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A Review on the Different Techniques of GaN Heteroepitaxial Growth: Current Scenario and Future Outlook

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ABSTRACT

Although metal-organic chemical vapor deposition (MOCVD) is the most common technique to grow III-nitride films for light-emitting diode (LED) application, there are still several open questions such as the dislocations in LED structures and low thermal conductivity. The solutions to such problems have been approached by various deposition techniques over the past few years. In this review, the properties of gallium nitride (GaN) grown using different techniques and the consequences of the heteroepitaxial layers are discussed. At first, the general properties of GaN and its application for optoelectronic devices are presented briefly. Then, for the purpose of having a better crystallinity of GaN, it is necessary to identify and evaluate the defects present in the heteroepitaxial layers, which lead to poor crystal quality of films, and eventually to find an approach to clear up these issues. Several approaches using various substrates that have been published are discussed here and, finally, the directions of a new potential method for GaN growth using the magnetron sputtering technique are described.

Keywords: Aluminium-Gallium Nitride, Film Defects, Gallium Nitride.

1. INTRODUCTION

Gallium arsenide phosphide (GaAsP) and gallium phosphide (GaP) were first commercialized for red and green light-emitting diodes (LEDs), respectively. Previously, from the periodic table, gallium nitride (GaN) could be employed to fabricate blue LEDs, according to Amano [1]. A high temperature (2530 °C) and a very high pressure (45,000 atm) are needed to grow GaN. Thus, a chemical reaction is used to lower down the temperature and pressure for the growth of GaN. However, there was a problem of finding a suitable substrate material to grow high crystal quality of GaN films. The lack of substrate material match with the thermal expansion coefficient (TEC) of GaN has long prevented the use of GaN [2,3]. In 1986, Amano et al. demonstrated that by using metal-organic chemical vapor deposition (MOCVD), the GaN layer is possible on sapphire [4] as a result of the stability at high temperature, as the melting point of sapphire is 2040 °C [5]. Although the GaN was successfully grown on sapphire, the surface of the GaN was rough and its quality was very poor. The problem that arises with the sapphire substrate is that the lattice mismatch of the sapphire (0001) plane and the wurtzite GaN differs by ~13.8%–

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16.0% [4,6]. The TEC difference between sapphire and GaN is also considerably large, which normally generates poor crystallinity and defects formation such as the cracking effect in the GaN layer. To overcome these problems, buffer layers are employed [6]. Akasaki and Amano fabricated a small amount of aluminium nitride (AlN) which acts as nucleation layers at a low temperature. The process is recognized as "low-temperature-deposited buffer layer technology" [7]. Since then, several ways have been developed to deposit perfect GaN films on different substrates.

Due to its advantageous properties, GaN film has been widely used in many applications. Hu *et al*. fabricated an ultraviolet light-emitting diode (UV LED) using GaN by a combination of sputtering and MOCVD methods. The UV LED was reported emitting at 375 nm on a sapphire substrate [8]. The advantages of UV LEDs in comparison to traditional mercury lamps have raised great interests in the exploration regarding its applications [9,10]. Using AlGaN/GaN heterostructure on a silicon carbide (SiC) substrate, Chauhan and Sunny demonstrated a metal-oxide-semiconductor field-effect transistor (MOSFET) using GaN for high-power applications [11]. Other methods, such as that employed by Lee *et al.*, grew GAN on a silicon (Si) substrate for high-electron-mobility transistors (HEMTs) [12]. Ramizy *et al.* grew GaN onto Si (111) using AlN as a nucleation layer grown by plasma-assisted molecular-beam epitaxy (PAMBE) [13]. The GaN layer was then further tested for hydrogen gas sensor applications. Nakamura *et al.* developed AlN and GaN layers using MOCVD on a sapphire substrate for water-splitting photocathode [14].

As mentioned by Wu *et al.,* due to the large mismatch in the crystal lattice and large difference in the thermal expansion coefficient between GaN and the substrates, the film will start to have large dislocation density, mosaic crystallinity, biaxial stress and wafer bonding [15]. In this review paper, the defects present in the GaN films and how they are reduced in most of the studied substrates [16] using different heteroepitaxial techniques are presented. The paper begin with the general properties of GaN in Section 2. In Section 3, GaN growth using different techniques on different substrates are presented. The paper concludes with the future outlook in Section 4.

2. GENERAL PROPERTIES OF GAN

Gallium nitride (GaN) is one of the promising materials for many applications, such as light-emitting diodes (LEDs) [17], photoelectric detectors and high-electron-mobility transistors (HEMTs), considering that its direct energy bandgap is 3.4 eV [18–20]. It has excellent thermal stability, thermal conductivity (2.1 W cm^{-1} K^{-1}) at 300 K [21] and high electron mobility (~400 cm^2 V^{-1} s^{-1}) at 300 K [22]. GaN can crystallize into two forms, either zincblende or wurtzite structure [23]. However, GaN is mostly found in wurtzite structure. Generally, the conductivity of undoped GaN is an n-type [24]. In order for GaN to have a p-type conductivity, it must be doped with other materials such as magnesium atoms [25,26], while silicon atoms will lead to an n-type conductivity [27]. The properties of GaN are summarized in Table 1.

Table 1 Some basic properties of GaN

Properties	Value	References
Mass density	6.15 g cm^{-3}	[28]
CTE at room temperature	$\Delta\alpha/\alpha$: 5.59 × 10^{-6} K^{-1} $\Delta c/c$: 3.17 × 10^{-6} K^{-1}	[28]

	$\Delta\alpha/\alpha$: 3.1 × 10^{-6} K^{-1} $\Delta c/c$: 2.8 × 10^{-6} K^{-1}	[23]
Lattice constant	a = 3.189 × 10^{-8} cm c = 5.185 × 10^{-8} cm	[28–31]

3. OVERVIEW OF EPITAXY TECHNIQUES

Regardless of applications, GaN requires good crystal quality films. Several epitaxial methods have been suggested to obtain better crystal quality of GaN, such as hydride vapor phase epitaxy (HVPE), metal-organic chemical vapor deposition (MOCVD), molecular-beam epitaxy (MBE), and pulsed laser deposition (PLD). Some were summarized by Denis *et al.* [32], Nasser et al. [33] and Qiang [34], and a summary of several methods to grow GaN is given in Table 2.

