Critical thickness for GaN thin film on AlN substrate

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***Abstract*—The critical thickness is the smallest thickness of a uniformly strained thin layer on a substrate for which it becomes energetically possible for a misfit dislocation to form spontaneously at the interface between the layer and the substrate. The critical thickness can be calculated by different criterions. The most used criterions assume that both the thin layer and the substrate isotropic with the same elastic properties. Recently a new criterion was developed to describe the formation of a misfit dislocation buried at a distance h below the free surface, assuming different elastic constants of the thin film and the substrate with hexagonal symmetry. We compared the results of this criterion for an $Al_{1-x}Ga_xN$ film on an AlN substrate with the result obtained by the isotropic criterions. After this, we calculated the critical thickness for the $Al_{1-x}Ga_xN$ on an AlN substrate for different temperatures.**

I. Introduction

III-nitrides are considered promising semiconductors for power applications. In particular, the performance of the devices based on III-nitride heterostructures, like InGaN/GaN and AlGaN/AlN, are influenced by the dislocation development during their epitaxial deposition. Dislocations are problematic as they can increase leakage currents in LEDs [1], decrease the lifetime of laser diodes [2], lower the electron mobilities in high electron mobility transistors [3], cause current collapse in GaN-based field-effect transistors used in RF applications [4], and produce overall degradation of the performance of high-power GaN-based devices [5].

One issue for the reliability of the III-nitride based device is the condition for the creation of misfit dislocations (MDs) during heteroepitaxy, when a thin epilayer is grown on a substrate with significantly different lattice parameters. This process has been observed experimentally in details. Below a certain epilayer thickness, called the *critical thickness* (CT), h_c, the epilayer is grown pseudomorphically on a substrate, which means that the epilayer is grown with the same lattice parameter of the substrate causing the straining of the epilayer and an increase of its internal energy. When the critical thickness h_c is reached, the relaxation of the misfit strain occurs via plastic flow. The most common mechanism of the plastic relaxation is the creation of the MDs along the interface between the film and the substrate.

Several models have been proposed to calculate the CT in the last decades. Matthews and Blakeslee [6] proposed a model based on force equilibrium for the bending of pre-existing threading dislocations. People and Bean [7] as well as Freund and Suresh [8] derived models for infinite MDs balancing the overall energy of straight, infinite MDs for a cubic symmetry. Jain *et al.* [9] developed a model assuming that both the film and the substrate are isotropic and considering the impact of the film thickness. Willis *et al.* [10] developed a general treatment for a dislocation buried at a distance h below the free surface of the thin layer and assuming different elastic constants of the thin layer and the substrate. Holec [11] corrected the previous treatment for the hexagonal symmetry of the material. This model can predict the CT of wurtzite structures, like $Al_{1-x}Ga_xN$/AlN heterostructure.

In this paper, we calculate the CTs for the $Al_{1-x}Ga_xN$/AlN heterostructure according to the criterions developed by Matthews and Blakeslee [6] and Jain *et al.* [9] for isotropic film and substrate with the same elastic properties. Then, we compared these results with the CT calculated according to the model developed by the Willis *et al.* [10] for film and substrate with different elastic properties adapted by Holec [11] for the hexagonal symmetry (WH model). We found that the CT calculated considering the hexagonal symmetry and different elastic properties of the film and the substrate results in lowering the CT by 30%-40% when compared with the values obtained for the isotropic film and substrate with the same elastic properties. This difference cannot be neglected for the prediction of the dislocation onset in ultra thin films, with thicknesses less than 20nm. After that, we evaluate the CT of the $Al_{1-x}Ga_xN$/AlN heterostructure according to the model presented in Willis *et al.* [10] which was later adapted by Holec [11] for different temperatures, from 300K to 1000K. We found that the temperature difference between 1000K and 300K decreases the CT by 10%-20% for $0.2 < x < 0.5$, while it can be neglected for $x < 0.2$ and $x > 0.5$, where x is the gallium content in the $Al_{1-x}Ga_xN$ alloy.

