Multistep fabrication of self-assembled unstrained quantum dashes

A. A. Ukhanov, a) A. S. Bracker, G. Boishin, and J. C. Culbertson

Naval Research Laboratory, Washington, DC 20375

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We describe a technique for molecular-beam-epitaxy-based fabrication of unstrained quantum dashes with AlInGaAs alloys lattice matched to InP substrates. Templates for lattice-matched quantum dash growth are obtained by combining molecular beam epitaxy with in situ etching by arsenic bromide. A seed layer of strained self-assembled InAs quantum dashes is converted into nanotrench templates through overgrowth followed by strain-enhanced etching. We have explored limitations on the accessible range of alloy compositions imposed by the etch process and found that strain-induced etching is limited to compounds with low Al content. Nanotrench templates can be filled with lattice-matched alloys of varied compositions to define barriers and quantum wires that could lead to optoelectronic devices in a spectral range around 1.5 μm. [DOI: 10.1116/1.2197515]

I. INTRODUCTION

Semiconductor quantum dashes (QDashes) are a promising material for optoelectronic devices such as lasers1–3 and amplifiers.4 For applications from 1.5 to 2.0 μm, the dashes typically consist of InAs grown by Stranski-Krastanow (SK) self-assembly. Intermixing often occurs during growth and capping of the strained QDashes, leading to changes in composition and shape. These drawbacks could be mitigated by growing lattice-matched In0.53Ga0.47As QDashes on an Al0.48In0.52As buffer and InP substrate, using a technique5 that relies on in situ etching of a strained seed layer to form a nanohole template for lattice-matched quantum dot growth. Here, we investigate the prospects for adapting this technique to the growth of unstrained QDashes.

Arsenic bromide (AsBr3) is capable of etching III-V semiconductor compounds such as GaAs, InAs, and AlGaAs in a molecular beam epitaxy (MBE) growth environment.6,7 This in situ technique allows etching with atomic precision and real time control via reflection high-energy electron diffraction (RHEED) oscillations. Importantly for device applications, previous work has shown that the etched material is very pure, with high electrical mobility and good optical properties.5–7 The etching is based on the formation of strongly bonded III-bromide compounds, resulting in layer-by-layer removal of group III surface atoms (In, Ga, or Al) to the gas phase.

Atomic-scale etch rate control using arsenic bromide has proven to be useful for nanostructure fabrication when combined with its sensitivity to strain and crystal facets. Schuler and Eberl demonstrated nanohole formation when a buried layer of strained self-assembled InAs quantum dots on GaAs is etched.8 Songmuang et al. subsequently used these nanoholes as nucleation sites for self-assembled lateral quantum dot molecules.9 More recently, Rastelli et al. used the nanoholes in GaAs as a template to form corrugated AlGaAs barriers, which were then filled with unstrained GaAs quantum dots.5

Here, we demonstrate growth and etching techniques required for fabrication of unstrained QDashes, including seed layer formation, nanotrench etching, and filling with lattice-matched barrier materials. In contrast to a previous work,5 we begin with a seed layer of InAs QDashes on a buffer layer matched to an InP substrate, followed by nanotrench etching of the capped seed layer dashes. In this material system, there is only a 3% lattice mismatch between InAs and InAlGaAs/InP, compared to 7% for InAs on GaAs, allowing us to explore strain-induced etching in a system with smaller strain. We also demonstrate strain-induced etching of QDash seed layers in ternary and quaternary alloy buffers such as InGaAs, AlInAs, and AlInGaAs. We compare them to InAs quantum dot seed layers in AlGaAs, in order to explore the limitation of strain-induced etching in compounds with high Al content.

Finally, we describe a simple method for obtaining In0.53Ga0.47As/Al0.48In0.52As/InP quantum dashes via multistep self-assembly using in situ AsBr3 etching that could be incorporated into a practical diode laser design based on unstrained InGaAs QDashes. The InGaAs heterostructure can be replaced with AlInGaAs and can be designed to emit light at 1.5 μm, which is attractive for optical communication. It may also be possible to grow unstrained InGaAs or AlInGaAs QDashes without a wetting layer and with more flexibility and control of QDash height than usual SK dots or dashes, which could have advantages for diode lasers, amplifiers, and modulators.