Table 2 Summary of GaN film growth using different techniques

Method/Type of substrate	Analysis	Growth temperature/pressure	References
MOCVD/Si (111) and etched (001)	Energy Dispersive Spectroscopy (EDX), X-ray Diffraction (XRD), Photoluminescence (PL) spectra	AlN: 1050–1200 °C; GaN: 1000–1150 °C/ atmospheric pressure (101.32 kPa)	[35]
Selective area growth (SAG) method in MOCVD/patterned Si (110)	Scanning Electron Microscopy (SEM), Atomic Force Microscopy (AFM), Cathode Luminescence (CL) spectroscopy	1060–1100 °C/ 1.33–6.67 × 10^4 Pa	[36]
HVPE/sapphire	SEM, XRD, CL	GaN: 1050 °C; Graphene; CVD: 1200 °C	[37]
MOCVD/Si (111)	AFM, XRD ω-scan	1100 °C/ GaN: 20 kPa; AlN: 5 kPa	[38]
HVPE GaN:Ge/ ammonothermal – GaN substrate	XRD, optical microscopy, Raman spectroscopy, Capacitance-Voltage (CV) technique, Hall measurements, Secondary Ion Mass Spectrometry (SIMS), PL	850 °C and 1045 °C	[39]
High temperature vapor phase epitaxy (HTVPE)/(0001) sapphire	Differential Interference Contrast (DIC) optical microscopy, SEM, XRD, PL, SIMS, Glow Discharge Mass Spectroscopy (GDMS)	1350–1400 °C/ 20–985 mbar	[40]
HVPE/sapphire	Electroluminescence (EL), PL, microCL, High-Resolution X-ray Diffraction (HRXRD)	1040 °C/ atmospheric pressure	[41]
MBE/sapphire	XRD rocking curve, SIMS, AFM	800–900 °C	[42]
MBE/HVPE-grown GaN substrate	Transmission Electron Microscopy (TEM), SIMS, AFM	660–76 °C	[43]
MBE/4H-SiC	Reflection High-Energy Electron Diffraction	700–800 °C	[44]

	(RHEED), Field-Emission Scanning Electron Microscopy (FESEM), AFM, X-ray Photoelectron Spectroscopy (XPS), HRXRD, PL		
MBE/HVPE-grown GaN substrate	TEM, AFM, SIMS	600 °C/ 10^{-6}–10^{-7} Torr	[45]

3.1 Halide vapor phase epitaxy

GaN can be grown using the halide vapor phase epitaxy (HVPE) method. HVPE was the first technique to grow GaN with single-crystal films [34]. HVPE is done in a quartz HVPE reactor which consists of two zone reactors (source and growth zones) for different temperature levels, as shown in Figure 1 [39]. The ammonia (NH_3) nozzle is placed at an equivalent level of the susceptor. Gallium chloride (GaCl) and germanium tetrachloride ($GeCl_4$) are supplied vertically through a spray-type nozzle placed above the substrate. The substrate is placed on a rotating susceptor disc on the right side of the HVPE reactor. GaCl is achieved through the reaction of hydrochloride (HCl) and Gallium (Ga) at 850 °C. In the growth zone, at 1045 °C, GaCl is transferred by the carrier gas and mixed with NH_3 to form GaN [45]. The most-reported papers used the HVPE technique to grow bulk GaN crystals and freestanding GaN substrate [46,47].

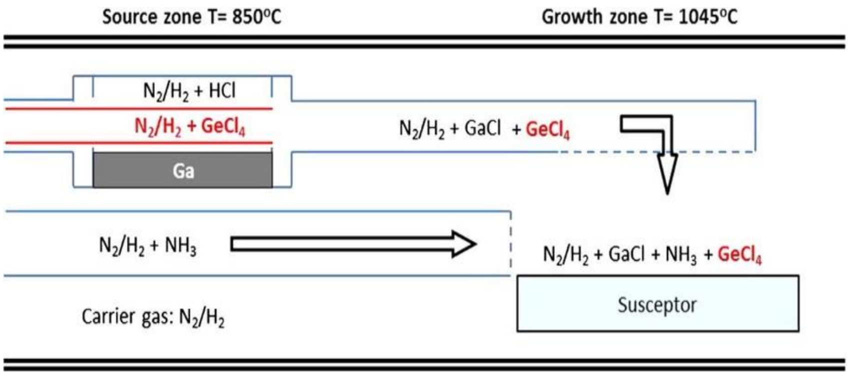

Figure 1. Schematic diagram of horizontal HVPE reactor [39].

3.2 Metal-organic chemical vapor deposition

Metal-organic chemical vapor deposition (MOCVD) is a long-established technique used for heterostructure growth to make electronic devices [48]. MOCVD is also called metal-organic vapor-phase epitaxy (MOVPE) [34]. This technique requires a constant flow of gases which will react chemically in a high-temperature chamber to form the GaN layer. Two types of MOCVD reactor that are readily available in the market are, namely, horizontal and vertical reactors. Figure 2 shows a schematic view of an MOCVD system [33]. Trimethylgallium (TMGa) reacts with NH_3 in a high-temperature reactor for the growth of GaN [31]. A carrier gas such as hydrogen (H_2) is needed to deliver the metal-organic precursors. Additional precursors such as trimethylaluminium (TMA) and methylsilane (SiH_3CH_3) are also used [43]. Several reactor concepts have been developed to improve the flow of gases on a wide surface area and within multi-wafer concepts.

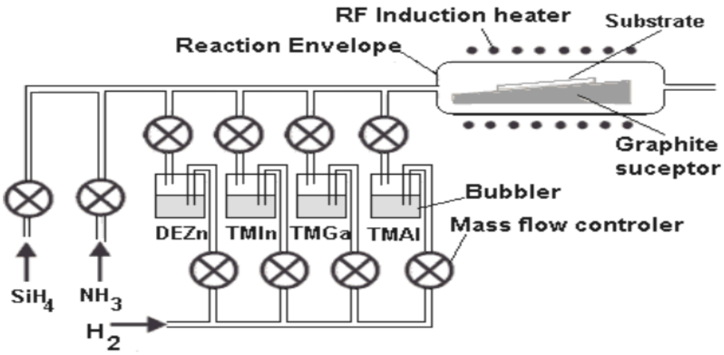

Figure 2. Schematic diagram of MOCVD technique [33].

