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Published in:
Applied Physics Letters

DOI:
[10.1063/1.117860](https://doi.org/10.1063/1.117860)

Published: 01/01/1996

Document Version
Publisher's PDF, also known as Version of record

Please cite the original version:
Sopanen, M., Lipsanen, H., & Ahopelto, J. (1996). Fabrication of InGaAs quantum disks using self-organized InP islands as a mask in wet chemical etching. *Applied Physics Letters*, 69(26), 4029-4031.
<https://doi.org/10.1063/1.117860>

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Citation: *Appl. Phys. Lett.* **69**, 4029 (1996); doi: 10.1063/1.117860

View online: <http://dx.doi.org/10.1063/1.117860>

View Table of Contents: <http://aip.scitation.org/toc/apl/69/26>

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Fabrication of GaInAs quantum disks using self-organized InP islands as a mask in wet chemical etching

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(Received 21 June 1996; accepted for publication 18 October 1996)

GaInAs quantum disks are fabricated by wet chemical etching from a GaInAs/GaAs near-surface quantum well using self-organized InP islands as an etch mask. InP islands are formed in coherent Stranski–Krastanow growth mode by metalorganic vapor phase epitaxy. The free-standing GaInAs/GaAs columns, produced by a three-step etching process, are overgrown at 550 °C. The luminescence efficiency per emitting area from the regrown quantum disks is one order of magnitude larger than that from a regrown reference quantum well. © 1996 American Institute of Physics. [S0003-6951(96)00452-4]

Quantum dot (QD) structures having feature sizes below 100 nm and high luminescence efficiency are difficult to fabricate by electron-beam lithography and dry etching techniques because of process-induced damage.¹ To improve the optical quality of the etched QD structures, wet chemical etching is preferred.² Furthermore, to reduce the proximity effect and to enhance the throughput of electron beam lithography, low-energy electrons have been used.³ Several alternatives to electron-beam lithography for mask fabrication have also been developed. Typically, in these methods nanoscale particles are introduced to the surface. For example, polystyrene spheres,⁴ aerosol Ag particles,⁵ CsCl islands,⁶ and Au islands⁷ have been employed. In addition, InAs islands grown in Stranski–Krastanow growth mode have been used as a mask in chloride gas etching.⁸ The advantages of a semiconductor island mask are *in situ* growth and uniform island size. It has been observed in several reports that also InP islands formed on GaAs^{9,10} and GaInP^{11–13} surfaces are uniform. The InP islands can act as stressors and induce quantum dots into an underlying near-surface quantum well (QW).¹⁴ However, in some optoelectronic device structures, e.g., edge-emitting lasers, the active area containing QDs must be far from the surface. If the stressors are overgrown, the QD confinement potential is severely reduced.¹⁵ Therefore, the stressor structure is not feasible for these applications. In this letter, self-organized InP islands are used as a mask to create GaInAs quantum disks from a GaInAs/GaAs near-surface QW by wet chemical etching. The overgrown GaInAs disks exhibit high luminescence efficiency.

Three samples were grown by atmospheric-pressure metal-organic vapor phase epitaxy on epitaxially semi-insulating $\pm 0.5^\circ$ (100) GaAs substrates. The growth was carried out at 650 °C using trimethylgallium, trimethylindium, tertiarybutylarsine, and tertiarybutylphosphine as precursors. Sample A consists of a 200-nm-thick GaAs buffer layer, a 6.5-nm-thick Ga_{0.9}In_{0.1}As QW, and a 10-nm-thick GaAs top barrier layer, on which nominally 3 ML of InP was deposited using a growth rate of 1.5 ML/s and a V/III ratio of 100.

With these growth parameters, uniform InP islands are formed.¹⁰ The reference QW sample B was similar to the sample A except that only 1 ML of InP was deposited. At this coverage, InP islands are not formed. It has been reported that 1 ML of InP passivates the surface states in GaAs increasing the luminescence efficiency of GaAs/AlGaAs surface QWs by a factor of at least 10^5 .¹⁶ We observed that InP passivation improves also the luminescence efficiency of a near-surface GaInAs/GaAs QWs. Sample C was otherwise similar to the sample A, but the GaInAs QW was omitted.

