Calculation of residual thermal stress in GaN epitaxial layers grown on technologically important substrates

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A detailed investigation of residual thermal stress and misfit strain in GaN epitaxial layers grown on technologically important substrates is performed. The thermal stress is low when GaN is grown on AlN, SiC and Si, and relatively higher when Al_2O_3 substrate is used. The stress is compressive for AlN and Al_2O_3 and tensile for Si and SiC substrates. Residual thermal stress analysis was also performed for three layer heterostructures of GaN/AlN/6H-SiC and GaN/AlN/Al_2O_3. The stress remains the same when a sapphire substrate is used with or without an AlN buffer layer but reduces by an order of magnitude when a 6H-SiC substrate is used with an AlN buffer layer.

1. Introduction

Recent successes of GaN compound semiconductor light emitting diodes [1] point to the future use of these materials in displays, optical data storage, underwater communications and so on. Their direct optical band gaps span the UV-orange spectral region. They are mechanically robust, with high melting temperature and good resistance to chemical attack. These materials offer several advantages over their main competitors, such as II–VI compounds and silicon carbide, in that they offer longer device lifetimes, higher efficiencies and higher powers.

The problems which stand in the way of realization of a laser diode made of GaN are: (1) very high densities of structural imperfections, that may be described either as dislocations [2] or grain boundaries [3], (2) relative difficulty of p-doping, ascribed to acceptor passivation and (3) presence of an uncontrolled deep center, that contributes to a strong yellow luminescence band in competition with the band-edge emission [4]. All of these problems relate, to a greater or lesser extent, to the lattice mismatch and thermal expansion mismatch that exist in between the GaN epilayer and underlying substrate. The most conventional substrate used to grow GaN thin film is sapphire. Single crystals of GaN on sapphire have been grown by vapor phase epitaxy (VPE) [5–7] and metalorganic vapor phase epitaxy (MOVPE) [8–10]. 6H-SiC is another potential substrate to deposit GaN. Methods that are used to grow GaN on 6H-SiC are metallo organic chemical vapor deposition (MOCVD) [11, 12], molecular beam epitaxy (MBE) [13, 14] and hydride vapor phase epitaxy (HVPE) [15–17]. Other technologically important substrates used are AlN [18], 3C-SiC [19–21], 4H-SiC [22], and Si [23–27].

Microscopic fluctuation of the crystalline orientation and surface roughness of GaN layers, which are caused by the large lattice mismatch can be eliminated by using a buffer layer. On the other hand, the serious problems due the difference in coefficient of thermal expansions (CTEs), such as wafer bending and cracking, can be hardly avoided in a hertoepitaxial structure. Until now, there have been only a few reports about the thermal stress or strain in GaN epitaxial layers grown on sapphire substrates [28–32]. Imperfections (cracking and stress) in thick GaN layers were investigated by Itoh *et al.* [28]. They found that the cracking occurs at layer thicknesses larger than 13 μm in crystals having epitaxial layers of good quality, while it does not appear even at a thickness of 30 μm in crystals with inferior layers. Compressive stress in the epitaxial layers varied between 1.6×10^9 to 3.7×10^9 dynes/cm^2. Thermal strains and stresses were studied by Hiramatsu *et al.* [29] by varying the film thickness of GaN from 0.6 to 1200 μm. The strain in GaN had a large value at a small film thickness, decreased as the thickness increased and completely relaxed at a thickness of 100 μm due to the formation of cracks. Kecks *et al.* [30] characterized residual stresses in GaN in the temperature range of 25–600°C. Their experimental results indicated a reversible change of stress from compressive to tensile and vice versa during thermal cycling. Ager *et al.* [31] predicted that the intrinsic stress states in GaN on sapphire and 6H-SiC at the growth temperature to

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be tensile and compressive, respectively, to be in agreement with the a-plane lattice CTE mismatch. Kozawa *et al.* [32] reported biaxial compressive stress in the GaN layer due the difference in CTE between GaN and sapphire.

