Horizontal Heterojunction Integration via Template-Assisted Selective Epitaxy

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ABSTRACT: We report on the successful integration of multiple atomically thin horizontal heterojunctions (HJs) epitaxially grown via metal organic chemical vapor deposition inside a confined template of dielectric material. InAs, GaAs, and InGaAs layers were included in laterally grown InP structures and characterized to show abrupt interfaces and crystalline material. The orientation of the templates and the substrate is chosen so that a flat vertical facet appears at the growth front allowing for the HJs to be horizontal, unlike typical planar epitaxy. This enables the design of recently proposed novel electronic HJ devices like triple-HJ tunnel field-effect transistors.

1. INTRODUCTION

Epitaxial growth in confined three-dimensional (3D) dielectric templates, referred to as template-assisted selective epitaxy (TASE), has recently generated increased interest for multiple purposes such as integrating highly lattice-mismatched materials, as a virtual substrate approach for III–V on silicon integration, and as means to directly integrate novel nanowire devices. TASE is a type of confined selective area growth (SAG) that involves epitaxy via metal organic chemical vapor deposition (MOCVD) of semiconductor materials within a confined structure that is formed with patterned dielectric films. The geometries of the confined structures used, shown in Figure 1, are so that gas-phase precursors can enter through a “source” hole in the dielectric and are exposed only to a small area of the substrate, referred to as a “seed,” where growth selectively initiates. Growth then proceeds laterally in the hollow channel and is thus confined in the template itself, allowing for an engineerable orientation and size of the epitaxial material.

With transistors reaching their scaling limit there is a need to move beyond traditional designs. Interesting novel devices have been proposed that base their theoretical superior electronic performances on specific heterostructures, particular crystal orientations and the anisotropy of III–V energy bands, planar gating enabled by lateral growth, and buried oxides. Energy filters that reduce injection into the channel of electrons having energy above the source Fermi energy have been proposed for nanowire field-effect transistor (FET) devices. Super lattice (SL) filters have also been proposed for planar and fin FETs with low subthreshold swings attractive for low-power logic applications.

Confined epitaxy via TASE is a method to achieve all of this, because it defines the device orientation arbitrarily via the fabricated template. The lateral growth allows planar gating of the structures, resolving one of the issues of other structures that require complex gating, such as mesa and nanowire tunneling-FETs (TFETs) show. Finally, direct integration of these horizontal heterojunctions is possible in a single step growth, with fast gas switching in the MOCVD allowing nanometer thin layers for quantum well structures to be grown directly in the confined channel.

In this work we demonstrate that TASE can be a novel and efficient way to grow various lateral horizontal heterojunctions for a number of applications, as future “building blocks” toward orientation-dependent and HJ-based devices. We start by showing a successful growth of an InP/InAs HJ, which is an important material system for telecom wavelength room-temperature lasers. This is followed by a TASE overgrowth containing InP/GaAs and InP/InAs HJ, which is a significant HJ system due to a type-II band alignment between these materials showing interesting photoluminescence and quantum-confinement properties. Then we demonstrate the growth of an InGaAs/InP triple quantum well for energy-filtering applications as well as a InP/InAs/GaAs/InP triple-heterojunction (3HJ) structure, which is proposed to be a high on-current TFET. We provide cross-sectional high-resolution transmission electron microscopy (TEM) analysis to show the high quality of the material grown. Lastly, we show how the

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multiple HJs provide clues to changing growth rates inside a TASE template.

2. MATERIALS AND METHODS

2.1. Template Fabrication. Templates were fabricated by depositing a 5 nm alumina etch stop layer and a 20 nm bottom dielectric SiO₂ via plasma-enhanced chemical vapor deposition (PECVD), lithographically defining the seeds, spin coating, and patterning a CSAR photoresist 50 nm sacrificial layer, and spin coating hydrogen silsesquioxane (HSQ) as the 100 nm top dielectric in which source holes are lithographically defined. The sacrificial layer was removed with 1-methyl-2-pyrrolidone stripper (NMP) followed by remote oxygen plasma at 350 °C. A final tetramethylammonium hydroxide (TMAH) wet etch removes the alumina layer exposing the seed, and a diluted HF dip is executed before growth. More details on the fabrication and a comparison with possible alternatives can be found in the references. 24

2.2. Epitaxy via Metal Organic Chemical Vapor Deposition. MOCVD was done in a horizontal reactor using trimethylindium (TMIn), trimethylgallium (TMGa), tertiarybutylphosphine (TBP), tertiarybutylarsine (TBA), H₂ as carrier gas. Growth parameters for the different materials are summarized in Table 1.