The etching procedure performed on the samples A and C was similar. First, the InP wetting layer was removed by dipping the samples in 1 HCl:3 H₂O for 15 s. The samples were then etched in a solution of 100 citric acid (50%):1 H₂O₂ (30%) for two minutes. The size of the InP islands was not affected by this process, but GaAs and GaInAs between the islands was etched at a rate of about 0.2–0.3 Å/s in accordance with previous results.¹⁷ Finally, the InP islands were removed by etching with concentrated HCl for 1 min. Atomic force microscopy (AFM) was used to image the surface of the samples. The optical properties were investigated by low-temperature photoluminescence (PL) measurements. The PL spectra were measured by a photomultiplier tube or a liquid-nitrogen-cooled Ge detector using standard lock-in techniques. The beam from an argon ion laser (488.0 nm) was focused into a spot having a diameter of 200 μm.

AFM was used to image the samples after the growth and also after each etching step. Figure 1(a) shows an AFM scan from the as-grown sample A. The uniform InP islands have an average height of 24 nm and an areal density of $1 \times 10^9 \text{ cm}^{-2}$. The islands in the as-grown sample C were identical to the sample A. No islands were observed in the sample B. Figure 1(b) shows an AFM scan from the sample A after the first etching step. The average height of the InP islands has been reduced to 12 nm by the HCl-based etch, but the density of the islands has remained at $1 \times 10^9 \text{ cm}^{-2}$. The island height variance is the same as in the as-grown sample, indicating remarkably homogeneous etching of the islands. Without this first step, i.e., removal of the wetting layer, the citric-acid-based etch had almost no effect on the structure. Figures 1(c) and 1(d) show AFM images of

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FIG. 1. AFM images from the sample A (a) after growth and (b) after the removal of the wetting layer and from (c) the sample A and (d) the sample C after the whole etching procedure. The scan size of the images is $2 \times 2 \mu\text{m}^2$.

the samples A and C, respectively, after the whole three-step etching procedure. Note, that the InP islands have already been removed from the surface. The citric acid has etched about 25 nm of GaAs in the sample C. However, in the sample A the etch depth is 30–35 nm, indicating higher etching rate for strained GaInAs.

The surface morphologies of Figs. 1(c) and 1(d) were investigated further from cross-sectional profiles of the AFM scans. Figure 2 shows a height profile recorded from the image of Fig. 1(c). Two types of columns are observed. The tall columns have a height of about 35 nm and an areal density of $1 \times 10^9 \text{ cm}^{-2}$, equal to the InP island density. The peaks of the tall columns correspond to the position of

FIG. 2. Cross-sectional profile from a $1 \times 1 \mu\text{m}^2$ AFM image of the as-etched sample A. The position of the GaInAs QW in the structure is indicated by dashed lines.

FIG. 3. PL spectra of the (a) as-grown and (b) regrown reference QW sample B and the (c) as-etched and (d) regrown sample A.

the as-grown GaAs surface. The position of the GaInAs QW is indicated by dashed lines in Fig. 2. The columns have typically a diameter of 20–60 nm, which is smaller than the original InP island diameter (70–80 nm) because of underetching. The short columns have an average height of 17 nm and their peaks are all below the level of the original GaInAs QW. Columns with a height of 18–30 nm are not detected in this image or in several other AFM images taken across the sample. We assume that the two types of columns are produced because of more rapid etching of GaInAs than GaAs. When the GaInAs layer is exposed somewhere, the etching proceeds also in horizontal direction in the GaInAs layer. Thus, the GaInAs layer is totally removed between the InP islands. Because of the larger etch rate of GaInAs, it is likely that the GaInAs layer has been underetched in the free-standing columns. Therefore, the QD diameters may be smaller than those of the columns. The columns in Fig. 1(d) have a continuous distribution of heights. There is no GaInAs layer in this sample C to accelerate the etching and to produce two column types.

