LATTICE-MATCHED AND MISMATCHED QUANTUM BOXES FABRICATED VIA SIZE-REDUCING GROWTH ON NONPLANAR PATTERNED SUBSTRATES

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The status of the growth and optical behavior of semiconductor quantum boxes realized via size-reducing epitaxical growth on the tops of appropriately patterned mesas in the substrate is reviewed. The first time-resolved cathodoluminescence results on such quantum boxes and some striking recent results of the behavior of the highly lattice mismatched InAs on nanoscale GaAs(001) mesa tops are reported.

1. Introduction

Realization of semiconductor quantum wires and quantum boxes via a variety of methods that range from post-growth nanolithography to purely growth-based is a subject of much activity as it provides opportunities to address challenging issues pertaining to growth, processing, and physical behavior in ultra small structures. In this paper we provide an overview of one particular approach that falls within the category of purely growth control-based approaches to fabrication of quantum boxes[1,2] . This particular approach is based upon the notion of *size-reducing epitaxy* on the top of appropriately chosen mesas, patterned onto substrates via conventional *ex-situ* approaches or via newer *in-situ* approaches but in both cases on length scales significantly larger than the final desired nanoscale features. We will not cover other growth-based approaches, such as growth through windows in mask layers[1,2] on planar substrates or the formation of three-dimensional island based quantum boxes (more popularly called quantum dots) in highly strained epitaxy on planar substrates such as for InAs on GaAs(001)[3], InAs on InP(001)[4], Ge on Si(001),[5] etc. The lattermost approach is the subject of entire sessions at this conference. The approach discussed here has the inherent advantage of realizing spatially regular arrays as compared to the random spatial arrangement of quantum dots due to their random initiation at the earliest stage of formation. However the control of growth on patterned substrates is complex while islands form relatively easily. We will show here that growth of highly lattice mismatched material on nanoscale mesa tops leads to some truly striking behavior revealing exciting physics of strain relaxation and potential self-limiting growth.

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2. Size -Reducing Epitaxy and Lattice-Matched Quantum Boxes

The basic idea of size-reducing epitaxy as a means of creating quantum boxes, described in detail in Ref. 1 and 2, derives from the work of a few different groups, starting from Tsang and Cho[6], who reported evolution of different growth profiles for growth on long mesa stripes of [110] and/or $[1\bar{1}0]$ orientation on GaAs(001) substrates[1]. Of these, growth profiles exhibiting mesa width reduction were exploited to create wire-like structures on stripe mesas[7,8]. Rajkumar et al observed that, to achieve quantum boxes, equivalent rates of size-reduction during growth from all sidewalls is necessary and this should be most easily testable for growth on mesas on GaAs(111)B substrates due to the inherent 3 fold symmetry of the triangular base truncated pyramidal shape with {100} sidewalls realized in conventional wet chemical etching[9]. Growth of GaAs/AlGaAs on such mesas was thus shown[9] to give size-reducing epitaxy. It was found that the pinching of the (111)B mesa occurs by either {110} or {211} planes instead of the as-patterned {100} sidewalls[9]. Photoluminescence and spatially-resolved cathodoluminescence (CL) studies showed good optical quality of quantum wells of shrinking lateral sizes on the mesa tops[9~10].

Following this first demonstration of purely growth controlled quantum box structures (and with good optical quality), attention shifted to the more conventional (001) surface orientation. In order to overcome the difficulty of achieving equal rate of size reduction from all sidewalls revealed by the different observed growth profiles for mesa edges along the [110] and $[1\bar{1}0]$ directions, square mesas with the edges along <100> directions were patterned since for these directions the angle between the As dangling orbitals and mesa edges are equivalent for all four sidewalls. Realization of GaAs/AlGaAs quantum boxes on such square mesas on GaAs(001) substrates, observed via transmission electron microscopy (TEM) studies, was reported by Rajkumar et al [1, 9]. Figure 1 (a) and (b) show respectively a scanning electron microscopy (SEM) picture and a cross-sectional TEM image of such growth. Contemporaneously, Lopez et al[11] reported CL results on GaAs/AlGaAs quantum confined structures grown on similar square mesas. Very