A. Critical thickness model

Here, the formulation of the WH model [10] [11] is described. It compares the energy E_d per unit length of a dislocation added to the system with the work W per unit length of dislocation done by the misfit stress field. This misfit stress field is created by the difference between the lattice parameters of the film and the substrate. As a consequence, the CT criterion is given by

$$W = -E_\mathrm{d}. \tag{1}$$

1) Work done by the misfit strain: the work W done by the misfit strain created by the lattice mismatch between the thin layer and the substrate can be calculated according to [8]

$$W = \int_h^0 \sum_{i,j} b_i \sigma_{ij}^m n_j \, dl, \tag{2}$$

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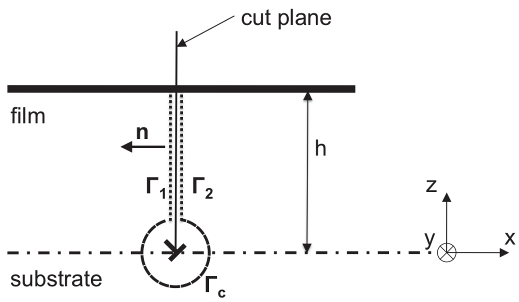

Fig. 1. To form the dislocation, the material is cut along the dotted line composed by the $\Gamma = \Gamma_1 + \Gamma_2$ and the core surface Γ_c. h is the film thickness.

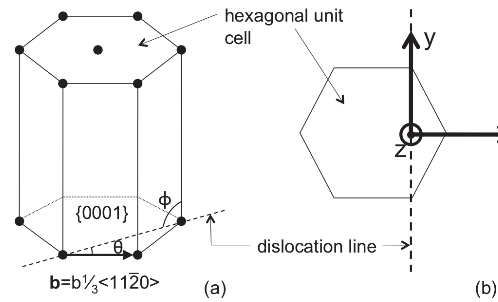

Fig. 2. (a) Dislocation geometry along the basal plane. (b) The considered slip system $\frac{1}{3}\left<11\bar{2}0\right>\{0001\}$ in the $\mathrm{Al}_{1-x}\mathrm{Ga}_x\mathrm{N}$ structure.

where l is the coordinate along the slip plane perpendicular to the dislocation line (the dislocation is brought from the surface to its actual position on the interface). According to the geometry shown in Fig. 1, the top surface $z = h$ of the specimen is traction free and therefore $\sigma^m_{xz}(x,h) = \sigma^m_{zz}(x,h) = \sigma^m_{yz}(x,h) = 0$. The only non-zero components of the uniform misfit stress are $\sigma^m_{xx} = \sigma^m_{yy} = \sigma_m$ within the thin layer. All directions within the xy-plane (the hexagonal plane) are equivalent and thus the mismatch strain components are $\varepsilon^m_{xx} = \varepsilon^m_{yy} = \varepsilon_m$, where ε_m is the misfit strain. ε_m is defined considering that the thin film's in-plane lattice constant a adjusts to the rigid substrate's lattice constant a^*. This is expressed as

$$\varepsilon_m = \frac{a^* - a}{a}. \tag{3}$$

Hooke's law gives

$$0 = \sigma^m_{zz} = c_{13}\epsilon^m_{xx} + c_{13}\epsilon^m_{yy} + c_{33}\epsilon^m_{zz} \tag{4}$$

which results in

$$\varepsilon^m_{zz} = -2\frac{c_{13}}{c_{33}}\varepsilon_m. \tag{5}$$

Then, using Hooke's law, the mismatch stress is

$$\sigma_m = \sigma^m_{xx} = \sigma^m_{yy} = \frac{(c_{13}+c_{12})c_{33} - 2c^2_{13}}{c_{33}}\varepsilon_m. \tag{6}$$