II. EXPERIMENT

The experiments were carried out in a solid source MBE system with an AsBr3 gas source etching unit (MBE Komponenten), consisting of a gas manifold and a heated injector. AsBr3 source liquid was kept at 30 °C, and the vapor was flowed through heated tubes to the injector unit at a flow rate of 1.5 SCCM (SCCM denotes cubic centimeter per minute at STP), giving a beam equivalent pressure of 5 × 10−7 torr in the MBE chamber. Etching rates were calibrated with RHEED oscillations. AsBr3 etching was carried out without an additional flux from the arsenic beam.

a)Electronic mail: ukhanov@bloch.nrl.navy.mil
Following substrate oxide desorption, we deposited buffer layers of 500 nm of GaAs at 610 °C on GaAs or 200 nm of Al_{0.48}In_{0.52}As or In_{0.53}Ga_{0.47}As at 450 °C on InP. Group III growth rates to provide lattice matching to InP substrates were calibrated by RHEED oscillations and fine tuned using x-ray diffraction.

Atomic force microscopy (AFM) in tapping mode (≈10 nm resolution) was performed to measure surface morphology following growth or etching.

III. RESULTS AND DISCUSSION

The fabrication procedure for unstrained InGaAs or AlInGaAs QDashes is based on the techniques of Ref. 5 for producing unstrained GaAs/AlGaAs quantum dots. A schematic sample structure and step-by-step fabrication sequence are shown in Fig. 1. First, a strained seed layer of InAs islands is grown on an In_{0.53}Ga_{0.47}As buffer lattice matched to an InP (001) substrate. InAs has a 3% lattice mismatch with In_{0.53}Ga_{0.47}As, and the bonding asymmetry of the zinc blende structure creates anisotropy in the indium atom surface migration distance,\(^{10}\) resulting in growth of finite length wires (dashes). In the second step, the InAs dashes are overgrown with a thin layer of InGaAs (or AlInGaAs), producing a flat surface. In the third step, the surface is exposed to AsBr\(_3\) etching gas, which etches the strained regions above the InAs dashes more rapidly than the regions of unstrained InGaAs, producing trenches in the cap surface that go all the way through the InAs dash layer. In the fourth step, the nanotrenches are covered with a thin layer of higher-band-gap Al_{0.48}In_{0.52}As that serves as the lower barrier for the final QDash layer. Because of the high aluminum content in this alloy, surface atom mobilities are low and the underlying trench structure is maintained at the surface. In the fifth step, the nanotrenches are filled with the active material (e.g., In_{0.53}Ga_{0.47}As), producing inverted dashes. In the final step the QDashes are overgrown with a top barrier of Al_{0.48}In_{0.52}As.

The right column of Fig. 1 shows AFM images following steps 1, 3, and 4 of this procedure, using InGaAs as the buffer layer and QDash material. We deposited 200 nm In_{0.53}Ga_{0.47}As buffer (T\(_{\text{sub}}\)=450 °C) lattice-matched to an InP (001) substrate. The seed layer dashes had width of 45±15 nm, length of 500±300 nm, height of 4.5±1 nm and were aligned along the [1–10] crystallographic direction [Fig. 1, step 1 and Fig. 2(c)]. Capping with 10 nm In_{0.53}Ga_{0.47}As produced a flat surface, as confirmed with AFM. Etching with AsBr\(_3\) for 61 s at 450 °C produced nanotrenches with width of 48±15 nm and depth of 5±1 nm [Fig. 1, step 3 and Fig. 2(c')]. Partial filling of the nanotrenches with 6 nm of Al_{0.48}In_{0.52}As alloy (T\(_{\text{sub}}\)=450 °C) produces somewhat narrower nanotrenches with width of 23±11 nm and depth of 4±1 nm (Fig. 1, step 4). We then filled the nanotrenches with 2 nm In_{0.53}Ga_{0.47}As to form inverted InGaAs dashes and followed with overgrowth of 100 nm of Al_{0.48}In_{0.52}As and a 3 min anneal at T\(_{\text{sub}}\)=450 °C. We did not achieve complete flattening after the final capping, which may require adjusting the anneal time and temperature for this alloy.

