

Realization of In-Situ Sub Two-Dimensional Quantum Structures by Strained Layer Growth Phenomena in the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ System

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We confirm that as the misfit strain in pseudomorphic epitaxial layer increases, surface thermodynamics controlled growth modes can change from a layer-by-layer to a three-dimensional (3-D) island mode. Both in-situ reflection high energy electron diffraction studies and in-situ scanning tunneling microscopy studies are utilized to demonstrate this transition to 3-D growth. This concept allows one to grow $\text{GaAs}/\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ heterostructures where the electrons in $\text{In}_x\text{Ga}_{1-x}\text{As}$ are possibly confined in lower dimensions.

Key words: InGaAs/GaAs, reflection high energy electron diffraction (RHEED), scanning tunneling microscopy (STM), strained layer growth

INTRODUCTION

Epitaxial growth techniques such as molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD) routinely provide two-dimensional (2-D) electronic systems by allowing fabrication of quantum wells. The realization of quantum wells is possible because of the layer-by-layer growth mode that is achievable under certain growth conditions.^{1,2} Based on thermodynamic considerations, the growth modes of an epilayer growing on a substrate can be classified as:

- Frank-van der Merwe (layer-by-layer);

- Vollmer-Weber (three dimensional islands [3-D]); and
- Stranski-Krastanov (where growth changes from layer-by-layer to 3-D island mode).³

The layer-by-layer growth mode has proven to be ideal for the growth of heterostructure systems.

Attempts to realize lower dimensional electronic systems are increasing, partly fueled by theoretical calculations of the superior electronic and optical properties of such structures.^{4,5} In this study, we have examined the potential of exploiting molecular beam epitaxy, under conditions where free energy considerations require a three-dimensional island surface structure, to realize "quantum boxes". In lattice matched systems, it is possible to achieve 3-D island growth, but this surface structure is random and leads to broadening or scattering rather than uniform sub 2-D confinement. For this reason, we use the strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ system. Since InAs and GaAs have a 7% misfit, we expect that as In composition in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ epilayer is increased, strain

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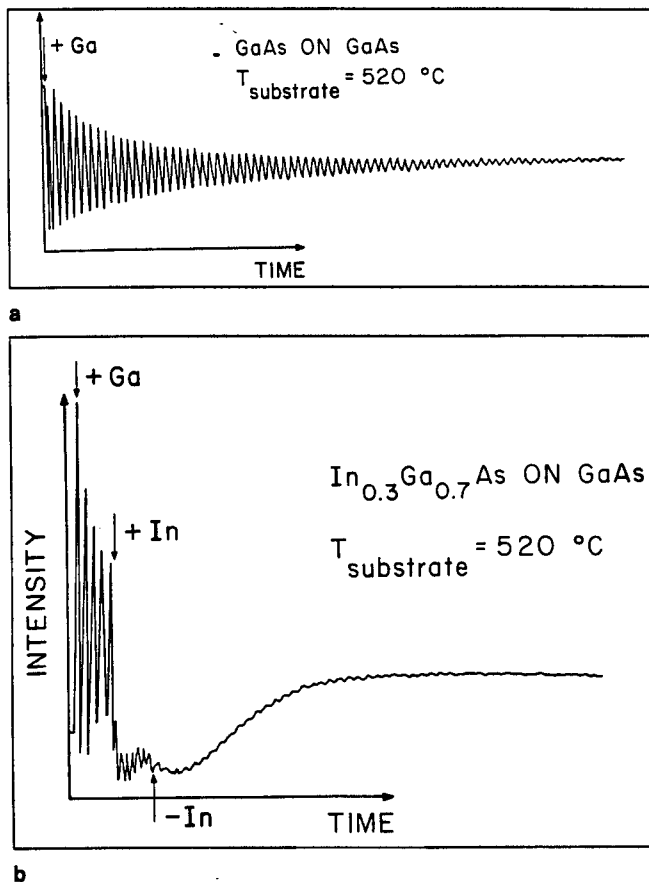


Fig. 1. RHEED oscillation data for MBE growth of (a) GaAs on GaAs, and (b) pseudomorphic $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$ on GaAs.