Also there have been few reports on the residual thermal stresses in GaN/AlN layer structures [33–35]. Magnitude and distribution of stresses generated in the lateral epitaxial overgrowth (LEO) of GaN layers grown on AlN/6H-SiC were modeled using finite element analysis by Zheleva *et al.* [33]. Their calculations showed that localized compressive stress field of 3 GPa occurred at the edges of the LEO GaN. Perry *et al.* [34] measured biaxial strains resulting from mismatches in CTEs and lattice parameters in GaN films grown on AlN buffer layers on 6H-SiC via changes in lattice parameter and found a compressive residual strain. Wang and Reeber provided a finite element modeling analysis of the residual stress distribution of multilayered GaN and AlN on 6H-SiC [35]. The effects of thickness and growth temperatures were considered in their analysis.

In this paper, we report residual thermal stresses as well as thermal strains in GaN epitaxial layers grown on different technologically important substrates. The residual stresses are calculated by using the model of Olsen and Ettenberg [36]. Until now residual thermal stresses available in the literature were calculated using an average thermal expansion coefficient between the growth and room temperatures. In this analysis, the variation of CTE with temperature is considered to obtain more accurate values. Additionally, residual stresses in three layered structure of GaN/AlN/6H-SiC and GaN/AlN/Al_2O_3 are calculated. These results provide an excellent guide for optimizing interfacial processing.

2. Analysis

Using the model of Olsen and Ettenberg [36], thermal stress in the GaN epitaxial layers grown on a substrate can be expressed as follows:

Fig. 1 shows the heterostructure multiple layer model with length L, width W, Young's modulus E_i, layer thickness t_i, moment M_i, coefficient of thermal expansion α_i, force F_i and curvature κ. Here, $i = 1$ represents the substrate and $i = 2, 3, 4 \ldots$ represents the epitaxial layers.

For one epitaxial layer, the following equations can be deduced

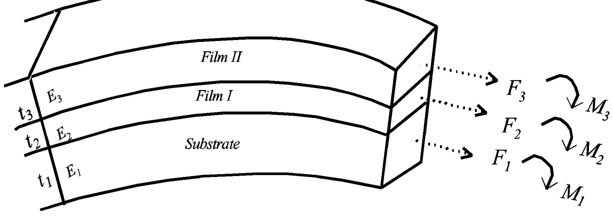

Figure 1 Sketch of 3-layer composite illustrating positive bending.

From equilibrium of forces

$$F_1 + F_2 = 0 \quad (1)$$

From equilibrium of moments

$$\frac{Wk}{12}\left(E_1 t_1^3 + E_2 t_2^3\right) + F_1 \frac{t_1}{2} + F_2\left(t_1 + \frac{t_2}{2}\right) = 0 \quad (2)$$

By solving for the strain at the interface between the film and the substrate

$$e = \frac{F_2}{E_2 t_2 L} - \frac{F_1}{E_1 t_1 L} - \frac{(t_1 + t_2)k}{2} \quad (3)$$

The strain is also taken from the difference between the CTEs of the substrate and film multiplied by the difference between growth and room temperature, ΔT.

$$e = \Delta T(\alpha_1 - \alpha_2) \quad (4)$$

Considering a small, and hence negligible, bending stress, the one-dimensional stress in the epitaxial layer is then taken as constant and given by

$$\sigma_2(1D) = \frac{F_2}{t_2 W} \quad (5)$$

where $\sigma_2(1D)$ is the one-dimensional stress.

Also, if a spherical bending is assumed with a square sample (L $\approx W$) then a two-dimensional stress can be obtained from the above one dimensional stress as follows

$$\sigma_2(2D) = \frac{\sigma_2(1D)}{(1 - \nu)^{-1}} \quad (6)$$

where $\sigma_2(2D)$ and ν are the two dimensional stress and Poisson's ratio, respectively.

