A NEW APPROACH FOR DETERMINING EPILAYER STRAIN RELAXATION AND COMPOSITION THROUGH HIGH RESOLUTION X-RAY DIFFRACTION

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ABSTRACT

We developed a new method of determining epilayer relaxation (along one direction) and composition using a symmetric and any single asymmetric high resolution x-ray diffraction scan. The previous use of small angle approximations can be very detrimental to calculated results and should be avoided. This new method does not employ small angle approximations or first order Taylor approximations, producing accurate results. The effect of x-ray geometry (glancing incident versus glancing exit) on the analysis of epilayer composition and strain is also reviewed. It is also shown that the glancing exit geometry is generally less susceptible to experimental error.

INTRODUCTION

High resolution x-ray diffraction is a proven technique for measuring epilayer composition and relaxation in lattice-mismatched single crystal systems. To measure both quantities, the perpendicular ($a_\perp$) and in-plane ($a_{||}$) lattice constants of the layer must be known.[1] The composition and relaxation are calculated from the lattice constants by the use of Poisson's ratio (ν) for the layer. A useful and accurate formalism for determining the relaxation and composition of epilayers has been done by Bartels and Nijman,[1] and more eloquently described by Swaminathan and Macrander,[2,3] and Leiberich and Levkoff.[4] In this model, the perpendicular and parallel strains are calculated from the peak splittings ($\Delta\omega_{gi}$ and $\Delta\omega_{ge}$ - the angular distance between the substrate and epilayer peaks in the glancing incident or glancing exit geometry (Fig. 1)). This allows the contribution of the change of the interplanar angle $\Delta\phi$ between layer and substrate to be differentiated from the peak splitting due to compositional changes ($\Delta\Theta$). In addition, tilt between an epilayer and substrate will apparently alter the peak splitting between the layer diffraction peak and the substrate diffraction peak. The effect of this tilt can be eliminated by averaging scans at 180° azimuthal positions. All subsequent calculations do not further address the effects of this tilt.

It is desirable, however, to calculate relaxation and composition of epilayers with as few scans as possible, e.g., a symmetric and perhaps one asymmetric diffraction scan (for example, the (004) and (115)-glancing incident diffraction planes). The (004) diffraction plane is generally the most intense in diamond and zinc-blende structures, and higher order 00*l* planes are inherently most accurate for measuring the perpendicular lattice constant in {001} crystal systems. Another advantage is that the symmetric scans may also be used to calculate epilayer and substrate tilts, noted above.

So far, attempts to utilize a symmetric scan to help determine relaxation have been hampered by the use of small-angle approximations in the calculations of the parallel lattice

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constant.[5] Herzog and Kasper have developed a useful set of relations, using low order Taylor expansions instead of small angle approximations, which allows for the parallel lattice constant to be determined from *any* two diffraction scans, with at least one being asymmetric.[6] However, this analysis still contains error, in both the relaxation and composition values, for samples exhibiting low relaxation, and at larger Bragg angles.

In this paper, we present a more accurate means to determine both relaxation and composition by making use of a symmetric and any single asymmetric scan. An analytical solution is given, as well as the format for an iterative method. Both solutions contain no error. In addition, we compare the results obtained by utilizing a glancing incident versus glancing exit diffraction plane, making recommendations on the best diffraction planes for relaxation analysis.

THEORY

Traditionally, epilayer strain and relaxation is calculated by performing two asymmetric diffraction scans from the same diffraction plane, rotated azimuthally 180°. The azimuthal rotation is necessary to differentiate the contribution of $\Delta\phi$ (the difference in angle between equivalent planes of the substrate and layer) and x (the composition) to the angular position of the epilayer peak. It is noted that $\Delta\phi$ is directly related to the amount of strain in the layer. In actual practice, it is more convenient to use (hkl) and $(\bar{h}\bar{k}l)$type of reflections, instead of rotating the sample 180°, as shown in Fig. 1.

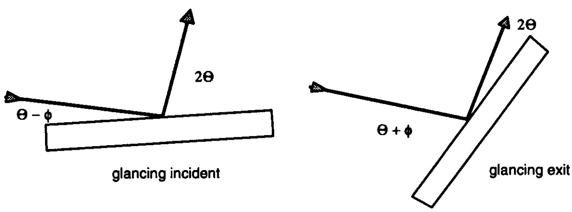

Figure 1. Schematic showing the definition of terms (a) glancing incident (gi) and (b) glancing exit (ge).