energy will dominate the growth modes in the pseudomorphic (no dislocation) regime. We have earlier discussed the possibility of a transition from a free energy minimum state which is atomically flat to one which has 3-D surface islands.⁶⁻⁹ In Fig. 1, we show in-situ reflection high energy electron diffraction (RHEED) oscillation data for lattice-matched and strained epitaxy on GaAs substrates. As can be seen in Fig. 1a, the oscillations persist due to the layer-by-layer growth mode; while during strained epitaxy, there is a much weaker specular reflection and a much weaker oscillation amplitude in which the surface smoothness does not recover, suggesting a 3-D growth mode. According to the free energy minimization criteria for (100) surface used by us,⁶ we have shown that an island height of n (in terms of monolayers) given by

$$n^3 \cong 2 \frac{W_1}{W_2} \left(\frac{R_0}{d_c} \right) \quad (1)$$

should occur, where d_c is the critical thickness and W_1 and W_2 are the nearest and second neighbor bond energies. In the InGaAs-on-GaAs systems, we expect a 2-D to 3-D surface transition when the In content exceeds $\sim 20\%$. To provide direct evidence of the growth modes, we have carried out in-situ scanning tunneling microscopy (STM) studies of the strained growth

of InGaAs on GaAs substrates. We have also made photoreflectance measurements for the confined structure which show high quality signals, signifying the fairly ordered nature of the 3-D growth.

MOLECULAR BEAM EPITAXIAL GROWTH

In this section, we describe the growth conditions for carrying out in-situ RHEED and STM measurements and ex-situ optical studies. For the STM measurements, MBE growth was carried out in an ultra high vacuum (UHV) system equipped with in-situ STM and RHEED. This system allowed us to obtain reproducible STM images of the various phases of InGaAs on GaAs (100) growth. Starting with a p^+ GaAs (100) substrate, a 400 nm nominally undoped GaAs buffer layer was grown at 570°C under arsenic-rich conditions. The growth was then interrupted, and the substrate temperature was lowered to 520°C and a few more monolayers of GaAs were grown. The growth of these layers was terminated by shuttering the Ga source, turning off the As furnace, and then slowly cooling the sample to room temperature under a decreasing As flux. The quality of the (2×4) RHEED pattern was used to manually control the rate of cooling. After 20–40 min of cooling, the pressure in the MBE chamber reached the 10^{-10} Torr range and the sample was transferred to the STM chamber. This growth procedure produced a buffer layer surface that was ordered and atomically flat, with only an occasional kink or step, for areas up to $100 \text{ nm} \times 100 \text{ nm}$. Having grown the buffer layer in this manner, we then grew a four monolayer $\text{In}_{0.35}\text{Ga}_{0.65}\text{As}$ film at a substrate temperature of 520°C, starting with a RHEED maximum. The growth was monitored by RHEED. Before the indium shutter was opened to grow the ternary compound, we observed strong specular oscillations and a sharp, streaked (2×4) pattern. Upon opening the In shutter, the pattern changed immediately to a streaked, diffuse (1×1) pattern¹⁰ and much weaker oscillations were observed. Simultaneous measurement of the integral order streak spacing showed a gradual 0.7% increase in the surface lattice constant up to the thickness of four monolayers at which point the growth was interrupted. A shuttering of the In and Ga sources, followed by (within 0.5 s) a reduction of the substrate temperature to 425°C, and enclosing the sample in a liquid nitrogen cooled shroud enabled us to preserve the (1×1) pattern and the lattice relaxation.

For an optical study of growth uniformity and for the observation of excitonic states, in these pseudomorphic layers, MBE growth of the heterostructures was carried out on (100) semi-insulating or n^+ (silicon doped) GaAs substrates. Growth was initiated with a 30 period undoped GaAs (20Å) / $\text{Al}_{0.30}\text{Ga}_{0.70}\text{As}$ (20Å) superlattice buffer followed by 2000Å of GaAs. These layers were grown at 620°C. This was followed by the growth of $\text{In}_x\text{Ga}_{1-x}\text{As}$ ($0.1 \leq x \leq 0.3$) at 520°C, at a rate $\leq 1.0 \mu\text{m/h}$. This temperature was chosen since our studies show that under these conditions, the system should be able to reach surface thermodynamic equi-

librium. The thickness of the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer was varied from 20 to 55 Å, keeping the total thickness below the critical thickness, in order to consistently compare with STM results. The substrate temperature was further lowered to 400°C and a 1500 Å GaAs layer was grown. The low substrate temperature was chosen to reduce the surface mobility of the In adatoms and reduce In interdiffusion. The top GaAs layer was also grown to provide quantum confinement heterostructures and to prevent surface depletion of the pseudomorphic layers.

