Buffer layer strain transfer in AlN/GaN near critical thickness

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X-ray diffraction has been employed to investigate the strain relaxation of both components of a GaN/AlN bilayer on sapphire (0001) as a function of the GaN layer thickness. Below a critical thickness, GaN and AlN both relax with the same in-plane lattice constant, consistent with the energy minimum condition of elasticity theory for a bilayer. Above the critical thickness, however, the strain relaxations in the two layers were different. We can fit this strain relaxation behavior with a free standing bilayer model with an additional term describing the interaction of dislocations. © *1999 American Institute of Physics.* [S0021-8979(99)00608-8]

The strain relaxation of lattice mismatched films is one of the areas of thin film physics which has not been completely understood yet. With the rapidly growing demand for thin films for industrial applications, more fundamental understanding of strain properties of thin films is desirable. The recent interest in III-nitride material for the fabrication of short wavelength optical devices[1–3] and high power electronic devices[4] has spawned interest in the interactions between adjacent layers in which the distribution of strain is involved.

Growing interest in GaN for blue and ultra-violet region optical devices requires high quality film growth. For many reasons, sapphire is an appropriate substrate material for GaN, but since the lattice mismatch between GaN and sapphire is 13.9% (without a buffer layer), misfit dislocations result in high defect density which degrade the quality of the GaN film. Some years ago there were reports that introduction of AlN buffer layer, which improves the crystallinity and surface morphology of the GaN,[5,6] enhanced the electrical and luminescent properties as well. Since then, AlN has been widely used for a buffer layer for GaN.

In a previous paper,[7,8] we investigated the strain relaxation of GaN grown on sapphire (0001) with a thin AlN buffer layer and demonstrated that the results could be explained with a theory in which the upper film was in mechanical equilibrium with the buffer layer. This analysis was based on an implicit assumption of an inert AlN buffer layer. However, in that work we did not test for the possibility that structure of the AlN buffer layer could have been influenced by the GaN on top of it.

Meanwhile, Huang and Wang[9] have shown that mutual elastic interaction between two thin films can be important especially when the buffer layer is very thin. Because the strain can be transferred between films, the critical thickness can be increased in certain cases. In the light of this theory, we decided to reexamine our previously measured samples for evidence of strain in the AlN layers, seen in their in-plane lattice constants. As before, a least-squares fit of multiple Bragg peaks was employed to attain an accurate three-dimensional determination of lattice constants.[7] For those samples in which the in-plane lattice constants of GaN and AlN were very close, reciprocal lattice mapping (RLM) was needed to unambiguously assign the peaks to GaN or AlN. In addition to those we measured previously, new samples were measured this time. The details regarding preparation and growth of the samples can be found in our previous publication.[7]

The x-ray diffraction experiments used a synchrotron radiation source of x rays at beamline X16C of National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory. A double crystal Si(111) monochromator produced x rays of 8.95 keV ($\lambda = 1.39$ Å). A four-circle diffractometer featuring *kappa geometry*[10] was used for measurements. Our experimental resolution was determined by 2 mm 2 mm slits in front of a scintillation detector. The angular positions in three dimensions of at least six Bragg peaks were used in a least-squares calculation to define the sample orientation as well as the six parameters describing the crystal lattice. In this paper we consider only the variation of the in-plane lattice parameter among the different samples.

The samples which we measured are listed in Table I with the resulting in-plane lattice constants. All samples were found to show good (001) epitaxial orientation with small mosaic spreads. Sample A consisted of just the buffer layer with 32 Å of AlN. All other samples had 32 Å of AlN followed by GaN with different thicknesses. Samples B and C were found to have the same in-plane lattice constants for AlN and GaN, as determined from the least-squares fit method, within experimental error. To be sure of pseudomorphic growth, reciprocal lattice maps of the region surround-

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TABLE I. Nominal thickness and measured in-plane lattice constants of the GaN on AlN samples used in this work.