We define the Burgers vector $\mathbf{b}$, the tensor of the misfit stress $\boldsymbol{\sigma}_m$, and the outer normal to the cut surface Γ denoted by $\mathbf{n}$ as:

$$\mathbf{b}=\begin{pmatrix}-b\sin\theta\sin\phi\\ b\cos\theta\\ b\sin\theta\cos\phi\end{pmatrix}, \boldsymbol{\sigma}_\mathrm{m}=\begin{pmatrix}\sigma_m & 0 & 0\\ 0 & \sigma_m & 0\\ 0 & 0 & 0\end{pmatrix}, \mathbf{n}=\begin{pmatrix}-\cos\phi\\ 0\\ \sin\phi\end{pmatrix}, \tag{7}$$

where θ is the angle between the dislocation line and the Burgers vector, and ϕ the angle between the slip plane and the normal to the film-substrate interface (see Fig. (2)). Then equation (2) yields

$$W = b\sigma_m h \sin\theta \sin\phi. \tag{8}$$

2) Dislocation energy: according to [8], the misfit dislocation may be produced by making a cut along the intended slip plane, displacing one cut surface relative to the other by the Burgers vector $\mathbf{b}$, and welding the material back together. The misfit dislocation is buried along the interface between the film, whose thickness is h, and the substrate at a distance h below the film surface. The work done by placing this dislocation is the energy of the misfit dislocation. The difference between the displacements created by the cut equals the length of the Burgers vector $\mathbf{b}$. The dislocation energy can be expressed as

$$E_\mathrm{d} = \frac{1}{2}\int_\Gamma \sum_{ij} b_i\sigma^d_{ij}n_j\, d\Gamma + \frac{1}{2}\int_{\Gamma_\mathrm{c}} \sum_{ij} b_i\sigma^d_{ij}n_j\, d\Gamma. \tag{9}$$

where n_j denotes the outer normal to the cut surface S (Fig. 1). The second integral of (9) is neglected because the thickness of the dislocation core is small compared to the film thickness h. The dislocation line lies in the hexagonal plane along the y-axis (Fig. 2). The considered dislocation is straight and extends to infinity. This assumption simplifies the problem to a plane strain problem where no variable depends on the y-coordinate ($\delta/\delta y \equiv 0$). The substrate fills the half space $z < 0$ and the thin film spreads in the region $0 < z < h$. It is assumed that the film and the substrate have wurtzite structures with hexagonal symmetry but different elastic properties. Across the interface at $z = 0$ between the film and the substrate the displacements and the tractions must be continous:

$$\begin{aligned} u_x(x,0) + b_x = u^*_x(x,0), \quad & \sigma_{x,z}(x,0) = \sigma^*_{x,z}(x,0), \\ u_z(x,0) + b_z = u^*_z(x,0), \quad & \sigma_{z,z}(x,0) = \sigma^*_{z,z}(x,0), \\ u_y(x,0) + b_y = u^*_y(x,0), \quad & \sigma_{y,z}(x,0) = \sigma^*_{y,z}(x,0). \end{aligned} \tag{10}$$

All quantities in the substrate have a $*$ superscript to distinguish them from quantities related to the thin film. The substrate is not influenced by the thin layer so all stress components must vanish for $z \to -\infty$. Hooke's law for an hexagonal structure tensor is

$$\begin{pmatrix}\varepsilon_{xx}\\ \varepsilon_{yy}\\ \varepsilon_{zz}\\ 2\varepsilon_{yz}\\ 2\varepsilon_{xz}\\ 2\varepsilon_{xy}\end{pmatrix} = \begin{pmatrix} s_{11} & s_{12} & s_{13} & 0 & 0 & 0\\ s_{21} & s_{22} & s_{23} & 0 & 0 & 0\\ s_{31} & s_{32} & s_{33} & 0 & 0 & 0\\ 0 & 0 & 0 & s_{44} & 0 & 0\\ 0 & 0 & 0 & 0 & s_{55} & 0\\ 0 & 0 & 0 & 0 & 0 & s_{66}\end{pmatrix} \begin{pmatrix}\sigma_{xx}\\ \sigma_{yy}\\ \sigma_{zz}\\ \sigma_{yz}\\ \sigma_{xz}\\ \sigma_{xy}\end{pmatrix}, \tag{11}$$

where $s_{11} = s_{11}$, $s_{12} = s_{21}$, $s_{13} = s_{23} = s_{31} = s_{32}$, and $s_{44} = s_{55}$ due to the hexagonal symmetry. The equation (11)