For two epitaxial layers the above equations are expanded to

$$F_1 + F_2 + F_3 = 0 \quad (7)$$

$$\frac{Wk}{12}\left(E_1 t_1^3 + E_2 t_2^3 + E_3 t_3^3\right) + F_1 \frac{t_1}{2} + F_2\left(t_1 + \frac{t_2}{2}\right) + F_3\left(t_1 + t_2 - \frac{t_3}{2}\right) = 0 \quad (8)$$

$$e_1 = \frac{F_2}{E_2 t_2 L} - \frac{F_1}{E_1 t_1 L} - \frac{(t_1 + t_2)k}{2} = \Delta T(\alpha_1 - \alpha_2) \quad (9)$$

$$e_2 = \frac{F_3}{E_3 t_3 L} - \frac{F_2}{E_2 t_2 L} - \frac{(t_2 + t_3)k}{2} = \Delta T(\alpha_2 - \alpha_3) \quad (10)$$

where e_1 and e_2 are the strains between the first epilayer and substrate, and the second and first epilayers, respectively. Then $\sigma_i(2D)$, the two-dimensional stress

TABLE I Room temperature lattice mismatch of GaN with other III-N compounds and substrates

Crystal	Lattice parameter (Å⁰)	Epitaxial relationship	In-plane direction GaN∥Substrate	Lattice misfit % (room temp.)	Lattice misfit % (growth temp.)	Thermal strain % (1000–25°C)
GaN	Wurtzite $a = 3.1878$[53] $c = 5.185$ Zincblende $a = 4.511$[53]	–	–	–	–	–
AlN	Wurtzite $a = 3.1129$[53] $c = 4.9819$	(0001)/(0001)	$[10\bar{1}0]//[\bar{1}0\bar{1}0]$	$\frac{a_{GaN}-a_{AlN}}{a_{AlN}} = 2.41$	2.35	−0.06
	Zincblende $a = 4.33$[53]	$(11\bar{2}0)/(11\bar{2}0)$	[0001]//[0001]	$\frac{c_{GaN}-c_{AlN}}{c_{AlN}} = 4.08$		
α-Al_2O_3	Hexagonal $a = 4.7589$[53] $c = 12.991$	(0001)/(0001)	$[2\bar{1}\bar{1}0]//[01\bar{1}0]$	$\frac{2a_{GaN}-\sqrt{3}a_{Al_2O_3}}{\sqrt{3}a_{Al_2O_3}} = -22.65$	−22.83	−0.18
			$[01\bar{1}0]//[2\bar{1}\bar{1}0]$	$\frac{\sqrt{3}a_{GaN}-a_{Al_2O_3}}{a_{Al_2O_3}} = 16.02$		
		$(01\bar{1}3)/(01\bar{1}0)$	$[03\bar{3}\bar{2}]//[2\bar{1}\bar{1}0]$	$\frac{x a_{GaN}-c_{Al_2O_3}}{c_{Al_2O_3}} = -6.09$ where $x = \sqrt{c^2 + (2\sqrt{3}a)^2}$		
			$[2\bar{1}\bar{1}0]//[0001]$	$\frac{4a_{GaN}-c_{Al_2O_3}}{c_{Al_2O_3}} = -1.85$		
		$(0001)/(2\bar{1}\bar{1}0)$	$[01\bar{1}0]//[01\bar{1}0]$	$\frac{\sqrt{3}a_{GaN}-\sqrt{3}a_{Al_2O_3}}{\sqrt{3}a_{Al_2O_3}} = -33.01$		
			$[2\bar{1}\bar{1}0]//[0001]$	$\frac{4a_{GaN}-c_{Al_2O_3}}{c_{Al_2O_3}} = -1.85$		
		$(2\bar{1}\bar{1}0)/(01\bar{1}2)$	$[01\bar{1}0]//[2\bar{1}\bar{1}0]$	$\frac{\sqrt{3}a_{GaN}-a_{Al_2O_3}}{a_{Al_2O_3}} = 16.02$		
			$[0001]//[0\bar{1}11]$	$\frac{3c_{GaN}-x_{Al_2O_3}}{x_{Al_2O_3}} = 1.19$ where $x = \sqrt{c^2 + (2\sqrt{3}a)^2}$		
6H-SiC	Hexagonal $a = 3.0806$[56,57] $c = 15.1173$	(0001)/(0001)	$[10\bar{1}0]//[10\bar{1}0]$	$\frac{a_{GaN}-a_{6H\_SiC}}{a_{6H-Sic}} = 3.48$	3.49	0.01
3C-SiC	Zincblende $a = 4.35997$[56,57]	(001)/(001)	[010]//[010]	$\frac{a_{GaN}-a_{3C\_SiC}}{a_{3C\_SiC}} = 3.46$	3.55	0.09
4H-SiC	Hexagonal $a = 3.07997$[56,57] $c = 10.083$	(0001)/(0001)	$[11\bar{2}0//[11\bar{2}0]$	$\frac{a_{GaN}-a_{4H-SiC}}{a_{4H-SiC}} = 3.50$	3.53	0.03
Si	Zincblende $a = 5.4309$[61]	(0001)/(111)	$[11\bar{2}0]//[1\bar{1}0]$	$\frac{2a_{GaN}-\sqrt{2}a_{Si}}{\sqrt{2}a_{Si}} = -16.99$	−16.8	0.19
		(001)/(111)	$[1\bar{1}0]/[10\bar{1}]$	$\frac{a_{GaN}-a_{Si}}{a_{Si}} = -16.93$		
		(001)/(001)	[010]//[010]			

