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Effect of graded layer on the X-ray double-crystal diffraction rocking curve

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The composition gradient in an InAlAs epitaxial layer on InP (0 0 1) substrate has been investigated using an X-ray double-crystal diffraction technique and a computer simulation method. Good agreement has been obtained between theoretical and experimental rocking curves when the correct graded layers are assumed in the samples. The results show that the graded layer introduces very sensitive asymmetric changes in layer peak and interference fringes. The intensities of the interference increase more strongly on the higher or lower angle side, while they are reduced on the other side, and the layer peak shifts to the higher or lower angle direction according to the positive or negative gradient. In all cases, however, the angle separations of the interference fringes do not change if the total thickness of the epilayer is unchanged.

1. Introduction

Multiple heterostructures are basic elements of optoelectronic devices. It is necessary to control and determine the actual parameters achieved in heterostructures, such as composition, thickness and the state of strains of individual layers. However, the occurrence of perfect heterostructures cannot be predicted. The depth-dependent strain (or composition) can occur in epilayers after diffusion and implantation processes or for other reasons [1–5]. To characterize the structure and quality of the epilayers, X-ray double-crystal diffraction (DCD) is one of the most powerful techniques. From DCD rocking curves (RCs) and their computer simulations, information about crystal perfectness and epilayer structure can be obtained [3, 6]. The rocking curve is sensitive to changes of epilayer composition, especially for large mismatch systems. For example, only 10^{-3} variation of x in $In_x Al_{1-x} As/InP$ would cause the layer peak to shift about 20 arcsec on the CuK_{α_1} 0 0 4 diffraction rocking curve. Therefore X-ray DCD provides a useful method to investigate composition fluctuation in epilayers. Usually variation of the composition in the epilayer is small; however, it obviously affects DCD rocking curves of the epilayers. Bensaussan *et al.* [4, 5] and Ferrari *et al.* [7] have reported evidence of this. In the present work the effects of changing composition in $In_xAl_{1-x}As/InP$ epilayers on X-ray rocking curves were investigated experimentally and theoretically.

2. Experiments and results

The $In_x Al_{1-x} As/InP$ (0 0 1) samples were grown by molecular-beam epitaxy. The RCs of the samples for the 0 0 4 reflection were recorded on a computer-controlled diffractometer with an RU-200 X-ray generator. Silicon with 1 1 1 symmetric diffraction was used as the first crystal. The double-crystal diffractometer was in the plus/minus setting with CuK_{α_1} radiation. Simulation of the rocking curves was performed on the basis of the X-ray scattering dynamical theory.

Fig. 1a shows the experimental 0 0 4 RC of sample 1 whose nominal structure is $In_{0.5} Al_{0.5} As$ (1 μm)/InP (0 0 1). From Fig. 1a one can see that the layer reflection splits into several peaks. Fig. 1b is the theoretical simulated RC, which agrees well with Fig. 1a. The parameters used in the simulation are listed in Table I. From Table I, five epilayers in Sample 1 were considered, in which compositions increase gradually from the interface to the surface. Thus Sample 1 has a composition graded structure.

Fig. 2a shows the experimental 0 0 4 RC of Sample 2 with nominal structure $In_{0.5} Al_{0.5} As$(0.4 μm)/InP (0 0 1), which has an asymmetric profile of the layer peak with stronger intensities of interference fringes on the lower angle side of the layer peak. This feature suggests a decreasing strain towards the substrate, which is contrary to that in Sample 1. Fig. 2b is the simulated RC, considering the epilayer

TABLE I The simulating parameters of sample 1

Sublayer	x	Thickness (μm)
1	0.4900	0.2
2	0.4905	0.2
3	0.4915	0.1
4	0.4920	0.1
5	0.4930	0.4

