Extrapolation of critical thickness of GaN thin films from lattice constant data using synchrotron X-ray

Chinkyo Kim[*], I. K. Robinson[*], Jaemin Myoung[**], Kyuhwan Shim[**], Kyekyoon Kim[***], and Myung-Cheol Yoo[****]
[*]Department of Physics, University of Illinois at Urbana-Champaign, 1110 W.Green St., Urbana, IL 61801
[**]Department of Material Sciences and Engineering, University of Illinois at Urbana-Champaign
[***]Department of Electrical and Computer Engineering, University of Illinois at Urbana-Champaign, 1406 W.Green St., Urbana, IL 61801, kevinkim@ux1.cso.uiuc.edu
[****]Samsung Advanced Institute of Technology, Suwon, Korea

ABSTRACT

In some materials, Van der Merwe's equilibrium theory of strain relief is believed to explain the sudden transition from pseudomorphic growth of a thin film to a progressively relaxed state. We show, for the first time for GaN, how an accurate estimate of the critical thickness of a thin film can be extrapolated from suitable measurements of lattice constants as a function of film thickness using synchrotron X-ray. We do this both for an elementary elastic energy function, in which the interactions between the dislocations are ignored, and for a more realistic energy estimate due to Kasper. The method is found to work quantitatively for thin films of GaN on AlN. The critical thickness is determined to be 29 ± 4 Å.

INTRODUCTION

Ever since molecular beam epitaxy has been used to grow materials, many attempts have been made to grow pseudomorphic epitaxial films on substrates with different lattice constants for many different combinations. However, due to the lattice mismatch it is not possible to grow pseudomorphic films with arbitrary thickness. In the initial stage of growth, the misfit can be accommodated by the strained film alone, but above a certain thickness, the so-called critical thickness, a spontaneous emergence of misfit dislocations cannot be avoided. Equilibrium theory generally predicts extremely small values for the critical thickness, which are sometimes almost impossible to measure directly. So we demonstrate here a method which allows us to extrapolate the critical thickness from a series of lattice constant measurements.

In thermodynamic equilibrium, misfit dislocations appear at the interface of strained layer heterostructures when the strained layer is thick enough that it is energetically favorable for the mismatch to be accommodated by a combination of elastic strain and interfacial misfit dislocations, rather than by elastic strain alone.[1] The equilibrium assumption does not always apply: in some well-studied systems, such as Ge_xSi_{1-x}, kinetic effects are more important.[2] In this work we show that the equilibrium theory is appropriate for thin GaN films on AlN buffer layer on sapphire (0001). Therefore, in the initial stage of growth, when the film is below the critical thickness, GaN is expected to grow pseudomorphically on AlN. Above the critical thickness, the strain begins to relax by spontaneous creation of dislocations at the GaN/AlN interface. Subsequently the lattice constant begins to approach the bulk value as the thickness of GaN increases further.

Mat. Res. Soc. Symp. Proc. Vol. 423 © 1996 Materials Research Society

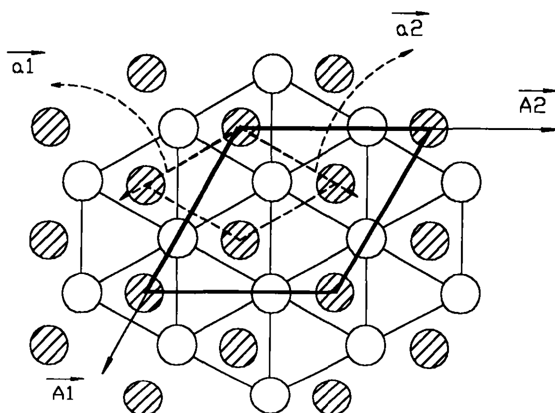

Fig. 1 Unit cells of sapphire (0001) and AlN on top of it. Solid line is for sapphire and dashed line for AlN. Open circles and hatched ones represent O in sapphire and Al in AlN, respectively.

EXPERIMENT

To test those ideas which will be explained in the next section we examined measurements of lattice constants of GaN thin film grown on AlN buffer layers on sapphire (0001).[3] The AlN buffer layer between the sapphire and the GaN was used because its in-plane lattice constant is intermediate, as used previously[4], which redistributes the substantial mismatch between two interfaces. GaN samples with thicknesses in the range from 50 Å to 1 μm were grown by MBE (Molecular Beam Epitaxy) on sapphire (0001) using a 32 Å AlN buffer layer. In order to have a workable signal from the thinnest films, X-ray beamline X16C at the National Synchrotron Light Source (NSLS) was employed to determine the lattice constant a of the GaN films using a least-squares fit method.[3] Fig.1 shows the known epitaxial arrangement of AlN on sapphire (0001)[5,6] and table I[7] lists the known bulk hexagonal lattice constants of the relevant materials. If there were no lattice mismatch, the in-plane lattice constant of AlN would be expected to be $4.758/\sqrt{3} = 2.747$ Å, considerably less than the bulk value of 3.112 Å. Instead, we found a = 3.084 Å indicating a partial compression of the buffer layer.

