Some Aspects of the X-ray Structural Characterization of 
(Ga_{1-x}Al_{x}As)n_{1}(GaAs)n_{2}/GaAs(001) Superlattices

BY J. KERVAREC, M. BAUDET, J. CAULET, P. AUVRAY, J. Y. EMERY AND A. REGRENY

Département Matériaux–Physique–Analyse, Centre National d'Etudes des Télécommunications, 22301 Lannion, France

(Received 27 October 1983; accepted 10 February 1984)

Abstract

The study of the physical properties of the superlattices (Ga_{1-x}Al_{x}As)n_{1}(GaAs)n_{2}/GaAs(001) requires the knowledge of their structural parameters n_{1}, n_{2} and x and of their crystalline state. X-ray diffraction enables the existence of such artificial superstructures, obtained by molecular-beam epitaxy, to be revealed and their characteristic parameters to be determined. Two experimental techniques are used: a standard powder goniometer and a double-crystal diffractometer. The diffraction diagrams directly yield the superperiod and the average Al concentration in the superlattice. The value of x is determined by refinement between observed and calculated structure factors. The results are even more accurate when the number of observed satellite peaks for a given periodicity is greater; this number depends at the same time on the Al composition x, the n_{1}/n_{2} ratio, the periodicity and its dispersion and the shape of the interfaces. This method is illustrated by a few examples with superlattice periods varying from 51 to 397 Å. The consequence on the X-ray diagrams of various defects (dispersion in n_{1} and n_{2}, superperiod gradient, Al diffusion) is discussed.

Introduction

The semi-conductor-based superlattices (SL) are new materials, the first ones having been built in 1970 (Esaki & Tsu, 1970), grown by epitaxy on a semiconductor. They are formed by the repeated growth of a pattern made of two layers of semi-conductors of different thicknesses and compositions. The superperiod obtained in this way and the nature of the two adjoining semi-conductors lead to interesting properties, particularly in optics and electrical transport, which should find applications in such fields as lasers and hyperfrequency (Palmier, 1981).

Most SLs are presently grown by molecular beam epitaxy (MBE) (Chang, Esaki, Howard & Ludeke, 1973): this method allows the thicknesses and compositions of the deposited materials to be fairly well mastered. The values of these parameters which govern the SL properties are then controlled by various methods.

X-ray and electron diffraction methods are successfully used to reveal the presence of these artificial superstructures and to determine their period and other characteristic parameters. X-ray diffraction has the advantage, however, of neither damaging the sample nor requiring any preparation of the samples.

The properties of (AlAs)n_{1}(GaAs)n_{2}/GaAs(001) type SLs (where n_{1} and n_{2} are the numbers of molecular layers of each material in a SL unit cell) have been much investigated. Two important publications have been devoted to their characterization by means of X-rays: one (Segmüller, Krishna & Esaki, 1977) dealing with the comparison of observed and calculated intensities for two SLs and the other (Fleming, McWhan, Gossard, Wiegmann & Logan, 1980) with the study of AlAs diffusion when annealed.

The (Ga_{1-x}Al_{x}As)n_{1}(GaAs)n_{2}/GaAs(001) type SLs have also been studied to a great extent but for their characterization other methods have been used: interferometry to measure thicknesses, and interband absorption-edge measurements for the determination of Al content (Dingle, Gossard & Wiegmann, 1975).

However, X-ray diffraction data contain all the information needed to determine these structural parameters since diffraction angles are a function of the lattice period and diffracted intensities depend on the nature and position of the atoms in the unit cell. In the following paragraphs we will show how the crystalline structure of (Ga_{1-x}Al_{x}As)n_{1}(GaAs)n_{2}/GaAs(001) type SLs can be determined from these data. The first part of this paper will be devoted to the description of experimental techniques, the second to the determination of structural unknowns and the third to some typical examples. In conclusion we will discuss the limits of this characterization.

Choice of experimental techniques

In order to obtain a maximum of experimental data, diffraction techniques must be carefully chosen. The diffraction diagram along (00l) of a SL is adequate for the determination of the structural unknowns. It is characterized by maxima which are often of slight intensity described as 'satellites' and situated more or less symmetrically in relation to maxima of greater
intensity close to the substrate peaks. It is noticeable that these more intense peaks coincide with those of an imaginary structure whose crystalline parameter in the [001] direction would be the average parameter of the unit cells which construct the SL; in fact, if the unit cell of the SL is composed of $n_1$ molecular layers of Ga$_{1-x}$Al$_x$As of parameter $c_1$ and of $n_2$ molecular layers of GaAs of parameter $c_2$ in the direction of growth (Fig. 1) the period of this SL is $C = (n_1c_1 + n_2c_2)/2$ and the average period of the unit cells of Ga$_{1-x}$Al$_x$As and of GaAs is $\bar{c} = (n_1c_1 + n_2c_2)/(n_1 + n_2)$.