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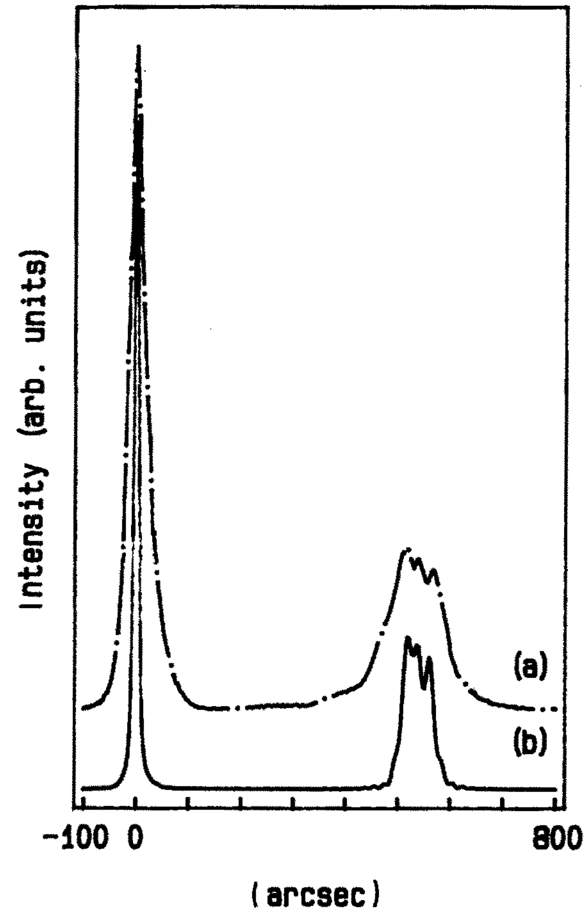

Figure 1 RC of Sample 1, (004) diffraction; (a) experimental, (b) simulated.

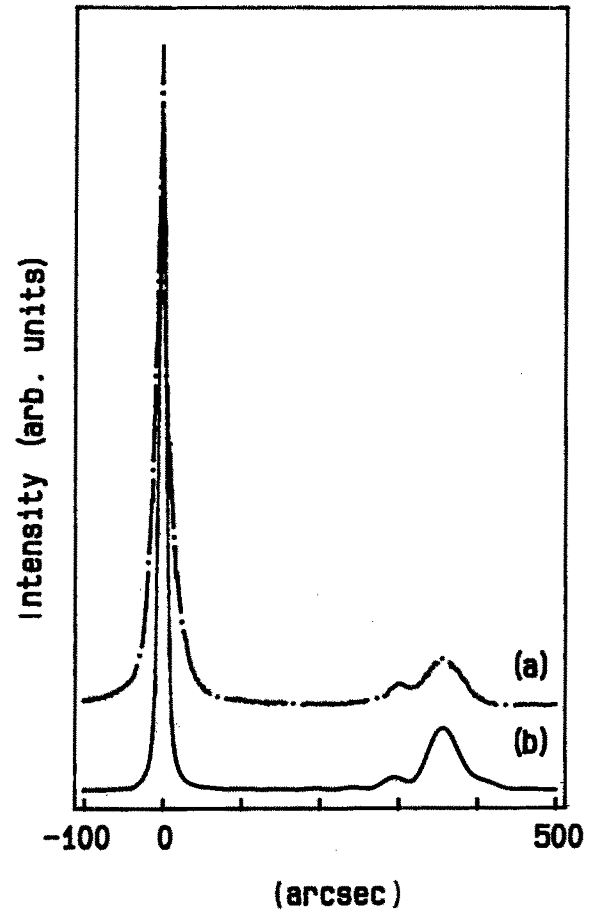

Figure 2 RC of Sample 2, (004) diffraction: (a) experimental, (b) simulated.

to be divided into six sublayers. The parameters used in the simulation are listed in Table II. This indicates that Sample 2 has the structure of a top layer and a graded layer. The variation of x in the graded layer is $\Delta x = 0.0025$.

A discrepancy between the experimental and theoretical rocking curves in Figs. 1 and 2 is seen in the full widths at half maximum (FWHMs) of the layer peaks on the experimental RCs, which are larger than those on the theoretical ones. This might be caused by various reasons such as the imperfections in samples which are ignored in the simulations.

The above results suggest that the composition variation layer might be brought during growth. Although the variation of composition is very small, it causes the X-ray DCD RCs to change obviously. The effect of the graded epilayer on the X-ray rocking curves by simulation, will be discussed systematically.