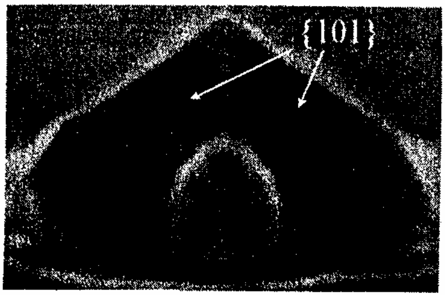

Fig. 1(a)

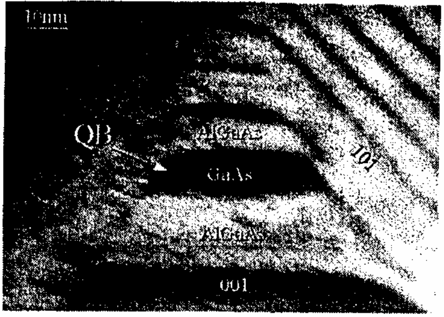

Fig. 1(b)

recently, such growth on *in-situ* patterned square mesas has been reported[12].

The fundamental issue of the physical mechanism driving the cations from the sidewalls to the mesa tops against the expected adatom concentration gradient due to the higher impinging flux on the mesa top over the sidewalls remains unresolved[1,7]. Guha and Madhukar suggested[13] that a driving force for such net migration might be related to step induced stress fields. This is based upon the observation of the evolving side facets, such as (113)/(114)A for the $[1\bar{1}0]$ oriented stripe mesas, providing a train of steps with decreasing separation between steps as one moves away from the mesa tops. Molecular dynamics based calculations of the step-step interaction energies (which are a manifestation of the elastic interactions and hence step-related stress fields) showed opposite signs for the [110] and $[1\bar{1}0]$ orientations[13], thus suggesting a possible connection. Recent simulated annealing calculations[14] showed that for the <100> orientation the sign of the step related elastic interactions is the same as for the $[1\bar{1}0]$ orientation which results in size-reducing epitaxy as mentioned above. We also speculate, however, that if the incorporation time of the cations on the sidewalls such as {111}, {113}, {101} planes is significantly longer than for the (001) planes, then the adatoms on the (001) plane getting incorporated relatively quickly will overcompensate for the higher impinging flux and give an overall lower adatom concentration on the mesa top. This would provide an explanation of the migration from the sidewalls to the mesa top without necessarily invoking surface stress as a contributory factor to the surface inter-facet migration.

Very recently utilizing growth interruption studies, we have found that on stripe mesas oriented along the $[1\bar{1}0]$ direction, the interfacet adatom migration takes place not only during deposition but even when the cation flux is turned off. Figure 2, panel (a), shows the cation shutter sequence, and panels (b) and (c) show TEM images of the mesa profile

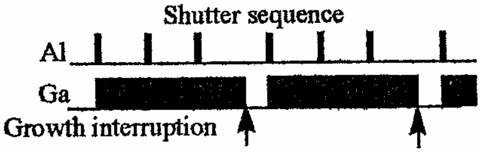

Fig. 2(a)

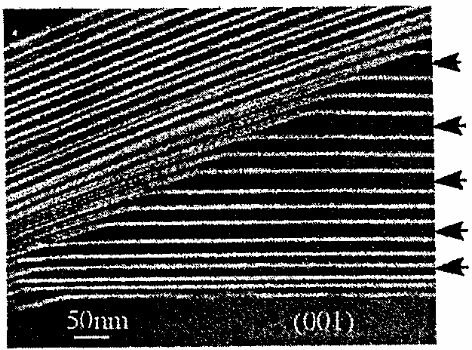

Fig. 2(b)

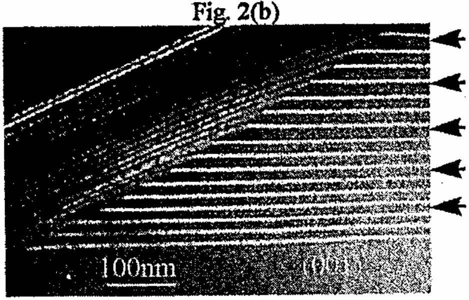

Fig. 2(c)

evolution revealed by AlGaAs markers (white lines in the images) for growths done at 610°C and 590°C, respectively. The arrows indicate the GaAs layers for which growth was interrupted. One can clearly see that such layers on the mesa top have a higher thickness than the preceding or following GaAs layers even though the amount of material delivered is the same. This is only possible if the Ga adatoms continue to migrate from the sidewall to the mesa top after the Ga shutter is turned off. The effect for the sample grown at 610°C is more evident than that grown at 590°C.