Sample	AlN (Å)	GaN (Å)	a_{AlN} (Å)	a_{GaN} (Å)
A	32	⋯	3.084	⋯
B	32	25	3.134	3.134
C	32	50	3.142	3.142
D	32	100	3.103	3.174
E	32	500	⋯	3.192
F	32	10^4	⋯	3.196

ing their (103) Bragg peaks were made. Figure 1 for sample C shows two peaks clearly separated along the surface-normal reciprocal direction l with the same in-plane h value. We chose to align on the reciprocal lattice of GaN for these measurements, so the peak exactly indexing at (103) arises from the GaN while the one above it is from AlN. This reciprocal lattice map clearly shows that the GaN is pseudomorphic with the AlN. The AlN is moreover substantially strained from its situation in sample A, containing just the buffer layer. This important fact was overlooked in our previous measurement[7] because of the weak intensity and its proximity to that of the GaN. In Fig. 2, we show an index scan along l direction through the (103) peak of the same sample. The two peaks are clearly resolved allowing a two-Gaussian fit to be made. From these results, it is unambiguous that both AlN and GaN have the same value of in-plane lattice constant. For sample B, pseudomorphic growth of GaN on AlN was similarly observed, but with both in-plane lattice parameters different from those of sample C.

Sample D had a sufficiently thick layer of GaN that its intensity dominates over that of AlN in such a way that it was not possible to find the AlN peaks by conventional searching, even though the peak separation and instrument resolution should have allowed this. Therefore reciprocal lattice mapping was used again to locate the weak AlN peak. In Fig. 3, the GaN peak is dominant and a very weak AlN peak is seen on its side. It is easily seen that the two films now have different in-plane lattice parameters. The lateral shift between the two peaks is clear evidence for nonpseudomorphic growth of GaN. Once located with the help of the reciprocal lattice map, we were able to make an index scan through (103) AlN peak, as shown in Fig. 4. Symmetry equivalent peak positions for the AlN were then input to the least-squares calculation[7] to obtain the in-plane lattice constant. Note that the h,k,l labels in Fig. 4 are with respect to the AlN reciprocal lattice. Even though the peak is very broad in l direction, as expected for such a thin film, the full width at half maximum is smaller in h,k direction, which gives a good determination of its in-plane position. The

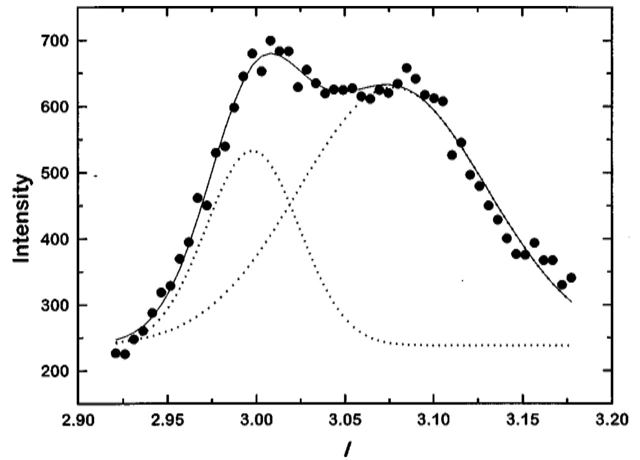

FIG. 2. Index scan along the surface-normal l direction through (103) peak. Gaussian functions have been used to fit the two peaks.