3. Calculations and discussion

To calculate the rocking curves, the Takagi–Taupin equations [8, 9] are used [6]. Random polarization is assumed in the X-ray incident beam. For convenience, we consider that a graded layer is divided into several sublayers in which compositions and structure factors are constant. A linear gradient is assumed in the graded layer. When the graded layer is divided into n sublayers, the composition–depth profile is a step-like function. For the $In_xAl_{1-x}As/InP$ (001) samples considered in our calculations, the mismatch between the epilayer and the substrate is negative. The gradient is positive if the composition becomes larger from the interface towards the surface. No other imperfection is assumed in the substrate and the layer except the composition grading.

3.1. Graded epilayer

If the composition varies through the whole epilayer, we define it as a graded epilayer. For the $In_xAl_{1-x}As(1\ \mu m)/InP$ (001) samples, a mean composition $\bar{x}_0 = 0.51$ and a total thickness $T = 1\ \mu m$ have been assumed. The composition gradient is positive and the variation of x *is* Δx. Fig. 3 shows a set of RCs of the graded epilayers with different values of Δx. From this figure one can see that the layer peak decreases in intensity and broadens in width of $\Delta x \neq 0$. As Δx increases, the rocking curves become complicated with the layer peak developing shoulder peaks beside it. The shoulders gradually grow as Δx increases and develop into separated peaks when Δx is large. This is

TABLE II The simulating parameters of sample 2

Sublayer	x	Thickness (μm)
1	0.5050	0.05
2	0.5045	0.04
3	0.5040	0.04
4	0.5030	0.04
5	0.5025	0.04
6	0.5010	0.20

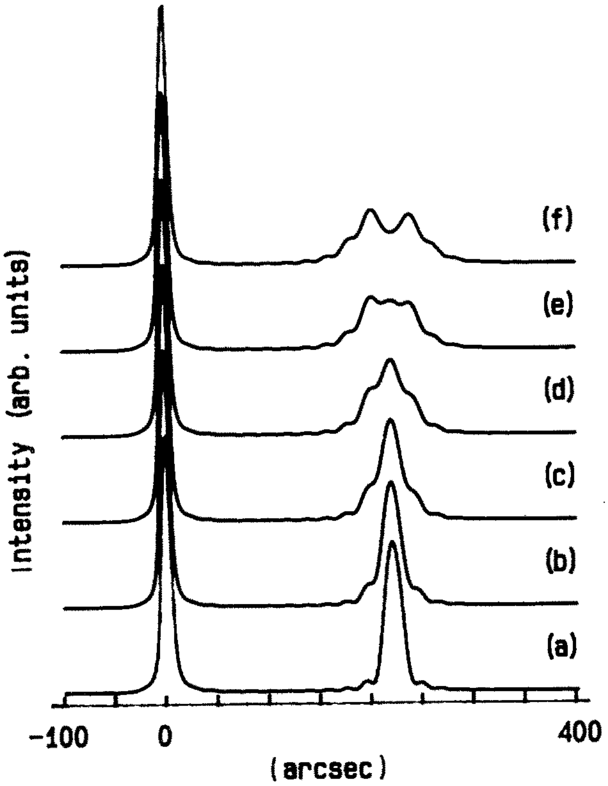

Figure 3 The calculated 004 RCs of In_xAl_{1-x} As/In P, positive graded epilayer with different Δx: (a) 0; (b) 0.002, $n = 15$; (c) 0.003, $n = 15$; (d) 0.004, $n = 20$; (e) 0.005, $n = 20$; (f) 0.006, $n = 30$.

