InAs/InP (001) quantum wire formation due to anisotropic stress relaxation: *in situ* stress measurements.

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Abstract

We report *in situ* and in real time quantitative measurements of stress along $[110]$ and $[1\bar{1}0]$ directions during the formation of InAs/InP(001) quantum wires (QWr) and consequent stress relaxation. Results show a strong stress anisotropy due to the distortion of As-In bonds along $[110]$ and As-As dimerization along $[1\bar{1}0]$. This anisotropy is claimed to be the origin of QWr formation instead of self-assembled quantum dots.

Anisotropic stress relaxation associated to QWr formation is shown to be characteristic of heteroepitaxial systems involving different group V elements grown by MBE under group V stabilized surface (2x4 reconstruction).

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Self-assembled InAs nanostructures grown on InP (001) are promising candidates for optoelectronic devices operating at $\lambda = 1.3$ and $1.55 \mu\text{m}$ wavelengths. Recently [1-3] it has been shown that is possible to obtain very uniform arrays of quantum wires (QWr) by solid source molecular beam epitaxy (MBE) deposition of InAs on InP. Other epitaxial techniques (MOVPE, CBE and GSMBE) generally provide well behaved standard quantum dots (QD) in this system. Surface effects and As-P exchange reactions on the surface [4,5] must be decisive to control the development of self-assembled nanostructures.

It is generally accepted that accumulated elastic stress ($\Sigma\sigma$) relaxation is the ultimate driving force for self-assembling of QD in highly mismatched epitaxial systems. However, only few works on *in situ* $\Sigma\sigma$ measurements during growth of SiGe/Si [6] and III-V compounds [7,8] have been performed that could provide direct experimental evidence of stress relaxation as a consequence of the formation of 3D nanostructures. In this paper we study InAs QWr formation by *in situ* and in real time monitoring of $\Sigma\sigma$ evolution. During the growth process we observe a large in-plane stress anisotropy that is responsible for the formation of QWr instead of Q-dots.

Furthermore, using also direct stress measurements we consider the As/P exchange process during As_4 exposure of the InP (001) surface. This process is unavoidable during the initial stages of InAs deposition at the temperatures of interest.

Our technique to measure, *in situ* and in real time, surface and interface stress evolution with extremely high sensitivity is based on the detection of laser deflection on a cantilever shape cut InP (001) substrate [7]. Epitaxial stress ($\Sigma\sigma$) can be quantitatively determined from the cantilever bending by means of Stoney's equation [9] that relates radius of curvature with elastic constants and set-up geometry. It is important to notice that this technique measures changes of $\Sigma\sigma$ from an initial tensile/compressive state. The zero is arbitrarily set.

Growth was carried out by solid source MBE. InP (001) substrates, cut along [110] and $[1\bar{1}0]$ directions, were back side mechanically polished to $80\text{-}100 \mu\text{m}$ thickness in order to prepare cantilevers, and In free clamped on the MBE substrate holder. Growth temperature (T_s)

was carefully determined by measuring desorption times of phosphorous surface by means of *in situ* reflectance anisotropy (RAS) signal [10,11]. Growth rate and 2D-3D transition were checked as usual by means of Reflection High Energy Electron Diffraction (RHEED).

Before InAs deposition, a 200 nm thick InP buffer layer is always grown in the 2x4 surface reconstruction at $T_s = 460^\circ\text{C}$ and under a P_2 flux (beam equivalent pressure $P_2\text{-BEP} = 5 \times 10^{-6}$ mbar) from a valved cracker solid phosphorous cell. Then, cantilever temperature is increased to 480°C under P_2 flux. In order to start growth, P_2 flux is interrupted during 2 s, and after that surface is exposed during 3 s to an arsenic flux $\text{As}_4\text{-BEP} = 2.5 \times 10^{-6}$ mbar. Then 2.5 InAs monolayers (ML) are deposited at a growth rate of about 0.7 ML s^{-1} . Finally, the apparition of a characteristic 3D RHEED pattern just after growth interruption and later *ex situ* AFM characterization confirms QWr formation.

Figure 1 shows $\Sigma\sigma$ evolution for both $[110]$ and $[1\bar{1}0]$ directions cut cantilevers. Stress evolution is clearly anisotropic. In the $[110]$ direction, after an initial $\Sigma\sigma$ transient associated to the exposure to As_4 flux followed by a linear increase corresponding to InAs deposition, a clear stress relaxation is observed in coincidence with the development of 3D RHEED pattern when growth is interrupted. Relaxation amounts $\sim 20\%$ value of the total accumulated stress during growth. On the other hand, along $[1\bar{1}0]$ no initial transient is observed and there is no measurable $\Sigma\sigma$ relaxation after growth interruption.

From the slope of the linear increase of $\Sigma\sigma$ (nearly identical for $[110]$ and $[1\bar{1}0]$ cantilevers except for small variations in the growth rate on successive growth runs), a totally isotropic mismatch stress of 0.9 Nm^{-1} per InAs monolayer can be extracted. This is in good agreement with the estimated value ($0.78 \text{ Nm}^{-1} \text{ ML}^{-1}$) using bulk elastic constants for InAs/InP.

However, the total accumulated stress at the onset of 2D/3D relaxation is clearly different for $[110]$ and $[1\bar{1}0]$ directions as a consequence of the large surface stress transient associated to the As_4 exposure, with an amplitude of the order of 1.2 Nm^{-1} . This strong surface stress anisotropy is clearly the origin of the asymmetric relaxation of total stress through the formation of 3D linear features aligned along $[1\bar{1}0]$ direction.

