN HEMTs on SiC and Si substrates: A review from the small-signal-modeling's perspective, Int. J. RF Microw. Comput. Eng. 24(3) (2014) 389 – 400.
- [4] S. Kang, U. Chatterjee, D. Y. Um, I. S. Seo, C. R. Lee, Growth and characterization of n-AlGaN 1-D structures with varying Al composition using u-GaN seeds, J. Cryst. Growth. 480 (2017) 108 – 114.
- [5] Y. Cordier, Gallium Nitride (GaN): Physics, Devices, and Technology, CRC Press. (2017) 45–61.
- [6] H. Xin Jing, C.A. Che Abdullah, M.Z. Mohd Yusoff, A. Mahyuddin, Z. Hassan, Structural and optical properties of AlN/GaN and AlN/AlGaN/GaN thin films on silicon substrate prepared by plasma assisted molecular beam epitaxy (MBE), Results Phys. 12 (2019) 1177 – 1181.
- [7] S. Choi, A.S. Ansari, H.J. Yun, H. Kim, B. Shong, B.J. Choi, Growth of Al-rich AlGaN thin films by purely thermal atomic layer deposition, J. Alloys Compd. 854 (2021) 157186.
- [8] X.Y. Liu, S.X. Zhao, L.Q. Zhang, H.F. Huang, J.S. Shi, C.M. Zhang, D.W. Zhang, AlGaN/GaN MISHEMTs with AlN gate dielectric grown by thermal ALD technique, Nanoscale Res. Lett. 10(1) (2015) 1 6.
- [9] A.H. Simon, Handbook of Thin Film Deposition, Fourth Edition, Applied Science, (2018) 195 – 230.
- [10] A. Mantarci, Role of RF power in growth and characterization of RF magnetron sputtering GaN/glass thin film, Emerg. Mater. Res. 8(3) (2019) 320 – 330.
- [11] A. Mantarcı, M. Kundakçi, Physical properties of RF magnetron sputtered GaN/n-Si thin film: impacts of RF power, Opt. Quantum Electron. 51(3) (2019) 81.

- [12] D.L. Ma, H.Y. Liu, Q.Y. Deng, W.M. Yang, K. Silins, N. Huang, Y.X. Leng, Optimal target sputtering mode for aluminum nitride thin film deposition by high power pulsed magnetron sputtering, Vacuum, 160 (5) (2019) 410 – 417.
- [13] S. Lin, J. Zhang, R. Zhu, S. Fu, D. Yun, Effects of sputtering pressure on microstructure and mechanical properties of ZrN films deposited by magnetron sputtering, Mater. Res. Bull. 105(5) (2018) 231 – 236.
- [14] L. Li, T. Xue, Z. Song, C. Liu, F. Ma, Effect of sputtering pressure on surface roughness, oxygen vacancy and electrical properties of a-IGZO thin films, Xiyou Jinshu Cailiao Yu Gongcheng/Rare Met. Mater. Eng. 45 (8) (2016) 1992 – 1996.
- [15] X. Zhang, X.H. Song, D.L. Zhang, Thickness dependence of grain size and surface roughness for dc magnetron sputtered Au films, Chinese Phys. B. 19(8) (2010) 6 – 10.
- [16] V. Perumal , U. Hashim, SC. Gopinath, R. Haarindraprasad, W. W. Liu, P. Poopalan, A.R. Ruslinda, Thickness dependent nanostructural, Morphological, Optical and impedometric analyses of zinc oxide-Gold hybrids: Nanoparticle to thin film, PLoS One. 10 (12) (2015) 1 24.
- [17] A. Safdar, H.Z. He, L.Y. Wei, A. Snis, L.E. Chavez De Paz, Effect of process parameters settings and thickness on surface roughness of EBM produced Ti-6Al-4V, Rapid Prototyp. J. 18(5) (2012) 401 – 408.
- [18] S. Mobtakeri, Y. Akaltun, A. Özer, M. Kılıç, E.Ş. Tüzemen, E. Gür, Gallium oxide films deposition by RF magnetron sputtering; a detailed analysis on the effects of deposition pressure and sputtering power and annealing, Ceram. Int. 47 (2) (2021) 1721 – 1727.
- [19] M. Ohtsuka, H. Takeuchi, H. Fukuyama, Effect of sputtering pressure on crystalline quality and residual stress of AlN films deposited at 823K on nitrided sapphire substrates by pulsed DC reactive sputtering, Jpn. J. Appl. Phys. 55(5) (2016).