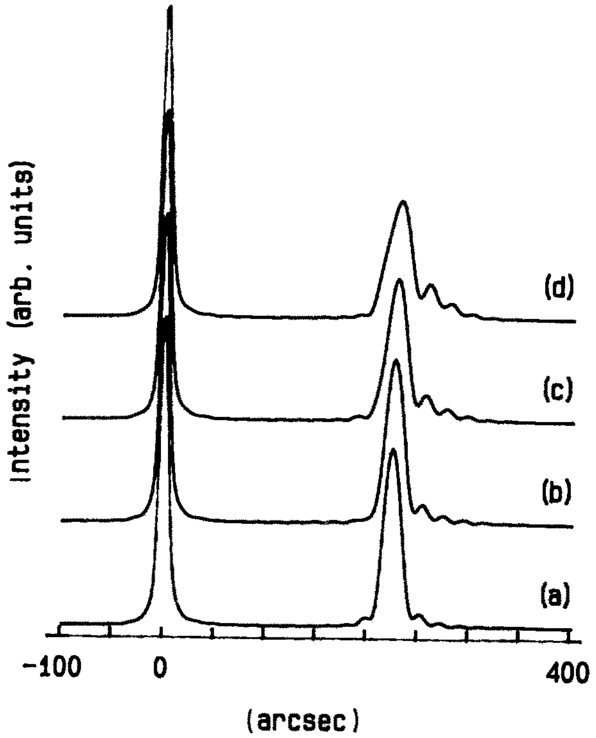

Figure 4 The calculated 004 RCs of $In_xAl_{1-x}As/InP$ with positive gradient transitional layer of different thickness, t: (a) 0.1 μm, (b) 0.2 μm, (c) 0.3 μm, (d) 0.4 μm.

expected from the interference of the graded sublayers. For a given layer thickness, the phase shifts of the X-ray waves increase with increasing Δx. The main peak intensity decays due to the overlapping between the large shifted X-ray waves, while the interference fringes are enhanced. In addition, in all curves, the epilayer peak is asymmetrical with the lower angle side steeper, and with interference fringes stronger in the lower angle side of the layer peak. This is to be expected because the intensities of interference fringes on the lower angle side increase with the interference of the epilayer and the substrate. However, from Fig. 3 one can see that the period of the interference fringes on each curve, $\delta\theta$, is unchanged with variation of composition. This indicates that the period of the fringes is dependent upon the thickness, instead of the composition, of the epilayer.

3.2. Graded transitional layer

Sometimes there is a thin region in the epilayer close to the substrate in which the composition changes gradually. We define this region as a graded transitional layer. The graded transitional layer changes in two ways: the change of interval, Δx, and/or thickness, t. Fig. 4 shows the RCs of $In_x\ Al_{1-x}$ As/InP with a graded transitional layer of different thickness, t. The total thickness of the epilayer $T = 1$ μm and the top layer is $In_{0.51}\ Al_{0.49}$ As with thickness $t_0 = (1 - t)$ μm. The composition gradient is positive, $\Delta x = 0.004$. From Fig. 4 one can see that as t increases, the intensity of the layer peak decreases and peak width increases. The positions of the epilayer peak and interference fringes shift to higher angle. At the same time, the intensities of the interference fringes increase in the higher angle side of the epilayer peak, while decreasing on the other side. This is because the fringes on the higher angle side correspond to a larger strain in the layer caused by a composition (also a lattice constant) positive gradient. The intensities of fringes are enhanced by interference of the graded sublayers with decreasing X-ray travel distance in the graded layer.

Similar to Fig. 4, the RCs of $In_x\ Al_{1-x}$ As graded layer with negative gradient are calculated as shown in Fig. 5. It also shows an asymmetric change in interference fringes and epilayer peak. In contrast to Fig. 4, the interference fringes shift to the lower angle range. This is because the reflections in the lower angle range correspond to the interference of the graded laminae with smaller values of strain. The enhancement of the interference fringes on the lower angle side is due to the X-ray phase shift increasing with t. From Figs 4 and 5 we find that the angle separation of the interference fringes on each curve, $\delta\theta = 19$ arcsec, is almost the same. This is different from the previous result of Ferrari and Franzosi [7] who supposed that the periods of the interference fringes change with variation of the lattice mismatch gradient.

In addition, the change of Δx in the graded layer is considered. Fig. 6 shows calculated RCs of $In_x\ Al_{1-x}$ As/InP with different graded layer in which the variation of composition Δx is 0.001–0.004. The composition gradient is positive and the thickness of graded layer $t = 0.5$ μm. The top layer is $In_{0.51}\ Al_{0.49}$ As(0.5 μm). In Fig. 6, an asymmetric change occurs in the layer peak and interference fringes on each curve.

