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B. A. Joyce

*Imperial College*, [b.joyce@ic.ac.uk](mailto:b.joyce@ic.ac.uk)

X. M. Zhang

*Imperial College*

J. H. Neave

*Imperial College*

P. N. Fawcett

*Imperial College*

M. R. Fahy

*Imperial College*

*See next page for additional authors*

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## Reflection High Energy Electron Diffraction (RHEED) Intensity Oscillations: Growth Modes and Growth Rates: A Critique

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## REFLECTION HIGH ENERGY ELECTRON DIFFRACTION (RHEED) INTENSITY OSCILLATIONS: GROWTH MODES AND GROWTH RATES: A CRITIQUE

B.A. Joyce\*, X.M. Zhang, J.H. Neave, P.N. Fawcett, M.R. Fahy, K. Sato<sup>1</sup> and I. Kamiya

Semiconductor Materials IRC, The Blackett Laboratory, Imperial College,  
Prince Consort Road, London SW7 2BZ, United Kingdom

<sup>1</sup>Permanent address: Japan Energy Corporation, 10-1 Toranomom 2-chome, Minato-ku, Tokyo 105, Japan

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### Abstract

The origin of and diffraction effects associated with reflection high energy electron diffraction (RHEED) intensity oscillations which occur during layer-by-layer growth of epitaxial thin films of III-V compounds by molecular beam epitaxy (MBE) are explained. It is shown that on (001) oriented substrates the period of the oscillations is in general a direct measure of the film growth rate which corresponds to the group III element flux. There are, however, exceptions to this simple concept including growth under group III rich-conditions, vicinal plane growth and growth from pulsed beams; each is considered.

On non-(001) low index orientations, the RHEED oscillation period only provides a measure of the growth rate over a very limited range of conditions. The fundamental reason appears to be the more restricted reactivity between the group III and V elements, so the oscillations are induced by the group V element, not the group III, which is quite different from (001) surfaces, at least for conventional growth conditions.

Finally, growth modes and strain relaxation differences between (001) and (110)-based growth of InAs on GaAs are illustrated. It is shown that there is no real relationship between strain and growth mode and it is suggested that adatom mobility is the essential parameter which determines growth mode.

In more general terms, it appears that kinetic factors rather than equilibrium considerations are responsible for the growth mode. Models based on purely equilibrium concepts are therefore unlikely to have general validity.

**Key Words:** Reflection high energy electron diffraction (RHEED), intensity oscillations, layer-by-layer growth, III-V compound semiconductor growth, epitaxy, strain relaxation.

\*Address for Correspondence:

B.A. Joyce, address as above

Phone: 44 171 594 6666 / FAX: 44 171 581 3817

Email: b.joyce@ic.ac.uk

### Introduction

Since they were first observed [14, 21, 23], intensity oscillations of the specular (and other) beams in reflection high energy electron diffraction (RHEED) patterns during film growth have generally been assumed to result from a two dimensional (2-D) layer-by-layer growth mode, with the period of the oscillations corresponding to the growth of a single monolayer (atomic or molecular), although some instances of bilayer growth have been reported [20, 24]. The same relationships have also been assumed for temporal intensity variation of other surface-sensitive probes, such as low energy electron diffraction [17], helium atom diffraction [12] and X-ray diffraction [11].

Optical techniques which probe surface effects, such as photoemission (PE) [6, 32] and reflectance difference spectroscopy (RDS), otherwise known as reflectance anisotropy (RA), also generate intensity oscillations [3, 19] for which the same assumptions of monolayer periodicity and growth modes are made. This in itself is worthy of further consideration, since diffraction techniques respond to long range order and morphology, while PE and RDS probe local electronic structure. It is not immediately obvious why such different effects should apparently produce the same temporal response during film growth.

The overwhelming majority of published work refers to growth by molecular beam epitaxy (MBE) on (001) oriented substrates (or vicinal surfaces a few degrees away from (001)), and much of that to III-V semiconductors in general, and GaAs in particular. There is nevertheless sufficient work on Si, Ge, metals and insulators to be able to conclude that there are no major material-dependent differences, at least for (001) growth.

It is only very recently that growth on orientations other than (001) has begun to be investigated systematically, and then only by RHEED, but it is already apparent [7, 8, 38] that there are significant differences from (001) and that the assumptions which previously seemed valid may no longer be appropriate.

In the first part of this paper, we will briefly

summarise our understanding of the temporal intensity behaviour as it has been developed for (001) growth, including the importance of diffraction effects. We will also indicate the excellent level of agreement which has been established between RHEED measurements and a Monte Carlo simulation of growth which ignores chemical effects [26]. The validity of this approach is strongly supported by scanning tunnelling microscopy (STM) observations of the final state film morphology at the atomic level [30].

In the second part of the paper, we will show that many of the simple ideas developed for (001) growth, especially of III-V compounds by elemental source MBE, break down once other orientations are considered. In particular, oscillation periods do not necessarily correspond to growth rates, even on singular surfaces, and growth modes may not follow the simple pattern of Frank-van der Merwe (2-D nucleation) [10], Volmer-Weber {three-dimensional (3-D) nucleation} [34] or Stranski-Krastanov (layer-by-layer growth for a limited number of monolayers, followed by three-dimensional nucleation and growth) [29].

In many covalently-bonded epitaxial systems where there is a significant misfit between layer and overgrowth it is observed that growth follows the Stranski-Krastanov mode and is initially pseudomorphic, or in extreme cases it is three-dimensional from the outset, whereas for similarly bonded systems with zero or very small amounts of misfit, the growth mode is two-dimensional. It has consequently been asserted that the growth mode is directly related to the misfit, and once the elastic strain imposed by pseudomorphic growth is relaxed by the formation of misfit dislocations, either in the initial or first few monolayers, growth becomes three dimensional. Again, it will be shown that this simple correlation is not universally applicable and indeed may have only limited validity, or even lack it entirely beyond pure coincidence.

### Origin of RHEED Intensity Oscillations During the Growth of III-V Compounds on (001) Surfaces

We have chosen a very specific starting point: RHEED intensity oscillations which occur during growth on singular (001) oriented substrates from beams of group III and group V elements. The reason for this specificity is that virtually all work aimed at establishing the origin and explanation of intensity oscillations has been carried out within this framework. It will be seen that it provides an adequate starting point for subsequent discussions.