in the ith epilayer, is given as:

$$\sigma_i(2D) = \frac{\frac{F_i}{t_i W}}{(1-\nu)^{-1}} \qquad (11)$$

3. Results and discussions

Table I shows the epitaxial relationship, in-plane directions, lattice parameter mismatch between GaN and different substrates, and thermal misfit strain. Lattice parameter mismatch has been calculated using the following equation:

$$\text{Lattice Misfit \%} = \frac{d_{film} - d_{substrate}}{d_{substrate}} \times 100\,\% \qquad (12)$$

where d_{film} and $d_{substrate}$ are the interplaner spacing of the film and substrate along the in-plane direction, respectively. The misfit strain was calculated from the difference of lattice misfit at growth temperature and room temperature along the in-plane direction.

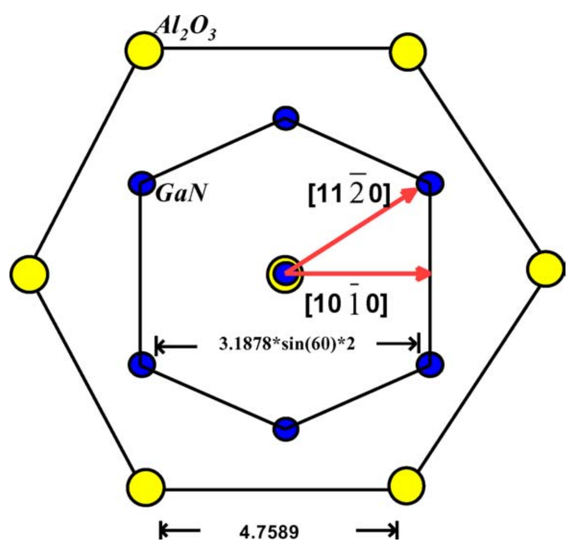

Figure 2 Orientation between GaN and Al_2O_3 in-plane directions.

TABLE II Properties of GaN and different substrates, and residual thermal stress of 1-micrometer thick epitaxial GaN film grown on different substrates

Crystal	Melting point (°C)	Elastic modulus (GPa)	CTE $\times 10^{-6}$/°C (room temp.)	Growth temp. (°C)	Residual thermal stress (Gpa)
GaN	1227	196[37]	$\alpha_a = 4.997$[38] $\alpha_c = 4.481$	–	–
AlN	2227	329.7[39]	$\alpha_a = 5.411$[40]	950,1050	−0.17
α-Al_2O_3	2030	425 [41]	$\alpha_a = 8.31$[42] $\alpha_c = 8.5$	450–1040	−0.99
6H-SiC	∼2700 sublimes	557[43]	$\alpha_a = 4.76$[44] $\alpha_c = 4.46$	950–1100	0.189
3C-SiC	1825 sublimes	444[43]	4.5[46]	–	0.14[a]
4H-SiC	2797	548[43]	4.75[46]	1000	0.183
Si	1415	112.4[47]	3.9[48]	600	0.35[a]

[a] CTE in a-direction is used (CTE of the cubic phase GaN is unavailable).