To achieve abrupt heterointerfaces, when switching materials during growth, the growth rate was slowed for a few seconds by reducing the molar flow of group III precursors in half, followed by a 1 s (approximate gas residence time in the reactor) purge in carrier gas only. All other parameters were kept unchanged throughout the growth.

2.3. Characterization. Success of growth is initially evaluated via top-down scanning electron microscopy (SEM) imaging as shown in Figure 1b, where contrast allows distinguishing between the empty cavity, the epitaxial material, the source hole, and the seed.

Table 1. Summary of Main Growth Parameters

<table>
<thead>
<tr>
<th>Material</th>
<th>Temperature (°C)</th>
<th>Pressure (torr)</th>
<th>Group III (mol/min)</th>
<th>V/III ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>InP</td>
<td>600</td>
<td>50</td>
<td>2.7 × 10⁻⁶</td>
<td>570</td>
</tr>
<tr>
<td>InAs</td>
<td>600</td>
<td>50</td>
<td>2.7 × 10⁻⁶</td>
<td>180</td>
</tr>
<tr>
<td>GaAs</td>
<td>600</td>
<td>50</td>
<td>1.9 × 10⁻³</td>
<td>25</td>
</tr>
<tr>
<td>(Figure 3)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>GaAs</td>
<td>600</td>
<td>50</td>
<td>4.75 × 10⁻⁶</td>
<td>100</td>
</tr>
<tr>
<td>(Figure 6)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>InGaAs</td>
<td>600</td>
<td>50</td>
<td>5 × 10⁻⁶</td>
<td>95</td>
</tr>
</tbody>
</table>

dielectric SiO₂ via plasma-enhanced chemical vapor deposition (PECVD), lithographically defining the seeds, spin coating, and patterning a CSAR photoresist 50 nm sacrificial layer, and spin coating hydrogen silsesquioxane (HSQ) as the 100 nm top dielectric in which source holes are lithographically defined. The sacrificial layer was removed with 1-methyl-2-pyrrolidone stripper (NMP) followed by remote oxygen plasma at 350 °C. A final tetramethylammonium hydroxide (TMAH) wet etch removes the alumina layer exposing the seed, and a diluted HF dip is executed before growth. More details on the fabrication and a comparison with possible alternatives can be found in the references. 24

3. RESULTS AND DISCUSSION

It has been previously observed that, under certain growth conditions (580–600 °C, 50 Torr, 570 V/III ratio), in...
templates fabricated on (110) InP substrates and oriented along the [1−10], growth can be terminated in flat and vertical (−110) and (1−10) facets. These facets were chosen to demonstrate the multiple HJs in this work. Additional development is needed to optimize the yield of these facets, which is now low (<30%) and varies between runs possibly because of minute differences in template fabrication, which, in turn, influences growth and growth initiation. Both our own trials and published work 25 show facet dependence on temperature and V/III ratio, leading us to believe yield can be increased by optimizing growth parameters.

3.1. InAs Heterojunction. To demonstrate the feasibility of horizontal HJs a single InP/InAs/InP HJ was integrated in the confined channel. The InAs HJ was achieved via a single step growth. Figure 2 shows cross-sectional STEM characterization of the structure, showing abrupt heterointerfaces on either side of the InAs layer. High-resolution imaging suggests crystallinity of the material, which is confirmed by the diffraction pattern. The InAs layer grown is ~1.5 nm thick, so although the lattice mismatch between the materials is high (~3%), it is below the ~2.1 nm Matthew Blakeslee critical thickness for InAs on InP.

3.2. GaAs Heterojunction. A single InP/GaAs/InP horizontal heterojunction was demonstrated. The structure was grown in a single step growth. Reproducibility of results was tested by including in the same structure multiple InAs layers grown with identical parameters used for the structure shown in Figure 2. The resulting InAs layers, while showing some variations, were mostly consistent in quality and thickness across the two trials. STEM characterization shown in Figure 3 shows abrupt, vertical interfaces, with crystalline order.