THEORY

First we consider the relationship between the film thickness and the lattice constant a using Van der Merwe's energy minimization theory.[8] From elementary elasticity theory the strain energy per unit area is proportional to $h(a_0 - a)^2$, where h is a film thickness, a_0 is the lattice constant of the (completely relaxed) material of the film and a is the actual lattice constant of the film. Assuming that the effective range of the dislocation field is constant, the energy due to

Table I Physical parameters of GaN, AlN and Sapphire

	a (Å)	c (Å)
GaN	3.189	5.185
AlN	3.112	4.982
Sapphire	4.758	12.991

dislocations depend on their density alone, which is proportional to $(a - a_s)$. Then, the total energy of the strained film is given by

$$E = \alpha h(a - a_o)^2 + \beta(a - a_s) \quad (1)$$

where α and β are constants. For the system to be in equilibrium, this energy should be a minimum: $\partial E/\partial a = 0$. Then, to obtain the value of h_c, the critical thickness, we simply need to consider $a = a_s$. Setting $\partial E/\partial a = 0$ also gives a relation between h and a so long as $a > a_s$. At $a = a_s$, h is equal to h_c below which the film is pseudomorphic and a will have the same lattice constant as the substrate, a_s. The derived relation between a and h from Eq.(1) is

$$a = a_o + \frac{h_c}{h}(a_s - a_o) \quad (2)$$

The method we have used to determine h_c experimentally is to fit lattice constant data for thicker films, and use Eq.(2) as an extrapolation formula.

Now we consider the total energy more rigorously. The elastic strain ε depends on the mismatch between a film and its substrate, m, as well as the average number of dislocations present at the interface.[9] The relationship is given by

$$m = |\varepsilon| + \frac{b'}{p} \quad (3)$$

where $m=(a_o-a_s)/a_s$, $|\varepsilon| = (a_o-a)/a_o$, a_s is the lattice constant of the substrate, b' is the active component of the Burgers vector and p is the average distance between dislocations. Assuming ideal misfit dislocations we obtain the following expression.

$$\frac{b}{p} \approx m - |\varepsilon| \approx \frac{(a - a_s)}{a_o} \quad (4)$$

Using the expressions from Kasper[2,9], a better estimate of the energy of the dislocations per unit length is given by

$$E_{ds} = \frac{\mu b^2}{4\pi(1 - \nu)p}\left[1 + \ln\left(\frac{Q}{q}\right)\right] \quad (5)$$

where q is the inner cut-off radius, given by[2] $q= \pi b/(2\sqrt{2}(1-\nu))$, μ is the shear modulus, ν is the Poisson's ratio, b is a Burgers vector. Q is the effective range of the misfit dislocation field, and has two limiting values that depend on the density of dislocations, p

$$Q = p/2 \qquad \text{when } h \gg p/2 \quad (6)$$

$$= \frac{p(4hp)}{2(p^2 + 4h^2)} \approx 2h \qquad \text{when } h \ll p/2 \quad (7)$$

We will use another approximation, $Q = hp/(2h + p/2)$, to interpolate smoothly between the two limits of Kasper. This then applies over the entire domain of h. With this approximation, the total energy of a film with thickness h is given by

$$E \approx \alpha(a - a_o)^2 h + \beta(a - a_s)\left(1 + \ln(\frac{2hp}{q(p + 4h)})\right) \tag{8}$$

Following the same calculation we used to obtain Eq.(2), we find,

$$(a - a_o)h = \left[1 + \ln\left(\frac{2hp}{q(p + 4h)}\right) - \frac{1}{p^2} + \frac{1}{p(p + 4h)}\right]\left[\frac{(a_s - a_o)h_c}{1 + \ln(2h_c / q)}\right] \tag{9}$$

where $p=ba_o/(a-a_s)$ as before. Since Eq.(9) is an implicit function, we must solve it numerically to obtain a as a function of h.

RESULTS

Our results for the lattice constant a of the GaN films are plotted in Fig. 2 as a function of their thickness. They show a progressive trend away from the bulk GaN lattice constant for thinner films. The in-plane lattice constants are found to lie between those of bulk GaN and the AlN buffer layer. It is clear that if the trend towards thinner films were extrapolated slightly, the GaN lattice constant would cross that of the AlN buffer layer. This is the situation that occurs at