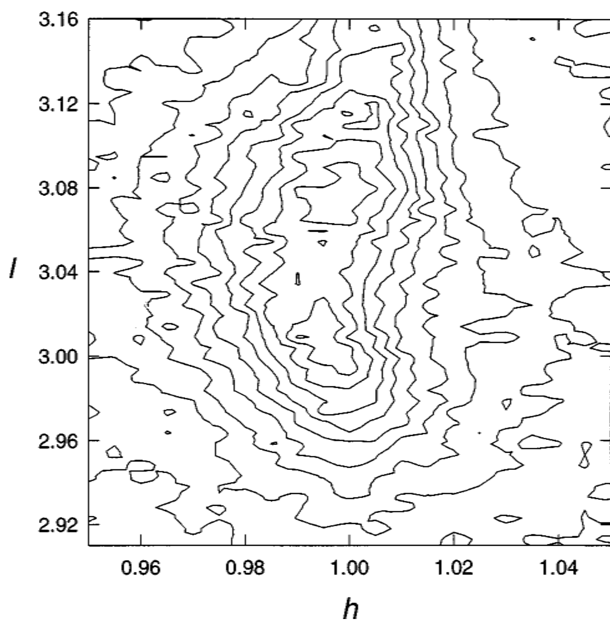

FIG. 1. Contour plot of a reciprocal lattice map near the (103) Bragg peaks of sample C. The GaN reciprocal lattice has been used for indexing. Two peaks with the same h value are clearly visible.

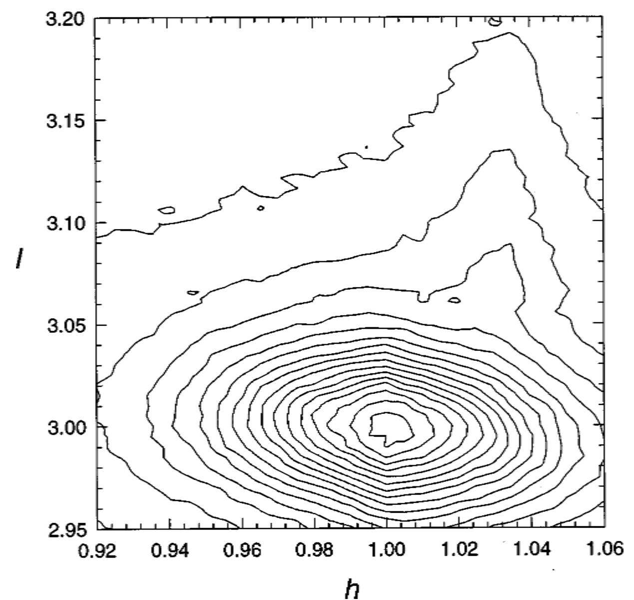

FIG. 3. Contour plot of a reciprocal lattice map near the (103) Bragg peaks of sample D. The GaN reciprocal lattice has been used for indexing. A very weak AlN peak is shown on the shoulder of the large GaN peak.

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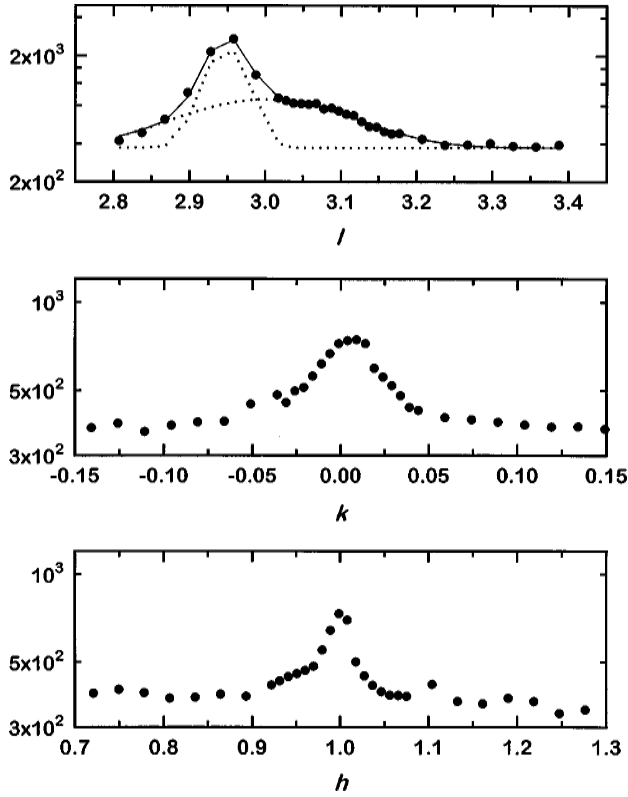

