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Fabrication of Quantum Dots in Twin-Free GaAs Nanopillars on Si

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1. Introduction

Free-standing nanowires are promising for future nano-scale devices. The VLS (vapor-liquid-solid) growth method enables us to make quantum dots in nanowires, which will contribute to the development of nanodevices such as transistors, nanolasers and nanosensors. We have recently reported core-multishell nanowires with flat tops and shown that air-gap structures can be formed just by using selective wet-etching [1,2]. These structures were grown by combining the VLS growth and MOVPE (metalorganic vapor phase epitaxy). Our fabrication methods are in general bottom-up approaches. Here, we report the fabrication of quantum dots in nanopillars. We found that twin-free zinc-blende GaAs can be formed over GaP nanowires, which will lead to refined heterostuructures. The pillars with flat tops are very suitable for light propagation. Further, the number of quantum dos can be defined in one pillar, which makes it easy to access quantum dots in an optical manner.

2. Fabrication procedures

Nanopillars were formed by combining the VLS growth mode and MOVPE mode. These growth methods were carried out in a low-pressure MOVPE reactor [1,2]. Trimethyl-gallium, trimethyl-aluminum and trimethyl-indium were the group-III sources, and AsH_3 and PH_3 were the group-V sources. The flow rate of the group III sources was 5-10×10^{-6} mol/min and that of the group V sources was 4×10^{-4} mol/min. Si(111) substrates were used in this study. Figure 1 shows the procedure for obtaining the target nanostructure. After the Au particles from the Au colloids had been dispersed on the surface, the temperature was raised to 520 ˚C to grow GaP nanowires for one minute. Then GaAs growth was performed at 550 ˚C (HT-GaAs) for 10 minutes, after which the nanostructures show pillars surrounded by {112} facets with tapered tops. Next, as shown in Fig. 1(b), InAs growth was performed at 460 ˚C for 3 seconds. The Au particles used here were 20 nm in diameter so quantum confinement is not strong enough in the lateral direction. In principle, we could make the InAs smaller in the lateral direction by using smaller Au particles. The thickness of the InAs was not optimized in this study but aimed it under 10 nm. Finally, HT-GaAs was grown again for 10 minutes and AlAs nanowires were grown at 460 ˚C for 20 seconds. The process from (a) to (c) was one continuous run in the chamber. After that, the sample was dipped in a wet-etchant ($H_2SO_4/H_2O_2/H_2O$) to remove AlAs nanowires together with the Au particles. The sample again loaded into the growth chamber and HT-GaAs was grown for 7 minutes to make flat-top pillars as shown in Fig. 1(e)

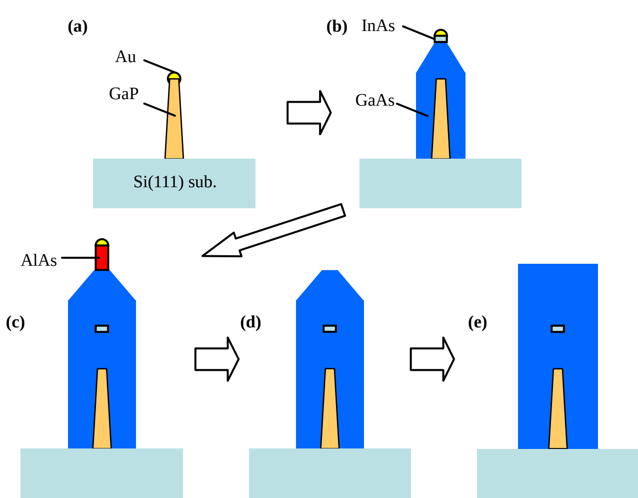

Fig. 1. Cross-sectional schematic images of nano-pillars explaining the fabrication process. (a) GaP nanowires on Si(111); (b) HT-GaAs and InAs growth; (c) HT-GaAs and AlAs growth; (d) after removal of AlAs together with Au particles; (e) HT-GaAs.