The reciprocal-lattice points 001 of this lattice $\bar{c}$ are close to the reciprocal-lattice points 001 of the GaAs substrate since $c_1 \sim c_2 = c_{GaAs}$; moreover, these points coincide with the reciprocal-lattice points 00$L = l(n_1 + n_2)/2$ of the superlattice. Subsequently these points will be called average lattice points.

As these peaks and the 001 peaks of the substrate are very close, the use of an apparatus which does not enlarge the diffraction profiles is essential, in order to separate them. A double-crystal diffractometer (Compton & Allison, 1949) in quasi-parallel setting serves this purpose. The apparatus we use (Fig. 2) is equipped with a Ge(001) monochromator which enables the recording of the diffraction diagram for the superlattice in the vicinity of the 004 reflection of GaAs. Sample rotation is operated by a stepping motor (elementary step = 1/4°), and computer controlled. The estimated error in the measurement of angular differences is of the order of 1° which corresponds to a relative error $Ad/d$ of $7 \times 10^{-6}$.

This assembly enables the recording of several diffraction peaks of the SL: the peak of indices 00$L = 2(n_1 + n_2)$ close to the 004 peak of the substrate and its satellites. As the beam impact on the sample is limited by a circular diaphragm, analysis is localized and the homogeneity of the sample can be verified. Most of the other 00$L$ reflections of the SL cannot be recorded with this assembly either because geometrical conditions are not favourable (absence of Ge reflections with indices 00$l \neq 4n$), or because their intensities are too weak (from ten to several thousand or so times weaker than those of the average lattice): in this case it is necessary to favour intensity at the expense of resolution in order to detect them.

To do this we use a classical powder goniometer functioning in $\theta$–$2\theta$ coupling and equipped with a post-sample curved quartz monochromator. As the X-ray beam has the dimensions 0.1 x 8 mm when it reaches the sample it can be considered that the recordings obtained correspond to an average along a whole line of the sample.

In practice the 00$L$ peaks close to $\theta = 0$ and to the 001 reflections of GaAs are recorded; in this way it can be established that it is in the vicinity of the 002 GaAs reflection that most of the satellites are detected; the positions and intensities of all peaks are measured.

**Determination of the structural parameters**

The classical model (Segmüller et al., 1977) chosen to describe the structure of a SL fixes the number of parameters to be determined. However, it is good to keep in mind the fact that, whatever this model, the results obtained correspond to an average in volume on the part of the sample irradiated, and that this average can vary from one sample area to another (Segmüller et al., 1977; Fleming et al., 1980).

In this model the unit cell of the SL is composed of $n_1$ molecular layers of Ga$_{1-x}$Al$_x$As followed by $n_2$ molecular layers of GaAs. The aluminium concent-

---

**Fig. 1.** Diagram of the unit cell in a (Ga$_{1-x}$Al$_x$As)$_{n_1}$(GaAs)$_{n_2}$/GaAs(001) superlattice.

**Fig. 2.** Geometrical characteristics of the X-ray double-diffraction set-up. S: X-ray source (0.4 x 0.8 mm², point focus). Collimator consisting of two diaphragms D1 and D2, 130 mm apart and having 0.3 mm diameter. $SD_2 = 190$ mm. O1: Crystal to be analysed. $O_1D_3 = 560$ mm. O2: Monochromator crystal. $SO_1 = 230$ mm. D3: Diaphragm limiting the size of the beam impact on the sample. $O_1D_3 = 620$ mm.

---
tration is a periodic step function, i.e., equal to \(x\) in the ternary molecular layers and to zero in those of GaAs. The \(2d_1\) thickness of a molecular layer of Ga\(_{1-x}\)Al\(_x\)As is the same as that of a heteroepitaxy of Ga\(_{1-x}\)Al\(_x\)As epitaxied under elastic stresses on a GaAs substrate. The \(2d_2\) thickness of a molecular layer of GaAs, in the same way, is the same as that of a molecular layer of GaAs in the bulk material. In these conditions the structural unknowns are \(n_1\), \(n_2\) and \(x\).

These unknowns are not obtained directly. The \(C\) period of the SL is first of all determined \((C=2n_1d_1+2n_2d_2)\) then \(\bar{x}\), average value of \(x\) in a SL unit cell \([\bar{x}=x(n_1+n_2)/(n_1+n_2)]\), and finally \(x\): the values of \(n_1\) and \(n_2\) resulting from it.

(a) Determination of \(C\)

The \(C\) parameter of the SL is linked to the angles of 00\(L\) reflections by Bragg’s law, \(2(C/L)\sin \theta(00L)=\lambda\). As the \(L\) indices of these reflections are not known a priori, they are determined from the Bragg angles of two reflections whose differences in indices are known: if the index \(L\) is given to the SL peak close to the 002 reflection of the substrate, \(L\) is calculated from the reflection angles of the satellites of indices \(L-i\) and \(L+j\) by

\[
\frac{d_{L-i}-d_{L+j}}{d_{L+j}} = \frac{i+j}{L-i}
\]

hence

\[
L = \frac{d_{L+j}}{d_{L-i}-d_{L+j}}(i+j) + i,
\]

from which, with the approximation \(\Delta d_{L-i} \sim \Delta d_{L+j}\) \(\sim \Delta d_L\), one can easily derive

\[
\frac{\Delta L}{L} = \frac{\Delta \theta_L}{\tan \theta_L} \frac{2L}{i+j}.
\]