Lattice misfits between GaN and AlN are only 2.41% (corresponding thermal strain −0.06%) and 4.08% for epitaxial relationships (0001)/(0001) and (11$\bar{2}$0)/(11$\bar{2}$0), respectively. To date, to deposit GaN on sapphire four different substrate orientations have been used; these are (0001), (01$\bar{1}$0), (2$\bar{1}\bar{1}$0) and (01$\bar{1}$2). The orientation between GaN and sapphire in-plane directions is shown in Fig. 2. The largest lattice mismatch between (0001) GaN and (2$\bar{1}\bar{1}$0) sapphire is −33.0% along the [01$\bar{1}$0]//[01$\bar{1}$0] in plane direction and smallest mismatch between (2$\bar{1}\bar{1}$0) GaN and (01$\bar{1}$2) sapphire (r-plane) is 1.19% along the [0001]// [0$\bar{1}$11] in-plane direction. From the view point of lattice mismatch sapphire r-plane is predicted to be the most suitable for GaN growth. A thermal strain of −0.18% exists in GaN (0001) when grown on sapphire (0001).

The next common substrate used to grow GaN is 6H-SiC. The lattice parameter misfits between GaN and 6H-, 3C- and 4H-SiC are very close to each other (i.e., 3.48, 3.46 and 3.50%, respectively) and the corresponding thermal strains are 0.01, 0.09 and 0.03, respectively. Cubic GaN films have been epitaxially grown onto (001) and (111) Si by electron cyclotron resonance microwave-plasma-assisted molecular-beam epitaxy with a lattice misfit of −16.93% and misfit strain of 0.19%.

Due to the difference in stress between the thin film and substrate, the composite of film and substrate bends. It is important to estimate the radius of curvature because the degree of curvature is directly proportional to the difference in strain between the film and substrate. In the calculation of curvature and stress instead of an