3.3 Molecular-beam epitaxy

Another method to grow GaN is molecular-beam epitaxy (MBE) [49]. It includes plasma-assisted MBE (PAMBE) and laser MBE [50]. The MBE process occurs when there are reactions among the molecular thermal energy, the atomic or ionized beams of each element in high-temperature and ultrahigh-vacuum environments [23]. The molecular beam of Gallium (Ga) comes from effusion cell sources. N radicals are achieved by radio-frequency (RF) plasma or ammonia source as a result of very high binding energy. Throughout the process, the substrate is typically rotated and the growth temperature is up to 800 °C. A basic MBE growth chamber is shown in Figure 3 [51].

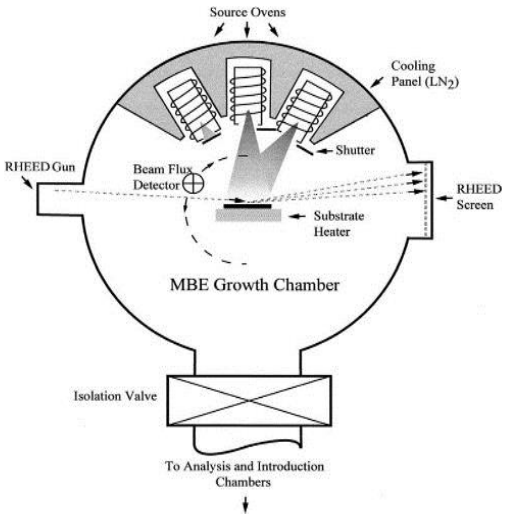

Figure 3. Schematic diagram of the top view of a simple MBE chamber [51].

4. RECENT PROGRESS ON THE GROWTH OF III-NITRIDE FILMS

Other than the fabrication techniques, the choice of substrate is very crucial in order to achieve good crystal quality of GaN. An experiment conducted by Yang *et al.* on the deposition of GaN on three different substrates of Si (111), sapphire (0001) and 4°-miscutting orientation by plasma-assisted molecular-beam epitaxy shows that the substrates affect the growth mechanism and physical properties of the GaN films [49].

Based on a previous study, AlN is one of the most suitable buffer layer materials for GaN growth [52]. The AlN layer serves as a barrier layer for silicon and gallium. Furthermore, in order to obtain GaN films with fewer defects (compressive strain), AlN layer is suggested, as it gives low lattice mismatch [53]. AlN is the binary material in the III–V nitride group, having a wurtzite structure in a hexagonal crystal form. AlN also is a potential material for LEDs, laser diodes [54] and insulating ceramics for piezoelectric sensors [55]. It has a large bandgap of 6.2 eV [18,28] and good thermal conductivity of 285 W m^{-1} K^{-1} [56]. The characteristics of AlN are shown in Table 3 and the fundamental crystal structure of AlN is shown in Figure 4.

Table 3 Some basic properties of AlN

Properties	Value	References
Melting temperature	2200 °C	[55]
Mass density	3.23 g cm^{-3}	[28]
Lattice constant	a = 3.1114 Å c = 4.9792 Å	[57]
CTE at room temperature	Δα/α: 4.2 × 10^{-6} K^{-1} Δc/c: 5.3 × 10^{-6} K^{-1}	[28–31]

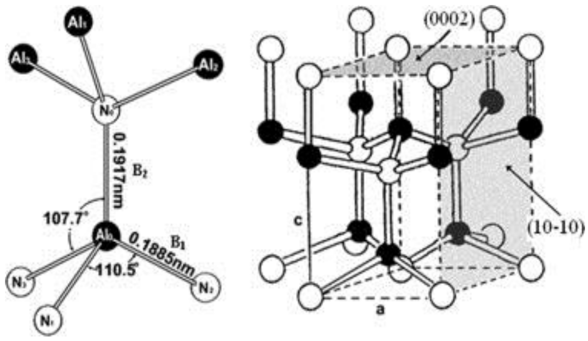

Figure 4. Schematic diagram of wurtzite AlN crystal structure [58].

4.1 Growth of GaN on sapphire substrate

Since sapphire (α-Al_2O_3) has a high melting point, it is good enough for the deposition of films at very high temperature. It also has high thermal conductivity (40 W m^{-1} K^{-1}) [24]. The schematic diagram in Figure 5 shows the principle of the growth of GaN on a sapphire substrate [31]. The difference in the lattice mismatch of sapphire and GaN is 13.6% [13].

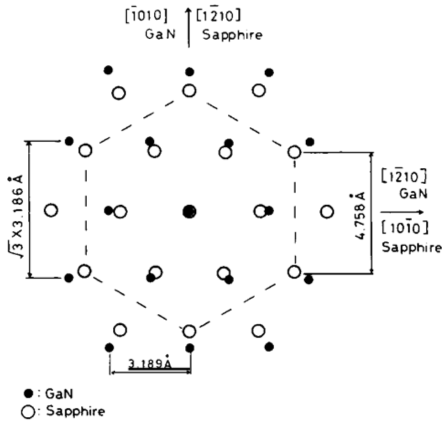

Figure 5. Crystal structure of GaN and sapphire [31].

4.1.1 Growth of GaN on sapphire substrate using HVPE

Hiramatsu *et al.* fabricated GaN using horizontal quartz reactor HVPE on a (0001) sapphire substrate [59]. They used a low-temperature buffer layer in this experiment. The temperature of the first region (source zone) was set at 850 °C, while the second region (growth zone) was at 1090 °C. They reported that when a stripe tungsten mask pattern is performed, the scanning electron microscope (SEM) image shows that the smooth surface can easily be obtained without pits compared to the case of SiO_2 mask where the surface has many pits and the absence of voids, as in Figure 6(a). Generally, most of the pits originate from threading dislocation [60,61], while the presence of the unintentional voids is from relaxed stress as a result of the thermal mismatch between the film and the substrate [62]. The measurement of X-ray rocking curve (XRC) also shows that GaN with SiO_2 mask has a broad full width at half maximum (FWHM), which indicates poor crystal quality. From the transmission electron microscope (TEM) images, there are no dislocations from the stripe tungsten mask, while for the SiO_2 mask, a dislocation comes from the center of the mask and goes through to the surface of the GaN layer.

