Plastic Relaxation In Single $In_xGa_{1-x}N/GaN$ Epilayers Grown On Sapphire

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***Abstract* — Plastic relaxation was observed in $In_xGa_{1-x}N/GaN$ epilayers grown on *c*-plane sapphire substrates. The relaxation obeys the universal hyperbolic relation between the strain and the reciprocal of the layer thickness. Plastic relaxation in this material system reveals that there is no discontinuous relaxation at critical thickness and once a layer starts to relieve, it follows the same strain-thickness dependence, unconstrained by the original misfit until the material system work hardens. From *x*-ray diffraction calibration, the in-plane and normal relaxation constants K_{P0} and K_{N0} for the $In_xGa_{1-x}N/GaN$ grown on sapphire were found to be −0.98 ± 0.03 and +0.51 ± 0.03 nm, respectively.**

I. Introduction

The strain relaxation mechanism in III-Nitrides that relieves the accumulated strain generally influences the growth and quality of the mismatched heteroepitaxial layers. At small epilayer thickness, elastic relaxation usually occurs through the formation of sinusoidal undulations, islands or pits [1]. Elastic relaxation in InGaN is a consequence of threading dislocation glide geometry and high Peierls forces [2], in which pinholes participate in the relaxation process [3,4]. Above the critical thickness, plastic relaxation is initiated by the formation of misfit dislocations. The issue of plastic relaxation has not been extensively investigated for nitride semiconductors due to the lack of understanding in the mechanisms involved. As such, the present work will concentrate on the study of plastic relaxation in $In_xGa_{1-x}N/GaN$ epilayers grown on *c*-plane sapphire substrates by metalorganic chemical vapor deposition (MOCVD). The strain relaxation, determined using *x*-ray diffraction, is found to be accurately hyperbolic and proportional to the reciprocal of the layer thickness, and thus in agreement with the force-balance model proposed by Matthews and Blakeslee [5]. Such relaxation process in InGaN/GaN also obeys the geometrical model of the critical thickness proposed by Dunstan *et al*. [6]. Following these theoretical models and taking into account the experimentally observed values, the in plane and normal strain relaxation constants for $In_xGa_{1-x}N/GaN$ epilayers grown on sapphire substrates were estimated.

In literature, various theoretical and experimental models that investigated the critical thickness of strained epilayers have been reported. The discrepancies between the models and their empirical fits are due to two important considerations. First, the growth kinetic (i.e. Peierls) barriers that limit the extent of strain relaxation in metastable structures for a given temperature and time affects the exact point at which plastic deformation begins. Exceeding the equilibrium critical thickness is only a necessary but not sufficient condition to determine any observable strain relaxation. Therefore, the data obtained from metastable structures may not match with the equilibrium theory. Second, the point at which strain relaxation is initiated may not be accurately determined due to experimental sensitivity. Analytical techniques frequently used to study dislocation-induced strain relief (e.g. x-ray diffraction, Raman spectroscopy, photoluminescence spectroscopy and reflection high-energy electron diffraction) are unable to detect dislocations with a density less than 10^4 cm^{-2}. Consequently, the apparent critical thickness measured is limited by the resolution and sensitivity of the analytical techniques.

Majority of the work in semiconductors that report strain relaxation mechanisms, follow the earliest theoretical models, which takes into account of the force and energy balance treatment. The force-balance model, first introduced by Matthews and Blakeslee [5], considered a mechanism for the generation of misfit dislocations and assumed that a sufficient density of pre-existing sources such as the substrate threading dislocations, are available. In this manner, the equilibrium critical thickness can be related to the first misfit dislocation that is introduced at the interface between the epilayer and the substrate. People and Bean [7] on the other hand described the critical thickness in GeSi/Si using an energy balance model that considered spontaneous homogeneous interfacial nucleation of misfit dislocations in the absence of pre-existing dislocations. This model defines an areal energy density for an isolated dislocation, which is physically invalid. Nevertheless, this model has been extensively used in the past for modeling the experimental critical thickness that apparently exceeds equilibrium predictions.