The basic observation we need to understand is a periodic intensity variation of all diffracted features (and the magnitude of the substrate current) during growth,

the period corresponding precisely to the growth of a single atomic or molecular layer on a singular surface. Such a response during thin film deposition is generally considered to be a manifestation of a two-dimensional, layer-by-layer growth mode and is common to most surface sensitive techniques. It is important to evaluate the temporal intensity variations in relation to both growth and diffraction conditions since, before we can study growth, the effects associated solely with diffraction must be assessed.

This can most readily be established by recording intensity oscillations as a function of polar and azimuthal angles for constant growth conditions. Typical data sets from such an investigation are shown in Figure 1. There is clearly a wide range of oscillation waveforms, and since the growth conditions were invariant, the differences must arise from diffraction-related rather than growth effects.

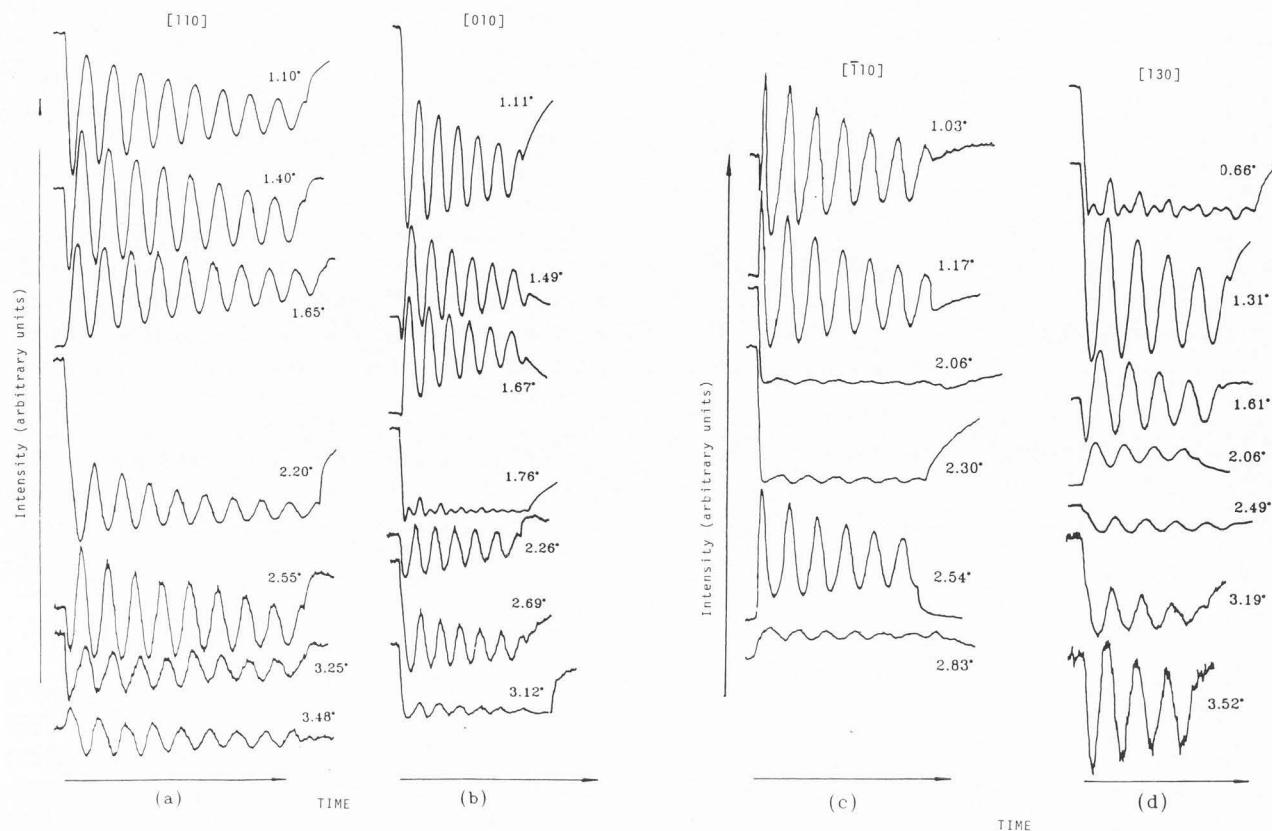
The most important feature is that the initial intensity response to the initiation of growth is quite varied, and consequently there is no fixed phase relationship with respect to polar and azimuthal angles, although in general, the steady state period is constant. This effect is illustrated in Figure 2, in which data points represent the time to the second oscillation minimum  $t_{3/2}$ , normalised by the steady state period  $T$ , plotted as a function of polar angles for different azimuths. Assuming the oscillations to be (damped) sinusoids, an ordinate value of 1.5 indicates the correspondence of oscillation maxima with incremental monolayers (ML). It is clear from Figure 2, however, that this is seldom the case.

Other oscillation waveform effects which must be accounted for in any theory include the following:

- i. A doubling of the oscillation period for certain azimuthal and polar angles.
- ii. A wide range of oscillation amplitudes.
- iii. The oscillations are damped.
- iv. Diffracted intensity is transferred between beams and background during growth.
- v. The intensity of the specular beam "recovers" to its initial value when growth is terminated, but the time constant is sensitive to both diffraction and growth conditions.

The predominant reason for this complex behaviour is the multiple scattering nature of electron diffraction, which results from the strong interaction between the incident electron beam and the crystal. Detailed accounts have been published elsewhere [36], but in summary, the measured intensity includes components from a range of diffraction processes, coherent and incoherent, elastic and inelastic. The precise nature of the components determines the waveform for any specific diffraction condition. Although not unequivocally proven, there is strong evidence from the agreement between Monte Carlo

## RHEED intensity oscillations

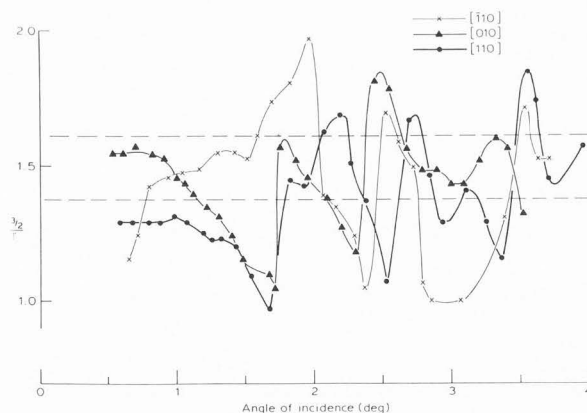


**Figure 1.** RHEED intensity oscillations of the specular spot on the 00 rod in (a) [110], (b) [010], (c)  $\bar{1}10$  and (d) [130] azimuths from a GaAs (001)-2x4 reconstructed surface during growth, as a function of polar angle with a linear ordinate scale. The primary beam energy was 12.5 keV and constant growth conditions were used throughout.  $T_s = 580^\circ\text{C}$ ,  $J_{\text{Ga}} = 1 \times 10^{14}$  atoms  $\text{cm}^{-2}\text{s}^{-1}$  and  $J_{\text{As}_2} = 2 \times 10^{14}$  molecules  $\text{cm}^{-2}\text{s}^{-1}$ .

simulations of growth and RHEED oscillation measurements [26] (see below), that when an intensity maximum corresponds to an incremental monolayer, step edge scattering is the dominant process.