Determination of \(L\) is more precise when \((i+j)\) is large and when \(L\) is small; Fig. 3 shows, for instance, that when \(i+j = 2\), for a given value of \(\Delta \theta_L/\tan \theta_L\), the error in \(L\) is greater than 1 for \(L > 60\) and greater than 2 for \(L > 80\). This is why extreme satellites of the 002 reflexion of the average lattice are used to determine \(L\): they are observed using the powder goniometer. The SL peaks are indexed by assigning to \(L\) the integer value closest to the calculated value.

The \(C\) parameter of the SL is obtained by using the value of \(L\) deduced from (1): \(C = Ld(00L)\). It is noticed that the error in \(C\) due to the angular difference of the average lattice of the substrate and the average lattice. In fact, this difference corresponds to a difference in reticular distance \(\Delta d\):

\[
\Delta d = d_{SL} - d_{sub} = (\bar{c} - c_{GaAs})/4
\]

\[
= \frac{n_1c_1 + n_2c_2 - (n_1 + n_2)c_2}{4(n_1 + n_2)} = \frac{n_1(c_1 - c_2)}{4(n_1 + n_2)}
\]

However, \(c_1 - c_2\), which is the parameter difference between a coherent epitaxy of Ga\(_{1-x}\)Al\(_x\)As and its substrate of GaAs, is proportional to \(x\): \(c_1 - c_2 = k_0x\) (Vegard’s law). Hence \(\Delta d = (k_0/4)[n_1/(n_1 + n_2)]x = (k_0/4)\bar{x}\) since \(\bar{x} = x(n_1 + n_2)/(n_1 + n_2)\); and as the corresponding angular difference, \(\Delta \theta\), is very small (less than 0.1°) it can be said that it is proportional to \(\Delta d\) and therefore to \(\bar{x}\).

\[
\Delta \theta = k_1\bar{x}
\]

with \(k_1 = 375°\) as determined experimentally (Baudet et al., 1983). For an estimated error of 1° on the measurement of this difference, the error in \(x\) is less than 0.003.

We must point out that \(k_0\) is the actual difference between the unit cells \(c_1 = 4d_1\) and \(c_2 = 4d_2\) perpendicular to the interface of an epitaxial layer of pure AlAs and a GaAs substrate; \(k_0\) is related to the unstrained parameters \(a_1\) and \(a_2\) and to the elastic constants \(c_{11}\) and \(c_{12}\) of AlAs by \(k_0 = (a_1 - a_2)\times(1 + 2c_{12}/c_{11}) = 1.8(a_1 - a_2)\) (Segmüller et al., 1977), which leads to a value of 377° for \(k_1\) in good agreement with the experimental value of 375°.

(c) Determination of \(x\)

The last structural unknown is determined by making adjustments between the modules of the calculated and observed structure factors \(F_c(00L)\) and \(F_o(00L)\) respectively, the latter being deduced from intensities

![Fig. 3. Variation of \(\Delta L\) (error in the index of the 00L reflection) as a function of \(L\) for various values of \(i+j\), with \(\Delta \theta/\tan \theta = 3.2 \times 10^{-4}\) [see (2)].](attachment:image.png)
$I_o(00L)$ measured on powder goniometer recordings: owing to the imperfections in the SLs and in the post-sample curved monochromator, $F_o(00L)$ and $I_o(00L)$ are linked by the relation

$$I_o(00L) \propto |F_o(00L)|^2 P \Lambda T$$  \hspace{0.5cm} (3)

with $L$, Lorentz's factor = $1/\sin 2\theta(00L)$, $P$, polarisation factor = $[1 + \cos^2 \theta_m \cos^2 2\theta(00L)]/2$, where $\theta_m$ and $\theta(00L)$ are respectively the angles of incidence on the reflecting planes of the monochromator and of the sample. $A$ is the absorption factor in the SL,

$$A = \left\{ \mu_{SL} \left[ 1 + \frac{\sin(\theta - \epsilon)}{\sin(\theta + \epsilon)} \right] \right\}^{-1} \times \left( 1 - \exp \left\{ -t_{SL} \mu_{SL} \left[ \frac{1}{\sin(\theta - \epsilon)} + \frac{1}{\sin(\theta + \epsilon)} \right] \right\} \right)$$

where $t_{SL}$ is the thickness of the SL, $\mu_{SL}$ its linear absorption coefficient (equal to that of the material of the same average composition where the Al and Ga atoms are randomly distributed) and $\epsilon$, the angle of the lattice planes with the surface of the sample. $A_e$ is the absorption factor in the encapsulation layer,

$$A_e = \exp \left\{ -t_e \mu_e \left[ \frac{1}{\sin(\theta - \epsilon)} + \frac{1}{\sin(\theta + \epsilon)} \right] \right\}$$

where $t_e$ is the thickness of the encapsulation, $\mu_e$ its linear absorption coefficient. $T$ is the thermal agitation factor, $T = \exp[-2B \sin^2 \theta(00L)/\lambda^2]$, where $B$ is the coefficient of thermal agitation assumed to be equal to that of GaAs at room temperature, $B = 0.595$ Å² (Matsushita & Hayashi, 1977).