Further work needs to be done to shed light on all the above aspects of importance to our understanding of the fundamental aspects of size-reducing epitaxy. A few phenomenological descriptions[15,16] of growth profile evolution in molecular beam epitaxy (MBE) on nonplanar patterned substrates have been introduced in the literature but have yet to reveal the origin of the forces that underlie the observations and provide independent justifications for the assumptions and approximations involved in realizing tractable analytical descriptions of potentially complex phenomena.

An aspect of quantum box behavior of some significance to applications is the kinetics of carrier relaxation in a 3D confined environment as the efficiency of carrier collection in the quantum box determines the quantum efficiency of the quantum box. Issues such as potential phonon bottleneck in an isolated quantum box with its discrete energy states and the associated δ-function like density of states have been discussed in the literature[17]. In real systems there can be means of communication between the quantum boxes and other spatial regions of the structure, such as the sidewalls in the present case, and other wells on the mesa top if included in the growth. To shed light on such aspects, time-resolved CL studies of quantum wells grown on square mesa tops utilizing size-reducing epitaxy have been undertaken by this group for the first time to our knowledge[18]. Figure 3 displays time-delayed CL spectra with various onset (Oi) and decay (Di) time windows for typical GaAs/AlGaAs multiple quantum well structures (as shown in Fig. 1(b)) grown on <100> oriented square mesas on GaAs(001). All the spectra are normalized to have about the same maximum peak height. The monochromatic CL images taken with peaks

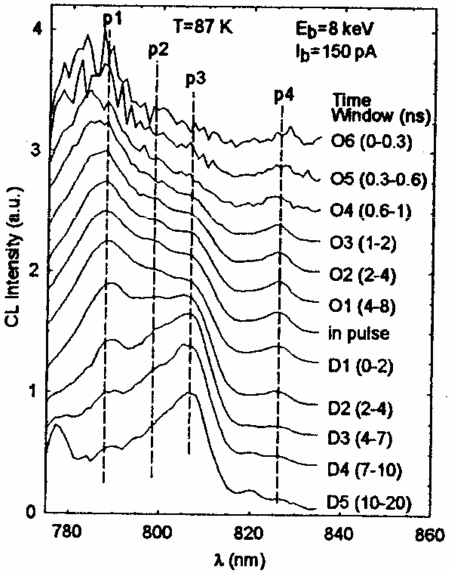

Fig. 3

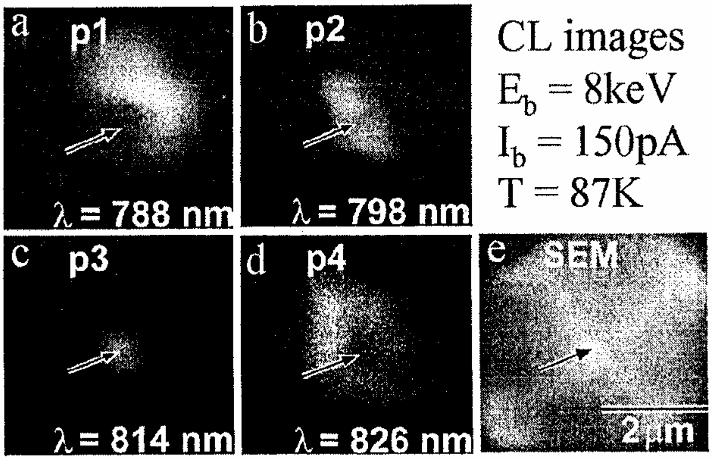

Fig. 4

p1 through p4 (indicated by vertical dashed lines in fig. 3) are shown in fig. 4 (panels a-d) along with the SEM image (panel e) of the mesa. The arrows in panels (a) through (e) denote the mesa top where pinch-off occurred. Emission at p2 and p3 is identified as from the confined structures on the mesa top while emission at p1 is from multiple quantum wells on the sidewalls. Emission at p4 is from the underlying bulk GaAs layers.