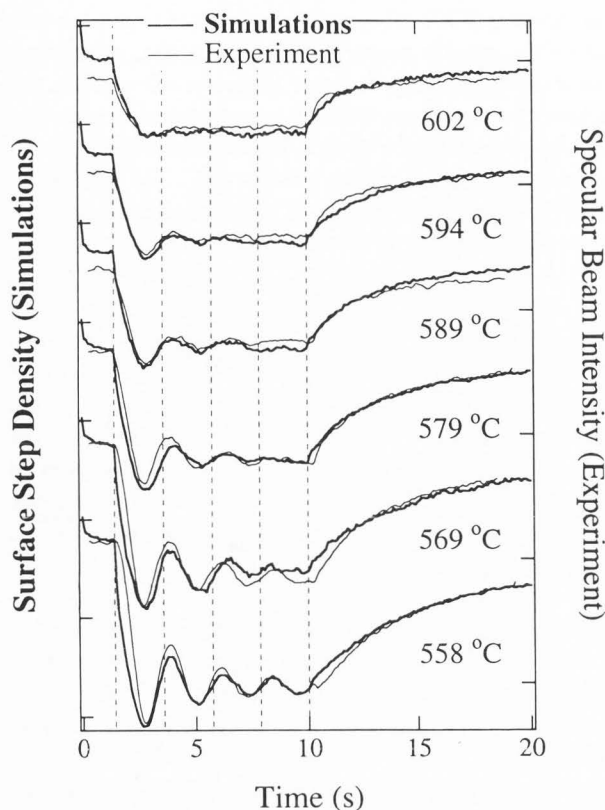
We will now consider in rather more detail the information on growth mode and kinetics which can be extracted from the oscillation waveform on singular (001) surfaces.

For MBE growth on a singular surface, the process of creation (2-D nucleation) and coalescence of islands is continuous, irrespective of the decay (damping) of the oscillations, so there is a variation of surface morphology on a microscopic scale throughout the whole growth period. This change is sampled by the incident electrons over an area (coherence area) defined by their coherence length and width. When growth is initiated on a singular surface, morphological changes occur in a correlated



**Figure 2.** Phase relationships of RHEED oscillations as a function of polar angle for [110], [010] and  $\bar{1}10$  azimuths from a GaAs (001)-2x4 surface. The growth conditions are defined in Figure 1. Phase is defined as the time taken to reach the second minimum normalised by the time of a complete period at steady state.

manner over regions of the surface much greater than the coherence area. The whole of the incident electron beam therefore samples the same periodic growth morphology, and intensity oscillations are observed. Widely



**Figure 3.** Direct comparison between measured RHEED intensity and the simulated surface step density on a GaAs (001) vicinal surface misoriented by  $2^\circ$  towards [010]. The growth rate was  $0.47 \text{ ML s}^{-1}$ . The ordinate scale is linear.

separated regions of the surface soon begin to lose their long range correlation, however, so different regions show different stages of growth, and the RHEED intensity is composed of contributions of widely correlated regions and the oscillations show a corresponding decaying envelope as the morphology gradually approaches a steady state value. Stated explicitly, the necessary condition for intensity oscillations is the periodic variation of step density over regions much greater than the coherence area of the incident beam. It is therefore apparent why the oscillation period is a measure of ML growth in the layer-by-layer growth mode and why the oscillations are damped. To progress further requires a more detailed growth model, and the most appropriate has been a Monte Carlo simulation based on the solid-on-solid model introduced by Gilmer and Weeks [35] but developed explicitly for MBE by Clarke and Vvedensky [1, 2]. This requires that some morphological feature on the growing surface can be related to the RHEED intensity and the step density has been found to

be the optimum parameter. Not only is there a remarkable qualitative correspondence, but there is also a quantitative relationship in that calculated step densities and measured intensities are related by a proportionality constant over a wide range of temperatures and fluxes. The relationship is nevertheless only phenomenological, and work is still in progress to develop a more fundamental basis of correlation.

Details of the Monte Carlo simulation studies have been published in a series of papers [1, 2, 26], so here we will only present a brief synopsis before illustrating the presently obtainable levels of agreement between simulation and experiments and their significance. A key feature is the assumption that growth kinetics are dominated by the group III species, i.e., that Ga-As<sub>x</sub> ( $x = 2$  or  $4$ ) reaction chemistry is not rate limiting. The processes which are considered are therefore (i) the random arrival of atoms (Ga) on the surface from the molecular beam at a defined rate and (ii) the surface migration of these adatoms, taken to be a nearest-neighbour (nn) hopping process, whose rate is given by:

$$k(T) = k_0 \exp(-E_D/k_B T) \quad (1)$$

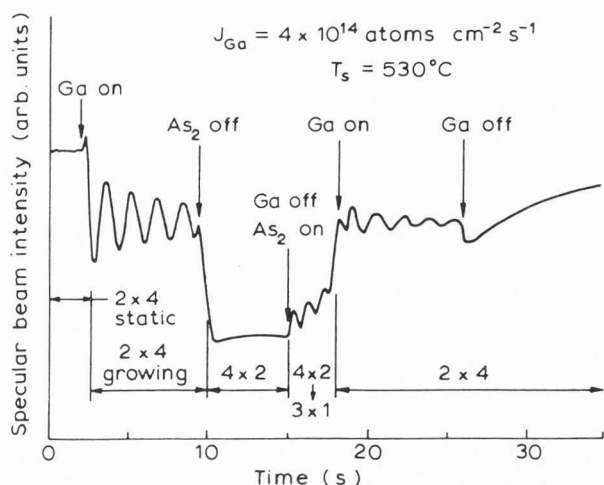
where  $k_0$  is the adatom vibrational frequency ( $k_0 = 2k_B T/h$ ), usually taken to be approximately  $10^{13} \text{ s}^{-1}$  and  $E_D$  is the barrier to hopping which is made up of two terms; a term  $E_S$  due to the substrate and a contribution  $E_N$ , from each nn to the adatom in the surface plane. The total barrier to hopping is thus:

$$E_D = E_S + nE_N \quad (2)$$

where  $n$  is the number of nn's ( $n = 0, 1, \dots, 4$ ). If  $k_0$  is assumed to be temperature independent over the comparatively narrow range involved, then the model has only one free parameter,  $E_N/E_S$ , since the effect of  $E_S$  is simply to renormalise the temperature.