For the model described above, the general expression for the structure factor of the 00L reflection of the SL is

$$F_o(00L) = \sum_{j=1}^{N} f_j \exp 2\pi Lz_j$$

where $N$ is the total number of atoms in the SL unit cell, and where $f_j$ and $z_j$ are respectively the atomic scattering factor and fractional coordinate of the $j$th atom. Developed in sums of geometric series this is simplified:

$$F_o(00L) = 2(f_{As} + f_{GaAl} R_1^{1/2}) (R_1^{1/2} - 1) / R_1 - 1$$

$$+ 2(f_{As} + f_{Ga} R_1^{1/2}) R_1^{1/2} R_2^{1/2} (R_2^{1/2} - 1) / R_2 - 1$$

where

$$R_1 = \exp(2\pi i L d_1/C); \quad R_2 = \exp(2\pi i L d_2/C)$$

and

$$f_{GaAl} = x f_{Al} + (1 - x) f_{Ga}.$$

The adjustment between $F_o(00L)$ and $F_c(00L)$ is computed by a method of minimum. The $F_c(00L)$ are calculated for values of $x$ (therefore of $n_1$ and $n_2$) taken between two extremes fixed from growth conditions; when $x$ converges towards its value in the SL, the agreement between the calculated and observed structure factors becomes optimum. This method of adjustment has been verified on a SL encapsulated by an epitaxy of Ga$_{1-x}$Al$_x$As whose directly measurable composition $x$ was precisely that of the subjacent SL. The calculations show that the relative variation of the $F_c(00L)$ with $x$ is very clear. The example of Fig. 4 shows that the intensities of satellites rapidly vary and are non-existent in the vicinity of specific values of $n_1$ and $n_2$ (therefore of $x$): thus for $n_1 = n_2 = 15$ the satellites of±2 order associated with the 002 reflection of the average lattice are absent. In the same way, the satellites of±3 order disappear for $n_1 = 20 = 2n_2$, and $n_1 = 10 = n_2/2$. More generally it is demonstrated that the intensities of satellites of±ith order are systematically non-existent for $i$ values given by the expression $i = k(1 + n_1/n_2)$, where $k$ is an integer.

**Examples of application**

The SLs that we describe have been obtained by MBE on substrates of laser type GaAs, containing less than $10^2$ dislocations cm$^{-2}$. The conditions of growth are: residual vacuum 13·3 nPa; As pressure 0·13 mPa; temperature of the substrate $\sim 870$ K; speed of growth 2 Å s$^{-1}$.

Fig. 4. Variation of the calculated intensity $I_{calc}$ of the first satellites of the 002 average lattice reflection of a superlattice with given $x$ and $n_1 + n_2$, as a function of $n_1$, or $x$. 

100. 6 10 15 20 25 30 $n_1$

1,00 0,60 0,40 0,30 0,24 0,20 $x$

1,00 0,60 0,40 0,30 0,24 0,20 $x$
Opening control of the aluminium oven is automatic for examples 1 and 3 and manual for example 2.

Example 1

This SL is composed of 50 unit cells. Its X-ray diffraction diagram in the vicinity of the 002 and 004 GaAs reflections (Fig. 5a) shows:

1. a great number of satellites which enable \( n_1 + n_2 \) and the C period to be determined with accuracy: \( n_1 + n_2 = 140.5 \pm 0.5 \) and \( C = 397.4 \pm 1.5 \) Å. The fact that \( n_1 + n_2 \) is not an integer reveals, as will be seen in the discussion, that the SL unit cells are not all identical: the X-ray diagram leads to the average value of \( n_1 + n_2 \) in the sample zone explored. For calculation of structure factors, the one or two closest integer values are used, here \( n_1 + n_2 = 140 \) and 141;

2. the absence, apart from one exception, of even-order satellites indicating that \( n_1 \) and \( n_2 \) are similar;

3. peaks whose width at half maximum is small which means, as will be seen further on, good repetitiveness of the unit cells in the thickness of the SL.

The diagrams obtained by X-ray double diffraction (XDD) in the vicinity of the 004 GaAs reflection (Fig. 5b) give \( x = 0.179 \pm 0.003 \) in various points of the sample. The \( x \) values obtained by refinement giving the SL indices 0,0,140 then 0,0,141 to the 002 reflection of the average lattice are identical: \( x = 0.345 \), which leads to

\[
\begin{align*}
n_1 + n_2 &= 140 \quad \left\{ \begin{array}{l}
n_1 = 71 \\
n_2 = 69
\end{array} \right. \quad \text{from which} \quad \left\{ \begin{array}{l}
e_1 = 206.5 \text{ Å} \\
e_2 = 189.4 \text{ Å}
\end{array} \right. \\
n_1 + n_2 &= 141 \quad \left\{ \begin{array}{l}
n_1 = 71 \\
n_2 = 70
\end{array} \right. \quad \text{from which} \quad \left\{ \begin{array}{l}
e_1 = 206.5 \text{ Å} \\
e_2 = 192.2 \text{ Å}
\end{array} \right.
\end{align*}
\]

with \( e_1 = 2n_1d_1 \) and \( e_2 = 2n_2d_2 \).