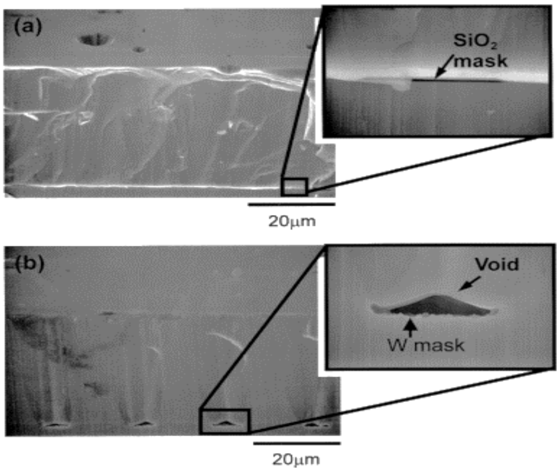

Figure 6. SEM cross-section images of GaN on (a) SiO_2 and (b) stripe tungsten mask [59].

Dwikusuma *et al.* fabricated GaN using a vertical HVPE reactor at a high temperature of 985–1100 °C [63]. The sapphire substrate is nitridated at 1100 °C before the deposition of GaN. The AlN layer comes from the nitrogen incorporation during the nitridation process [64], as proved in Figure 7. Comparing the suitability of the lattice crystal in growing GaN on sapphire (~16%), AlN is used for the deposition because of its lower lattice mismatch (~3%). The smaller energy barrier for the growth of GaN is used due to the presence of the smaller lattice mismatch.

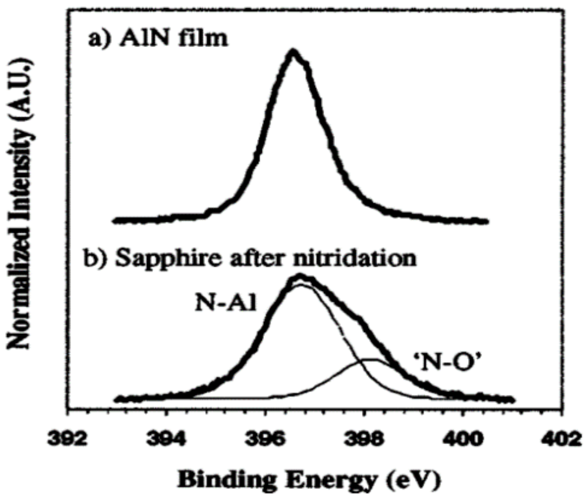

Figure 7. XPS peaks of (a) standard AlN film and (b) sapphire substrate after nitridation [64].

4.1.2 Growth of GaN on sapphire substrate using MOCVD

Huang *et al.* successfully fabricated GaN on sapphire using closed-coupled showerhead (CCS) MOCVD for high-electron-mobility transistor (HEMT) [6]. AlN with double-step growth at high temperature (HT-AlN) acts as the buffer layer. GaN film obtained by depositing on HT-AlN has better crystalline quality for HEMT application. However, the film has mixed polarity domains, resulting in poor surface morphology and decreased resistivity [65]. This may be due to the occurrence of unintentional nitridation at high growth temperature, resulting in the formation of amorphous AlN layer. Therefore, to enhance the crystal quality and to eliminate polarity domains, a two-step AlN layer is fabricated. As a result, the grown GaN layer has a low density of threading dislocations and leakage current. Besides, the result of the HEMT device made from the usual photolithography and lift-off process shows that the GaN layer grown on a two-step-temperature AlN buffer is capable of producing a high-resistance film for HEMT device application.

The presence of a large density of threading dislocation (TD) is commonly known as the result of large differences in the lattice crystal and TEC of both the GaN films and the substrate. The TDs result in bad performance of GaN devices. To reduce the TDs on GaN films, different approaches have been studied, including microscale SiO_2-patterned mask [36], epitaxial lateral growth (ELOG) [65], defect selective passivation and patterned sapphire substrate. Recently, Chen *et al.* reported that when a GaN layer is deposited on sapphire and the sputtered AlN film is made as a buffer layer, a low-TD film is obtained [66]. The reduction in TDs is due to the development of basal plane stacking faults (BPSFs) at different heights, which prevent the TD extension during the growth. A large area of nucleation islands achieved using the sputtering process and the deposition GaN films leads to the formation of the stacking faults, hence proving that the AlN layer deposited using sputtering for the buffer layer improves the crystal quality of GaN.

Aida *et al.* grew a 3-µm thin film of GaN on a sapphire substrate [67]. The surface roughness on the back of the sapphire substrate was studied by them regarding the bowing characteristic of GaN at temperature ranging from 25 °C to 800 °C. Throughout the epitaxy process, the bowing phenomenon of the sapphire substrate can occur due to the presence of heteroepitaxial strain coming from the differences in lattice constant and TEC. In their study, the change of the substrate bowing is measured from the FWHM of the rocking curve measurement. The study shows that GaN grown with a smooth back surface roughness of the sapphire substrate results in a big amount of bowing. The GaN on the sapphire substrate is bowed in the convex direction at room temperature and, with increasing temperature, the bowing reduces, becoming flat and then becomes concave. These results are consistent with a previous study which shows that temperature will affect the bowing of the substrate [68].

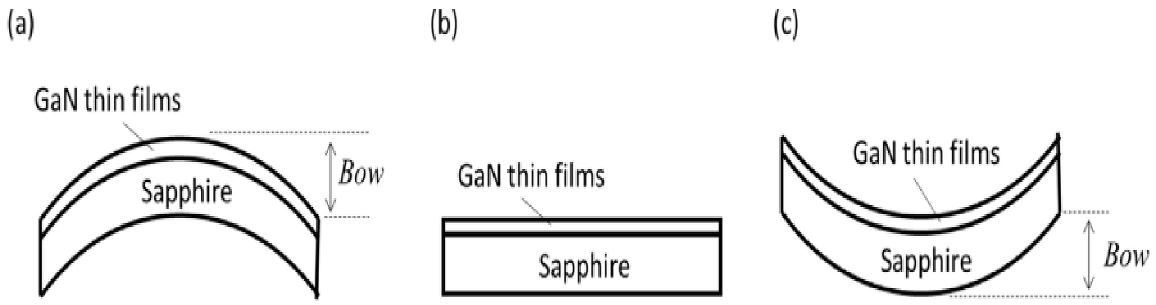

Figure 8. Bowing characteristics of (a) convex, (b) flat and (c) concave shapes [67].