The simulation then gives the step density as a function of Ga flux and substrate temperature for chosen values of  $E_D$  based on experimental measurements using a vicinal plane technique [23] in which the temperature where growth becomes dominated by step advancement is determined. This is a reasonably well defined quantity which can be simulated with the step density and provides a method to evaluate the parameters of the model [26].

A direct comparison of simulation and experiment is shown in Figure 3, in which the same rescaling was used for each temperature for a given Ga flux. These experiments were carried out on a vicinal plane, in which the growth mode changes from 2-D nucleation on the terraces to step propagation as the temperature is raised and the mobility of the adatoms increases (23).



**Figure 4.** Specular beam intensity for GaAs growth following 5 ML Ga predeposition on a GaAs (001)-2x4 substrate surface. The ordinate scale is linear.

It provides a very stringent test for the SOS model of MBE, but introduces one complication in respect of the oscillation period (see below). It can be seen that the step density closely matches qualitatively and quantitatively the RHEED waveform, but it is essential to re-emphasise that this only occurs provided diffraction conditions are chosen so that step-edge scattering is the dominant process. This can, however, only be done empirically by determining, for a particular azimuth, the polar angle at which maxima in the RHEED oscillations correspond to incremental monolayers as discussed by Zhang *et al.* [36] and illustrated in Figure 2.

Areas of common response between simulation and experiment are: (i) the initial decrease in intensity as a function of temperature at the start of growth; (ii) the amplitude of the oscillations at low temperatures; (iii) the steady state intensities at all temperatures; (iv) the gradual disappearance of the oscillations; (v) the decay envelope of the oscillations; (vi) the shapes of the oscillations; (vii) the increasing period with increasing temperatures (viii); the recovery of intensity at the cessation of growth, although for agreement at this stage it is necessary to introduce an activation barrier to hopping at step edges [28].

By appropriate choice of diffraction conditions and application of the vicinal plane technique, we have established the relationship between RHEED oscillations and growth mode for the ideal case. In the same context, the oscillation period provides a direct measure of growth rate on a singular (001) surface and at conventional growth temperatures this equates with the group III element flux for III-V compounds.

There are, however, several situations, even with (001) substrates where this is not the case. They include growth under excess group III element conditions, growth on vicinal planes and growth from pulsed beams. We will discuss each of these in turn.

#### Growth under excess group III conditions

When an excess of the group III element is formed at the surface, either by predeposition, surface segregation during growth or growth using a high III:V flux ratio, the oscillation period corresponds to the growth of a ML, but it is not related to the group III element flux. Instead it relates to the group V element flux ("arsenic induced oscillations") but only as the product of this flux and the sticking coefficient of the group V element [22]. It is effectively a measure of the reactivity of the group V element toward the group III element and does not imply the simple condensation of the group V element; the excess group III element must be "consumed" (i.e. undergo reaction to form the III-V compound).

A typical example is shown in Figure 4, where ~5 ML of excess Ga was deposited on GaAs(001)-2x4 surface in the absence of an arsenic flux. The Ga flux was then terminated, and an As<sub>4</sub> flux initiated. Oscillations induced by the As flux then continued until the excess Ga was consumed.

#### Growth on vicinal substrates

Neave *et al.* [23] first reported the observation of a growth mode transition on a vicinal GaAs (001) substrate. At a constant Ga flux the growth mode changed from two-dimensional nucleation on the terraces to step propagation as the temperature was increased. Below some critical temperature ( $T_c$ ), RHEED intensity oscillations were observed because growth involved 2-D nucleation, but at temperatures  $>T_c$ , they were absent since the step propagation growth mode produced no statistical change in surface morphology. The concept of growth mode change with temperature as the result of increased migration of adatoms has subsequently been confirmed by STM observations [18, 30, 31].

In the context of the present discussion, however, the important result is the increase in the period of the oscillations as  $T_c$  is approached from below, as shown in Figure 5. The explanation is simple and is shown schematically in Figure 6. There is not a step function change in growth mode at  $T_c$ , but with increasing substrate temperature there is a gradual reduction in the number of adatoms involved in 2-D nucleation as an increasing number are able to migrate to step edges. RHEED oscillations, however, only record the proportion involved in the 2-D process, so the period increases, or the apparent growth rate decreases. In other words, the oscillation period does not provide a simple measure of the growth rate. Shitara *et al.* [27] have

shown how the changing period can be used to determine the kinetics of step propagation, but that need not concern us here.

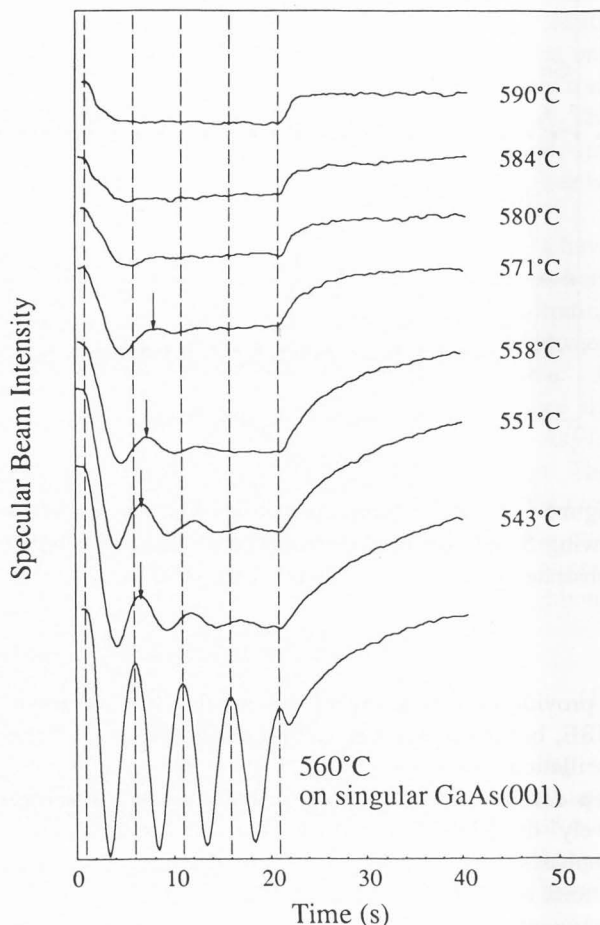
#### Growth from pulsed beams (migration enhanced epitaxy, MEE)