The values of calculated and observed structure factors are given in Table 1. It must be noted that the diffraction peaks which are not observed have a great influence on the \( x \) value obtained.

Example 2

This SL is made from 120 unit cells and is overlaid by an encapsulation layer of around 0.3 µm.

As before, the X-ray diagrams in the vicinity of the 002 and 004 GaAs reflections (Figs. 6a and b) lead to: \( n_1 + n_2 = 40 \pm 0.1 \), from which \( C = 113.1 \pm 0.3 \) Å and \( x = 0.3 \pm 0.003 \) at various points of the sample. The adjustment between calculated and observed structure factors gives \( x = 0.48 \) from which \( n_1 = 25 \), \( n_2 = 15 \), \( e_1 = 70.7 \) Å and \( e_2 = 42.4 \) Å. Table 2 gives the values of calculated and observed structure factors.

It is noticeable that the XDD diagram (Fig. 6b) presents an oscillating phenomenon between the substrate peak and that of the average lattice, whose period \( \omega \) is linked to the total thickness of the SL by

\[
t = \lambda /2\omega \cos \theta
\]

(Bartels & Nijman, 1978).

The beat period \( \omega \) is \( 13 \pm 1'' \), which leads to a calculated thickness \( t \) of 1.45 µm which is in good agreement with the real thickness of the SL (1.38 µm). This kind of phenomenon is frequently observed; when the sample is made of several epitaxial layers (the encapsulating layer, the SL itself and a buffer layer) each of them leads to a fringe system and their superimposition can mask, when the total thickness of the SL is slight, the peak coinciding with the 004 reflexion of the average lattice which prevents the determination of \( x \) and that of the structure.

Example 3

This consists of a SL of 200 non-encapsulated unit cells. The X-ray diagrams in the vicinity of the 000 and 002 GaAs reflections (Figs. 7a and b) give \( n_1 + n_2 = 18.2 \pm 0.14 \), \( C = 51.4 \pm 0.4 \) Å and \( x = 0.144 \pm 0.003 \).
Table 1. Observed and calculated structure factors in the vicinity of the 002 and 004 reflections of the average lattice for the refined values (example 1)

(a) $x = 0.345$, $n_1 = 71$ and $n_2 = 69$;
(b) $x = 0.345$, $n_1 = 71$ and $n_2 = 70$.

<table>
<thead>
<tr>
<th>Satellite order</th>
<th>H</th>
<th>K</th>
<th>L</th>
<th>$F_o$</th>
<th>$F_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(-9)</td>
<td>0</td>
<td>0</td>
<td>131</td>
<td>0.72</td>
<td>0.75</td>
</tr>
<tr>
<td>(-8)</td>
<td>0</td>
<td>0</td>
<td>132</td>
<td>non-obs.</td>
<td>0.06</td>
</tr>
<tr>
<td>(-7)</td>
<td>0</td>
<td>0</td>
<td>133</td>
<td>0.96</td>
<td>0.97</td>
</tr>
<tr>
<td>(-6)</td>
<td>0</td>
<td>0</td>
<td>134</td>
<td>non-obs.</td>
<td>0.03</td>
</tr>
<tr>
<td>(-5)</td>
<td>0</td>
<td>0</td>
<td>135</td>
<td>1.30</td>
<td>1.36</td>
</tr>
<tr>
<td>(-4)</td>
<td>0</td>
<td>0</td>
<td>136</td>
<td>non-obs.</td>
<td>0.03</td>
</tr>
<tr>
<td>(-3)</td>
<td>0</td>
<td>0</td>
<td>137</td>
<td>2.57</td>
<td>2.28</td>
</tr>
<tr>
<td>(-2)</td>
<td>0</td>
<td>0</td>
<td>138</td>
<td>non-obs.</td>
<td>0.22</td>
</tr>
<tr>
<td>(-1)</td>
<td>0</td>
<td>0</td>
<td>139</td>
<td>8.01</td>
<td>7.34</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>140</td>
<td>(+002 GaAs)</td>
<td>17.29</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>141</td>
<td>6.29</td>
<td>5.78</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>142</td>
<td>0.47</td>
<td>0.48</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>0</td>
<td>143</td>
<td>2.10</td>
<td>2.06</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>0</td>
<td>144</td>
<td>non-obs.</td>
<td>0.32</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>0</td>
<td>145</td>
<td>1.37</td>
<td>1.24</td>
</tr>
<tr>
<td>6</td>
<td>0</td>
<td>0</td>
<td>146</td>
<td>non-obs.</td>
<td>0.26</td>
</tr>
<tr>
<td>7</td>
<td>0</td>
<td>0</td>
<td>147</td>
<td>0.81</td>
<td>0.88</td>
</tr>
<tr>
<td>8</td>
<td>0</td>
<td>0</td>
<td>148</td>
<td>non-obs.</td>
<td>0.23</td>
</tr>
<tr>
<td>9</td>
<td>0</td>
<td>0</td>
<td>149</td>
<td>0.54</td>
<td>0.68</td>
</tr>
<tr>
<td>(-3)</td>
<td>0</td>
<td>0</td>
<td>277</td>
<td>non-obs.</td>
<td>0.22</td>
</tr>
<tr>
<td>(-2)</td>
<td>0</td>
<td>0</td>
<td>278</td>
<td>non-obs.</td>
<td>0.13</td>
</tr>
<tr>
<td>(-1)</td>
<td>0</td>
<td>0</td>
<td>279</td>
<td>8.72</td>
<td>7.89</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>280</td>
<td>(+004 GaAs)</td>
<td>143.54</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>281</td>
<td>18.30</td>
<td>17.64</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>282</td>
<td>non-obs.</td>
<td>1.49</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>0</td>
<td>283</td>
<td>2.56</td>
<td>2.94</td>
</tr>
</tbody>
</table>