This geometry is typically referred to as glancing incident (gi) and glancing exit (ge). We will use a subscript following any asymmetric diffraction plane (hkl) to differentiate between the two geometries in the rest of the text.

In the conventional approach, the perpendicular ($a_\perp$) and parallel ($a_\parallel$) lattice constants can be calculated from the glancing incident and exit peak splittings ($\Delta\omega_{gi}$ and $\Delta\omega_{ge}$, respectively), utilizing the equations:[3]

$$a_\perp = a_s \cdot \frac{\sin\Theta_B}{\sin(\Theta_B + \Delta\Theta)} \cdot \frac{\cos\phi}{\cos(\phi + \Delta\phi)} \tag{1}$$

$$a_\parallel = a_s \cdot \frac{\sin\Theta_B}{\sin(\Theta_B + \Delta\Theta)} \cdot \frac{\sin\phi}{\sin(\phi + \Delta\phi)} \tag{2}$$

where a_s represents the substrate lattice constant, Θ_B is the substrate Bragg angle, $\Delta\phi = \Delta\omega_{ge} - \Delta\omega_{gi}/2$ and $\Delta\theta = \Delta\omega_{ge} + \Delta\omega_{gi}/2$.

However, the perpendicular lattice constant can be independently calculated from a symmetric scan, i.e. the (004) diffraction plane of a (001)-oriented semiconductor crystal. This is accomplished without approximation by:[1]

$$a_{\perp} = \left[\frac{\sin\Theta_B}{\sin\left(\Theta_B + \Delta\Theta_{sym}\right)} - 1 \right] \cdot a_s + a_s \qquad (3)$$

where $\Delta\Theta_{sym}$ is the angular distance between the substrate and epilayer peak. Using this calculated value for the perpendicular lattice constant, Eqn. (1) can be rewritten such that there is only one unknown ($\Delta\phi$), which can be determined from the $\Delta\Theta_{(gi\ or\ ge)}$ from any asymmetric scan.

$$\sin(A + \Delta\phi) \cdot \cos(\phi_{substrate} + \Delta\phi) = \frac{\lambda \cdot l}{2a_{\perp}} \qquad (4)$$

where $A = \Theta_B + \Delta\theta_{(gi\ or\ ge)}$ (depending on the geometry of the performed asymmetric scan), l is the index from the asymmetric plane, and λ is the radiation wavelength. Solving for $\Delta\phi$, however, is not a trivial process, due to the fact that it is embedded into both *sine* and *cosine* terms. Small angle approximations can be used to simplify the process,[5] however too much error is introduced. Thus, to obtain a reasonable $\Delta\phi$ value at all levels of relaxation, better approximations, or an iterative process such as the secant method, should be used. Although the iterative method is superior in nature, we have also developed an analytical technique which solves for $\Delta\phi$ with a negligible amount of error, given by:

$$\Delta\phi = \frac{\sin(A)\sin(\phi) - U \cdot \cos(A)\cos(\phi) + V \cdot \sqrt{\left(\sin(A)\sin(\phi) + U \cdot \cos(A)\cos(\phi)\right)^2 + 4\sin^2(A)\cos^2(\phi) + 4DE}}{2E} \qquad (4)$$

where $U = \pm 1$ for g.i. and g.e. respectively, $V = -1$ for all g.e. and any g.i. scan where $\phi > 45°$, +1 otherwise, and $E = -\sin(A)\cos(\phi) + U\cos(A)\sin(\phi)$. The parallel lattice constant of the epilayer is calculated from $\Delta\phi$ by:

$$a_{\parallel} = \frac{a_{\perp} \cdot \tan(\phi)}{\tan(\phi + \Delta\phi)} \qquad (5)$$

Note that finding $\Delta\phi$ also will predict the $\Delta\omega_{ge\ (or\ gi)}$ peak splitting (whichever was not determined in the initial measurement). The misfit lattice constant (i.e. the lattice constant if the layer is fully relaxed) is calculated from the Poisson's ratio (ν) of the epilayer:

$$a_r = \frac{-\left(a_{\perp}(\nu - 1) - 2\nu a_{\parallel}\right)}{1 + \nu} \quad , \quad \%\ \text{Relaxed} = \frac{a_{\parallel} - a_s}{a_r - a_s} \cdot 100\% \qquad (6)$$

The composition is obtained through the misfit lattice constant from Vegard's Law (or a more appropriate polynomial equation if applicable). Since the Poisson ratio is composition dependent, the calculated composition should then be used to recalculate a new Poisson's ratio, from which a new a_r is iterated. We have found that only one iteration is needed with a reasonable initial guess value ($0 < x < 1$) for the composition. Finally, the percent relaxation is defined in Eqn. (6).