In Fig. 2, we compare nanotrench etching for an InAs QDash seed layer with two different buffer alloy compositions to etching of nanoholes in GaAs with an InAs seed layer. For nanoholes in GaAs, we deposited ≈1.7 ML InAs on a GaAs buffer (T\(_{\text{sub}}\)=500 °C) to form the seed layer of quantum dots with diameter of 30±15 nm and height of 6±2 nm [Fig. 2(a)]. The substrate temperature was lowered to 470 °C and a 10 nm GaAs cap was grown while ramping the temperature back to 500 °C. GaAs capping produced a flat surface, as observed with AFM. AsBr\(_3\) etching gas was applied for 65 s at 500 °C to produce nanoholes with diameter of 40±20 nm and depth of 4±1 nm [Fig. 2(a')]. These results are consistent with previous reports.\(^{58}\)
Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As, a higher gap material that could itself be...it has been seen that etch pits can be created directly in Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As, a higher gap material that could itself be used as a barrier. A natural question is whether the aluminum content can be increased even further to provide stronger confinement. We have investigated etching of quantum dots and quantum dashes that are capped with alloys containing ~50% aluminum. The seed layer dots or dashes [Figs. 3(a) and 3(b)] were grown and capped with 10 nm Al$_{0.48}$Ga$_{0.52}$As (for dots) or Al$_{0.48}$In$_{0.52}$As (for dashes). Following etching with AsBr$_3$, we see an unexpected result. Instead of holes and trenches, we observe protrusions with similar lateral dimensions to the seed layer but heights reduced by 20%–25%. This effect probably results from lower surface mobility for atoms on the 50% aluminum alloys. The growth front of the aluminum-containing cap layer simply follows the contour of the underlying dot or dash seed layer, projecting their shape upward instead of flattening out (similar to filling of nanoholes with barrier material in Ref. 5). Subsequent etching is then insufficient to remove the added surface topography, even with strain-enhanced etching produced by the seed layer.

The capping and etching process clearly imposes an upper limit on the aluminum content that can be used for etching holes or trenches. Nevertheless, trenches in a material such as Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As could prove useful for optical devices. The growth and etching sequence described by Fig. 1 places a barrier layer between the QDashes and a low-band-gap buffer layer. For an efficient diode laser or amplifier, one would prefer that the quantum dash layer has the lowest band gap in the structure and that no additional parasitic potential barriers exist. The possibility of etching nanotrenches in Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As would allow us to improve and simplify the unstrained QDash fabrication [Fig. 4, left column]. Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As trenches can be filled with In$_{0.53}$Ga$_{0.47}$As directly, creating quantum confinement with no need for an additional Al$_{0.48}$In$_{0.52}$As barrier as shown in Fig. 1.

In Fig. 4, we present an unstrained In$_{0.53}$Ga$_{0.47}$As/Al$_{0.20}$Ga$_{0.27}$In$_{0.53}$As quantum dash laser design that should emit light near 1.5 μm. The same heterostructure was previously used for a diode laser with strained InAs quantum dashes and showed excellent properties such as low threshold current (410 A/cm$^2$) and high modal gain (22 cm$^{-1}$). Here we propose to use this heterostructure with...
an unstrained $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{Al}_{0.20}\text{Ga}_{0.27}\text{In}_{0.53}\text{As}$ QDash active region. The wavelength of the unstrained quantum dash laser could be adjusted by fine tuning the Al/Ga ratio or even the In/Ga ratio, at the expense of a small amount of strain. Alternatively, the QDash size may be modified by changing the etching depth. The proposed fabrication procedure has other benefits as well. The wetting layer could be etched away, eliminating carrier traps to improve temperature dependence and carrier-induced optical properties in diode lasers and amplifiers. Deeper etching may allow one to adjust the QDash aspect ratio to produce a polarization-independent active medium for semiconductor optical amplifiers.

**IV. CONCLUSION**

We have created a type of unstrained $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ quantum dash by combining self-assembly with *in situ* AsBr₃ etching. We demonstrated strain-induced etching for InAs QDash seed layers in buffers with the InP lattice constant (3% mismatch) and found that the etching is not substantially altered compared to the InAs/GaAs system (7% mismatch). We explored seed layer etching in ternary and quaternary alloy buffers InGaAs, AlInAs, and AlInGaAs and found that effective etching is limited to compounds with low Al content. An unstrained QDash structure could be incorporated in a realistic diode laser operating in the midinfrared wavelength range.

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