Horikoshi *et al.* [15] have developed a modified growth technology which is different from conventional MBE in that the group III and V fluxes are not co-incident, but arrive alternately from pulsed sources, with or without time intervals in which there is no incident flux. If the RHEED intensity is monitored throughout these cycles, it is also found to oscillate, but the oscillations continue throughout the duration of growth; moreover, they are not damped. Their origin is, however, completely different from those previously discussed. They in fact arise from transient changes in surface reconstruction which occur as the surface stoichiometry changes with the pulsed fluxes. They are not a measure of the growth rate in the conventional sense, but are related more to the local electronic structure (i.e., reconstruction) than to long range order, so in that sense, their origin is more akin to oscillations observed by optical techniques such as RDS.

#### Relationship Between RHEED and RDS Oscillations

It has been well established that real-time oscillations with identical periods can be obtained simultaneously from RHEED and RDS during the MBE growth of GaAs and AlAs (001) oriented films [13, 19]. Comparability of the phase of the oscillations is not a meaningful issue, since as we have clearly shown, the phase of the RHEED oscillations is a function of diffraction conditions. An important factor however, is that RDS oscillations are obtained over a very wide range of incident wavelengths, despite the spectral response of a static surface, where strong absorption peaks associated with surface dimers (As or Ga, depending on the reconstruction) are observed [3].

The layer-by-layer growth mode generates RHEED oscillations through a temporal cycle of island formation and coalescence, it is a long range phenomenon and the system evolves to a dynamical steady state characterised by a constant step density. When this state is reached, there are no further oscillations. The RDS response is presumably derived from the local electronic structure and shows much less tendency to damping. Nevertheless the period indicates that this local state changes continuously and systematically as each new ML is deposited. The absence of any strong dependence of the incident beam wavelength implies a lack of chemical specificity in the response, however, which makes it very difficult to determine the basis of the RDS oscillations.



**Figure 5.** Changing oscillation period during growth on a vicinal GaAs (001) surface misoriented by  $2^\circ$  towards [010]. The bottom trace was measured on a singular GaAs (001) surface with the same incident angle and azimuth as on the vicinal surface. The ordinate scale is linear.

Their origin is still not established, but some form of transient reconstruction change cannot be ruled out.

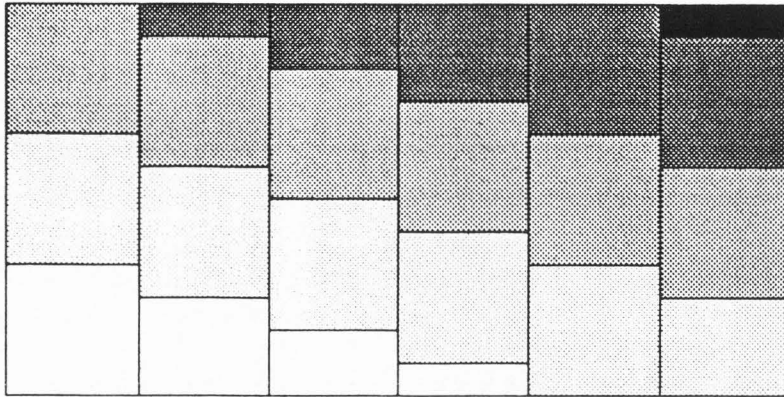
#### Growth of GaAs on non-(001) Oriented GaAs Substrates

The extension of RHEED studies to other low index orientations, i.e., (110) and  $\{111\}$ , has only occurred comparatively recently; consequently, there is not the same body of systematic work as on (001). The incentive to consider other orientations largely derives from the increased activity in growth on patterned substrates in pursuit of the fabrication of various mesoscopic structures with increased degrees of carrier confinement.

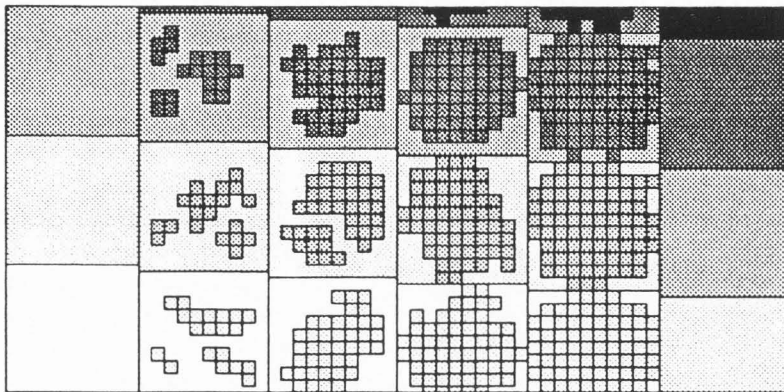


**Figure 6.** Schematic illustration to explain the growth mode transition which occurs on vicinal surfaces and the origin of the increasing oscillation period. The grey scale represents the height of the surface.