FIG. 4. Index scan along in all three directions of the weak AlN peak found in Fig. 3. A fit of Gaussians has been done for the l scan.

shape and intensity of AlN peak in samples C (pseudomorphic) and D (nonpseudomorphic) were about the same which confirms that the peak is truly due to the AlN layer. This locates the critical thickness for pseudomorphic growth in between samples C and D.

Our experimental results show an interesting trend among the samples in Table I, whose in-plane lattice parameters are plotted in Fig. 5. The in-plane lattice constant of AlN *increases* up to the critical thickness and then begins to decrease as soon as the pseudomorphic growth regime ends, rather than just staying constant. From the microscopic perspective, the critical thickness is the threshold below which dislocations are prevented from forming at the interface between the layers. It is therefore clear that, once misfit dislocations are introduced, the GaN continues to relax normally, in the same direction as below the critical thickness, but the AlN reverses its trend and shrinks back.

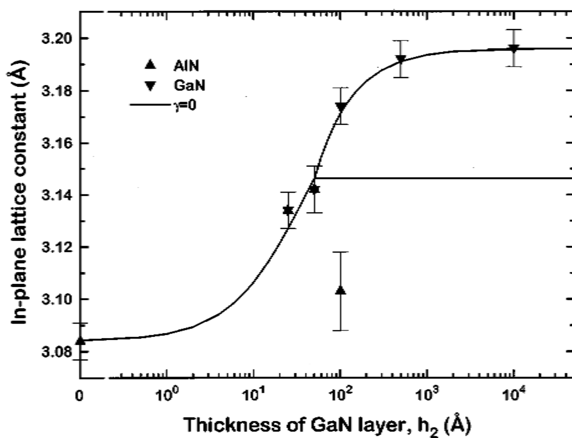

FIG. 5. In-plane lattice constants of the AlN and GaN films with fit curves based on Eqs. (2) and (6).

We now turn to a discussion of a possible explanation of the results. In all our samples, 32 Å of AlN buffer layer were grown on the sapphire substrate followed by a subsequent GaN layer. The in-plane lattice mismatch between sapphire and AlN is so large that misfit dislocations are expected to form at the interface to accommodate it even at a few layers growth. The dislocation densities at a sapphire-AlN interface are expected to be much higher than at an AlN-GaN interface. Because of this higher dislocation density, the response of AlN layer to the stress coming from sapphire can be assumed to be less important than that to GaN. It is therefore reasonable to make the approximation that AlN and GaN form a *free standing bilayer*. The only effect of sapphire substrate is a slight modification of the AlN in-plane lattice constant to 3.084 Å in the following calculation, through epitaxial strain well beyond its critical thickness. This value is consistent with a calculation for 32 Å of bare AlN on sapphire and the bulk elastic constants of AlN.

Based on this assumption, and following the strain transfer model of Huang and Wang,[9] the total energy of AlN and GaN bilayer in the pseudomorphic (thin film) limit consists only of elastic strain energy which can be expressed as[9]

$$E=\alpha_1 h_1(a-a_{10})^2+\alpha_2 h_2(a-a_{20})^2, \tag{1}$$

where a is the common in-plane lattice constant of two films. It should be noted that this description is only valid up to a certain thickness in h_2 (GaN) for which the lattice mismatch between two films is fully accommodated by strain so that both films have the same in-plane lattice constant. Within the assumption of growth of nitride films in mechanical equilibrium, the minimization condition, $\partial E/\partial a=0$, gives us

$$a=\frac{\alpha_1 h_1 a_{10}+\alpha_2 h_2 a_{20}}{\alpha_1 h_1+\alpha_2 h_2}=\frac{h_1 a_{10}+\dfrac{\alpha_2}{\alpha_1}h_2 a_{20}}{h_1+\dfrac{\alpha_2}{\alpha_1}h_2}. \tag{2}$$