Chen *et al.* developed a two-step temperature process to achieve homogeneous nucleation layer and better crystallinity of GaN nanostructure deposited on top of a patterned sapphire [69]. Comparison with a single-step temperature growth shows that although there is a huge change in the crystal quality and crystal orientation with rising temperature, the homogenous nucleation layer is degraded. In the first step, the temperature was set at 950 °C, resulting in the advanced nucleation layer which is a thin AlN, and the second step was set at 1040 °C for the GaN growth. The TEM image in Figure 9(a) shows that two TDs with the screw type are observed and no edge-type TD is observed. Furthermore, a few stacking faults are present in this case, as shown in Figure 9(b). This result indicates that the crystal quality of SAG of the GaN nanostructure is significantly improved using a very thin mask (5 nm).

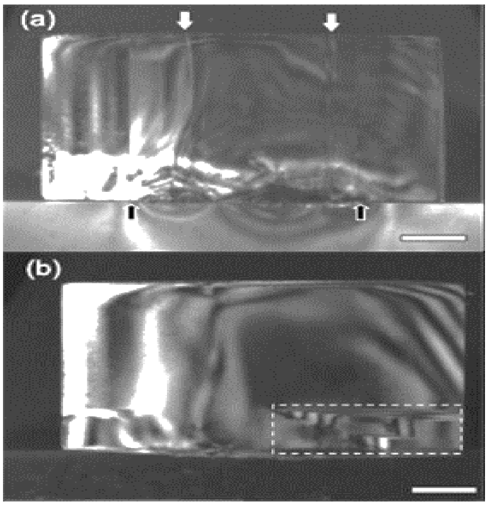

Figure 9. TEM images of GaN films [69].

4.1.3 Growth of GaN on sapphire substrate using MBE

Dixit *et al.* grew GaN films on a pre-nitridated sapphire substrate using a laser molecular-beam epitaxy (LMBE) system that comes with *in situ* reflection high-energy electron diffraction (RHEED) and a radio-frequency (RF) nitrogen plasma source [50]. They studied the deposition temperature (500–700 °C) of the grown layer of GaN. They estimated the presence of stress in the GaN layer using omega-2 theta, as in Figure 10, and found that the GaN epitaxial layer has a large in-plane compressive stress at low growth temperature, while at high growth temperature (700 ̊C), the strain and stress in the layer are drastically reduced. They believed that the deposition temperature changes the growth mode of GaN. At high growth temperature, GaN grows from grain to an island growth, resulting in the reduction of biaxial strain.

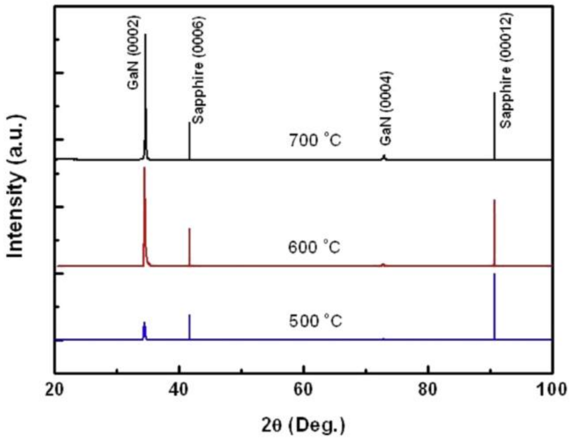

Figure 10. HRXRD 2θ/ω spectra of GaN layers [50].

4.2 Growth of GaN on silicon carbide (SiC) substrate

Due to the advantages of its high strength, low thermal expansion, good thermal shock resistance and high thermal (4.5 W cm^{-1} K^{-1}) and electrical conductivities [29], silicon carbide (SiC) substrate is one of the good candidate materials for high-power LEDs [70,71]. Additionally, the difference in the thermal expansion between SiC and GaN is as low as ~3.5% [72]. The difference in the coefficients of the thermal expansion of GaN and SiC is also low, which is ~3.2% [70,72].

4.2.1 Growth of GaN on SiC substrate using MBE

Tian *et al.* successfully fabricated high-quality GaN crystal on 6H-SiC using HVPE with a two-step growth process, which was then directly self-detached from the SiC for the use of GaN wafers [74]. The experiment was conducted at atmospheric pressure in a home-built vertical HVPE reactor. The first zone temperature was set at 500–800 °C, while the second zone temperature was at 1050 °C. From the PL spectra measurement in Figure 11, the freestanding GaN layer has good optical quality since, at a wavelength of 500–600 nm, the presence of a very weak yellow luminescence indicates a low density of native defect.

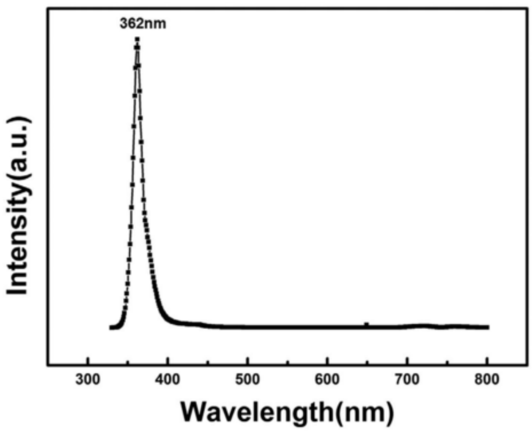

Figure 11. PL peak of the freestanding GaN [74].

4.2.2 Growth of GaN on SiC substrate using MOCVD

Helali *et al.* grew GaN on a 4H-SiC substrate using MOCVD [16]. A Si_3N_4 is fabricated as the passivation layer for the HEMT device performance. The passivation layer is used to overcome the trapping effects existing at the surface and in the GaN buffer layer. The trapping effects reduce the breakdown voltage and the output current. The passivation layer using Si_3N_4 is believed to reduce the electrically active surface traps, resulting in the increase of the maximum power of the RF power recovery. The high crystalline quality of GaN material was successfully grown on SiC substrates by di Forte-Poisson *et al.* [75]. A low-pressure MOCVD was used to grow the GaN. They reported that the structural quality of the GaN improves when AlN nucleation layer is employed. A comparison of the HR-XRD measurements has been made with GaN grown directly on SiC, as shown in Figure 12. Figure 12(a) shows the rocking curve of FWHM of GaN with AlN as the nucleation layer and Figure 12(b) shows the GaN grown directly on SiC substrate.