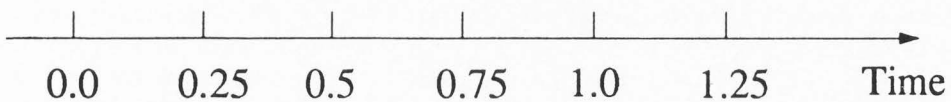
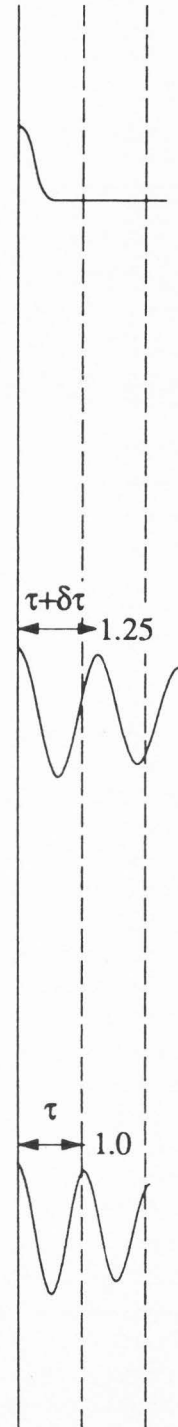
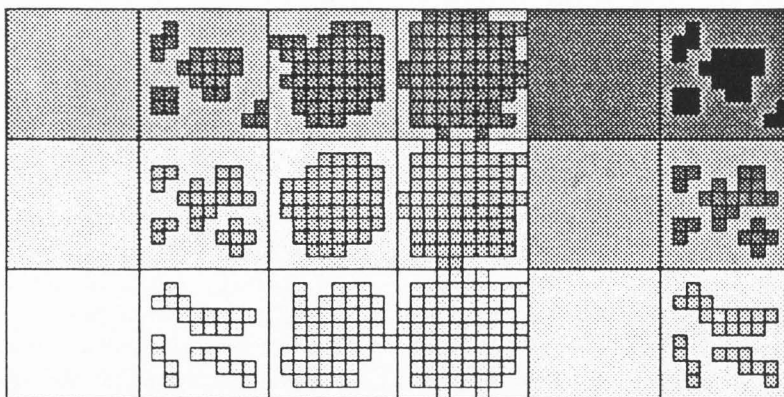
100% Step-flow

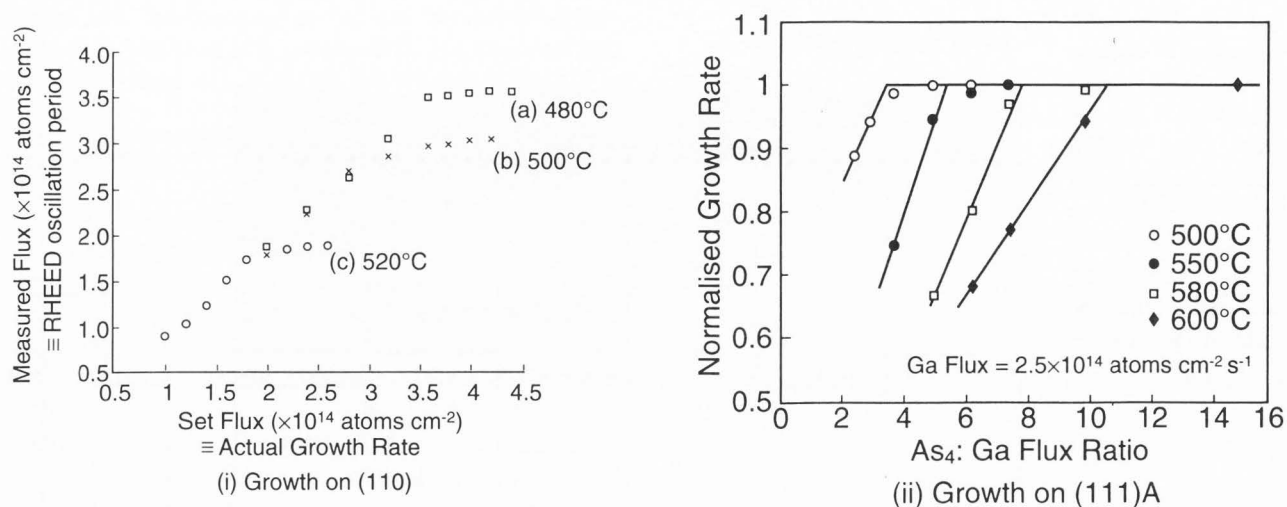


80% 2D-nucleation + 20% Step-flow



100% 2D-nucleation





**Figure 7.** (i) Growth on a GaAs (110) substrate. The apparent Ga flux, as measured from the period of the RHEED oscillations plotted as a function of the actual Ga flux (equivalent to the growth rate) at substrate temperatures of (a) 480°C, (b) 500°C and (c) 520°C at a constant  $\text{As}_2$  flux of  $4 \times 10^{14}$  molecules  $\text{cm}^{-2} \text{s}^{-1}$ . Note that at each temperature an  $\text{As}_2$ :Ga flux ratio is reached where the actual growth rate is determined by the period of oscillations. (ii) Growth on a GaAs (111)-2x2 substrate. The variation of apparent growth rate, derived from the RHEED oscillation period at growth temperature of 500°C, 550°C, 580°C and 600°C as a function of the  $\text{As}_4$ :Ga flux ratio at a fixed Ga flux.

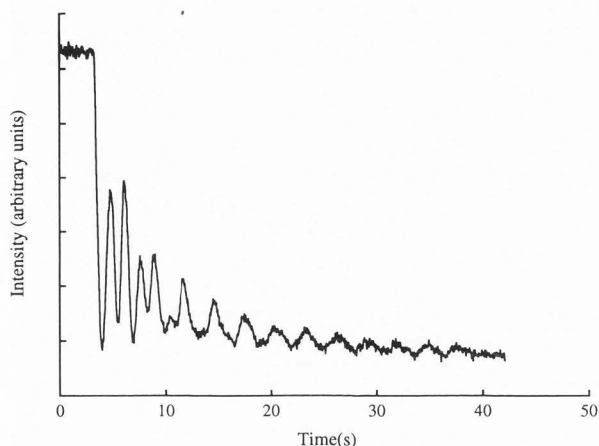
It had indeed been claimed that RHEED oscillations could not be observed at all for conventional growth conditions, but this is certainly not the case, and we will first summarise our recent results [7, 8, 9] for the growth of GaAs on GaAs (110) and GaAs (111)-2x2 substrates cut as close as possible to the singular surface.

Stated simply, the most important point is that in general, the oscillation period no longer provides a measure of the growth rate (i.e., of the Ga flux). The results for the two surfaces are illustrated in Figure 7 (i and ii) for (110) and (111) respectively. The (110) surface does not reconstruct, but the (111) surface shows a 2x2 reconstruction. The behaviour in each case is rather similar. There is a substrate temperature - flux ratio regime in which the oscillation period indicates a growth rate less than the actual value. The true value in each case was shown to be constant and the same as for an (001) substrate mounted next to the (111) and (110) substrates in the MBE system. This was confirmed by growing identical single quantum well structures on each substrate and establishing by cross-section transmission electron microscopy (XTEM) that all thicknesses were the same, even though RHEED oscillations indicated that the (111) and (110) wells should have been thinner. The growth rate is therefore still determined by the Ga flux, but this is not in general measured by RHEED. What then is the origin of RHEED intensity oscillations in this case? We believe it to be closely related to the effect

seen on (001) surfaces where an excess of Ga is present, which generates As-induced oscillations. On both (110) and (111) surfaces there is apparently a much lower reactivity between  $\text{As}_x$  ( $x = 2$  or  $4$ ) and Ga than on (001) surfaces, so that unless As is supplied in very large excess, some free Ga is able to form on the surface, at least during the initial stages of growth, and the oscillations are then As-induced. The critical flux ratio at which real and apparent growth rates become equal increases with increasing temperature, which suggests that the fundamental effect is the very short surface lifetime of arsenic molecules, rather than their direct interaction with Ga.