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Mathematically, the force-balance model treats the critical thickness as the layer thickness at which the force on a dislocation line due to elastic strain exactly balances the internal tension of the dislocation line [5]. For a layer of thickness *L*, the strain force on the dislocations is given by

$$F_S = 2G\left(\frac{1+v}{1-v}\right)bL\,|\varepsilon\,|\cos\gamma \qquad (1)$$

while the internal tensile force in the dislocation line can be written as

$$F_l = \frac{Gb^2}{4\pi}\left(\frac{1-v\cos^2\beta}{1-v}\right)\left(\ln\frac{L}{b}+1\right) \qquad (2)$$

where G is the shear modulus, v is the Poisson ratio, b is the magnitude of Burgers vector, β is the angle between the Burgers vector and dislocation line, and γ is the angle between the glide plane and the interface. The critical thickness, $L = h_c$ is obtained by using $F_s = 2F_l$. The factor 2 arises because two threading dislocations per layer are required, one for the upper interface and another for the lower interface. Using Eqs (1) and (2), the equation that must be solved to obtain the critical thickness is given by

$$h_c = \frac{b(1-v\cos^2\beta\,)}{4\pi\,|\varepsilon\,|(1+v)\cos\gamma}\left(\ln\frac{h_c}{b}+1\right) \qquad (3)$$

This expression shows that h_c is inversely proportional to the strain ε if the smaller magnitude logarithmic term is treated as a constant with the others. This leads to the empirical critical thickness model proposed by Dunstan *et. al* [6] that postulates that h_c is given by

$$h_c = \frac{k_1 b}{\varepsilon} \qquad (4)$$

where ε is the misfit strain in a pseudomorphic layer and b is the Burger's vector of the misfit dislocation. The model treats a partially relaxed layer the same manner as a pseudomorphic unrelaxed layer, and deduces that a certain amount of strain will be relaxed as the layer thickness passes through the critical thickness. This model also shows that the relation between the strain ε and the thickness h above the critical thickness will show reverse relation and the strain ε during plastic relaxation is given by

$$\varepsilon(h) = \frac{k_2 b}{h} \qquad (5)$$

The constants of proportionality k_1 and k_2 are both expected to be in the order of unity. When k_1 and k_2 have the same value, there will be no discontinuous relaxation at critical thickness where the discontinuous relaxation will correspond to a reduced value of k_2. Once a layer begins to relax, it follows the same strain-thickness relation independent of its original misfit caused by heteroepitaxy.

II. EXPERIMENT

In order to probe plastic relaxation in the $In_xGa_{1-x}N$ /GaN system, a set of single layer $In_{0.10}Ga_{0.90}N$ samples of five different thicknesses was grown by metalorganic chemical vapor deposition (MOCVD) on *c*-plane sapphire substrates with a 25 nm thick low temperature GaN nucleation layer. During the growth, the reactor pressure was kept at 200 Torr. A high temperature 1000 ^{0}C GaN layer of thickness 1 μm was grown on top of the nucleation layer before single $In_{0.10}Ga_{0.90}N$ layers were grown at 750 ^{0}C. X-ray diffraction (XRD) measurements were performed using a Philips X'Pert MRD system equipped with a PW3050/20 goniometer and $Cu_{K\alpha 1}$ radiation source. In order to estimate the lattice constants and strain in all the samples, 2θ-ω scan and *x*-ray rocking curves were obtained for both the (00.2) and (10.2) reflections.

III. RESULTS

The (00.2) reflection XRD spectra recorded from the $In_{0.10}Ga_{0.90}N$ samples are shown in Figures 1. Figure 2 shows the $In_{0.10}Ga_{0.90}N$ (00.2) and (10.2) reflections 2θ peaks for the samples, fitted with Lorentzian functions.

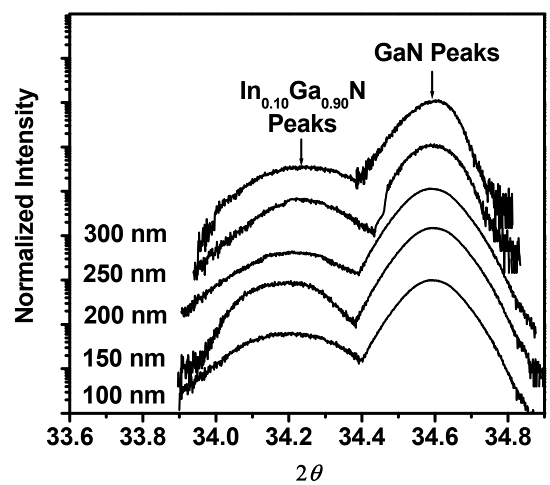
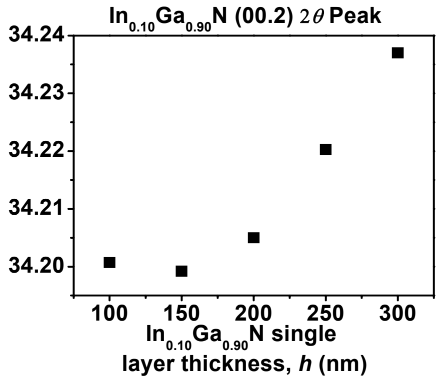
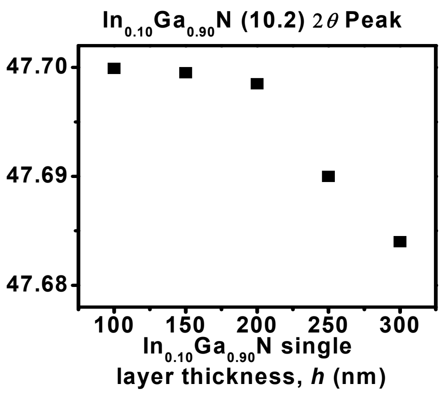