3.3. InGaAs Superlattice. A three-well InP/InGaAs structure, resembling SL energy filters, was here demonstrated.
By design all InGaAs layers and the first two layers of InP are supposed to be identical (3 nm), while the third InP is thinner (2 nm). The resulting growth somewhat deviates from the design as shown in Figure 4. After the SL itself, a thick InGaAs layer is included, to mimic a possible contact layer in a final device. The structure was grown in a single step. The facets are flat, and the interfaces are quite abrupt even though less so than the previous structures. The reason why the interfaces are not as abrupt is unclear. We think As/P intermixing, a common problem with these materials, while a possibility, is not inherent to the confined growth, since it is not present in the other HJs shown here. Damage of the lamella is also a possibility with focused ion beam (FIB) induced gallium damage causing “blurriness” in the image, and so are small variations in thickness of the lamella.

When growing bulk ternary material via confined epitaxy, there is the possibility that a compositional gradient exists along the structure due to differences in the diffusion between the group III precursors. We executed an energy-dispersive spectroscopy (EDS) measurement to verify this in a confined InGaAs growth. The apparent change in In composition along the cavity shown in Figure 5, suggested by measuring the InL/AsL and the GaK/AsK, is 13% and 10%, respectively, measured from seed to growth front across 350 nm. The presence of a compositional gradient has also been observed in other works.26 It is reasonable to believe that, once the gradient is known, progressively adjusting the molar flow ratio of precursors during the growth should lead to a constant composition within the cavity.

3.4. Triple Heterojunction (3HJ). A 3HJ structure, comprised of an InP/GaAs/InAs/InP sequence, was demonstrated and is shown in Figure 5. The structure was achieved in a single step growth using the same parameters used in the structures described above. The 3HJ itself was repeated twice, and InAs layers were included as markers.

The entire structure is not symmetrical, with the left side being longer than the right, as clearly seen in Figure 6e. Though comparing the left and right sides of the template, seen in Figure 6a,b, the same HJs are present with similar appearance and spacing. It is possible that the growth initiated asymmetrically but ultimately

Figure 5. TEM EDS data of an InGaAs growth via TASE. The plot shows InL and GaK counts normalized to AsL and GaK, respectively. The green area indicates the position of the InGaAs inside the template. (inset) Schematic diagram of the structure.

Figure 6. Cross section of an InP growth with InAs marker layers and InP/InAs/GaAs triple HJs. (a, b) HAADF-STEM imaging. (c) Schematic diagram of the structure.

Figure 7. Average lateral growth rates for the individual InP layers between the HJs.
proceeded with some consistency among sides. In fact, if one was to measure the distance between the first InAs layer and an offset virtual point at the seed, the entire structure appears symmetrical. This “single point” nucleation hypothesis is corroborated by results from another horizontal HJ trial, where growth initiated nonuniformly and only on one side of the seed. This asymmetric growth behavior is seen in other SAG literature but needs further investigation.

3.5. Growth Rate Inside the Template. Including HJs allows to track the progression of growth and observe possible changes in growth rate ($R_g$) within the confined cavity. The presence of different materials does change the surface energy at the growth front, and thus the growth initiation, but on first approximation measuring the thickness of the InP spacers between the HJs will provide an estimate on $R_g$ in the cavity.

The length of the InP layers between the HJs shown in Figure 3 and Figure 6 has been measured and divided by the individual growth time for that layer to obtain the average growth rate for each. This average growth rate is then plotted versus distance from seed in Figure 7. When observing the InP spacers in the 3HJ sample in Figure 6, each grown under the same conditions and for the same time, it is clear that the thickness of each decreases with the distance to the source hole. This is in contrast with expectations: during the confined epitaxy the growth front advances toward the source hole, reducing the distance between them, and since MOCVD growth is driven by diffusion, growth rates are then expected to increase with time, as has been observed in previous studies. A possible cause of this reduction of $R_g$ is the presence of nonselective growth, that is, parasitic nucleation on the dielectric. While selectivity is almost perfect for InP nonselective growth, that is, parasitic nucleation on the different materials does change the surface energy at the growth front, and thus the growth initiation, but on first approximation measuring the thickness of the InP spacers between the HJs will provide an estimate on $R_g$ in the cavity.

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