Since h_2 was the variable in our experiment, Eq. (2) can be plotted as the curve through the first three data points in Fig. 5 where α_2/α_1 was set to 0.8 for the best fit. This value roughly agrees with the ratio of the calculated shear modulus of GaN and AlN, which is 0.88.[11,12] $a_{10}=3.084$ Å is the effective in-plane lattice constant of the 32 Å film of AlN and we used $a_{20}=3.196$ Å for the bulk value of GaN as was determined in our previous work.[7]

Once GaN reaches the thickness where misfit dislocations are introduced at the interface of two films, a dislocation term must be added to the equation for the total energy in Eq. (1). The new term represents the dislocation energy,[8] which is proportional to the density of dislocations,

$$E=\alpha_1 h_1(a_1-a_{10})^2+\alpha_2 h_2(a_2-a_{20})^2+\beta\, a_1-a_2\,, \tag{3}$$

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where a_1 is the in-plane lattice constant of AlN and a_2 is that of GaN. Since AlN behaves as a virtual substrate for GaN and vice versa, the dislocation energy term is proportional to $a_2 - a_1$, which is a quantity proportional to the density of dislocations. In our case, a_2 is always larger than a_1, $a_1 - a_2 = a_2 - a_1$. In the same way as above, the minimization condition gives us

$$\frac{\partial E}{\partial a_1} = 2\alpha_1 h_1 (a_1 - a_{10}) - \beta = 0, \tag{4}$$

$$\frac{\partial E}{\partial a_2} = 2\alpha_2 h_2 (a_2 - a_{20}) + \beta = 0. \tag{5}$$

So,

$$(a_1 - a_{10}) h_1 = \frac{\beta}{2\alpha_1}, \quad (a_2 - a_{20}) h_2 = -\frac{\beta}{2\alpha_2}. \tag{6}$$

An attempt to fit these equations to the lattice constant data is shown as the remaining solid curves on the right side of Fig. 5. The equation of the curve for a_2 (GaN) is identical to the function used in our previous work,[7] which fits the GaN data quite well, as shown. However, in our new experiment, h_1 (AlN) was not varied, so a_1 should just remain constant according to this theory. This disagrees with our empirical observation that the AlN lattice parameter relaxes back towards its bulk value above the critical thickness, so we must question our assumption that the energy due to the dislocations is only proportional to their density.

We therefore attempted to improve the theory by including a higher-order term for the dislocation energy, which might be related to the *interactions* between dislocations, for example. The simplest form of interaction would vary as the square of their density. With this higher-order correction, the new form of total energy is

$$E = \alpha_1 h_1 (a_1 - a_{10})^2 + \alpha_2 h_2 (a_2 - a_{20})^2 + \beta (a_2 - a_1) + \gamma (a_2 - a_1)^2, \tag{7}$$

and the minimization conditions are

$$\frac{\partial E}{\partial a_1} = 2\alpha_1 h_1 (a_1 - a_{10}) - \beta - 2\gamma (a_2 - a_1) = 0, \tag{8}$$

$$\frac{\partial E}{\partial a_2} = 2\alpha_2 h_2 (a_2 - a_{20}) + \beta + 2\gamma (a_2 - a_1) = 0. \tag{9}$$