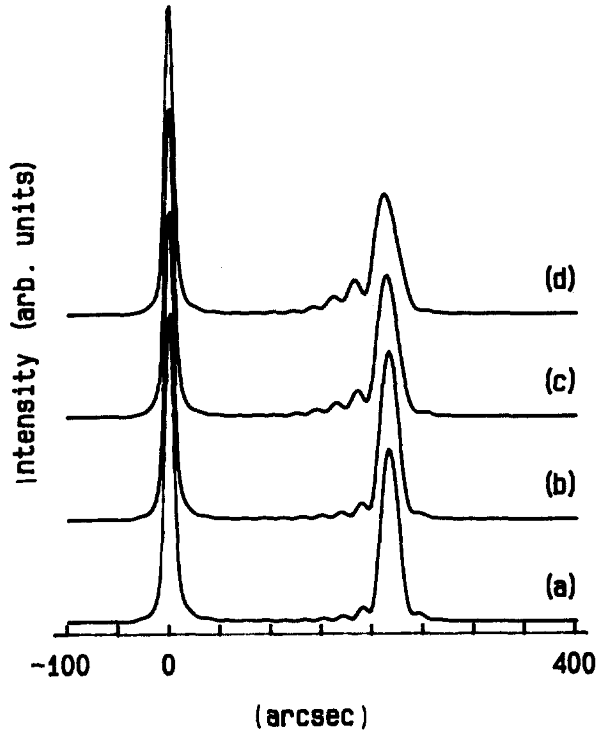

Figure 5 The calculated 0 0 4 RCs of In_xAl_{1-x} As/InP with negative gradient transitional layer of different thickness, t: (a) 0.1 μm, (b) 0.2 μm, (c) 0.3 μm, (d) 0.4 μm.

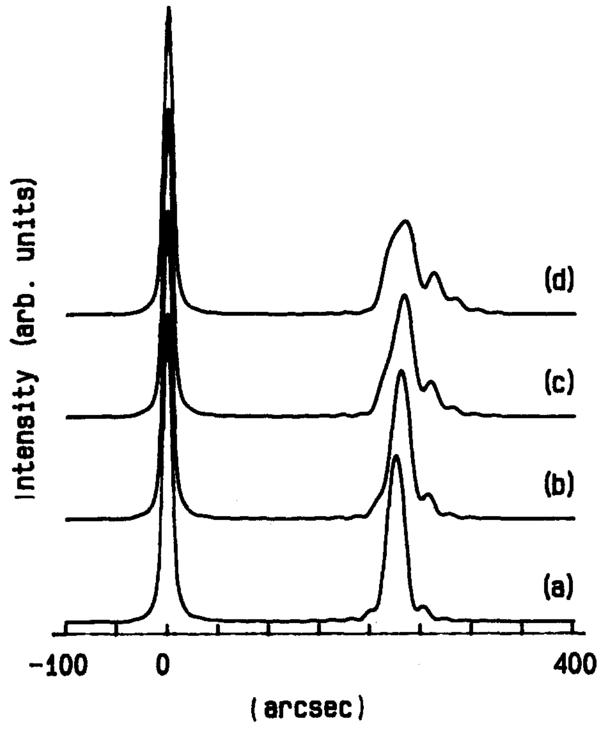

Figure 6 The calculated 0 0 4 RCs of In_xAl_{1-x} As/InP with positive gradient transitional layer of different Δx: (a) 0.001, (b) 0.002, (c) 0.003, (d) 0.004.

As Δx increases, the intensity of the layer peak gradually decreases and its position shifts to the higher angle direction. The interference fringes shift to the higher angle side and their intensities become stronger in the higher angle range and weaker in the lower angle side. The larger is Δx, the more greatly the peaks change and the rocking curve tends to develop a new peak. These phenomena are also due to the increasing phase shifts of the X-ray waves in the graded layer with increasing Δx.

If the gradient is negative, similar results could be obtained from RCs of $In_x Al_{1-x}$ As/InP, as shown in Fig. 7. With respect to Fig. 7 the interference fringes increase more strongly in the lower angle side and shift to the lower angle side.

Therefore we come to the conclusion that with a graded layer in the epilayer:

(a) the maximum layer peak shifts, broadens and decreases on RC and the positions and intensities of interference fringes are sensitive to t or Δx of the graded layer;

(b) When the value of t or Δx is larger, the rocking curves become complicated and new peak(s) appear;

(c) the angle separation of the interference fringes will be unchanged if the total thickness of the epilayer remains constant.