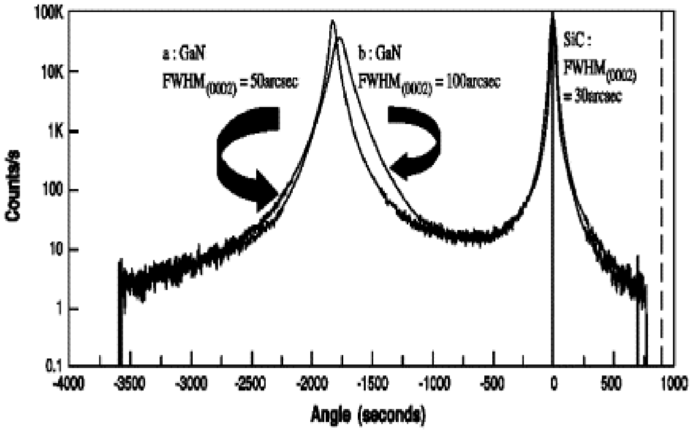

Figure 12. X-ray rocking curve of a GaN/SiC heterostructure on (a) treated SiC and (b) pure SiC.

4.2.3 Growth of GaN on SiC substrate using MBE

Yun *et al.* deposited GaN on a porous SiC using MBE by employing NH_3 as the nitrogen source [76]. They believed that by using a porous template, the nanopatterned porous structure can promote the growth of the film, resulting in less presence of defect density. They also studied the effect of the skin layer (~60 nm) present at the surface of the substrate. The skin layer in this work is defined as a layer where most pores are buried. From the TEM measurement, they found that the dislocation distribution is significantly dependent on the skin layer. The dislocation density with the skin layer is about $\sim 5 \times 10^9$ cm^{-2}, while when the skin layer is detached, the dislocation density is reduced to $\sim 1 \times 10^9$ cm^{-2}. This shows that the crystal quality of GaN is better without the skin layer. Also, PL spectra shows that the FWHM of the GaN films without the skin layer is smaller compared to GaN with the skin layer.

4.3 Growth of GaN on silicon (Si) substrate

Silicon is the most used semiconductor and substrate material due to its cost efficiency [77], large size [17,21], excellent thermal stability [21] and high thermal conductivity (130 W/m^{-1} K^{-1}) [77,78]. However, there are several problems, such as the huge difference in the lattice constant and the difference in the CTE with GaN [21,77], resulting in material defects and quality and device reliability. However, the silicon substrate itself is defect-free. To overcome these problems, several epitaxial procedures have been developed. Several works have been published about the deposition of GaN on a silicon substrate [80]. It has been done by changing parameters such as the thickness of the buffer layer, deposition temperature, material concentration and flow rate of the gases.

4.3.1 Growth of GaN on Si substrate using HVPE

Bessolov *et al.* grew a semipolar AlN and GaN using HVPE on a planar Si (100) substrate [81]. A SiC layer with thickness of ~100 nm as a buffer layer was deposited on a silicon substrate by solid-phase epitaxy. The SiC layer was then followed by an AlN buffer layer with a thickness of ~300 nm and the GaN layer. The SiC was deposited at 1280 °C, AlN at 1080 °C and GaN at 1050 °C. Before deposition, the Si (100) surface was cleaned with a chemical etching agent. The silicon was misoriented by angles 2°, 4° and 7° in the < 011> direction. However, the study shows that the presence of defects such as stacking faults and threading dislocations are not dependent on the substrate misorientation.

4.3.2 Growth of GaN on Si substrate using MOCVD

A single crystalline of GaN deposited using MOCVD on an n-type silicon (111) substrate has been claimed by Uen *et al.* [82]. The effect of the substrate nitridation temperature where the silicon nitride (SiN_x) was formed by a nitridation process in the MOCVD reactor was studied. The silicon wafer was cut by 4° towards the <011> direction. They reported that when appropriate nitridation temperature is given, which is higher than 950 °C, a defect resulting in a broad emission of yellow luminescence is not present in the photoluminescence (PL) spectra, as shown in Figure 13. The yellow luminescence (YL) is present due to defects which act as deep acceptors [82,83] and is known as the dominant defect related to the PL bands [85]. Two defects that can be observed from the PL can be found at 3.4 eV, which belongs to the hexagonal GaN, and the broad YL transition at 2.3 eV, which may come from deep-level impurities and/or lattice defects [24,82]. These induced defects can greatly disturb the performance of devices, especially for laser diodes.

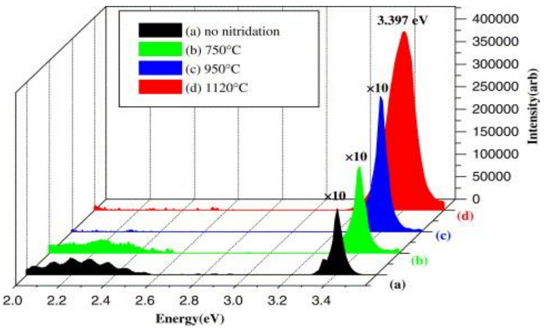

Figure 13. PL spectra of GaN epilayer [82].

Abd Rahman *et al.* obtained high quality of single crystalline of GaN grown on silicon (111) using MOCVD [86]. The influence of the nitridation times (40, 220 and 400 s) on GaN crystalline quality was studied. In order to remove the native oxide on the surface, the substrate is annealed inside a chamber at 1125 °C for 10 minutes under H_2 ambience before the growth process is conducted. It is reported that the crystallinity and surface morphology of the GaN layer become much better after the silicon is nitridated for 400 s at 1000 °C. AFM image shows that the sample is uniform and homogenous with small grain size of 4.247 nm and the RMS roughness of the sample is 1.475 nm. After the nitridation, the process is then continued by depositing a thin AlN nucleation layer as the buffer layer and 40 pairs of AlN/GaN multilayer as the strain-compensation layer. The XRD spectra for phi-scan analysis shows that the GaN has good single-crystal quality with a hexagonal structure. A previous study stated that, by adding super-lattice interlayers as the strain-compensating layer, the substrate bowing could be reduced [87]. The bowing of the Si wafers is caused due to the large mismatch between the CTEs of GaN and Si. The bowing of the sample recorded is 51.99 µm. A smooth layer structure with no crack and pits is observed from the FESEM image.