In fig. 5(a) are shown the CL decay curves for the peaks p1 through p4 and a linear fit for the initial 6ns of decay. The resulting initial CL decay times of 2.1ns for p1 to 1.7ns for p4 are indicated. The CL onset curves for the first ~2ns, along with the onset rates r(ns^{-1}) for peaks p1 through p4 are shown in Fig. 5(b), and the CL peak intensity ratios for peaks p2,p3, and p4 with peak p1 for onset times ranging from 150ps to 6μs, in fig. 5(c). These results show that relaxation of hot carriers into the largest 3D confined regions occurs on a time scale of a few hundred ps during the onset of luminescence. Moreover, owing to the thermal re-emission from quantum wells, diffusion across the $Al_xGa_{1-x}As$ barriers, and carrier feeding from the thinner quantum wells on the sidewalls, the luminescence decay time also increases for the larger confined regions. Interestingly, the thinner quantum wells on the sidewalls, apart from acting as markers for the grown structure, act as optical markers by providing key signatures in the wavelength and time domain that allow for a visualization of the carrier relaxation.

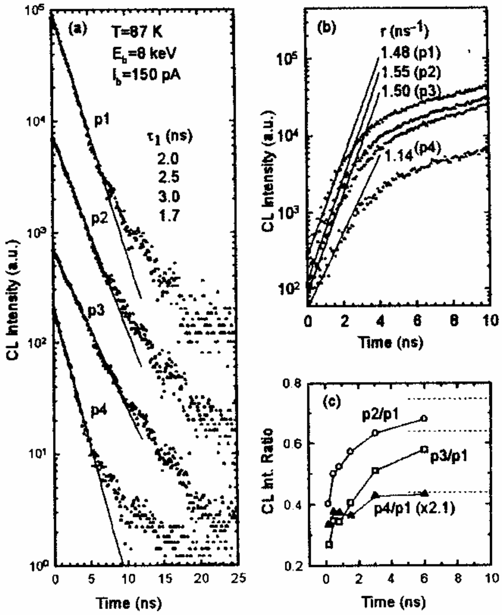

Fig. 5

3. Highly Lattice-Mismatched Quantum Boxes

Konkar et al initiated[19] the first examination of the growth behavior of highly lattice mismatched overlayer on nanoscale (<100nm) mesa top using the InAs/GaAs(001) system. As is well known, on a planar GaAs(001) surface, InAs deposition in excess of ~1.57ML leads to strain-induced formation of 3D islands which act as quantum boxes[3]. There is considerable (~10%) fluctuation in the island sizes and their arrangement lacks regularity. The motivation for examining InAs on GaAs nanoscale mesas is thus two fold: (i) mesas provide spatial regularity, and (ii) strain relief provided by the mesa free-surfaces should lead to a mesa size dependent enhancement in the critical thickness for initiation of 3D islands, in analogy with the enhancement of the critical thickness for misfit dislocation formation at low lattice misfits demonstrated some time ago[20].

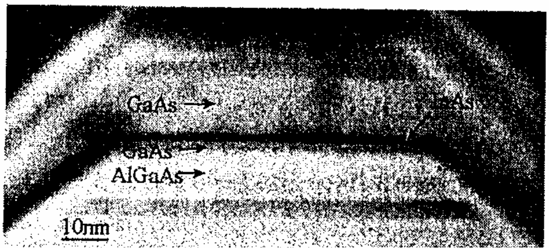

Fig. 6

In fig. 6 is shown a TEM image of InAs on ~70nm wide GaAs(001) square mesa. The delivered amount of InAs was 3.5ML but the measured thickness on the mesa top is (6 ± 1 ML), the enhancement being a consequence of In migration from the sidewall in accordance with size-reducing epitaxy. Note that InAs on the mesa top is planar. No extended defects within the resolution of the TEM were detected. Figure 7 shows the TEM determined InAs thickness on top of 75 ± 5 nm mesas as a function of the delivered amount of InAs. Note the self-limiting growth behavior signified by the thickness saturation value of ~11±1 ML. In all the cases the InAs layer showed flat morphology. Clearly, the strain relief in the nanoscale square mesas is, on one hand, sufficient to prevent 3D island formation and, on the other, it is the build-up of the lattice misfit strain with film thickness which, synergistically and self-consistently, impacts the direction of the mesa profile induced-stress driven interfacet migration. It reverses the migration direction (i.e. makes it become from the mesa top to the sidewall) as the film thickness increases, thus giving self-limiting growth. The good optical quality of such InAs mesa top confined volumes has been established via CL studies. Figure 8 shows a typical CL behavior observed for the mesa with 3.5ML InAs delivery.