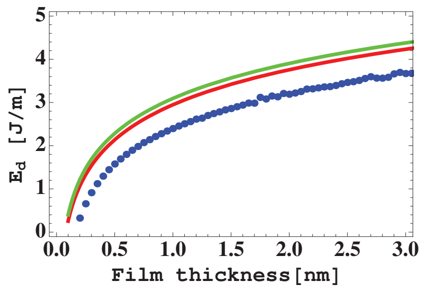

Fig. 3. Dislocation energy E_d as a function of the film thickness. The red curve corresponds to CT criterion [6], the green curve corresponds to CT criterion [9], the blue dotted curve corresponds to the CT criterion [10] [11].

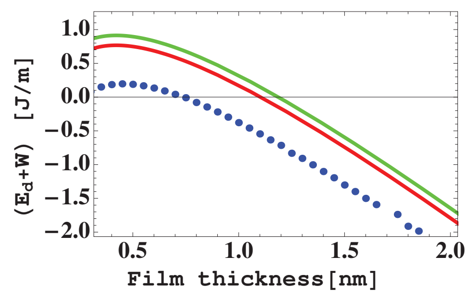

Fig. 4. Total energy $E_d + W$ as a function of the film thickness. The red curve corresponds to CT criterion [6], the green curve corresponds to CT criterion [9], the dotted curve corresponds to the CT criterion [10] [11].

can be solved with the boundary conditions (10) to derive all the stress and displacement components necessary for the calculation of $E_\mathbf{d}$.

B. Critical thickness for GaN film on AlN

The calculation of the dislocation energy E_d was performed numerically assuming that the most favourable slip system in GaN is $\frac{1}{3}\langle 11\bar{2}0\rangle\{0001\}$. The result is shown in Fig. 3 as a function of the film thickness. It is compared with the solution for the dislocation energy calculated according to the Matthews and Blakeslee criterion [6] as well as the Jain *et al.* criterion [9]. Fig. 4 shows the total energy calculated solving (1) with the parameter values in Table I and II and the dislocation energy calculated with (8), (9), and (10). The result is compared again with the models for the isotropic case. The intersections between the curves and the x-axes in Fig. 5 are the different CTs. The calculated results for the critical thickness are 1.1 nm according to the Matthews and Blakeslee's model [6], 1.2 nm according to isotropic Jain *et al.* model [9], and 0.7 nm according to the WH model [10] [11]. It is concluded that the hexagonal symmetry of the III-nitride crystal structure, the difference between the elastic properties between the substrate and the film, and the impact of the thin film thickness on the misfit dislocation creation lowers the CT by 30%-40% when compared with the isotropic values in GaN/AlN.

C. Critical thickness calculation for $Al_{1-x}Ga_xN$ film on AlN substrate for different temperatures

The same procedure described in section A was applied to calculate the CTs of the $Al_{1-x}Ga_xN$ film on AlN substrate with $0.1 < x < 1$, where x is the gallium content in the $Al_{1-x}Ga_xN$ alloy. The calculation was performed numerically assuming that the most favorable slip system in $Al_{1-x}Ga_xN$ for the MDs is again$\frac{1}{3}\langle 11\bar{2}0\rangle\{0001\}$. The characteristics of the slip system are in general a function of the gallium content x but this fact is neglected because the variation of the angle ϕ with x is very small. The lattice parameters of the $Al_{1-x}Ga_xN$ alloy for each fraction x were calculated by Vegard's law, which is an approximate rule that states that a linear relationship exists between the crystal lattice parameter of an alloy and the concentrations of the constituent elements. As such the lattice parameter of the $Al_{1-x}Ga_xN$ alloy is calculated by:

$$a_{\mathrm{Al}_{1-x}\mathrm{Ga}_x\mathrm{N}} = a_{\mathrm{AlN}}(1-x) + a_{\mathrm{GaN}}x. \tag{12}$$