Alternatively, an iterative method can be used to find the roots of (1) or (4). We have utilized the secant method and have found that any reasonable guess value $\Delta\phi_o$ (we use ($\Delta\phi$ = ($\Delta\Theta_{004,ge}$-$\Delta\omega_{gi,004}$) will suffice. Then $\Delta\phi_1$ is set equal to $\Delta\phi_o + TOL$, where TOL is the tolerance limit (~ 1 x 10^{-6}). The converging sequence described by the iteration formula

$$\Delta\phi_{k+1} = \Delta\phi_k - \frac{\Delta\phi_k - \Delta\phi_{k-1}}{f(\Delta\phi_k) - f(\Delta\phi_{k-1})} \cdot f(\Delta\phi_k) \tag{7}$$

which is repeated until $|f(\Delta\omega_k)| < TOL$. A second iteration involving Poisson's ratio will give the final values for relaxation and composition.

Asymmetric diffraction planes and relaxation

It is important to use a diffraction peak that is most sensitive to the effects of relaxation.[7] We define the change in $\Delta\omega_{(ge\ or\ gi)}$ versus the amount of relaxation as the gauge for relaxation sensitivity. Fig 2(a, b) show the relative change in $\Delta\omega_{gi}$ and $\Delta\omega_{ge}$ respectively, between a fully strained and fully relaxed epilayer for various diffraction planes. The calculation was that of a misfit $In_{0.1}Ga_{0.9}As$ layer on a GaAs substrate. We found the same basic trends for other material systems as well. For glancing incident, the (026) shows the greatest sensitivity, followed by the (115) and the (224). The $(115)_{gi}$ is especially convenient, since the samples can be easily aligned using the cleavage planes and/or wafer flats. Furthermore, the incident x-ray angle is similar to that of the (004) peak, so that one samples nearly the same volume in both cases. For glancing exit, the (353), (444), (044) and the (224) planes are most sensitive to relaxation. Of these, the $(224)_{ge}$ (hinged on a [110]) or $(044)_{ge}$ (hinged on [001]) are most easily aligned.

Fig. 2(c) shows the product the relative relaxation sensitivities of Figs. 2(a) and 2(b). This provides a figure of merit for the various diffraction planes when both glancing exit and glancing incident scans are used. In this case, the (026) will inherently be the most sensitive, followed by the (044), (335) and (224) diffraction planes. However, the (044) and (224) planes are an all-around good choice, due to ease of alignment.

In practice, it is hard to achieve much better than ~5-10 arc sec accuracy in the position of the epilayer peak., especially when the peaks are broadened by mosaic tilt and diffuse scatter from misfit and edge dislocations. Even the most careful measurements have some uncertainty.

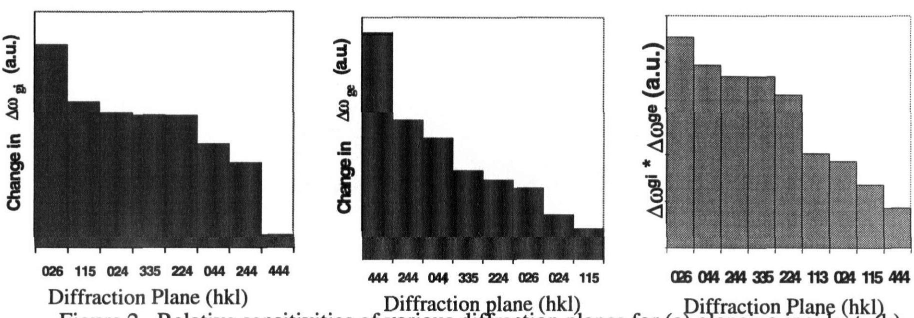

Figure 2. Relative sensitivities of various diffraction planes for (a) glancing incident, (b) glancing exit, and (c) the product of glancing incident and exit.

These effects are quantified in Fig. 3, which shows the calculated percent relaxation with a shift in epilayer peak position of +15 arc sec, for several diffraction planes . Chart (a) in each graph represents the case when a glancing incident scan is used, while curve (b) is that from the glancing exit (both combined with a symmetric scan). The calculated was done with a 50% relaxed $In_{0.1}Ga_{0.9}As/GaAs$ structure. The results are rather dramatic. For instance an uncertainty of 15 arc sec in the epilayer peak position will be an uncertainty of ~5% in the calculated percent relaxation for the combination of a symmetric and the $(115)_{gi}$, $(224)_{gi}$, and $(026)_{gi}$ and $(044)_{gi}$ diffraction planes. The error can be reduced by using a small slit which reduces broadening due to mosaicity.