EXPERIMENTAL RESULTS

Scanning Tunneling Microscopy Studies

Shown in Fig. 2a is a typical STM image of buffer layer surface. The dark rows running diagonally across the image in the $[\bar{1}10]$ direction are spaced 1.6 nm apart, corresponding to the 4x spacing. These are the rows of missing As dimers. Higher resolution, smaller

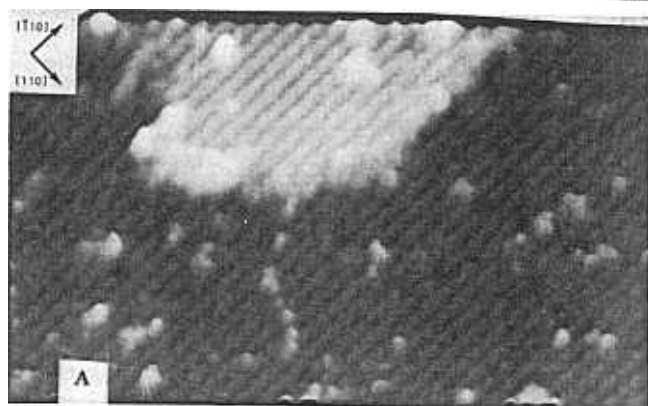


Fig. 2. (a) STM image of the surface of a GaAs (100) buffer layer. The tunneling voltage was 2.8 V, applied to the sample, and the tunneling current was 70 pA. The As terminated surface shows rows of missing As dimers along the $[\bar{1}10]$ direction that are spaced 1.6 nm apart, corresponding to the 4X spacing. The corrugation is 0.2 nm, and (b) 90 nm \times 50 nm STM scan of the surface after four monolayers of $\text{In}_{0.35}\text{Ga}_{0.65}\text{As}$ have been deposited. The crystal axes are the same as in (a). The tunneling voltage was 3.0 V, applied to the sample, and the tunneling current was 30 pA. The surface is covered with islands roughly 3.2 nm in diameter and with an average corrugation of 0.6 nm. Examination with RHEED shows this surface to have a diffuse (1×1) pattern and a 0.7% relaxation of the surface lattice constant.

scale images show the expected coexistence of the (2×4) and $c(2 \times 8)$ reconstructions.^{11,12}

Figure 2b shows a typical STM image of the surface of this InGaAs film. Note the 3-D character of the surface even in the pseudomorphic phase of growth. We note that Fig. 2b represents a typical growth front observed under high strain (In content $>20\%$) conditions, which is independent of the phase of RHEED oscillations at the start of InGaAs growth. It is important to observe that although there is some disorder in the sizes of the InGaAs islands, it appears that they have a fairly narrow size distribution. Theoretically, we expect the free energy minimum conditions to lead to islands with uniform sizes if entropy considerations are ignored. Although STM can only examine fairly small regions of the substrate, within these regions the root mean square (RMS) height of the islands is ~ 0.6 nm, and on the average their lateral extent is $\sim 3.2 \pm 0.5$ nm. The uniformity of these island sizes on a larger scale is confirmed by photoreflectance (PR) and piezoreflectance (PzR) studies to be described later.

Optical Studies

Initial investigation of the optical properties consisted of low temperature photoluminescence (PL) measurements. Since $\text{In}_x\text{Ga}_{1-x}\text{As}$ has a lower bandgap than GaAs, we expect confined states in this region. Shown in Fig. 3 is the PL spectra of the GaAs/ $\text{In}_{0.30}\text{Ga}_{0.70}\text{As}/\text{GaAs}$ heterostructure with a 3.5 nm pseudomorphic well. A peak with a linewidth of ~ 13 meV is observed. The energy position of the emission confirms that it originates from the $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer

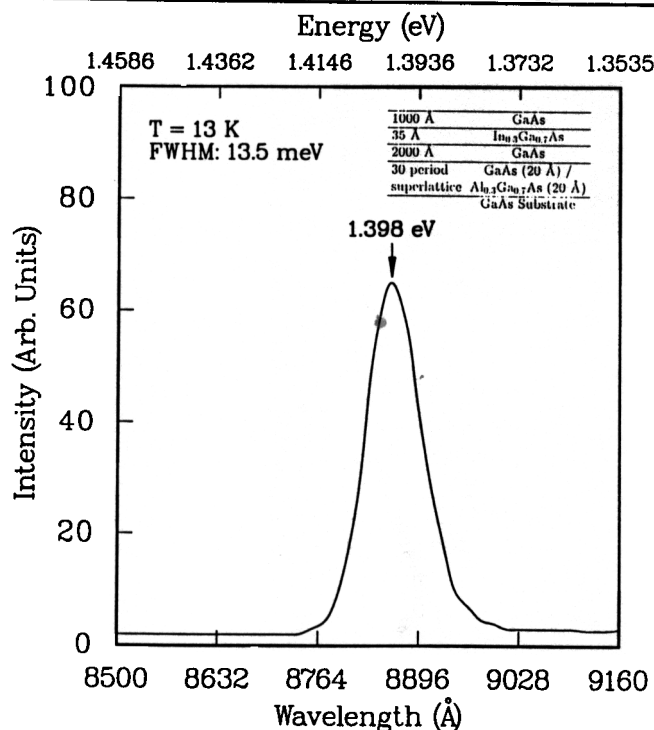
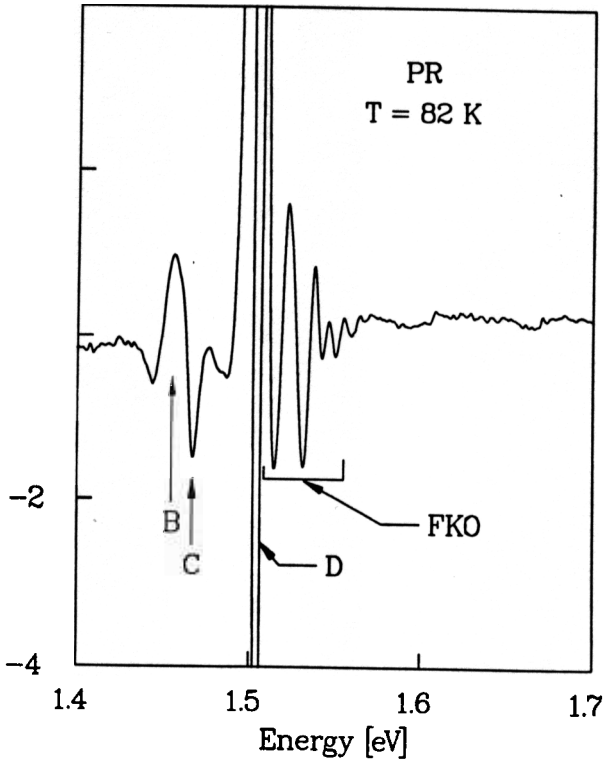
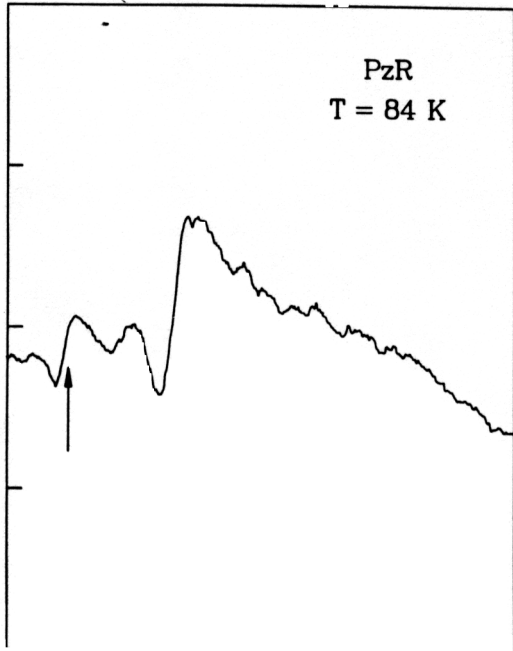


Fig. 3. Low temperature photoluminescence spectra of GaAs/ $\text{In}_{0.30}\text{Ga}_{0.70}\text{As}/\text{GaAs}$ pseudomorphic heterostructure. Inset shows the structure of the grown layer.



b

Fig. 4. Piezoreflectance (a) and photoreflectance (b) data measured at 84 and 82K, respectively, for the GaAs/In_{0.3}Ga_{0.7}As/GaAs pseudomorphic heterostructure.

and we believe it is excitonic in nature.

To shed further light on the spectra, we used modulation spectroscopy techniques to probe the electronic