This mesa size dependence of the remarkable enhancement in the "critical" thickness for 2D to

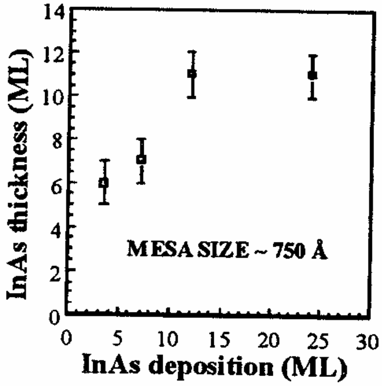

Fig. 7

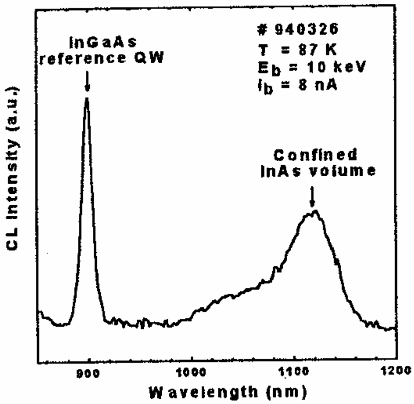

Fig. 8(a)

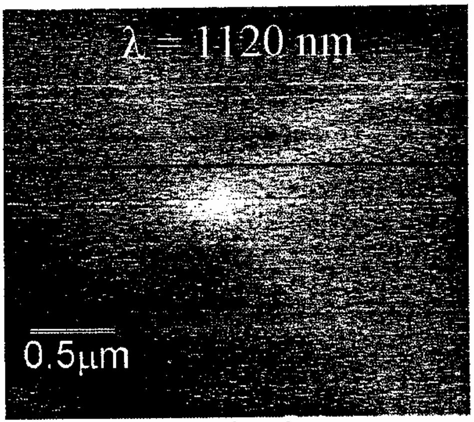

Fig. 8(b)

3D morphology transition and the self-limiting growth behavior are currently under examination in our group. Hobart et al have initiated studies of the SiGe on Si(001) system[21]. We end this section with an example of InAs on a 46nm square mesa (see Fig. 9). The delivered amount was 24ML and the measured thickness is (12 ± 1) ML, still planar and defect-free.

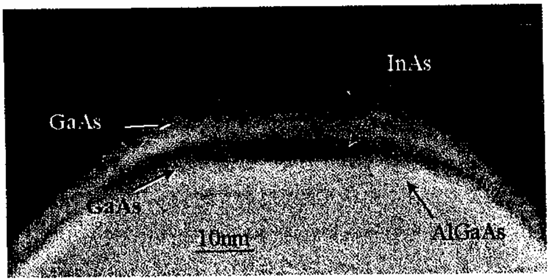

Fig. 9

4. Conclusions

Size-reducing growth of lattice matched and mismatched overlayers on mesa tops is a promising technique for realization of quantum box arrays. Indeed, the first high optical quality quantum boxes were obtained via this approach (as were quantum wires though they were grown in the V-grooves[1] rather than mesa tops). The driving mechanism for size-reducing growth has been suggested to be related to the nature of the surface stress tensor. It is proposed that mesa orientation may be chosen such that the surface stress tensor provides a driving force for adatom migration from the sidewalls to the mesa tops. In this case, the technique has been called *substrate-encoded size-reducing epitaxy* (SESRE) since the directionality of migration can then be engineered into the pattern chosen for the nonplanar patterned substrate. It is seen that such surface stress can be made to act in concert with the lattice mismatch induced stress fields to overcome some of the limitations otherwise imposed by the essentially random evolution of such strain fields at the early stages of strained epitaxy. Thus, high structural and optical quality quantum box structures of highly lattice mismatched combination can be fabricated on nanoscale mesa tops via purely growth control. The dramatic enhancement of the critical thickness for 2D to 3D morphology change and the self-limiting thickness behavior are encouraging features that could be helpful for achieving arrays with size uniformity required for system applications. Much, however, needs to be done, experimentally and theoretically, to both understand the true nature of the phenomena and assess the device potential of such quantum box structures.

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