The refinement has been carried out by means of the 001, 002 and 003 SL reflections, the only two satellites $-1$ and $+1$ observed in the vicinity of the 002 reflection of the average lattice not being sufficient for the refinement because their intensities vary in the same way with $n_1$ (see Fig. 4). This leads to $x = 0.29$, $n_1 = 9$ and $n_2 = 9$. The complete results are given in Table 3.

The X-ray diagrams obtained at other points of the sample lead to different values of $n_1 + n_2$ and reveal an inhomogeneity which has been frequently observed (Segmüller et al., 1977; Fleming et al., 1980).

**Discussion**

In the preceding examples (as in most of the SLs that we have characterized) it can be considered that the agreement between the observed and calculated X-ray diagrams is satisfactory. However, there are several anomalies: in example 1, one cannot approximate the oscillating phenomenon occurring between the substrate peak and the average lattice has a period $\omega = 13^\circ$. 

![Fig. 6. Second sample: 120 unit cells ($n_1 = 25$, $n_2 = 15$, $x = 0.480$) with a 0.3 μm GaAs encapsulation layer. (a) X-ray diffraction diagrams in the vicinity of the 002 and 004 GaAs reflections. (b) X-ray double-diffraction diagram in the vicinity of the 004 GaAs reflection. The oscillating phenomenon occurring between the substrate peak and the average lattice has a period $\omega = 13^\circ$.](image)
Table 2. Observed and calculated structure factors in the vicinity of the 002 and 004 reflections of the average lattice for the refined x values (example 2)

\[ x = 0.475, n_1 = 25 \text{ and } n_2 = 15 \]

<table>
<thead>
<tr>
<th>Satellite order</th>
<th>H</th>
<th>K</th>
<th>L</th>
<th>( F_o )</th>
<th>( F_c )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(-4)</td>
<td>0</td>
<td>0</td>
<td>36</td>
<td>1.12</td>
<td>2.30</td>
</tr>
<tr>
<td>(-3)</td>
<td>0</td>
<td>0</td>
<td>37</td>
<td>0.61</td>
<td>1.27</td>
</tr>
<tr>
<td>(-2)</td>
<td>0</td>
<td>0</td>
<td>38</td>
<td>2.86</td>
<td>3.09</td>
</tr>
<tr>
<td>(-1)</td>
<td>0</td>
<td>0</td>
<td>39</td>
<td>8.64</td>
<td>9.57</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>40</td>
<td>(+002 GaAs)</td>
<td>24.11</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>41</td>
<td>7.51</td>
<td>7.67</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>42</td>
<td>2.78</td>
<td>3.15</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>0</td>
<td>43</td>
<td>non-obs.</td>
<td>0.98</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>0</td>
<td>44</td>
<td>1.08</td>
<td>2.16</td>
</tr>
<tr>
<td>(-2)</td>
<td>0</td>
<td>0</td>
<td>78</td>
<td>1.23</td>
<td>1.39</td>
</tr>
<tr>
<td>(-1)</td>
<td>0</td>
<td>0</td>
<td>79</td>
<td>non-obs.</td>
<td>1.76</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>80</td>
<td>(+004 GaAs)</td>
<td>13.92</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>81</td>
<td>10.34</td>
<td>10.53</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>82</td>
<td>3.42</td>
<td>3.23</td>
</tr>
</tbody>
</table>

Table 3. Observed and calculated structure factors in the vicinity of the 000 and 002 reflections of the average lattice for the refined x value (example 3)

\[ x = 0.29, n_1 = 9 \text{ and } n_2 = 9 \]