3. Structure and optical analysis

Figure 2(a) shows a scanning electron microscopy (SEM) image of the nanopillars on a Si substrate. We could obtain flat-top nanopillars vertically grown on Si substrates. Figures 2(b) and (c) show a high-resolution transmission electron microscopy (HR-TEM) image and selective area diffraction (SED) patterns at the indicated regions. No twins are seen in the upper region. This region is apparently a zinc-blende structure. From the evaluation of the height, the boundary is the top of the GaP nanowires so that the quantum dot is located in the twin-free region. Further, in order to determine the quantum dot position more precisely, we performed energy dispersive X-ray spectrometry (EDS) analysis, as shown in Fig. 3. However, no clear sign of an InAs quantum dot was detected. The InAs seems to be so thin that the luminescence from the K line is under the detection limit. We also performed low-temperature photoluminescence measurement. Figure 4 shows the PL spectrum at 4 K obtained from a single pillar. Only one peak with shoulders was seen in the range from 750 to 1600 nm using Si-CCD and InGaAs diode array (the input light was 710 nm in wavelength). The peak at 821 nm (1.51 eV) is attributed to zinc-blende GaAs [3,4]. A shoulder at 832 nm (1.49 eV) is also seen, which is attributed to carbon-related emission [4]. Possible InAs-related emission around 1.46 eV as a shoulder is seen. The intensity is very week, although InAs is located in the twin-free region. Possible reasons are high concentrations of impurities and non-radiative centers. In addition, InAs dots are about 20 nm in diameter, which may cause stress around the dot region due to large lattice mismatch and this stress would influence the emission. As the next step, we have to reduce the impurities, especially carbon, by optimizing the growth conditions and use low-strain quantum-dot material like GaInAs.

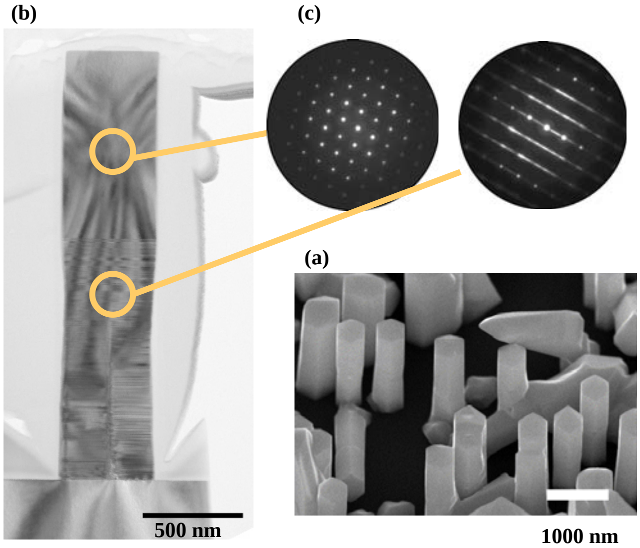

Fig. 2. (a) SEM image of the sample observed from 38 degrees from the normal direction; (b) HR-TEM image of the nanopillar; (c) SED patterns.

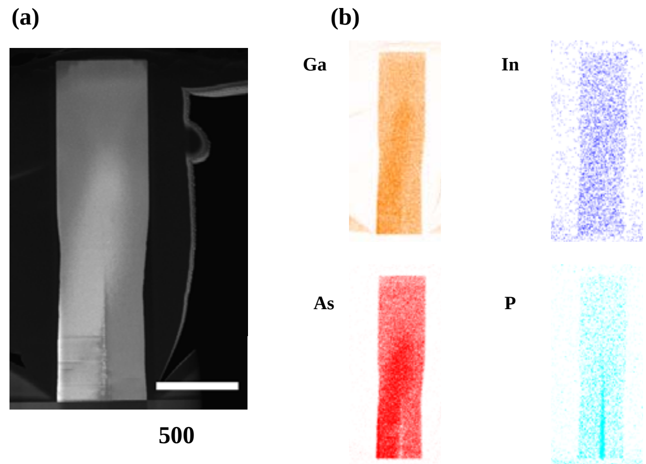

Fig. 3. (a) HAADF-STEM image; (b) elemental mapping by EDS obtained from K lines.

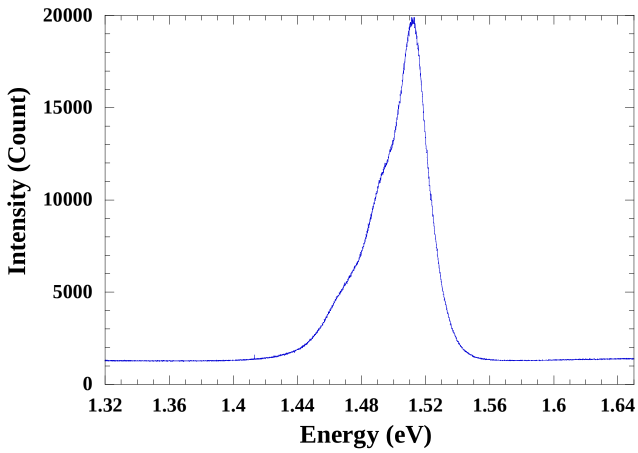

Fig. 4. PL spectrum of one nanopillar at 4 K.

4. Conclusions

We have demonstrated vertically grown nanopillars with flat tops on Si substrates and were able to make twin-free GaAs regions in the upper part of the pillars. An InAs quantum dot was positioned in the twin-free regions. InAs was difficult to detect by EDS and the clear emission from InAs was not seen in PL measurement. In the future, we would like to study the growth conditions to reduce impurities and try low-strain GaInAs quantum dots.

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