Dabiran *et al.* [4] have observed identical effects during the growth of GaAs on GaAs (111) surfaces, which they have also attributed to a reduction in Ga incorporation. They were, however, only able to observe oscillations over a very limited range of growth conditions on (111) oriented substrates.

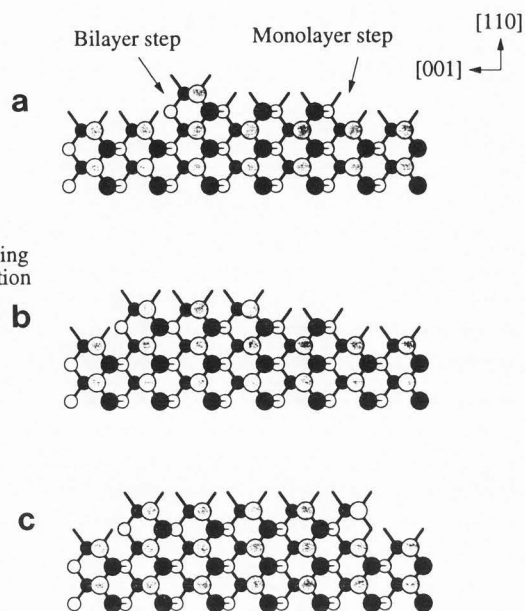
This behaviour has obvious implications for the growth mode, and under conditions where the RHEED oscillations are not measuring the actual growth rate, it is observed that significant surface roughening occurs and the oscillations damp very rapidly. This is consistent with some limited accumulation of Ga on the surface and is perhaps equivalent to growth on (001) substrates under Ga-rich conditions, which also produces poor surface morphology. Provided due allowance is made for the different chemistry, however, it is possible



**Figure 8.** RHEED intensity oscillations in the  $[\bar{1}10]$  azimuth during growth of GaAs on GaAs (110) at 450°C. The beam energy was 14 keV and the polar angle  $\sim 0.3^\circ$ .  $J_{\text{Ga}} = 1.6 \times 10^{14}$  atoms  $\text{cm}^{-2}\text{s}^{-1}$  and  $J_{\text{As}_2} = 4 \times 10^{14}$  molecules  $\text{cm}^{-2}\text{s}^{-1}$ . The ordinate scale is linear.

to grow GaAs films of high morphological perfection on both (110) and (111) GaAs substrates. It is necessary, however, to use lower temperatures to increase the arsenic molecule surface lifetime and high As:Ga flux ratios to increase the population of adsorbed arsenic molecules.

An additional feature of growth on (110) oriented substrates is that under certain conditions there is a transition from monolayer to bilayer (BL) growth after a few ML have been grown [8]. The relevant conditions are low temperature and high arsenic fluxes (high  $\text{As}_2$ :Ga flux ratios), and the evidence is directly from RHEED oscillations where the period is observed to double. A typical example is shown in Figure 8, taken in the  $[\bar{1}10]$  azimuth, but it is important to note that a similar doubling of the period is observed in the [001] azimuth at the same polar angle. This probably rules out the possibility that the changing period is a diffraction effect, which is an important consideration. Horio and Ichimiya [16] have shown that a BL to ML transition can be accounted for in terms of the interference between electron waves reflected from a top BL surface and the underlying BL surface, but that does not seem appropriate here. A possible mechanism for the transition we observe is that at the commencement of growth under As-rich conditions, ML islands nucleate initially, but stable double layer steps can form at the edge of the ML islands, as shown in Figure 9, due to reconstruction-induced energy lowering. As further material is deposited, an island with a double layer step at one



**Figure 9.** Schematic diagram showing the formation of a BL island during growth on a GaAs (110) surface. In (a) a BL step is formed at the edge of a ML island. In (b) and (c) subsequent propagation of the ML step on the uppermost terrace to the edge of the island produces a second BL step and results in the formation of a BL island.

at one boundary will develop into a BL island, since the BL step propagates more slowly than the ML step (twice as many atoms need to be incorporated at the BL step to achieve the same step velocity). Consequently, the ML step will propagate to the edge of the island, where another stable BL step will be formed, and further growth will occur by propagation of these BL steps. It is a necessary condition of this model, however, that ML islands continue to nucleate as the precursors of BL island growth.

### Growth of Mismatched Structures

It will be convenient to treat the GaAs/InAs system as typical. The extent of misfit is comparatively large, 7%, so the degree of strain rapidly becomes high. If we consider first the growth of InAs on GaAs (001), the behaviour follows a fairly conventional pattern. At most, the first ML is formed by a process of 2-D nucleation but after that the growth rapidly becomes 3-D, nominally following the Stranski-Krastanov mode. There is evidence, however, that even the first ML does not retain its integrity with subsequent growth, but that during that process it breaks up into 3-D clusters of variable thickness [5]. As far as RHEED oscillations are concerned,

essentially only a single period is seen following the initiation of growth. Growth is initially pseudomorphic, but strain relief begins at or before the completion of the first ML with the formation of misfit dislocations and is largely isotropic. The change in growth mode has been equated with the onset of strain relief, i.e., when the so-called critical thickness has been exceeded.

If we now consider growth on (110) oriented substrates, we find the behaviour to be completely different from that on (001), despite there being the same misfit. The first major difference is that growth occurs in a 2-D layer-by-layer mode over a large range of film thicknesses (from 1 to  $> 500$  ML), although there is probably no upper limit. RHEED oscillations are sustained during growth, and both plan view and cross-section TEM confirm the growth mode to be layer-by-layer [37, 38].