In contrast, Fig. 3(b) shows that the combination of a symmetric and glancing exit diffraction planes are generally less susceptible to experimental error (adding 15 arc sec to the epilayer peak position has a reduced effect on the calculated percent relaxation). The case of utilizing both the glancing exit and incident scans (not shown) is nearly identical to Fig. 3(b). Thus the combination of a glancing exit and symmetric scan will lower the experimental uncertainty, while maintaining the convenience of using only one asymmetric scan.

As a second consideration, it should be noted that the $\Delta\phi$ term in Eqn. (1) will greatly effect the Bragg conditions for the epilayer. For instance, a $(224)_{ge}$ diffraction scan of a fully strained $In_{0.1}Ga_{0.9}As$ layer on GaAs, requires a $\Theta/10\Theta$ scan (as opposed to the customary $\Theta/2\Theta$ scan) to go through the center of both the substrate and layer peaks in reciprocal space.[8] A $\Theta/2\Theta$ scan will include both peaks only if the detector aperture is wide enough. However, a wide detector aperture decreases the signal-to-noise ratio. Only symmetric peaks (barring severe substrate misorientation) or asymmetric diffraction planes of *completely relaxed* epilayers will have a sample/detector angular scanning ratio of exactly $\Theta/2\Theta$. Thus one must be careful in using a detector slit for most glancing exit diffraction planes.

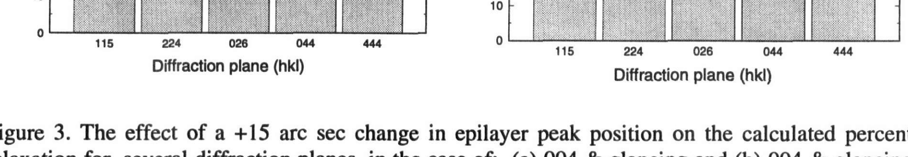

Figure 3. The effect of a +15 arc sec change in epilayer peak position on the calculated percent relaxation for, several diffraction planes. in the case of: (a) 004 & glancing and (b) 004 & glancing exit. Without shifting the peak position, the calculated value is 50%. A large deviation from 50% represents a diffraction geometry susceptible to experimental error.

In contrast, for $(224)_{gi}$ peak, a $\Theta/1\Theta$ scan is required for a fully strained layer. Thus, the range of uncertainty for the correct $\Theta/x\Theta$ scan ratio is reduced, allowing the use of a smaller detector slit. The $(115)_{gi}$ diffraction plane is very ideal in this sense, requiring a $\Theta/1.6\Theta$ scan for a fully strained layer. Therefore, one can use a narrow a detector slit with a $\Theta/2\Theta$ scan, without blocking diffraction from the layer. This will reduce the experimental uncertainty, so that one can avoid the problems in the glancing incident geometry shown in Fig. 3(a).

CONCLUSION

In summary, we introduce for the first time, a correct format for determining composition and relaxation in heterostructures using a symmetric and one asymmetric X-ray diffraction scan. It is also shown that using small angle approximations or low order Taylor series approximations can produce significant errors. To assure correctness for all geometries, the equations given in this text, or an iterative method, should be used.

We also show that uncertainties in epilayer peak position (due primarily to peak broadening) can produce considerable error when the combination of a symmetric and glancing incident scan is used. This error can be reduced in the $(115)_{gi}$ diffraction plane by using a detector slit and increasing the scan time across the epilayer peak. In general, the glancing exit geometry is less susceptible to experimental error.

We also show that the $\Delta\phi$ term can have a large effect on the positions of the peaks in reciprocal space, and will change the exact sample/detector angular scanning ratio to something other than $\Theta/2\Theta$. This effect is especially dramatic in glancing exit geometry. Since the $(026)_{ge}$ and $(115)_{gi}$ diffraction planes have a very low deviation from $\Theta/2\Theta$ with increasing strain, these planes are highly suitable for measuring structures whose relaxation is unknown.

In summary, epilayer composition and relaxation can be measured with the combination of a symmetric and any asymmetric diffraction scan. For highest accuracy, the combination of a symmetric and glancing exit asymmetric diffraction scan is recommended, for instance the (004) and $(224)_{ge}$ or $(044)_{ge}$ diffraction planes. When speed and convenience are a priority, the combination of the (004) and $(115)_{gi}$ (with a rotating detector and detector slit) is recommended.

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