Fig. 1. (00.2) 2θ-ω XRD rocking curves of the $In_{0.10}Ga_{0.90}N$/GaN samples for the thickness indicated.



(a)



(b)

Fig. 2. The $In_{0.10}Ga_{0.90}N$ (00.2) and (10.2) reflections 2θ peaks for the samples, where the spectra were fitted with Lorentzian functions.

To estimate the normal plane lattice constants, c(GaN) and $c(In_xGa_{1-x}N)$, i.e. equivalent interplanar spacing, we use Bragg's law

$$d_{hkl} = \frac{l\lambda}{2\sin\theta} \tag{6}$$

where d_{hkl} is the interplanar spacing for any allowed symmetric (00.l) reflection, λ is the wavelength of the radiation and θ is the relevant Bragg angle estimated from the peak of the XRD distribution. Similarly, a scan through the asymmetric (10.l) reflection provides the values of the in plane lattice constants, a(GaN) and $a(In_xGa_{1-x}N)$ using the expression,

$$\frac{l^2}{d_{hkl}{}^2} = \frac{4}{3}\frac{h^2+k^2+hk}{a^2} + \frac{l^2}{c^2} \tag{7}$$

In the case of biaxially strained wurtzite structures such as InGaN/GaN, distortion of the hexagonal unit cell occurs. In order to separate the influence of strain and composition, the parameters of the wurtzite lattice, $c(In_xGa_{1-x}N)$ and $a(In_xGa_{1-x}N)$, should be measured and compared to their respective relaxed values. By definition, the ratio of the normal to the in plane strain, $\xi(x)$ is given by

$$\xi(x) = -\frac{c(In_xGa_{1-x}N) - c_0(x)}{c_0(x)}\frac{a_0(x)}{a(In_xGa_{1-x}N) - a_0(x)} \tag{8}$$

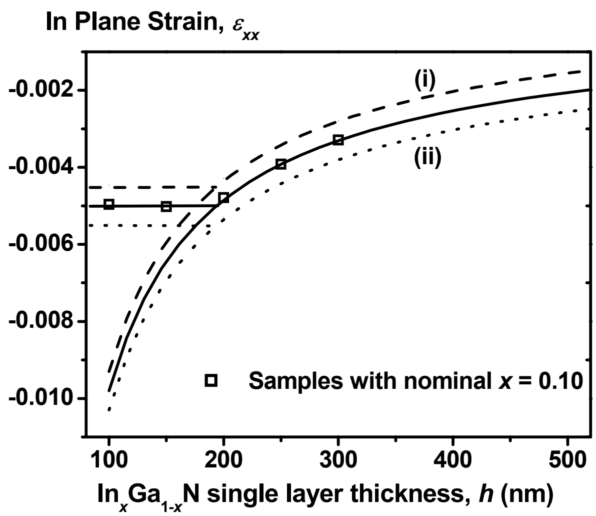
where c_0(x) and a_0(x) are the completely relaxed normal and in plane lattice constants of $In_xGa_{1-x}N$, respectively. c_0(x), a_0(x) and $\xi(x)$ for $In_xGa_{1-x}N$ has been obtained from the linear interpolation according to the Vegard's law. The completely relaxed lattice constants of GaN and InN are obtained from the reported results of Qian *et al.* [8] and Zubrilov *et al.* [9], while the widely accepted theoretical estimation of ξ of GaN and InN are taken from the values reported by Wright [10]. From the values of $c(In_xGa_{1-x}N)$ obtained from Eq (6) and the values of $a(In_xGa_{1-x}N)$ obtained from Eq (7) for a given composition x, the corresponding normal and in plane strain can be determined by solving Eq (8). For symmetry consideration, for uniaxial strain along the c-axis or biaxial strain in the plane normal to the c-axis, $\varepsilon_{xx} = \varepsilon_{yy}$ and $\varepsilon_{xy} = \varepsilon_{yz} = \varepsilon_{xz} = 0$, where ε_{ij} denotes the strain tensor components.