From Eqs. (8) and (9),

$$\begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \frac{1}{h_1 h_2 + \gamma\ h_1 + \epsilon\gamma\ h_2} \begin{pmatrix} h_2 (h_1 a_{10} + \epsilon\beta\ + \epsilon a_{20}\gamma\ ) + \gamma\ h_1 a_{10} \\ h_2 (\epsilon a_{20}\gamma\ + h_1 a_{20}) + \gamma\ h_1 a_{10} - h_1 \beta \end{pmatrix}, \tag{10}$$

where $\gamma\ = \gamma/\alpha_2$, $\beta\ = \beta/(2\alpha_2)$ and we used $\epsilon = \alpha_2/\alpha_1 = 0.8$. This provides explicit functional forms of a_1 and a_2 as functions of h_2 with two adjustable parameters, γ and β. The functional form of Eq. (10) is shown for different values of γ in Fig. 6. This time the h_2 dependence of a_1 can be explained and its direction depends upon the sign of γ. In Fig. 7, the best fit with $\gamma = -18$ and $\beta = 2.49$ is shown along with our data.

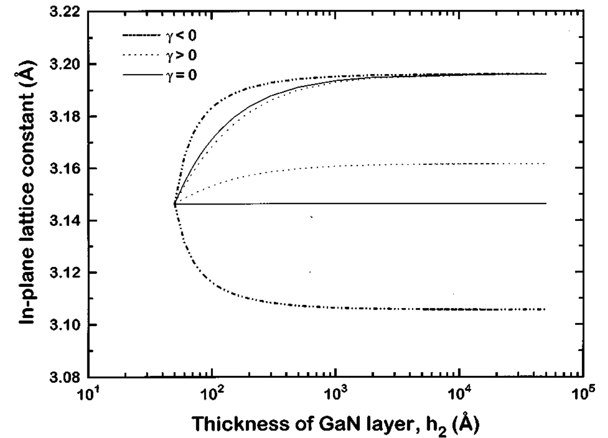

FIG. 6. Variation of the in-plane lattice constants of GaN and AlN predicted for different γ values.

The coefficient γ represents the interaction between dislocations. To explain the downwards relaxation of the AlN lattice constant back towards its unstrained value, γ needs to be negative. This is necessary so that it becomes energetically favorable for the system to increase the density of dislocations once the critical thickness has been reached. It implies an *attractive* interaction between the dislocations. While dislocations can indeed in some circumstances be attracted together, for example to form tilt boundaries,[12] this is not expected at an interface between two layers. Nevertheless the experimental observation of the rebounding of the AlN lattice constant, once dislocations have started to form, at least makes intuitive sense. Furthermore, a careful comparison between the quality of the fit curves in Fig. 5 and Fig. 7 shows that the simpler theory fits the GaN lattice constant data better. These two observations may expose a weakness in the second theoretical approach given, which invites further discussion.

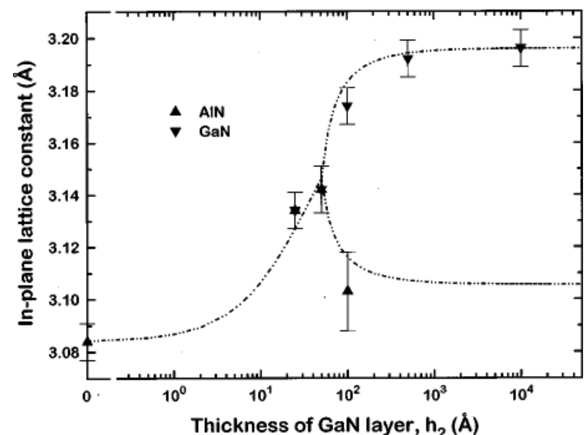

FIG. 7. In-plane lattice constants of the AlN and GaN films with fit curves based on Eqs. (2) and (10).

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In summary, the parallel strain of a GaN on AlN bilayer structure grown on a sapphire (0001) substrate has been investigated using x-ray diffraction as a function of the GaN layer thickness. It was found that *both* films are strained, with the AlN layer dilated and the GaN layer compressed. Below the critical thickness, both films have the same in-plane lattice constant which increases with GaN thickness. This can be explained by the elastic strain-transfer model due to Huang and Wang.[9] Beyond the critical thickness, however, the in-plane lattice constants split up so that each film relaxes back towards its unstrained in-plane lattice constant value.

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