To understand (a), we turn to the Bragg law. The angle difference between the substrate peak and the peak for the ith sublayer is given approximately by

$$\Delta\theta_i = -(\Delta d/d)_i^{\perp}\tan\theta_B \quad (1)$$

If composition changes, $(\Delta d/d)_i^{\perp}$ and $\Delta\theta_i$ change. The effect of all laminae interference is to cause the mean layer peak intensity to decrease and the FWHM to broaden and its position to shift. Also, the intensities of the interference fringes vary. The larger is Δx or t of the graded layer, the more different are $(\Delta d/d)_i^{\perp}$ and $\Delta\theta_i$, and the more greatly the peaks change.

It is necessary to point out that with a graded layer, one cannot calculate the true strains and compositions in epilayers from Equation (1). To obtain the correct

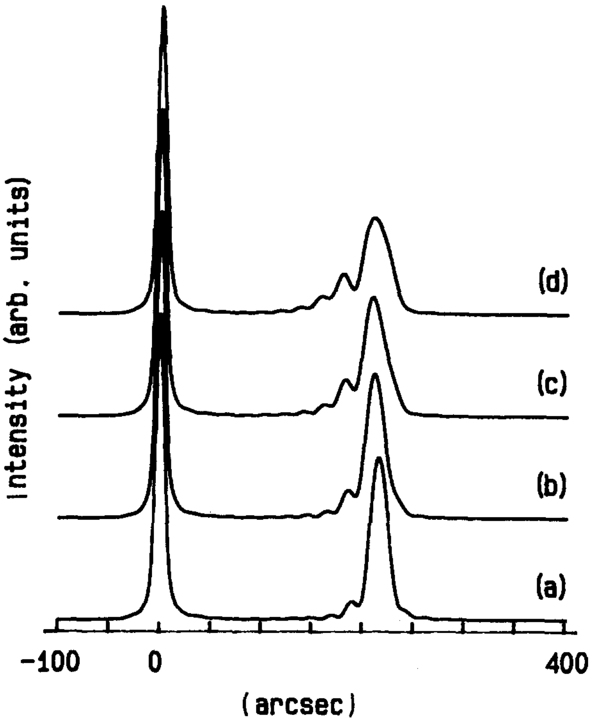

Figure 7 The calculated 0 0 4 RCs of In_xAl_{1-x} As/InP with negative gradient transitional layer of different Δx: (a) 0.001, (b) 0.002, (c) 0.003, (d) 0.004.

structural parameters, a powerful method is to simulate the experimental rocking curves by using X-ray scattering dynamical theory.

From Condition (c) it is suggested that the angle separations between the interference fringes are not affected by the change in composition of the epilayer. This can be interpreted by X-ray diffraction kinematical theory. The angle separation, $\delta\theta$, is given by [10]

$$\delta\theta = \lambda|\gamma_h|/T\sin 2\theta_B \qquad (2)$$

where $\gamma_h = \sin(\theta_B - \phi)$ and T is the total thickness of epilayer. If T is constant, $\delta\theta$ would be unchanged. This is consistent with our results. Therefore, the thickness of the epilayer could be determined from the angle separation of interference fringes on the rocking curves.

4. Conclusion

The graded layer introduces an asymmetrical change in the layer peak and interference fringes on the rocking curve of the epilayer. For a graded epilayer, as the composition gradient increases, the intensity of the layer peak decreases and the peak width increases. Shoulder peaks develop beside the layer peak and the shoulders change into separated peaks when the gradient is large. For a graded transitional layer, as the variation of composition or thickness of the graded layer increases, the interference fringes shift to the higher angle side and their intensities become stronger in the higher angle range when the gradient is positive and vice versa. A new diffraction peak appears when the variation of composition or thickness of the graded layer is large. The larger the variation of composition or thickness, the more greatly the peaks change. However, with a constant thickness of epilayers, the angle separations between the interference fringes are unchanged. By fitting the experimental rocking curves of $Al_{0.5}In_{0.5}As/InP(001)$ samples using the X-ray scattering dynamical theory, evidence of graded layers has been found in two samples. Moreover, X-ray DCD and computer simulations of the rocking curves provide a powerful method to investigate the structures of graded layers.

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