We assumed that Vegard's law is also valid for the calculation of the elastic constants for the $Al_{1-x}Ga_xN$ alloy:

$$c_{ij,\mathrm{Al}_{1-x}\mathrm{Ga}_x\mathrm{N}} = c_{ij,\mathrm{AlN}}(1-x) + c_{ij,\mathrm{GaN}}x. \tag{13}$$

The results for the composition $0.2 < x < 1$ are plotted in Fig. 5. They show again that the hexagonal symmetry of the III-nitride crystal structure, the difference between the elastic properties between the substrate and the film and the impact of the thin film thickness on the misfit dislocation creation lowers the CT by 15% for $x = 0.2$ and 40% for $x = 1$ when compared to the isotropic values. In order to investigate the impact of the temperature on the CT, it is necessary to calculate the elastic constants and the lattice parameters as functions of temperature. Therefore, the mismatch strain ε_m is also a function of the temperature. As a consequence, the CT depends on the temperature too. The temperature dependence of GaN and AlN thermal expansion coefficients are determined experimentally [12]:

$$\alpha_l(T) = \sum_{\mathrm{h}=0}^{3} l_h \left(\frac{T}{1\mathrm{K}}\right)^h, \tag{14}$$

where l is the lattice parameter a and c, l_h are the coefficients in Table III, and T is the absolute temperature. The temperature dependence of the lattice constants is then given by theformula:

$$l = l_{300\mathrm{K}} \left(1 + \int_{300\mathrm{K}}^{T} \alpha_l(T)\right) dT, \tag{15}$$

where $l_{300\mathrm{K}}$ are the lattice parameters at 300K (see Table II). The functions of the temperature dependence of the elastic constants c_{ij} have been extrapolated using the experimental data of [13] within the range of temperatures of the data (200K-800K for GaN and 300K-1200K for AlN). For GaN and AlN they are obtained in GPa:

$$c_{ij}(T) = \sum_{h=0}^{3} v_h \left(\frac{T}{1\mathrm{K}}\right)^h, \tag{16}$$

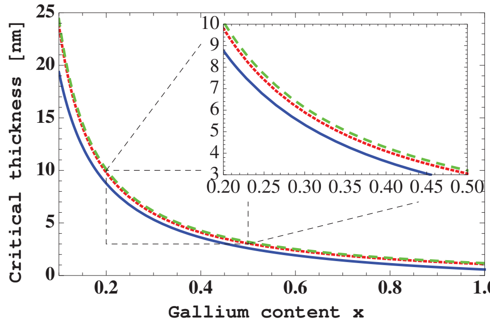

Fig. 5. The critical thickness (CT) as function of the Ga content x in the $Al_{1-x}Ga_xN$ alloy. The dotted red curve corresponds to CT criterion [6], the dashed green curve corresponds to CT criterion [9], and the solid blue curve corresponds to the CT criterion [10] [11].

where v_h are the coefficients in Table IV, and T is the absolute temperature. The results for the composition $0.2 < x < 0.5$ are plotted in Fig. 6. They show that the temperature has an impact on the CT because the critical thickness decreases by 10%-20% in this composition range.

II. Conclusion

We have shown that the differences between the CT result of an $Al_{1-x}Ga_xN$ film on AlN substrate with $0.1 < x < 1$ obtained with the procedure described here according to the WH [10] [11] and the results obtained with the isotropic CT criterions [6], [9] are about 15%-40%. The WH model [10] [11] is the most restrictive criterion to predict the dislocation onset in III-nitrides so no MDs can develop below this value. Therefore the crystal anisotropy cannot be neglected in ultra thin films with thicknesses less than 20nm in order to predict exactly the film thickness range for which the structure is MDs free. In III-nitride heterostructures, the overall degradation of the performance of electronic devices caused by dislocations occurs at thicknesses lower than those predicted by [6] [9]. It is also shown that the temperature has an impact on the CT calculation that can be neglected during the epitaxial deposition, whose standard temperature is often in the range between 800K and 1000K.