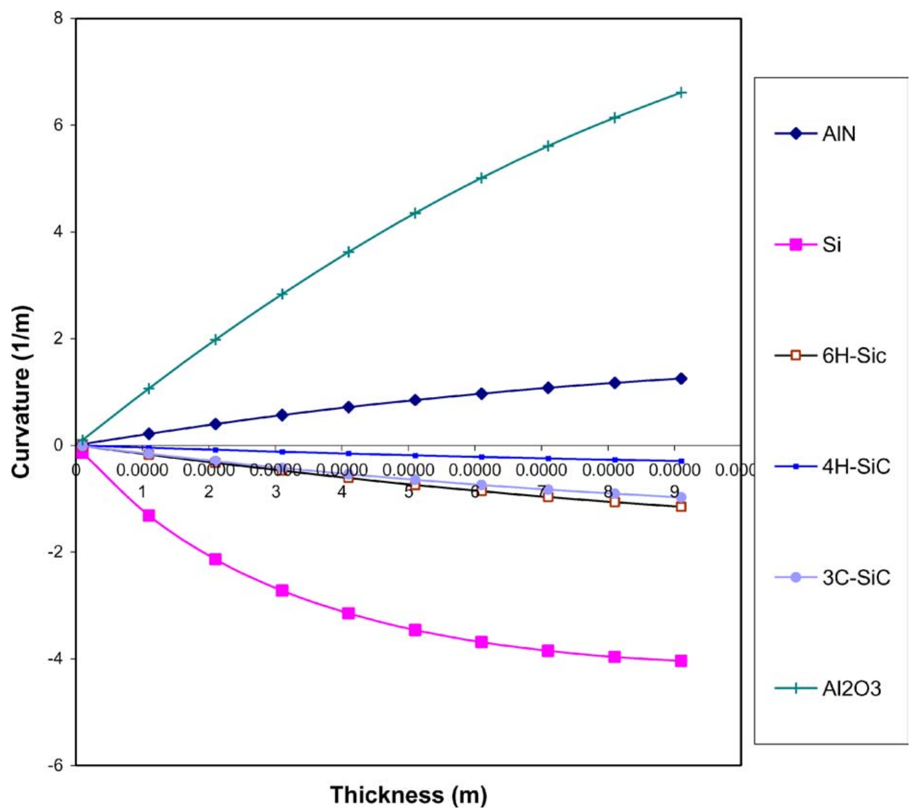

Figure 3 Comparison of curvature in epitaxial GaN layers on technologically important substrates.

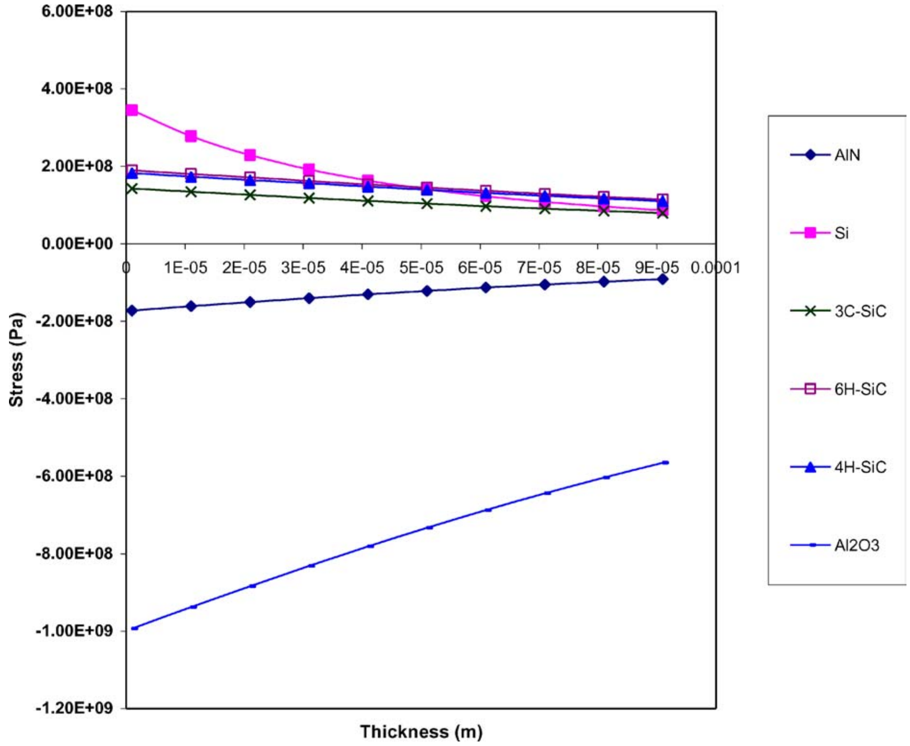

Figure 4 Comparison of residual thermal stresses in epitaxial GaN layer on technologically important substrates.

average CTE over the entire range of temperature, the variation CTE with temperature was considered. The final curvature and stress is then the accumulation of values at different intervals from growth temperature to room temperature. Table II shows the properties and thermal stress of 1-micrometer thick epitaxial GaN film grown on different potential substrates. Figs 3 and 4 show curvature and residual thermal stresses in epitaxial GaN layers, respectively. The thermal stress is low when GaN is grown on AlN, SiC and Si while in case of Al_2O_3, it is much higher. Also the thermal stress is tensile in GaN when grown on Si and SiC while it is compressive when AlN and Al_2O_3 substrates are used.

Figs 5 and 6 show variation of residual thermal stress in GaN versus thickness of GaN with an AlN buffer layer of thickness of 0.1 micrometer for three layer heterostructures GaN/AlN/6H-SiC and GaN/AlN/Al_2O_3, respectively. There is very little change in thermal stress when GaN is grown on Al_2O_3 with or without a buffer layer of AlN. On the other hand the stress decreases by an order of magnitude by using a buffer layer of AlN while growing GaN on 6H-SiC.