Yamada *et al.* demonstrated the deposition of GaN on a Si (111) substrate with an AlN intermediate layer deposited at 350 °C using a reactive sputtering method [17]. The GaN layer is grown by the facedown horizontal MOVPE system at 1100 °C and at low pressure. Compared with GaN films grown with AlN intermediate layer deposited at 1200 °C using the MOVPE method, the initial nucleus density of GaN is about three times smaller than that of GaN films grown with AlN intermediate layer deposited at 350 °C. The small pit density of the deposited film when AlN is used as the intermediate layer is due to the small nucleus density of GaN. By introducing AlN as the nucleation layer, a smooth surface of GaN with Ga-polar is observed. Apart from that, the direct growth of GaN suffers from the high possibility of the presence of amorphous Si_3N_4 [52].

Wang *et al*. developed AlN templates on a Si (111) substrate using PLD at 850 °C [21]. They reported that the quality of the single crystal of AlN template with smaller surface roughness results in good crystal quality of GaN films. From the XRC measurement, according to the XRC results, and comparing the results with GaN grown without the AlN nucleation layer, the screw-type threading dislocations including edge and mixed types are reduced extremely. A high density of TDs in the order of 10^9–10^{10} cm^{-2} exist in the GaN film on the silicon substrate due to a large lattice mismatch. The TDs are the most defects found in the GaN films and are present between the substrate and the epitaxial layer [88]. The TDs significantly affect the performance of the GaN-based devices. The TDs present on the growth of GaN on Si include the pure edge, the pure screw and mixed dislocations [79]. The advantages of the *ex situ* low-temperature growth include overcoming the chemical reactions between the Si substrate and the films, decreasing the kinetic energy that will diffuse between Si and nitride, preventing thermal diffusion, lowering power consumption and enhancing productivity. Results obtained by Wośko *et al.* show that the LT-AlN interlayers during the growth of GaN on Si (111) have advantages in lowering the stress of the deposited GaN layer and thus extending its critical thickness [89].

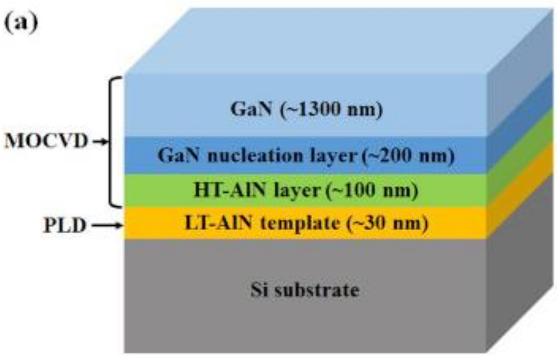

Figure 14. Schematic diagram of samples grown by both PLD and MOCVD [21].

Crack-free GaN grown using MOCVD on an n-type Si (111) substrate is reported by Li *et al.* [78]. By using different pressures to grow the AlN buffer layer of 40-nm thickness per layer, high quality of GaN with the absence of crack is obtained. The GaN film is grown without the AlN nucleation layer because the roughness of the nucleation layer when doped decreases the electrical performance of devices. The study also shows that a 3D-grown GaN can lessen the tensile stress and, when the 3D growth time is prolonged, the residual stress in the top layer of GaN decreases, resulting in the reduction of cracks. The threading dislocation (TD) densities of the sample obtained by AFM is about 5.2×10^8 cm^{-2} and the RMS roughness is 0.238 nm.

Iwata *et al.* fabricated a GaN layer using MOVPE on a (111) Si substrate [90]. They reported the influence of rapid thermal annealing (RTA) on the crystal defects. The RTA is conducted inside an infrared gold image furnace. The AlN nucleation layer is grown at 970–1220 °C, while the AlInN buffer layer is deposited at 720 °C. According to the study, the RTA does not affect the yellow emission band (YL), as shown in Figure 15. However, using the TEM measurement, they learned that threading dislocations with the half-look screw type shrink/move when RTA is done at 600–700 °C. Nevertheless, no change is observed regarding the edge and mixed types of dislocations.

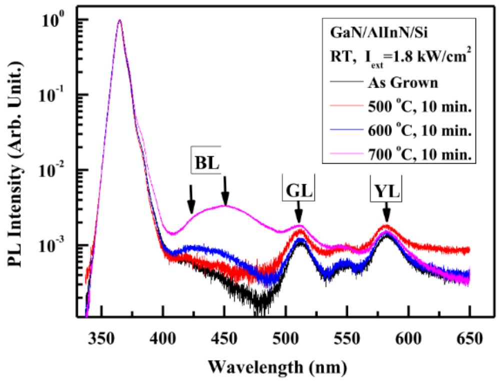

Figure 15. PL spectra of the GaN films [90].

Ji et al. grew high-quality GaN using MOCVD assisted with *in situ* NH_3 cleaning process on a (111) Si substrate [91]. They prepared two samples of GaN, of which one sample is without the cleaning process and the other one is *vice versa*. The new type of cleaning process was invented to clear up the particles of AlN, GaN, *etc.* and metal droplets of Al, G, *etc.* because it is hard to clean these using the normal cleaning process. The difficulty is that, at high temperature, the coating of the metal droplets on the post-growth flange and reactor will vaporize, resulting in the deposition onto the Si substrate together with the trimethyl-aluminium (TMAl) preflow. By observing two different samples using optical microscopy, they found a lot of pits present on the surface of the GaN layer without the new cleaning process, which is related to the intersection of threading dislocations. On the other hand, the sample that undergoes the new cleaning process has very smooth surfaces with a clear sample, as shown in Figure 16. This result is supported with FWHM of HRXRD, which shows that samples with the new cleaning process have better crystal quality.

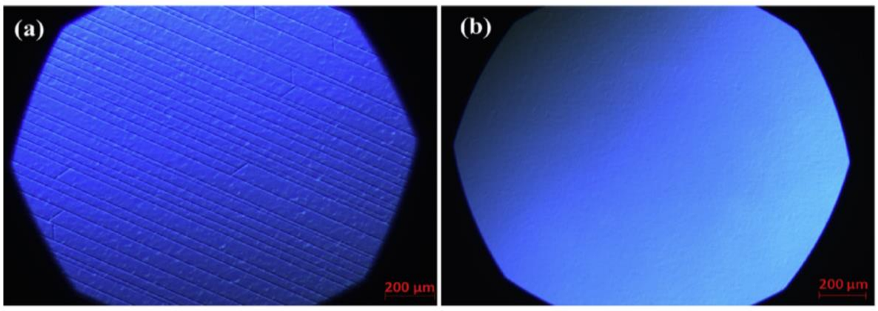

Figure 16. GaN surface layer (a) without new cleaning process and (b) with new cleaning process[91].