<table>
<thead>
<tr>
<th>Satellite order</th>
<th>H</th>
<th>K</th>
<th>L</th>
<th>( F_o )</th>
<th>( F_c )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(-2)</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>6.62</td>
<td>6.06</td>
</tr>
<tr>
<td>(-1)</td>
<td>0</td>
<td>0</td>
<td>15</td>
<td>non-obs.</td>
<td>0.00</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>16</td>
<td>(+002 GaAs)</td>
<td>15.66</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>17</td>
<td>5.39</td>
<td>5.48</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>18</td>
<td>non-obs.</td>
<td>0.03</td>
</tr>
</tbody>
</table>

The value of \( n_1 + n_2 \) to a single integer. It is established on the other hand that the satellite peaks become broader as they are further from the average peak (Figs. 5a and 6a) and that the agreement between \( F_o \) and \( F_c \) is much better as the SL period becomes larger.

These disagreements are a supplementary source of information. In fact, it can be expected that the real structure of a SL is different from the theoretical model used for the following reasons:

1. The growth of the SL is not uniform on the whole surface of the substrate as a result, for example, of the arrangement of ovens in the growth chamber or because the temperature of the substrate is not strictly uniform.

2. As all the atoms which constitute a molecular layer are not deposited simultaneously the interfaces between GaAs and Ga\(_{1-x}\)Al\(_x\)As are not planar. It allows that the crystalline unit cells of the SL are not identical (Segmüller et al., 1977). If it is admitted that the defects of surface evenness do not exceed the thickness of a molecular layer, the number of molecular layers in a SL unit cell can vary at most by ± 1.

The non-uniformity of the SL in the epitaxial plane can be understood by a more or less continuous variation of the diffraction diagrams as a function of the sample zone analysed, and the existence on certain samples of a gradient leading to a period variation of 1 to 2 molecular layers cm\(^{-1}\) has been revealed.

The uneven surfaces of the interfaces can be taken into account by assuming that the SL is made of a juxtaposition of perfect crystallites of the same composition \( x \) whose period varies between the extreme values of \( 2n_1d_1 + 2n_2d_2 \). In this hypothesis, the experimental diagrams is the sum of the X-ray diagrams given by each crystallite; the value of \( n_1 + n_2 \) deduced from such a diagram is an average value in the sample zone analyzed, therefore not necessarily integer, although it is found to be integer most of the time (see examples 1 and 3).

The calculations of \( d(00L) \) and \( I(00L) \) show in addition that dispersion in \( n_1 \) and \( n_2 \) has the effect of (1) broadening and deforming the satellite peaks and this occurs the further they are from the peak of the average lattice and (2) decreasing their intensities: if this dispersion is important, it leads to the disappearance of satellites beginning with those of highest order. It must be noticed that it is the relative dispersion which is important. So a uniform disper-

![Fig. 7. Third sample: 200 unit cells (\( n_1 = 9, n_2 = 9, x = 0.29 \)). (a) X-ray diffraction diagrams in the vicinity of the 000 and 002 GaAs reflections. (b) X-ray double-diffraction diagram in the vicinity of the 004 GaAs reflection.](image-url)
sion of 1 on \( n_1 + n_2 \) has little consequence when \( n_1 + n_2 = 140 \) (satellites of ±5 order broadened by 15°) but leads in practice to the disappearance of most satellites for \( n_1 + n_2 = 18 \) (satellites of ±1 order broadened by 190°, and satellites of ±2 order broadened by 378°) (Fig. 8).

In addition to these differences from the theoretical model due to the growth process, which can be taken into account by making the hypothesis of juxtaposed monocrystalline blocks, another type of fluctuation can be envisaged which concerns the distribution of aluminium at the interfaces Ga\(_{1-x}\)Al\(_x\)As/GaAs. At growth temperature of these layers, diffusion of aluminium is practically non-existent; Dernier, Moncton, McWhan, Gossard & Wiegmann (1977) found that the interface regions between essentially pure GaAs and AlAs are quite narrow and do not exceed two monolayers, that is to say 6 Å. In the calculation of structure factors we allow the \( x \) concentration to be constant in the Ga\(_{1-x}\)Al\(_x\)As layer of thickness \( e_1 \) and non-existent in the GaAs layer of thickness \( e_2 \). The hypothesis of a step variation of the \( x(z) \) function is perfectly justified in so far as \( \Delta n_1 + \Delta n_2 < 1/10 \) for the three above mentioned examples.

However, different interfaces can be created either at the time of growth, by introducing a modulation of aluminium concentration at each layer change, or by annealing under well defined conditions a SL corresponding initially to the step model. In order to estimate the influence of such a modification on the intensities of satellites it is assumed that the \( x(z) \) function at the interfaces is a sinusoid in which the half period is the width of the interface. This modification leads to a decrease in the number of the observable satellites which is more apparent as the interface is broader. So, Fig. 9 shows that the intensity ratio between the 002 peak of the average lattice and its satellites increases with the interface width; now the higher this ratio, the less visible the satellites.