4. Conclusions

GaN is a wide band gap (=3.39 eV) semiconductor and a promising candidate for the manufacture of light emitting diodes (LED's) and laser diodes in the short wavelengths from blue to ultraviolet. Single crystals of GaN are grown on different substrates using different

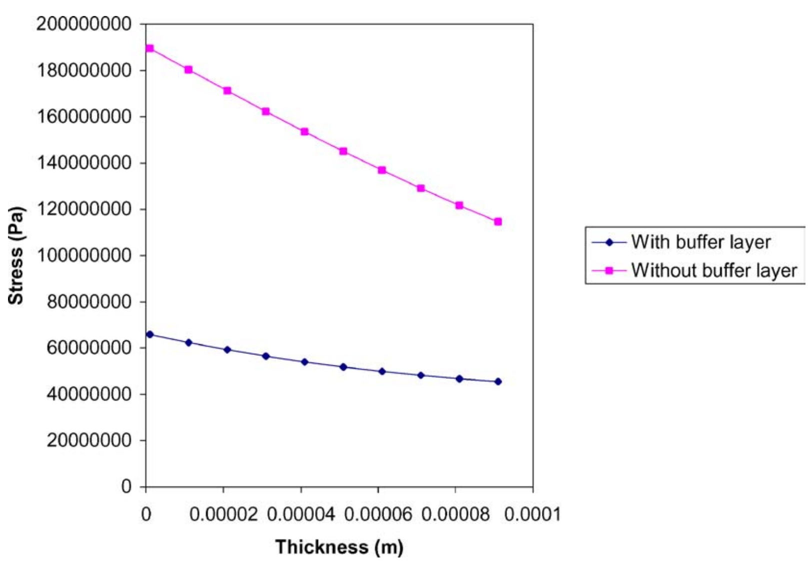

Figure 5 Residual thermal stress in GaN for multilayer GaN/AlN/6H-SiC.

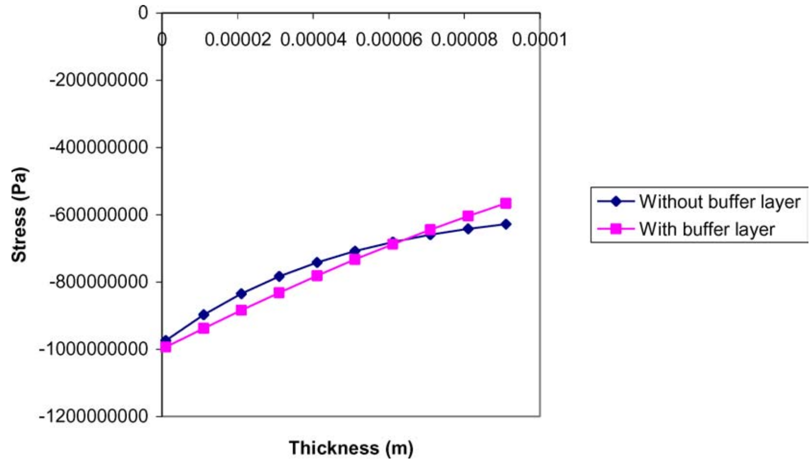

Figure 6 Residual thermal stress in GaN for multilayer GaN/AlN/Al_2O_3.

epitaxial growth techniques. To utilize this semiconductor efficiently, crystals should be grown free of residual stresses. As a result, it is imperative to perform a true characterization of the residual thermal stresses in these crystals. In this paper, curvature, residual thermal stress and misfit strain in GaN grown on different technologically important substrates are reported. GaN grown on AlN, SiC and Si has residual thermal stress lower by a factor compared to that in GaN grown on Al_2O_3. Also the thermal stress is tensile in nature when Si and SiC substrates are used while it is compressive for AlN and Al_2O_3.

Using a buffer layer of AlN reduces the thermal stress by an order when GaN is grown on 6H-SiC while it stays the same when a sapphire substrate is used.

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