Bridging the gap between the nanometer-scale bottom-up and micrometer-scale top-down approaches for site-defined InP/InAs heterostructure nanowires

Guoqiang Zhang^{1,2}, Masato Takiguchi^{1,2}, Kouta Tateno^{1,2}, and Hideki Gotoh¹

¹ NTT Basic Research Laboratories, NTT Corporation, 3-1 Morinosato-Wakamiya, Atsugi, Kanagawa, 243-0198, Japan

Phone: +81-46-240-2827 E-mail: zhang.guoqiang@lab.ntt.co.jp

² NTT Nanophotonics Center, NTT Corporation, 3-1 Morinosato-Wakamiya, Atsugi, Kanagawa, 243-0198, Japan

This work presents a method that bridges the gap between the nanometer-scale bottom-up and micrometer-scale top-down approaches, which has long been a significant challenge for applications that require low-cost and high-throughput manufacturing processes. We realized the bridging by controlling the seed indium nanoparticle position through a self-assembly process. Site-defined InP nanowires were then grown from the indium-nanoparticle array in the vapor-liquid-solid mode through a "seed and grow" process. We also grew the uniform InP/InAs heterostructure nanowire array by the process and controlled the emission wavelength in 1.15-1.35 µm by tuning the thickness of InAs segments. Our work establishes a new paradigm of nanowire growth allowing integration into both a large-scale and low-cost process.

1. Introduction

The field of microelectronics has been at grips with the issue of miniaturization for decades as demand for ever-smaller products pressures manufacturers to make smaller components [1]. Photolithography, a top-down method, is limited by the diffraction limit of light. This issue has opened the door to the development of other methods for fabricating devices. Another competing method, self-assembly [2], is to rely on a bottom-up process driven energetically as the total system reaches a lower energy state. In theory, this approach could very well define the future of electronics manufacturing.

III-V compound semiconductor nanowires (NW) are successful example of the bottom-up approach [3-5]. Site-defined growth of III-V NWs, whereby NWs are grown in pre-determined areas on a substrate, is a common method. It usually entails growth in open windows on a mask made by electron-beam lithography and has been achieved. For many methods previously demonstrated, the diameter of the NWs is the same as the width of the window [6], therefore to achieve NWs with a uniform nanometer-scale diameter top-down techniques like photolithography are not feasible.

Our approach is to realize site-defined growth with high reproducibility using a two-step "seed and grow" process. An indium particle is formed on the exposed substrate windows and InP NWs are then grown from the seed particles [7]. The seed particle's size can be adjusted during the process once it is contained in the exposed substrate window. The ability to control this parameter means the particle can be made smaller than the substrate window in which it is seeded, thus making it feasible to prepare larger substrate windows within the limits of photolithography. It is particularly significant because it acts as a bridge of the gap between the conventional micrometer-scale top-down method (photolithography) and a common nanometer-scale bottom-up method like vapor-liquid-solid (VLS) approach.

2. Experiments, results, and discussion

Experiments

The InP substrates with circular open windows defined by SiO₂ masking film were fabricated by the photolithography technique. We synthesized the InP/InAs NWs in a metalorganic vapor phase epitaxy (MOVPE) system. Indium particles were formed on InP substrate by introducing trimethylindium (TMIn) source material. NW growth was initiated by introducing TMIn and tertiarybutylarsenic (TBA), or tertiarybutylphosphine (TBP) simultaneously. *Results and discussion*

Our procedure takes two main steps: a seeding step performed by diffusion followed by a growth step in the VLS mode. The first step of the growth process involves the breakdown of trimethyl-indium into indium and its other constituents. The Indium adatoms then diffuse into areas of the substrate that aren't covered by the SiO_2 film (Fig. 1). The two steps allow better control over the location and size of the grown NWs. This first step is crucial because the success of the growth step is determined by the morphology of the window region and location of the indium droplets on the substrate. Diffusion on the substrate surface will be temperature dependent following an Arrhenius-type relationship.

The ideal seeding conditions were found by varying temperature and the window spacing on the substrate while keeping deposition time constant at 10 minutes (Fig. 2). The best conditions found for seeding were 480 and 500 degrees with window spacing of 3 μ m. At these temperatures the indium adatoms had enough kinetic energy to diffuse to the open windows of exposed InP substrate, and eventually accumulated into the indium particles in the open windows. Site-defined InP NWs were then grown from the indium-nanoparticle array in the VLS mode through the "seed and grow" process (Fig. 3).

Finally, we confirmed that the developed method is applicable to grow the uniform InP/InAs

axially-heterostructured NW array by the established synthesis technology for the heterostructure NW [8-9] (Fig. 4a). We studied the optical property of the InP/InAs NW by the micro-PL set-up at 4 K [5]. The emission wavelength from the embedded InAs quantum disks was modulated to 1.15-1.35 μ m by the thickness of the InAs segments through quantum confinement effect (Fig. 4c). The uniformity of the emission sepctrum of 4 NWs suggests the high controllability of the technology developed in this work.

3. Conclusions

In summary, we present a method bridging the gap between the nanometer-scale bottom-up and micrometer-scale top-down approaches for the realization of site-defined InP/InAs heterostructure NWs. Successfully combining a controllable bottom-up growth technique with a top-down substrate preparation technique greatly improves the potential for mass-production and wide-spread adoption of this technology.

References

- [1] G. A. Brown, et al. Materials Today 7 (2004) 20.
- [2] G. M. Whitesides, B. Grzybowski, Science 295 (2002) 2418.
- [3] M.-H. Bae, et al. Crystal Growth & Design 14 (2014) 1510.
- [4] K. Tateno, et al. Nano Lett. 12 (2012) 2888.
- [5] M. D. Birowosuto, et al. Nature Mater. 13 (2014) 279.
- [6] D. Dalacu, et al. Nanotechnology **20** (2009) 395602.
- [7] G. Zhang, et al. Applied Physics Express 5 (2012) 055201.
- [8] G. Zhang, et al. AIP Advances 3 (2013) 052107.
- [9] G. Zhang, et al. Nanotechnology 26 (2015) 115704.



Figure 1. (a) Schematic diagram of absorption and decomposition of source materials, surface diffusion and accumulation of indium adatoms on the surface. (b) InP (211)B substrates with open windows of exposed InP area. The diameter of the circular open window, D, is 2 μ m. The window spacing (L) is 5 μ m in the image.



Figure 3. Top view SEM images of InP NWs grown from the indium particles deposited at 500 °C formed on InP (211)B substrate with the D of 2 μ m and L of 3 μ m. The roots of all NWs are located in the window region, indicating the seeding from the indium particles.





Figure 2. Indium particles deposited at various temperatures on InP (211)B substrate with varied window spacing (L). At low deposition temperature, there are indium particles formed on the SiO_2 film. With increasing temperature, the density of the indium particles formed on the SiO_2 film decreases because of the increased diffusion length. The distance between the adjacent windows (L) has marked effect on the distribution of indium particles. With decreasing distance (L), more indium atoms can diffuse and reach the InP open window area.

Figure 4. (a) SEM image (top view) of InP NWs with two InAs quantum disks grown on InP (211)B. The NWs are inclined to the substrate because of the <111> direction. (b) Schematic diagram of the NW structure in (a) and the band structure diagram of the NW. (c) Spectroscopy of 4 single NWs selected from the sample. The excitation laser power is 1 μ W with a laser spot diameter of 2 μ m. Two peaks (E₁ and E₂) are originated from the two InAs quantum disks, as indicated in (b).