Flow rate modulation epitaxy of high-quality V-shaped AlGaAs/GaAs quantum wires using tertiarybutylarsine as the arsenic source

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Abstract

We report the first successful growth of high-quality V-shaped AlGaAs/GaAs quantum wires (QWRs) using tertiarybutylarsine (TBAs) as the arsenic source by flow rate modulated metalorganic vapor-phase epitaxy. We found that the fundamental structural qualities of QWRs grown using TBAs (for example, the growth selectivity, the sharpness of the V-groove bottom, and the uniformity of the QWR surface) are comparable to or better than those of QWRs grown using AsH\textsubscript{3}. A characteristic defect was observed mainly in the intersecting region of the (1 1 1)A side wall and the (0 0 1) flat facets at low growth temperatures, but it would not have an important influence on the size of the grown QWRs for a substrate with a sufficiently long (11 1)A side wall facet. In a preliminary optical investigation of a 4.5 nm thick QWR, we observed an energy separation as large as 58 meV between the ground and the \textsuperscript{1}\textsuperscript{st} excited state and a ground state Stokes shift as small as 3.9 meV at 5 K. Both of these results are much better than the values of the sample grown using AsH\textsubscript{3}. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Flow rate modulation epitaxy (FME) of GaAs on patterned substrates has been proven to be a very successful method for the fabrication of high-quality quantum wires (QWRs) \cite{1,2}. Using V-shaped AlGaAs/GaAs QWRs grown by this technique, we have succeeded in investigating many important optical characteristics of QWR structures \cite{3-5}. We have also succeeded in the first demonstration of QWR lasers emitting from ground state at room temperature with a threshold current as low as 6 mA \cite{6}. On the other hand, replacing AsH\textsubscript{3} and PH\textsubscript{3} with less toxic organic materials is still an important challenge for the metallocorganic vapor-phase epitaxy (MOVPE) technique. Tertiarybutylarsine (TBAs) seems to be the most promising replacement gas for AsH\textsubscript{3}; various two-dimensional structures with optical and
electronic qualities comparable to or better than those of structures grown using AsH$_3$ have been realized by using TBAs [7]. However, few experiments have been conducted on the MOVPE selective growth on patterned substrates using TBAs. In this paper, we report the first successful growth of high-quality V-shaped AlGaAs/GaAs QWRs by FME selective growth on V-grooved substrates using TBAs as the arsenic source.

2. Experimental procedure

Epitaxial growth was carried out using a low-pressure (76 Torr) horizontal MOVPE reactor, which is a newly installed system specially designed for the growth of ultrahigh-purity structures. The substrates used were 2 or 4 μm pitch V-grooved GaAs substrates formed on (001) ± 0.1° GaAs substrates by photolithography and wet chemical etching using a NH$_4$OH:H$_2$O$_2$:H$_2$O = 1:3:50 solution. Before growth, the substrates were further etched by the same NH$_4$OH:H$_2$O$_2$:H$_2$O solution for 10–20 s to reduce the surface roughness produced during the V-groove preparation processes [8]. Trimethylaluminum (TMAI) and triethylgallium (TEGa) were used as the group III sources. The growth temperature was changed in the range of 630–660°C. V/III ratios of 40 and 80 were used for GaAs and AlGaAs, respectively, in the MOVPE continuous growth mode.

The FME growth cycle consisted of a 1 s TEGa supply period, a 1 s H$_2$ purge period, a 1 s TBAs supply period, and a 1 s H$_2$ purge period; this was the same as that used in previous growth with AsH$_3$ [1,2]. The amount of TEGa and TBAs supplied in one pulse was 0.092 and 17.8 μmol, respectively. A small amount of bias TBAs (about 12 μmol/min) was flowed throughout the growth period to prevent the evaporation of arsenic atoms and the incorporation of impurities during TEGa supply and H$_2$ purge periods. The above conditions resulted in a QWR growth rate of about 1.09 ML/cycle at the V-groove center for the 4 μm pitch V-grooved substrate.

The sample structure typically consisted of a 0.2 μm GaAs buffer layer, a 0.9 μm Al$_{0.425}$Ga$_{0.575}$As upper barrier layer, and a 10 nm GaAs cap layer. Here the Al composition was the value for the (1 1 1)A side wall facet measured by photoluminescence (PL). The structural properties of the grown samples were characterized by transmission electron microscopy (TEM) and atomic force microscopy (AFM). The optical qualities were evaluated by PL and PL excitation (PLE) measurements.

3. Results and discussion

3.1. Structural characterization

Fig. 1 shows a cross-sectional TEM image of a vertically stacked 4-QWR sample grown on a 4 μm pitch V-grooved substrate at 630°C. As can be seen in this figure, crescent-shaped QWRs were successfully formed at the V-groove bottom. The growth selectivity (ratio of growth rate at V-groove center to that at (1 1 1)A side-wall), in the range of 4–5, was found to be as high as that of growth using AsH$_3$. Also, as in the case of growth using AsH$_3$, the QWR upper interface showed a clear facet evolution; one (001) facet was surrounded by two GaAs QWR layers, a 0.2 μm Al$_{0.425}$Ga$_{0.575}$As upper barrier layer, and a 10 nm GaAs cap layer. Here the Al composition was the value for the (1 1 1)A side wall facet measured by photoluminescence (PL). The structural properties of the grown samples were characterized by transmission electron microscopy (TEM) and atomic force microscopy (AFM). The optical qualities were evaluated by PL and PL excitation (PLE) measurements.