4.3.3 Growth of GaN on Si substrate using MBE

Wang *et al.* studied the effect of Si doping of GaN on Si (111) [92]. Using a buffer layer of SiC, the GaN:Si and GaN films are grown using MBE. From the AFM measurements, the undoped GaN film contains plateau-valley morphology with large, atomically flat terraces on its surface. This kind of valleys is believed to disturb the device performance. The study shows that the dislocation density decreases from about 1×10^{10} to 1×10^{9} cm^{-2} when the GaN film is doped with Si. From the TEM images shown in Figure 17, it is found that the density of stacking faults (SFs) and cubic phase increase It is also found that the main dislocations present in the doped GaN are pure-edge dislocations, while in the undoped GaN the types of the dislocation present are mixed and edge dislocations. The decrease of the dislocation density in the doped GaN may be due to the small density of threading dislocations with the mixed type because of the existence of a large density of SFs.

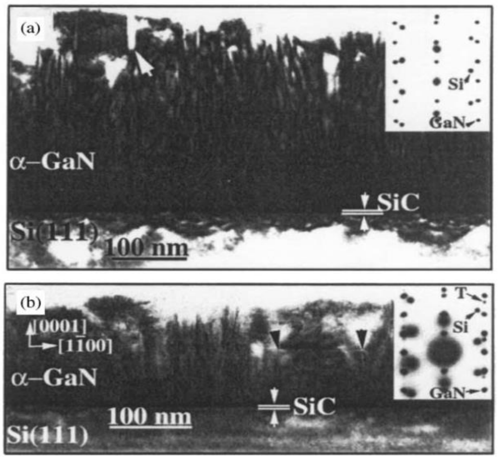

Figure 17. TEM cross-section images of (a) undoped GaN and (b) Si-doped GaN [92].

As stated by Ji *et al.* [91] and reviewed by Li *et al.* [93], it can be summarized that direct growth of GaN on Si substrate still faces reproducibility and reliability issues because of:

1. The chemical reaction between GaN and Si (meltback etching phenomenon).
2. The presence of dislocation density with values of more than 10^{8} cm^{-2} due to the high difference of lattice mismatch between GaN and Si.
3. The large difference in CTE that leads to the generation of tensile stress during the cooling down, causing the cracking effect.

5. OVERVIEW OF SPUTTERING TECHNIQUES

Sputtering is one of the methods to deposit III-nitride films. Different types of sputtering available, such as DC and RF magnetron sputtering, pulsed sputtering and currently the high-power impulse magnetron sputtering (HiPIMS), are widely used to deposit the III-nitride films. These kinds of sputtering use different power sources and can sputter different types of materials. The sputtering process is based on the ion bombardment of the target towards the substrate, resulting in the formation of films, as shown in Figure 18. The process is done in a high-vacuum chamber (less than 1×10^{-6} Torr) to get high crystal quality. Different parameters can be controlled during the sputtering process, such as sputtering pressure, bias voltage of substrate, temperature, target-to-substrate distance, flow rate of process gas, deposition time and sputtering power.

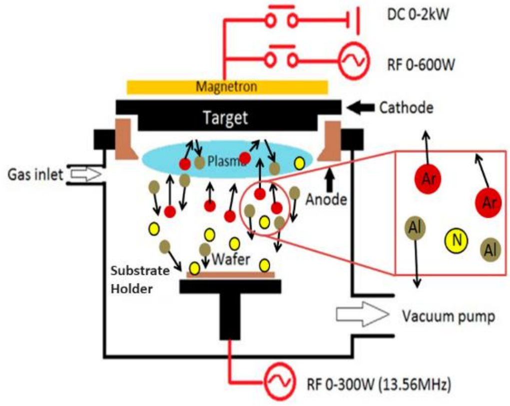

Figure 18. Schematic diagram of sputtering process [94].

6. SUMMARY AND FUTURE OUTLOOK

From this review, it can be concluded that the properties of GaN films are dependent on the parameters and the methods used to grow GaN. The reviewed GaN growths use AlN as the buffer layer deposited at high temperature (500–1045 °C) using MBE, MOCVD and HVPE. However, the high temperature is incompatible with the industrial back-end processes [55]. Thus, a much lower deposition temperature is preferable as it also can prevent thermal diffusion into the substrate from the Al [17]. Recently, the sputtering method is proposed for industrial applications as it offers low cost, produce no harmful waste and can easily manipulate the growth parameters [95]. DC or RF reactive sputtering has been commonly used to grow AlN films due to its simplicity and high productivity [96,97]. Furthermore, high-quality AlN film can be deposited using sputtering at low temperature (room temperature) [56]and on unheated silicon substrates[97]. Several authors also reported successful GaN growth using sputtering, without any substrate heating or post-deposition annealing temperature [98], at room temperature [99] and using substrate temperatures of 400 °C and 700 °C [100].

By using DC magnetron sputtering, some problems may occur that result in the difficulty in obtaining high crystal quality of the films, such as arcing [101], where its presence due to abnormal electrical discharge comes from the high resistance of the Al target surface during nitridation [102], as well as many impurities especially oxygen would be present in the GaN layer when using sputtered AlN as the buffer layer, resulting in leakage currents in the buffer layer, which highly deteriorate the breakdown character and prevent the use of GaN for a wide variety of electronic applications [103]. However, Ait Aissa *et al.* demonstrated that DC magnetron sputtering and HiPIMS can grow AlN film on a silicon substrate with dense and smooth properties and at low deposition temperature, and that the stoichiometry and structural quality can be easily controlled [101]. Sato *et al.* found that by using pulsed plasma as an excitation source instead of using an excimer laser for pulsed sputtering deposition, the crystalline quality of the films can be improved [56]. Hu *et al.* also demonstrated that by optimizing the technique/systems to grow the GaN-related LED layers, good crystal quality of the fabricated films can be achieved [104]. These studies indicated that by optimizing the sputtering conditions, high crystal quality of GaN can be obtained.

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