This effect has been observed by Cabon (1983) on a sample whose interfaces have been modified by diffusion of aluminium under the influence of annealing. Fig. 10 shows the X-ray diagrams before and after thermal processing. The characteristics of this SL before annealing are: \( n_1 = 22, n_2 = 19, x = 0.47 \). After annealing (in conditions specified in Fig. 10) a decrease of the intensity of the satellites is observed which becomes more distinct as their order is increased. By introducing a sinusoidal modulation of the interfaces to the expression of the calculated structure factor we obtain a transition layer between the two materials which, after refinement, amounts to eight molecular planes (Fig. 11). These results are in good agreement with those of Fleming et al. (1980).

It is seen that the dispersion in \( n_1 \) and \( n_2 \) and the non-abrupt variation of \( x(z) \) at the interfaces have similar effects, i.e. the decrease in the number of observable satellites. However, the effect of dispersion is distinguished from the effect of the \( x(z) \) variation because it produces a decrease in intensity and a broadening of all the satellite peaks. Figs. 8 and 9 show that the relative dispersion of \( n_1 \) and \( n_2 \) and

![Fig. 8. Broadening of satellite peaks (≤0 order) in the vicinity of the 002 average reflection for a dispersion of 1 on \( n_1 + n_2 \). For satellites of positive order, the curves obtained would be almost identical.](image)

![Fig. 9. Variation of the ratio \( I(002)/(000L-i) \) between the intensity of the 002 reflection of the average lattice and that of various satellites \( (i = 1, 3, 5) \) as a function of the relative amplitude of the interface \( \Delta n/(n_1 + n_2) \) with \( n_1 + n_2 = 18 \) and \( x = 0.3 \). Similar results would be obtained for satellites of positive order.](image)
relative amplitude of the \( x(z) \) variation at the interfaces have limits beyond which the diffraction diagrams can no longer reveal the existence of the SL.

Another factor which is inherent to the nature of the \((\text{Ga}_{1-x}\text{Al}_x\text{As})_n(\text{GaAs})_{n_2}/\text{GaAs(001)}\) SLs limits the intensities of the satellite reflections and therefore their visibility: this factor is the aluminium concentration \( x \). In fact, (4) can be rewritten

\[
F_c(00L) = 2[x(f_{\text{Al}} - f_{\text{Ga}})R_1^{1/2}\frac{R_1^{n_1} - 1}{R_1 - 1} + 2f_{\text{As}}\left(\frac{R_1^{n_1} - 1}{R_1 - 1} + R_1^{n_1}\frac{R_2^{n_2} - 1}{R_2 - 1}\right) + 2f_{\text{Ga}}\left(R_1^{1/2}\frac{R_1^{n_1} - 1}{R_1 - 1} + R_1^{n_1}\frac{R_2^{n_2} - 1}{R_2 - 1}\right)]
\]

(5)

showing that, for the satellite peaks, \( F_c(00L) \) is more or less proportional to \( x \). This is because the last term of (5) corresponds to a GaAs structure in which an artificial modulation of the distance between atomic planes would have to be introduced, a term which, outside the reflexions of the average lattice, is not as important as the first term in (5).

Therefore, the smaller the value of \( x \) and the greater that of \( \Delta n/(n_1 + n_2) \), the weaker the intensities of the satellite peaks, the more difficult it will become to reveal the existence of the SL.

**Conclusion**

It has been shown that the average parameters \( n_1, n_2 \) and \( x \) of \((\text{Ga}_{1-x}\text{Al}_x\text{As})_n(\text{GaAs})_{n_2}/\text{GaAs(001)}\) SLs can be determined using classical X-ray diffraction techniques provided that a sufficient number of satellites are observed. In addition, the widening of satellites and the weakening of their intensities when their order increases is consistent with a dispersion in the number \( n_1 + n_2 \) of molecular layers in the unit cells of the SL.

This method of characterization is even more effective when the ratio \( \Delta n/(n_1 + n_2) \) is smaller and \( x \) is greater. Modification of the Al concentration at the interfaces between \( \text{Ga}_{1-x}\text{Al}_x\text{As} \) and \( \text{GaAs} \) also decreases the satellites intensities beginning with those of highest order.

So SL imperfections [i.e. variations in \( n_1 + n_2 \) and modifications in the original \( x(z) \) step function] gradually result in the disappearance of all satellites in the diffraction pattern; the question 'which degree of imperfection will produce such a disappearance?' has been partially answered by showing that a \( \pm 5\% \) relative dispersion in \( n_1 + n_2 \) results in such a widening of the \( \pm 1 \) order satellite peaks that they cannot, in practice, be detected any more.

However, an improvement of the techniques used (working with more powerful X-ray sources, for instance) ought to be sought in order to obtain more information from the diffraction patterns, particularly when the number of observed satellites is not sufficient to allow a full structural characterization.

**References**


Cabon, P. (1983). Contribution à l'Étude Cristallographique des Interfaces dans les Superréseaux \((\text{Ga}_{1-x}\text{Al}_x\text{As}, \text{GaAs})/\text{GaAs}\). CNET Report, 22301 Lannion, France.