The regrowth of the samples A and B was performed in the same growth run. Before the regrowth, the passivating InP layer in the sample B was removed by etching one minute in HCl. Both samples were rinsed in 1 HF:10 isopropanol and then in isopropanol before the samples were transferred into the reactor. About 50 nm of GaAs and 1 ML of InP was deposited at 550 °C.

Figure 3 shows PL spectra from the (a) as-grown and (b) regrown reference QW sample B and from the (c) as-etched and (d) regrown sample A. The peaks at 1.515 and 1.495 eV correspond to bound exciton and carbon-related band-to-acceptor emission from the GaAs layer. The peak at 1.440 eV in Fig. 3(a) originates from the QW. The full-width at half-maximum (FWHM) of the peak is 5 meV. After the regrowth, the width of the QW peak (at 1.447 eV) has increased to 6 meV. The luminescence intensity has decreased by almost two orders of magnitude indicating presence of nonradiative recombination centers at the regrowth interface.

FIG. 4. PL spectra from the regrown sample A at various excitation intensities.

Figure 3(c) shows the PL spectrum from the as-etched sample A. A peak at 1.438 eV originating from the GaInAs QDs is observed. This assumption is confirmed by the fact that no peaks are observed at 1.37–1.49 eV in the PL spectrum of the as-etched sample C (not shown here). Due to several effects, the QD transition energy is nearly the same as that of the QW peak in the reference sample B. Quantization in the disk plane shifts the peak to blue, but the partial relaxation of strain in the GaInAs layer and the surface effects shift the peak to red. After regrowth, the QD peak is detected at 1.457 eV. The FWHM of the QD peak increases from 9 to 16 meV in the regrowth. The broadening results from regrowth-induced mixing in the interfaces and from a relative larger increase in the luminescence efficiency of the smallest QDs. The blueshift of the QD peak by 19 meV after regrowth is realized by increased strain in the GaInAs disks by the GaAs matrix and by the elimination of the surface effects. Preliminary calculations indicate that the band gap in the GaInAs disks raises by 10–40 meV after regrowth. The increase is largest at the edge of the disk and smallest at the center of the disk. The diameter of 50 nm was used for the GaInAs disk in this calculation. In addition to the peaks discussed above, weak luminescence bands at about 1.36 and 1.03 eV are observed in all the etched and regrown samples. Therefore, these bands must arise from defect and impurity centers introduced to GaAs surface during etching and regrowth.

The integrated intensity of the QD peak in the regrown sample A is only five times lower than in the regrown reference QW sample B. However, if the luminescence intensity

is normalized by the area of the emitting region, about one order of magnitude higher luminescence efficiency is achieved from the overgrown QDs than from the overgrown QW. For this calculation, a diameter of 50 nm has been used for the GaInAs disks, producing a fill factor of 2%.

Figure 4 shows PL spectra from the regrown sample A measured at various excitation intensities. As the excitation intensity is increased, the high-energy side of the QD peak rises indicating possible excited-state luminescence. Also, the saturation of the QD peak intensity relative to the GaAs peaks indicates filling of the states in the QDs.

In summary, GaInAs quantum disks have been fabricated by wet chemical etching from a GaInAs/GaAs quantum well structure using self-organized InP islands as an etch mask. The three-step etching procedure produces free-standing columns containing GaInAs disks with an areal density of $1 \times 10^9 \text{ cm}^{-2}$, equal to the density of the InP islands. Photoluminescence studies indicate that the luminescence efficiency per emitting area of the overgrown GaInAs disks is one order of magnitude higher than that of the overgrown quantum well at the same depth from the surface.

This work was supported by the Academy of Finland under Grant No. 34158. The assistance of T. Aalto in the sample processing is acknowledged.

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