IV. DISCUSSION

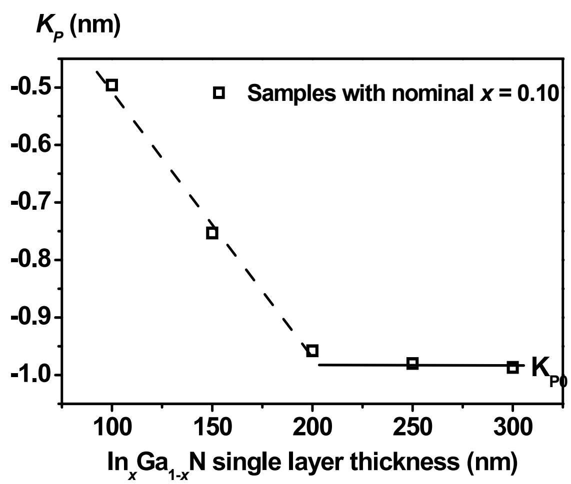
Figure 3 depicts the relation between the in-plane strain, ε_{xx} for the $In_{0.10}Ga_{0.90}N$ layers and their corresponding thicknesses. ε_{xx} is compressive in nature. For thicknesses above 200 nm, the compressive strain decreases hyperbolically with respect to the thickness, in accordance to the relation

$$\varepsilon_{xx} = \frac{K_{P0}}{h} \tag{9}$$

where K_{P0} is termed as the in-plane relaxation constant for $In_xGa_{1-x}N$/GaN epilayers grown on sapphire, as described in Eqs (4) and (5). Eq (9) therefore forms a universal relation; once a layer initiates relaxation, it follows the same strain-thickness relation independent of its original misfit until the dislocation motion becomes difficult due to the interaction and entanglement among dislocations, which is also known as work hardening. The solid hyperbolic curve in Figure 3(a) is the best fitted data for the $In_{0.10}Ga_{0.90}N$ samples using Eq (9), which gives a value of $K_{P0} = -0.98 \pm 0.03$ nm for the general $In_xGa_{1-x}N$/GaN epilayers grown on sapphire. The solid horizontal line in Figure 2(a) shows the constant compressive strain below the critical thickness. Therefore, the critical thickness can be obtained from the intersection between the solid horizontal line and the solid hyperbolic curve. Further illustration is shown in Fig. 3(b), which denotes the various values of K_P (= ε_{xx} x h) at different thicknesses. Below the critical thickness of approximately 200 nm for $In_{0.10}Ga_{0.90}N$, the magnitude of K_P increases linearly with thickness (dashed line) due to the constant compressive strain. Above this critical thickness, the $In_{0.10}Ga_{0.90}N$ layer relaxes plastically and K_P saturates to the solid line with a value of K_{P0} = −0.98 ± 0.03 nm. As relaxation continues, the magnitude of the compressive strain decreases, and when it reaches the elastic limit completely, no further plastic relaxation is expected to occur.



(a)



(b)

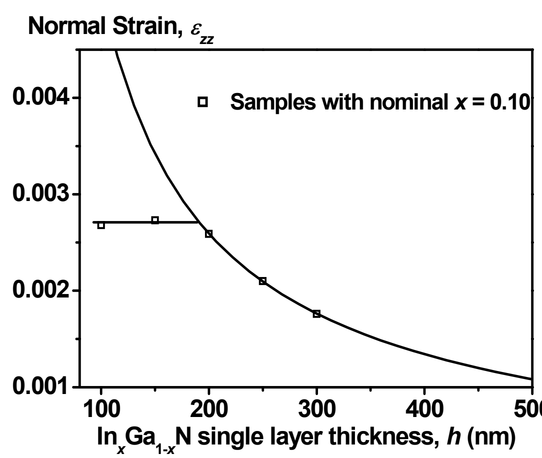
Fig. 3. (a) Variation of the in plane compressive strain ε_{xx} in the single $In_{0.10}Ga_{0.90}N$/GaN epilayers grown on sapphire with different thickness. The fitted solid hyperbolic curve is the universal theoretical hyperbolic relation for in plane compressive strain and thickness, $\varepsilon_{xx} = K_{P0}/h$ where K_{P0} = −0.98 ± 0.03 nm. The solid horizontal line shows the in plane compressive strain below critical thickness. The dashed (i) and dotted (ii) curves display the upper and lower bound of the strain measurement, affected by the resolution limit of the *x*-ray diffraction (XRD). (b) Variation of K_P (= ε_{xx} x *h*) in the single $In_{0.10}Ga_{0.90}N$/GaN epilayers grown on sapphire with different thickness. Below the critical thickness, K_P follows the linear relation with thickness, as shown by the linear dashed line. Above the critical thickness, K_P saturates to K_{P0} = −0.98 ± 0.03 nm, as illustrated by the solid horizontal line.