Fig. 1. TEM image of a vertically stacked 4-QWR sample grown using TBAs on a 4 μm pitch V-grooved substrate at 630°C.
(311)A facets. Further, the two side-wall facets with an intersecting angle of about 78° were closer to the exact (111)A facets (intersecting angle: 72°) as compared with structures grown using AsH₃ (intersecting angle: 83°).

Next, we investigated the surface roughness of the grown QWR using AFM, which was found to be a very effective technique for the structural characterization of V-grooved QWRs [8–10]. Fig. 2 shows the topography AFM images of two 80 nm thick QWR samples grown at 630 and 660°C, respectively. These samples were not covered with the upper AlGaAs barrier and the GaAs cap layers for AFM observation. In this case, a V-groove period of 2 μm was used in order to enable the AFM cantilever to reach the V-groove bottom easily. These images were flattened along the [011] direction in order to put all facets on the same average height level. The scan area is 5 μm × 5 μm. The appearances of the long-range height modulation on the (111)A side wall facets and the quasi-periodic step arrays on the small (311)A facets formed in the intersecting region of the (001) flat and the (111)A side wall facets are similar to those observed in the growth using AsH₃ [8]. A significant difference from the growth using AsH₃ was that a characteristic defect was observed at 630°C as indicated by black arrows in Fig. 2(a). This defect probably originated from remaining crystalline imperfections on the initial V-groove surface. This kind of defect appeared mainly in the region where the (111)A side wall and the (001) flat facets intersected, and also on the (001) flat facet, but with a smaller size and a smaller density. Fortunately, these defects were not observed on the (111)A side wall facets or at the V-groove bottom. Therefore, the influence of these defects on the size of QWRs would not be an important problem, provided that the (111)A side wall was long enough. A line scan of AFM data showed that these defects have a “V”-shaped profile in both the [011] and the [011] directions with a typical depth of 100–200 nm. Further, these defects were found to be sensitive to growth temperature; they disappeared almost completely when the temperature was increased to 660°C as is clear from Fig. 2(b).

In Fig. 3, we show the enlarged AFM images of the region indicated by the black square in Fig. 2(a), where Figs. 3(a) and (b) represent the flattened topography and the deflection image, respectively.

![Fig. 2. Flattened topography AFM images of two 80nm thick GaAs QWR samples grown using TBAs on 2μm pitch V-grooved substrates at (a) 630°C and (b) 660°C. The scan area is 5μm × 5μm.](image-url)
The deflection image is more sensitive to small surface roughness, but we could not obtain the exact value of surface roughness from this image because the deflection value was not calibrated with the absolute amplitude of height variation. The QWR surface was observed as a stripe with a width of about 130 nm as marked by two horizontal black lines. In the case of growth using AsH$_3$, we usually observe step-bunching-induced quasi-periodic step arrays on the two $(3\,1\,1)A$ QWR facets, especially in the deflection image, with an average height of about 7 Å and an average length of about 700–800 Å [8]. However, the surface of QWRs grown by TBAs showed very smooth morphology without any observable step arrays, even in the deflection image, as is clear from Fig. 3. This observation suggests that step bunching on the $(3\,1\,1)A$ QWR facets seems to be greatly suppressed by the use of TBAs, probably due to differences in surface reaction dynamics between TBAs and AsH$_3$.

3.2. Optical properties

We also made preliminary characterization of the optical quality of the grown QWRs by PL and PLE spectroscopy. Fig. 4 shows the 5 K PL and PLE spectra of a 4.5 nm thick single QWR grown on a 4 µm pitch V-grooved substrate at 630°C. The polarization of the laser was set parallel to the QWR direction in PLE measurements.
direction in PLE measurements. The full-width at half-maximum of the PL peak of about 8 meV was comparable to that of the sample grown using AsH$_3$. In the PLE spectrum, well-resolved heavy hole-related subband structures were clearly observed. An energy separation as large as 58 meV between the ground and the first excited QWR states and a ground state Stokes shift as small as 3.9 meV were obtained. These values were about 12 meV larger and 2.3 meV smaller than those of samples grown using AsH$_3$ (46 and 6.2 meV), respectively [11]. The small Stokes shift cannot be attributed to a pseudo-flat interface, i.e., an interface with roughness smaller in length than the exciton Bohr radius. We also measured micro-PL for this sample, and the spectrum was split into several sharp peaks at 10 K as in the case of the sample grown using AsH$_3$ [12]. One possible reason for the small Stokes shift might be the suppression of step bunching on the (311)A QWR facets as shown in Fig. 3. The small Stokes shift is also a good indication that the specific defects observed in Fig. 2 do not affect the uniformity of the QWR, at least for the present 4 μm pitch substrate. Therefore, the results of this study suggest that high-quality QWRs with stronger lateral quantum confinement and smoother heterointerfaces than structures grown using AsH$_3$ can be produced by using TBAs.

4. Conclusion

In conclusion, we have investigated for the first time the FME selective growth of AlGaAs/GaAs QWRs on V-grooved substrates using TBAs as the arsenic source. Preliminary results show that TBAs can produce QWRs with structural and optical qualities higher than those of samples grown using AsH$_3$, indicating the possibility that TBAs could replace AsH$_3$ for the selective growth of low-dimensional quantum structures.

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References