TABLE I
The film and substrate parameters (in GPa) at 300K are taken from [13].

c_{11}^{GaN}	c_{12}^{GaN}	c_{13}^{GaN}	c_{33}^{GaN}	c_{44}^{GaN}	c_{66}^{GaN}
390	145	106	398	105	$(c_{11}-c_{12})/2$
c_{11}^{AlN}	c_{12}^{AlN}	c_{13}^{AlN}	c_{33}^{AlN}	c_{44}^{AlN}	c_{66}^{AlN}
411.7	148.9	99.4	385.6	124.1	$(c_{11}^*-c_{12}^*)/2$

TABLE II
The parameters used are taken from literature.

a_{GaN}	c_{GaN}	a_{AlN}	c_{AlN}	b	θ	ϕ
3.22 Å	5.2 Å	3.11 Å	4.98 Å	3.22 Å	$\pi/6$	$\pi/2$
[14]	[14]	[14]	[14]	[11]	[11]	[11]

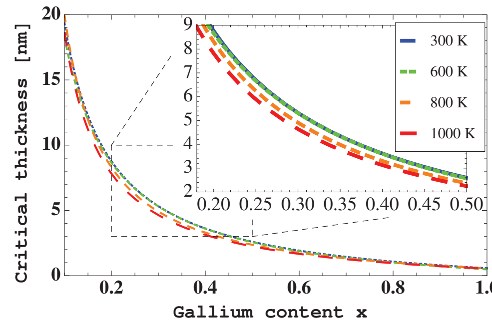

Fig. 6. The critical thickness (CT) as function of the Ga content x in the $Al_{1-x}Ga_xN$ alloy for different temperatures, from 300K to 1000K.

TABLE III
The film and substrate parameters (in K^{-1}) used are taken from [12].

a_0^{GaN}	a_1^{GaN}	a_2^{GaN}	a_3^{GaN}
3.2×10^{-6}	1.2×10^{-9}	10^{-12}	0
c_0^{GaN}	c_1^{GaN}	c_2^{GaN}	c_3^{GaN}
0.9×10^{-6}	9×10^{-9}	-3×10^{-12}	0
a_0^{AlN}	a_1^{AlN}	a_2^{AlN}	a_3^{AlN}
-8.7×10^{-8}	1.9×10^{-10}	3.4×10^{-13}	-7.9×10^{-17}
c_0^{AlN}	c_1^{AlN}	c_2^{AlN}	c_3^{AlN}
-7×10^{-8}	1.6×10^{-10}	2.7×10^{-13}	-5.8×10^{-17}

TABLE IV
The film and substrate parameters used are taken from [13].

	$v_0[K^{-1}]$	$v_1[K^{-1}]$	$v_2[K^{-1}]$	$v_3[K^{-1}]$
c_{11}^{GaN}	376.4	9×10^{-4}	-3×10^{-5}	10^{-8}
c_{12}^{GaN}	142.4	2×10^{-4}	-10^{-5}	6×10^{-9}
c_{13}^{GaN}	99.1	4×10^{-4}	-10^{-5}	7×10^{-9}
c_{33}^{GaN}	387.1	7×10^{-4}	-4×10^{-5}	2×10^{-8}
c_{44}^{GaN}	98.5	4×10^{-4}	4×10^{-6}	2×10^{-9}
c_{11}^{AlN}	411.7	7×10^{-4}	-2×10^{-5}	5×10^{-9}
c_{12}^{AlN}	148.9	10^{-4}	-6×10^{-6}	2×10^{-9}
c_{13}^{AlN}	99.4	7×10^{-4}	-5×10^{-6}	2×10^{-9}
c_{33}^{AlN}	385.6	3×10^{-5}	-2×10^{-5}	6×10^{-9}
c_{44}^{AlN}	124.1	2×10^{-4}	-2×10^{-6}	9×10^{-10}

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