The dashed (i) and dotted (ii) curves in Figure 3(a) indicate the upper and lower range of the strain measurement due to the resolution limitation of the *x*-ray diffraction (XRD). This takes into consideration that the minimum composition of In that the equipment can distinguish is *x* = 0.005 at its most optimum operation condition, translating to an equivalent strain of approximately 5 x 10^{-4} for an $In_xGa_{1-x}N$/GaN grown on sapphire. Due to the hyperbolic shape, the disparity in thickness between curve (i) and (ii) becomes greater as the magnitude of the strain becomes lower. Hence, the two curves clearly justify one of the primary reasons for the force-balance model [5] to have a large discrepancy in the estimated critical thickness at low strain level, when compared to empirical results reported in the literature.

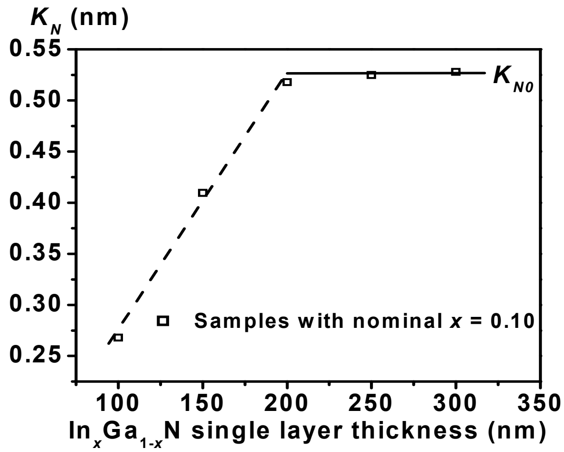
The normal tensile strain ε_{zz} of the single $In_{0.10}Ga_{0.90}N$ layers shows an identical relation to Eq (9) with respect to the thickness, as illustrated in Figure 4(a). Below the critical thickness of approximately 200 nm, ε_{zz} stays constant according to the solid horizontal line in Figure 4(a). Above this critical thickness, the normal tensile strain decreases hyperbolically with respect to the thickness, following the relation,

$$\varepsilon_{zz} = \frac{K_{N0}}{h} \tag{10}$$

where K_{N0} is the normal relaxation constant as described in Eqs (4) and (5). The relaxed strain for $In_{0.10}Ga_{0.90}N$ samples can be best fitted with the solid hyperbolic curve in Fig. 4(a), giving a value of K_{N0} = +0.51 ± 0.03 nm. The variation in K_N (= ε_{zz} x *h*) is depicted in Figure 4(b). Below the critical thickness, K_N decreases linearly with thickness (dashed line) due to the constant tensile strain. Above this critical thickness, K_N converges to the solid line with a value of K_{N0} = +0.51 ± 0.03 nm due the plastic relaxation.



(a)



(b)

FIG.4. (a) Variation of the normal tensile strain ε_{zz} in the single $In_{0.10}Ga_{0.90}N$/GaN epilayers grown on sapphire with different thickness. The fitted solid hyperbolic curve is the universal theoretical hyperbolic relation for the normal tensile strain and thickness, $\varepsilon_{zz} = K_{N0} / h$ where K_{N0} = +0.51 ± 0.03 nm. The solid horizontal line shows the normal tensile strain below the critical thickness. (b) Variation of K_N (= ε_{zz} x h) in the single $In_{0.10}Ga_{0.90}N$/GaN epilayers grown on sapphire with different thickness. Below the critical thickness, K_N follows the linear relation with thickness, as shown by the linear dashed line. Above the critical thickness, K_N saturates to K_{N0} = +0.51 ± 0.03 nm, as illustrated by the solid horizontal line.

V. Conclusion

From these experimental observations, we can conclude that single $In_xGa_{1-x}N$/GaN epilayers at any In composition grown on sapphire will exhibit an identical strain relaxation characteristic as shown in this study for x = 0.10. Also these layers will follow the same relaxation process and will give rise to the same K_{P0} and K_{N0} values after the critical thickness. We believe all III-Nitride and other semiconductor material systems will show the same strain relaxation feature as well. Knowledge of the relaxation constants would be useful to predict the strain component in multilayers and graded structures [11].

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