The most effective mechanism of strain relief in semiconductor systems with a large mismatch is through the generation of Lomer-type dislocations. On a (001) surface, the Lomer dislocations of two orthogonal Burgers vectors ( $b = a/2[110]$  and  $a/2[1\bar{1}0]$ ) can nucleate easily so long as the deposit has an island structure. Furthermore, strain relief is more or less isotropic. This is completely consistent with the 3-D growth mode on this orientation. On the (110) surface, however, only one set of Lomer dislocations satisfying the strain relief requirement can be generated. They are along  $[001]$  and have  $b = a/2[1\bar{1}0]$ , relieving strain in the  $[1\bar{1}0]$  direction. These dislocations have to be nucleated before the 2-D islands in the first InAs ML coalesce to form a continuous film. RHEED observations of lattice parameter change have indicated that within experimental error all strain in the  $[1\bar{1}0]$  direction is indeed relieved during the formation of the first ML.

Relaxation in the orthogonal  $[001]$  direction has to rely on generation of  $60^\circ$  dislocations which glide along the inclined (111) and  $(1\bar{1}\bar{1})$  planes, resulting in a dislocation line direction of  $[110]$  and  $b = a/2\langle 101 \rangle$  types. Since strain relief occurs more easily in the  $[110]$  direction by the generation of Lomer dislocations, the initial strain relief is hugely anisotropic. The microstructure of InAs on GaAs is illustrated schematically in Figure 10.

The important point, however, is that with the same degree of strain, growth modes are completely different in (001) and (110) oriented films, which means that there is no simple relationship between growth mode and strain.

At this stage, it is only possible to speculate on the reasons for this difference, or more fundamentally, what physical parameter controls the growth mode. One observation we have made perhaps provides a clue. This involved the growth of InAs on a vicinal GaAs (110) substrate, misoriented by  $1.5^\circ$  in the  $[110]$  direction.

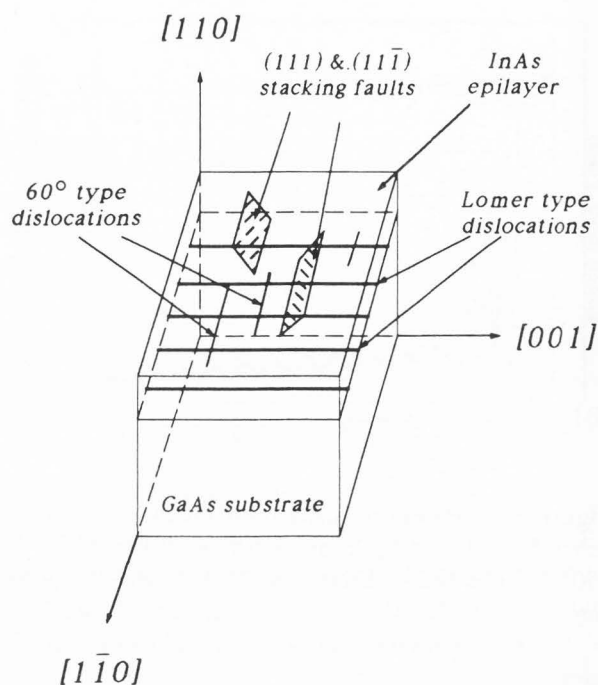
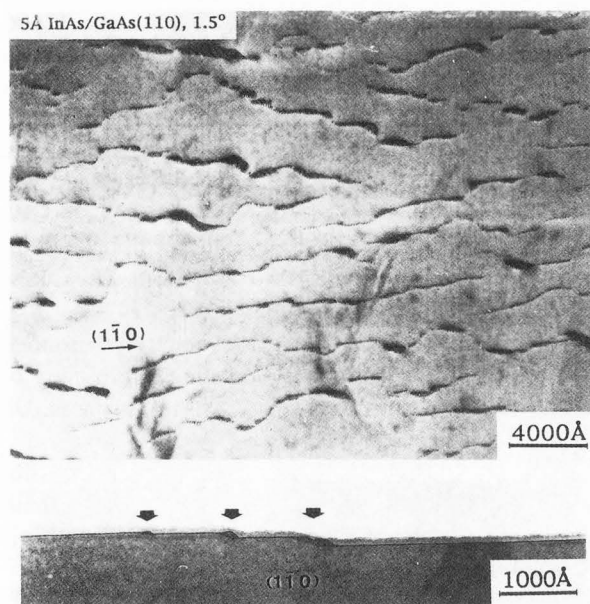


Figure 10. Schematic illustration of the microstructure of InAs layers on GaAs (110).

This produced steps lying along  $[1\bar{1}0]$ , but instead of being ML steps, as in the case of (001), substantial bunching occurred so that the separation was of the order of  $2000 \text{ \AA}$ , rather than  $100 \text{ \AA}$  had the steps been of ML height. Nevertheless, growth of InAs films of nominal thickness from  $5$  to  $60 \text{ \AA}$  resulted solely in decoration of the widely spread macro-steps by the deposit, as shown in Figure 11. There was no evidence of 2-D growth on the terraces even for the comparatively low substrate temperature of  $420^\circ\text{C}$ . A possible interpretation is that In adatoms are extremely mobile on this surface; it is this mobility which determines the growth mode, not the presence of strain in the system. The more general implication is that the growth mode is determined by kinetic factors rather than strictly thermodynamic considerations, although some interplay may well occur. This would suggest that growth modes could be controlled by easily accessible experimental parameters such as growth rates, flux ratios and temperatures. There is some minimal evidence for this in work by Schaffer *et al.* [25], who claim that even on GaAs (001) surfaces, a more 2-D mode can be imposed on InAs growth by using very cation-rich growth conditions. The complete generality of this kinetics vis-a-vis thermodynamics concept is, however, very far from being established.

In this paper, we have shown that considerable caution must be exercised in the interpretation of RHEED



**Figure 11.** Plan-view and cross-section transmission electron micrographs showing the decoration of macroscopic steps on a GaAs (110) surface by InAs. The nominal thickness of the InAs layer was 5 Å.

intensity oscillation data. The period only provides a direct measurement of the ML growth rate under rather ideal conditions and there are many examples where major differences occur. These include growth under group III rich conditions, growth on vicinal planes and growth from pulsed beam sources (MEE). On non-(001) low index orientation substrates, the reduced reactivity between, for example, Ga and As compared to that on (001) surfaces, means that growth effectively takes place under Ga-rich conditions, so the oscillations are As-induced and do not equate with the growth rate. The origin of RDS oscillations during growth is still obscure. Finally, we have also shown, by comparing the growth of InAs on GaAs (001) and (110) oriented substrates, that there is no simple relationship between strain relief and growth mode in strained layer systems, but that the effects which determine the growth mode are probably dominated by adatom mobility.

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**Editor's Note:** All of the reviewer's concerns were appropriately addressed by text changes, hence there is no **Discussion with Reviewers**.