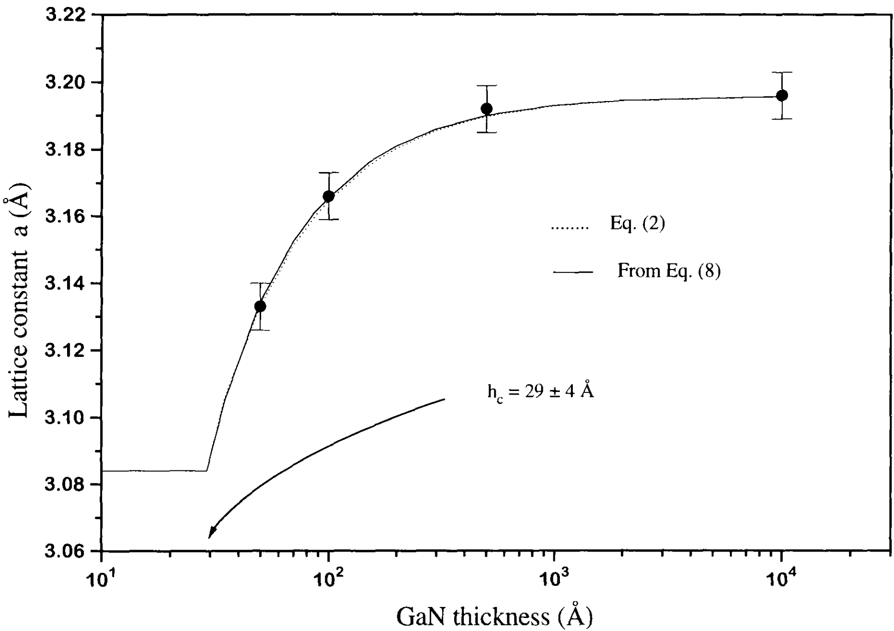

Fig. 2 Fit of lattice constant a as a function of GaN layer thickness

the critical thickness, h_c of GaN; thinner films would simply have a pseudomorphic epitaxial relationship with the AlN substrate. In order to establish a quantitative estimate of h_c, it is necessary to make a theoretical fit to the data for the trend of lattice constant with thickness, which we explained above. We fitted our data to our extrapolation formula, Eq.(2) and obtained the best fit over the entire region of film thickness with $h_c = 28 \pm 4$ Å.

With this initial knowledge of the relevant parameters we find that the thickness crossover between the two limits of Kasper (Eqs.(6) and (7)), h=p/2, falls at h= 72Å, right in the middle of the range of our data. We are therefore justified in using the more accurate treatment of Eq. (9). We therefore fit the data again using Eq.(9) with q = 5.525 Å, obtained from $\nu = 0.38$. This gave the second curve in Fig.2 and $h_c = 29 \pm 4$ Å.

SUMMARY AND CONCLUSIONS

We extrapolated the lattice constants of GaN film and obtained the critical thickness of GaN on AlN buffer layer with a sapphire substrate. The lattice constant a was observed in the region between 3.133 Å and 3.196 Å. The critical thickness of GaN was determined to be 29 ± 4 Å. This value agrees extremely well with the value of h_c predicted by the theory of Fischer et al,[3,10] 31.5 Å Since this theory is also based on energetic equilibrium arguments, it appears to support the conclusion that the behavior of GaN thin films, unlike Ge_xSi_{1-x} films for example, can indeed be described by equilibrium models.

Our second conclusion is a practical one. The logarithmic correction for the interaction between dislocations, which was used to obtain the more accurate interpolation formula, Eq.(9), is apparently not very important for the determination of h_c. This can be seen by comparing the two fit curves in Fig.2, which are almost identical. The simple analytic form of Eq.(2) is apparently sufficient to describe the thickness trend of the lattice constant and thereby allow the accurate determination of the critical thickness from its fit.

ACKNOWLEDGMENTS

We thank Dr. Mauro Sardela for useful discussions. This work was supported by the United States Department of Energy under grant DEFG02-91ER45439 and by Samsung Electronics Co. Ltd.. NSLS (National Synchrotron Light Source) is supported by the U. S. DOE under grant DE-AC012-76CH00016.

REFERENCES

[1]G. C. Osbourn, IEEE J. Quantum Electron. QE-22, 1677 (1986)
[2]S.C. Jain, J. R. Willis, and R. Bullough, Adv. in Physics **39**, 127 (1990)
[3]C. Kim, I. K. Robinson, J. Myoung , K. Shim, K. Kim, M. Yoo, to be published
[4]H. Amano, K. Hiramatsu and I. Akasaki, Jpn. J. Appl. Phys. **27**, L1384 (1988)
[5]R. C. Powell, N. -E. Lee, Y. -W. Kim, and J. E. Greene, J. Appl. Phys. **73**, 189 (1993)
[6]P. Kung, C. J. Sun, A. Saxler, H. Ohsato, and M. Razeghi, J. Appl. Phys. **75**, 4515 (1994)
[7]H. Morkoc, S. Strite, G. B. Gao, M. E. Lin, B. Sverdlov, and M. Burns, J. Appl. Phys. **76**, 1363 (1994)
[8]J. H. van der Merwe, Surf. Sci. **31**, 198 (1972)
[9]E. Kasper and H. -J. Herzog, Thin Solid Films **44**, 357 (1977); E. Kasper, Surf. Sci. **174**, 630 (1986)
[10]A. Fischer, H. Kuhne, and H. Richter, Phys.Rev. Lett. **73**, 2712 (1994)