On figure 2 a topographic image by AFM of the QWr grown on the cantilevers is shown. A periodicity of 20 nm and 1.5 nm peak to valley amplitude can be obtained from the AFM profile.

Due to the important role of the surface stress associated to the As/P surface exchange process in QWr formation, we have carried out a detailed study of this process under a wide range of substrate temperatures. We have used both our sensitive laser deflection technique and RAS, in order to obtain simultaneous information on surface dimer population.

As/P exchange experiments during As/P switching have been performed for a range of cantilever temperatures between 370 °C and 480 °C. A detailed study of the relationship between surface stress and surface coverage of P and As dimers as determined by simultaneous RAS measurements will be published elsewhere. Its most relevant result in respect to this work is that, within the range of surface temperatures investigated, during As/P switching impinging arsenic ($\text{As}_4\text{-BEP} = 2.5 \times 10^{-6}$ mbar) does not actively displace P from the surface. Instead, it bonds on the available In sites left vacant by phosphorous desorption at the corresponding temperature, reaching an As surface coverage that is therefore determined by substrate temperature and arsenic flux.

Figure 3 is an example of the $\Sigma\sigma$ behavior as a consequence of As/P exchange during As/P switching. We have chosen the experiments performed at cantilevers temperature $T_s = 430^\circ\text{C}$ for a clearer description of the processes involved. A positive change of $\Sigma\sigma$ along $[1\bar{1}0]$ when P_2 flux is switched off is observed. This is the expected $\Sigma\sigma$ behavior [7] as the $2\times 4\text{-InP}$ surface is compressively strained due to surface P dimerization in $[1\bar{1}0]$ direction. Notice that dimerization distorts the P-In bonds by trying to reduce P-P interatomic distance, originating a compressive negative sign $\Sigma\sigma \approx 0.5 \text{ Nm}^{-1}$ stress. In absence of P_2 flux, in the range of temperatures under study, phosphorous desorbs from the surface and consequently P-P dimer density decreases, producing a reduction in compressive stress. Later, when As_4 flux is switched on, As-As dimers rapidly form on the highly reactive In terminated surface (figure 4a). As a

consequence a fast increase of compressive stress, responsible for the negative sign peak, is observed (fig. 3).

Along [110] changes in P-P dimer population do not contribute to $\Sigma\sigma$ when P_2 is switched off. However, $\Sigma\sigma$ behavior along [110] under As_4 flux reflects the apparition of a large surface tensile stress of 1.2 Nm^{-1} . At the interface between InAs and InP, In atoms are bonded to P at the bottom side and to As at the top side. Due to the 3.2% InAs/InP lattice mismatch, the In-As bonds are distorted to accommodate to the InP lattice parameter, and this angular distortion of highly directional covalent bonds originates a tensile stress in [110] (see fig. 4b).

An isotropic tensile contribution to the observed stress cannot be discarded, originating probably on the formation of monolayer InAs islands on the As terminated surface as a consequence of the reaction of As_4 with free In left on the surface by previous phosphorous desorption. This isotropic contribution must be responsible for the progressive shift towards positive values of both [110] and $[1\bar{1}0]$ stress evolution (fig.3). This isotropic tensile stress component will add to the tensile stress along [110] and is probably an important fraction of the large total stress value observed (1.2 Nm^{-1}) in this direction. On the contrary, on the $[1\bar{1}0]$ direction, this positive sign isotropic stress counteracts the negative sign (compressive) stress due to As-As dimers, giving rise to a resultant small total stress.

On figure 3 we plot also the strain behavior of the InP cantilevers under a longer time As_4 exposure and its recovery after switching back to P_2 flux (notice change in time scale). We would like to point out the reversible character of the As/P exchange process, shown by the coincidence of the initial and final strain levels.

An important consequence can already be drawn from present observations: the usual concept of critical thickness for 2D/3D transition during the formation of QD does not strictly apply in the case of InAs/InP QWr formation. Instead, the concept of anisotropy of total stress, including an important surface stress component, has to be considered. Consequently, relaxation takes place in one direction only. This process is characteristic of MBE growth mode under group V stabilized (2x4 reconstruction) surface for heteroepitaxial systems with different group

V elements. Growing under other surface conditions, MOCVD, CBE, and GSMBE, results in the usual QD formation.

In summary, using *in situ* strain induced laser deflection technique, we obtain experimental results that explain QWr formation. Associated anisotropic stress relaxation is shown as a characteristic of heteroepitaxial systems involving different group V elements grown by MBE under group V stabilized conditions.

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Figure captions:

Figure 1. *In situ* stress evolution during InAs/InP(001) growth. $\Sigma\sigma$ is larger along [110] direction than along the $[1\bar{1}0]$ direction. Relaxation of $\Sigma\sigma$ can be observed only along [110] due to quantum wire (QWr) formation.

Figure 2. 250x250 nm² atomic force microscopy (AFM) topographic image of InAs quantum wires (QWr) grown on an InP(001) cantilever. A periodicity of 20 nm and 1.5 nm peak to valley amplitude can be obtained from the AFM profile along [110] direction.

Figure 3. *In situ* stress evolution during As/P switching. A different $\Sigma\sigma$ behavior for cantilevers cut along [110] and $[1\bar{1}0]$ is observed. The recovery of $\Sigma\sigma$ after switching back to P₂ flux is also shown (notice the change in time scale).

Figure 4. Schematic view of InAs/InP(001) interface along $\langle 110 \rangle$ directions. a) As-As dimer formation induces compressive stress along the $[1\bar{1}0]$ direction; b) distorted In-As bonds due to 3.2% InAs/InP lattice mismatch produce a tensile stress along [110].

Figure 1 J.M. García et al.

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Figure 4 J.M.García et al. J. Crys. Growth